CRYSTALLISATION RATE OF PENTAERYTHRITOL

by

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SUMMARY

The growth rate of Pentaerythritol crystals in aqueous solution has been measured over the temperature range 30 to 75°C by following the decrease in solution concentration of stirred crystal suspensions by refractometry. Measurements have also been made of the crystal mass increase in a fluidised bed for comparison at low relative crystal/solution velocities.

Two methods of crystal size analysis have been used : the Coulter Counter method and the image shear microscope method. The Coulter Counter theory has been corrected to allow for crystal shape and size, and the seed size distributions determined allowed for in the computation of crystal growth rates.

Commercial material containing two major impurities Di-P.E. and a Formal, exhibited an unusual transient overlap effect in the equilibrium solubility determination. The solubility approached from dissolution first obtained a peak enhanced concentration before attaining equilibrium. The equilibrium relationship for purified material was found to be represented by :

$$\log_{10} x = 4.980 - \frac{1242}{T}$$

where x = % mass fraction and T is in °K.

Growth rates (g), which ranged from about 10^{-8} cm/min to 10^{-3} cm/min were correlated with supersaturation (s) by the equation $g = k_L s^b$. It was found that b varied with the amount of impurity and temperature but had an average value of ca. 2. Values of k_L increased with temperature and the activation energy for commercial material was

found to be about 30 kcal/gmol. It was concluded that surface integration was the rate controlling process.

Heterogeneous particles were found to enhance growth rates but the absence of particles >0.45 micron resulted in brittle crystals. Although it was found that purification from Formal also enhanced the growth rate, the rate was very sensitive to traces (<0.1ppm level) of an unidentified third impurity which could not be easily removed.

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SECTION ONE

INTRODUCTION

Pentaerythritol, $C(CH_2OH)_4$, (referred to hereafter as P.E. for convenience) is a polyhydric alcohol produced as a white crystalline compound by the reaction of acetaldehyde and formaldehyde in the presence of an alkaline condensing agent. First discovered in 1882 it was not manufactured on a commercial scale until the early 1930's when it was used for the production of the explosive PETN (pentaerythritol tetranitrate). It was used extensively for this purpose during World War II, but its use has since grown rapidly in the manufacture of resins for surface coatings.

Side reactions often occur in the manufacture of P.E. yielding a product containing up to about 2% of the ether, Di-Pentaerythritol, and about 4% of an unidentified complex formal derivative, labelled the 'Formal' hereafter. These impurities are acceptable for the present market requirements, but are suspect in causing crystallisation difficulties during manufacture. During the final production stages the P.E. process solution is cooled in a batch crystalliser and this results in a P.E. product consisting of agglomerates of a few large crystals and a large number of small ones. The amount of this fine material is such (18 mass % $<$350 mesh) that it is very "dusty" and unpleasant to handle. The object of the present study was to obtain the data necessary for the design of a crystalliser capable of producing a dust free uniform sized product.

The usual procedure for the study of a crystallisation process is

to study the pure analar material, occasionally with the effects of known additions of impurities. The nature of this project is such that it necessitates a reverse approach.

A number of batches of P.E. were obtained containing varying amounts of the impurities di-P.E. and Formal. In the earier work (1) on P.E. crystallisation it was suggested that the 'formal' inhibited crystal growth below about 67°C. However no account was taken of the di-P.E. impurity, and it was realised that many parameters would have to be investigated before an understanding of the process to the extent of the evaluation of design criteria could be achieved.

SECTION TWO

SURVEY OF LITERATURE ON CRYSTALLISATION

2. 1. Solubility

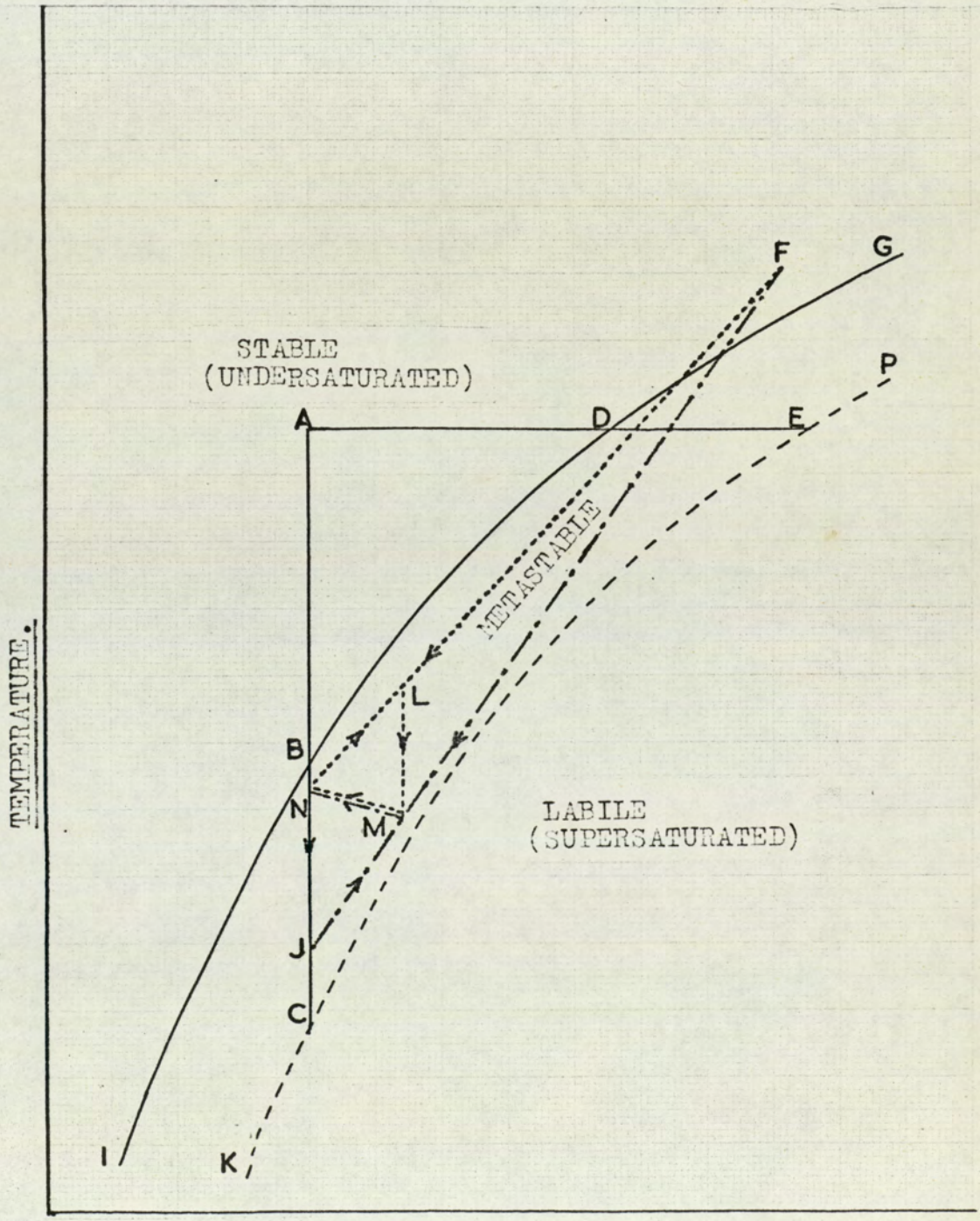
The first requirement for the study of any crystallisation process is a knowledge of the phase equilibrium diagram. A typical example of the type of diagram obtained for the system pure solute dissolved in pure solvent is shown by the line IBDG, called the "solubility" curve, in Figure 2.1. This curve defines the mass of solute which is in equilibrium with a given mass of solvent at various temperatures; the solvent is then said to be saturated with respect to the solute. For systems of more than one solute each must be studied individually.

A solution which contains more dissolved solute than that represented by the saturation composition is termed "supersaturated". Ostwald (2) seems to have been the first to introduce the terms "Labile" and "Metastable" zones which refer to the supersaturated solutions in which homogeneous nucleation may, and may not occur respectively.

Miers (3) did considerable research on this subject by studying the refractive index of solutions. Although he realised that factors such as the rate of cooling had an effect on the limits of supersaturation, he believed that supersolubility was a real property of solutions and melts under ordinary conditions. The supersolubility curve is shown in Figure 2.1 by the broken line KCEP.

FIGURE 2.1.

CONDENSED PHASE DIAGRAM.



For the purpose of illustration Figure2.1is considered as a "conserved property diagram" to a first approximation so that straight lines and the "lever-rule" may be used to show the effect of mixing solutions. This strictly requires that temperature is proportional to enthalpy per unit mass of mixture, and that the units of concentration are mass fraction (Spalding (4)).

If a solution in state A is cooled it remains undersaturated until temperature B is reached on the solubility curve. If further cooled it becomes supersaturated until temperature C on the super-solubility curve is reached; any further attempted cooling will produce spontaneous nucleation. The region between the solubility and supersolubility curves is the metastable supersaturated solution in which crystals (if present) are able to grow, but homogeneous nucleation does not normally occur. It is possible to produce supersaturation not only by cooling but also by evaporation or sometimes by the addition of another solute soluble in the solvent. The concentration then follows line A D E, Figure 2.1. The supersaturation curve is affected by many variables and is now considered to be a region of supersaturation rather than a definite curve, which is roughly parallel to the solubility curve.

Systems with appreciable increase of solubility with temperature are often crystallised industrially by means of a continuous cooling crystalliser. Two methods of adding the concentrated feed to a continuous cooling crystalliser are shown in Figure 2.1. where the solution conditions entering and leaving the crystalliser bed are represented by points M and N respectively. The conventional method

is the feed addition to the mother liquor leaving the bed (point N) before entering the cooler (dashed line LM). Alternatively the feed may be added after the mother liquor has been cooled (point J) prior to the mixture entering the bed (point M). This latter method gives the shortest residence time for what is probably the highest supersaturation. For the successful control of a continuous industrial crystalliser additional equipment is also needed for the removal (and redissolving) of fines generated in the system.

The phenomenon of supersolubility can be explained by the enhanced solubility of fine particles. Ostwald (2) found that if a solute was finely ground before dissolving in water, a solubility greater than the normal solubility was obtainable. He derived the equation which was later corrected by Freundlich (5) to the Ostwald-Freundlich equation:

$$\ell n \frac{c_r}{c_\infty} = \frac{2\sigma V_m}{R' T r} \qquad \ldots\ldots\ldots \quad 2.1$$

where c_r and c_∞ are the solubilities of the spherical particles of radius r and ∞ respectively, σ is the surface energy of the solid particle in contact with the solution, V_m is the molar volume of solute, T is the absolute temperature and R' is the gas constant. In the derivation of this equation it was assumed that the particles were spherical, the dissolved solid obeyed the gas laws, and that σ and V_m were independent of particle size. A number of workers have postulated corrections to the Ostwald-Freundlich equation; e.g. consideration of the energy contributions of edges and corners to

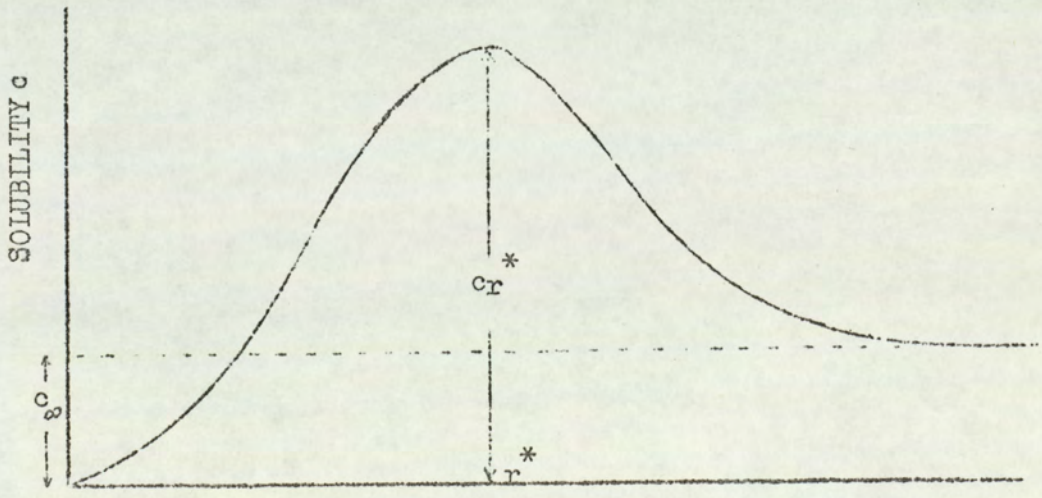
the total surface energy; allowance for the degree of dissociation or ionisation of the dissolved solid; and the variation of surface energy with particle size. However the equations deduced all postulated a continual increase in solubility with reduction in particle size.

Knapp (6) showed that, if the opposing effect of the electric charge on the surface tension of a particle was considered, the particles being assumed to be isolated charged spheres and their charge independent of size, then the Ostwald-Freundlich equation was modified to:-

$$\ell n \frac{c_r}{c_\infty} = \frac{V_m}{R' T} \left(\frac{2\sigma}{r} - \frac{q^2}{8 \pi \psi r^4} \right) \quad \ldots\ldots\ldots \quad 2.2.$$

where q is the particle charge and ψ the dielectric constant of the medium in which they are dispersed. From 2.2 the solubility can be shown to have a maximum when

$$r^* = \left(\frac{q^2}{4 \pi \psi \sigma} \right)^{\frac{1}{3}} \quad \ldots\ldots\ldots \quad 2.3$$



where r^* is the critical radius for maximum solubility c_r^*

However according to Helmholtz's theory there arises at the interface of disperse particles and the dispersion medium an electrical "double-layer". If then each particle is regarded as a double layer condenser, its electrical energy is given by $\frac{q^2 d}{2\psi\, r\,(r+d)}$

where q is the electrical charge on each layer and d is the distance between the layers. If d is negligible compared with r this reduces to $\frac{q^2 d}{2\psi\, r^2}$. The Ostwald-Freundlich equation then becomes:-

$$\ell n \frac{c_r}{c_\infty} = \frac{V_m}{R'T}\left(\frac{2\sigma}{r} - \frac{q^2 d}{2\,\Pi\psi r^5}\right) \quad \ldots\ldots \quad 2.4$$

and the solubility is then a maximum when:-

$$r^* = \left(\frac{5q^2 d}{8\,\Pi\psi\sigma}\right)^{\frac{1}{4}} \quad \ldots\ldots\ldots \quad 2.5$$

Dundon (7) found appreciable increases in the solubilities of 0.2 μ to 0.5 μ diameter particles of PbI_2, $Ag_2Cr\,O_4$, PbF_2, $SrSO_4$, $BaSO_4$, CaF_2, and he found that the solubility rose to a maximum on decreasing the particle size further. Roller (8) studied the solubility of gypsum and found that the solubility rate was proportional to the specific area at sizes above 25 μ, that between 25 μ and 2.8 μ the solubility rate increased more rapidly than the surface exposed and that below 2.8 μ the solubility rate began to decrease again.

2.2. Nucleation.

Crystallisation is a two step process involving first nucleation and then the growth of the nucleus to macro size. Nucleation involves the activation of smaller unstable particles called embryos. An embryo formed in the metastable region is very small and will dissolve on account of the increased solution potential. As the degree of supersaturation is increased the size of the embryo which can be tolerated by the solution decreases to a critical size where the embryo becomes a nucleus possessing sufficient excess surface energy to form a new phase and growth begins.

Two types of nucleation are apparent, Primary nucleation and Secondary nucleation. Most Primary nucleation processes occur heterogeneously as it is extremely difficult if not impossible to avoid extraneous nuclei. Van Hook and Frulla (9) found that by carefully preparing samples of 1 to 5 cm^3 of sucrose solution the metastable limit was raised to a supersaturation of about $s = 0.6$, where $s = \frac{c - c_\infty}{c_\infty}$ at ordinary temperatures as compared with the previously accepted limit of 0.2. The samples had to be prepared by careful dissolution followed by deactivation of latent nuclei by heating at temperatures at least 20°C above saturation, and sealing in closed tubes. They also averaged the observations of at least 50 droplets of solution and found that nucleation rate decreased to a limiting value of about one half the rate observed in the carefully prepared larger samples in the sealed tubes. These results seem to imply that this phenomenon is due to the diminished probability of smaller

samples containing foreign nuclei, indicating that heterogeneous nucleation probably occurs in most practical cases. The primary nuclei then grow in the supersaturated solution.

The growth of the primary nuclei in the supersaturated solution may be accompanied by the formation of fresh nuclei which is then termed secondary nucleation. This phenomenon of secondary nucleation has also occasionally been observed during the growth process of seeded supersaturated solutions, at a supersaturation below that required for primary nucleation.

2.2.1. Primary Nucleation.

2.2.1.1. Homogeneous Nucleation.

When nucleation occurs the transition from the metastable phase to the stable phase represents a decrease in the degree of molecular mobility, a decrease in the free energy of the system and so demands expenditure of energy to create the stable phase. The total quantity of work required to form the stable nucleus is the sum of the work required to form the surface and the work required to form the bulk of the particle.

Gibbs (10) was the first to show that the work of formation of a droplet from its vapour equals one third of that required to form the surface of the droplet. He showed that the total work required to form a droplet from its vapour, $w = a_p \sigma - v_p \Delta P$ where σ is the surface energy per unit area of a droplet of radius r

a_p = surface area of the droplet = $4\pi r^2$

v_p = droplet volume = $\frac{4\pi r^3}{3}$

ΔP = Pressure difference in the droplet = $\frac{2\sigma}{r}$

$$\therefore \quad w = \frac{4\pi r^2 \sigma}{3} \qquad \ldots\ldots\ldots \qquad 2.6$$

Similarly, for the homogeneous nucleation of a small particle from a solution, the excess free energy ΔG between the particle and the solute in solution is equal to the sum of the surface excess free energy ΔG_s, i.e. the excess free energy between the surface of the particle and the bulk of the particle, and the volume excess free energy. If ΔG_r is defined as the excess free energy per unit volume between a very large particle and the solute in solution,

then, $$\Delta G = \Delta G_s - v_p \Delta G_v$$

and for the spherical particle,

$$\Delta G = 4\pi r^2 \sigma - \frac{4}{3}\pi r^3 \Delta G_v \qquad \ldots\ldots\ldots \qquad 2.7$$

The maximum value of ΔG, designated ΔG^*, occurs at a critical size r^* (i.e. a critical number of molecules in the embryo) and represents the free energy of formation of the critical nucleus.

From equation 2.7, when $\frac{d \Delta G}{dr} = 0$

$$r^* = \frac{2\sigma}{\Delta G_v} \qquad \ldots\ldots\ldots \qquad 2.8$$

and $\Delta G^* = \frac{4}{3} \pi \sigma (r^*)^2$

This derivation assumes a spherical nucleus and consequently an isotropic σ. Cormia et al (11) have modified this assuming a cylindrical nucleus with different surface energies for the side and end. A spherical nucleus appears more reasonable in most cases, however, and for this the Ostwald-Freundlich equation (2.1) relates the supersaturation expressed as $\frac{c_r}{c_\infty}$ to the radius by:

$$\ell n \frac{c_r}{c_\infty} = \frac{2 \sigma V_m}{R' T r}$$

and so for the critical nucleus:

$$r^* = \frac{2 \sigma V_m}{R' T \ell n \left(\frac{c_r}{c_\infty} \right)} \quad \ldots\ldots\ldots \quad 2.9$$

and the free energy of formation of the critical nucleus becomes:

$$\Delta G^* = \frac{16 \pi \sigma^3 V_m^2}{3 (R')^2 T^2 \left(\ell n \left(\frac{c_r}{c_\infty} \right) \right)^2} \quad \ldots\ldots \quad 2.10$$

Kinetically the formation of nuclei can be assumed to be a series of bimolecular reactions of the form

$$A_1 + A_1 \rightleftharpoons A_2$$

$$A_i + A_1 \rightleftharpoons A_i + 1$$

$$A_{i+1} + A_1 \rightleftharpoons \text{critical nucleus.}$$

The rate of nucleation, j, i.e. the number of nuclei formed per unit time per unit volume may, since it is due to a random process, be expressed in terms of an Arrhenius type velocity equation:

$$j = C \exp\left(\frac{-\Delta G}{R' T}\right)$$

where C is a constant of proportionality.

Therefore for spherical nuclei:

$$j = C \exp\left(\frac{-16 \Pi \sigma^3 V_m^2}{3(R')^3 T^3 \left(\ln \frac{c_r}{c_\infty}\right)^2}\right) \quad \ldots\ldots\ldots \quad 2.11$$

Becker and Doering (12) proposed an equation of the form of Equation 2.11 with an analysis of the frequency factor or pre-exponential term C. They assumed that embryos of all sizes up to critical size achieve a non-equilibrium steady-state distribution by growth and decay processes, and introduced a non-equilibrium factor into the term C to allow for the backflux and decrease in embryo population caused by the growth of nuclei. This theory has been summarised by Van Hook (13).

Becker (14) proposed the nucleation rate equation:

$$j = C \exp\left(\frac{-\Delta G_d}{K_b T} - \frac{\Delta G^*}{K_b T}\right) \quad \ldots\ldots \quad 2.12$$

Where K_b is the Boltzmann constant and ΔG_d is the free energy of activation of diffusion. The alternative equations for the nucleation rate have been reviewed (13) (15) (16), but are generally

of the form of equation 2.11, ΔG_d being assumed constant over a limited temperature range.

2.2.1.2. Heterogeneous Nucleation.

The presence of a solid impurity in a supersaturated solution can act as a catalyst for nucleation and it has been shown that homogeneous nucleation is very difficult if not impossible to produce in practice. However, not all impurities in a particular system will act as accelerators and it is in fact possible for some to act as nucleation inhibitors.

The free energy of formation of the critical nucleus for heterogeneous nucleation $\Delta G^{*'}$ is related to the free energy of formation of the critical nucleus for homogeneous nucleation ΔG^* by:

$$\Delta G^{*'} = \beta \Delta G^*$$

where β is a factor less than unity.

Volmer (17) has related β to α (the angle of contact between the crystalline deposit and the foreign solid surface) which is analogous to the angle of wetting in liquid-solid systems:

$$\beta = \frac{(2 + \cos\alpha)(1 - \cos\alpha)^2}{4} \quad \ldots\ldots 2.13$$

When $\alpha = 180^\circ$, $\beta = 1$ and $\Delta G^{*'}$ is the same as for homogeneous nucleation.

When $\alpha = 0^\circ$, $\beta = 0$ and $\Delta G^{*'} = 0$ and nucleation is spontaneous.

When α lies between 0° and 180° $\Delta G^{*'} < \Delta G^*$ and so the impurity acts as a nucleation accelerator.

Preckshot and Brown (18) have studied the effect of

crystallographically similar, but insoluble, ionic crystals in nucleating quiet supersaturated solutions of potassium chloride. The time required for nucleation for various fixed degrees of supersaturation were measured conductometrically. They found that for the same time necessary for nucleation, lead sulphide promoted nucleation at a lower degree of supersaturation than an unseeded solution; lead telluride required even less supersaturation; and lead selenide was the most effective.

Telkes (19), working on the nucleation of supersaturated inorganic salt solutions, has contributed data to strengthen the theory that an additive will accelerate nucleation only if its crystallographic structure and that of the salt to be crystallised agree to within 15%.

2.2.1.3. Induced Nucleation.

Nucleation can be induced in supersaturated solutions free of extraneous material, below the supersaturation necessary for homogeneous nucleation. This can be done by the effects of external influences such as electric and magnetic fields, ultra-violet light, X-rays, sonic and ultrasonic radiation, cavitations produced by stirring and even the mechanical impact of a stirrer with the vessel walls. Of these ultrasonic radiation seems to be the most effective nucleator.

Ultrasonics, when applied to liquids, cause cavitations in the liquid alternately producing areas of high and low pressure. The frequency and power of the ultrasonic waves have to be carefully controlled for a particular process, as while the low pressure areas cause embryo coagulation, high intensity ultrasonics break up suspended

particles.

Van Hook and Frulla (9) found this effect in the nucleation of sugar solutions. They found that at a supersaturation ratio of 1·1 for which homogeneous nucleation would not occur, a sugar solution would nucleate on momentary irradiation of ultrasonics at a frequency of 8 k.c. and a minimum power input of $10W/cm^2$, yielding a prolific crop of crystals. However, at 340 k.c. very few crystals developed in the same time.

Mullin and Raven (20) also showed this phenomenon with stirred solutions. They found that the degree of supersaturation necessary for nucleation decreased with increasing stirrer speed only over a limited range, after which there was an increase before again decreasing with further increase in stirrer speed. They suggested that this increase was probably due to the fracture of nuclei at this critical stirrer speed yielding fragments of less than nucleic size.

2.2.2. Secondary Nucleation.

Miers (21) in 1911 observed that if a crystal was introduced into a supersaturated solution which was in a metastable state below that required for spontaneous nucleation, it may cause crystals to grow not only in contact with it but also at some distance from it. This phenomenon of nucleation occuring at a supersaturation below that required for spontaneous nucleation by the presence of other growing crystals is termed secondary nucleation.

Strickland - Constable and Mason (22) working on $MgSO_4 \cdot 7H_2O$ distinguished four classes of breeding of nuclei:-

(i) "Initial breeding" which occured when a seed crystal yielded a shower of small crystals, which were originally attached to it, after immersion in a supersaturated solution.

(ii) "True breeding" which resulted from broken portions of the dendritic or needle-like growth on the original seed.

(iii) "Splinter breeding" which occured when a needle broke off a mother crystal accompanied by a shower of small crystallites.

(iv) "Attrition breeding" which was that which resulted from agitation.

McCabe (23) investigating the growth of stirred, seeded supersaturated solutions of potassium chloride and copper sulphate found that the size distribution plots of seed and product crystals differed only by a displacement along the diameter axis, indicating no agglomeration or secondary nucleation.

Melia and Moffitt (24) however observed secondary nucleation with potassium chloride solutions although it did not normally occur until dendritic type growths appeared on the crystal surface. They suggested that it was produced by the shearing action of the solution on the crystals. This hypothesis was further strengthened when they worked on sodium chloride solutions in the presence of additives which promoted dendritic growth of the sodium chloride crystals. A large increase in the number of secondary nuclei produced was observed in the presence of the additives. They also found that these secondary nuclei were themselves capable of producing fresh nuclei.

Ting and McCabe (25) investigated the secondary nucleation characteristics of continuously cooled, stirred seeded solutions of

magnesium sulphate. They found that on cooling a solution a first crop of nuclei were produced at a certain temperature with insufficient heat release to change the cooling rate; then on further cooling a temperature was reached when prolific nucleation occured, completely obscuring the stirrer and accompanied by sufficient heat release to appreciably retard the rate of cooling. It appears therefore that there is a "supersaturation curve" for both primary and secondary nucleation. Both of these "supersaturation curves" were affected by seed size and weight, stirring and cooling rate.

Cayey and Estrin (26) studied the secondary nucleation period for magnesium sulphate and counted nuclei photographically after different conditions of seeding. They suggested (in contradiction to Melia and Moffitt (24)) that the secondary nucleation was dependent on the number and size of seed crystals, and on the level of supersaturation.

Secondary nucleation is a phenomenon which is still not fully understood. It appears that it only occurs for some particular systems and then only if a certain supersaturation level is exceeded. The experimental data available is limited and in certain cases conflicting.

2.3. Crystal Growth.

The following consecutive steps are required in any heterogeneous reaction:-

1. Transport from the medium to the reaction environment.
2. Absorption on the surface.

3. Orientation in the surface (reaction).

4. Desorption of products of reaction.

5. Dissipation of products of reaction.

For crystal growth from solution the last two steps consist of the dissipation of the heat of crystallisation which will be rapid compared with the relatively slow growth rate, and so step 1 is more likely to be rate controlling with respect to 4 or 5. A molecule on arriving at the crystal surface is not necessarily immediately incorporated into the crystal lattice because it may either diffuse away or it may not be at a favourable site. As the orientation of the molecule for the greater part of the entropy change step 3 is more likely to be rate controlling than step 2. So the two most likely rate controlling steps are:

1. Transport from the medium to the growing environment.

2. Orientation in the surface.

2.3.1. Diffusion Theories.

Noyes and Whitney (27) assumed that the liquid in contact with the crystal was saturated, and that crystallisation was the reverse of dissolution. They assumed that the rate at which a substance dissolves in its own solution was proportional to the difference between the concentration of that solution and the concentration of the saturated solution. Nernst (28) assumed that the crystal was surrounded by a laminar film of liquid of thickness, δ, through which the solute had to diffuse. Then:

$$\frac{dm}{dt} = \frac{D_L A}{\delta} (c - c_\infty) \quad \ldots\ldots\ldots \quad 2.14$$

where m = mass of solute deposited in time t

A = surface area of the crystals.

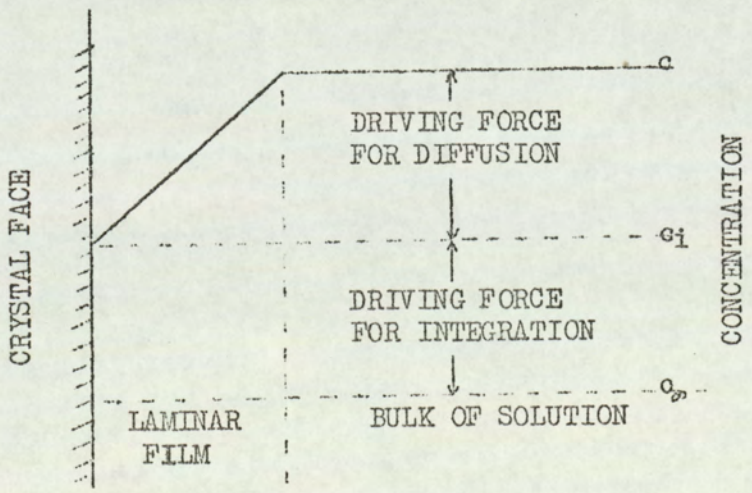
c = solute concentration in the bulk of the solution.

c_∞ = solute concentration of saturated liquor.

D_L = coefficient of diffusion of the solute.

However, this equation suffers from the defect that it assumes the liquid in contact with the crystal is saturated, whereas it was found by Miers (29) to be supersaturated.

Mullin (30) has shown how Berthoud (31) and Valeton (32) suggested that there were two steps involved in crystal growth: diffusion to the crystal surface and then an "integration" reaction (assumed to be first order) when the solute was incorporated into the crystal lattice.



These two stages can then be represented by

$$\frac{dm}{dt} = K_d A \left(c - c_i\right) \quad \ldots\ldots\ldots \quad 2.15$$

and $$\frac{dm}{dt} = K_r A \left(c_i - c_\infty\right) \quad \ldots\ldots\ldots \quad 2.16$$

where K_d is a coefficient of mass transfer by diffusion, K_r the integration rate constant and c_i the solute concentration at the crystal/solution interface.

c_i can be eliminated to present an overall crystal growth equation:

$$\frac{dm}{dt} = \frac{A \left(c - c_\infty\right)}{\frac{1}{K_d} + \frac{1}{K_r}} \quad \ldots\ldots\ldots \quad 2.17$$

or $$\frac{dm}{dt} = K A \left(c - c_\infty\right) \quad \ldots\ldots\ldots \quad 2.18$$

where $$\frac{1}{K} = \frac{1}{K_d} + \frac{1}{K_r}$$

Marc (33) found that as the stirring rate was increased the velocity of growth increased until after a critical rate was reached it remained constant. He considered that at this stage the crystal was covered with an adsorbed layer of molecular dimensions. Crystal growth is then assumed to be controlled by the surface integration step, equation 2.16, and $K \simeq K_r$, equation 2.18.

2.3.2. Surface Integration.

The following theories have given significance to the "K_r rate constant", equation 2.17, which makes allowance for the facility

with which a surface may incorporate a particle adjacent to it.

The first theories on crystal growth concerned the morphology of the crystals and an historical account of these earlier theories has been made by Buckley (34), from which the following has been abstracted:

" Curie (35) proposed that there was an intimate connection between the crystalline form and the surface energy of the solid. Each face has a specific free energy and the crystal will assume the habit giving minimum surface energy, such that $\sum_{i=1}^{i=n} A_i \sigma_i = \text{minimum.}$

" Volmer (36) based his theory on the existence of an absorbed layer around the crystal surface of molecular dimensions. While studying the growth of mercury crystals from the vapour state at low temperatures he observed the crystals growing layer by layer. He proposed that a molecule arriving at a crystal surface lost only a portion of its latent heat and was thus bound to the surface but had complete mobility on the surface. The adsorbed layer consists then of such molecules frequently colliding with each other forming larger two dimensional particles. When a particle becomes of nucleus dimensions it would attach itself to the crystal lattice. This is called Two Dimensional Nucleation. Volmer assumed that the transfer of the particle from the adsorbed layer to the lattice would be instantaneously made up from the solution. He proposed that the relationship between the growth velocities g_1 and g_2 of two differing lattice planes was given by:

$$\frac{g_1}{g_2} = C \exp\left(\left(H_1' - H_2'\right) n \Big/ R' T\right) \ldots\ldots \quad 2.19$$

where C is a constant, n a factor $\geqslant 1$, and H_1' and H_2' are the heats of adsorption of the two planes. He assumed that the heat of adsorption of a particular lattice plane was proportional to the specific surface energy.

" Brandes (37) making similar assumptions to Volmer considered the surface free energy to have little influence on crystal growth. He considered that the work of formation of the two dimensional nucleus was the controlling factor for growth, since the growth of the nucleus to complete the lattice plane was very rapid compared with the nucleus formation. The ratio of the growth velocities g_1 and g_2 on planes 1 and 2, where the work of formation of the nucleus is w_1, and w_2 was given by:

$$\frac{g_2}{g_1} = \exp\left(-\left(w_2 - w_1\right) \Big/ K_b T\right) \quad \ldots\ldots\ldots 2.20$$

where K_b is the Boltzmann constant.

" The work of formation was derived on a basis analogous to three dimensional nucleation.

" Bravais (38) postulated that the velocities of growth on lattice planes depends on the densities of the lattice points on the planes. However there are many criticisms to this theory.

" Both Kossel (39) and Stranski (40) proposed theories to account for the way in which atoms or molecules attach themselves to the crystal face. Kossel (39) assumed the crystal to build itself up by the indefinitely continued repetition of the most probable equivalent steps. He showed that it was immaterial to his theory whether the molecular attachments occured in rows parallel to a cube

edge or the diagonal. He expressed the attachment energy ∇_o as being made up of three components

$\nabla_o = \nabla' + \nabla'' + \nabla'''$ of which ∇' and ∇'' were tangential to the growth direction and ∇''' was at right angles to the lattice plane. Thus for the original two dimensional nucleus on a new plane the energy release was that due to ∇''' only. For this particular nucleus he found that for a homopolar crystal the most probable position of attachment is the interior of the plane, followed by the edge and the corner in lower degrees of probability; whereas for an ionic crystal the probability was in the reverse order, i.e. corner $>$ edge $>$ interior. Kossel stated that once the initial nucleus was attached the plane would build up rapidly to completion.

" Stranski (40) working independently and considering the relative work of separation necessary to remove molecules from various positions in the lattice plane came to the same basic conclusions as Kossel".

Nielsen (41) also considers this two-dimensional nucleation mechanism of growth. He classified two types of mechanism:

(i) The"mononuclear layer" mechanism where the time between two consecutive nucleations is greater than the time it takes for a surface nucleus to grow such that it covers the crystal surface.

(ii) The "polynuclear layer" mechanism where the surface nucleation is so fast that each molecular layer of the crystal is the result of intergrowth of numerous individually nucleated surface crystals.

However the probability of the formation of these two-dimensional nuclei is a very sensitive function of supersaturation,

and Burton, Cabrera and Frank (42) have shown that if reasonable values of the edge energy of the two-dimensional nuclei are assumed, a critical supersaturation of about $s = 50\%$, where $s = \frac{c - c_\infty}{c_\infty}$ is necessary for the formation of two-dimensional nuclei. Whereas growth has been known at very low supersaturations of $s = 1\%$ and lower.

Frank (43) recognised that growing crystals are not perfect flat plane faces, and that their imperfections will provide the steps required for growth making two-dimensional nucleation unnecessary. The face containing a "screw dislocation", i.e. the one in which the displacement is parallel to the dislocation line will then grow perpetually "up a spiral staircase". If there are two such dislocations on a face, growth will occur if the supersaturation is raised to a value such that the size of the critical two-dimensional nucleus correctly orientated will pass between two points in the positions of the two dislocations.

Burton, Cabrera and Frank (42) based their theory on the existence of these screw dislocations and considered the crystal growth process from the vapour to be a result of three separate processes, (i) exchange of molecules between adsorbed layer and vapour, (ii) diffusion of adsorbed molecules towards the steps and exchange with them and (iii) diffusion of adsorbed molecules in the edge of the steps toward the kinks (or growth sites along the steps) and exchange with them.

Strickland - Constable (44) has summarised this theory of Burton et al (42) for crystal growth from the vapour and shown that a

similar approach can be made for growth from solution, and the flux J' (mass transferred per unit area and unit time) is then given by:

$$J' = \frac{\beta' D_L}{a'} \frac{\tanh\left(\frac{y_o}{\sqrt{2}\,\gamma_s}\right)}{\left(\frac{y_o}{\sqrt{2}\,\gamma_s}\right)} \left(c - c_\infty\right) \quad \ldots\ldots\ldots \quad 2.21$$

where β' is a reflection coefficient of the molecules, D_L the diffusion coefficient, a' is the molecular spacing in the adsorbed layer, y_o is the distance apart of the steps and γ_s is the average diffusion distance of adsorbed molecules.

If y_o is assumed inversely proportional to $\left(c - c_\infty\right)$ then for high supersaturations, y_o is small, and $J' \rightarrow \frac{D_L \beta'}{a'} \left(c - c_\infty\right)$ whereas for low supersaturations, y_o is large, and $J' \rightarrow \frac{D_L \beta'}{a'} \left(c - c_\infty\right)^2$

Strickland - Constable (44) has reported that Reich (45) found a second order growth rate dependence on supersaturation for well stirred seeded solutions of $MgC_2C_4 \cdot 2\,H_2O$, $BaC_2O_4 \cdot 2\,H_2O$, and TℓBr.

Chernov (46) adopted a similar approach to Burton et al (42) and came to the same conclusion for the dependence of the normal (perpendicular) growth velocity, g, of a crystal face growing from a vapour being proportional to s^2 for low supersaturations and being linearly dependent on s for higher supersaturations. He showed the non-linear dependence on supersaturation to be obtained for $0.05 < s < 0.80$ when crystals of β - methyl naphthalene and p-toluidene were grown from the vapour. For the normal growth velocity from

solution Chernov assumed that matter is transferred to the crystal only on the end faces of steps and only by diffusion within the volume of a fixed boundary layer of thickness, δ adjacent to the crystal. He found that for the interval $0.01 < s < 0.2$ the following approximation was true:

$$g = k_{\delta} s^{b} \quad \ldots\ldots\ldots \quad 2.22$$

and at very low supersaturations b = 2. The exponent b increased as δ decreases, and the region of thequadratic equation is enlarged as the solution is stirred more vigorously.

Burton et al (42) have observed that in some cases a crystal surface will not grow at all, in spite of the fact that it is in contact with a supersaturated solution of $s \simeq 0.1$. This could either be due to the absence of dislocations in the crystal surface, or else to the presence of so many of them that the mean distance between them is too small for the particle integration. As this would require of the order of 10^{12} dislocations per cm^2 which is high, the former explanation is more probable.

2.3.3. Experimental Data.

In order to assess the contribution of the surface integration and the diffusional resistance respectively on the growth process it is necessary to try to eliminate one of them. As the relative crystal/solution velocity is increased the laminar film thickness, δ decreases until in the limit the surface integration step should control. Marc (33) found this effect with stirred suspensions of potash alum,

NH_3alum $K_2Cr_2D_7$ and $AgOA_c$ when after a certain stirrer speed was attained no further increase in growth rate was observed. He then found the growth rate to be proportional to the square of the degree of supersaturation.

Bransom et al (47) however found no such limit with the growth rate of seeded solutions of magnesium sulphate heptahydrate stirred between 100 revs/min. and 1000 revs/min. They found a continual increase in the mass transfer coefficient, up to an experimental limit of 30°C, with stirrer speed which was independent of seed size. They attributed this to "homogeneous isotropic turbulent eddies".

Cartier et al (48) modified an equation by Amelinckx (49) for the resistance to crystallisation due to the surface integration. The particle integration rate was based on a statistical determination of the rates of particle attachment and detachment at a crystal face. They obtained the equation

$$\frac{dm}{dt} = K_r' A \left(\exp \left(Z \left(c_i - c_\infty \right) \right) - 1 \right) \quad \ldots\ldots 2.23$$

where $$Z = - \frac{1}{k_b T} \frac{d \nabla}{dc}$$

K_r' = Particle Integration Factor

∇ = Attachment energy of the crystallising particles.

c_i = Solution concentration at the interface %m/v

k_b = Boltzmann constant.

Hence a plot of $\ell n\left(\frac{dm}{dt} + K_r' A\right)$ against $\left(x_i - x_\infty\right)$ which are the concentrations in mass %, should give a straight line of slope ρZ where ρ is the solution density. A value of K_r' has to be determined by trial and error which will give an intercept of $\ell n\, K_r' A$. It was found that K_r' and Z could be expressed in terms of the absolute temperature:-

$$K_r' = \alpha\, T^{\frac{3}{2}} - \beta$$

$$\rho Z = B - \frac{A}{T}$$

where α, β, A and B are constants.

Cartier et al studied the effect of relative crystal/solution velocity (u) on growth rate by direct measurements of a single crystal with a microscope. They found for citric and itaconic acids the diffusional resistance was insignificant at sufficiently high relative velocities and equation 2.23 then satisfactorily correlated their results.

Mullin and Garside (50) worked on the crystallisation of aluminium potassium sulphate using single crystal measurements as used by Cartier et al and measurements of weight and sieve analysis in a fluidised bed. They found a good agreement with the growth rates obtained by the two methods. They found that the growth rate was proportional to $u^{0.65}$ and that the dependence on supersaturation $(c - c_\infty)$ varied between $(c - c_\infty)^{1.4}$ and $(c - c_\infty)^{1.62}$ within the supersaturation limits $0.003 < (c - c_\infty) < 0.015$. However the exponent 1.62 remained fairly constant above a certain relative velocity.

Davis and Jones (51) used conductivity measurements to

determine the rate of growth of seeded stirred suspensions of silver chloride. They found that the growth rate was independent of stirrer speed between 100 r.p.m. and 500 r.p.m. and so assumed integration control, for which they found the dependence on supersaturation to be second order. No account was taken of the increase of crystal area during these experiments, but conditions were chosen such that the total change was only 0.1% of the original total surface area of the seed crystals.

Schierholtz (52) nucleated stirred solutions of calcium sulphate at 25°C and followed the decrease in concentration using titration measurements of samples of solution at regular intervals. He found that the crystallisation rate was first order with respect to $(c - c_\infty)$ with the exception of the early and final stages. No allowance was made for the effect of change of crystal area, but as the product crystals were of an acricular nature he suggested that the effective area available for crystal growth throughout this period would be constant.

McCabe (53) has proposed a method for the prediction of the size analysis of a mass of crystals grown from a mass of crystals of known size analysis, which is known as "McCabe's ΔL law". This states that if a known mass of seed crystals of known size distribution is grown under given conditions of supersaturation then the size analysis of the product is given by:

$$M_2 = \int_0^{M_1} \left(1 + \frac{\Delta D}{D_1}\right)^3 d M_1 \quad \ldots\ldots \quad 2.24$$

where M_2 is the product mass obtained from M_1 seed, D_1 is the seed size and ΔD the increase in size.

Solution of equation 2.24 involves tedious trial and error methods. A nomograph by Hooks and Kerye (54) has eased the solution by giving the ratio $\left(1 + \frac{\Delta D}{D_1}\right)^3$ for trial values of ΔD for any seed size D_1.

McCabe (23) added experimental validification to equation 2.24 by crystallising potassium chloride and copper sulphate by means of a seeded water cooled crystalliser tube agitated by rubber strips attached to a central shaft and revolved inside the centre of the tube. He showed that for these seeded suspensions over a wide number of variables, e.g. agitator speed, weight ratio of product to seed, temperature, etc., the size distribution plots of cumulative number percentage over size, D, for product and seed crystals, obtained from a sieve analysis differed only by a displacement along the diameter axis. This indicated that neither agglomeration nor secondary nucleation occured during the growth process, and that the linear velocity of growth was independent of crystal size. However this has since been shown to be true only for surface integration control.

As temperature has a greater influence on the kinetics of a reaction than on the physical property of the solution, the extent of the resistance to crystallisation presented by the surface integration can be seen by studying the effect of temperature on the growth rate. The effect is indicated by the value of the activation energy involved.

Van Hook (55) compared the activation energies of sugar solutions of viscosity and diffusion with growth with the conclusion

that the former two were considerably less than the third over the normal temperature range. The comparison was made at a constant supersaturation of s = 0.05 and the three values approached a common low level only at high temperatures. The high activation energy associated with growth was of the order normally associated with purely chemical reactions rather than physical processes (i.e. greater than 10 K cal/g. mole).

Rumford and Bain (56) determined the rate of growth of sodium chloride crystals in a fluidised bed for different supersaturations over the range 26 to 73°C. Below 50°C the rate of growth plotted against supersaturation was non-linear showing the growth rate to be surface integration control. Above 50°C the growth rate was linearly dependent on supersaturation. This could either be a first order surface integration or diffusion controlled growth. As the activation energy for crystallisation was found to be 5.4 K cal/mole, they considered the growth rate to be diffusion controlled above 50°C. Cooke, however, in a discussion (56) disagreed with this conclusion and suggested that the growth rate was diffusion controlled at all temperatures, but the contribution of the surface integration is greater at lower temperatures and supersaturations. He suggested that if the authors had continued their work for higher supersaturations the curves would have become linear for all temperatures.

McCabe and Stevens (57) found that equation 2.24 is inapplicable with low relative velocities in a diffusion controlled process with a crystal size distribution. They studied the rate of growth of copper sulphate pentahydrate crystals in an agitated solution following the

decrease in solution concentration using a conductivity cell. They found that the rate of growth g could be expressed in terms of relative velocity, u, between crystals and solution, the interfacial growth rate g_i, and the growth rate at zero velocity, g_o, by the empirical equation:

$$\frac{1}{g} = \frac{1}{g_o + Cu} + \frac{1}{g_i} \qquad \text{..... 2.25}$$

where C is a constant.

They found that the growth rate was not affected directly by crystal size, but, at low values of u, g is markedly influenced by the crystal - solution relative velocity. As u increases, the effect of velocity on growth rate diminishes and finally becomes negligibly small. This is consistent with the view that the growth process consists of a diffusion process and a surface integration in series.

Hixon and Knox (58) found the rate of growth coefficients to depend both upon the mass transfer coefficients which varied with fluid velocity and the rate coefficient of the surface integration. They measured the increase of weight of single crystals of copper sulphate and magnesium sulphate and calculated the product area from

$$A_2 = \left(\frac{M_2}{M_1}\right)^{\frac{2}{3}} A_1$$

They correlated their results on a dimensionless basis to allow the mass transfer coefficients to be compared with mass transfer coefficients or heat transfer coefficients of other systems:

$$\frac{K_d D}{D_m} = C\left(\frac{\rho u D}{\mu}\right)^{0.6} \left(\frac{\mu}{M_m D_m}\right)^{0.3} \quad \ldots\ldots \quad 2.26$$

These are the Sherwood, Reynolds and Schmidt numbers respectively:

where D_m = Molar Diffusivity.

D = Equivalent Diameter of the crystal.

K_d = Coefficient of mass transfer by diffusion.

M_m = mean molicular weight of the solution.

ρ = density of the solution.

μ viscosity of solution.

u = crystal / solution relative velocity.

C = a constant.

To correlate the data on this basis, it was necessary to assume that a resistance i.e. $\left(\frac{1}{K_r}\right)$ was being presented by a surface reaction of first order for magnesium sulphate and of second order for copper sulphate.

Bransom (59) has shown that for a fluidised bed the growth velocity g can be correlated in terms of a modified Reynolds number Re′:

$$g = \frac{dr}{dt} = C \left(Re'\right)^n \left(c - c_\infty\right)^b$$

where $Re' = \frac{2r \rho u}{\mu'}$, r = crystal equivalent radius, ρ = solution density and C = specific growth rate.

Using the data of Hixon and Knox (58) he found b = 1 for both copper and magnesium sulphate, n = 0.65 for copper sulphate and n = 0.3 for magnesium sulphate. He further showed that for a given

continuous crystallisation process, μ and ρ are constant and u varies very little so that:

$$g = C\ r^n \left(c - c_\infty\right)^b \qquad \ldots\ldots \qquad 2.27$$

and when the growth rate of crystals is expressed in this form most of the important operating parameters can be predicted.

Bransom and Palmer (60) working on an Oslo type of crystalliser found the exponent n = 1.0 when calculating average growth rates for a bed of crystals. However when size analyses were done on the individual beds before and after growth it was found that n = 1.5. This was explained by the size classification occuring in the bed.

Bennett (61) has used the data of Rumford and Bain (56) to obtain a value of n = 0.171 for the same correlation, equation 2.27.

However the exponents b and n will vary according to the type of system and the material used.

Bennett and Fentiman (62) found for the crystallisation of sucrose crystals that the rate constant differed for three different size fractions of seed used by a factor of two. After washing the seed in aqueous methanol the rate constants were reduced and the three size fractions then gave a similar value. When the specific surface area of each type of seed was measured, however, by the krypton adsorption method this showed that the difference was due to the surface roughness of the seed caused by adhering dust particles. The ratio of surface areas then measured for each type of seed, before washing, was similar to the ratio of the rate constants. They also pointed out that the relationship between total mass, or total volume,

and total surface area for a polydisperse system of normal distribution undergoing crystal growth is probably not $M \propto A^{1.5}$ but is closer to $M \propto A^{1.3}$. However this relationship will depend on the size distribution of the original seed.

2.3.4. Impurities in Crystal Growth.

2.3.4.1. Effect of Impurities.

The interaction of growing crystals with impurities is thought to give rise to effects such as change of growth rate, impurity capture and crystal defect formation, e.g. internal strains and dislocations.

The presence of an impurity usually causes a lowering of the growth rate. Buckley (34) has stated that the strongest effect is usually produced by organic substances of high molecular weight. An important characteristic of this kind of impurity is its inability to influence appreciably the dissolution rate of crystals. Certain inorganic ions are also effective when present in very small quantities. Increases in growth rate have been experienced in a few cases, and accounted for either by the catalytic effect of an impurity or by lowered surface energy, the smaller size of the critical nucleus and the enhanced probability of two-dimensional nucleation.

McCartney and Alexander (63) worked on the crystallisation of calcium sulphate by following the concentration change of seeded solutions using a dip-type conductivity cell. They found a second order dependence on supersaturation for both pure and impure solutions, regardless of the amount of inhibition of growth. They also found

that the additives which interact with calcium sulphate are those with polar groups on a chain structure, particularly proteinaceous and polycarboxylic materials. Also the retarding power increased markedly with molecular weight. Polyacrylic acid, for instance, completely inhibited growth at a concentration of 1.3 p.p.m., and even in 0.13 p.p.m. it retarded the growth rate by 74% compared with that of pure calcium sulphate. They found that if a strong acid was added to one of the polycarboxylic materials, the latter's ionization was suppressed and the retarding action largely destroyed. Although for HCl and H_2SO_4 the crystallisation rate was still retarded by the action of these acids themselves.

Chernov (46) attempted to explain the effects of impurities on the spiral growth of crystals, and based his treatment on the layer growth of crystals resulting from the motion of steps. He considered two possible mechanisms of inhibited crystal growth:

(a) Strongly adsorbed impurities captured by a growing crystal.

(b) Impurity poisoning of active growth sites (kinks).

He suggested that an impurity slows the advance of elementary steps whose heights are comparable with that of the adsorbed impurity molecules, and that this effect becomes less noticeable as the step height increases. The influence of impurities on the normal growth rate must therefore depend particularly on the mean step height.

Chernov stated that when adsorbed impurities have a short lifetime on the crystal surface, the impurity poisoning of active sites becomes important and it is then practically impossible to

incorporate new particles into the crystal in the poisoned kinks. He mentioned that kinks in a step are poisoned by impurities in both growing or dissolving cystals.

Impurity molecules captured by a growing crystal differ in size from the crystal constituents and therefore induce tensile strains in the lattice.

2.3.4.2. Equilibrium and non-equilibrium capture of impurities in crystal growth.

Chernov (46) stated that when the system consisting of a crystal and the surrounding medium contains an impurity, the impurity concentration x_s in the crystal and x_w in the medium, under equilibrium conditions are related to the phase diagram. The equilibrium coefficient of distribution (or capture) K' is then given by:

$$K' = \frac{x_s}{x_w}$$

If a crystal grows very slowly its impurity concentration is determined by the equilibrium capture coefficient, and by x_w^* at the interface. When K' is less than unity (crystal rejects impurity) x_w^* will increase with growth rate. Therefore the equilibrium capture coefficient increases effectively with the growth rate.

Chernov mentioned that the equilibrium impurity concentration is not constant throughout a crystal, since a difference exists between its value near the surface and that in the bulk. He added that the equilibrium concentration can be characterized approximately by three quantities: the concentration in the bulk of the crystal,

that in the surface layer, and that in the steps, and that these can differ considerably from each other. If a crystal grows very slowly an equilibrium concentration exists in all three quantities. At high growth rates equilibrium is not established in the bulk, but it may be in the surface layer and the steps, or only in the steps. However each surface layer becomes an interior layer with an equilibrium impurity concentration. This also applies to the line of atoms forming the end face of a crystal. At still higher growth rates none of the three equilibrium concentrations is achieved. Therefore when new layers are deposited on the crystal surface the impurity concentration in these layers will not generally be in equilibrium, and impurity diffusion from or to the crystal will begin.

Although it is usually assumed that in the solution the ratio of impurities to the substance being crystallised is greater than in the crystal (i.e. "purification") Botsaris et al (64) showed that this is not always the case. They investigated the incorporation of lead molecules in crystals of KCl and concluded that a possible mechanism is one of non-equilibrium capture of impurity, the magnitudes of the distribution coefficient depending on the rate of crystal growth, and the rate of diffusion of impurity through the lattice structure.

SECTION THREE

PREVIOUS WORK DONE ON PENTAERYTHRITOL

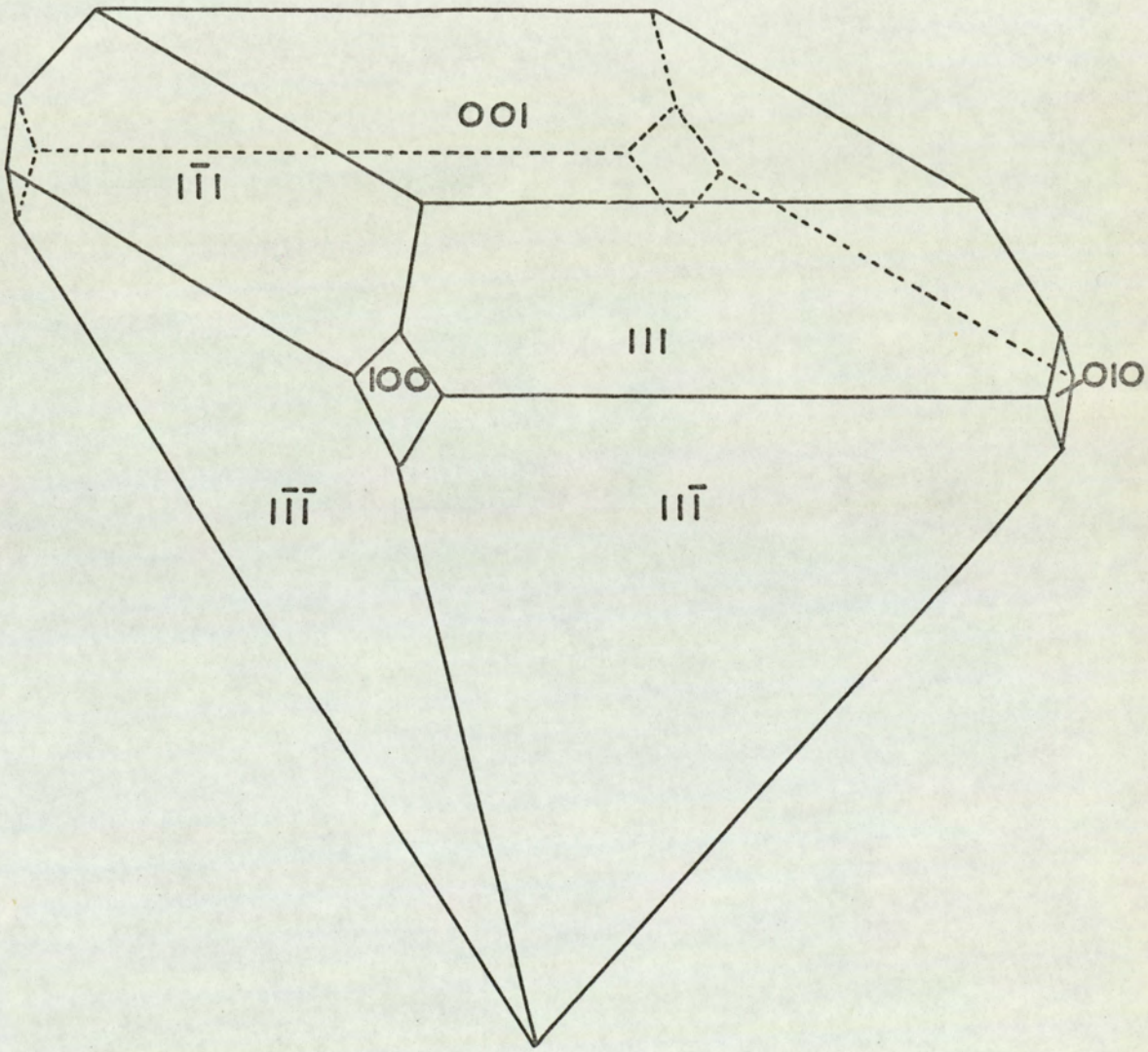
3.1. Crystal Structure.

Berlow, Barth and Snow (65) state that P.E. has a body centred lattice of tetragonal symmetry with two molecules in the unit cell. The crystal has a four fold alternating axis of symmetry parallel to its c- axis. The central carbon atom of one molecule in the unit cell is at (o, o, o) and that of the other at $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$. The molecular units are so arranged that the oxygen atoms are in planes perpendicular to the vertical c- axis. The oxygen atoms of four neighbouring molecules are arranged in the form of a square whose sides are inclined 10° to the a and b axes. In P.E. the C-C bond length is 1.50 Å, the C-O bond length is 1.46 Å and the O-O distance is 2.69 Å.

Von Groth (66) states that P.E. crystals should be tetragonal bipyramids on (001). He showed the morphology as figure 3.1. where the Miller indices of the main faces are marked. This is a 1 st order (h h l) class 4/mmm tetragonal bipyramid, as shown by Bishop (67). Von Groth gave the ratio of the unit cell axes as $\frac{c}{a} = 1.0236$, and this ratio is also recorded in the Barker Crystal Index (68). If however the bipyramid base is orientated at 45° to the a, b axes, this 2nd order orientation gives the equivalent axes ratio of $\frac{c}{a} = 1.447$, which is almost in agreement with Berlow et al (65) who recorded the values a = 6.10, c = 8.73 and hence $\frac{c}{a} = 1.43$. Wyckoff (69) also records values which give a similar ratio, of a = 6.083 ± 0.002 Å and c = 8.726 ± 0.002 Å and hence $\frac{c}{a} = 1.4345$. The melting point has been recorded

FIGURE 3.1.

P. E. CRYSTAL MORPHOLOGY..... VON GROTH (66)



as 260.5°C by Wyckoff (69) which compares favourably with the value of 259°C found for Pure P.E. in the previous work (1), whereas Von Groth (66) records the melting point as 253°C and so the data of Wyckoff is taken to be more accurate.

Figure 6.1.A shows a sketch of the tetragonal bipyramid in the 2nd order (h o l) orientation with the indices of the main faces marked and also of the minor ones which sometimes appear. For the Wyckoff ratio of $\frac{c}{a}$ = 1.4345 if the bipyramid base is taken as unit length, the length of the remaining (1 0 1) sides become 1.0074 and the normal angle between the (1 0 1) and (1 0 $\bar{1}$) faces is 69.67°. For practical purposes and within experimental error therefore, the external geometry may be considered as if the crystal is of the cubic system with each (1 0 1) face an equilateral triangle and the normal angle between any two faces being 70.53°. This has been done in section 6, figure 6.1.B.

Wyckoff (69) also states that there is a phase change at 179.5°C above which the unit cell becomes cubic and tetramolecular with a = 8.963 Å. This cubic modification of P.E. was also found by Nitta and Watanabé (70) at 180°C using X-ray diffraction measurements.

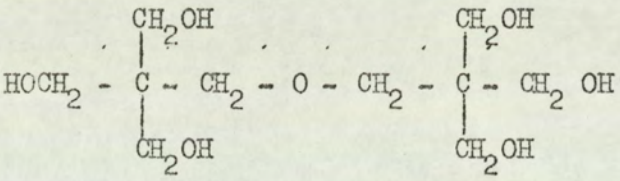
3.2. Impurities

3.2.1. Nature of the impurities

Two main impurities (>0.1%) were analysed chromatographically in the commercial Pentaerythritol. These were the ether Di - Pentaerythritol, and a complex unidentified by-product of

formaldehyde and P.E. which, for the purpose of this work is labelled the "Formal". The amount of Di- P.E. present varies from 0% to about 2% whereas the Formal present is usually about 4%. As the Formal identity is unknown this analysis figure is only comparative in relation to Di - P.E., obtained by giving the Formal the same chromatograph response factor as Di - P.E.

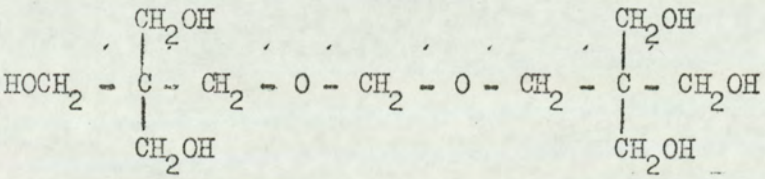
Di - Pentraerythritol is a white odourless crystalline ether, having the formula:



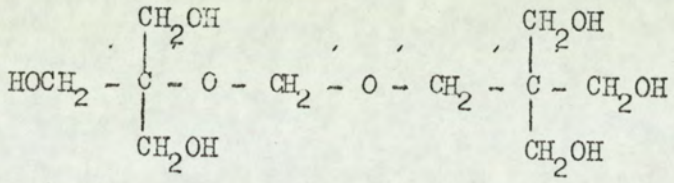
with a molecular weight of 254.

Similar compounds to the unidentified Formal have been reported in the literature as being present in commercial P.E.

Salkind et al (71) and Wiersma et al (72) consider this to be the compound Bis-pentaerythritol monoformal with a molecular weight of 284 and a hydroxyl value of 35.9%.



Barth and Snow (73) however have reported the detection of a similar impurity which they identified by carbon and hydrogen determinations, hydroxyl value, molecular weight and saponification value as formaldehyde bipentaerythritol acetal having the formula:



with a molecular weight of 270 and hydroxyl content of 37.8%

3.22. Preparation of Formal

The degree of polymerisation of formaldehyde in the aqueous solution can be expressed in the form of the following reactions:

$CH_2O + H_2O \rightleftharpoons HOCH_2OH$
Methylene glycol

$2\ HOCH_2OH \rightleftharpoons HO\ (CH_2O)_2H + H_2O$
dimer

$HOCH_2OH + HO\ (CH_2O)_2H \rightleftharpoons HO\ (CH_2O)_3H + H_2O$
trimer

Salkind et al (71) have proposed two possible sequences of reactions which account for the formation of di- P.E. and bis - P.E. monoformal. The first is based on the fact that very little formaldehyde exists in aqueous solution as free H C H O but it is mostly in the hydrated form. The reaction proceeds by splitting out water between these polymers and acetaldehyde. In the sequence of reactions methylene glycol forms P.E., the hydrated dimer forms Di - P.E. and the trimer by a similar sequence forms bis - P.E. monoformal. The other proposed sequence recognises the low concentration of H C H O in aqueous solutions but considers its high reactivity compared to that of its polymers. The formaldehyde reacts with acetaldehyde to form acrolein, two molecules of which then react with methylene glycol to form bis - P.E. nonoformal.

In the previous work (1) the attempted extraction of Formal with n - propanol using the method of Barth and Snow (73) gave a product which analysed chromatographically as containing only 15% Formal. In view of Salkind's (71) suggested sequence of reactions for the formation of the impurities, an attempt was made (1) to synthesise the Formal using P.E. and Formaldehyde. At first the formaldehyde was used in the form of the commercial 40% aqueous Formaldehyde solution, but after refluxing with impure P.E. (Batch A) this resulted in a decrease in Formal content. This was attributed to an inhibiting effect of the methanol present as a stabliser in the formaldehyde solution.

The formaldehyde was therefore added in the form of paraformaldehyde (a mixture of low molecular weight polyoxymethylene glycols) to aqueous P.E. solutions. Walker (74) recorded various equilibria occuring in dilute and concentrated aqueous formaldehyde solutions for different pH ranges, and reported that good yields of formals were obtained by heating alcohols with paraformaldehyde at 100°C in the presence of ferric chloride. Experiments were therefore carried out (1) under different conditions of pH and formaldehyde concentration, and with ferric chloride sometimes added in an attempt to synthesise the compound. All solutions were found to be cloudy at first but cleared after about 20 minutes, which was attributed to the slow depolymerisation rate of the paraformaldehyde. During the reaction the solubility of P.E. was found to be greatly enhanced and concentrations of ca. 58 mass % were often used. The pH of the solution did not appear to effect the formation of the Formal although the presence of ferric chloride

seemed to favour the formation of Di - P.E. It was found that the critical condition for the synthesis of the formal by the reaction of P.E. with formaldehyde is a long refluxing time. 70 g of Batch A (with 4.73% Formal and <0.1% Di - P.E.) refluxed with 10 g of Paraformaldehyde in 50 cm^3 water for 40 hours yielded on cooling to room temperature and filtering, 21 g of product crystals analysed chromatographically as containing 25% Formal and 0.5% Di - P.E. However this product contained other by-products ofreaction and it was decided to study the effects of Formal on the crystallisation of P.E. by using mixtures of the batches of material with known impurities, with the purified P.E., thus limiting the impurity content usable to about 5% Formal.

3.2.3. Purification of P.E.

The commercial batches of P.E. were purified by dissolving sufficient material in 10% (w/v) HCl to form a saturated solution at its boiling point. The solution was refluxed for 1 hour, cooled to 0^oC, filtered and washed in ice cold water. This procedure was then repeated and the resulting P.E. recrystallised from distilled water. The product was then washed with successive quantities of ice cold water and dried in an oven. The chromatographically "Pure P.E." analysed as containing <0.1% Di - P.E. and <0.1% Formal will be referred to as Pure P.E. hereafter.

3.3. Physical Properties

PENTAERYTHRITOL : $HOCH_2 - C(CH_2OH)_3$

Berlow et al (65) state that P.E. is a polyhydric alcohol with four primary hydroxyl groups arranged compactly around a central carbon atom. It is an odourless, white crystalline compound which is non-hygroscopic, practically non-volatile and stable in air. Its density is 1.396 g/cm^3. The entropy of transition of P.E. is 22.8 e.u., its entropy of fusion is 3.2 e.u., and its entropy of sublimation is 60.8 e.u.

The diffusion coefficient of P.E. in water at 20°C is 0.573 cm^2/s at a normality of 0.4 and 0.589 cm^2/s at a normality of 0.2.

P.E. is moderately soluble in cold water and freely soluble in hot water. Values of 5.6 mass% and 30.5 mass% were found in the previous work (1) at 20°C and 80°C respectively. The solubilities in aqueous solution found for Pure P.E. and for impure P.E. above 50°C were in good agreement with Cooke's data (75) and were correlated by the equation $\log_{10} x = 5.072 - \frac{1266}{T}$ where x = mass% and T = degrees Kelvin, which is shown in figure 3.2. Berlow et al (65) report that P.E. is only slightly soluble in alcohols and other organic liquids. The nucleation correlations (figure 3.2) of P.E. in aqueous solution were found to be (1) :

Pure P.E. $\log_{10} x = 2.289 - \frac{633}{T}$

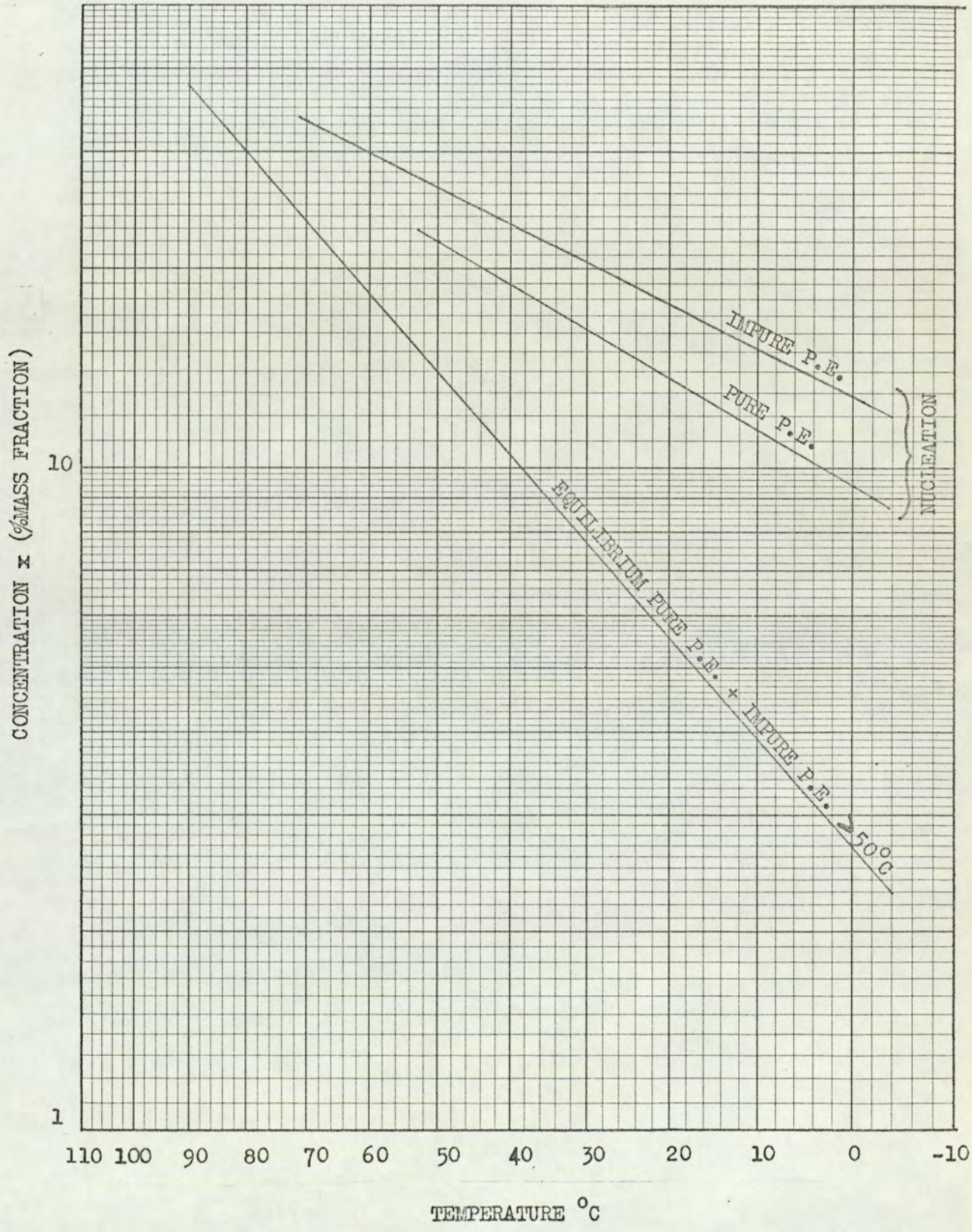
Impure P.E. $\log_{10} x = 3.112 - \frac{545}{T}$

I.C.T. (76) has reported the heat of combustion as 661 Kcal/mol and the equivalent conductance as 1.71 at 25°C and 0.06 g mol. / dm^3.

According to Bradley and Cotson (77) the vapour pressure of P.E. ranges from 2.12×10^{-5} cm H g at 106.4°C to 52.4×10^{-5} cm H g at 135.1 °C and is represented by the equation $\log p = 15.17 - \frac{7528}{T}$.

FIGURE 3.2.

EQUILIBRIUM AND NUCLEATION OF P.E. IN AQUEOUS SOLUTION (1)



Nitta et al (78) stated the vapour pressure of P.E. is given by $\log p = 14.525 - \frac{6861.}{T}$. Bright and Carson (79) give the heats of solution of P.E. in water as:

g mol P.E./500 g mol Water	Differential molar heat of solution K cal/g.mol solute
0.381	- 5.45
1.216	- 5.17
2.117	- 5.25
3.025	- 5.34

Where the thermochemical sign convention is used, i.e. the minus sign means absorption of heat.

Berlow et al (65) have recorded the variously reported melting points of P.E. as ranging from 256°C to 265.5°C. They state that P.E. exhibits a polymorphic transformation variously reported between 180°C and 192°C. Wyler and Wernett (80) report that P.E. forms eutectic with 35% Di - P.E. melting at 190°C.

In the previous work (1) this eutectic was found with 40% Di - P.E. at 185.5°C, and the binary melting system is shown in figure 3.3. where the temperature (°C) is the correct temperature after the thermometer calibration. The discrepancy in the eutectic point could possibly be due to the 4% Formal analysed chromatographically as being present in the Di - P.E. source.

The Binary melting system Formal/Pure P.E. studied in the previous work (1) after extraction with n - propanol to obtain a 15% Formal composition is shown in figure 3.4. It was found that the Pure P.E. containing $< 0.1\%$ Formal and $< 0.1\%$ Di P.E. melted at 259°C, and with

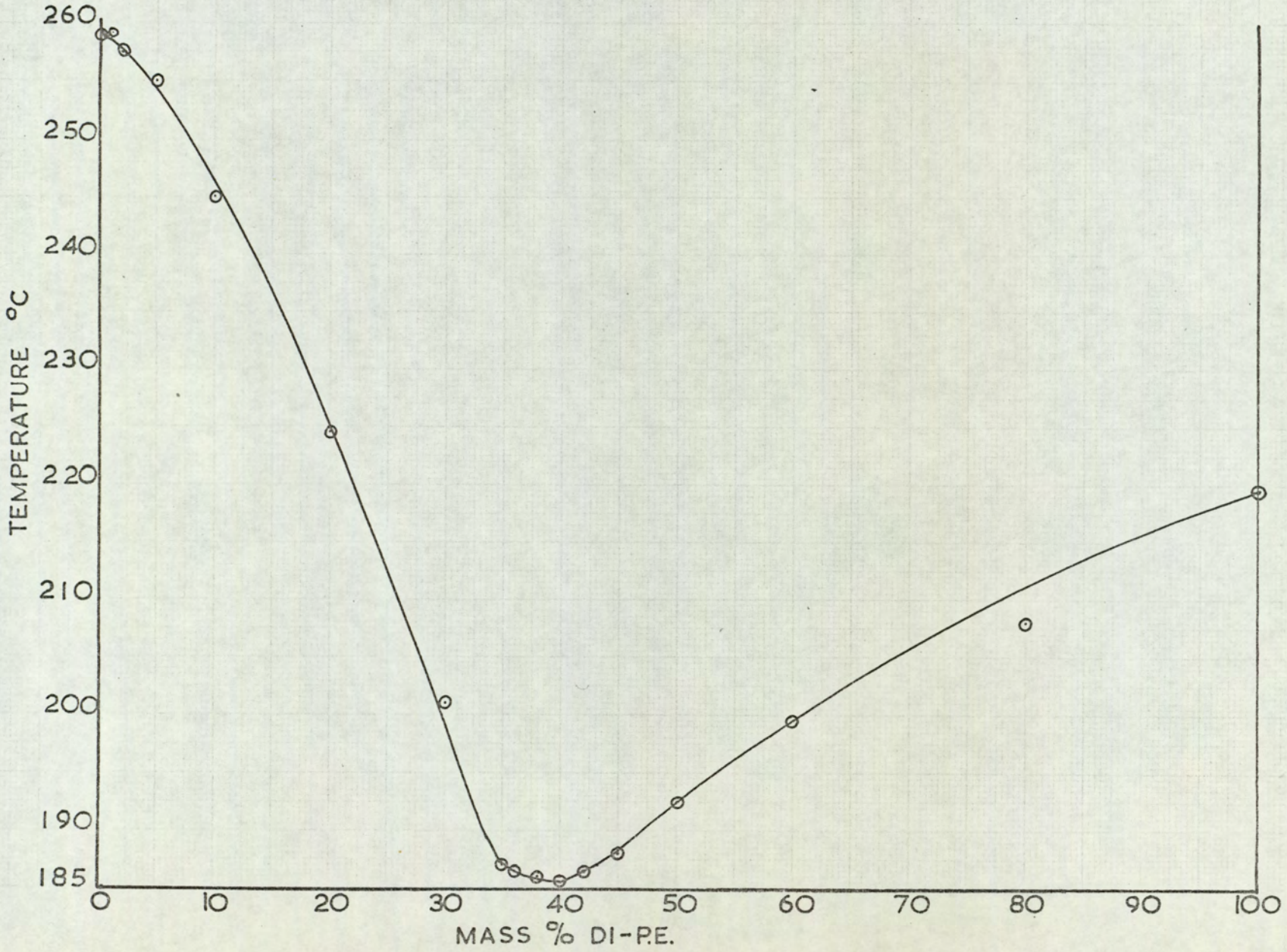
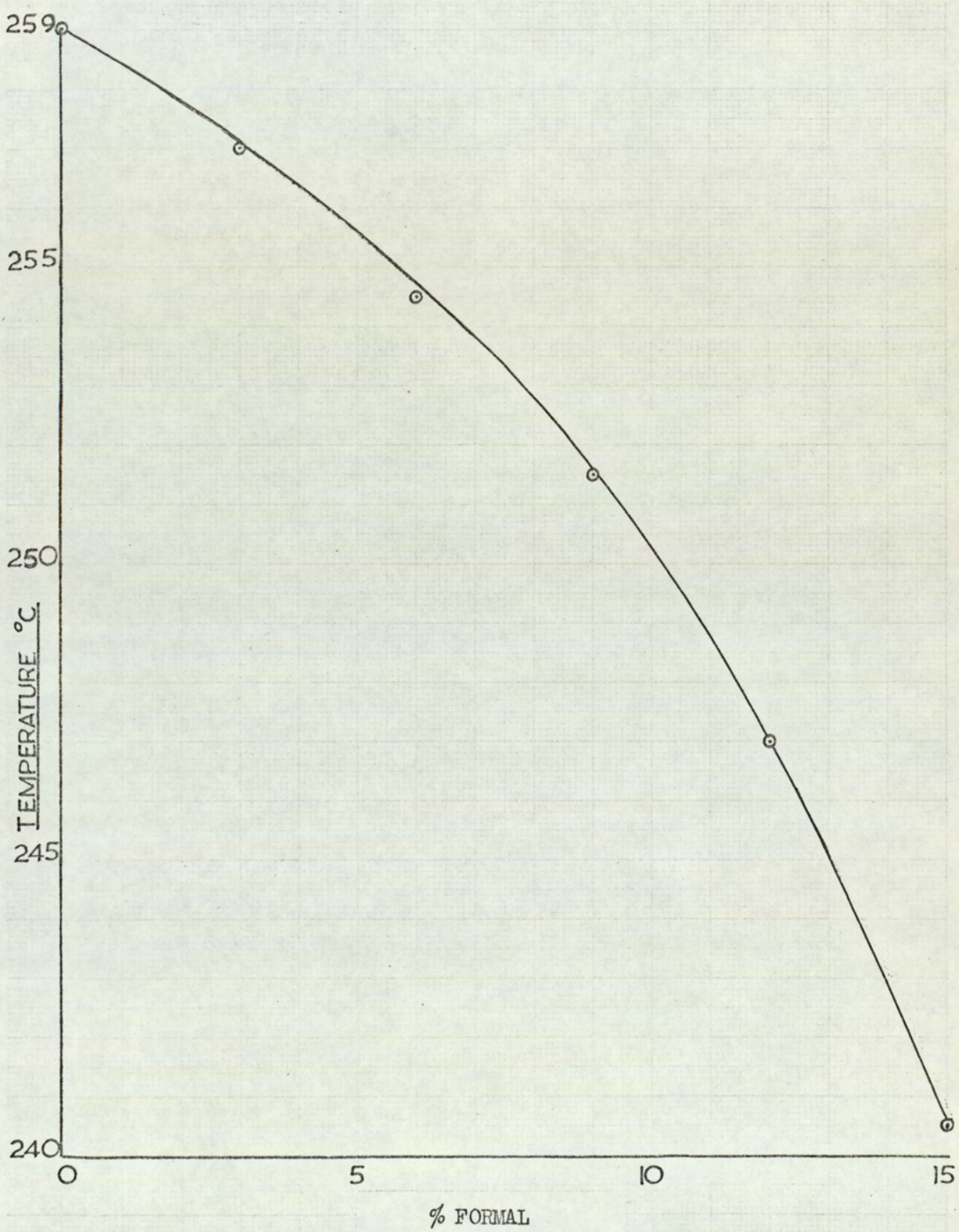


FIGURE 3.3. DI-P.E./P.E. BINARY MELTING SYSTEM

FIGURE 3.4.

FORMAL/P.E. MELTING SYSTEM



the 15% Formal present the melting point was depressed to 240.5°C.

Figure 3.5 shows the theoretical fluidisation and transport velocities for monodisperse spheres of density 1.4g/cm^3 in water. Although corrections are necessary to allow for the P.E. crystal shape and the solution properties, it serves as an indication of P.E. crystal fluidisation characteristics.

3.4 Chemical Analysis

3.4.1. Introduction

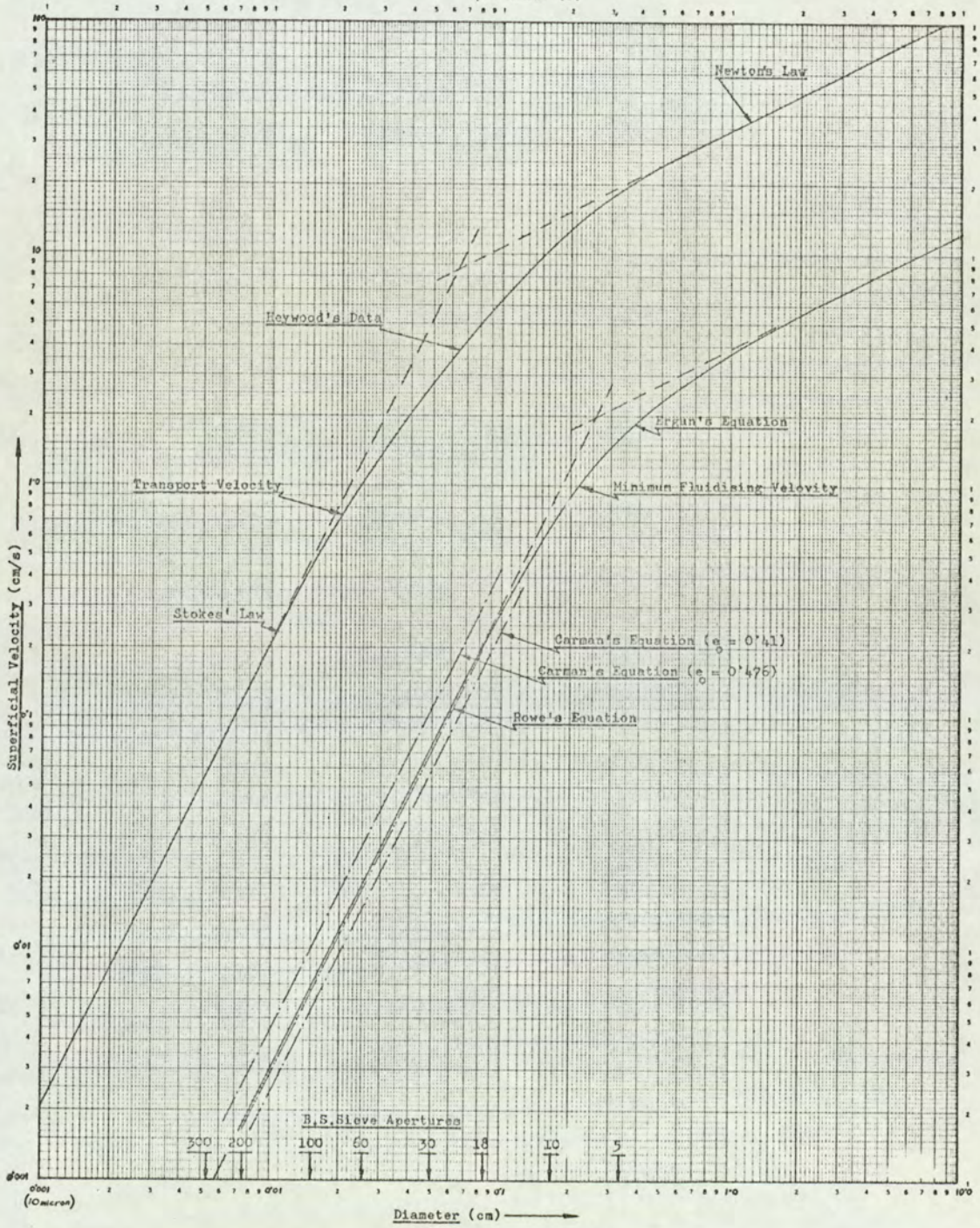
Berlow et al (65) state that the analysis of technical P.E. generally includes the following determinations: P.E. content, melting range, hydroxyl content, ash content, acidity, moisture content, water solubility, colour and physical state. The method given for the determination of P.E. content is the Benzal method based on the formation of the di- benzylidene acetal, a crystalline compound which is relatively insoluble in a dilute aqueous methanolic solution of hydrochloric acid containing benzaldehyde. The acetal precipitate is flitered off and the % P.E. calculated from the weight of filtrate. However the method is known to be inaccurate, and the analysis obtained for P.E. from the manufacturer's normally includes the main impurity concentrations Di - P.E. and Formal found by the acetal chromatographic method (section 3.4 2) and the P.E. content is then found by difference.

3.4.2. Acetal Chromatographic Analysis

This method of analysis involved acetylation of the P.E. and examination of the resulting product by high temperature gas

FIGURE 3.5 Theoretical Fluidisation and Transport Velocities for Spheres (Monodisperse)

$\rho_s = 1{\cdot}4$ (g/cm^3)
$\rho = 1{\cdot}0$ (g/cm^3)
$\mu = 0{\cdot}01$ (P)



chromatography. Acetylation of the P.E. is carried out by first refluxing the sample with anhydrous sodium acetate and acetic anhydride, washing with warm distilled water and then extracting the product with analar benzene. The benzene extracts are then dehydrated by shaking with anhydrous sodium sulphate, filtered and the benzene evaporated off. The resulting crystalline product is ground, dried in a desiccator and transferred to a gas-liquid chromatograph for examination of the isolated acetylated impurities. The chromatograph has a preheater temperature of 300°C and a column packed with silicone gum E 301/Embacel, at a temperature of 305°C. The analysis gives a direct measurement of the Di - P.E. content, but as the Formal has not been obtained in a pure state to calibrate the chromatograph, the response factor is taken to be the same as that for Di - P.E. and so the amount present stated in terms of the equivalent amount of Di - P.E. (Table1Appendix A).

3.4. 3. Formaldehyde Content

In the absence of a high temperature chromatograph a method was established in the previous work (1) for analysing the Formal content of P.E. in the absence of other formal impurities. This was done by finding the 'formaldehyde content' of P.E. by a colorimetric method based on the reaction of formaldehyde with chromotropic acid in concentrated sulphuric acid when, on heating, an intense violet colour is formed. As this reaction is the result of both free formaldehyde adsorbed on the solid, and also of combined formaldehyde in the Formal an allowance was made for the effect of the adsorbed free formaldehyde by using the Sodium Sulphite method for the formaldehyde determination. As this

method involves the use of a very dilute acid only in the final titration, and as no heat is applied it was considered that this would not be sufficient to break down the Formal present.

The total formaldehyde content was first obtained by accurately weighing 1.000 g of the P.E. sample and dissolving in 100 cm^3 of distilled water. 1 cm^3 of this solution was transferred with a pipette to a 50 cm^3 volumetric flask. 1 cm^3 of the chromotropic reagent was added and then 10 cm^3 of 95% sulphuric acid while cooling the flask in ice. The flask was heated in an oil bath at 90°C for 1 hour, cooled in cold water and made up to 50 cm^3 with distilled water, continually mixing and cooling during dilution. The effect of heat was found critical and the calibration was standardised for a heating time of 1 hour at 90°C.

The optical density was measured by comparison with a reagent blank treated in the same way, measured at a wavelength of 570mμ in ½ cm cells using a "SPEKKER" absorptiometer with Kodak No. 6 filters.

The absorptiometer was calibrated by treating 1 cm^3 of known concentration formaldehyde solutions in the same way as the 1 cm^3 of P.E. solution. The calibration is shown in figure 3.6.

The adsorbed free formaldehyde was then found using the Sodium Sulphite method. 50 cm^3 of the sodium sulphite solution were placed in a 500 cm^3 Erlenmeyer flask. A few drops of thymolphthalein indicator were added and the solution neutralised with $\frac{N}{100}$ hydrochloric acid until the blue colour had disappeared. The 99 cm^3 of the original 1 g P.E./100cm^3 H_2O solution remaining after the colorimetric test were transferred to the flask. The formaldehyde present reacts with the sodium sulphite to

FIGURE 3.6.

SPEKKER ABSORPTIOMETER FORMALDEHYDE CALIBRATION

λ = 570mμ $\frac{1}{2}$cm cell

form the formaldehyde - bisulphite addition product :

$$HCHO + Na_2SO_3 + H_2O \longrightarrow NaOH + CH_2(NaSO_3)OH$$

The resulting mixture was titrated slowly with the standard $\frac{N}{100}$ hydrochloric acid to complete discoloration. One cm^3 of normal acid is equivalent to 0.03003 g Formaldehyde. The per cent formaldehyde in the sample is hence given by the equation:

$$\% HCHO = \frac{\text{acid titer} \times \text{Normality of acid} \times 3.003}{\text{weight of sample}}$$

and HCHO mg/cm^3 solution = % HCHO X $\frac{1}{10}$

The formaldehyde equivalent of the optical density obtained for the total formaldehyde content in the colorimetric test is known from the formaldehyde calibration of the instrument, figure 3.6. The effect of the adsorbed formaldehyde, found from the sodium sulphite method, on the optical density is the product of theratio of the absorbed formaldehyde to equivalent formaldehyde and the optical density. If this is subtracted from the optical density the corrected optical density is assumed to be due to Formal composition only, in the absence of other formals. The calibration of the Formal in terms of the corrected optical density is shown in figure 3.7.

This method however relies on the manufacturer's chromatographic analysis for Formal content for calibration, which itself is not a true concentration, being relative to Di - P.E. Also any other formal impurities present in the P.E. would give a reaction which would be attributed to the presence of the usual Formal.

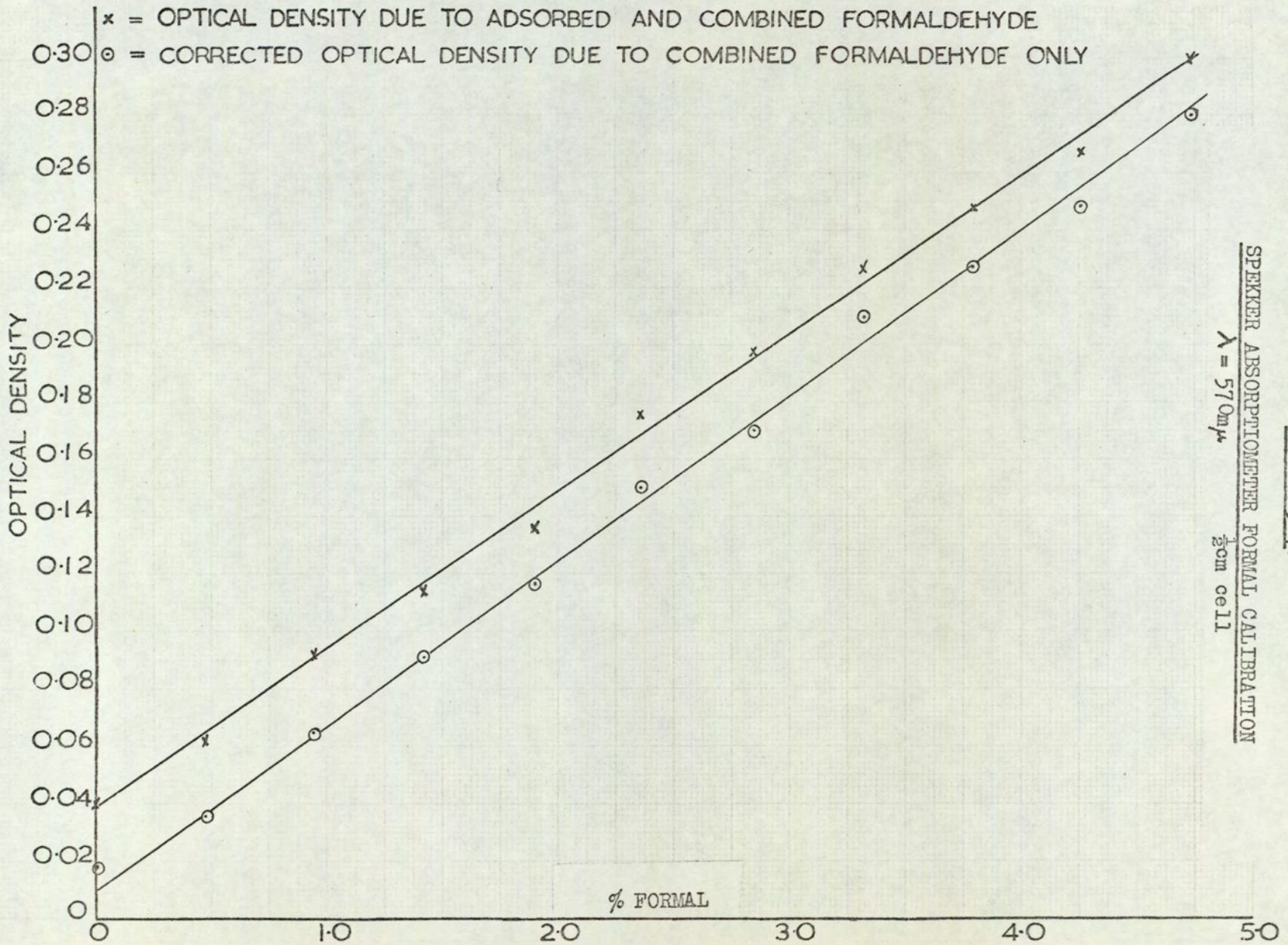


FIGURE 3.7.

SPEKKER ABSORPTIOMETER FORMAL CALIBRATION

λ = 570mμ $\frac{1}{2}$cm cell

3.4. 4. T. M. S. Chromatographic analysis

Suchanec (81) has proposed an improved chromatographic analysis for P.E. using the trimethylsilyl (T.M.S.) ether derivatives. The P.E. sample is mixed with a known weight of mannitol and placed in a flask together with pyridine and hexamethyldisilazane. The flask is placed on a hot plate and heated almost to boiling for 10 minutes. After cooling below 50°C trimethylchlorosilane is added, and mixed while warming before cooling to room temperature. The precipitate of ammonium chloride and pyridinium chloride is allowed to settle, and a sample is taken and introduced into the chromatographic column at 125°C. The column temperature is programmed at a rate of 13°C per minute to a maximum temperature of 326 °C, and held isothermally for 3 minutes at 326°C. Although the 'Formal' in the P.E. sample has not yet been isolated the response factor for the chromatograph can be found by using the known additions of mannitol as a reference, and thus finding the mass % composition of Formal impurity. A similar impurity was found by Suchanec which he labelled bis- P.E. monoformal (as did Salkind et al (71) and Wiersma et al (72).

3.5. Crystal Growth

During the final production stages in the manufacture of P.E. the process solution is cooled in a batch crystalliser. This results in a product consisting of agglomerates of a few large crystals and a large number of very small ones, making it 'dusty' and unpleasant to handle. In the previous work (1) it was found that the product could be greatly

improved even with batch crystallisations by controlling the growth temperature. This was developed to obtain good crystals with few agglomerates which were classified into sieve fractions for use as seed in the seeded stirred cell growth experiments. The method used was to cool a hot concentrated solution to its nucleation temperature, stir at this temperature for 2 hours and filter off the product crystals. These were washed with ice cold water and then with acetone before drying in an oven at 100°C.

Attempts to obtain growth rate data from small scale fluidised beds in the previous work (1) failed, mainly because of agglomeration occuring during the experiments. This was probably because too small a seed size was used, (64 - 75μ) for which it was difficult to obtain satisfactory fluidisation conditions in the $\frac{5}{8}$ in I.D. crystalliser tube. The method used, therefore, was to follow the decrease in solution concentration of stirred cells seeded with crystals of a known weight and size analysis. This was done using an immersion refractometer graduated in % sugar in intervals of 1% (which could be estimated to 0.1% with ease). The growth rate constants, K, for each interest were calculated assuming a first order growth with respect to supersaturation $(c - c_{\infty})$, where $\frac{dc}{dt} = -KA\,(c - c_{\infty})$. For this purpose the approximation $A_2 = A_1\left(\frac{M_2}{M_1}\right)^{\frac{2}{3}}$ was used which is only true for a monodisperse system, where the average crystal size equals the size of the crystal of average area, which also equals the size of the crystal of average weight. Although a narrow sieve fraction of crystals was used as seed, the initial size analysis used was that after the first five minutes attrition under the

conditions of the experiment in a saturated solution: It was found that for the seed crystals in a saturated solution all the attrition occured during this first five minutes. The size analysis carried out with the Coulter Counter then showed a fairly wide size distribution, so the area determinations for each interval can only be regarded as an approximation. The results for the Batch A and Purified Batch A solutions seeded with 2 g of seed stirred at 500 r.p.m. are shown in Table 49 , Appendix E .

The values of the growth rate constant K were averaged with the exception of:

a) Results including the first 25% increment on the initial mass, because these were considered due to a repairing process of damaged attrited crystals.

b) Results where the refractive index scale, $n < n_{\infty} + 0.5$ Indicated % Sugar, as the accuracy would be too dependent on the accuracy of the equilibrium value.

c) Results where the time, t, $>$ 1500 minutes as the crystals could not be assumed to undergo no further attrition after this time.

Average results, $\bar{K}$, are plotted as log $\bar{K}$ vs $\frac{1}{T}$ in figure 3.8. and are correlated by the following equations for $80^{\circ}C > T_o > 50^{\circ}C$:

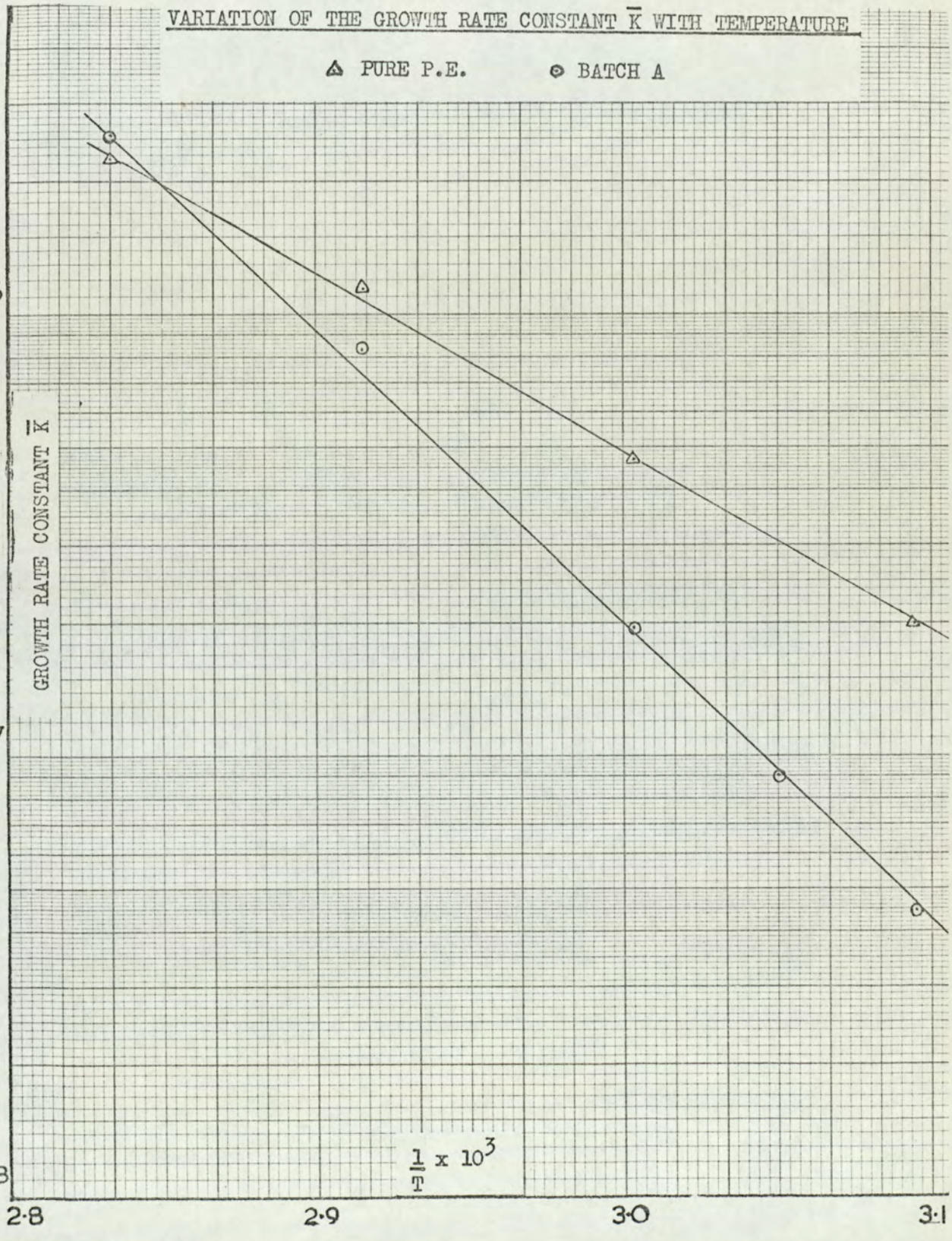
Batch A (4.7% Formal, < 0.1% Di - P.E.)

$$\log_{10} \bar{K} = 13.401 - \frac{6710}{T}$$

where T is in degrees Kelvin.

with an activation energy for growth of 30.65 K cal /g mol, and

FIGURE 3.8.


VARIATION OF THE GROWTH RATE CONSTANT $\bar{K}$ WITH TEMPERATURE
PURE P.E.
BATCH A
GROWTH RATE CONSTANT $\bar{K}$
10^{-6}
10^{-7}
10^{-8}
$\frac{1}{T} \times 10^3$
2·8
2·9
3·0
3·1

Purified Batch A ($< 0.1\%$ Formal, $< 0.1\%$ Di. - P.E.)

$$\log_{10} \bar{K} = 5.770 - \frac{4025}{T}$$

with an activation energy for growth of 18.4 K cal/ g mol.

These indicated surface integration control for both materials under these conditions.

The limitations of this work were realised however as being mainly due to the limitations of the refractometer, and a more accurate refractometer would be necessary to obtain more reliable results. It was also realised that many parameters needed to be studied, particularly low crystal/solution relative velocities, temperatures $< 50^{\circ}C$, effect of seed size distribution, and impurity content, and effect of Di - P.E. in the mother liquor.

SECTION FOUR.

BENCH SCALE CRYSTALLISATION APPARATUS.

4.1. Introduction.

In most crystallisation processes the crystal growth proceeds by the two consecutive steps: diffusion of molecules to the crystal surface, and integration of the molecules into the crystal lattice. In order to study these steps individually it is necessary to try to eliminate one of them. If the solution is stirred sufficiently vigorously the repeated acceleration and deceleration in the turbulent eddies can give a laminar film around the crystal considerably thinner than that due to terminal velocity under gravity, so that in the limit the growth process should be controlled only by the surface integration step. Stirred cells (section 4.4) were used to examine this process.

The diffusional resistance when significant can be studied by varying the crystal/solution relative velocity in a controlled manner. One method of doing this used by Mullin and Garside (50), is to hold a single crystal stationary in a tube and to meter the solution rate through it. However it was not found possible to grow single crystals of reasonable size from the impure mother solutions of interest. This problem with P.E. crystals was also encountered by Whetstone (82). The method used, therefore, to study the effect of the relative crystal/solution velocity on the growth rate was to meter the solution flow through a fluidised bed of crystals (Section 4.2.)

As indicated above crystals in a vigorously stirred suspension are not at a relative velocity equal to their terminal falling velocity, and

Bransom et al (47) found that this increase in growth rate with stirrer speed in the diffusion controlled regime was independent of seed size. This was attributed to "homogeneous isotropic turbulent eddies". In a transition from diffusion to surface integration growth rate control in a stirred cell, a critical stirrer speed might be obtained for this transition point for which the particular crystal/solution relative velocity would be unknown. Although this transition point might be observed also in the fluidised bed of crystals, the relative velocities studied will, in general, be considerably lower than in a stirred vessel and if the transition occurs at a high relative velocity a large seed size would be necessary with a fluidised bed. A possible method of obtaining higher known relative velocities is to utilise the terminal velocity of crystals under "free fall" conditions and to study the growth rate with respect to crystal size which is a known function of velocity (figure 3.5.). A critical crystal size equivalent to the terminal falling velocity at the transition relative velocity should then be obtained. Ideally this would require an extremely long sedimentation tube, but in practice an attempt was made to overcome this problem by constructing a "Repetitive Inversion Sedimentometer" (section 4.3.).

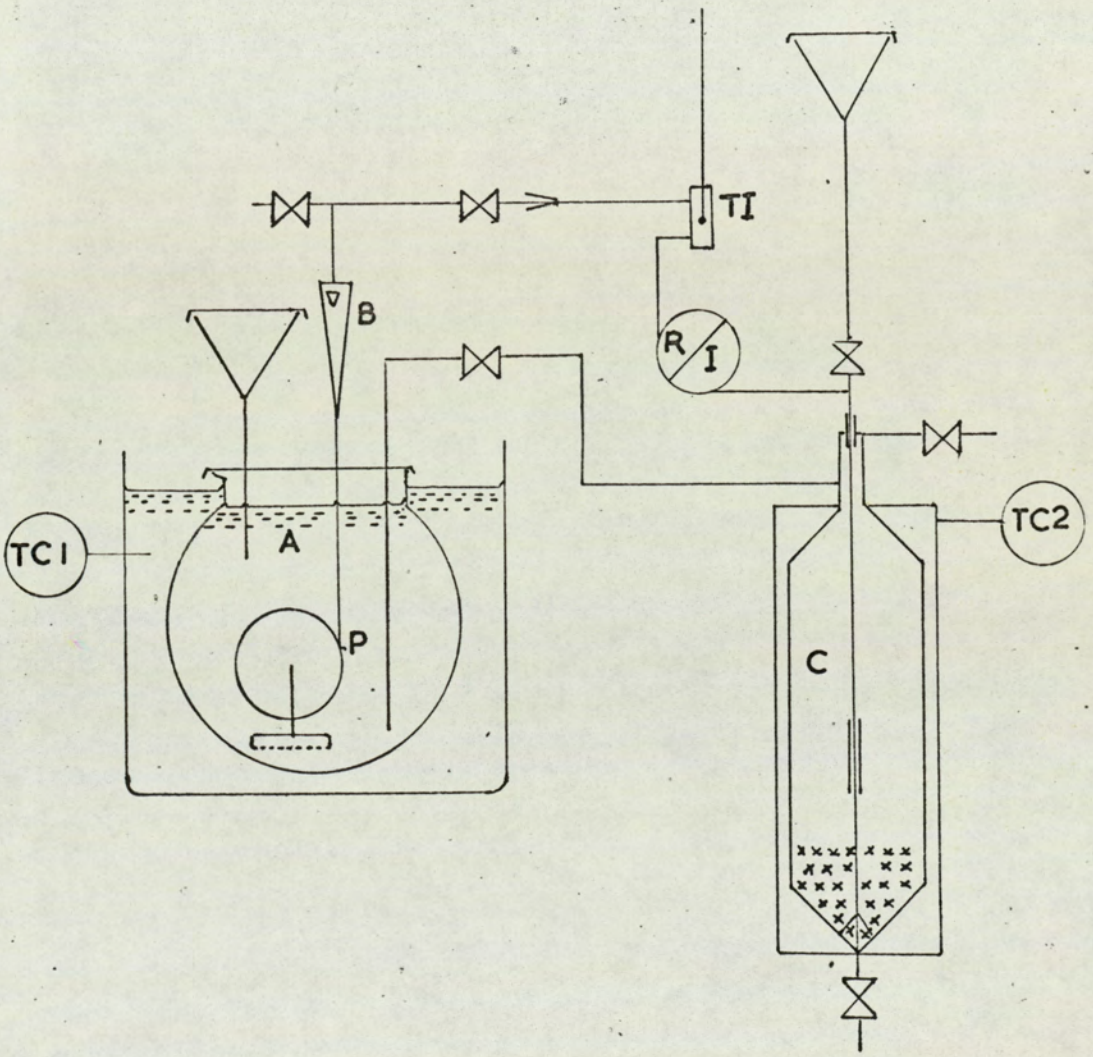
4. 2. Fluidised Bed Apparatus.

4. 2. 1. Preliminary Design

The apparatus is shown in figure 4.1. and as far as possible was built from "Quick-fit" ground glass parts. The internal diameter of the

FIGURE 4.1.

Preliminary Fluidised Bed Apparatus



crystalliser (C) was 1.80 in and the included angle of the cone base was ca. 90°. The downcomer in the crystalliser was fitted with a flexible seal and knee joint at Run No. F.C.19. which enabled agglomerations of crystals to be broken up. The capacity of the feed tank (A) was 1 dm^3 and contained a submerged glandless pump (P) fitted with a nylon filter over its intake. The solution flow rate was indicated by a Metric Series Rotameter (7K) and a Bellingham and Stanley Immersion Refractometer (R./I.) indicated the solution concentration at the temperature measured by a thermometer (T.I) on the inlet line to the crystalliser. Temperature controlled water was circulated around the feed tank with the Townson and Mercer T.E.3 thermostat circulator (T.C.1) which controlled to ± 0.01°C and around the crystalliser body (C) with a second T.U.3 circulator (T.C.2).

A Hoffman clip on a rubber tube fitted to the crystalliser base allowed the crystal product to be discharged for examination, in order to measure the growth obtained.

4. 2. 2. Solution concentration decay.

Because of the success of the refractive index change method for the growth rate study used in the agitated cells, and the previous difficulty of establishing a method for the increase in crystal size in a fluidised bed (1), it was thought that a method of concentration change using the property of refractive index should be possible using a fluidised bed with a closed system of circulating mother liquor. For this a crystal/mother liquor ratio of about 10g/500 cm^3 would be necessary to obtain a reasonable rate of concentration decrease, with

the slow growth rate of P.E. The system shown in figure 4.1 was therefore adapted to reduce the mother liquor hold up. The feed vessel (A) was changed to a jacketed vessel of 500 cm^3 capacity and the crystalliser (C) to a jacketed glass tube of $1\frac{1}{4}$ in I.D. with a sintered glass disc as a support and distributor. The immersion (glandless) pump was used as before so as to prevent any contamination with grease from pump glands. The system was unsuccessful however due to the difficulty of having an air tight system with an immersion pump. It was found that with such a small hold up and slow growth rate, the evaporation loss was such that it more than compensated for the change in concentration due to crystal growth.

4. 2. 3. Modified Design.

The fluidised bed apparatus after final modification is shown in figure 4. 2. and 4. 3. Although measurement of growth rates from the solution concentration change proved unsuccessful it showed the advantage of using a glass sintered disc as a fluidised bed support and fluid distributor as opposed to the downcomer method used in the preliminary experiments. A uniform flow was obtained through the crystal bed with the sintered glass disc, whereas the downcomer had produced uneven flow with a moving bed portion at the walls making the system more susceptible to agglomeration. The preliminary experiments showed the method of measuring the crystal mass increase preferable to measuring the increase of crystal size, but the discharge of product crystals after an experiment had proved inefficient. Cell (C), figure 4. 2. was therefore designed so that all limbs could be easily detached and the product crystals could be

FIGURE 4.2.

Modified Fluidised Bed Apparatus

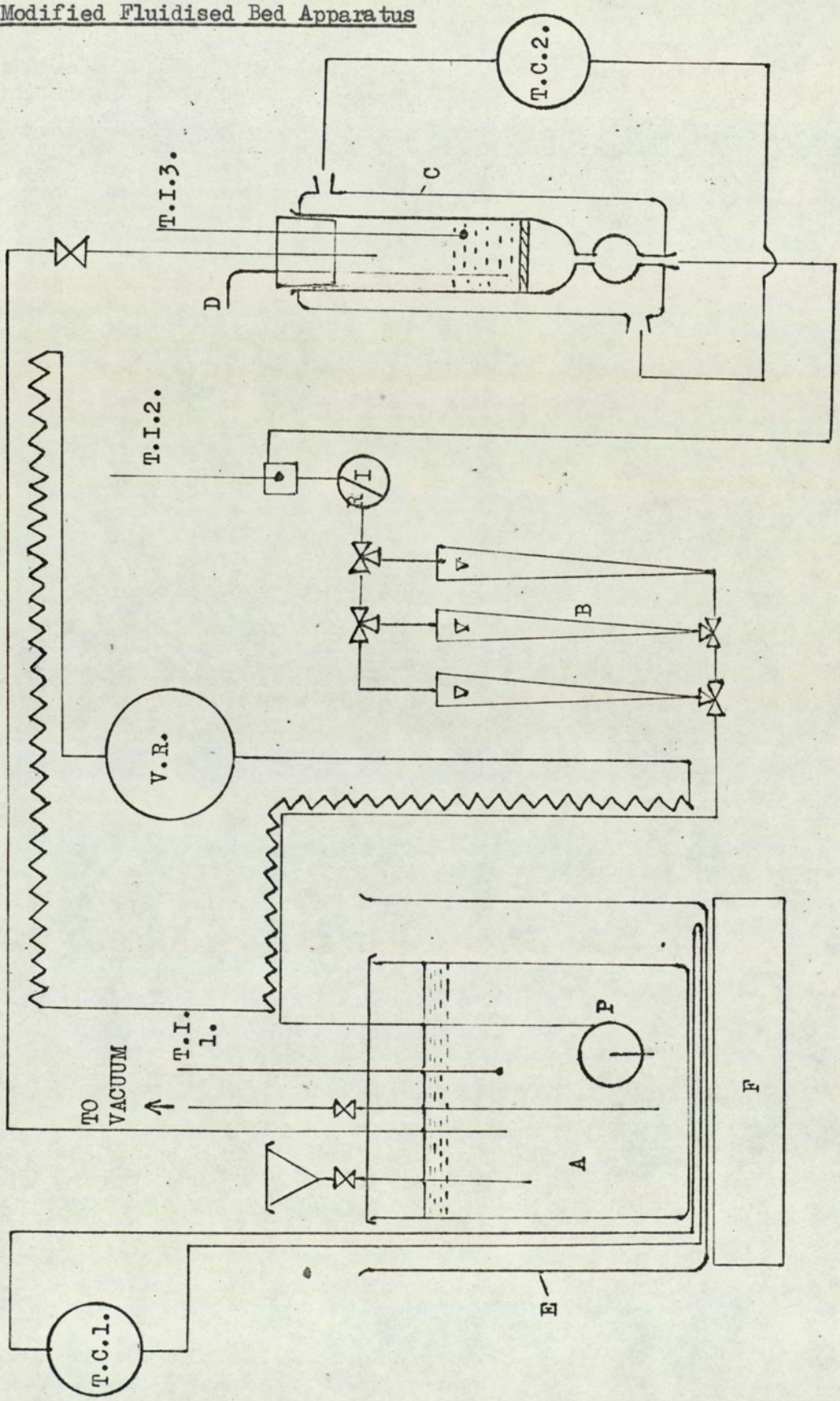
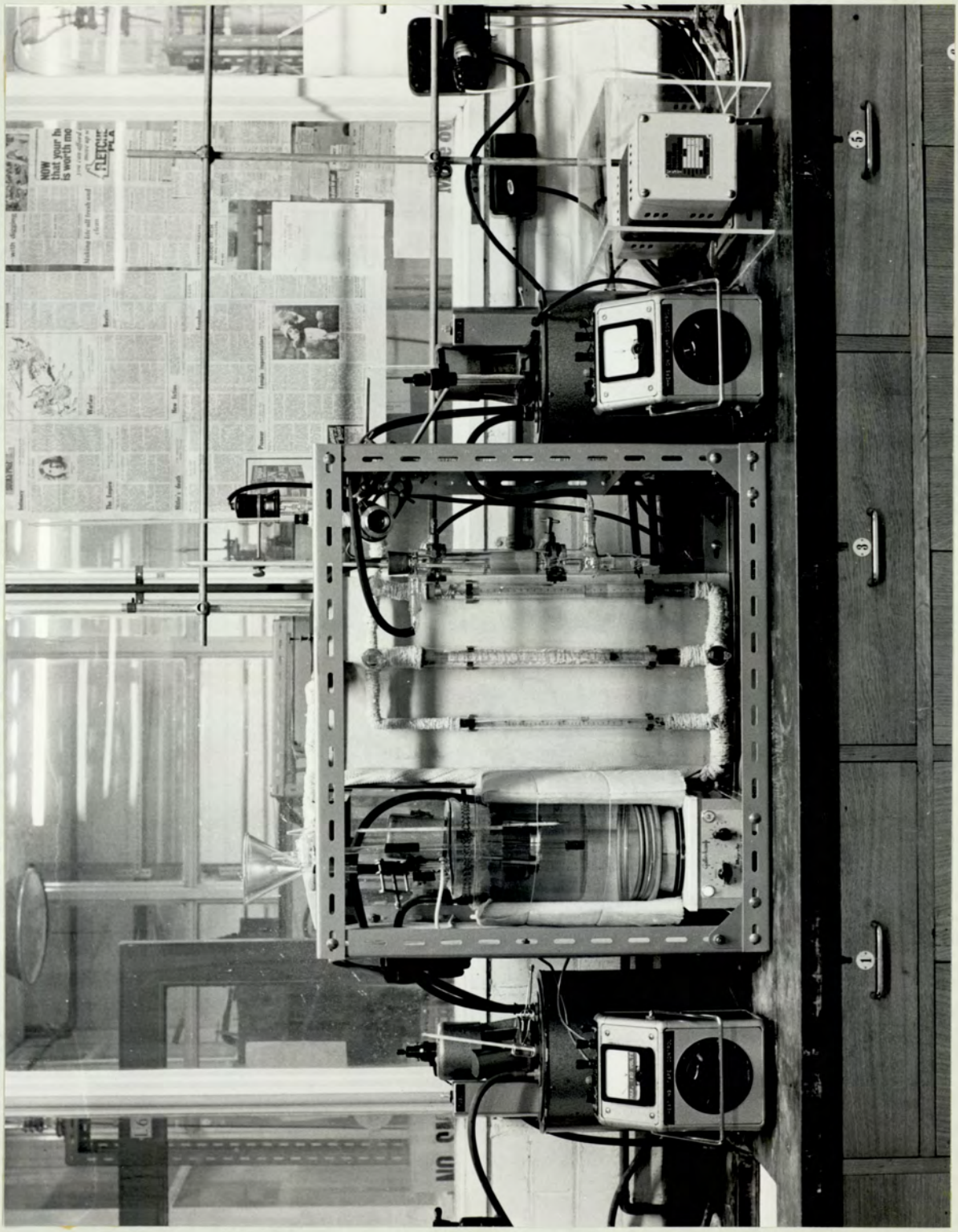


FIGURE 4.3.

MODIFIED FLUIDISED BED APPARATUS



weighed in-situ. The cell was jacketed and fitted with three B.14 ground glass sockets; two in the cell jacket and one for the solution inlet to the tube. A rubber bung was fitted into the top of the tube, containing the outlet line, a stainless steel probe (D) to prevent agglomeration and a thermometer (T.I.3). A small reservoir was made within the cell jacket for the feed solution to attain the required cell temperature, which was controlled by a Townson and Mercer T.U.3 circulating thermostat unit (T.C.2).

The feed vessel (A) of 2dm^3 capacity was contained in a water bath (E) controlled with an immersion coil attached to a Townson and Mercer T.U.3 circulating thermostat unit (T.C.1). The water bath (E) was placed on a magnetic stirrer and hot plate unit (F) which maintained circulation in the bath and provided supplementary heat. The feed vessel (A) was fitted with a feed funnel, a vacuum line, an inlet line, a thermometer (T.I.1) and an immersion pump (P). Three metric rotameters (B)584345/E, 7K and 14K were fitted with glass air jackets and three way glass valves served to direct the solution flow. A Bellingham and Stanley immersion refractometer (R/I) with prism 1B, indicated the solution concentration at the temperature measured by the thermometer (T.I.2). Heating tape controlled with a voltage regulator was used on lines between A and B, and B and C and all lines were well lagged. The immersion pump (P) was connected via a voltage regulator to a voltage stabiliser improving the flow control.

4. 3. Repetitive Inversion Sedimentometer.

A jacketed brass cell was built, 2 in. internal diameter and

12 in. long with a refractometer fitted centrally through the side of the cell, to measure the change in concentration of a supersaturated solution containing growing crystals maintained essentially at their terminal falling velocity. As the water jacket was connected to a thermostatically controlled circulator it was necessary to invert the cell reversibly, and for this the pneumatic control, figure 4.4. was thought most suitable.

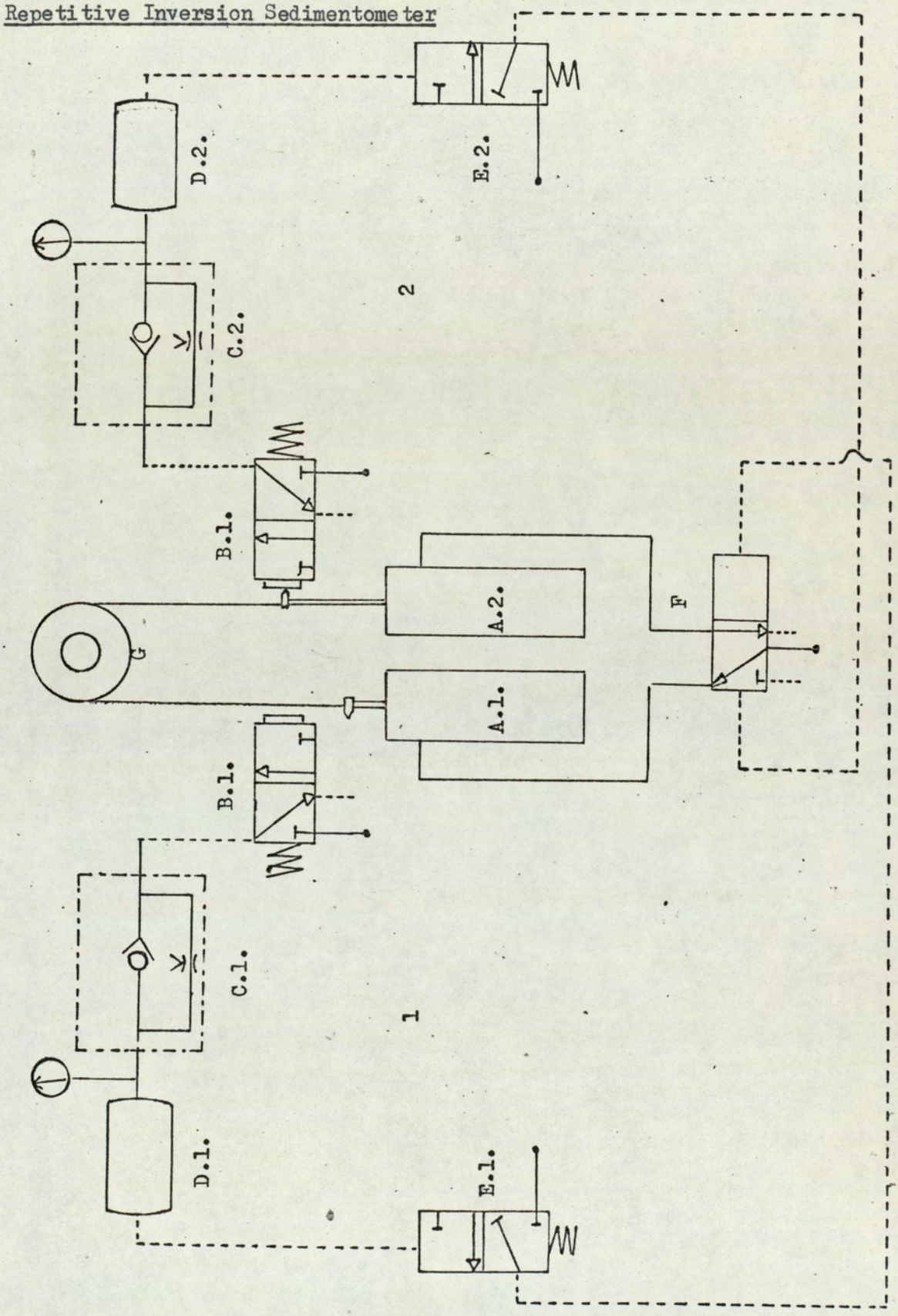
The operation of Cycle 1 is initiated when the tappet of the three way poppet valve B.1. is struck by the piston rod A.1. The main line air previously exhausting through B.1. is directed via the air flow regulator C.1. to dwell unit D.1. When the pressure in D.1. builds up to about 50 p.s.i.g. it is sufficient to operate the three way poppet valve E.1. This in turn directs the main line air previously exhausting through E.1. to operate the pilot piston of the Four Way Piston Valve, F.

Air enters the pilot chamber of F moving the piston over and thereby reversing the main valve. Main line air is then directed to the rear-cushioned piston cylinder A.1. Operation of A.1. closes Cycle 1 and the main air lines in this cycle exhaust to atmosphere.

As the chain attached to the piston rod A.1. passed via a 4 in. pulley to rod A.2, the 6 in. stroke of the piston is sufficient to reverse the pulley one half a revolution and to operate the tappet valve B.2. and subsequently Cycle 2. The pulley was attached to a 12 in. cylindrical jacketed vessel of 2 in. bore with a refractometer fitted centrally. This enabled repetitive inversion of the cell at time intervals controlled by the air flow regulators C.

FIGURE 4.4.

Repetitive Inversion Sedimentometer



4. 4. Stirred Cells.

4. 4. 1. Cell A.

This consisted of the standard Bellingham and Stanley in-line refractometer housing of Cell A of the previous work (1), which was a stainless steel cone frustum internal design mounted with the axis horizontal and having a water jacket fitted round this housing supplied with water from a Townson and Mercer T.U.3 thermostat circulating unit. The refractometer used in the previous work (1) which indicated 0-40% sugar and estimated to 0.1% sugar, was replaced with the more accurate Bellingham and Stanley immersion refractometer fitted with a 1A prism with an arbitrary scale 0-105 in intervals of 0.1 divisions (range n_D = 1.3254 to 1.3664 which made it possible to read solution concentrations to ca. 0.025% P.E. without the need for estimation. The refractometer was screwed into a circular brass plate, designed so that the prism was off-centre in the cell and the light source from the illuminating window at the back of the cell was incident to the plane of the prism face and clamped to the front of the cell. It was found that water could leak past the prism/stainless sleeve joint and condense on the scale making it impossible to read. The joint was therefore sealed with a layer of epoxy resin glue. The cell was mounted in a frame with a sodium lamp behind. The lower port of the cell was sealed with a rubber bung having a glass tube and plug for draining purposes, fitted flush with the inside of the cell casting. A rubber bung fitted with a thermometer and with a hole large enough to accommodate a small stirrer with a three blade marine impeller 1 in. diameter was inserted in the top

port of the cell. The capacity of the cell was about 300 cm^3.

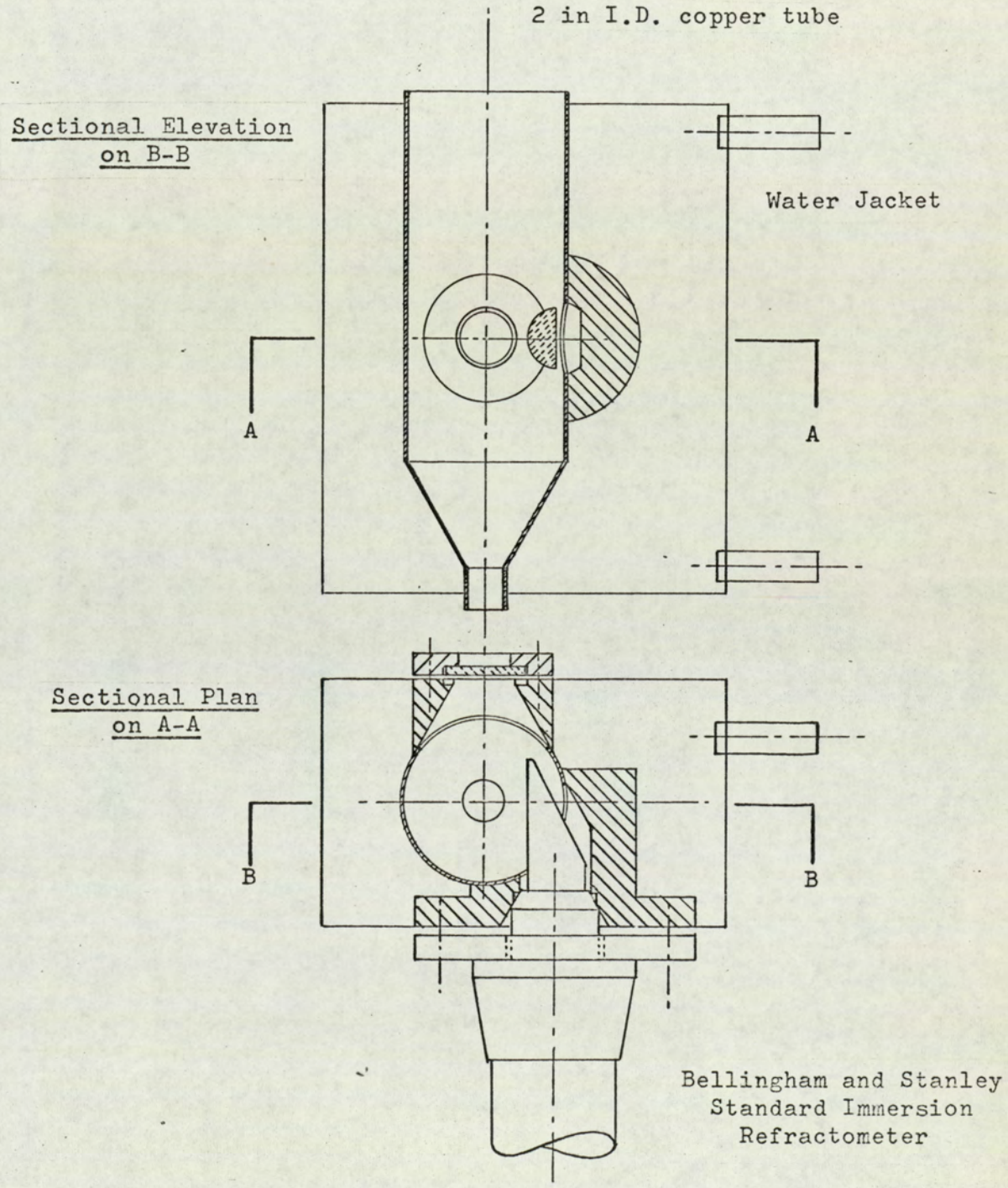
4. 4. 2. Cell C

Although Cell A reproduced the hydrodynamic conditions of the previous work (1) and care was taken to obtain the optimum angle of incidence of the light source, it suffered from the defect of having a long light path and consequently the image became obscured even with relatively low suspension densities. Also crystals could settle on the horizontal ledge by the illuminating window thus reducingthe effective surface area available for growth. Cell C was therefore constructed (figure 4.5) with the smallest practical light path through the suspension to avoid obscurity of light being caused by crystals, and the only horizontal surface being a small part of the annular gap where the glass prism joins the stainless steel sleeve. The cell was constructed of a 2 in. internal diameter copper tube with a conical base surrounded with a water jacket through which water was pumped from a Townson and Mercer T.U.3. thermostat circulating unit. A Bellingham and Stanley immersion refractometer was used with a 1B prism sealed to the stainless sleeve with an impact adhesive. The windows and refractometer were sealed to the tube by O-rings of neoprene. A plug was used to permit drainage of the cell, and a rubber bung fitted with a thermometer with a teflon bush for a stirrer fitted with a three blade marine impeller 1 in. diameter, was placed in the top.

4. 4. 3. Cell S

Repeated heating and cooling of Cell C frequently weakened the seams of the water jacket causing leaking. CellS was therefore constructed,

FIGURE 4.5.



MODIFIED GROWTH RATE CELL C

(Half Full Size)

FIGURE 4.6

CELL S

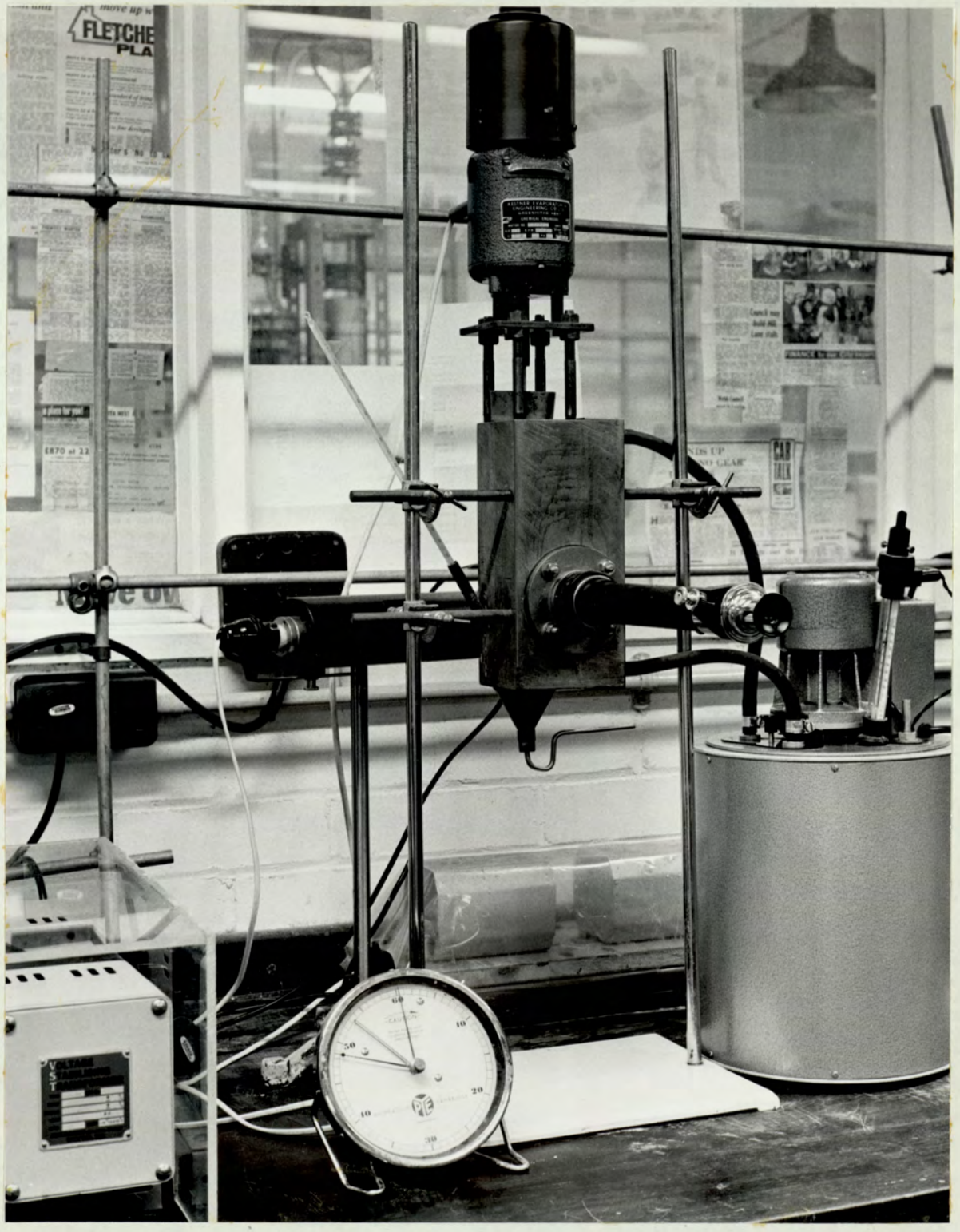
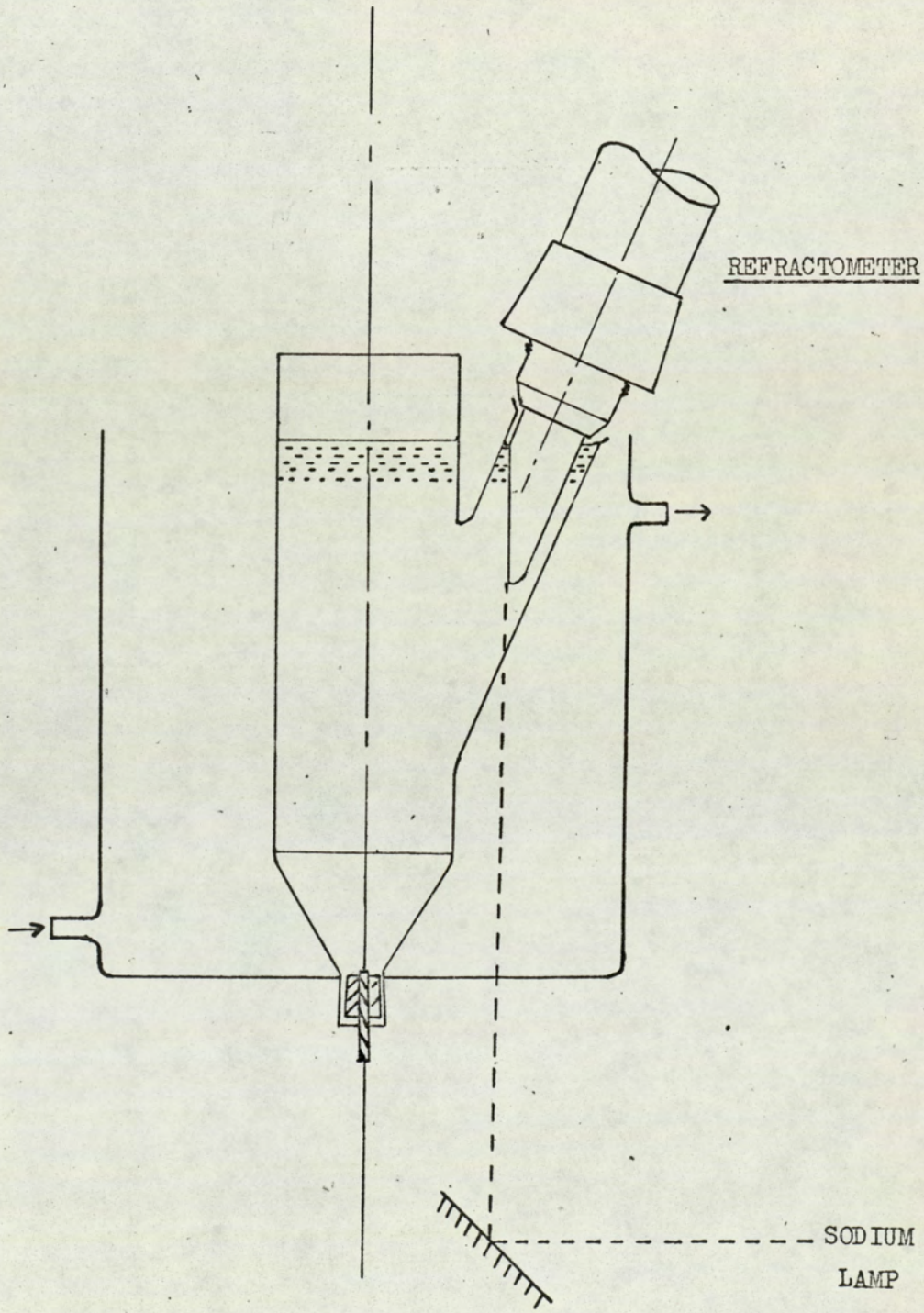


figure 4. 6., from solid brass of the same design as Cell C but of an overall length of 12 in. to accommodate larger volumes of solution if required. The water jacket consisted of a slot about 2in x 1 in bored down one side of the cell through which water was circulated from a Townson and Mercer T.U.3 thermostat circulating unit. A thermometer pocket was sealed into the side of the cell opposite the water jacket, and this, as with the refractometer and illuminating window, was placed about 3 in. from the base so as to be covered when 250 cm3 volumes of solution were used. A conical copper base with a draining plug was soldered to the cell base. A rubber bung was placed in the top with a teflon bush to accommodate a stirrer with a three blade marine type impeller of about 1 in. diameter. The stirrer motor was fixed rigidly to the cell brass housing. A Bellingham and Stanley immersion refractometer was used with a 1B prism sealed to the stainless sleeve with an epoxy resin. The cell was well lagged.

4. 4. 4. Cell G.

Cell G, figure 4.7, was made with similar dimensions to Cell C to obtain the same hydrodynamic conditions to observe suspension characteristics. However, because of the difficulty of clamping the refractometer horizontally to the glass cell a Y piece was made, as shown, to accommodate the prism such that the prism face was vertical and had the minimum of suspension to reduce the light transmission to the prism face. The height of the Y piece was such that the 250 cm3 of solution covered the prism but did not reach the top, so that the prism/cell junction had only to minimise evaporation and need not be

FIGURE 4.7.



GLASS CELL G

HALF FULL SIZE

water tight. Allowance therefore had to be made for the solution expansion with temperature and also the hydrodynamic fluctuations with stirrer speeds up to 2000 r.p.m. The optimum position was found by trial and error.

Because of the construction difficulties involved with an enclosed glass water jacket, an open water jacket was used with a Churchill constant volume thermostat circulator. A Bellingham and Stanley immersion refractometer was used with a 1A prism sealed to the stainless sleeve with an epoxy resin. A rubber bung was placed in the top of the cell with a thermometer fitted and a teflon bush to accommodate a stirrer with a 1 in. diameter 3 blade marine type impeller. Illumination was supplied by a sodium lamp and reflected at the optimum angle through the base of the glass water jacket by means of a mirror.

4. 5. Discussion.

Although it was originally thought necessary to construct the inverting sedimentometer to investigate the effect of the crystal terminal falling velocity on the crystal growth rate, it was later found however that results with the fluidised bed apparatus were in reasonable agreement with those found in the stirred cells. This was so even at the higher temperature up to 70°C where the integration rate/diffusion rate ratio would be greatest. As diffusion control would be immediately apparent with a slower growth rate for the lower relative velocities in the fluidised bed, this indicated surface integration rate control for all conditions studied. So although the sedimentometer was constructed it was not used in this project as no extra information was thought to be obtainable by this method for P.E.

SECTION FIVE

PENTAERYTHRITOL EQUILIBRIUM IN AQUEOUS SOLUTION

5. 1. Previous Work.

As the "Formal" impurity could be synthesised with formaldehyde and P.E. in aqueous solution, and in view of the sequence of reversible reactions proposed by Salkind et al (71) for the formation of bis - P.E. monoformal, the stability of the Formal in aqueous solution was studied (1). Any decomposition might be expected to affect the equilibrium (solubility) results. This was done by purging a concentrated P.E. (Batch A containing 4.73% Formal) aqueous solution, held at 90°C, with a steady stream of nitrogen at 3 ft^3/h to drive off any formaldehyde formed. The solution concentration was kept constant by periodically adding water. Samples of the solution were taken after 40, 60 and 200 ft^3 of nitrogen had been used respectively (i.e. after about 70 hours total) and analysed by the "Formaldehyde content" method described in section 3.4.3. The results all agreed to within the limits of analytical accuracy (i.e. approx. $\pm$ 0.2% Formal), whence it was concluded that the Formal does not readily decompose in aqueous solution up to at least 90°C.

Equilibrium solubility of solute in solvent can either be achieved from undersaturation or from supersaturation. Attempts at achieving equilibrium quickly using ultrasonic irradiation in the previous work (1) failed because of the heat evolved from the ultrasonic probe raising the solution temperature. The ultrasonic

probe could not be used, therefore, to attain equilibrium from undersaturation, but it was found to be a very effective nucleator for supersaturated solutions. Supersaturated solutions were therefore nucleated by ultrasonic irradiation and then stirred. Three methods of measurement of solution concentration were used: Specific gravity; refractive index; and weighing before and after evaporation of solvent.

Specific gravity measurements were found to be the most sensitive although they were limited to measurements essentially at room temperature. This technique was used to study the effect of the impurities on the rate of approach to equilibrium of a solution in a jacketed stirred vessel held at 25.0°C. Aqueous solutions of ca. 17 mass % were made up and cooled to 25°C. The supersaturated solutions were nucleated, stirred and periodic measurements of the solution specific gravity showed that equilibrium for both Pure P.E., and Pure P.E. + 2.0% Di-P.E. solutions was achieved in less than 2 hours. It also indicated an enhanced solubility effect of Di-P.E. at this temperature of about 0.5 mass % increase for 2.0 mass % Di-P.E. Batch A (4.73% Formal, $<$0.1% Di-P.E.) solutions, however, nucleated and stirred in the same way took at least 8 hours to attain equilibrium and the value obtained differed by about 0.1 mass % concentration from the value obtained from undersaturation. It was difficult to decide which was the actual equilibrium concentration as the values obtained overlapped according to whether they were approached from undersaturation or supersaturation. But it was noted that the presence of this 4.73% Formal enhanced the solubility at this temperature by about 2% compared with Pure P.E.

This "overlap" phenomenon was further studied using refractive index measurements of nucleated, stirred solutions using an immersion refractometer capable of measuring concentrations of 0.1% P.E. with ease and estimating to ± 0.05% P.E. The overlap of about 0.1% solution concentration was again observed with Batch A solutions approached from undersaturation and supersaturation, at 70°C and 80°C. However attempts to correlate the results using the 4 component system P.E. / Di-P.E. / Formal / Water were unsuccessful and the results appeared to depend on the nucleation temperature, the degree of supersaturation, and whether the equilibrium was approached from supersaturation or undersaturation.

The other method used for the determination of the equilibrium solution concentration was the more conventional method of weighing the amount of solvent evaporated from a known weight of solution. Supersaturated solutions were nucleated and stirred in a thermostatically controlled oil bath for about 12 hours. Approximately 10 cm^3 of solution was withdrawn through an immersion filter, weighed, evaporated in an oven, and reweighed. The method was unsatisfactory at high temperatures due to crystallisation occuring on transference to the crucible. Also readings were inaccurate due to the inherent difficulties involved in weighing hot liquid samples.

The equilibrium results for the three methods of solution concentration measurement were collected and plotted as log x vs $\frac{1}{T}$ where x = mass % and T = degrees Kelvin. A good agreement was found between the results for Pure P.E. with those of Cooke (75) who used P.E. of "better than 99.6% purity". A favourable agreement was also

found with the results of the impure P.E. $> 50^{\circ}C$. These results could be correlated by the equation (figure 3.2.)

$$\log_{10} x = 5.072 - \frac{1266}{T}$$

However below about $50^{\circ}C$ the results for impure P.E. showed a marked deviation from the above correlation indicating an enhanced solubility compared with Pure P.E. although the scatter was too great to obtain any correlation for the impurity effect.

5. 2. Refractometer Calibrations.

5.2.1. Reading Accuracy.

In the previous work (1) the refractometers were capable of measuring solution concentrations to ca. 0.1% P.E. and estimating to ca $\pm$ 0.05% P.E. The refractometers used in this work however were the more accurate Bellingham and Stanley immersion refractometers with an arbitrary scale 0 - 105 in intervals of 0.1 divisions (range n_D = 1.3254 to 1.3664). This enabled concentrations of ca. 0.025% P.E. to be measured ($\equiv$0.1 divisions) and of ca. 0.012% P.E. to be estimated. In order to take full advantage of these new refractometers a new calibration was carried out.

The thermometers used were graduated in 0.1 deg. C and could be estimated to about 0.05 deg. C which was sufficiently accurate compared with the refractometer accuracy and the slope of the calibration curve (figure 5.2)

The steam point and the transition point of hydrated sodium sulphate were checked for one test thermometer and this found to be

within the reading accuracy of the expected values for total immersion of the thermometer. All thermometers used in the experimental work were checked against this test thermometer. The correction for partial immersion of the thermometers, as used with the refractometers, is shown in figure 5.1., the results having been found experimentally in a stirred cylinder of water by comparison of a partially immersed and a totally immersed thermometer. Also shown (figure 5.1) is the calculated correction obtained from the equation

$$y = e' \left(T_o - T_s\right) L'$$

assuming T_s varies between 22°C and 30°C. Where T_s is the mean temperature of the emergent stem; T_o = observed temperature °C; e' = 0.000156 (apparent expansion of Hg in glass); L' = length of emergent mercury column expressed in degrees; and y is the correction to be added to T_o.

As T_s had to be estimated, corrections, y, for converting T_o to actual temperature are obtained by interpolating from the curve drawn through the experimental points.

5. 2. 2. Calibration.

Batch D material containing 1.0% Di-P.E. and 5.5% Formal was used for the calibration of "impure P.E.". Known concentrations (% m/v) were made up by accurately weighing out $\left(\angle \pm 0.0005g\right)$ the required amount of P.E. and washing this into a 250 cm^3 pyrex volumetric flask. The volume was made up to 250 cm^3 with distilled water taking care to eliminate air bubbles. The flask was warmed

until the P.E. had dissolved and cell C was brought up to above 80°C. The hot solution was transferred to the cell and stirred at above 80°C to ensure dissolution of any nuclei formed during the addition of the solution. The solution was cooled while stirring at a rate of about 1 deg. C/min and readings of the refractometer scale (S) and temperature (T_o) were taken at intervals of about 0.5 scale divisions.

Solution concentrations (c, % m/v) were used in multiples of 2.5% for Batch D material and the experimental points plotted as observed, temperature T_o, vs Scale S are shown in figure 5.2. Interpolated data at 5 deg. C intervals of T_o from 20 to 75°C were computed to obtain isothermal correlations by the least mean squares method, the correlations found together with the standard deviations of the points are shown in table 2, appendix A. It can be seen that the second order polynomial, $c = F + B.S. + G.S^2$ fits the data well for these temperatures with an average standard deviation of ca. ± 0.04% m/v. Linear equations could be used with little loss of accuracy at the lower temperatures but the standard deviation at the higher temperatures increased to ca. 0.09%. The second order polynomial was therefore considered necessary and sufficiently accurate for the present work. These calibrations were used with all the impure commercial batches of P.E. used. The chemical analyses of these materials are shown in table 1, appendix A and are considered sufficiently comparable not to require individual calibrations.

A general overall equation for use at intermediate temperatures which fitted the data well in the metastable zone was obtained, but being inverted it is necessary to solve a quadratic in

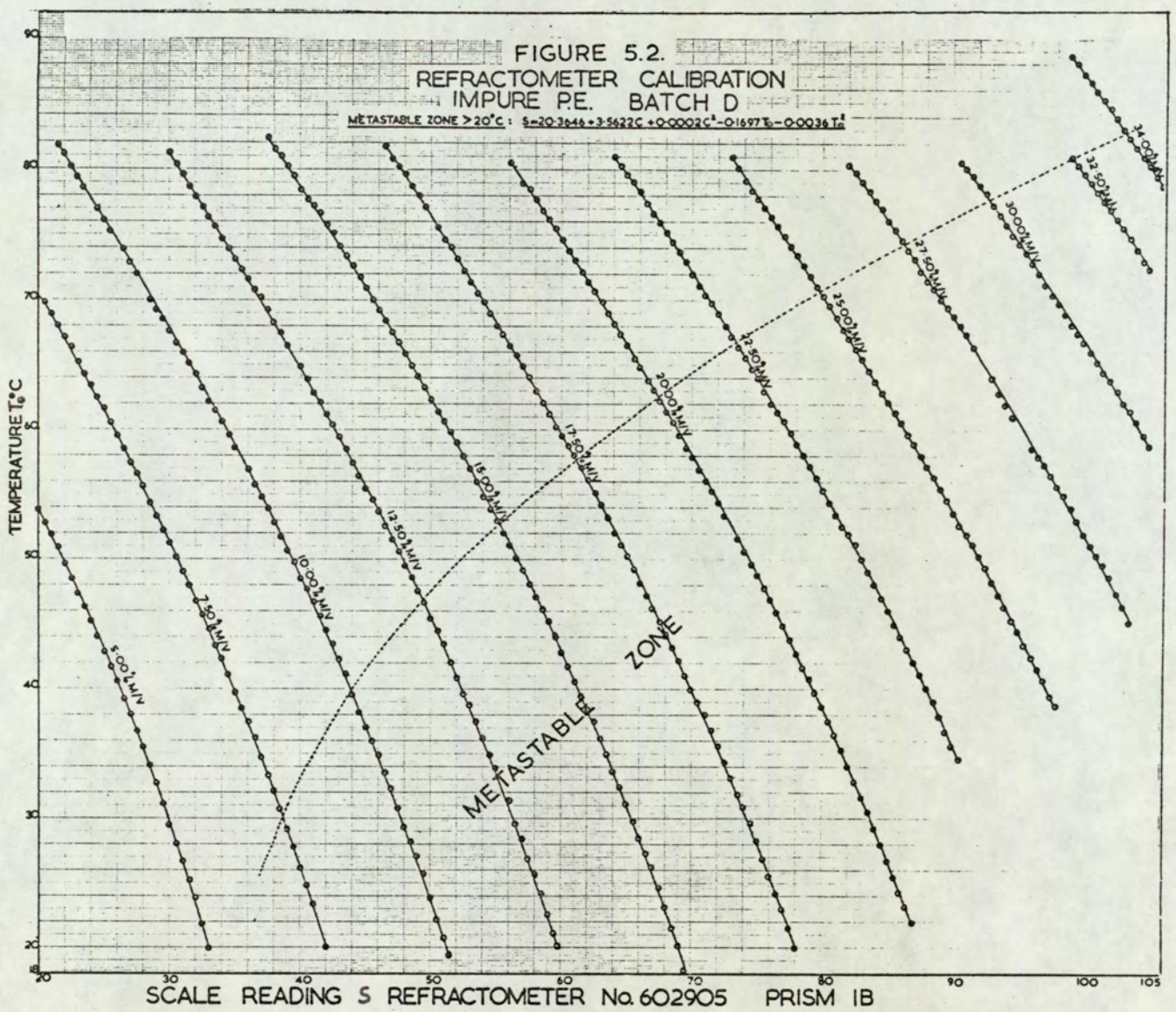


FIGURE 5.2.
REFRACTOMETER CALIBRATION
IMPURE P.E. BATCH D
METASTABLE ZONE > 20°C : S=20·3646+3·5622C+0·0002C²-0·1697T₀-0·0036T₀²
TEMPERATURE T°C
SCALE READING S REFRACTOMETER No. 602905 PRISM IB
METASTABLE ZONE
5·00% M/V
7·50% M/V
10·00% M/V
12·50% M/V
15·00% M/V
17·50% M/V
20·00% M/V
22·50% M/V
25·00% M/V
27·50% M/V
30·00% M/V
32·50% M/V
34·00% M/V

order to extract c:

$$S = 20.3646 + 3.5622c + 0.0002313\,c^2 - 0.1697\,T_o - 0.0035888\,T_o^2$$

where T_o is the observed temperature (^oC), S is the refractometer scale of cell C, and c is the solution concentration (% m/v).

The standard deviation of this expression from the data points in the metastable zone is $\pm$ 0.171 scale divisions, or ca. $\pm$ 0.04 % m/v.

The calibration was repeated with purified P.E. ($<$ 0.1% Di-P.E., $<$ 0.1% Formal) using solution concentrations in 5.0% m/v intervals. The number of points $<$ 40°C were limited due to the higher nucleation temperature of the pure solutions, so the impure P.E. calibration was considered more accurate below this temperature. The results were plotted as T_o (^oC) vs refractometer scale (S) (figure 5.3) and the isothermal data at 5 deg. C intervals of T_o interpolated. These interpolated values were used to compute the first and second order polynomial equations, by the method of least mean squares. The results are shown in table 3, appendix A.

It should be noted that all calibrations are in terms of the observed temperature T_o °C (i.e. partially immersed thermometer) and scale S (i.e. the refractometer scale of cell C). This was found to be the most convenient for use with the growth experiments, and corrections for partial immersion of the thermometer were carried out where necessary.

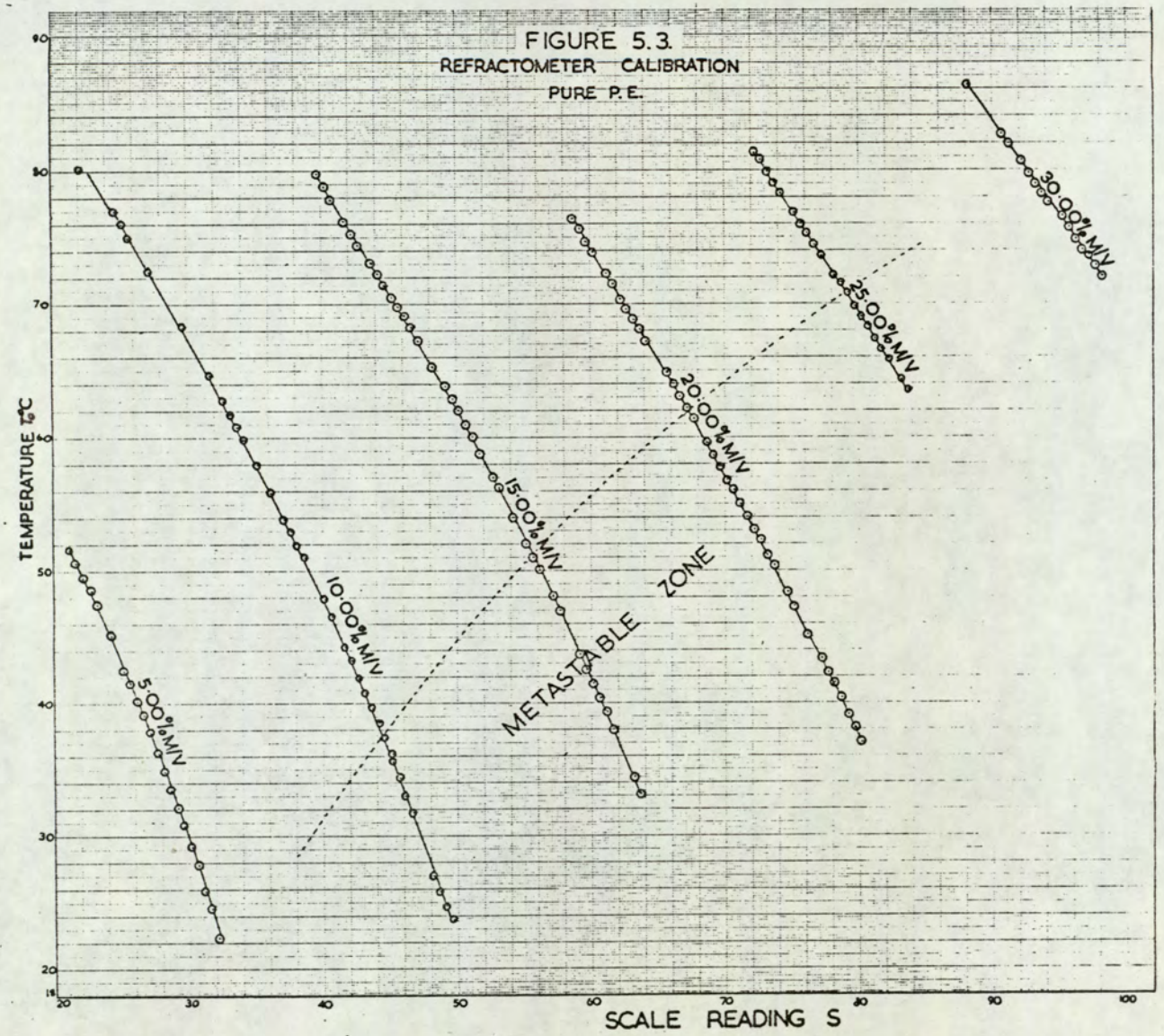


FIGURE 5.3.
REFRACTOMETER CALIBRATION
PURE P.E.
TEMPERATURE T°C
SCALE READING S
5.00% M/V
10.00% M/V
15.00% M/V
20.00% M/V
25.00% M/V
30.00% M/V
METASTABLE ZONE

5. 2. 3. Instrument Calibrations.

The refractometer calibration in terms of % m/v P.E. was carried out using cell C with a 1 B prism in refractometer No.602905, i.e. scale S. In order to calibrate the readings in terms of actual refractive index to make them more universally applicable further calibration was necessary. The refractometer with the 1 B prism of cell C was calibrated against water (n_D at 20.0°C = 1.33300) and acetone (n_D at 19.4°C = 1.35890) and found to have a 1.20 scale division zero displacement, i.e. Scale S ≡ Zeroed 1 B + 1.20.

All refractometers used on other apparatus were zeroed using distilled water and then readings converted to scale S using the equivalence table 4, appendix A. This then enabled use of the refractometer calibrations for conversion to P.E. concentration. Prisms used for the experimental apparatus are shown below:-

APPARATUS	PRISM
Cell C	Scale S ≡ Zeroed 1B + 1.20
Cells A and G	Zeroed 1 A
Cell S and Fluidised bed apparatus	Zeroed 1 B

5. 3. Equilibrium Results.

5.3.1. Impure P.E.

In view of the anomalous results obtained with the impure P.E. in the previous work (1), where equilibrium values obtained from undersaturation and supersaturation appeared to overlap, this

investigation was continued with the more accurate refractometers simulating growth experiments (section 7.2) by using the same total quantities of P.E. and water, and approaching equilibrium from dissolution of the P.E. No "surface reaction" of the type encountered by the integration of the molecules into the crystal lattice during crystal growth would be expected in a dissolution process. It was often observed during growth experiments that growth apparently ceased some 0.5% m/v from the equilibrium concentration, and these "apparent equilibrium values" were dependent on the growth rate of the experiment. Dissolution, however, is thought to be only a diffusional process, and equilibrium should therefore be attained more rapidly than from growth, without the inhibiting surface reaction effect.

Tests were done primarily to find the equilibrium value relevant to a particular growth experiment and for this purpose the exact amounts of solute and solvent are required to obtain the right impurity concentration. Batches D (1.0 % Di-P.E., 5.5% Formal) and F (< 0.1 % Di-P.E., 5.5% Formal) were used to find the effects with and without Di-P.E. respectively. All solutions in the seeded growth experiments were made up to 250 cm^3 total volume with distilled water at 20°C. The required mass of P.E. for the dissolution test was therefore found from the equivalent initial growth run concentration, c_o% m/v, $\left(\text{i.e. } m_o = 2.50 \times c_o\right)$ and adding the seed mass used for that particular growth test (usually 2.0 g).

$$\therefore \quad \text{Mass of P.E.} = 2.50\, c_o + \text{Seed.}$$

The required volume of water to simulate the growth experiment

is then found from $$V = 250 - \frac{2.50\ c_o}{1.396}\ cm^3$$

where 1.396 is the P.E. density.

The exact weight of P.E. was washed into cell S held at the required temperature using the measured volume of water required (Vcm^3). The mixture was stirred and readings of the refractometer scale against time were taken after the solution attained the temperature of the experiment (this usually took about 20 min.). The readings were usually found to go through a maximum before attaining an equilibrium value. This was studied in more detail at 60°C. The results are shown in figure 5.4. Batch F with $<$0.1 % Di-P.E. is shown to attain a markedly enhanced solution concentration initially which then decreases slowly to an equilibrium value. The enhanced value obtained and the equilibrium value were both found to depend on the solute concentration present (cf.D.F.2 and D.F.3.). However with Batch D containing 1.0% Di-P.E. the degree of initial enhancement was very much reduced and the equilibrium value was attained very much more rapidly. Again the equilibrium concentration was found to depend on the total P.E. concentration present, but it was found that the equilibrium value attained was the same for both Batches D and F for equivalent total P.E. concentrations. It was therefore decided to use Batch D for all equilibrium concentrations required.

An isothermal dissolution test was therefore carried out, D.F.4, a repeat of D.F.2, by first warming the water to the test temperature and then adding the P.E. However a very similar result was obtained to D.F.2. As the P.E. could not be washed into the

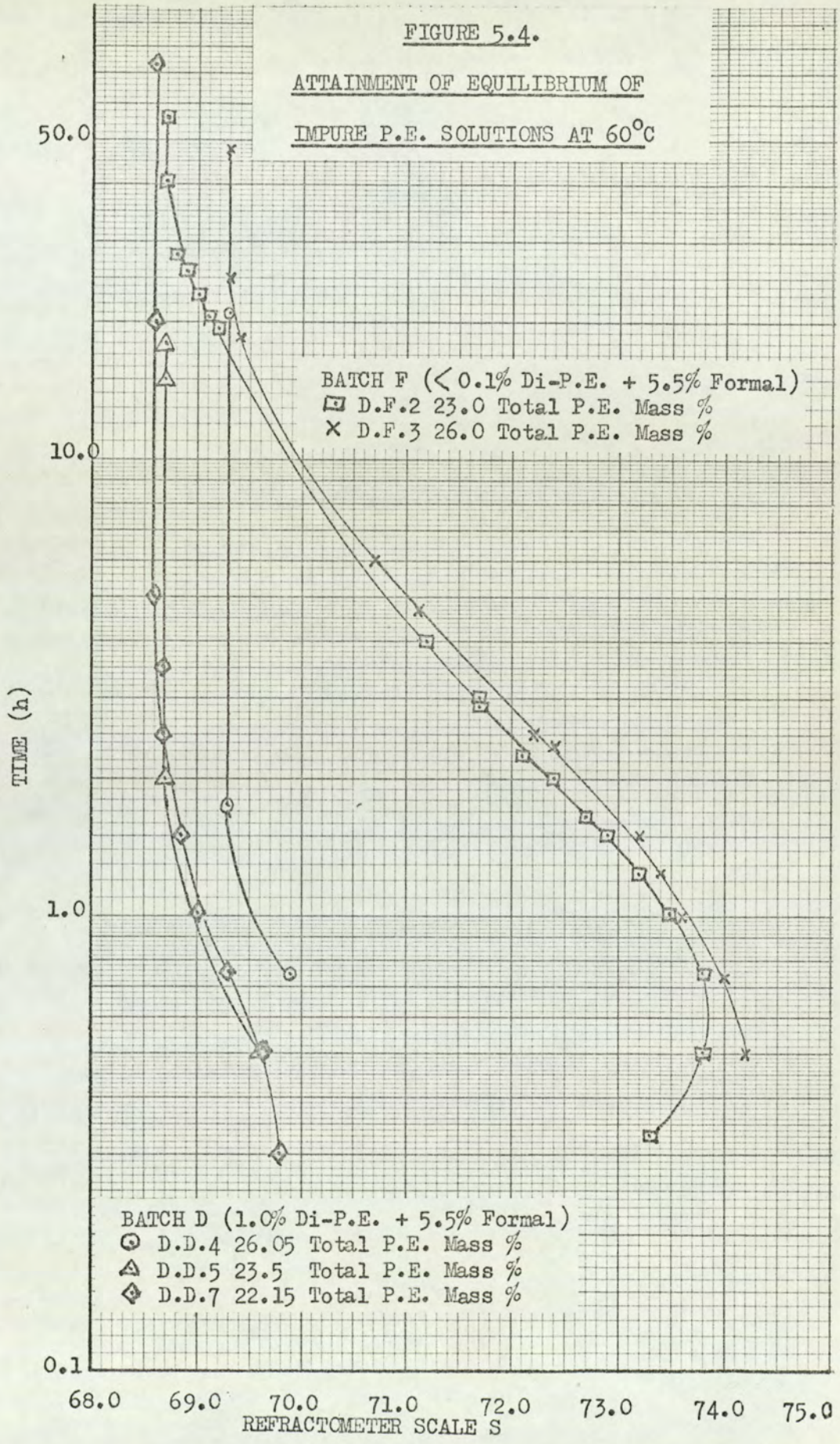


FIGURE 5.4.
ATTAINMENT OF EQUILIBRIUM OF
IMPURE P.E. SOLUTIONS AT 60°C
BATCH F (< 0.1% Di-P.E. + 5.5% Formal)
D.F.2 23.0 Total P.E. Mass %
D.F.3 26.0 Total P.E. Mass %
BATCH D (1.0% Di-P.E. + 5.5% Formal)
D.D.4 26.05 Total P.E. Mass %
D.D.5 23.5 Total P.E. Mass %
D.D.7 22.15 Total P.E. Mass %
TIME (h)
50.0
10.0
1.0
0.1
68.0
69.0
70.0
71.0
72.0
73.0
74.0
75.0
REFRACTOMETER SCALE S

cell with the isothermal test it was not as accurate as the previous method and dissolution tests were continued as before using Batch D material to obtain equilibrium quickly. At the lower temperatures this concentration enhancement was greater than at 60°C (for Batch D) and the maximum values attained (where recorded) are given in table 5, appendix A.

The equilibrium values obtained at 60°C, recorded as refractometer scale S, are plotted vs total P.E. % mass fraction in figure 5.5. Other values of the equilibrium scale, for use with the growth rate experiments at this temperature have been interpolated. The equilibrium results for all the impure P.E. dissolution tests are shown in table 6, appendix A. As the equilibrium values depend on the amount of P.E. present the results for an equivalent initial growth run supersaturation of about $c_o - c_\infty \simeq 4\%$ (the usual supersaturation used) have been plotted in figure 5.6. It can be seen that below about 48°C the results are greater than that expected from the $\log_{10} x$ vs $\frac{1}{T}$ correlation. The results $\geqslant 50°C$ only, for a supersaturation of about $\Delta c \simeq 4\%$, were correlated using a least mean squares analysis by the equation:

$$\log_{10} x = 5.073 - \frac{1265}{T}$$

where x = equilibrium concentration mass %; T = degrees Kelvin.

5. 3. 2. Pure P.E.

Although results in the previous work (1) were in good agreement with Cooke's data (75), a more accurate determination was

FIGURE 5.5

EQUILIBRIUM VALUES OF IMPURE P.E. SOLUTION AT 60°C

(Solution Equilibrium Concentration ≏ 20% m/v)

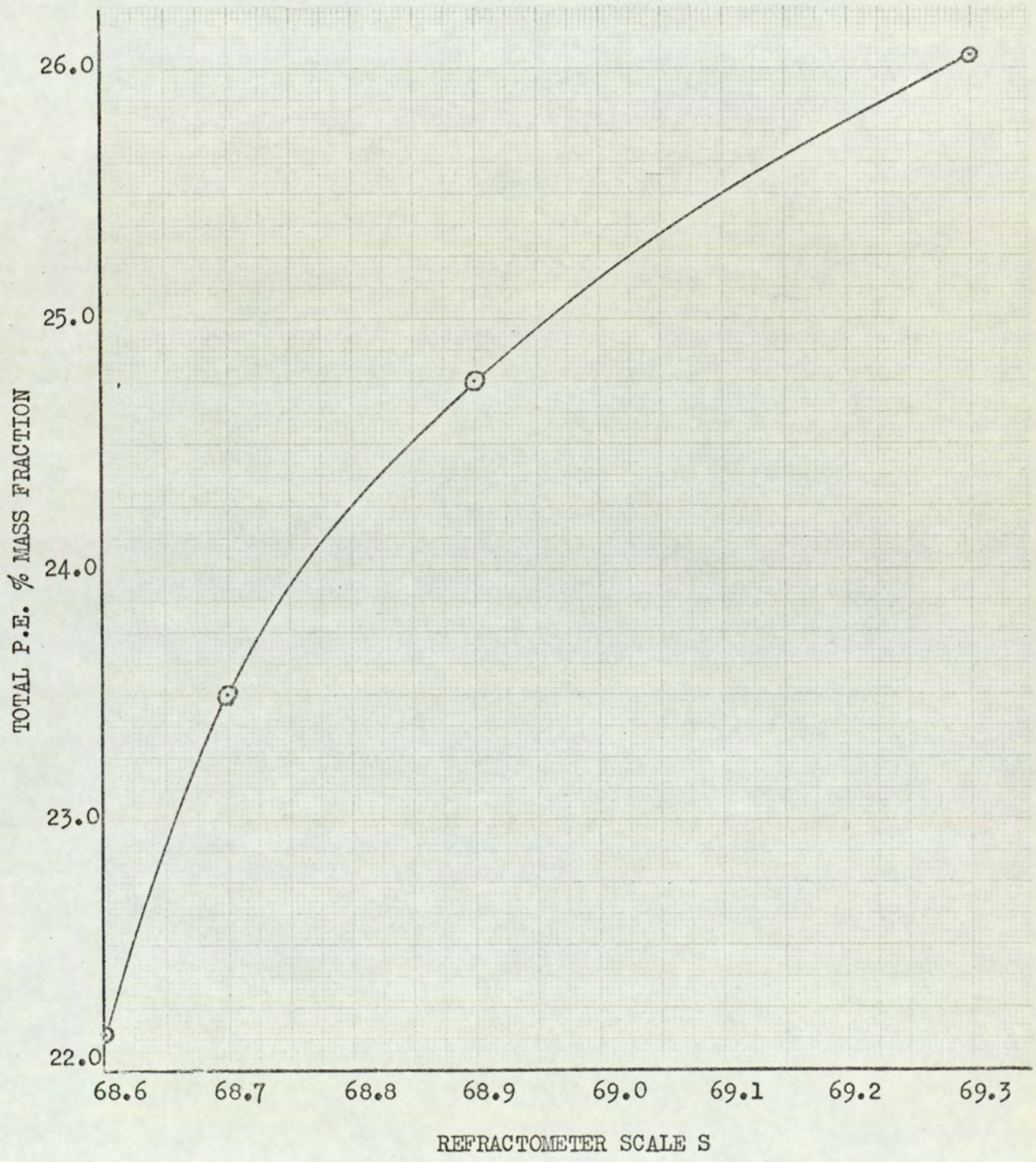
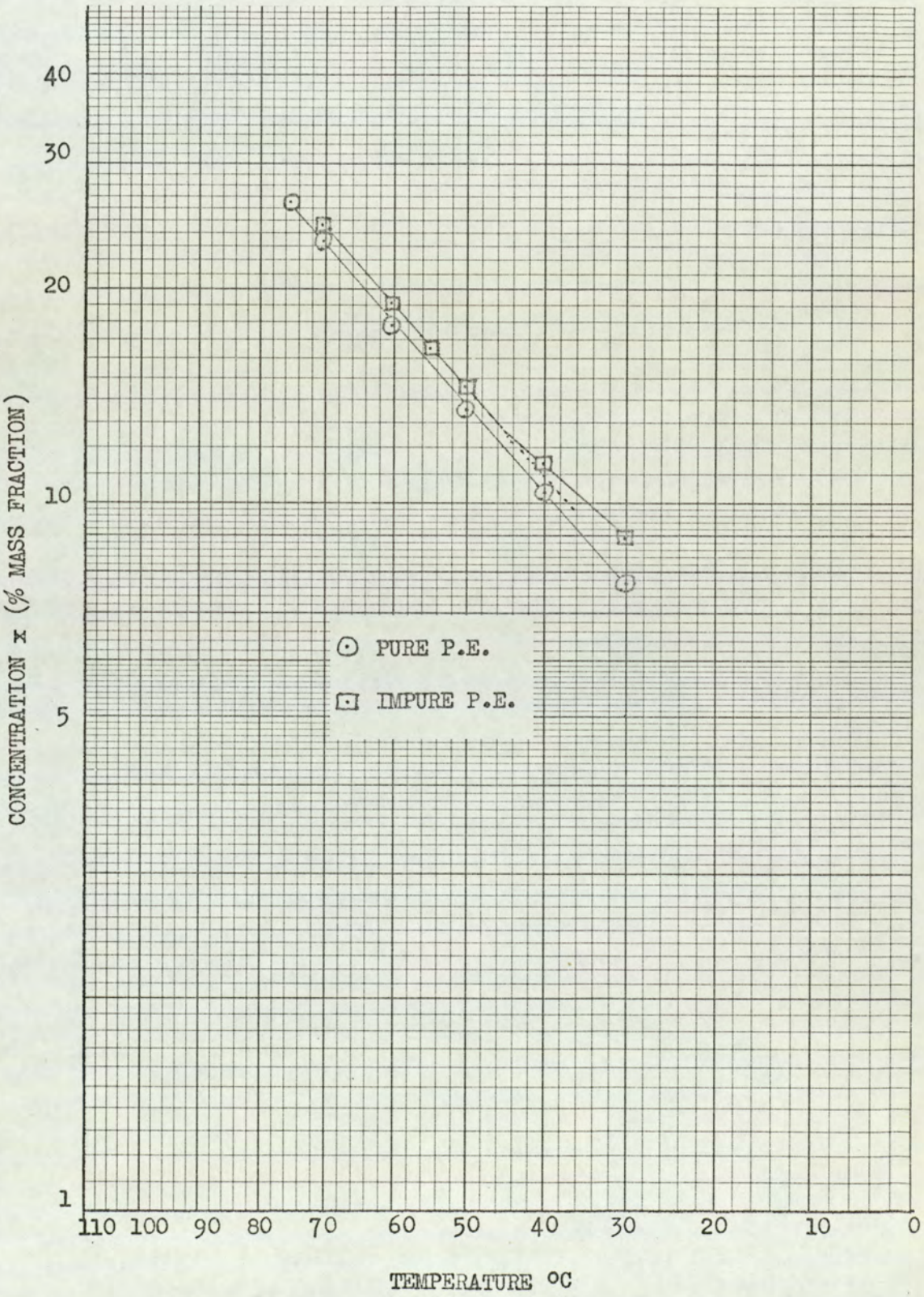


FIGURE 5.6.

P.E. EQUILIBRIUM IN AQUEOUS SOLUTION



necessary for use with the more accurate refractometers used in this work. At first dissolution tests were done as for the impure material but the equilibrium results obtained were found to apparently depend on the amount of material used, which was inexplicable.

A method was devised, therefore, carried out on the same samples as used for the Pure P.E. calibration, using the nucleated solutions and approaching equilibrium first by growth at one temperature and then by dissolution at a higher temperature. This was done by lowering the solution temperature after nucleation to the nearest decade (e.g. 70, 60°C etc.) and utilising the high surface area of the prolific nuclei to rapidly deplete the available supersaturation. After equilibrium was attained the temperature was raised and the equilibrium values at higher temperatures obtained by dissolution. By repeated tests from growth and dissolution at different temperatures using the calibration solutions (at 5% concentration intervals) it was shown that the equilibrium value was the same approached from growth or dissolution and regardless of the amount of P.E. used. The original dependence on the amount of material found in the early dissolution tests was thought to be due to an "Ostwald ripening" effect, i.e. an enhanced solubility of fine particles (section 2.1.). This was likely as the Pure P.E. used was finely ground with a pestle and mortar for easy handling, creating a lot of fine material. Whereas the nucleation tests would produce a fairly uniform macro-crystal product.

The results are shown in table 7, appendix A, and have been

correlated using a least mean squares analysis (figure 5.6):

$$\log_{10} x = 4.980 - \frac{1242}{T}$$

where x = equilibrium concentration, mass %; T = degrees Kelvin.

SECTION SIX

EXPERIMENTAL AND COMPUTATIONAL METHODS

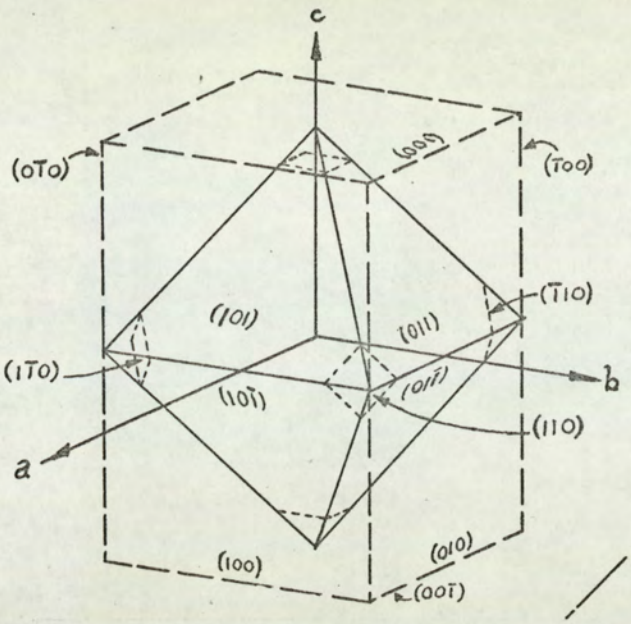
6. 1. Shape Factor.

As there is some discrepancy in the literature on the crystal structure of P.E., it was thought necessary to grow and measure single P.E. crystals and to show the effect of the occasionally occuring (001) faces and of the impurities on the shape factors. Figure 6.1A shows a sketch of a P.E. crystal with the indices of the main faces marked and also of the minor ones which sometimes appear.

6. 1. 1. Measurements on Pure P.E. Crystals.

There was little difficulty in growing P.E. crystals on the end of a wire in mother solution which was chromatographically pure, and measurements were made on crystals of ca. 0·25 inches grown under various conditions of super saturation and temperature. These crystals were transparent and often had a slightly rectangular, as opposed to square, bipyramid base which resulted in rectangular (001) faces which were often observed. When the (001) faces were absent these crystals then had knife edges at their tops and bottoms. The (110) faces were hardly ever observed and the (100) and (010) and (111) faces, although theoretically possible, were never observed. It was assumed that trace impurities were the cause of these erratic occurrences.

A typical crystal grown in "pure" P.E. at 60°C is shown in



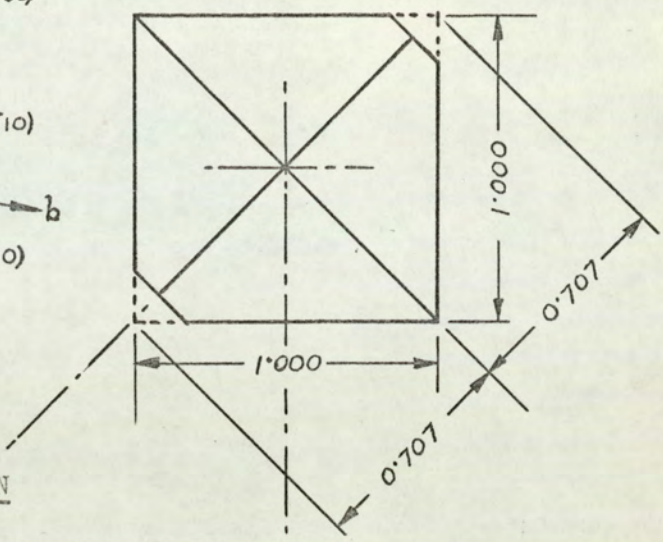


FIG. 6.1.A: PICTORIAL PROJECTION

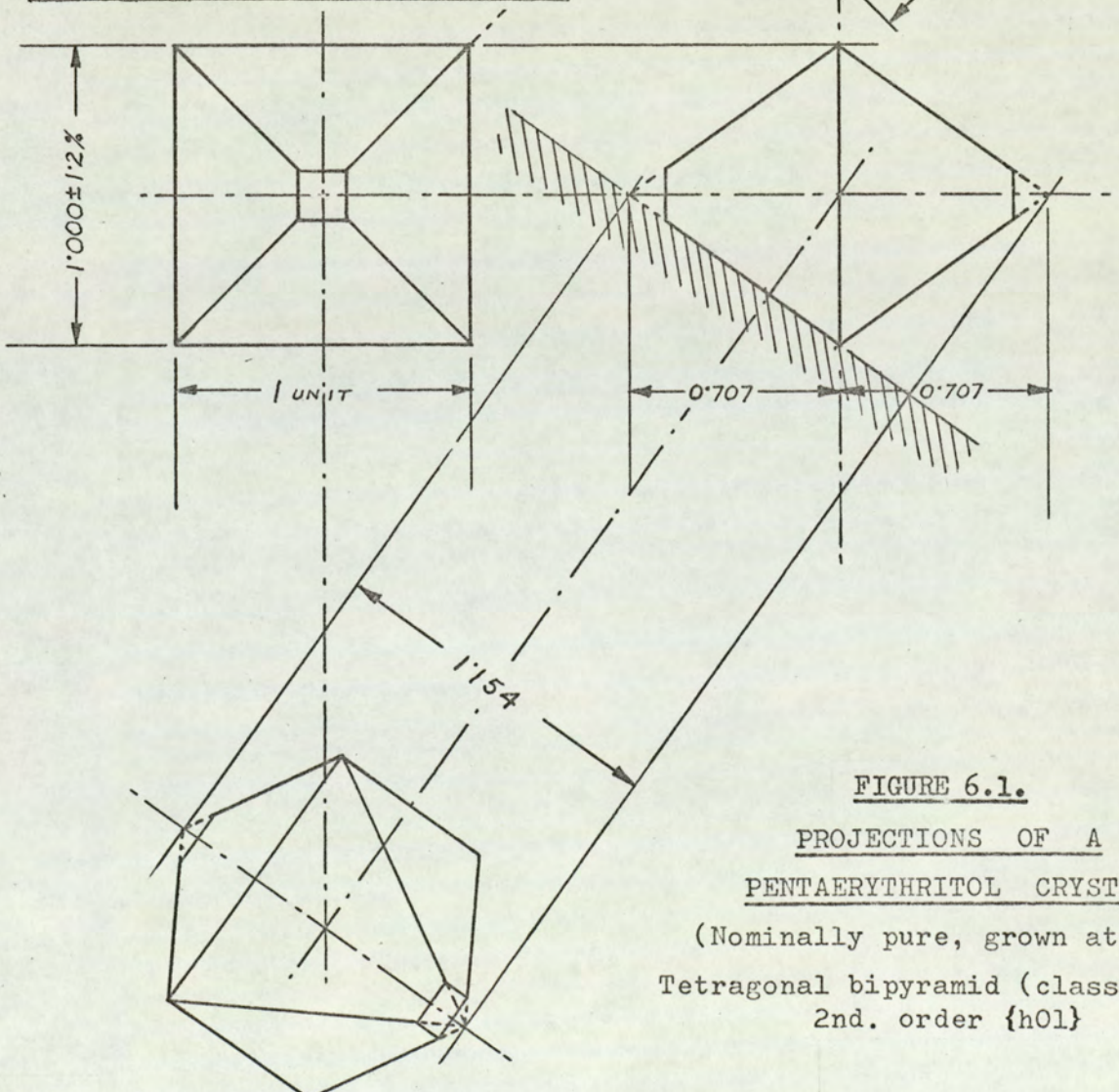


FIGURE 6.1.

PROJECTIONS OF A

PENTAERYTHRITOL CRYSTAL

(Nominally pure, grown at 60°C)

Tetragonal bipyramid (class 4/mmm)
2nd. order {h0l}

IG. 6.1.B : Appearance as seen by microscope showing true shape of one face

Figure 6.2. The departure from squareness of the base was ± 1.2% and c/a = 1·39 ± 2·5% (i.e. 1·356 to 1·426). This gives the lengths of the pyramid edges as 0·99 and the (normal) angle between the (101) and (10$\bar{1}$) faces as 71·5 ± 1·3°. For practical purposes and within experimental error the crystal may therefore be considered to be of the cubic system section 3. 1., with the (100) faces taken as true equilateral triangles with the normal angle between any two faces of 70·53°. This has been done in Figure 6.1B which also gives the crystal lying in its stable position as it would appear under a microscope: i.e. a regular pentagon of unit size across the pyramid base and 1·154 units across any pair of opposite corners.

6. 1. 2. Impure P.E. Crystals.

When crystals grown from impure (Batch C) P.E. were examined under the microscope there was no apparent departure from the square base bipyramid shape. Crystals larger than ca. 0·1 mm of good quality were rare and the faces of these usually appeared to have smaller crystals growing from them (as opposed to simple agglomeration). This is presumably due to faults in the lattice caused by impurity inclusions. An attempt to grow an impure crystal to reasonable size proved to be very difficult; the product was always opaque and it was not possible to prevent prolific outcrops of small crystals from the faces by temperature cycling.

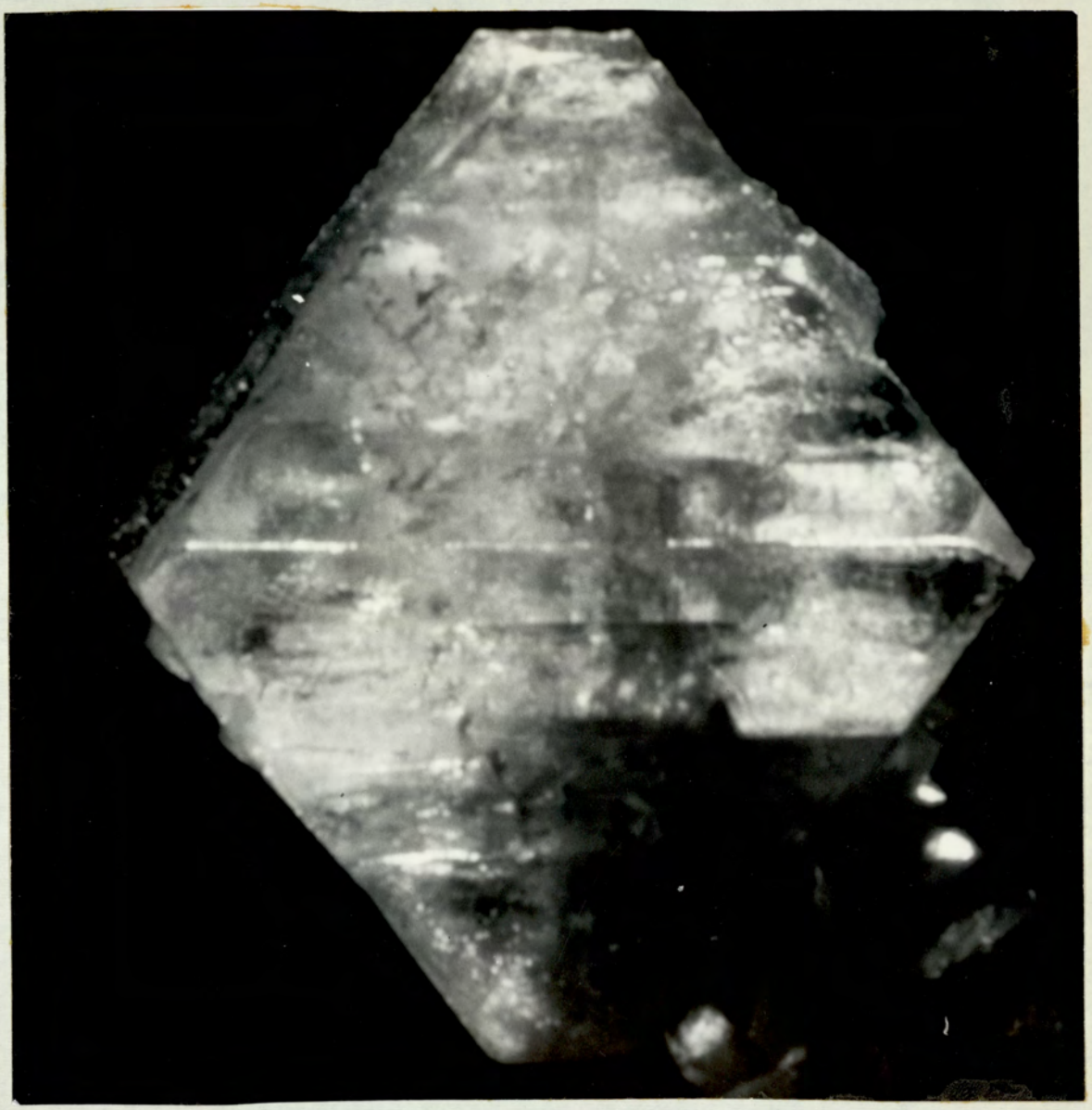
6. 1. 3. Shape Factors.

Taking the characteristic crystal dimensions, L , as the

FIGURE 6.2.

"PURE" P.E. CRYSTAL

(grown at 60°C)



length of the pyramid base, (Figure 6.1B), then for the ideal crystal:

$$\text{crystal volume, } v_p = \sqrt{\frac{2}{3}}\, L^3 = 0.4714\, L^3$$

$$\text{crystal surface, } a_p = 2\sqrt{3}\, L^2 = 3.4644\, L^2$$

For the crystal Figure 6.2, allowing for the missing points(which average ca $0.12L$ at each end of the c - axis):

$$\text{crystal volume, } v_p = (0.4714 - 0.0023)\, L^3$$

$$= 0.4691\, L^3$$

$$\text{crystal surface, } a_p = (3.4644 - 0.0422)\, L^2$$

$$= 3.4222\, L^2$$

Hence the missing points caused by the (001) faces represent 0.5% of the "ideal" crystal volume and 1.2% of the surface area. As this error is variable but small, shape factors will hereafter be based on the ideal crystal.

The Volume shape factor, ϕ, will be defined by:

$$v_p = \overline{11}\, \phi\, \frac{L^3}{6}$$

$$\text{Then } \frac{\overline{11}}{6}\, \phi = 0.4714$$

$$\underline{\phi = 0.900}$$

The Area shape factor, θ , will be defined by:

$$a_p = \overline{11}\ \theta\ \angle^2$$

Then $\overline{11}\ \theta = 3.4644$

$$\theta = 1.101$$

6. 2. Size Analysis.

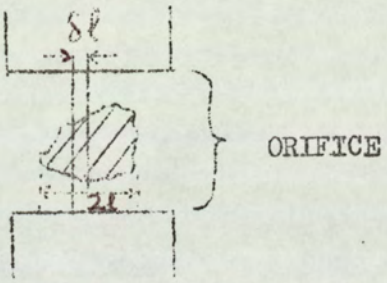
The Coulter Counter size analyser as used in the previous work (1) proved to be a useful method of size analysis. The basic assumption in its operation being that if the particle diameter is kept $<$ 40% of the orifice diameter, then the electrical response due to the particle is directly proportional to the volume of the particle. However, in fact it is shown, section 6.2.1.2., that a significant correction is necessary depending on the particle / crifice diameter ratio even below the stipulated limit. It is also shown in 6.2.1.2. that the effect of the particle shape is very small, and the error involved in obtaining an equivalent spherical diameter for attrited seed crystals from a stirred growth cell due to variable shape factors is considered minimal. This instrument also had the advantage of being able to measure small samples of suspension without the need for filtration of the suspension. However for crystal products it was often necessary to examine samples to determine the amount of agglomeration, and in the event of secondary nucleation or excessive attrition to obtain a size analysis over a wide range. For this purpose an Image Shear Microscope was used. As this involved a visual measurement of some chosen characteristic dimension the method

was unsuitable for attrited seed crystals with morphological variations. The Coulter Counter size analyser was therefore used for seed crystals and the Image Shear Microscope for product crystal measurements.

6. 2. 1. The Coulter Counter size analyser.

6. 2. 1. 1. Description.

The Coulter Counter determines the number and size of particles suspended in an electrically conductive liquid by passing a measured volume of suspension through a small orifice having an immersed electrode on either side of it.



Consider an element of cross-sectional area, a, thickness $\delta\ell$, of a particle length 2ℓ as it is orientated in the axis of the orifice. Then the change in resistance $\delta\Delta R_c$ due to this element is given by (83):

$$\delta\Delta R_c = -\frac{\Omega_o\, a\, \delta\ell}{A_c^2} \Big/ \left(1 - \frac{a}{A_c}\right) \quad \ldots\ldots \quad 6.1.$$

where A_c is the orifice area normal to the flow axis, and Ω_o is the electrolyte resistivity.

The basic assumption of the Coulter Counter principle is that if the particle diameter is kept $<40\%$ of the orifice diameter then the

electrical response can be considered to be directly proportional to the particle volume, v_c

$$\text{i.e. } \Delta R\Omega = \frac{\Omega_o}{A_c^2} v_c \quad \ldots\ldots \quad 6.2.$$

and an equivalent spherical diameter D_c can then be calculated. The necessary correction for this simplifying assumption is shown, section 6.2.1.2.

As the particles pass through the orifice the voltage pulses they produce are amplified and fed to a threshold circuit having an adjustable threshold level. If this level is reached or exceeded by a pulse, the pulse is counted. By pre-calibrating the threshold circuit and taking a series of counts at selected threshold levels, data are obtained for plotting a cumulative size distribution. Before plotting the counts are corrected for coincident particle passages and for the background count due to the extraneous particles already present in the electrolyte. To keep the coincidence corrections at a moderate level it is necessary to have a very high dilution. For the P.E. crystals counted this was found to be approximately 0.01g of crystals in 250 cm^3 of electrolyte. The electrolyte used was first filtered through a 0.45 μ porosity Millipore membrane filter to keep the background count as low as possible. An aqueous saline solution saturated with P.E. at 25°C was used as electrolyte and the calibration done over a temperature range of 14°C to 25°C (appendix B) to allow for room temperature variations.

6.2.1.2. Particle Size Effect.

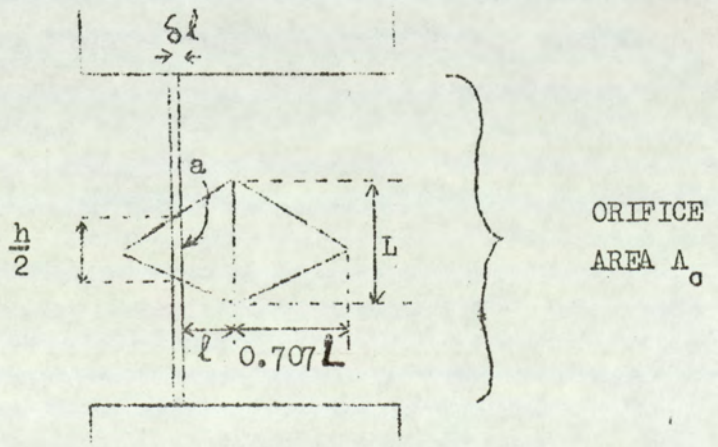
The simplified integration of equation 6.1.to give equation 6.2. gives rise to an error dependent on the particle size and shape. Allen (84) has shown that for a rod-shaped particle integration of equation 6.1. gives:

$$\Delta R = - \frac{\sim_o \; v_p}{A_c^{\,2}} \Big/ \left(1 - \frac{a}{A_c}\right) \qquad \ldots\ldots \qquad 6.3$$

where v_p is the volume of the rod $\left(v_p = 2a\ell\right)$.

The true equivalent spherical diameter, D, of the rod given as $D_c = 40\mu$ with a 100μ orifice (i.e. the 40% diameter ratio limit) is therefore 37.1μ, i.e. 7.8% error. Similarly for spherical particles Allen (84) showed an error in D of 3.6% at the 40% diameter limit with the 100μ orifice diameter tube. The following adjustment is therefore made to the calculated Coulter Counter diameter D_c to allow for the size and shape of the P.E. tetragonal bipyramid crystal. Two possible crystal orientations were considered:

ORIENTATION A: Pyramid Base perpendicular to Orifice Axis.



Consider an element of side h, area a, and thickness $\delta\ell$ perpendicular to the orifice axis at a distance ℓ from the pyramid base.

$$\frac{h}{L} = \frac{0.707L - \ell}{0.707L}$$

$$\therefore\ h = \frac{0.707L - \ell}{0.707}$$

$$a = h^2 = \left(L - \frac{\ell}{0.707}\right)^2$$

From equation 6.1 $\Delta R = \dfrac{-2\Omega_o}{A_c^2} \displaystyle\int_{\ell=0}^{\ell=0.707L} \frac{h^2}{1 - \frac{h^2}{A_c}}\, d\ell$

i.e. $\Delta R = \dfrac{-2\Omega_o}{A_c^2} \displaystyle\int_{\ell=0}^{\ell=0.707L} \frac{\left(L - \frac{\ell}{0.707}\right)^2 d\ell}{\left(1 - \frac{\left(L - \frac{\ell}{0.707}\right)^2}{A_c}\right)}$

$$\therefore\ \Delta R = -\frac{\Omega_o}{A_c^2}\left(0.707\, A_c^{\frac{3}{2}} \ln\left(\frac{1 + \frac{L}{\sqrt{A_c}}}{1 - \frac{L}{\sqrt{A_c}}}\right) - 1.414\, A_c L\right) \quad \ldots\ 6.4$$

and equating with equation 6.2

$$v_c = 0.707\, A_c^{\frac{3}{2}} \ln \left(\frac{1 + \frac{\angle}{\sqrt{A_c}}}{1 - \frac{\angle}{\sqrt{A_c}}} \right) - 1.414\, A_c \angle \quad \ldots\ldots \quad 6.5$$

$$\therefore \frac{v_c + 1.414\, A_c \angle}{0.707\, A_c^{\frac{3}{2}}} = \ln \left(\frac{1 + \frac{\angle}{\sqrt{A_c}}}{1 - \frac{\angle}{\sqrt{A_c}}} \right)$$

$$\text{Put } Y = \frac{v_c + 1.414\, A_c \angle}{0.707\, A_c^{\frac{3}{2}}}$$

$$\text{Then } 1 + Y + \frac{Y^2}{2!} + \frac{Y^3}{3!} + \ldots\ldots = \frac{1 + \frac{\angle}{\sqrt{A_c}}}{1 - \frac{\angle}{\sqrt{A_c}}}$$

As $v_c = \frac{\pi D_c^3}{6}$, the solution of this equation for any given D_c and orifice area A_c is conveniently found by computation of the Newton-Raphson approximation method (85). This involves equating the function to zero, differentiating the function, $f'\angle$, and obtaining the more approximate solution $\angle(2)$ from $\angle(2) = \angle(1) - \frac{f(\angle)}{f'(\angle)}$

In this case the initial $\angle$ tried is the original Coulter Counter

diameter D_c which serves as an approximate solution. The polynomial is terminated at the 9th term.

$$\text{Hence } f(\angle) = \frac{1 + \frac{\angle}{\sqrt{A_c}}}{1 - \frac{\angle}{\sqrt{A_c}}} - 1 - Y - \frac{Y^2}{2!} - \ldots\ldots \frac{-Y^8}{8!}$$

$$\text{If B, a constant} = \frac{1.414\ A_c}{0.707\ A_c^{\frac{3}{2}}}$$

$$f'(\angle) = \frac{\frac{2}{\sqrt{A}}}{\left(1 - \frac{\angle}{\sqrt{A}}\right)^2} - B - YB - \left(\frac{Y^2}{2!}\right) B - \left(\frac{Y^3}{3!}\right) B \ldots\ldots \left(\frac{Y^7}{7!}\right) B$$

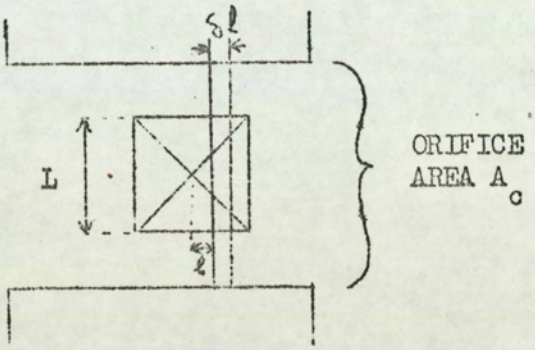
$$\angle(2) = \angle(1) - \frac{f(\angle)}{f'(\angle)}$$

which is repeated until $\frac{f(\angle)}{f'(\angle)} < 0.01\mu$. The computer program for a 280 μ orifice to find $\angle$ from values of D_c at 1μ intervals is shown appendix C.

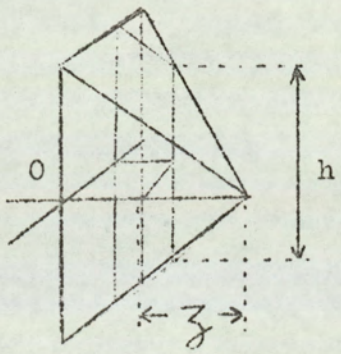
Hence assuming ϕ = 0.900

D, the true equivalent spherical volume diameter = $\left(0.900\ \angle^3\right)^{\frac{1}{3}}$

ORIENTATION B: Pyramid Base parallel to Orifice Axis and one edge perpendicular to it.



Consider A Quarter segment of the crystal; with the origin at the crystal centre as below:-



$$\frac{a}{2} = \frac{1}{2} L \cdot 0.707 L - \frac{1}{2} ʒ h$$

$$\frac{ʒ}{h} = \frac{0.707 L}{L} \quad \therefore \quad h = \frac{ʒ}{0.707}$$

$$\frac{ʒ}{0.707 L} = \frac{\ell \cdot 2}{L} \quad \therefore \quad ʒ = 1.414 \ell$$

$$\therefore \quad ʒ h = 2.828 \ell^2$$

$$\therefore \quad a = 0.707 L^2 - 2.828 \ell^2$$

From equation 6.1 $$\Delta R = \frac{-2\sim_o}{A_c^2} \int_{\ell=0}^{\ell=\frac{1}{2}L} \frac{a\, d\ell}{1-\frac{a}{A_c}}$$

$$= \frac{-2\sim_o}{A_c^2} \int_0^{\frac{L}{2}} \frac{0.707L^2 - 2.828\ell^2 \; d\ell}{1 - \frac{(0.707L^2 - 2.828\ell^2)}{A_c}}$$

$$\therefore \Delta R = -\frac{\sim_o}{A_c^2}\left\{ -A_c L + \frac{A_c^2}{0.707L\sqrt{\frac{A_c}{0.707L^2}-1}} \tan^{-1}\left(\frac{1}{\sqrt{\frac{A_c}{0.707L^2}-1}}\right)\right\}$$

and equating with equation 6.2:

$$V_c = -A_c L + \frac{A_c^2}{0.707L\sqrt{\frac{A_c}{0.707L^2}-1}} \tan^{-1} \frac{1}{\sqrt{\frac{A_c}{0.707L^2}-1}}$$

.... 6.6.

As $V_c = \frac{\pi D_c^3}{6}$ the solution of this equation for any given D_c and orifice area A_c is again conveniently found by computation of the Newton-Raphson approximation method as used with Orientation A.

Hence $$f(L) = \frac{0.707\, V_c\, L}{A_c^2}\left(\frac{A_c}{0.7L^2}-1\right)^{\frac{1}{2}} + \frac{0.707\, L^2}{A_c}\left(\frac{A_c}{0.707L^2}-1\right)^{\frac{1}{2}} - \tan^{-1}\frac{1}{\left(\frac{A_c}{0.707L^2}-1\right)^{\frac{1}{2}}}$$

$$\therefore f'(L) = \frac{0.707V_c L}{A_c^2}\left(\frac{-A_c}{0.707L^3}\right)\left(\frac{A_c}{0.707L^2}-1\right)^{-\frac{1}{2}} - \frac{0.707V_c}{A_c^2}\left(\frac{A_c}{0.707L^2}-1\right)^{\frac{1}{2}}$$

$$+ \frac{0.707}{A}L^2\left(\frac{-A_c}{0.707L^3}\right)\left(\frac{A_c}{0.707L^2}-1\right)^{-\frac{1}{2}} + \frac{1.4L}{A_c}\left(\frac{A_c}{0.707L^2}-1\right)^{\frac{1}{2}}$$

$$+ \frac{1}{L}\left(\frac{A_c}{0.707L^2}-1\right)^{-\frac{1}{2}}$$

which reduces to:

$$f'(L) = \frac{\left(\frac{A_c}{0.707L^2}-1\right)^{\frac{1}{2}}}{A_c}\left(\frac{0.707V_c}{A_c} + 1.414L\right) - \frac{\left(\frac{V_c}{A_c L}+2\right)}{L\left(\frac{A_c}{0.707L^2}-1\right)^{\frac{1}{2}}}$$

If D_c is put as the initial approximate solution for L

then $$L_{(2)} = L_{(1)} - \frac{f(L)}{f'(L)}$$

which is repeated until $$\frac{f(L)}{f'(L)} < 0.01\,\mu.$$

The computer program, 4, for a 280 μ orifice to find L from values of D_c at 1 μ intervals is shown appendix C. D can again be found from

$$D = \left(0.900L^3\right)^{\frac{1}{3}}$$

The comparison of corrected sizes using Orientation A and Orientation B respectively, with the 280 μ orifice, is:

	ORIENTATION A		ORIENTATION B	
D_c μ	L μ	D μ	L μ	D μ
10.0	10.33	9.98	10.36	9.99
50.0	51.34	49.57	51.37	49.60
100.0	100.03	96.58	100.27	96.81
150.0	143.36	138.66	144.54	139.56

As would be expected from the theory the variation of D from D_c becomes more significant at the larger values of the ratio particle diameter, D_c / orifice diameter D'. Similarly the larger this diameter ratio, the greater the difference between orientations A and B.

Because of the difficulty of growing a large perfect P.E. crystal it was decided to use a perspex model crystal in oil to simulate conditions of a small P.E. crystal in electrolyte, in order to find the stable orientation of a settling P.E. crystal.

For a P.E. crystal of D = 0.01 cm.

Terminal Falling velocity, u_t = 0.2 cm/s.

$$\therefore \quad Re \simeq \frac{0.2 \times 0.01 \times 1.0}{0.01}$$

$$= \frac{0.2}{}$$

Simulation with large model:

$$u_t = \frac{981\ D^2(\rho_s - \rho)}{18\mu}$$

$$\frac{u_t\ D\ \rho}{\mu} = 0.2$$

$$\therefore \quad \frac{981\ D^3\ (\rho_s - \rho)\ \rho}{18\mu^2} = 0.2$$

$$\therefore \quad \frac{D^3\ (\rho_s - \rho)\ \rho}{\mu^2} = 0.00367$$

If a perspex model crystal is used $\rho_s = 1.2$

If Shell Voluta Oil 45 is used at about 70°F

$\mu = 4.13$ Poise $\qquad \rho = 0.906$ g/cm^3

$$D = \left(\frac{0.00367 \times 4.13^2}{(1.2 - 0.906)\ 0.906} \right)^{\frac{1}{3}}$$

$$= 0.62 \text{ cm.}$$

Due to the acceleration conditions in a stirred beaker during a Coulter Counter size analysis, the crystal/solution relative velocity will be greater than u_t and therefore Reynolds number will be rather larger than 0.2, permitting a slightly larger model crystal.

A perspex model crystal was made having a pyramid base length

$\angle$ of about 1 cm. Each pyramid was painted a different colour and the model repeatedly and randomly dropped into a 1 dm^3 graduated cylinder filled with Shell Voluta Oil 45. The model invariably orientated on settling into the stable orientation B, with the pyramid base parallel to the cylinder axis.

Corrections for D_c were therefore computed for each of the Coulter Counter orifices used i.e. 50 μ, 280 μ and 560 μ, using the Orientation B program, 4, with the appropriate Ac values. The results are shown Tables 15 , 16 , 17 , appendix B.

6.2.1.3. Discussion.

Allen (84) did considerable work on the accuracy of a Coulter Counter. Investigation of the coincidence correction showed a tendency to overcount at low dilutions for counts of less than 5% of the maximum permitted count for 10% coincidence. However on a numerical basis the error was small. Edmundson (86) suggested a more accurate method of coincidence correction, but this involved serial dilutions of the sample at every threshold level, for each size analysis. The theoretical improvement of accuracy was thought unlikely to compensate for the practical errors involved. Allen (84) has also criticised this method on the basis that the linear correlation assumed is not necessarily correct. The coincidence correction method of Mattern (87) recommended by Coulter Electronics Ltd. is therefore used with the appropriate dilution limits.

Allen (84) showed a discrepancy of experimentally determined F_c factors with those published by Coulter Electronics (83).

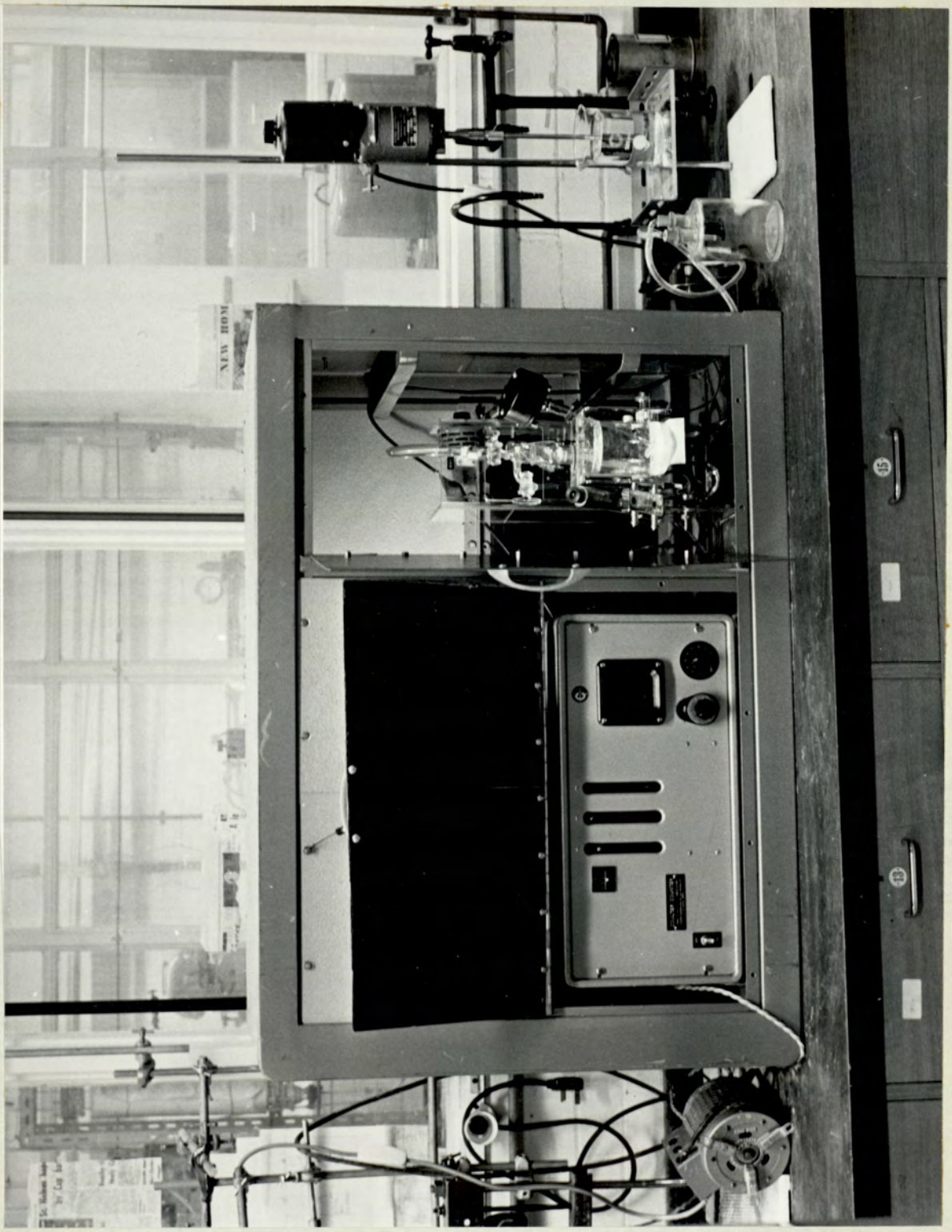
Lines (88) considered this to be a result of experimental error and also a symptom of a badly adjusted set, which seemed to need a zero adjustment of 2 or 3 threshold divisions . This was also indicated by the fact that Allen showed a variation of F_c factors with Gain setting which is not feasible. Lines considered the published F_c factors to be accurate to better than 1%. As the model A Coulter Counter used was regularly serviced, the published F_c factors were used.

Allen further found that the thermal effects during analysis and background noise made a lower size limit of 0.8 μ or even higher, impossible. This had also been experienced in the previous work (1) when it had been found necessary to switch off all external electrical motors in the vicinity. The Coulter Counter was therefore placed in a metal cabinet, figure 6.3, which acted as an effective shield from external interference.

In the previous work (1) an ultrasonic probe was used for sample dispersion. This had been compared with shaking the sample and dispersing in a paste with a brush and found to give reproducible results for Batch A seed crystals. This method was further investigated with Batch D seed crystals, 1.0% di-P.E., and found to cause crystal attrition. A comparison of stirring and ultrasonics, table 18 , for various degrees of agitation and for various periods of time showed an increase of the amount of attrition with both time and power using the ultrasonic probe, but a good reproducibility, regardless of time or stirrer speed, using a marine impeller type stirrer. Stirring was therefore used for sample dispersion for all Coulter Counter size analyses throughout this work, and the ultrasonic

FIGURE 6.3.

THE COULTER COUNTER PARTICLE SIZE ANALYSER



disperser dispensed with.

Size analyses of three samples of Batch E prepared seed sieve fraction 89 - 105 μ are shown tables 19 , 20 , 21 , appendix B. The collected and averaged sample analyses together with the corrected diameters are shown table 22 . It can be seen that the maximum deviation of $\sum$No % x D from the average is about 7% and the maximum deviation of $\sum$No % x D^2 from the average is about 12%. Because of the practical difficulties involved in using a two-tube technique, as in the previous work (1), with small seed crystals, a larger seed size was in general used throughout this work. The collected size analyses of seed materials after attrition at 2000 r.p.m. in the stirred cell C are shown table (24). A size analysis comparison of Batch C seed after attrition in cells A and C at different stirrer speeds is shown table (23). Collected size analyses of seed materials used in the modified fluidised bed experiments are shown table (25), and some crystal product size analyses are shown table (26), after growth in the stirred cell C. All these respective size analyses are the averaged result of three samples analysed with the Coulter Counter, each sample itself being the averaged result of three samples.

Calibration of the Coulter Counter, diameter corrections and all size analyses are shown appendix B.

6.2.2. The Image Shear Microscope.

The Image Shear Miscroscope described by Dyson (89) is an attachment which may be fitted to any microscope eyepiece and contains

two prism blocks which are rotated with respect to each other about a vertical axis by means of a micrometer drive. This results in the image, as seen through the eyepiece, being split into two complete images which may be sheared across each other in any direction by operation of the micrometer drive. The amount of shear involved is strictly proportional to the micrometer reading. If the two images are set edge to edge, the shear is proportional to the distance across that particular axis of the object, and can be calibrated using a microscope graticule. As this is only an eyepiece modification no restriction is placed on the method of microscopy or the magnification used.

A size analysis with the image shear microscope involves measuring a characteristic dimension for a number of particles. Figure 6.1B shows a P.E. tetragonal bipyramid crystal lying in its stable position as it would appear under a microscope. A convenient characteristic dimension is the size across any pair of opposite corners, $= 1.154\mathcal{L}$ where $\mathcal{L}$ is the length of the pyramid base. This is then readily converted to the equivalent spherical volume diameter, D, using the shape factor $\phi = 0.900$.

$$\text{Then} \quad D = \left(0.900\ \mathcal{L}^3\right)^{\frac{1}{3}}$$

To avoid selective sampling of good crystals, the microscope slide was moved at regular intervals under the microscope and all crystals under each view measured until a total of 50 had been counted for that particular slide. The slide sample was then renewed and the procedure repeated for four slide samples until a total of 200 crystals

had been sized. The calibration of the micrometer scale with a microscope graticule showed a linear relation over the range of the scale of 2.28 micrometer scale divisions = 10.0 μ.

Size analysis of attrited seed used for the stirred cells showed a poor agreement with that done with the Coulter Counter. This was attributed to the morphological variations caused by attrition a constant shape factor being essential for this method of analysis. However reasonable agreement was found . with the product of pure P.E. growth with little attrition, and the results of the product size analyses done are shown tables 27 , 28 , appendix B.

6. 3. Fluidised Bed Experiments.

6.3.1. Preliminary Fluidisation Experiments.

The apparatus is described in section 4.2.1. The temperature control units were switched on: T.C.2 being set to the required test temperature and T.C.1 to a temperature found from experience, which depended on the setting of T.C.2., the ambient temperature and the flow rate to be used in the particular test. The apparatus which contained diluted solution from a previous test took about 1 h to reach equilibrium and T.C.1. was then adjusted until the thermometer T.I. indicated the same temperature as T.C.2. The dilute solution was then purged from the system and made up with fresh concentrated solution until mother liquor of the required temperature and supersaturation (indicated by the refractometer) was obtained.
A known weight of carefully sieved seed crystals (prepared in small

batch crystallisations) was added to cold saturated solution in the feed funnel and the slurry quickly run into the crystalliser. The superficial velocity was usually of the order of 0.2 cm/s, representing for most seed sizes a very high degree of fluidisation (Figure 3.5.). However agglomeration of the growing crystals always occured and complete solidification of the bed had to be prevented by fitting the downcomer with a flexible seal and knee joint which could be used to break up agglomeration. After a suitable time depending on the temperature and supersaturation used the bed was dropped out of the crystalliser onto a Buchner filter and immediately washed with acetone. The product was weighed and examined. The size of the seed and product crystals was measured with a vernier microscope as the Image Shear Microscope was not available at this time. The characteristic dimension measured in this case was the distance across the crystal pyramid base, i.e. L. The average of 10 crystals was taken in each case with a standard deviation of the seed from the average being about 7 μ. This was considered reasonable as the vernier only read to 10 μ. The increase of bed mass was usually a more accurate method although it was often difficult to remove all the bed from the crystalliser.

6.3.2. Fluidisation experiments with modified apparatus (section 4.2.3)

6.3.2.1. Procedure.

T.C.1. was set to a temperature about 20°C above the required cell operating temperature. The system was filled with distilled

water and the glass taps set for flow through the appropriate rotameter. T.C.2. was set to a temperature slightly above the required cell temperature, and the immersion pump then switched on. The pump was controlled with a voltage regulator to give the estimated velocity required for fluidisation, indicated by the rotameter. The magnetic stirrer and hot-plate were switched on and controlled to give sufficient supplementary heat to the feed vessel to maintain the feed in an undersaturated condition about 15°C above the cell temperature. The heating tape on the transfer lines was then used as a fine control of the temperature in the cell. When the conditions were set with distilled water, the system was emptied using a vacuum pump and the hot undersaturated solution poured into the feed vessel. This solution of known concentration having previously been prepared in a 2 litre pyrex graduated flask.

When temperature conditions were stable with the circulating solution, the pump was stopped, the jubilee clip on the outlet line of the cell closed, the rubber bung removed and the seed crystals poured into the cell. The pump was immediately started and the heating tape temperature raised to compensate for the cold crystals. The bed was fluidised for a specific time with any hint of agglomeration being removed with the probe. As the crystals grew, the solution velocity had to be increased to maintain fluidisation, and each change in solution flow necessitated a corresponding change in heat input with the heating tape to maintain the cell temperature.

At the end of a run, the pump was stopped, the inlet and outlet lines to the cell removed and a vacuum line applied to the

inlet cone to remove the solution. The jacket connections were then removed, the jacket drained, acetone rinsed and finally air blown to dryness. The cell complete with crystals wet with the mother liquor was then weighed. The cell was dried for 24 h in an oven at 100°C and reweighed. This enabled the weight of the solid due to the mother liquor to be calculated and knowing the weight of the empty cell, the crystal product weight derived. The rate of mass increase for this particular supersaturation and temperature is thus calculated. As the initial size distribution is known from a Coulter Counter size analysis, this can be converted to linear growth velocity, g, as shown below.

6.3.2.2. Analysis of Results.

In the previous work (1) it was assumed that for a group of crystals of narrow size distribution, the average crystal size equals the size of the crystal of average area, which also equals the size of the crystal of average weight.

∴ Assuming a constant shape factor throughout growth:

$$\frac{A_2}{A_1} = \left(\frac{M_2}{M_1}\right)^{\frac{2}{3}}$$

However, this is invalid for a crystal size distribution. Although a narrow sieve fraction of seed crystals was used with a sieve aperture ratio of approximately 1.19, a more accurate size analysis is obtained using the Coulter Counter size analyser.

McCabe's ΔL law (53) states that if a known mass of seed

crystals of known size distribution is grown under given conditions of supersaturation, then the size analysis of the product is given by:

$$M(2) = \int_{0}^{M(1)} \left(1 + \frac{\Delta D}{D(1)}\right)^3 \, d\, M(1)$$

where M(2) is the product mass obtained from M(1) seed, D(1) is the seed size and Δ D the increase in size. The assumptions made are (a) a constant shape factor throughout growth and (b) the crystal size has no effect on growth rate.

For the present study the assumption of a constant shape factor appears reasonable. If, however, the rate of diffusion to the crystal surface controls the growth rate process, the variation of the relative crystal/solution velocity throughout the fluidised bed with crystal size distribution would cause a faster growth rate for the larger crystals, making this law invalid. As the preliminary experiments in a fluidised bed indicated integration rate control under the conditions examined, integration rate control will be assumed for these experiments in order to apply McCabe's ΔL law to obtain a more accurate growth velocity, g, for comparison with the stirred cell experiments.

From the Coulter Counter size analysis, the % No (J) of a mean equivalent spherical volume diameter D (1,J) is obtained for the crystal seed size distribution J, where J = 1 : P, the number of mean diameters in the analysis.

Volume of a crystal of diameter $D(1,J) = \frac{\pi}{6} \times D(1,J)^3$

Total volume of seed $= \frac{M(1)}{\rho_s}$

$\therefore$ Actual No. of crystals, No.(J), of diameter D(1,J)

$$= \%\,\text{No.}(J) \times \frac{M(1)}{\rho_s \times \sum\left(\%\text{No.}(J) \times \frac{\pi}{6} \times D(1,J)^3\right)} \quad \ldots\ldots 6.7$$

$$M(1) = \sum \rho_s \times \text{No.}(J) \times \frac{\pi}{6} \times D(1,J)^3 \quad \ldots\ldots\ldots 6.8$$

Now assuming McCabe's ΔL law, and that no attrition or agglomeration occurs during growth: D (2,J), the product size = $D(1,J) + \Delta D$

$$M(2) = \sum \rho_s \times \text{No.}(J) \times \frac{\pi}{6} \times \left(D(1,J) + \Delta D\right)^3$$

$$\therefore\ M(2) - M(1) = \rho_s \times \frac{\pi}{6} \sum \left[\text{No}(1)\left\{\left(D(1,1) + \Delta D\right)^3 - D(1,1)^3\right\} + \ldots\right.$$

$$\ldots + \text{No.}(2)\left\{\left(D(1,2) + \Delta D\right)^3 - D(1,2)^3\right\} + \ldots\ldots$$

$$\left.\ldots\ldots + \text{No.}(P)\left\{\left(D(1,P) + \Delta D\right)^3 - D(1,P)^3\right\}\right] \ldots\ldots 6.9$$

$$= \rho_s \times \frac{\pi}{6}\left[\Delta D^3 \sum \text{No}(J) + 3\,\Delta D^2 \sum\left(\text{No}(J) \times D(1,J)\right)\right.$$

$$\left. + 3\,\Delta D \sum\left(\text{No}(J) \times D(1,J)^2\right)\right] \ldots\ldots 6.10$$

Let $U = \sum \text{No.}(J)$

$$R = \sum\left(3 \times \text{No.}(J) \times D(1,J)\right)$$

$$Y = \sum\left(3 \times \text{No.}(J) \times D(1,J)^2\right)$$

$$Q = \frac{M(2) - M(1)}{\rho_s \times \frac{\pi}{6}}$$

Then $U\left(\Delta D\right)^3 + R\left(\Delta D\right)^2 + Y\left(\Delta D\right) - Q = 0 \quad \ldots\ldots\ldots 6.11$

This can be solved for ΔD by the Newton-Raphson approximation method where $f(\Delta D)$ = Equation 6.11

$$\text{Then } f'(\Delta D) = 3U(\Delta D)^2 + 2R(\Delta D) + Y \quad \ldots\ldots \quad 6.12$$

Try as a first approximation $\Delta D = 0.0001$ cm.

If $\frac{f\,\Delta D}{f'\Delta D}$ is less than 0.0000001 cm, the solution is sufficiently accurate, and $D(2,J) = D(1,J) + \Delta D$ for each seed diameter.

Otherwise a more accurate solution $\Delta D(2)$ is obtained from:

$$\Delta D(2) = \Delta D(1) - \frac{f(\Delta D)}{f'(\Delta D)}$$ where $\Delta D(1)$ is the previous value. The iteration is repeated until the required accuracy is obtained.

The growth velocity of a crystal face, g, is then obtained from:

$$g = \frac{\Delta D}{2(\Delta t)}$$

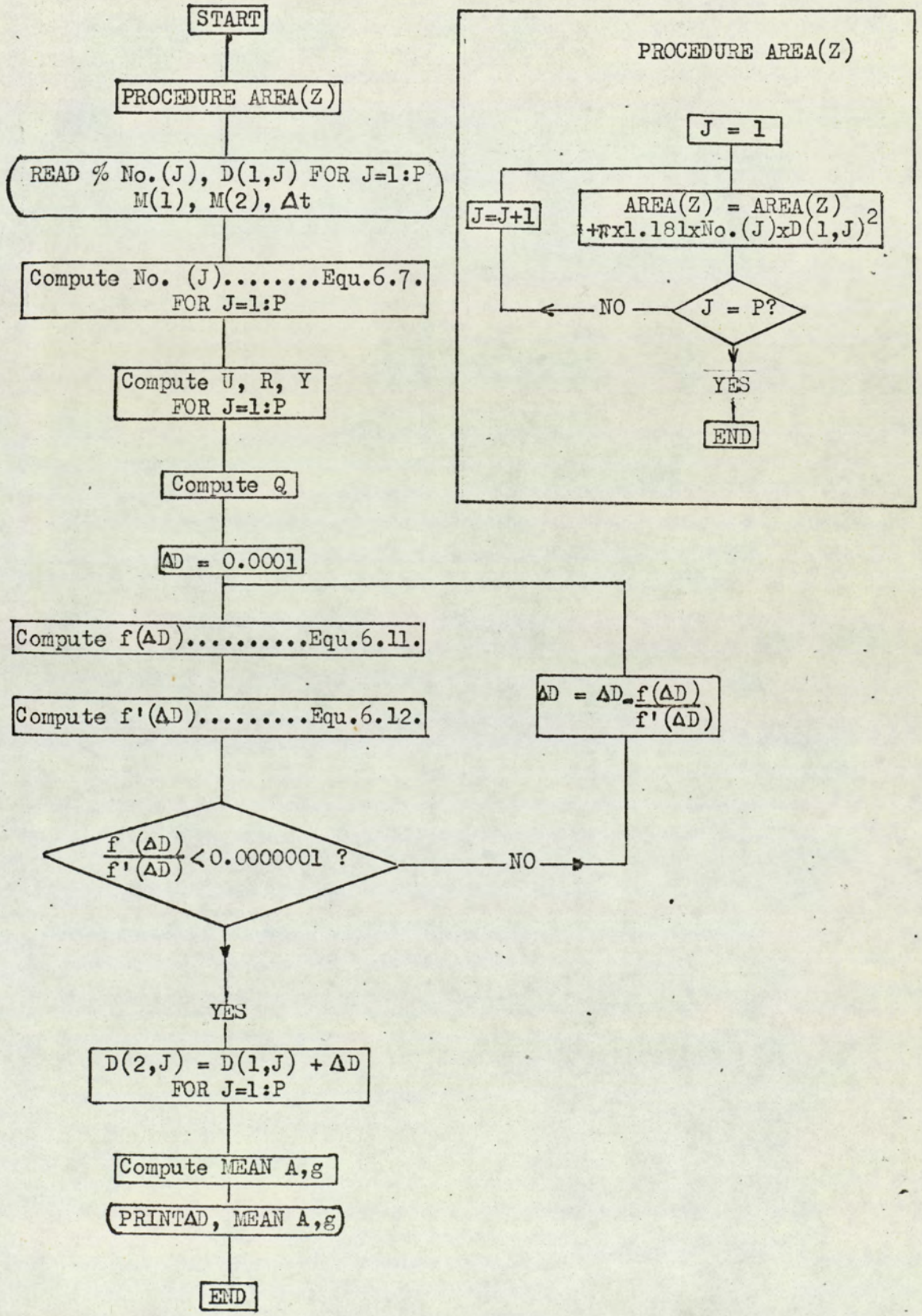
where Δt is the time of the experiment in minutes.

In order to find a rate constant, Km, for the rate of crystal mass increase it is necessary to know the mean area during growth. This can now be obtained as the final size distribution is known:

Volume of a single crystal, $v_p = \pi \phi \frac{L^3}{6} = \frac{\pi D^3}{6}$

$$\therefore \quad L = \frac{D}{\phi^{\frac{1}{3}}}$$

PROGRAM 1 : CRYSTALLISATION IN A FLUIDISED BED



Surface area of a single crystal, $a_p = \pi \theta L^2$

$$= \pi \frac{\theta}{\phi^3} D^2$$

For a tetragonal bipyramid crystal of P.E. $\frac{\theta}{\phi^3} = 1.181$

$\therefore$ Total Seed Area, $A(1) = \sum \pi \times 1.181 \times \text{No.}(J) \times D(1,J)^2$

Total Product Area, $A(2) = \sum \pi \times 1.181 \times \text{No.}(J) \times D(2,J)^2$

Mean Area, $A = \frac{A(1) + A(2)}{2}$

As the mother liquor concentration, c, can be found from the refractometer reading and calibration, the rate constant K_m can be found from $\frac{dm}{dt} = -K_m A \left(c - c_\infty\right)$ assuming a first order integration rate. c_∞ is the equilibrium concentration.

The computation of this analysis of the fluidisation results is shown in computer program 1 (appendix C) written in Elliott 803 version of Algol 60.

6.4. Stirred Cell Experiments.

6.4.1. Introduction.

In the previous work (1) growth rates were obtained by following the decrease in concentration of seeded solutions by refractometry. Some preliminary results with nucleation tests seemed to suggest that the growth rate was dependent on seed source. This was thought to be due to the possible variation of impurity partition

coefficient with temperature and hence a variation of seed impurity concentration. A simplified method of studying this phenomenon was tried by cooling a hot solution until nucleation occured, then stirring the suspension at this temperature, following the growth of the nuclei with the decrease in concentration of the solution. The product was discharged into acetone, filtered and size analysed. A mono-sized product was expected from the growth of the nuclei which would enable a growth rate constant to be calculated. However, size analysis of the product with both the Image Shear microscope and the Coulter Counter showed not only agglomeration of the crystals but also a surprisingly wide size distribution. This was attributed to non-instantaneous nucleation of the solution. Attempts to avoid this product size distribution by cooling the mother liquor into the metastable zone and inducing prolific nucleation with an ultrasonic probe, and also by adding a small amount of the dispersant NONIDET P40, were unsuccessful.

6.4.2. Seeded Solutions.

6.4.2.1. Seed.

Because of the possible sensitivity of P.E. growth rate to seed impurity concentration, care was taken to record conditions of all seed preparations. It had been found in the previous work (1) that a better quality seed containing many discreet well formed crystals in the lower size ranges was obtained by controlling the growth temperature in the preparation and washing the product with acetone. In general

this was done with the material to be used for the growth rate experiments and a 30% m/v solution was cooled to the nucleation temperature and stirred at this temperature for 3h. The resulting crystals were filtered, acetone washed and dried in an oven at 60°C. The dry product was then sieved and classified into close-cut sieve fractions. A sample of the sieve cut to be used as seed was placed in the growth cell filled with saturated solution at room temperature. This was then stirred under the hydrodynamic conditions to be used in a growth run and the attrited seed size analysed with the Coulter Counter after 15 minutes attrition. The analysis of each was checked after a further 12 h or so, stirring and it was found, as in the previous work (1), that all the attrition occured in the first few minutes of stirring. The smallest sieve fraction, 44 - 64 μ, was often found unsatisfactory because of the necessity to use a two-tube technique for analysis with the Coulter Counter with its inherent practical difficulties, and a larger sieve fraction was then used. The size analyses of the attrited seed used in the growth rate experiments are shown in appendix B.

The optimum mass of seed crystals was chosen as 2g after the following considerations:

(a) This involved a relatively small solid/solution ratio for a period of a run and consequently did not over obscure the refractometer image.

(b) The heat of crystallisation was small and the solution temperature could be easily controlled with the thermostatically

controlled circulator.

(c) As the refractometer scale could only be read to 0.1 divisions and estimated to 0.05 divisions, the smallest interval considered consistent with the required accuracy of the experiment was 0.5 divisions. The average time taken for this concentration change using 2g of seed was usually of the order of 5 to 50 minutes. This is considered to be the optimum time interval for the experiment as at least half a minute is necessary to accurately read a scale partly obscured by suspension and longer runs would be unnecessarily laborious.

6.4.2.2. Procedure.

The cells used were as shown in section 4.4. In order to obtain a more accurate analysis of results especially as the concentration approached equilibrium, evaporation tests were done for cells C and S under the stirred experimental conditions for growth by measuring the concentration decrease of an unsaturated solution over a period of time at constant temperature. The results for various temperatures are shown Fig.6.4. Appendix C on a log evaporation vs $\frac{1}{T}$ plot with linear correlations drawn according to the Clausius - Clapeyron relation. Interpolated values from these correlations for the required growth experiment temperatures are shown table 29

Although instantaneous nucleation would not be expected (Fig.3.2.) with a supersaturation less than about 7% m/m for pure P.E. and 10% m/m for impure P.E., it was found in the previous work (1) that in an agitated cell a 6% m/m supersaturation impure solution would nucleate in about 9 h and a 4% m/m supersaturation solution would

nucleate in about 50 h. It was therefore considered safe to work at initial supersaturations of about 4% m/m. The required amount of P.E. was accurately weighed out and washed into a 250 cm^3 pyrex volumetric flask with a screw cap which had been selected for having the volume line very low on the neck to allow for solution expansion on heating. The volume was made up to 250 cm^3 with distilled water taking care to eliminate air bubbles, the flaskwas warmed until the P.E. had dissolved the cap secured and the solution shaken. The solution was poured into the dry growth cell held at the temperature of the run. The cell had previously been well washed with hot distilled water. The solution was stirred at the required stirrer speed for the run, set with a tachometer, until the required run temperature was obtained and the refractometer reading was then taken. The required weight of seed crystals was weighed into a clean sample bottle and poured into the solution simultaneously starting the stirrer and the stop clock. The sample bottle was reweighed and the seed weight derived. Readings of time for fixed intervals of concentration change were taken over the period of the run.

An optimum was required of the two conflicting considerations:

1. The larger Δc taken the more accurate that particular reading for the mean concentration.
2. The smaller Δc taken the easier to detect the growth rate dependence on supersaturation.

It was decided to use intervals of 0.5 divisions scale reading, i.e. ca 0.14 % P.E. As the refractometer scale could only

be estimated to 0.05 divisions, reading errors of ± 10% can be expected, but should be largely self-compensating for any individual run as results ar e taken consecutively.

At the end of a run the drain plug was removed and the suspension dropped into a beaker of acetone. The product was filtered, dried and occasionally size analysed with the Image Shear microscope.

6.4.2.3. Analysis of Results:

The actual number of crystals, No.(J), of diameter D(1,J) can be found from the intial size distribtuion of the attrited seed, as in the fluidised bed experiments from equation 6.7, where M(1) is the initial seed mass.

Now assuming McCabe's ΔL law, which should be true for a well stirred vessel, and provided no attrition, agglomeration or new nuclei occur during a growth experiment, then the number of crystals of any diameter D(I,J) at any time t(I) is No.(J) and the size distribution can be calculated from $D(I,J) = D(1,J) + \Delta D(I)$

	TIME I COUNTER		
	D (1,1)	D (I,1)	D(N,1)
	.	.	.
	.	.	.
SIZE	.	.	.
DISTRIBUTION			
J	D (1,J)	D (I,J)	D(N,J)
COUNTER	.	.	.
	.	.	.
	.	.	.
	D (1,P)	D (I,P)	D(N,P)

Where N is the number of readings of time.

P is the number of mean diameters of the size distribution.

A crystal growth velocity can therefore be determined by computation of the results of refractometer scale, hence solution concentration, against time in a similar method to that used for the fluidised bed experiments, for each time interval.

As a number of readings are obtained for different supersaturation for each run, the rate constant K assuming a first order integration rate as suggested by the previous work (1) can be calculated from $\frac{dc}{dt} = -K A (c - c_\infty)$ which when integrated gives:

$$K = \frac{1}{\Delta t(I)\ \overline{A}\ (I)} \ln \frac{\left(c(I) - c_\infty \right)}{\left(c(I+1) - c_\infty \right)} \quad .... \quad 6.13$$

where $\Delta t(I)$ is the time interval $t(I + 1) - t(I)$ for the concentration decrease $c(I)$ to $c(I + 1)$, $\overline{A}(I)$ is the mean crystal area over this time interval, and c_∞ is the equilibrium concentration at the temperature of the run.

K can be related to K_L of the first order growth velocity equation:

$$g = \frac{dr}{dt} = K_L \left(c - c_\infty \right) \quad ..... \quad 6.14$$

where r is the equivalent spherical volume radius, by the conversion shown appendix C

As the final time interval to reach equilibrium is not measurable, the computation is terminated at the penultimate concentration value of c(N). The equilibrium value c_∞ is calculated from the input refractometer scale data for c(N + 1).

All concentrations are derived from the isothermal refractometer correlations for the particular material and temperature, tables 2 and 3

$$c(I) = F + B\ S(I) + G\ S(I)^2 \qquad \ldots\ldots\ 6.15$$

As the total volume of solution used initially is 250 cm^3 at ca.25°C and the only loss of solvent throughout a run is through evaporation, the volume of solvent V(I) at any time (I) is given by:

$$V(I) = 250 - \frac{m(1)}{\rho s} - t(I) \times \text{Evapn.}$$

where m(1) is the mass of solute initially in solution and the evaporation rate in cm^3 / min is given in table 29 appendix C

But $m(1) = 2.5\ c(1)$ and $\rho s = 1.396$ for P.E.

$\therefore\ V(I) = 250 - 1.7908\ c(1) - t(I) \times \text{Evapn} \quad \ldots\ 6.16.$

$$c = \frac{100\ m}{\frac{m}{\rho s} + V}$$

$$\therefore\ dm = \frac{dc}{100\ V}\left(\frac{m}{\rho s} + V\right)^2$$

But $m(I) = c(1) \times 2.5 - \left(M(I) - H\right)$

where H is the intial seed mass = M(1)

$\therefore$ After any time interval $\Delta t(I)$, knowing M(I) which when I = 1, equals H, $M(I+1)$ can be calculated from:

$$M(I+1) = M(I) + \frac{c(I) - c(I+1)}{100\ V}\left(\frac{c(1) \times 2.5 - M(I) + H}{1.396} + V(I+1)\right) \qquad \ldots\ 6.17.$$

$\Delta D(I)$ for this time interval $\Delta t(I)$ can now be calculated

using a similar equation to 6.10:

$$M(I+1) - M(I) = \rho_s \times \frac{\pi}{6}\left[\Delta D(I)^3 \sum \text{No.}(J) + 3\,\Delta D(I)^2 \sum \text{No.}(J) \times D(I,J) + 3\,\Delta D(I) \sum \left(\text{No.}(J) \times D(I,J)^2\right)\right] \quad \ldots\ldots 6.18$$

This is solved for $\Delta D(I)$ using the Newton-Raphson approximation method as before:

$$f\ \Delta D(I) = U\,\Delta D(I)^3 + R(I)\,\Delta D(I)^2 + Y(I)\,\Delta D(I) - Q(I) = 0 \quad \ldots\ldots 6.19$$

where $U = \sum \text{No.}(J)$

$$R(I) = \sum \left(3 \times \text{No.}(J) \times D(I,J)\right)$$

$$Y(I) = \sum \left(3 \times \text{No.}(J) \times D(I,J)^2\right)$$

$$Q(I) = \frac{M(I+1) - M(I)}{1.396 \times \frac{\pi}{6}}$$

$$f'\Delta D(I) = 3\,U\,\Delta D(I)^2 + 2\,R\,\Delta D(I) + Y(I) \quad \ldots\ldots\ 6.20$$

The first approximation used is $\Delta D(I) = 0.0001$ cm and the iteration repeated until $\frac{f\ \Delta D(I)}{f'\Delta D(I)} < 0.0000001$

Then $D(I+1, J) = D(I,J) + \Delta D(I)$

The growth velocity $g(I)$ is now obtained from

$$g(I) = \frac{\Delta D(I)}{2 \times \Delta t(I)} \quad \ldots\ldots 6.21$$

Mean Area $\overline{A}(I)$ over this growth interval is given by

$$\overline{A}(I) = \frac{A(I+1) + A(I)}{2}$$

where $A(I+1) = \sum \pi \times 1.181 \times No.(J) \times D\left(I+1,J\right)^2$

$A(I) = \sum \pi \times 1.181 \times No.(J) \times D\left(I,J\right)^2$

Hence K(I) can be calculated from equation 6.13.

In order to compare results with Chernov's correlation (46) equation 2.22, where:

$$g = k_L \, s^b$$

the mean supersaturation $s(I)$ of this interval is given by:

$$s(I) = \frac{\frac{c(I+1) + c(I)}{2} - c(N+1)}{c(N+1)}$$

the computation of this analysis for each interval of refractometer scale has been done, computer program 2, using the Elliott 803 version of Algol 60.

PROGRAM 2 : CRYSTALLISATION RATE OF P.E. IN A SEEDED CELL

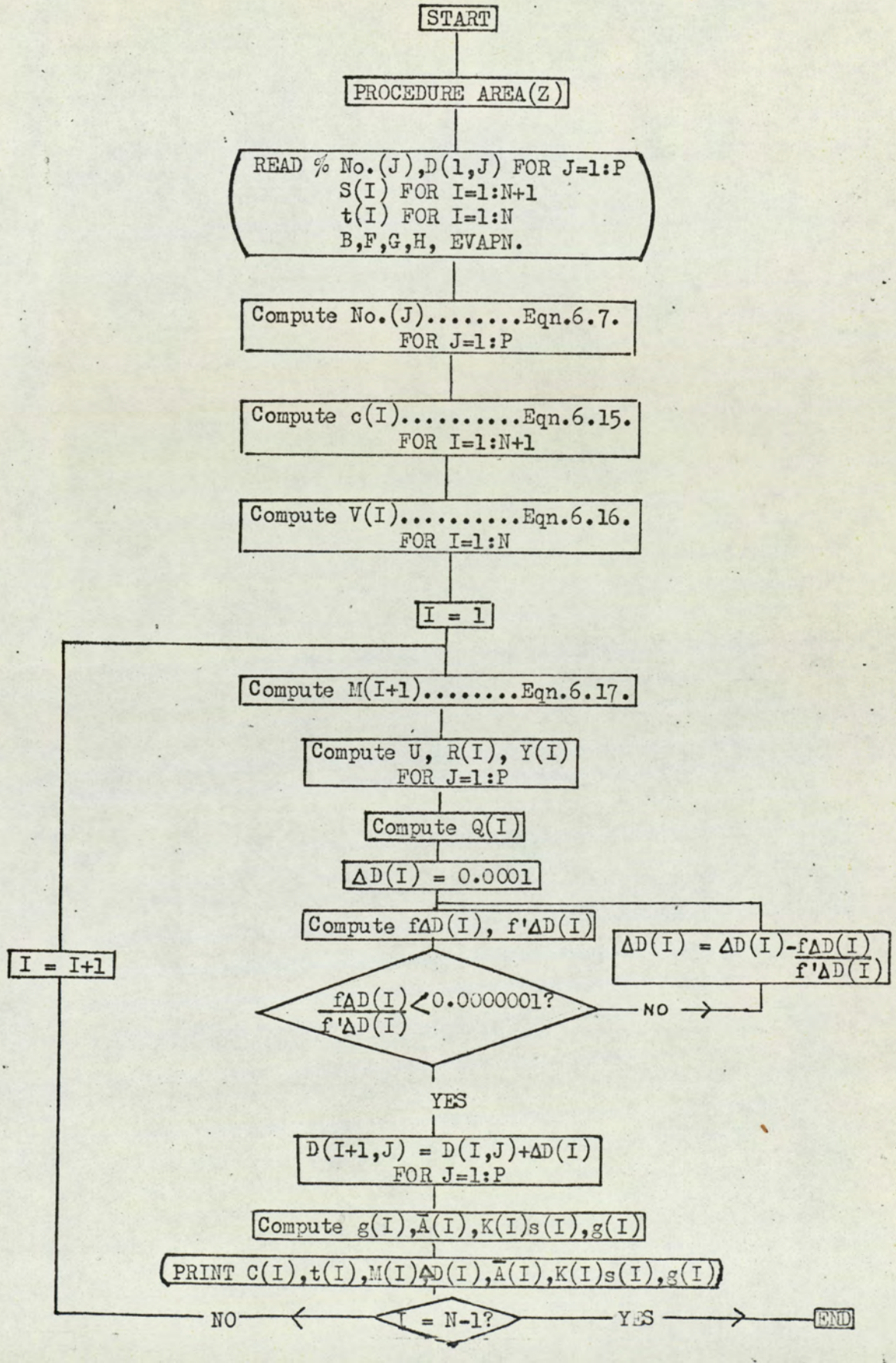


TABLE: 30

EXPERIMENTAL LEGEND

SUFFIX	SOLUTION	ADDITIONAL TREATMENT OF MATERIAL	EXPLANATION
D			DISSOLUTION experiments simulating growth experimental conditions to obtain equilibrium values.
F			FLUIDISED bed experiments to obtain growth rate data.
R			RUNS of seeded solutions in a stirred cell to obtain growth rate data.
	A to G		Different bags and batches of production material labelled Batches A to G.
	P.A. to P.G.		Batches A to G purified of di-P.E. and 'formal' by HCl reflux.
	D.P.E.		A synthetic mixture of di-P.E. and Purified Batch E.
		F	The preceeding material having been made into a 10% solution, filtered through a 0.45μ filter, and evaporated to dryness before use .
		M	The preceeding material having been made into a 10% solution and stirred for 2 hours with 10% m/v Molecular Sieve Type 13X before filtering and evaporating to dryness before use.

VARIOUS COMBINATIONS OF THE ABOVE HAVE BEEN USED.

EXAMPLES:

RUN No.	EXPLANATION
D.D. 4	The fourth dissolution experiment using BATCH D solution.
F.E. 1	The first fluidised bed experiment using BATCH E solution.
R.P.G.M. 2	The second run of a seeded solution of purified BATCH G in a stirred cell. The purified Batch G having previously been stirred in solution with Molecular Sieve Type 13X.
R.E.F. 3	The third run of a seeded solution of BATCH E in a stirred cell. The BATCH E having previously been filtered in solution through a 0.45μ filter.

SECTION SEVEN

RESULTS

The experimental legend is shown on the previous page.

7. 1. Growth Rates in a Fluidised Bed.

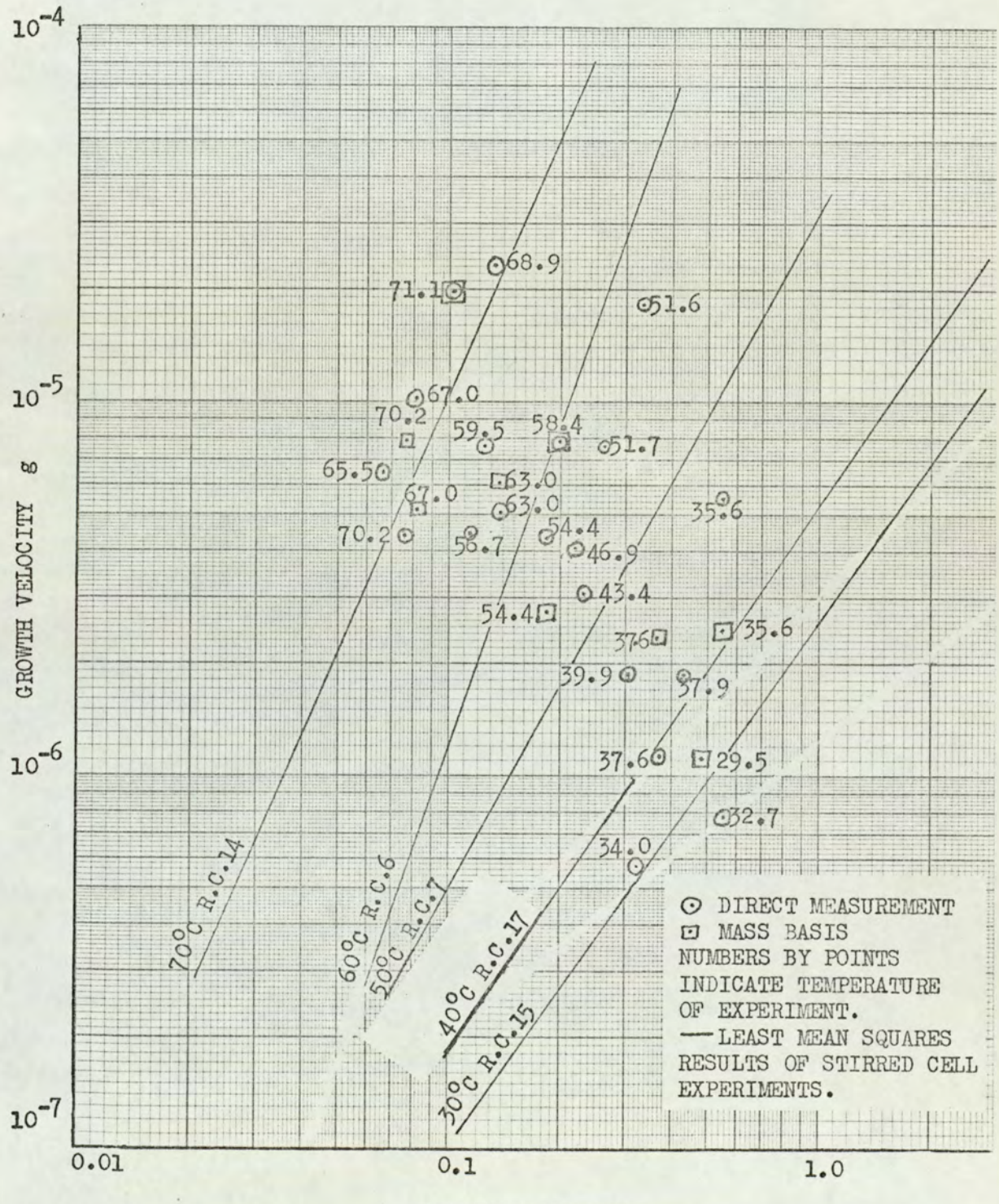
7. 1. 1. Preliminary Experiments.

Runs F.C.1 to F.C.6 were unsuccessful as too small a seed size was used, presenting difficulties of agglomeration and transport of small crystals into the feed vessel. From Runs F.C.7 about 4g. of seed crystals (-120+150 mesh) were added as a slurry in a small amount of cold saturated solution. After a suitable time the product crystals were discharged and examined. The crystal size, L, measured before and after growth was the average of 10 crystals measured across the pyramid base and gave a standard deviation from the average of about 7μ. As the vernier microscope only read to 10μ this was considered reasonable. The size L was converted to the equivalent spherical volume diameter, D, using a $\phi L^3 = D^3$ and so $D = 0{\cdot}9655L$ and the growth velocity, $g = \frac{dr}{dt}$, was calculated.

With the exception of F.C. 1 3. it was not possible to remove all the bed for weighing until F.C.19. when the flexible seal and knee joint was fitted to the downcomer. With this modification it was possible to give the bed a stir periodically, to prevent agglomeration building up and permit the ready discharge of the crystal product. As the average seed size was known, the growth velocity could then be derived by calculating the product size from the increase of crystal mass assuming

FIGURE 7.1.

RESULTS OF PRELIMINARY FLUIDISED BED EXPERIMENTS



spherical crystals.

The results for these preliminary fluidised bed experiments, with the growth velocity, g, derived both from the direct measurement and using the mass increase basis are shown, Table 31 , Appendix D . As the growth rate is later shown (Section 7.2) to deviate from first order with respect to supersaturation, the results are shown in figure 7. 1 with growth velocities, g, plotted against supersaturation, s, and compared with the least mean squares correlation found for Batch C material in the seeded cell experiments stirred at 2000 r.p.m. The numbers beside points indicate the temperatures of the fluidised bed experiments.

In comparison with the stirred cell experiments, results on average give growth velocities as would be expected from experiments about 7° C higher than that indicated. This could be because of an unconscious tendency towards selective sampling of good large crystals for product measurements. In general the results calculated on a mass basis are nearer to that expected from the stirred cell experiments, assuming no diffusional effect. Although these results, particularly those measured directly, are of limited accuracy they indicate a faster growth rate than with the stirred cells. This is not feasible even if the system was diffusion controlled and it appears that the surface integration step is rate controlling even for the low relative solution/crystal velocities and high temperatures of the fluidised bed.

7. 1. 2. Results with Modified Fluidised Bed Apparatus.

Assuming surface integration rate control the computer program 1 using McCabe's ΔL Law (Section 6.3.2.2.) was used to calculate the

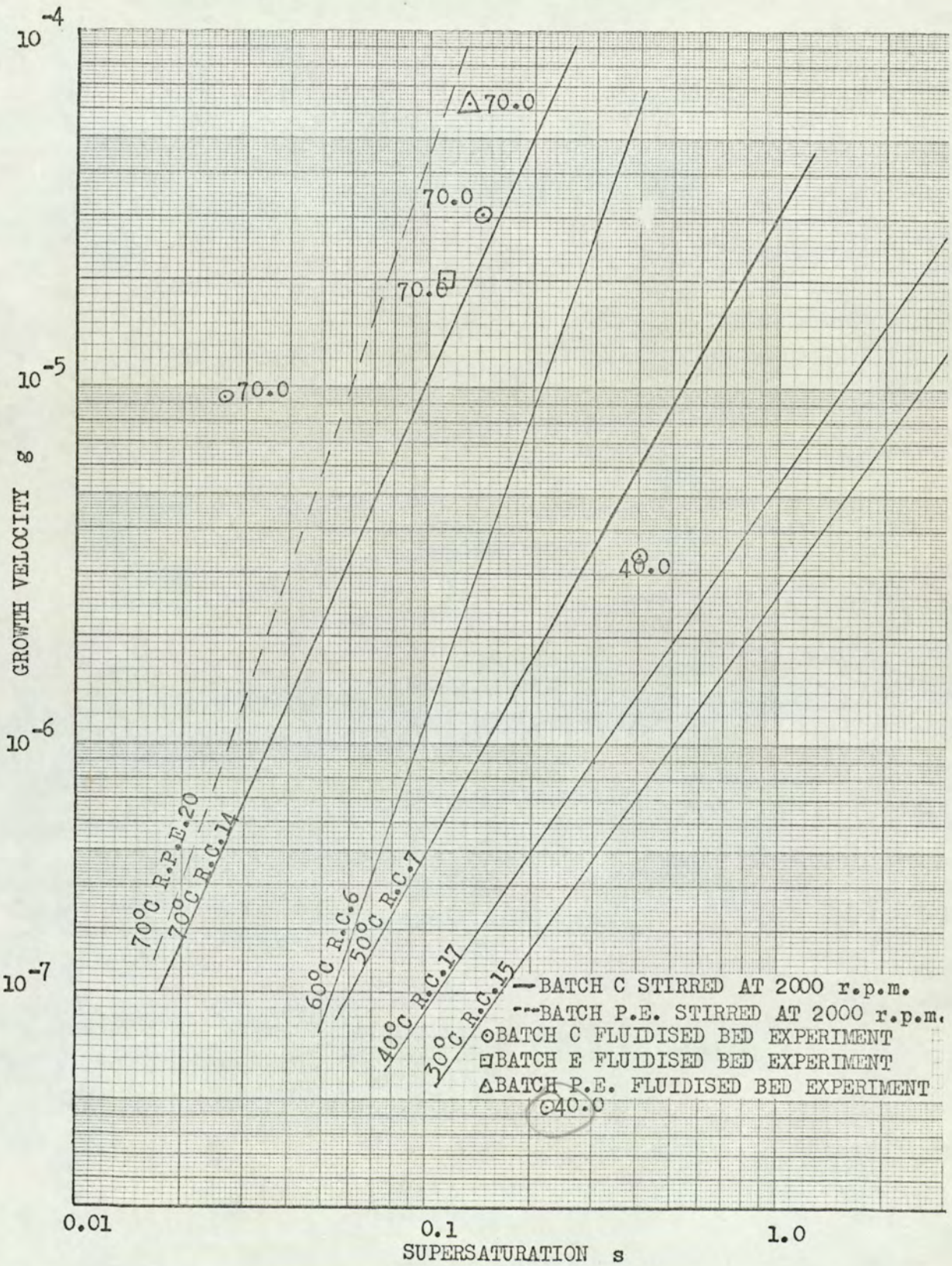
results of the experiments with the modified fluidised bed apparatus shown in section 4.2.3. An example experimental data sheet is shown for run R.E.1 in Table 32 , Appendix D . The size analyses of the seed batches used are shown in Table 25 . Appendix B . Although the mean srystal area $\bar{A}$ has been calculated Km, the rate constant, based on a first order growth rate is not shown as the stirred cell experiments showed a first order assumption to be invalid. The following results were obtained:

RUN No.	T_o °C	SEED	SEED MASS(g)	PROD. MASS(g)	TIME MINS	s	ΔD μ	$\bar{A}_2$ cm^2	g cm/min.
FC28	70.0	124-150μC	8.0	11.26	90	0.026	16.71	3308	9.283×10^{-6}
FC29	70.0	124-150μC	8.0	14.55	50	0.141	30.48	3662	3.048×10^{-5}
FC30	40.0	124-150μC	8.0	8.03	90	0.224	0.17	2926	9.636×10^{-8}
FC31	40.0	124-150μC	8.0	8.60	50	0.406	3.39	2997	3.388×10^{-6}
FE.1	70.0	124-150μE	7.96	14.04	60	0.110	24.21	4284	2.018×10^{-5}
FPE1	70.0	124-150μPE.	7.91	17.63	35	0.129	43.43	3841	6.204×10^{-5}

These results have been plotted as g vs s in figure 7.2. and compared with the least mean squares correlations obtained from the stirred cell experiments. As the crystal seed dispenser was not reweighed after use with runs F.C.28 to F.C.31 these results are not as accurate as they could otherwise have been. F.C.30 and F.C.31 at 40°C are particularly susceptible to this error as the crystal mass increase was so small.

FIGURE 7.2.

RESULTS OF MODIFIED FLUIDISED BED EXPERIMENTS



F.C.28 shows a growth rate very much higher than expected. However, for runs F.C. 29, F.E.1 and F.P.E.1, where the crystal mass was about doubled throughout an experiment, all the growth rate velocities were very much the same as the stirred cells with the same materials at 70°C. As this was the highest temperature practical with this apparatus no further experiments were attempted. This indicated surface integration control under all the experimental conditions up to 70°C for Batches C, E and purified E.

TABLE: 33

PREVIOUS WORK(1) - APPENDIX E

SEEDED CELL A EXPERIMENTS AT 500 R.P.M.

RUN No.	RUN No.	SOLUTION		SEED	
PREVIOUS WORK(1)	PRESENT TEXT	TEMP °C	CONC. %m/v	SIEVE CUT μ	BATCH
R.14	R.A.1	60.0	24.0	44 - 53	A*
R.20	R.A.2	70.0	30.0	44 - 53	A*
R.21	R.A.3	55.0	21.5	44 - 53	A*
R.23	R.A.4	80.0	37.0	44 - 53	A*
R.27	R.P.A.1	60.0	24.0	44 - 53	P.A*
R.29	R.P.A.2	70.0	29.0	44 - 53	P.A*
R.30	R.P.A.3	80.0	36.0	44 - 53	P.A*
R.31	R.C.1	60.0	24.0	44 - 53	C*
R.32	R.P.A.4	50.0	18.5	44 - 53	P.A*
R.33	R.A.5	50.0	19.5	44 - 53	A*
R.34	R.P.A.5	80.0	36.0	44 - 53	A*

TABLE: 34

CHRONOLOGICAL SUMMARY OF EXPERIMENTS IN SEEDED CELLS - APPENDIX D

ALL EXPERIMENTS AT 2000 R.P.M. UNLESS OTHERWISE STATED

EXPT. No.	RUN No.	SOLUTION		SEED		CELL	COMMENT	TABLE No.
		T_o °C	c%m/v	SIEVE CUT (μ)	BATCH			
1	R.B.1	60.0	24.0	44 - 64	B*	A	NO GROWTH 4½h.	-
2	R.B.2	60.0	24.0	44 - 64	B*	C	NO GROWTH 4½h.	
3	R.B.3	60.0	24.3	-	B	C		36
4	R.B.4	60.0	25.3	-	A	C		
5	R.A.6	60.0	24.0	-	A	C		
6	R.B.5	60.0	24.0	-	B	C		

* SPECIALLY PREPARED SEED

TABLE: 34 (CONT.)

EXPT. No.	RUN No.	SOLUTION T_o °C	SOLUTION o%m/v	SEED SIEVE CUT(μ)	SEED BATCH	CELL	COMMENT	TABLE No.
7	R.C.2	60.0	24.0	-	C	C		36
8	R.C.3	60.0	24.0	75 - 89	C*	C		38
9	R.C.4	60.0	24.0	75 - 89	C*	C	NONIDET ADDN.	39
10	R.C.5	60.0	24.0	44 - 64	C*	A	POOR IMAGE	40
11	R.C.6	60.0	24.0	44 - 64	C*	C		41
12	R.C.7	50.0	19.5	44 - 64	C*	C		42
13	R.C.8	40.0	17.5	44 - 64	C*	C		43
14	R.C.9	60.0	24.0	44 - 64	C*	A	500 R.P.M.	44
15	R.C.10	60.0	24.0	44 - 64	C*	C	500 R.P.M.	45
16	R.C.11	40.0	14.0	44 - 64	C*	C	500 R.P.M.	46
17	R.C.12	70.0	30.0	44 - 64	C*	A	500 R.P.M.	47
18	R.C.13	70.0	30.0	44 - 64	C*	C	500 R.P.M.	48
19	R.C.14	70.0	30.0	44 - 64	C*	C	2000 R.P.M.	49
20	R.C.15	30.0	12.5	44 - 64	C*	C	2000 R.P.M.	50
21	R.C.16	70.0	30.0	44 - 64	C*	C	1000 R.P.M.	51
22	R.C.17	40.0	1525	44 - 64	C*	C	2000 R.P.M.	52
23	R.D.1	70.0	30.0	44 - 64	D*	C		53
24	R.D.2	70.0	30.0	44 - 64	D*	A		54
25	R.D.3	70.0	30.0	44 - 64	D*	A		55
26	R.D.4	70.0	30.0	75 - 89	C*	S		56
R7	R.D.5	70.0	30.0	75 - 89	D*	S		57
28	R.D.6	70.0	30.0	75 - 89	C*	C		58

* SPECIALLY PREPARED SEED

TABLE: 34 (CONT.)

EXPT. No.	RUN No.	SOLUTION		SEED		CELL	COMMENT	TABLE No.
		$T^{\circ\circ}_{o}C$	c%m/v	SIEVE CUT(μ)	BATCH			
29	R.D.7	70.0	30.0	64 - 75	P.G.*	S		59
30	R.P.G.1	70.0	30.0	64 - 75	P.G.*	C	NO GROWTH 5h.	
31	R.P.G.2	70.0	30.0	64 - 75	P.G.*	S	NO GROWTH 5h.	
32	R.P.G.3	70.0	30.0	89 - 105	P.G.	S		60
33	R.E.1	70.0	30.0	89 - 105	E*	C		61
34	R.E.2	70.0	27.5	89 - 105	E*	C		62
35	R.E.3	60.0	27.5	89 - 105	E*	C		63
36	R.E.4	60.0	24.5	89 - 105	E*	C		64
37	R.E.5	60.0	26.0	89 - 105	E*	C		65
38	R.E.6	50.0	24.0	89 - 105	E*	C		66
39	R.E.7	70.0	30.0	89 - 105	E*	S		67
40	R.P.G.4	70.0	30.0	89 - 105	P.G.*	C		68
41	R.G.1	70.0	30.0	-	G	C	QUALITATIVE	37
42	R.G.2	70.0	30.0	-	G	S	QUALITATIVE	37
43	R.P.G.5	70.0	30.0	89 - 105	P.G.*	C		69
44	R.P.G.6	70.0	30.0	89 - 105	P.G.*	S		70
45	R.P.D.1	70.0	30.0	89 - 105	P.G.*	C		71
46	R.F.1	70.0	30.0	75 - 89	F*	C	NO GROWTH $\frac{3}{4}$h.	
47	R.F.2	70.0	30.0	89 - 105	F	C	QUALITATIVE	37
48	R.E.8	70.0	30.0	89 - 105	E*	C		72
49	R.F.3	70.0	30.0	89 - 105	F*	C		73

* SPECIALLY PREPARED SEED

TABLE: 34 (CONT.)

EXPT. No.	RUN No.	SOLUTION		SEED		CELL	COMMENT	TABLE No.
		T_o °C	c%m/v	SIEVE CUT(μ.)	BATCH			
50	R.F.4	70.0	30.0	89 - 105	F*	C		74
51	R.F.5	70.0	30.0	89 - 105	F	C	QUALITATIVE	37
52	R.E.9	70.0	30.0	89 - 105	E*	C		75
53	R.P.G.7	70.0	30.0	89 - 105	P.G.*	C		76
54	R.G.3	70.0	30.0	89 - 105	P.G.*	C		77
55	R.G.4	70.0	30.0	89 - 105	E*	C		78
56	R.P.E.1	70.0	30.0	89 - 105	P.G.*	C		79
57	R.P.G.T.1	70.0	30.0	89 - 105	P.G.*	C		80
58	R.P.E.2	70.0	30.0	89 - 105	P.G.*	C		81
59	R.P.E.3	70.0	27.5	89 - 105	P.G.*	C		82
60	R.P.G.8	70.0	27.5	89 - 105	P.G.*	C	NO GROWTH 5h.	
61	R.P.E.4	70.0	27.5	89 - 105	F*	C	cf. 59 seed	83
62	R.P.G.K.1	70.0	27.5	89 - 105	E*	S	CELL SCOURED	84
63	R.P.G.B.1	70.0	27.5	89 - 105	E*	S		85
64	R.P.D.2	70.0	27.5	89 - 105	E*	C		86
65	R.P.G.M.1	70.0	27.5	89 - 105	E*	S		87
66	R.P.E.5	70.0	27.5	89 - 105	E*	C		88
67	R.P.E.6	70.0	27.5	89 - 105	E*	S		89
68	R.P.E.7	70.0	27.5	89 - 105	E*	C		90
69	R.P.E.8	70.0	27.5	89 - 105	E*	S		91
70	R.P.D.3	70.0	27.5	89 - 105	E*	S		92
71	R.E.10	70.0	30.0	89 - 105	E*	C		93

* SPECIALLY PREPARED SEED

TABLE: 34 (CONT.)

EXPT. No.	RUN No.	SOLUTION		SEED		CELL	COMMENT	TABLE No.
		T_o °C	c%m/v	SIEVE CUT(μ)	BATCH			
72	R.E.11	70.0	30.0	89 - 105	E*	S		94
73	R.P.E.M.1	70.0	27.5	89 - 105	E*	S		95
74	R.P.D.4	70.0	27.5	89 - 105	E*	A		96
75	R.P.D.5	70.0	27.5	89 - 105	E*	C		97
76	R.P.D.6	70.0	27.5	89 - 105	E*	S		98
77	R.E.12	70.0	30.0	89 - 105	E*	S		99
78	R.E.13	70.0	30.0	89 - 105	E*	C		100
79	R.E.14	70.0	30.0	89 - 105	E*	C	CELL CLEANED	101
80	R.E.15	70.0	30.0	89 - 105	E*	C	CELL POLISHED	102
81	R.E.16	70.0	30.0	89 - 105	E*	C	0.5g Zn ADDN.	103
82	R.E.17	70.0	30.0	89 - 105	E*	C	0.5g Cu ADDN.	104
83	R.E.18	70.0	30.0	89 - 105	E*	-	QUALITATIVE	
84	R.E.19	70.0	30.0	89 - 105	E*	C	CELL POLISHED	105
85	R.E.20	70.0	30.0	89 - 105	E*	C	2500 R.P.M.	106
86	R.E.21	70.0	30.0	89 - 105	E*	C		107
87	R.E.22	70.0	30.0	89 - 105	E*	G	DISSOLUTION	
88	R.E.23	70.0	30.0	89 - 105	E*	G		108
89	R.E.24	70.0	30.0	89 - 105	E*	G		109
90	R.E.25	70.0	30.0	89 - 105	E*	C		110
91	R.E.26	70.0	30.0	89 - 105	E*	C		111
92	R.E.27	70.0	33.5	89 - 105	E*	C		112
93	R.E.28	60.0	27.5	89 - 105	E*	G		113

* SPECIALLY PREPARED SEED

TABLE: 34 (CONT.)

EXPT. No.	RUN No.	SOLUTION T_o °C	SOLUTION o/m/v	SEED SIEVE CUT(μ)	SEED BATCH	CELL	COMMENT	TABLE No.
94	R.E.29	60.0	27.5	89 - 105	E*	C		114
95	R.E.30	60.0	27.5	89 - 105	P.E.*	C	cf. 94 seed	115
96	R.P.G.M.2	70.0	27.5	89 - 105	P.E.*	C	NO GROWTH 24h.	
97	R.P.G.M.3	70.0	27.5	89 - 105	P.E.*	C	Zn+Cu ADDN. NO GROWTH 6h.	
98	R.P.E.9	70.0	27.5	89 - 105	P.E.*	C		116
99	R.P.G.9	70.0	27.5	89 - 105	P.E.*	C	NONIDET ADDN. NO GROWTH	
100	R.P.G.10	70.0	27.5	89 - 105	P.E.*	C	0.5g BRASS ADDN. NO GROWTH 2½h.	
101	R.P.G.11	70.0	27.5	89 - 105	P.E.*	G	HCl ADDN. NO GROWTH	
102	R.E.31	70.0	30.0	89 - 105	E*	C		117
103	R.P.E.10	30.0	12.5	89 - 105	P.E.*	C	NUCLEATION?	118
104	R.E.32	60.0	27.5	89 - 105	E*	C	1.0g SEED	119
105	R.E.33	60.0	27.5	89 - 105	E*	C	0.5g SEED	120
106	R.E.34	60.0	26.0	89 - 105	E*	C	1.0g SEED	121
107	R.E.35	60.0	26.0	89 - 105	E*	C	0.5g SEED	122
108	R.E.36	60.0	24.5	89 - 105	E*	C	1.0g SEED	123
109	R.E.37	60.0	24.5	89 - 105	E*	C	0.5g SEED	124
110	R.C.F.1	60.0	27.5	89 - 105	E*	C		125
111	R.C.18	60.0	27.5	89 - 105	E*	C		126
112	R.P.E.11	60.0	24.0	89 - 105	P.E.*	C	NUCLEATION?	127
113	R.P.E.12	60.0	22.0	89 - 105	P.E.*	C		128
114	R.P.E.13	60.0	20.0	89 - 105	P.E.*	C	NO GROWTH 8½h	

* SPECIALLY PREPARED SEED

TABLE: 34 (CONT.)

EXPT. No.	RUN No.	SOLUTION T_o °C	SOLUTION c%m/v	SEED SIEVE CUT(μ)	SEED BATCH	CELL	COMMENT	TABLE No.
115	R.P.E.14	60.0	22.0	89 - 105	P.E.*	C	1.0g SEED	129
116	R.P.E.15	60.0	20.0	89 - 105	P.E.*	C	5.0g SEED NO GROWTH 23h.	
117	R.P.D.M.1	60.0	20.0	89 - 105	P.E.*	C	NO GROWTH 5h.	
118	R.P.E.16	50.0	17.75	89 - 105	P.E.*	C		130
119	R.P.E.17	40.0	13.75	89 - 105	P.E.*	C		131
120	R.P.E.18	75.0	31.5	89 - 105	P.E.*	C		132
121	R.P.E.19	40.0	13.75	89 - 105	P.E.*	C		133
122	R.P.E.20	70.0	28.5	89 - 105	P.E.*	C		134
123	R.P.E.F.1	70.0	28.5	89 - 105	P.E.*	C	NO GROWTH 7h.	
124	R.E.38	50.0	19.5	89 - 105	E*	C		135
125	R.P.E.F.2	70.0	28.5	89 - 105	P.E.*	C		136
126	R.E.39	60.0	27.5	75 - 89	D*	C		137
127	R.A.7	70.0	30.0	89 - 105	P.E.*	C		138
128	R.E.F.1	70.0	30.0	75 - 89	D*	C		139
129	R.E.F.2	60.0	27.5	89 - 105	E*	C		140
130	R.E.F.3	40.0	15.25	75 - 89	D*	C	NO GROWTH 24h.	
131	R.E.F.4	60.0	24.5	39 - 105	E*	C		141
132	R.E.F.5	50.0	19.5	75 - 89	D*	C		142
133	R.P.E.F.3	40.0	13.75	89 - 105	P.E.*	C	NO GROWTH 22h.	
134	R.P.E.F.4	70.0	27.5	89 - 105	P.E.*	C		143

* SPECIALLY PREPARED SEED

TABLE: 34 (CONT.)

EXPT. No.	RUN No.	SOLUTION		SEED		CELL	COMMENT	TABLE No.
		T_o °C	o%m/v	SIEVE CUT(μ)	BATCH			
135	R.P.E.F.5	60.0	22.0	89 - 105	P.G.*	C		144
136	R.P.E.F.6	70.0	27.5	89 - 105	P.E.*	C		145
137	R.P.E.D.1	70.0	28.5	89 - 105	P.E.*	C		146
138	R.P.E.D.2	50.0	17.75	89 - 105	P.G.*	C		147
139	R.E.40	70.0	30.0	89 - 105	E*	C	0.5gBRASS FILINGS ADDN.	148

* SPECIALLY PREPARED SEED

7. 2. Seeded Solutions in stirred cells

7. 2. 1. Experimental Data.

An example experimental data sheet for a stirred cell run is shown in Table 35 , Appendix D , for R.P.E.16. The size analyses of the specially prepared seeds of the various materials and sieve fraction, after attrition in a saturated solution under the experimental conditions as done by the Coulter Counter size analyser are shown in Table 24 , Appendix B . The computer data being presented as the number percentage of a particular mean diameter, D, cm. The refractometer readings are presented as scale S as would be obtained in Cell C. Scale readings obtained in other cells were converted to scale S, which was the calibration scale, by means of the Table 4 in Appendix A . The computer data for this refractometer scale array included the equilibrium refractometer scale as the final reading as found for the particular equivalent P.E. concentration in the equilibrium experiments, Appendix A . The equivalent time readings for the refractometer scales were recorded in minutes. The reading of the 'apparent equilibrium' value after which no further concentration decrease was observed was rejected for computation purposes as the time to reach this value was indeterminate.

The calibration coefficients for the concentration based on the refractometer scale, S, i.e. B,F and G where $c = F + B.S. + G.S^2$ were recorded for the particular solution material and temperature involved (from Tables 2 and 3 , Appendix A). The initial seed mass, H_g was also recorded, and although the seed dispenser was not reweighed

after use until Experiment 25, the error involved should be very small. Finally the evaporation rate in cm^3/min. for the particular cell and temperature used was recorded from Table 29 Appendix C . As Cells A and G were only used for growth rate comparisons, and the evaporation rate only becomes effective as c approaches c_∞ , evaporation losses for these cells are assumed zero.

All temperatures are recorded as the observed temperatures T_o° C and can be converted to actual temperatures by means of Figure 5.1., Appendix A . This has been done in Section 8 for the purpose of the Activation Energy calculations.

7. 2. 2. Pentaerythritol materials.

The chemical analyses of the materials used are shown in Table 1 , Appendix A .

7. 2. 2. 1. Batch A. (<0·1% Di - P.E and 4.73% Formal)

This was the material used in the previous crystallisation work (1). The growth rate constants K were calculated then assuming a first order growth rate with respect to supersaturation and using the approximation $\frac{A_2}{A_1} = \left(\frac{M_2}{M_1}\right)^{\frac{2}{3}}$ for the area calculations for each interval of growth. A summary of the results is shown in Table 149 , Appendix E . This work has now been analysed using the more accurate method which allows for a crystal size distribution, (program 2), and with the refractometer readings converted to refractometer scale S for use with the more accurate calibrations. The runs at 80°C could not be analysed by this method as the calibrations in this work were not carried out at this temperature. The effects of the Coulter Counter size correction and

the new equilibrium values are also shown (Appendix E).

7. 2. 2. 2. Batch B (<0·1% Di - P.E., 4·98% Formal)

In order to compare this material with the essentially similar Batch A material of the previo us work (1), Experiment 1 was carried out in the old Cell A with the more accurate refractometer. This unexpectedly showed no growth for the 4½ hours of the test. At first this was thought to be due to an impurity incorporated in the cell, but when the experiment was repeated (R.B.2) in the new Cell C there was again no growth for 4½ hours. The most probable cause was then thought to be an impurity picked up in the seed preparation or sieving process. Tests R.B.3 and 4 were carried out with 2g of unclassified Batches B and A seed respectively and indicated growth rates similar to each other but very slow in comparison with the previo us work (1). A check on this Batch A material containing <0·1% Di-P.E and 4.73% Formal, (R.A.6) with 2g unclassified Batch A seed indicated a growth rate very much faster than R.B.3 or 4. To check that this was not because the impurity had been washed out of the cell, R.B. 5 was carried out, a repeat of R.B.3, with a similar result. To check that this very slow growth rate could not be a Di-P.E. effect not shown in the chemical analysis R.C.2. was carried out with 2g unclassified Batch C seed (containing 1·0% Di-P.E. and 4·3% Formal). This gave a growth rate even faster than R.A.6. This indicated an unknown additional impurity present in Batch B far more effective in slowing growth than either Di-P.E. or Formal, and no further tests were carried out with this material.

7. 2. 2. 3. Batch C (1·0% Di-P.E., 4·3% Formal)

Experimental work was started on Batch C material for comparison with Batch A of the previous work (1) to find the effect of the Di-P.E. impurity. Using R.C.3. as a control an incidental test (R.C.4.) was carried out with the addition of 5 drops of a non-ionic detergent dispersing agent NONIDET P.40. As there was no apparent effect on the growth rate this additive was then tried, without success, in nucleation tests to attempt to restrict the product size distribution, thought to be due to agglomeration.

It was realised at this stage that the stirrer speed was set at 2000 r.p.m. for experiments 1 to 9, due to an error in the stroboscope reading, whereas the previous work (1) had been carried out at 500 r.p.m. This was checked hereafter with a portable tachometer. R.C.5 was carried out at 2000 r.p.m. in Cell A for comparison with the previous work; the image however was poorly defined and R.C.6 in Cell C was found more accurate.

To find the temperature effect R.C.7 and R.C.8 were carried out at 50°C and 40°C in Cell C with the well-defined image, at 2000 r.p.m. R.C.8. shows a very high growth rate compared with R.C.17 (which had a lower initial supersaturation). This phenomenon appeared completely anomalous until the detailed study on Batch E, (Section 7.2.2.5) was performed.

Tests R.C.9 and R.C.10 were carried out at 500 r.p.m. in Cells A and C respectively for speed and cell comparison with R.C.5 and R.C.6. The image of R.C.9 (CellA) was again poorly defined, but R.C.10 indicated a

slower growth rate in the latter stages. Test R.C. 11. at 40°C was carried out at a low initial concentration and the growth was very slow. Test R.C.12 in Cell A at 70°C showed a better refractometer image and indicated a growth rate similar to R.C.13 in Cell C. from This it was concluded that cells A and C gave essentially the same results.

The size analyses of B atch C seed material attrited under the different hydrodynamic conditions of Cells A and C both stirred at 500 r.p.m. and 2000 r.p.m. showed very similar degrees of attrition (Table 23 , Appendix B). On this evidence R.C.16 at 1000 r.p.m. was calculated using the size analysis for 2000 r.p.m. which would be within the limits of experimental error. Experiments 18, 21 and 19 at 500, 1000 and 2000 r.p.m. respectively showed the effect of stirrer speed to be that of maintaining the larger crystals in suspension. (as described in Section 7. 2. 4) and 2000 r.p.m. was considered necessary to achieve this.

All further experiments were done at 2000 r.p.m. and experiments 11, 12, 19. 20 and 22 show the effect of temperature on the crystal growth of Batch C maintained at this stirrer speed in Cell C.

7. 2. 2. 4. Batch D (1·0% Di-P.E., 5·5% Formal)

Experiments 23 to 29 were carried out with 30% m/v Batch D material, (which contained 28% more Formal than Batch C), at 70°C to compare the cells at 2000 r.p.m. and to find the effect of seed size and composition Two dm^3 batches of 22% m/v C, D and P.G. solutions were cooled and nucleated at 40°c. The solutions were stirred for 2 hours and sieve fractions of the resulting crystals, after filtering with an acetone

wash and drying, used as seed. A comparison of seed Batches C and D with approximately 4·3% and 5·5% Formal respectively should indicate the sensitivity of the growth rates to the impurity in the seed material or possibly preparation nucleation temperature, if the impurity partition coefficient varies significantly with temperature.

In order to compare cells, test R.D.2 in Cell A was carried out but found difficult to follow because of the poorly defined image. However, R.D.3 (a repeat of R.D.2) showed an overall growth very similar to R.D.1 in Cell C, although individual readings were more erratic. Test R.D.4. in Cell S also showed a very similar growth rate to R.D.1. indicating that cell type is not critical in determining the crystal growth rate.

R.D.5. in Cell S with 75 - 89μ Batch D seed showed a faster growth rate overall than R.D.4. Because of the good comparison between R.D.4. and R.D.1., this was attributed to an incorrect size analysis of the attrited seed. Size analyses of the attrited seed for the computation of results with Cell S were taken as those found for Cell C as both cells had similar internal dimensions. This was because sampling of attrited seed suspensions was very difficult due to the Cell S construction. The high growth rate of R.D.5 could be due to additional attrition of this seed in Cell S, possibly because of a different stirrer position.

Runs R.D.1. and R.D.6. with 44 - 64μ Batch D seed and 75 - 89μ Batch C seed respectively, in Cell C showed a very good agreement, indicating that small differences with impure seed composition are not

critical to crystal growth rates. Therefore nucleation temperatures with impure seed preparation need not be noted. Test R.D.7. with 64 - 75μ P.G. seed however showed a faster growth rate than R.D.4. particularly in the early stages. This could be due to an impurity diffusion into the seed to achieve an equilibrium partition with the mother liquor, enchancing the rate of growth.

A comparison of Runs R.C.14. and R D.1. shows a favourable agreement between Batches C and D, with Batch D showing a slower growth rate as would be expected from the chemical analysis.

7. 2. 2. 5. 1. Batch E. (0.9% Di - P.E. 5.2% Formal)

A considerable amount of Batch E seed was prepared from Batch E mother liquor in a number of batch crystallisations by nucleating 22% m/v solutions. The collected sieve fraction 89 - 105μ was then size analysed, after attrition at 2000 r.p.m. in Cell C, for a more detailed study with Batch E mother liquor.

R.E.1. at 70°C, Experiment 33, showed a very similar growth rate to R.C.14. but rather faster than R.D.1. This indicated complete suspension of the rather larger 89 - 105μ sieve fraction crystals. The suspension characteristics were studied in more detail in later experiments (Section 7.24).

Using R.E.1. (30%) as control R.E.2 with 27.5% initial concentration was carried out to find the effect of starting the growth experiment at a different initial concentration. Although it indicated a slower growth than R.E.1. for equivalent supersaturations, the number of readings obtainable were too limited to obtain an accurate analysis.

R.E. 3, 4 and 5 were therefore carried out at 60°C to study this effect further. Equivalent supersaturations for these runs should give the same growth rate regardless of the initial supersaturation. The fact that these results do not concur must be due to one of the following reasons:-

(a) The normal growth velocity of a crystal is dependent on the crystal size (i.e. contrary to McCabe's ΔL law)

(b) The initial seed size distribution is incorrect.

(c) The increase in crystal area throughout a run is not simply due to the increase in crystal size due to growth.

Of these possibilities (c) seemed to be the most feasible. It was thought possible that a surface "roughness" effect might exist which would be proportional to the mass of solute deposited per unit crystal seed area, possibly caused by growth of numerous screw dislocations. This was therefore studied further in Experiments 104 to 109 with different initial seed masses at these different initial supersaturations in an attempt to obtain the quantitative effect. The runs at first seemed to further substantiate the "roughness" hypothesis, with smaller initial seed masses undergoing faster normal growth velocities under equivalent conditions. However on examination of the crystal products (Table 28 , Appendix B) for these runs it could be seen that in fact the increase in surface area throughout these runs, in addition to that calculated, was due to attrition occuring during growth. Although a quantitative attrition effect is not possible from the limited data obtained, it appears that there exists a critical time (i.e. diameter

increase) for each growth velocity (which is dependent on supersaturation) which if exceeded will cause excessive brittleness of the growing crystal resulting in attrition under certain hydrodynamic conditions. Examination of R.E.15. product using 2g of seed grown in 30% m/v initial concentration solution at 70°C, where the high temperature will produce the fastest growth velocity and weakest crystals, showed a product size almost as predicted (Table 26 , Appendix B) with some apparent agglomeration but no sign of any attrition. It was therefore considered safe to work with solution supersaturations up to about $\Delta c = 4\%$ with 2g of seed.

Batch E was also used to establish the effect of heterogeneous particles on the growth velocity (Section 7.2.3.) and to study the effect of the stirrer position on the suspension characteristics of a run (Section 7.2.4.). Having established that absolute cleanliness of a cell is essential, and found the optimum stirrer position, R.E.31, Experiment 102, was carried out in this position showing a good comparison with R.E.19. and ascertaining an efficient cleaning of Cell C after the previous particle additions.

R.E. 6 at 50°C was carried out at too high an initial concentration while establishing the attrition effect, resulting in erroneous high results. However tests R.E.19, R.E.4 and R.E.38 at 70°C, 60°C and 50°C respectively, compare favourably, although with somewhat faster growth rates, with R.C.14, R.C.6 and R.C.7 at the same temperatures. As it was possible that each batch of material might have contained some small amount of contaminant as was found in Batches B, F and G, this suggested that Batch E was the batch which contains the least.

7. 2. 2. 5. 2. Filtered Batch E

As an enhanced growth rate was found with the presence of heterogeneous particles (Section 7.2.3), the effect of filtering production material through a fine filter was studied. A 10% m/v solution of the material was prepared, filtered through a 0.45μ membrane filter, and slowly evaporated to dryness. The filtered material amounted to 0.030 mass% of the P.E. As the stability of Formal in solution at high temperatures has been demonstrated in the previous work (1) the impurity content should remain constant. Experiments 110 and 111 were carried out on Batch C material to find the effect of filtration. However as the products of Experiments 104 to 109 had not been size analysed at this time it was anticipated that the effect would be more pronounced with the higher initial concentration of 27.5% m/v at 60°C. Although this effect is masked therefore by attrition occuring during growth, it was surprising to note that R.C.F.1 with the filtered material showed a faster growth than R.C.18 . This was contrary to expectations, having established (Section 7.2.3) that heterogeneous particles enhance crystal growth.

It was considered that a study of filtered Batch E material would give more relevent growth rate and activation energy data for the impure material than the normal production material with an unknown quantity of heterogeneous particles. Experiments 128 to 132 were therefore carried out with the filtered material. Test R.E.F. 2 with 27.5% initial concentration is again irrelevent as it was carried out for a comparison with R.E.F.4. to find the proposed"roughness" effect of using different initial supersaturations and simply illustrates the amount of attrition

which occured during growth. However tests R.E.F.1. and R.E.F.4 showed growth rates ca. 75% faster than R.E.19 and R.E.4 (at 70°C and 60°C respectively) whereas R.E.F.5 at 50°C showed a growth rate essentially the same as R.E.38 at the same temperature. Product examination of R.E.F.4.(Table 27 , Appendix B) showed this to be due to attrition occuring during the growth process and it is to be assumed therefore that heterogeneous particles are beneficial in establishing a less brittle crystal when grown at higher growth rates (i.e. in this case at higher temperatures). Test R.E.F.3 (at 40°C) exhibited no growth for the 24 hours of the experiment and this was at first thought to be completely anomalous. However test R.P.E.F.3 at 40°C, Experiment 133, also showed complete growth inhibition in contrast to R.P.E.19 under the same conditions. This appears a real effect at this temperature, therefore, and it is possible that heterogeneous particles are necessary for growth at a lower temperature, possibly to produce the required dislocations on the crystal surface.

7. 2. 2. 5. 3. Purified Batch E.

Tests R.P.E.1. to R.P.E.9 at 70°C were carried out for a comparison with purified Batch G material, and also to establish the enhancing effect of heterogeneous particles (Section 7.2.3.) Test R.P.E.10. (at 30°C) showed a faster growth rate than R.P.E.9 (at 70°C) which must have been due to nucleation induced at the start of the run (although the initial Δc was only about 4.3%). Test R.P.E.11 at 60°C with an initial supersaturation at $\Delta c = 5.4\%$ indicated a growth velocity more than an order of magnitude faster than R.P.E.12 at the same temperature with an

initial supersaturation of $\Delta c=3.2\%$ which again must have been due to nucleation. Test R.P.E. 13 at 60°C however with an initial supersaturation of only $\Delta c=1.4\%$ showed complete growth inhibition for the $8\frac{1}{2}$ hours of the test. There was therefore a very limited metastable region within which this system could be studied, and a comparison with different initial concentrations to observe the 'brittleness' effect of the Pure P.E. crystals was impractical. Test R.P.E.14 however was done with 1g of seed with $\Delta c=3.2\%$ and resulted in a growth rate slower by about half than R.P.E.12. This would either indicate agglomeration during growth or an impurity in the solution being absorbed on the crystal surface more slowly with the smaller amount of seed. This latter explanation would also account for the growth inhibition of test R.P.E.13., since growth at this low supersaturation was observed (tests R.P.E.12. and R.P.E.14.) once started at a higher supersaturation.

A seed mass of 5 g was therefore used for R.P.E.15, which would have approximately the same crystal surface area as R.P.E.12 when the supersaturation had fallen to $\Delta c=1.4\%$. However growth inhibition persisted. The possibility of contaminant could not be overuled as R.P.E.12 would have a depleted contaminant concentration in solution, assuming the contaminant built into the crystal lattice, after the growth to a solution supersaturation of $\Delta c=1.4\%$.

A comparison of tests R.P.E.7 and R.P.D.5 at 70°C Experiments 68 and 75, show a similar growth rate but with R.P.D.5. apparently about 20% slower until about $\Delta c=1.2\%$ when approximately 4g of solute had been deposited. This could be due to Batch D being slightly contaminated,

which would also account for the slow growth of R.D.1. compared with R.E.19.

An extraction of purified Batch D was therefore carried out with Molecular Sieve Type 13X in the method described in Section 7.2.2.7.2. Experiment 117 with a 20% solution of this material (Δc=1.4%) however still showed complete growth inhibition. It was considered possible that a much higher degree of purity was necessary to initiate growth at this very low supersaturation; not only would this be very difficult to obtain however, but a means of analysing very low impurity concentrations had not been established nor had the critical impurity been isolated.

Experiments 118 to 122 inclusive were carried out, which together with Experiment 113 at 60°C show the effect of temperature on the growth rate for this nominally pure material, purified Batch E.

As anticipated the results show purification to improve the growth rate at all temperatures; for example at 60°C, R.P.E.12 has a rate ca. x 5 faster than R.E.4 and of the same order as R.E.19 at 70°C. However it can be seen that the increase of growth rate with temperature increases with supersaturation, with little difference below about 0.05 supersaturation. R.P.E.17 at 40°C (Δc=3%) showed a low growth rate initially, but the rate then increased. This test was repeated (R.P.E.19) to obtain intermediate valves with the same result. It can be seen that the growth rate has a maximum at a supersaturation, $\hat{s}$, of about 0.1 where the value of approximately 1.8×10^{-5} is higher than that obtained with R.P.E.12 at 60°C, at an equivalent supersaturation. There seems to be no logical explanation for this other than it being the progressive depletion of some contaminant in the mother solution.

7. 2. 2. 5. 4. Filtered Purified Batch E

A 10% m/v solution of Batch P.E. was filtered through a 0.45μ membrane filter and evaporated to dryness. Test R.P.E.F.1. at 70°C showed no growth for the 7 hours of the test. The filtration process was therefore repeated using a completely new set of apparatus washed with distilled water. Test R.P.E.F.2. (a repeat of R.P.E.F.1) now showed a similar growth rate to R.P.E.20 under the same conditions. The first filtered batch must have, therefore, picked up some contaminant in the filtering process. T est R.P.E.F.3 was carried out at 40°C and showed no growth for 22 hours, which, in view of the growth inhibition of R.E.F.3 at this temperature, appeared to be a real effect.

A comparison of R.P.E.7. and R.P.E.20 (70°C) with initial supersaturations of Δc=2.5% and Δc=3.8% respectively showed a faster growth rate with R.P.E.20. As no attrition is expected under these conditions, it is possible that the discrepancy is due to a 'roughness' effect caused by heterogeneous particles. Test R.P.E.F.4. (Δc=2.7%) was therefore carried out and showed a similar growth rate to R.P.E.7 indicating that filtration had no effect. It was therefore unlikely that this could be a roughness effect due to heterogeneous particles and appeared to indicate a contaminant being adsorbed at the beginning of the test with the higher supersaturation (R.P.E.20).

R.P.E.F.5. was then carried out at 60°C and showed a growth rate much slower than R.P.E.12 at this temperature, suggesting that catalytic particles had been removed.

Test R.P.E.F.4 was repeated, (R.P.E.F.6) to find the reproducibility and it was found that the growth rate was much slower initially but after

about 0.7g of solute had been deposited the growth rate became faster than R.P.E.F.4. This is possibly due to the adsorption of contaminant. Reproducibility with filtered material proves to be very poor therefore, whereas with the "naturally occuring" heterogeneous particles present the reproducibility was good. This is possibly due to growth occuring by adsorption on dislocations in the crystal surface which are readily 'poisoned' by traces of contaminant, whereas heterogeneous particles help recreate adsorption sites.

7. 2. 2. 6. Batch F (<0.1% Di-P.E, 5.5% Formal)

Test R.F.1. showed no growth for $\frac{3}{4}$ hour. To check whether an impurity had been picked up in the seed preparation, unprepared Batch F was sieved and the same sieve fraction used for the qualitative experiment 47. This still showed a very slow growth considering far more attrition would be expected with this unprepared seed which would provide a relatively large surface area available for growth.

Control test R.E.8. was therefore carried out which gave a similar growth rate to R.E.1. confirming that there was no impurity in the cell or glassware used for solution preparation.

R.F.3. a repeat of R.F.1 was left overnight to see the extent of the growth inhibition and by morning a reasonable growth had been obtained. The more detailed study R.F.4. showed that in fact most of the inhibition was in the first $\frac{3}{4}$ hour, although growth after this was still slow in comparison with R.A.2. which could hardly be accounted for by the difference in Formal composition.

To check the possibility of sieve contamination as found for the

finer sieves 200-350 mesh in the earlier experiments with P.G. material (Section 7.2.2.7) another qualitative experiment was carried out, 51, using a completely different set of sieves to those used in Experiment 47, but the result was the same.

To confirm that no impurity could have been picked up in the seed preparation, a small batch of Batch E seed was prepared in the same way as the Batch F seed using the same apparatus. The sieve fraction 89 - 105μ was used as seed, and in the computation of this test (R.E.9) the same size analysis was assumed as found for the bulk of the prepared seed of this Batch. This appears to be a reasonable approximation in view of the rate of concentration decrease found. This test R.E.9. gave growth rates an order of magnitude greater than test R.F.4 which confirmed that Batch F must contain some impurity more effective than that attributed to Di-P.E. or Formal, possibly the same impurity as that inhibiting growth in Batch B.

7. 2. 2. 7. 1. Purified Batch G

Batch G was purified by the HCl refluxing method described Section 3.2 and this material then investigated in order to obtain a more complete and accurate study than the previous work (1). Experiment 30 (R.P.G.1) exhibited no growth over a period of five hours. This was repeated in Cell S with the same result it was realised that this anomalous phenomenon could possibly be due to an impurity contracted from the sieves. The 200, 240, 300, 350 mesh sieves had inadvertently been used to sieve a catalyst of vanadium pentoxide on a silica base . The material $<$350 mesh was remixed with the bulk material before use,

and although the sieves had been washed with detergent they could still have contaminated the material.

To check this R.P.G.3 was carried out with material collected in the larger sieves used for the solution and sieve cut 89 - 105μ, i.e. $>$170 mesh, used for seed. This gave a growth rate of the expected order in comparison with previous work (1), and so these four contaminated sieves were discarded and a new set used.

A further quantity of Batch G material was purified. Because of the hazards involved with chipping crystallised material from a glass vessel, a high density polyethylene bucket was now used for the recrystallisations involved. Experiment 40 with this new batch of purified material showed a slower growth than R.P.G.3. In case this could be an unevenly distributed impurity in Batch G, qualitative experiments were carried out, 41 and 42, with Batch G solution and unclassified seed in Cells C and S, which being of the same order, confirmed that the cells were not causing the inhibition. Also as the unclassified seed would be expected to have a larger specific surface than the sieve fractions usually used for seed, this growth appeared to be normal. A repeat experiment R.P.G.5 again gave a similarly slow growth to R.P.G.4. To check Cell C again, this was repeated in Cell S and found to show an even slower growth rate. However, it was noted that some insoluble pieces of material were present in this experiment suspected to be chips from the polyethylene bucket. A small batch of purified Batch D was therefore purified using all glassware and Experiment 45 with this material showed a very fast growth. Although this confirmed that the polyethylene bucket had affected an inhibition of growth in Experiments 40, 43 and 44,

this growth was far faster than R.P.G.3., which had not involved polyethylene. Another batch of P.G. was therefore prepared using all glassware. R.P.G.7 with this material showed a growth rate of the same order as R.P.G.3. i.e., faster than Experiments 40, 43 and 44 but still not as fast as R.P.D.1. The discrepancy between R.P.G.3 and R.P.G.7 is possibly because of the higher initial supersaturation used in R.P.G.3. causing preferential adsorption of an impurity into the crystal lattice.

Runs R.G.3 and R.G.4. were carried out with Batch G solution using 89 - 105μ sieve fraction P.G. and E seed respectively. These showed an inhibited growth of the same order as R.F.4. Both Batches F and G contained $<0.1\%$ Di-P.E. and a high Formal concentration and so although, (as with Batch B,) it seemed inconceivable that this growth inhibition could be an effect of Di-P.E. or Formal, the only conclusive proof of an unknown impurity lay with the comparison of the purified material with R.P.D.1. To confirm that this was not a freak run Batch E was carefully purified and test R.P.E.1 again showed a very fast growth. The rate of decrease in concentration was so great however that it appeared that nucleation had been induced. This theory was strengthened when R.P.E.1. was repeated, Experiment 58 showing a very poor reproducibility. R.P.E.3, therefore, was done using a lower initial supersaturation and still showed a relatively fast growth rate. R.P.G.8 however, with the same conditions and the same seed showed no growth at all for five hours. This confirmed that Batch G. contained some other impurity than Di-P.E. or Formal, which was a very effective crystal growth inhibitor, was not destroyed by refluxing with hydrochloric acid,

and was not detected by the usual gas chromatographic analysis.

7. 2. 2. 7. 2. Attempted Extractions of unknown impurity

As any extraction of production material would be likely to change the concentrations of the known impurities Di-P.E. and Formal and hence the growth rate, P.G. was used for the extractions where any increase of growth rate after an extraction would indicate the effectiveness of the extraction. As this impurity had seemingly not been obvious in the chromatographic analysis, and as previous work (1) had shown that a trace amount of oil could completely inhibite crystal growth, the nature of this impurity was suspected to be an oil or grease. A possible source is from silicone grease used in plug cocks and valves on the production plant. The recommended solvent (90) for silicone grease was Toluene.

One litre of 30% m/v P.G.aqueous solution was made up and stirred in a flask, fitted with a water condenser, with one litre of toluene at 80°C for two hours. The aqueous layer was separated, cooled, filtered and the P.G.T. material dried in an oven. However, Experiment 57 with this material showed a slower growth rate than R.P.G.7. The only explanation for this is that a further impurity was added from the toluene. As only G.P.R. toluene was available, and trace amounts of 'oil' are apparently very effective inhibitors this was quite possible.

At this stage to make completely sure that no impurity was being presented by the cells, possibly as the result of previous impure experiments, Cell S was thoroughly cleared by scouring with a wire

brush on an electric motor. The cell was then washed with a water hose and finally by filling with distilled water and stirring under experimental conditions for about an hour.

One litre of 12 mass% P.G. aqueous solution was made up and held at 50°C in a one litre flask by means of an isomantle. 5.0g of acid washed Kiesulguhr were added and stirred for 3 hours. The solution was then filtered first through a No. 1. Whatman paper (approximately 100μ porosity) and then through a 0.45μ Millipore membrane filter. The P.G.K. material was carefully evaporated to dryness and Experiment 62 in Cell S with this material under the same conditions as R.P.G.8. which had shown no growth for five hours, grew very slowly. However the growth rate was still much slower than R.P.E.3.

One litre of 12% mass fraction P.G. aqueous solution was again made up and stirred in a 3 litre flask fitted with a water condenser with one litre of analar benzene at 50°C, maintained by an isomantle, for two hours. The aqueous layer was then carefully separated off and the P.G.B. material carefully evaporated to dryness. This showed a growth rate, Experiment 63, faster but of the same order as R.P.E.3. after the initial stages. But these initial stages were very much slower than R.P.E.3. As these experiments, 62 and 63, were carried out with Batch E seed, a check test R.P.D.2. was done in Cell C with Batch E seed. This showed a growth rate of the same order as R.P.E.3.

One litre of 10% mass fraction P.G. aqueous solution was stirred in a one litre flask, maintained at 40°C, with 10g Molecular Sieve (Type 13X) for two hours. The solution was then filtered first through a

No. 1 Whatman paper and then a 0.45μ Millipore membrane filter. The P.G.M. material was carefully evaporated to dryness and Experiment 65 at 70°C showed a growth rate slower in the initial stages but subsequently faster than R.P.E.3. Test R.P.E.5 with Batch E seed in Cell C again showed a growth rate of the same order as R.P.E.3 and R.P.D.2. Test R.P.E.6 was carried out under the same conditions in Cell S and showed a growth rate about an order of magnitude greater than R.P.E.5. This was eventually shown Section 7.2.3.1. to be due to an enhancing effect of extraneous particles in Cell S. Reproducibility, however, was shown to be very good (Experiments 67 and 69) under these enhanced conditions. So although Experiments 62, 63, 65 and 67 do not show realistic growth rates under normal conditions, they are comparable in themselves and Experiment 65 shows adsorption in aqueous solution with 10% molecular sieve type 13X to be the most effective method tried of extraction of the unknown impurity. However, comparison with Experiment 67 shows the extraction to be by no means complete. Experiment 73, with purified Batch E material after adsorption with molecular sieve, showed on comparison with Experiment 69 that molecular sieving had no effect on the purity of purified Batch E material. Therefore six successive extractions with 10% m/v molecular sieve type 13X were done on a 10 mass %. P.G. aqueous solutions as described above.

In an attempt to obtain a quantitative estimate of the amount of impurity present, a known weight of sieve was taken from each extract, dried in an oven at 100°C, reweighed and then calcined in an open muffle furnace at about 850°C for three days. The resulting sieve

was again weighed enabling the amount of impurity burnt off to be calculated. Blank tests showed a loss in weight due to the dry sieve only and also with adsorbed purified Batch E which could not be accounted for with the 10% m/m mother liquor allowance. This was attributed to molecular sieve composition and preferential adsorption of P.E. molecules respectively. Compensation for both was made in the impurity content estimates. The results of these extractions are shown Appendix F and although only the first two extractio ns showed a measurable decrease in impurity content, within the accuracy of this method, the theoretical impurity content after six extractions, based on a constant partition coefficient consideration has been calculated as 0.0329ppm(AppendixF). The original impurity concentration was estimated to be 0.26%.

Run R.P.G.M.2, Experiment 96, after two molecular sieve extractions showed no growth for 24 hours, and R.P.G.M.3, Experiment 97, with purified Batch G material after six successive extractions and an estimated.03ppm.impurity content showed no growth for six hours. Attempts at inducing growth by the addition of zinc and copper particles failed. Although these were under conditions of low initial super - saturation (Δc=1.2%) Experiment 98 with purified Batch E material under the same conditions showed a reasonable growth. Complete extraction of the impurity was therefore considered unsuccessful.

7. 2. 2. 7. 3. Attempted neutralisation of the unknown impurity

On the assumption that the impurity might be grease or oil an attempt was made to neutralise the effect by adding detergent

to separate the 'grease' molecules. This was done, Experiment 99, by adding successive 0.1 cm^3 aliquots of 10% non-ionic detergent (Nonidet P.40) which had previously been shown (R.C.4.) to have no effect on the growth of Batch C material, in twenty minute intervals. For 0.3% contaminant present in 27.5% P.E. solution this would require about 1.8 cm^3 of the diluted detergent to give an equal mass. 2.5cm^3 of 0.1 cm^3 aliquots were added with no effect on the growth inhibition. 1cm^3 of undiluted 'Nonidet' was then added and finally when no growth was apparent after a further hour, 1.5 cm^3 of 'Teepol' detergent was added. This was stirred for a further 14 hours, but again complete growth inhibition persisted.

As it had been found that heterogeneous particles enchanced the growth rate in Cell S, it was thought possible that this might be due to the effect of the particles adsorbing the impurity. Experiment 100 was therefore carried out in Cell C with the addition of 0.5 g brass filings cut from Cell S. However there was still complete growth inhibition for 2½ hours.

McCartney and Alexander (63) found that the crystal growth of calcium sulphate was markedly retarded by the presence of polycarboxylic materials but that the retarding action was largely destroyed in the presence of HCl. As the impurity was unidentified in this case, HCl was added in an attempt to nullify the growth inhibition. This experiment 101 was carried out in the glass cell, G and as the required pH necessary was unknown 0.1 cm^3 aliquots of 10%HCl were added in 20 minute intervals until a total of 1.0 cm^3 10% HCl had been added. Five successive aliquots of 0.5 cm^3 of 10% HCl were then added every half hour, and finally 1cm^3,

2 cm^3 and 5 cm^3 of concentrated HCl added at hourly intervals, however this had no effect on the growth inhibition.

As complete removal or neutralisation of the contaminant seemed extremely difficult it was decided to work on Batches C, D and E only - these being comparable with each other and apparently not contaminated (as indicated by Experiment 73 where it was found that molecular sieving, the most effective extraction process found for the contaminant, had no effect with purified Batch E).

7. 2. 2. 8. Purified Batch E + 1.0% Di-P.E. (P.E.D)

A synthetic mixture was prepared of purified Batch E with 1.00% Di - P.E. added in the form of commercial Di-PE chromatographically analysed as containing 4.0% Formal and no detectable quantity of Pentaerythritol. Tests R.P.E.D.1 and 2 were carried out, (Experiments 137 and 138,) at 70°C and 50°C respectively. The calibrations and equilibrium concentrations were taken to be the same as for Pure P.E. which seemed a reasonable approximation as only 1.0% impurity was added. In each case the growth rate was found to be slower than for Purified Batch E, R.P.E.20 and R.P.E.16, but of the same order as found for Batch E, R.E.19 and R.E.38. However, as the Di-P.E. could also contain some contaminant as effective as that in batch G, this is not necessarily an inhibiting effect of Di - P.E. As no analytical method, other than an indication from the effect on growth rate was available for this unknown contaminant it could not be definitely ascertained that the reduced growth rates were due entirely to the Di - P.E.

7. 2. 3. Enhancing effect of heterogeneous particles.

7. 2. 3. 1. Establishing the effect.

As indicated above, it was found that heterogeneous particles present in Cell S after cleaning with a wire brush just prior to Experiment 62, caused a very effective growth rate enhancement. The experiments leading to this conclusion are outlined below.

Experiments 62, 63 and 65 all indicated a partial removal of the impurity causing growth inhibition of P.G. material. However Experiment 67, R.P.E.6, in Cell S, gave a growth rate very much faster than R.P.E.5, under the same conditions in Cell C. R.P.E.5 was therefore repeated, Experiment 68, with a similar result. R.P.E.6 was now also repeated, Experiment 69, in Cell S, again with the same result and very much faster than R.P.E.5 and 7 in Cell C. R.P.D.3 in Cell S also showed a very much faster growth than R.P.D.2, in Cell C under the same conditions.). At this stage it was thought possible that Cell C had aquired some impurity which was inhibiting growth, possibly from the impure Batches G and F in the previous Experiments 40 to 60.

Experiment 71, R.E.10, with 30% m/v Batch E solution at 70°C was carried out in Cell C to compare the result with R.E.1, Experiment 33 under the same conditions in Cell C. R.E.10 showed a similar growth rate although it was faster in the final stages. This was also similar to R.E.7, Experiment 39, carried out in Cell S. The discrepancy in the final stages of growth with R.E.1 being slower than both R.E.7 and R.E.10, could possibly be explained by different hydrodynamic conditions.

Experiment 72, R.E.11. in Cell S confirmed that either both cells C and S were previously inhibited and the inhibiting purity had been removed from Cell S when cleaned prior to Experiment 62, or Cell S now contained a crystal growth accelerator.

A comparison was made R.P.D.4 in Cell A which was first dismantled and thoroughly cleaned and polished. This then showed a growth rate at first slower, but of the same order as R.P.D.2, but nevertheless much slower than R.P.D.3. Although accurate reproducible results were difficult with Cell A because of a poorly defined image, this run showed a definite enhancing effect of Cell S. Cell C was now cleaned with a wire brush and then polished with water soaked paper. R.P.D.5 again showed a growth rate of the same order as R.P.D.2. It was realised that although Cell S had been cleaned with a wire brush prior to Experiment 62, and washed out with a jet of water under experimental conditions, the interior had not been polished.

Cell S, therefore, was again cleaned with a wire brush attached to an electric motor and this time polished with paper soaked in distilled water. The cleaning operation was made difficult, however, by the thermometer pocket projection into the cell interior. Efficient cleaning around this pocket was impractical. Experiment 76, R.P.D.6 with this cell now showed a growth rate faster, but of the same order as R.P.D.5. although very much slower than R.P.D.3 in the same cell before polishing. Also R.E.12 in Cell S and R.E.13 in Cell C with the same conditions as R.E.1 and R.E.7 before the cells were cleaned, showed very similar growth rates. This was conclusive proof that the heterogeneous particles of swarf from Cell S, on the first cleaning had very

effectively enhanced the subsequent crystal growth experiments. This could possibly be a catalytic effect of the metal ions from the brass, or creation of secondary nuclei or else adsorption of the heterogeneous particles into the crystal surface creating "growth sites" for crystal growth.

Cell C was now given the same treatment as Cell S prior to Experiment 62 i.e. it was thoroughly cleaned with a wire brush on an electric motor, washed with a jet of water and stirred with distilled water. Experiment 79, R.E.14 in Cell C after this treatment showed a growth rate enhanced by about 15%. To confirm that this was due to the presence of copper particles, and obviate the possibility of it being due to the removal of trace amounts of impurity, Cell C was thoroughly polished with paper, soaked with distilled water, and R.E.15 carried out after this polishing showed a similar growth to R.E.13 before cleaning with the wire brush.

0.5 g zinc powder having an average particle size of about 3μ was added to Cell C at the start of R.E.16 and resulted in growth of seed approximately 20% faster than R.E.15. 0.5 g copper powder having an average size of about 50μ but containing many particles $<10\mu$ was added to R.E.17 resulting in a growth of seed slower than R.E.16 but still about 10% faster than R.E.15.

Although this confirmed that heterogeneous particles enhanced crystal growth, the degree of enhancement was considerably more in Cell S (e.g. R.E.11 about three times as fast as R.E.15). This could be either due to the larger number of heterogeneous particles in the swarf produced by the wire brush, or else an additional catalytic effect of some metallic ion, other than copper or zinc present in the brass. To

distinguish between these possibilities, o.5 g of brass filings were taken from Cell S, consisting of flat jagged plates which when measured across an average dimension on a microscope slide and assumed spherical, gave a surface area of 215 cm^2 for 0.5 g which would be of the same order as the smooth internal surface of Cell S in contact with the mother liquor during a run. However due to the particle shape the actual surface area of filings would be well in excess of this figure. These filings were added in Experiment 139, R.E.40 in Cell C giving a growth rate of seed about 30% faster than R.E.15. On examination of products after these metallic particle additions it was observed that they were incorporated into the crystals. Also with each of these runs, the particle additions were made before the introduction of seed, and after stirring for about 15 minutes no decrease in concentration was observed. It was therefore concluded that heterogeneous particles present during crystal growth enhance the growth rates, possibly by adsorption onto the crystal surface causing dislocations in the crystal structure and making available more growth sites for deposition of solute molecules.

7. 2. 3. 2. Absolute cleanliness of Cell C

In view of this enhancing effect of extraneous particles it was essential to have a cell completely free of foreign particles. This was impractical with Cell S due to the thermometer pocket obstruction making absolute cleanliness around this pocket very difficult. At this stage it was considered necessary to check Cell C with a clean glass cell. As a first approximation a jacketed beaker thoroughly cleaned with chromic acid, benzene and distilled water, of approximately 400 cm^3 capacity was

used. It was fitted with a large rubber bung, a thermometer, and a stirrer positioned centrally.

Experiment 83 (R.E.18) was carried out with this beaker filled with 250 cm^3 of 30.0% m/v Batch E solution and 2 g of Batch E 89-105μ seed and stirred for 3 hours at 70.00C. The resulting solution was poured into Cell C held at 70.0°C and the refractive index read. The result was of the order expected from R.E.15.

Cell C was again dismantled and thoroughly cleaned and polished. As the prism on this cell had a contact adhesive sealing the refractometer prism and the Cell S had an epoxy resin seal, to obviate the possibility of either effecting crystallisation, the contact adhesive was removed and replaced with epoxy resin. R.E.19 done with this cell showed a very similar growth to R.E.15 except in the final stages below 0.06 super - saturation where it was rather faster, but this is accounted for in the next section.

A glass cell, with similar dimensions to Cell C, Cell G, was made with the light path through a suspension kept to a minimum for ease of use. An enclosed jacket proved difficult in construction so a constant volume type water circulator was used and the jacket left open. Experiment 87 with the first glass cell at first indicated a very fast growth rate with a rapid decrease in concentration. However this proved to be due to an interchange of suspension and circulating water around the lip of the glass refractometer socket, and eventual dissolution of crystals. Further investigation showed the height of this lip to be critical. A water-tight seal was difficult, involving a refractometer clamped to the glass cell, and undesirable. So the refractometer socket

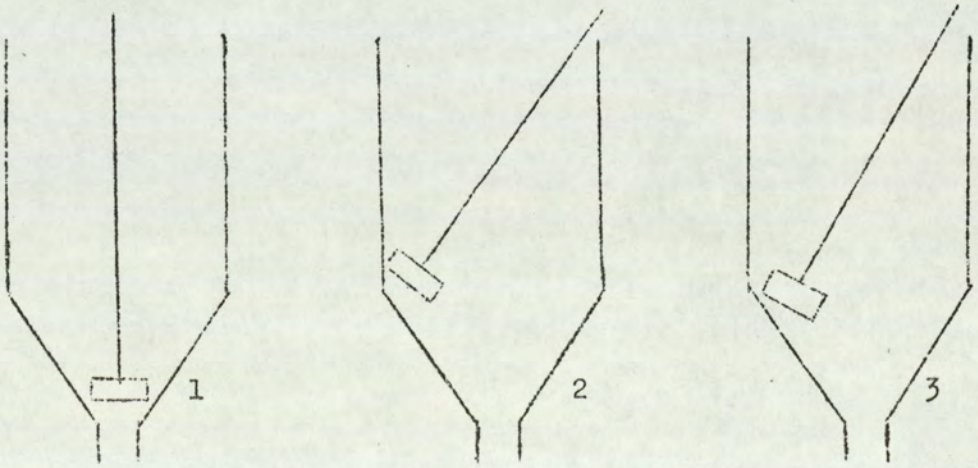
lip had to be of such a height to allow for solution expansion with temperature, the hydrodynamic fluctuations at 2000 r.p.m. stirrer speed but still to largely cover the prism face. Such a cell was constructed but difficulty was experienced (R.E.23) in obtaining a well defined image, so the accuracy of this run was suspect, although, overall of the same order as R.E.19. The optimum position of the light source was found to be beneath the cell and R.E.24 (a repeat of R.E.23) was more successful and showed a similar growth to R.E.19 in Cell C. A further comparison of Cells G and C, R.E.28 and R.E.29 respectively, at 60.00C again showed very similar growth rates. So as the Cell C was more robust and easier to use than Cell G, all further experiments were carried out in Cell C. Experiments enhanced by heterogeneous particles were: Experiment No: 62, 63, 65, 67, 69, 70, 72, 73, 76, 77, 79, 81, 82, 139.

7.2.4. Hydrodynamic Considerations

Experiment 18, 21 and 19 with Batch C at 500 r.p.m, 1000 and 2000rpm. respectively, showed the effect of stirrer speed to be that of holding the particles in suspension. Each showed a similar growth rate until 0.09 supersaturation when the growth rate of R.C.13 at 500 r.p.m. (with a crystal mass at this point of about 6g) started to fall off in comparison with R.C.14 at 2000 r.p.m. However, R.C.16 at 1000 r.p.m. remained the same until a supersaturation of about 0.075 when (at a crystal mass of about 8g) it started to fall off in comparison with R.C.14. These critical crystal masses seemed to indicate the point at which settling of crystals began for a particular stirrer speed. It is of interest to note that although an increase of stirrer speed maintained the crystal suspension, the calculated growth rate constant K based on

first order integration assumption, still decreased progressively with supersaturation.

On the basis of these results 2000 r.p.m. seemed quite adequate to maintain the crystal suspension and so all runs after Experiment 22 were done at this stirrer speed. It was noted however on comparison of runs with Batch E material with different cells and particularly after reassembly after the various cleaning operations, that there was an unexpected variation in the final stages of the runs as the concentration approached equilibrium (e.g. R.E.19 was faster than R.E.13 below about 0.06 supersaturation). This was attributed to different hydrodynamic conditions. Experiment 85, R.E.20 was therefore carried out at 2500 r.p.m., the maximum speed attainable with the particular motor used. However growth appeared much faster throughout the run, using the size analysis after attrition at 2000 r.p.m. for the basis of calculation, even in the initial stages. In view of the comparison of Experiments 18, 19 and 21 this must have been due to additional attrition. The stirring was also quite violent at this speed, so 2000 r.p.m. was again used and a study of the stirrer position made.



The above three positions were tried with the rubber bung in each

case made to accomodate the stirrer shaft and thermometer. In position 1 the shaft was placed vertical and central such that the impeller blades had about ¼ inch clearance with the cone of the cell. This was about as low as vibrations of the stirrer would allow, without the blades touching the sides. R.E.21 in this position at 2000 r.p.m. showed an increased swirling of solution than in previous experiments, which caused seepage in the mother liquor arround the rubber bung. Growth rates were calculated to be about 10% faster in this run down to a supersaturation of about 0.05, which was attributed to this seepage of mother liquor and hence a greater crystal/solution ratio than calculated. However below 0.05 supersaturation when the crystal mass had increased to about 9 g the growth rates were much slower than in previous experiments (c.f. R.E.19). This was thought to be due to the crystals having grown large enough to fall past the stirrer blade settling in the 'dead-pocket' below.

Run R.E.24 in Cell G was carried out with the stirrer in an off-set position as in R.E.19 and previous experiments with Cell G. Growth rate results were shown to be similar with no settling observed in the glass cell.

Although it had been noted that the stirrer had been off-set in the previous experiments in Cell C, the exact position had not been noted.

R.E.25 was carried out in Cell C with the stirrer in position 2 i.e. the stirrer was off-set with the impeller/shaft union being on a level with the cone/cylinder junction of the cell, and the impeller blade about ¼ in from the cell wall. The growth rate results were similar to R.E.19 until 0.04% supersaturation but slower after this value when the seed mass was about 9.5g. The stirrer

was again changed, with the impeller moved lower and off-set further, so the tip was on a level with and almost touching the cone/cylinder junction, as shown - position 3. R.E.26 done at 2000 r.p.m. in this position showed a growth rate faster than R.E.25 and similar to R.E.19.

To test the suspension of large crystals a 33.5% m/v Batch E solution was made up for test R.E.27 at 70°C. The initial refractometer reading was off-scale but estimated at 110 which gave an initial concentration of 33.3% m/v. Growth rates for this test below 0.08 supersaturation were of the same order as R.E.19 indicating complete suspension even with 20 g of crystals of which about 20 No% $>110\mu$ equivalent spherical diameter.

R.E.28 with 27.5% m/v Batch E solution at 60°C in a glass cell was examined carefully for any settling towards the end of the run. But crystals remained in complete suspension. R.E.29 under the same conditions in Cell C showed a very similar growth rate. This stirrer position was therefore adopted as producing satisfactory hydrodynamic conditions for suspension of crystals of the size to be examined at 2000 r.p.m.

Although the off-set stirrer position had not been noted accurately, in experiments prior to Experiment 85, a comparison of R.E.3 and R.E.29 shows some scatter in individual growth rate values due to the small time intervals involved but similar rates overall even for low supersaturations with a crystal mass of 20 g. The earlier experiments in Cell C must have, therefore, been under similar hydrodynamic conditions.

SECTION EIGHT

DISCUSSION.

8.1. Equilibrium in aqueous solution.

Equilibrium of a solute in contact with a solvent may either be achieved by growth of the solute in a supersaturated solution of the solute in solvent (i.e. crystallisation) or dissolution of excess solute in the solvent. Crystallisation processes are thought to proceed by the two consecutive steps: diffusion of molecules to the crystal surface and then integration of the molecules into the crystal lattice. This latter integration step is often so slow as to be rate controlling. Dissolution however is thought to proceed only by a diffusion process of molecules into solution. Equilibrium values for use with the growth experiments were therefore obtained from dissolution in the absence of the possibly inhibiting surface integration step. It was observed in the growth experiments that growth usually ceased at a concentration greater than the equilibrium concentration found from the dissolution tests. Burton et al (42) also reported this phenomenon of a lack of crystal growth even when the solution in contact with the crystal had a supersaturation as high as $s \simeq 0.1$. They attributed this either to the absence of dislocations in the crystal surface, or else to the presence of so many of them that the mean distance between them is too small for the particle integration. As this would require of the order of 10^{12} dislocations per cm^2, they favoured the former explanation. In this

work it was observed that this "apparent equilibrium" value, where the crystal growth ceased, depended on the rate of growth in the particular experiment.

Although no account was taken of these final readings for use with the growth experiments, occasionally the experiment was continued to ascertain that there was no further decrease in solution concentration. These final readings representing an "apparent equilibrium obtained from crystal growth are recorded in the table on p. 162 , together with the time for which this particular concentration value was observed. The longer times sometimes include an increase in solution concentration due to evaporation.

Growth Run.	T_o °C	Apparent Equilibrium.		Time observed at this conc. Min.	Equilibrium obtained from dissolution c_∞ % m/v.
		Scale S	c % m/v.		
R.D. 5.	70.0	87.3	26.998	660	26.310
R.E. 1.	70.0	86.9	26.886	1200	26.310
R.E. 7	70.0	85.8	26.576	1460	26.310
R.G. 1	70.0	88.2	27.251	840	26.310
R.E.11.	70.0	84.9	26.323	2400	26.310
R.E.16.	70.0	85.4	26.464	760	26.310
R.P.G.3.	70.0	81.6	25.597	720	24.771
R.P.E.6.	70.0	80.2	25.184	110	24.771
R.P.E.7.	70.0	81.4	25.538	720	24.771
R.E. 5.	60.0	70.5	20.523	6000	20.067
R.E.29.	60.0	70.3	20.466	880	20.181
R.P.E.14	60.0	65.0	19.011	720	18.634
R.C. 7	50.0	59.0	15.676	1500	15.106
R.E. 6	50.0	59.0	15.676	2400	15.277
R.P.E.16	50.0	53.7	14.729	600	14.116
R.P.E.17	40.0	46.2	10.812	720	10.610

It can be seen that R.G.1. with a relatively slower growth rate due to contamination of Batch G material indicated a higher "apparent equilibrium" value than experiments with Batches D and E at the same temperature. It can also be seen that R.E.11 carried out in the scoured cell S with the enhancing effect of the heterogeneous particles

indicated a much lower equilibrium value than R.E.7. under the same conditions in the clean cell C. However when 0.5 g Zn dust was added to the solution in cell C, R.E.16, the growth rate was enhanced and the apparent equilibrium attained was lower than R.E.7. although not as low as R.E.11. This effect was noted also with purified Batch E, being the "purest" material obtained. Growth still ceased at some value higher than the equilibrium value and the enhancing effect of the heterogeneous particles in cell S depressed the "apparent" equilibrium" value obtained (R.P.E.6.) compared with that in the clean cell C (R.P.E.7.). The actual equilibrium values obtained for Pure P.E. (section 5.) were obtained both by dissolution and by growth of nucleated solutions where a large surface area is available for growth, and found to agree. It appears therefore that the cessation of growth depends on the growth rate and is probably due to the lack of dislocation sites. However heterogeneous particles enhance crystal growth, possibly by increasing the number of growth sites, and permit growth at lower supersaturations.

As the presence of Formal enhances the P.E. solubility the exact concentration of impure P.E. solutions is required for the equilibrium (solubility) determination. Nucleation of the solutions of the required concentration would be difficult if an attempt was made to obtain equilibrium from growth, and so equilibrium was achieved from dissolution of the required solute concentration simulating a growth experiment. The results obtained were considered to be more reliable than any determination from growth. The results were well correlated by the conventional log x vs $\frac{1}{T}$ plot

above 50°C, but below this temperature the solubility was enhanced (by about 1 mass % at 30°C) compared with that expected from the correlation. Although this might be due to a sudden change in the impurity partition coefficient the difference at 30°C could hardly be accounted for by the amount of impurity present unless the chromatographic chemical analysis (table 1) is considerably different from the actual mass concentrations.

An unusual phenomenon was observed in the dissolution results of the impure materials. It was found that an enhanced solubility was obtained initially which then decreased gradually to a limit at which it remained constant. This was assumed to be the equilibrium value. The same value was attained whether the impure material contained Di-P.E. or not, but the degree of initial enhanced solubility was severely restrained with the presence of Di-P.E. (figure 5.4).

A possible cause of this temporarily enhanced solubility effect could be the non-homogeneous distribution of the Formal within the crystal in the initial batch of P.E. material used. If this contained more Formal in the outer extremes of the crystal than in the core (possibly as a result of the method of preparation) then since the diffusivity in the liquid would be expected to be greater than in the solid an initially high Formal concentration in the liquid would give an enhanced solubility. Equilibrium is then achieved by diffusion of the excess Formal into the remaining crystals from the solution thus resulting in a slightly lower solubility due to the purer mother liquor. This might also account for the enhanced values at $< 50^{\circ}C$ where the solid diffusivity and mobility in the crystal

lattice might be too low to attain impurity equilibrium, resulting in mother liquor of relatively high Formal concentration.

The effect of the Di-P.E. appears to be that of arresting the initial enhanced solubility which might be the effect of the Di-P.E. on the equilibrium system at this temperature, or possibly the effect on the original crystal preparation creating a more homogeneous impurity distribution.

It would be interesting to note the effect of annealing the original solid P.E. at a high temperature. This might allow impurity diffusion in the solid creating a homogeneous crystal and prevent the initial solubility enhancement on dissolution.

8. 2. Results of Previous Work (1) on Crystal Growth.

In the previous work (1) growth rates were calculated from measurements of solution refractive index change in seeded cells, assuming a first order dependence on supersaturation $\left(c - c_{\infty}\right)$ such that $\frac{dc}{dt} = - K A \left(c - c_{\infty}\right)$. In order to do this the area after each concentration change had to be calculated and the approximation $\frac{A_2}{A_1} = \left(\frac{M_2}{M_1}\right)^{\frac{2}{3}}$ was used. Results for K excluding the initial and final few readings were averaged and the activation energies found. A summary of the results found for Batch A and purified Batch A are shown in table 49 appendix E. The activation energies for growth of Batch A and purified Batch A were found to be 30.65 and 18.4 K cal / g mole respectively. A number of improvements to this

method have been used in this work: A more accurate refractometer has been used; the area determination has been made allowing for the crystal size distribution; the Coulter Counter size analysis has been corrected to obtain a true equivalent spherical volume diameter; the calibrations and equilibrium determinations have been redone for greater accuracy.

The refractometer readings of the previous work were as accurate as the apparatus allowed. These were converted to the equivalent refractometer scale S readings of this work to enable the more accurate calibrations to be used and for use with the computer program 2 to allow for the size distribution of the attrited seed for the area determinations. This has been done, tables 150 and 151 with R.P.A.1. and R.A.5. using the equilibrium refractometer readings of the previous work and the uncorrected Coulter Counter size analyses (i.e. D_c) for a direct comparison. It can be seen that the effect of the average area approximation used in the previous work is accumulative throughout the calculation and R.P.A.1. shows an error of about x 2 in the final K determination with a final area of 9164 cm^2 as opposed to 5030 previously calculated. However readings are too few and over too limited a supersaturation range in these two experiments to accurately show the dependence on supersaturation. Tables 152 and 153 show the same two experiments calculated using the corrected equilibrium values found from dissolution in this work (section 5). The growth velocities g are, of course, as calculated in the previous tables, but when plotted against the

supersaturation s showed the sensitivity of the correlation on the equilibrium value. A comparison of the growth rate constants K also showed the sensitivity of these values on the equilibrium concentration which becomes more pronounced for the lower supersaturations. The correction was made for the Coulter Counter equivalent spherical diameters (i.e. D as opposed to D_c) and all the results for Batch A and purified Batch A (P.A.) calculated using the new equilibrium values and computation for area determination. The results are shown tables 154 to 160 , appendix E. It can be seen from a comparison of R.P.A.1. and R.A.5. before and after correction that the Coulter Counter diameter correction makes only about 1% difference in the growth rates in this case. The most significant difference of these corrections is that of allowing for the crystal size distribution of the attrited seed as opposed to calculating the attrited seed area and assuming a monodisperse crystal system.

The growth rate velocities, g (cm / min) have been plotted figures 8.1 and 8.2 as log g vs log s and the values of k_L and b found for the correlation $g = k_L s^b$ by the method of least mean squares. The values are shown below. The results at 80°C could not be calculated by this method as the calibrations had not been done at this temperature.

BATCH A.

Run No.	T_o °C	Corrected Temp. °C.	k_L	b
R. A. 1.	60.0	60.1	0.000183	2.701
R. A. 2.	70.0	70.2	0.000458	1.977
R. A. 3.	55.0	55.1	0.00116	4.375
R. A. 5.	50.0	50.1	0.0000357	3.408

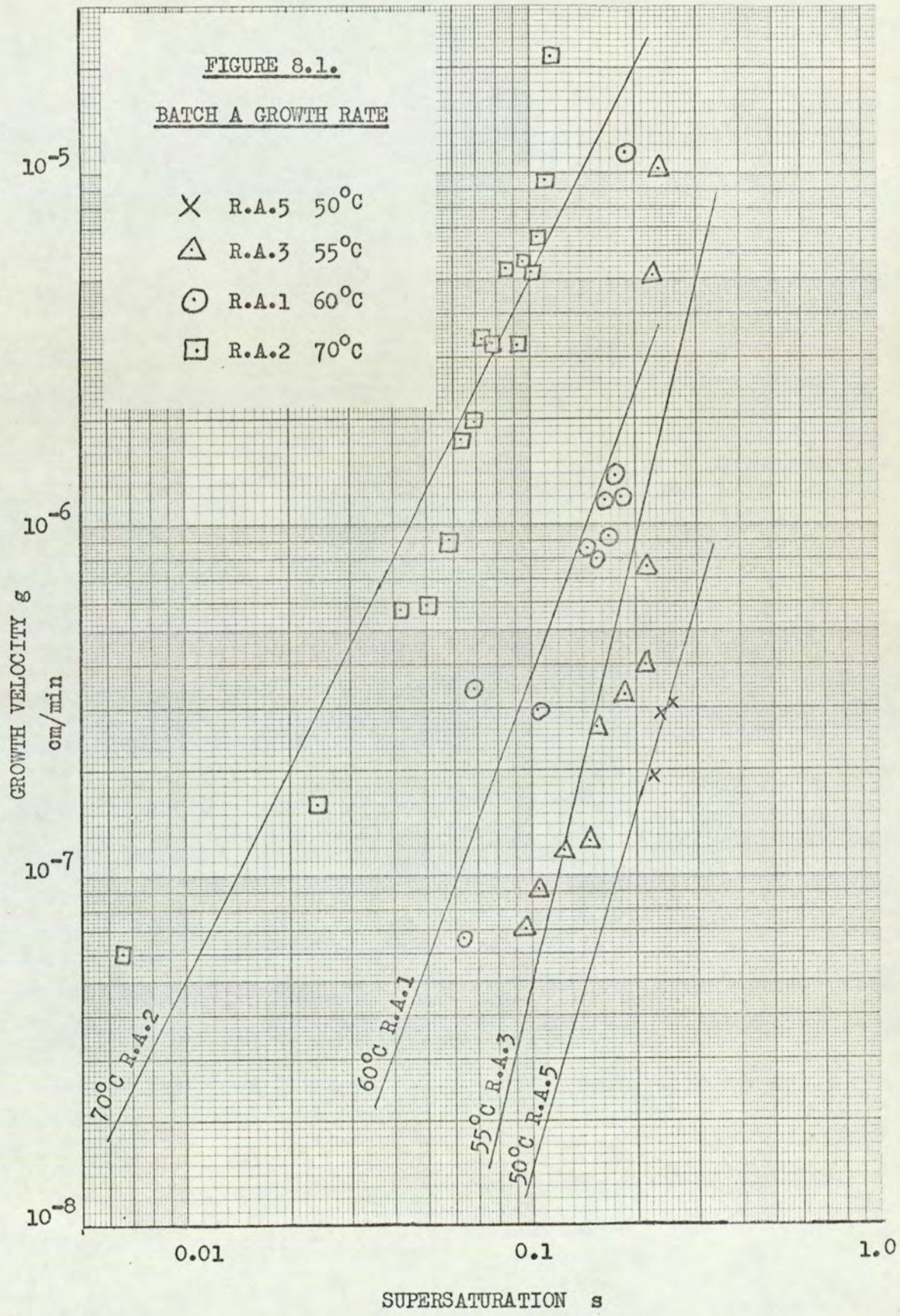
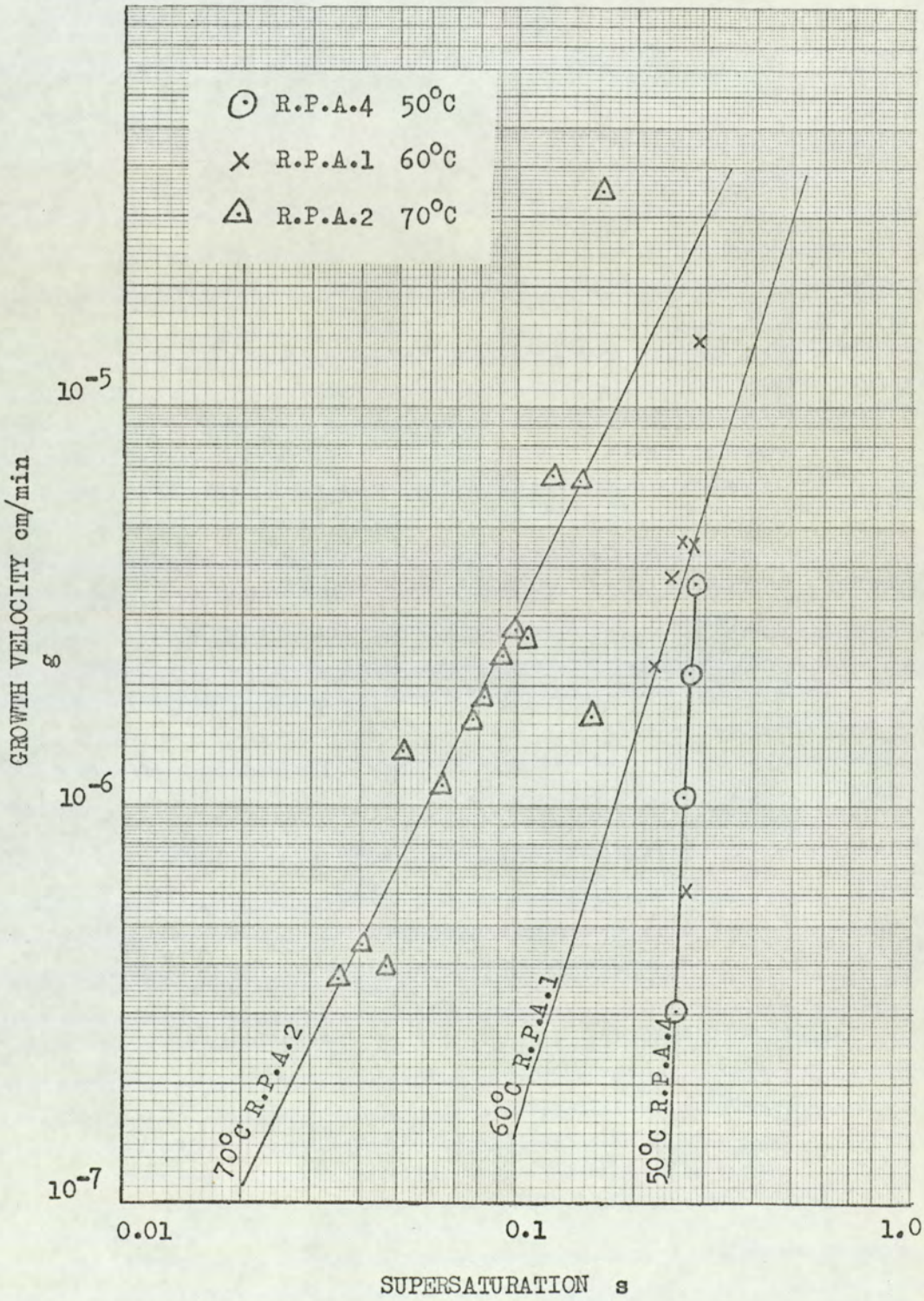


FIGURE 8.1.
BATCH A GROWTH RATE
R.A.5 50°C
R.A.3 55°C
R.A.1 60°C
R.A.2 70°C
10-5
10-6
10-7
10-8
GROWTH VELOCITY g cm/min
0.01
0.1
1.0
SUPERSATURATION s
70°C R.A.2
60°C R.A.1
55°C R.A.3
50°C R.A.5

FIGURE 8.2.

PURIFIED BATCH A GROWTH RATE



Purified BATCH A (P.A.).

Run No.	T_o °C	Corrected Temp. °C.	k	b
R.P.A.1	60.0	60.1	0.000303	3.319
R.P.A.2	70.0	70.2	0.000395	2.115
R.P.A.4	50.0	50.1	8.3×10^8	25.97

It should be possible to correlate the growth rate constants k_L using the Arrhenius correlation of $\frac{d \ln k_L}{dT} = \frac{E}{R' T^2}$

and hence $\ln k_L = \ln A - \frac{E}{R' T}$

where T is degrees Kelvin; R' the Universal Gas Constant; A is a constant and E the activation energy for growth. This was done with apparent success in the previous work (1) for the averaged growth rate constants $\bar{K}$ based on a first order supersaturation dependence.

However values of k_L are extremely sensitive to the slope (exponent of s) b. In the previous work as the growth rate was assumed to be first order with respect to supersaturation, not many readings were taken and the supersaturation range was often very limited. Very few readings were taken at low supersaturations as the results were known to be more sensitive to the equilibrium value which, for the impure material, was suspect. It is fortuitous that R.A.5. at 50°C with only three readings over a very limited supersaturation range gives an exponent b of the same order as the

other runs. Only four results were obtained with R.P.A.4 however and due to the very limited supersaturation range they gave an exponent b of 26 and consequently an extremely magnified k_L of 8.3×10^8. The data for most of this previous work is therefore too limited to obtain overall correlations of the type $g = k_L s^b$, and the Arrhenius correlation cannot be applied to k_L values determined. However over the range studied the actual growth velocities, g, are as accurate as the previous refractometer readings allowed, and can be compared with the present work.

Although the stirrer speed was only 500 r.p.m. in the previous work as compared with 2000 r.p.m. used in this work, the seed used (especially after attrition) was smaller than that used for most of this work, and as it has been shown that the function of stirrer speed is merely that of maintaining the crystals in suspension the results should be comparable in this respect. A visual suspension test had also been carried out with this seed in the previous work, and 500 r.p.m. had appeared satisfactory.

8. 3. Crystal Growth.

8.3.1. Relative Velocity Effect.

The effect of the relative crystal / solution velocity was investigated using a fluidised bed of crystals and also by varying the stirrer speed in a suspension. In the fluidised bed experiments growth rate determinations were made using direct measurements of seed and product crystals, and also by measuring the crystal mass increase.

Attempts were also made to follow the concentration decrease of the circulating mother liquor of the fluidised bed experiments. However, because of the slow growth rate of P.E. and the difficulty of obtaining an air-tight seal using a submerged impeller pump, the evaporation rate tended to compensate for the solution concentration decay due to growth. This method might be developed to overcome this problem, possibly by using a magnetically operated pump to avoid grease from glands. Measurements of the crystal mass increase in situ in the fluidised bed cell proved to be the most reliable used, and it was found that results were in reasonable agreement with those of the stirred cell experiments. It was also found that the only effect of stirrer speed in the stirred cell experiments, apart from that of crystal attrition, was that of maintaining the crystals in suspension. It was therefore concluded that the crystal growth rate of P.E. was surface integration rate controlled for all conditions investigated up to 70°C. A "repetitive inversion sedimentometer" built to investigate the relative velocity effect of crystals under terminal velocity conditions, although constructed was therefore not used as no more useful information was thought to be obtainable by this method for P.E.

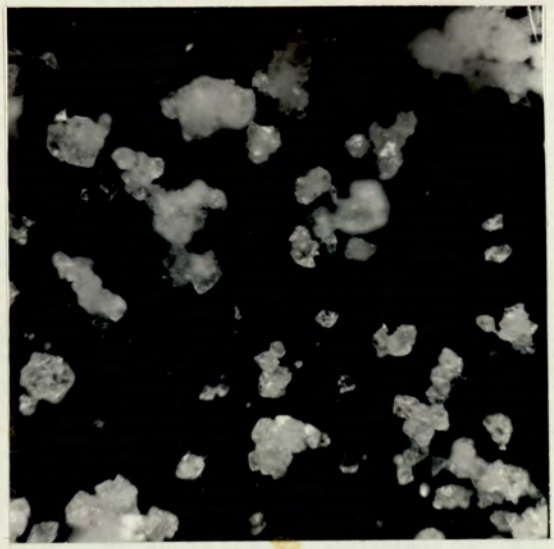
8.3.2. Seeded Solutions in stirred cells.

8.3.2.1. Experimental Testing of Mathematical Model.

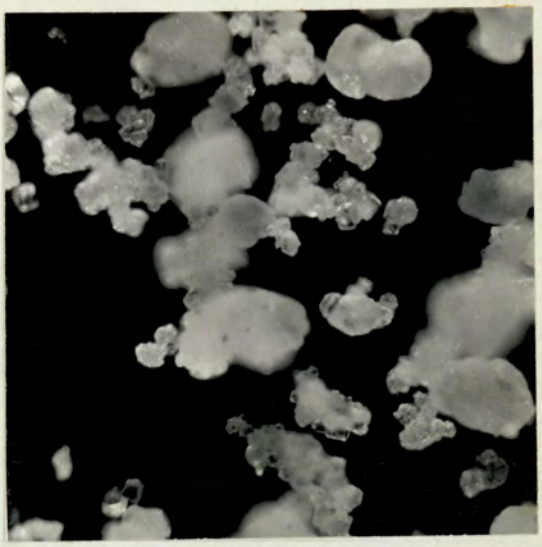
Figure 8.3. shows a selection of product crystals after growth in the stirred cells, obtained by filtering the suspension after an experiment through a No.1 Whatman filter and washing with

FIGURE 8.3.

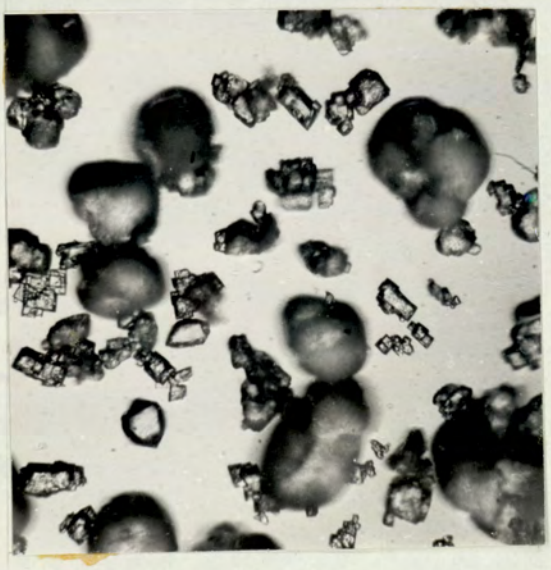
Magnification x 240



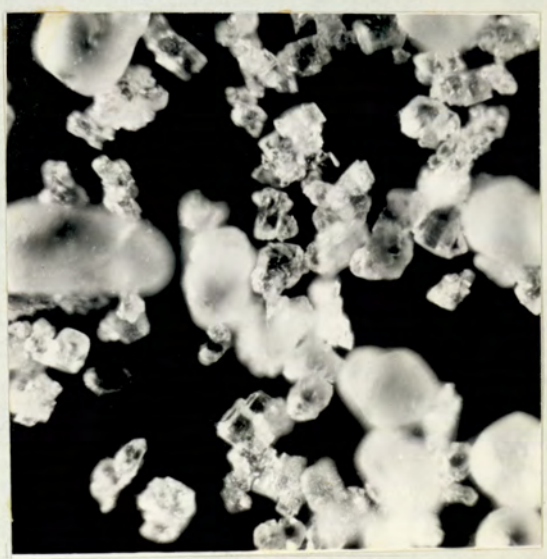
8.3.a Attrited Batch E Seed
89 - 105μ



8.3.b R.E.15 Product 2g Seed
70°C Initial $\Delta c \simeq 4\%$



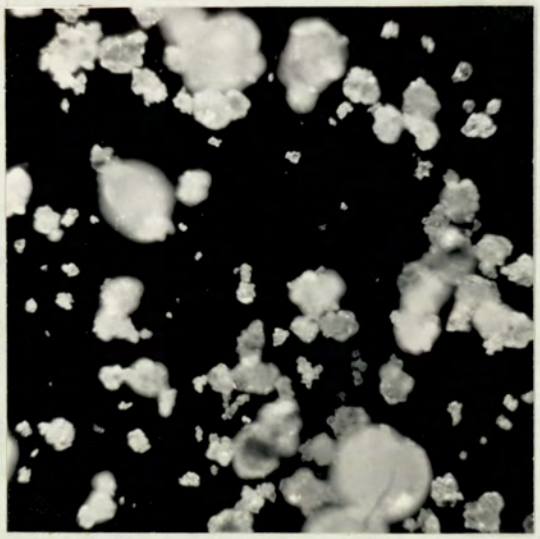
8.3.c R.P.E.12 Product 2g Seed
60°C Initial $\Delta c \simeq 3.2\%$



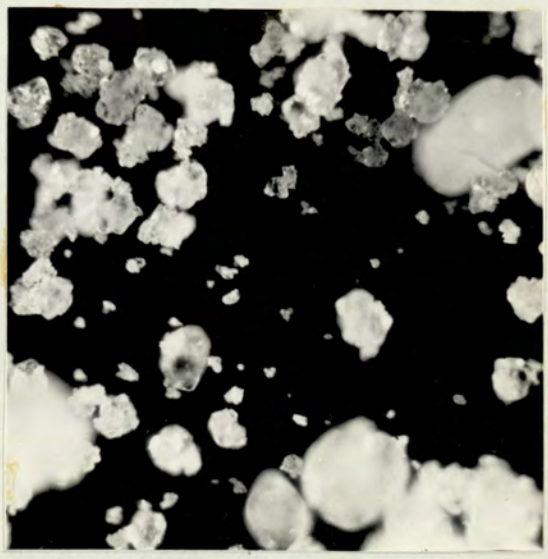
8.3.d R.P.E.12 Product 2g Seed
Initial $\Delta c \simeq 3.2\%$ 60°C

FIGURE 8.3. (CONT.)

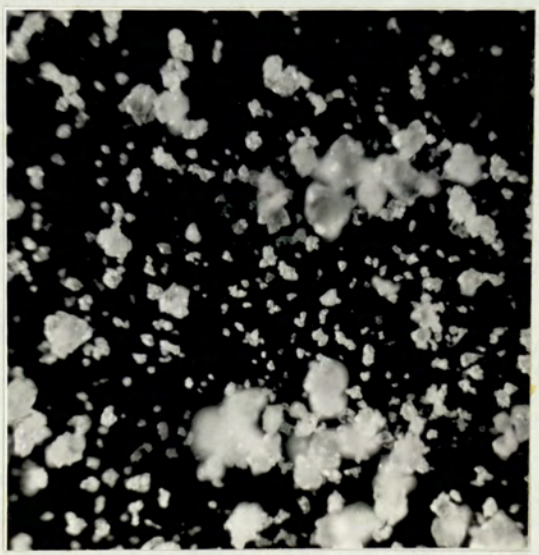
Magnification x 240



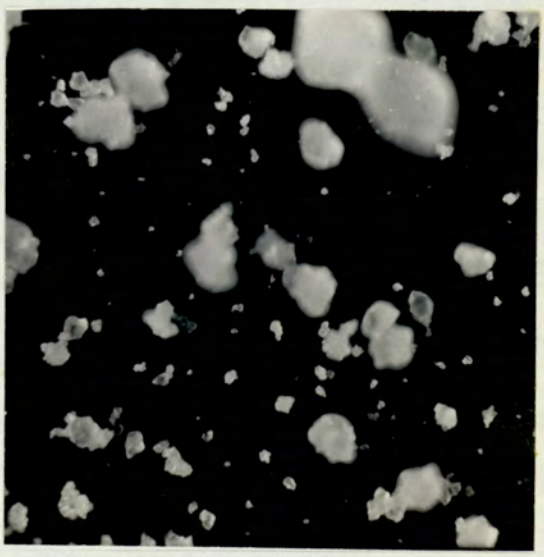
8.3.e R.E.36 Product 1.0g Seed
60°C $\Delta c \triangleq 4.5\%$



8.3.f R.E.37 Product 0.5g Seed
60°C $\Delta c \triangleq 4.5\%$



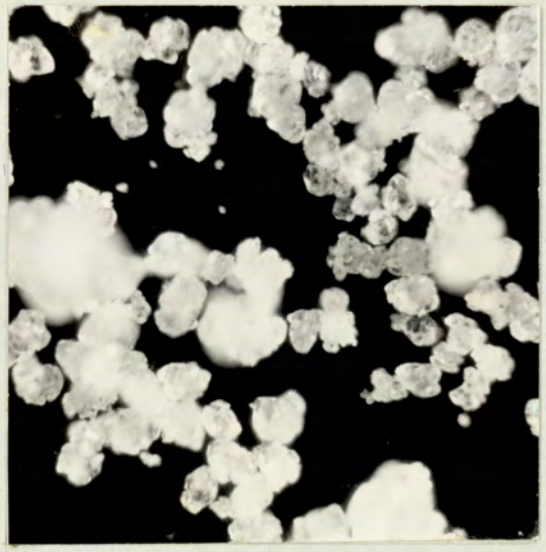
8.3.g R.E.34 Product 1.0g Seed
60°C $\Delta c \triangleq 6\%$



8.3.h R.E.35 Product 0.5g Seed
60°C $\Delta c \triangleq 6\%$

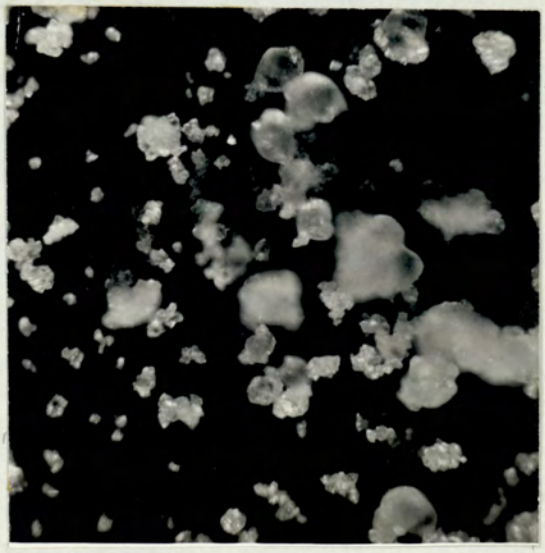
FIGURE 8.3. (CONT.)

Magnification x 240

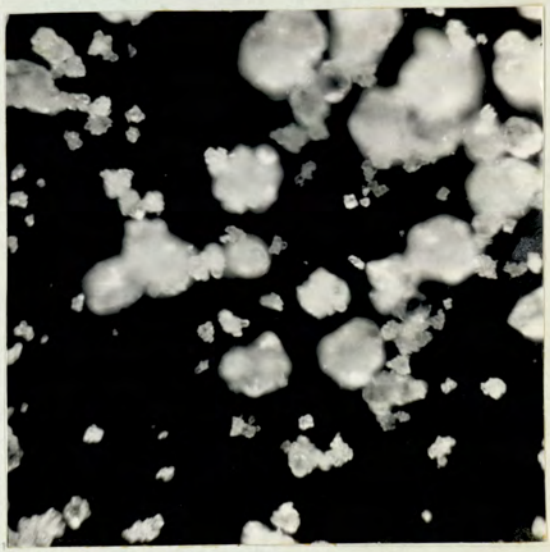


8.3.i R.E.33 Product 0.5g Seed
60°C $\Delta c \simeq 7.5\%$

8.3.j R.E.32 Product 1.0g Seed
60°C $\Delta c \simeq 7.5\%$



8.3.k R.E.16 Product 2g Seed
70°C $\Delta c \simeq 3.5\%$
0.5g Zn DUST ADDED



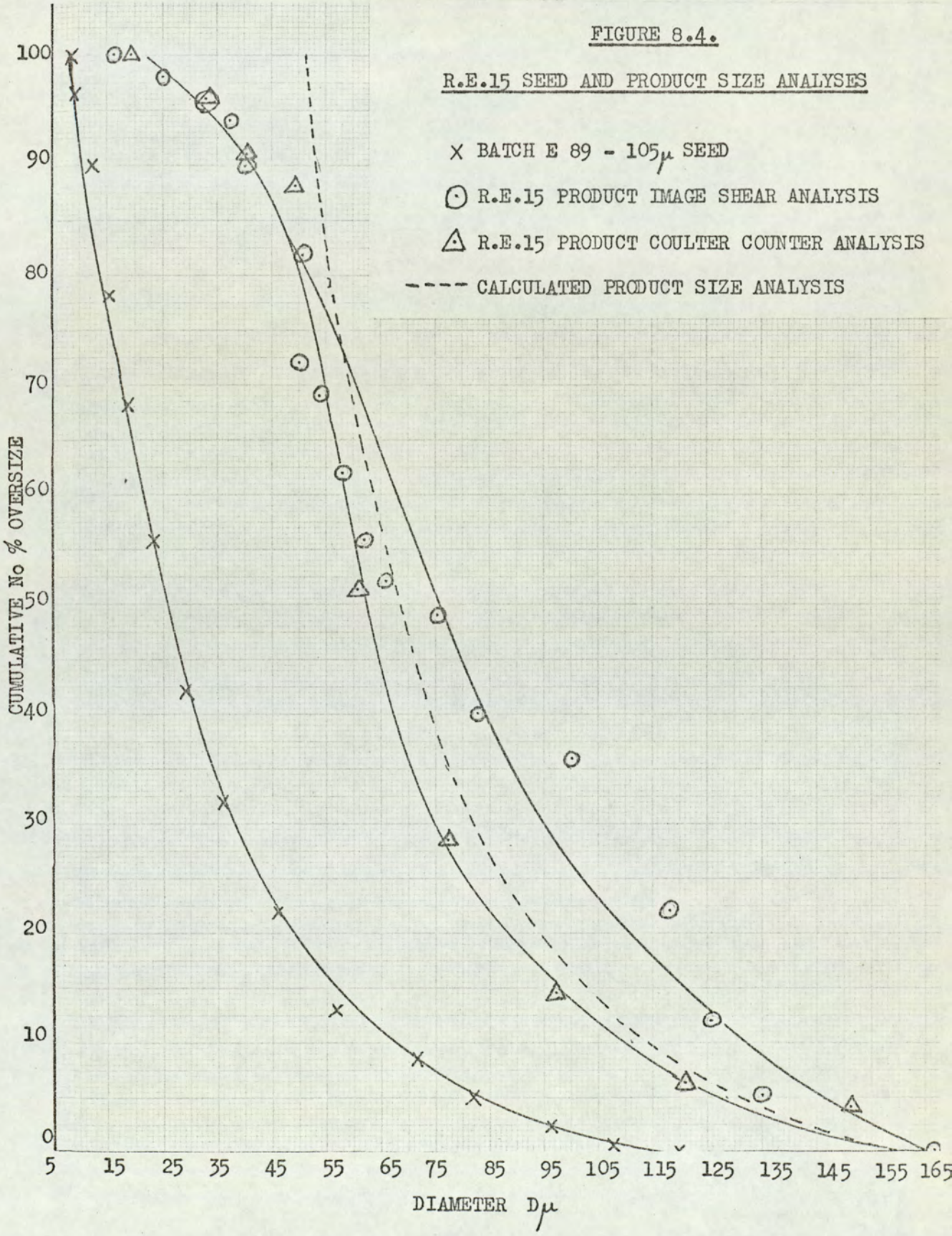
8.3.l R.E.F.4 Product 2g Seed
60°C $\Delta c \simeq 4.5\%$

acetone to avoid agglomeration. 8.3a shows a typical seed (Batch E 89 - 105 μ sieve fraction) after attrition in a saturated solution with a stirrer speed of 2000 r.p.m. As expected the attrited crystals were irregular with a wide size distribution. Figure 8.3b shows a typical Batch E product grown under the usual experimental conditions in the stirred cells of initial $\Delta c \simeq 4\%$ with 2 g of seed. As this had been carried out at 70°C, growth was faster than at lower temperatures and crystals would be expected to be weaker than if grown at a lower temperature. However, although the product was irregular in shape, as might have been expected after the attempts to grow impure single crystals, there appears to have been very little attrition. Figure 8.4. shows graphically the analyses of attrited Batch E seed and R.E.15 product which was analysed by both the Image Shear and Coulter Counter techniques. The Coulter Counter analysis of the attrited seed only is shown as the Image Shear analysis (table 27) showed a marked deviation attributed to the irregular crystal shapes. The Coulter Counter analysis of the product was also considered more accurate than the Image Shear method because of the dependence of the Image Shear method on a characteristic dimension. The largest diameter visible on the microscope slide was normally taken with agglomerates which would account for the oversizing of the larger crystals. The dotted line indicates the calculated product size analysis with an overall diameter increase of 43 μ (from R.E.15).

Figures 8.3 c and d show the product crystals of R.P.E.12 using refracted and reflected light respectively. The crystals are more regular than with the impure material and are transparent.

FIGURE 8.4.

R.E.15 SEED AND PRODUCT SIZE ANALYSES



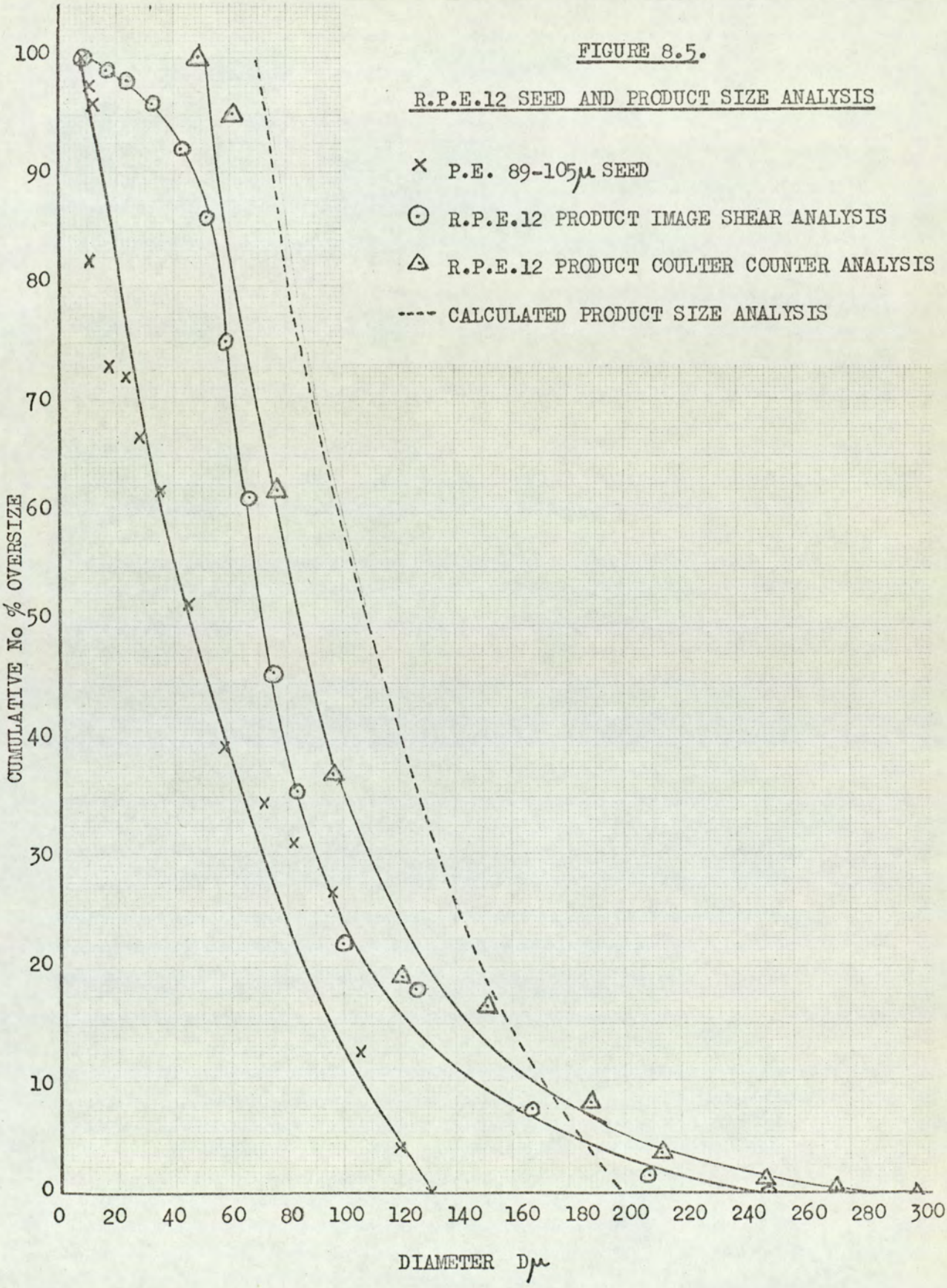
Some large agglomerates are formed which were also indicated in the size analyses (figure 8.5). As the analysis is on a number % oversize basis, agglomerates deplete the number % of the smaller sizes and might account for the deviation from the calculated product size analysis. Although agglomerates look well formed as though a result of growth, agglomeration might also occur in the filtration process. However there was no method of distinguishing between these possibilities.

For an absolute experimental proof of the method of computation of results runs were carried out at different initial supersaturations and with different amounts of seed, at 60°C. However, results did not concur at equivalent supersaturations: faster growth rates being indicated with higher initial supersaturations and also with smaller amounts of seed. At first this was attributed to a surface roughness effect proportional to the mass deposited per unit surface area, but examination of the products shows it in fact to be a result of attrition. It appears that there is a critical time or size increase (for a particular growth velocity) beyond which the crystals grown become too fragile for the particular hydrodynamic conditions, resulting in attrition. The products of some of these experiments with size analyses shown in table 28 are shown in figures 8.3 e to j. It was therefore concluded necessary to limit initial supersaturations to ca. $\Delta c \simeq 4\%$ and use at least 2 g seed.

R.E.16 was carried out with the addition of 0.5 g Zn particles of about 3 μ average size and resulted in a 20% increase in growth rate . Examination of the product, figure 8.3 g, showed these particles to be integrated into the crystals. It was thought

FIGURE 8.5.

R.P.E.12 SEED AND PRODUCT SIZE ANALYSIS



that this growth rate enhancement was a result of an increased number of adsorption sites caused by the particles since the particles did not themselves create nucleation. The effect of filtering solutions through a 0.45 μ filter (the finest available) was therefore examined. Results were anomalous and showed a poor reproducibility. Examination of R.E.F.4 product, figure 8.31, table 27, showed this to be due to attrition. It was concluded that brittle crystals were caused by growth free of heterogeneous particles.

8.3.2.2. Batches B, F and G.

These three materials were all found to show crystal growth inhibition far greater than that expected due to the known impurities indicated by the gas chromatographic analysis. The conclusive proof of an unknown contaminant was made by comparison of P.G. and P.E. materials (i.e. purified of known impurities) when P.G. was found to be greatly inhibited. Attempts to neutralise the contaminant were unsuccessful and although benzene was found to extract a certain amount of the contaminant the most efficient extraction was made using Molecular Sieve Type 5X. An estimation of the contaminant content made by burning off the molecular sieve extract and finding the loss in weight with six successive extractions indicated an original contaminant concentration of 0.26% in Batch P.G., and 0.0329 p.p.m. contaminant left in solution after the sixth successive extraction. However even after these 6 extractions, experiment 97 showed complete growth inhibition with $\Delta c = 1.2\%$ at 70°C whereas purified Batch E grew at a reasonable rate under these same conditions. A comparison

of purified Batch A with purified Batch E, indicates that Batch A was also contaminated, and it seems fortuitous that these materials (A, B, F and G) all contain $< 0.1\%$ Di-P.E. It was thought possible that the contaminant might inhibit this side reaction for the formation of Di-P.E. and therefore be present early in the manufacturing process.

The nature of this contaminant is still unknown. Although it was found in the previous work (1) that a trace of oil completely inhibited crystal growth, P.E. has since been found to be hypersensitive to other impurities. Further growth inhibition was found (a) when the material was sieved through a sieve which had been used for a SiO_2/V_2O_5 catalyst, and (b) when a polythene bucket was used in the preparation of pure P.E.

As the exact amount of contaminant was unknown for these materials, growth rates obtained were only relative and were not studied in detail.

8.3.2.3. Batches C. D and E.

These materials had reasonably comparable chemical analyses. The growth rates obtained were of the same order and the differences could possibly be attributed to the differences in the known impurity contents. The results were fitted to the correlation $g = k_L s^b$ by the method of least mean squares, and the following results obtained:-

BATCH D:

Run No.	T_o °C	Corrected Temp. °C.	k_L	b
R. D. 1	70.0	70.2	0.00161	2.386
R.D.1 + 3 + 4 + 6	70.0	70.2	0.00126	2.266
R. D. 7	70.0	70.2	0.0127	2.723

BATCH C:

Run No.	T_o °C	Corrected Temp. °C.	k_L	b
R. C. 6	60.0	60.1	0.000847	2.851
R. C. 7	50.0	50.1	0.0000325	1.814
R. C.14	70.0	70.2	0.00193	2.261
R. C.15	30.0	30.0	0.00000267	1.438
R. C.17	40.0	40.0	0.00000541	1.497

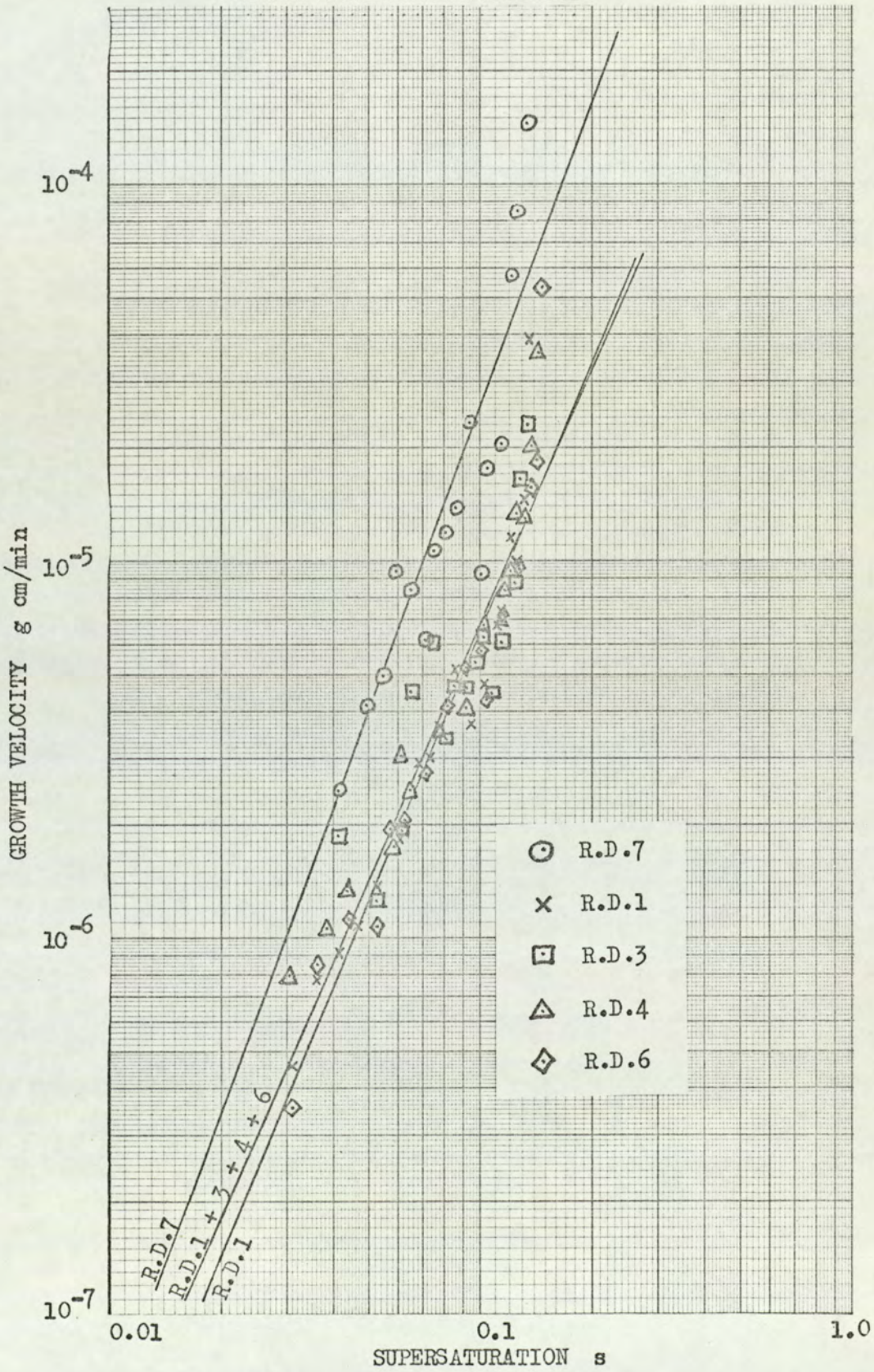
BATCH E:

Run No.	T_o °C	Corrected Temp. °C.	k_L	b
R. E. 4	60.0	60.1	0.000194	1.908
R. E.19	70.0	70.2	0.000623	1.693
R. E.38	50.0	50.1	0.0000403	1.696

R.D.1. with Batch D 44 - 64 μ seed at 70°C showed a comparable growth rate with R.D.3, 4 and 6 using different seed size and Batches, and carried out in different cells. The results are

FIGURE 8.6

BATCH D GROWTH RATE AT 70°C



shown in figure 8.6. The k_L values for R.D.1. and the combined tests R.D.1, 3, 4 and 6 compare better than appears at first sight, e.g. g values calculated from the given correlations at s = 0.1 are 6.6×10^{-6} and 6.8×10^{-6} respectively. The exaggerated k_L difference is due to its sensitivity to the exponent b. For better accuracy a wider supersaturation range should be used, but this is not possible for P.E. without causing attrition of seed crystals or creating nucleation.

R.D.7 with P.G. seed is also shown for comparison but has a very much higher growth rate than with the impure seed materials. This phenomenon of enhanced growth using pure seed with impure solution was also found with Batch G solution (experiments 54 and 55). It could possibly be due to both depleted impurity content in the solution due to the impurity diffusion into the crystal to achieve equilibrium and this causing dislocations in the crystals.

Growth results of Batch C over the temperature range 30°C to 70°C are shown in figure 8.7 together with the least mean squares correlations. With the exception of R.C.6 at 60°C the exponent b decreases progressively with temperature. This could possibly be due to a contaminant having a different equilibrium partition coefficient at different temperatures. Batch C however showed a similar growth rate to Batch E which was thought to be contaminant free, as extraction with molecular sieve had shown no effect on the growth rate. The growth results for Batch E are shown in figure 8.8. The sensitivity of the k_L value on b is further demonstrated by a comparison of Batches C, D and E at 70°C. Although Batch E has the

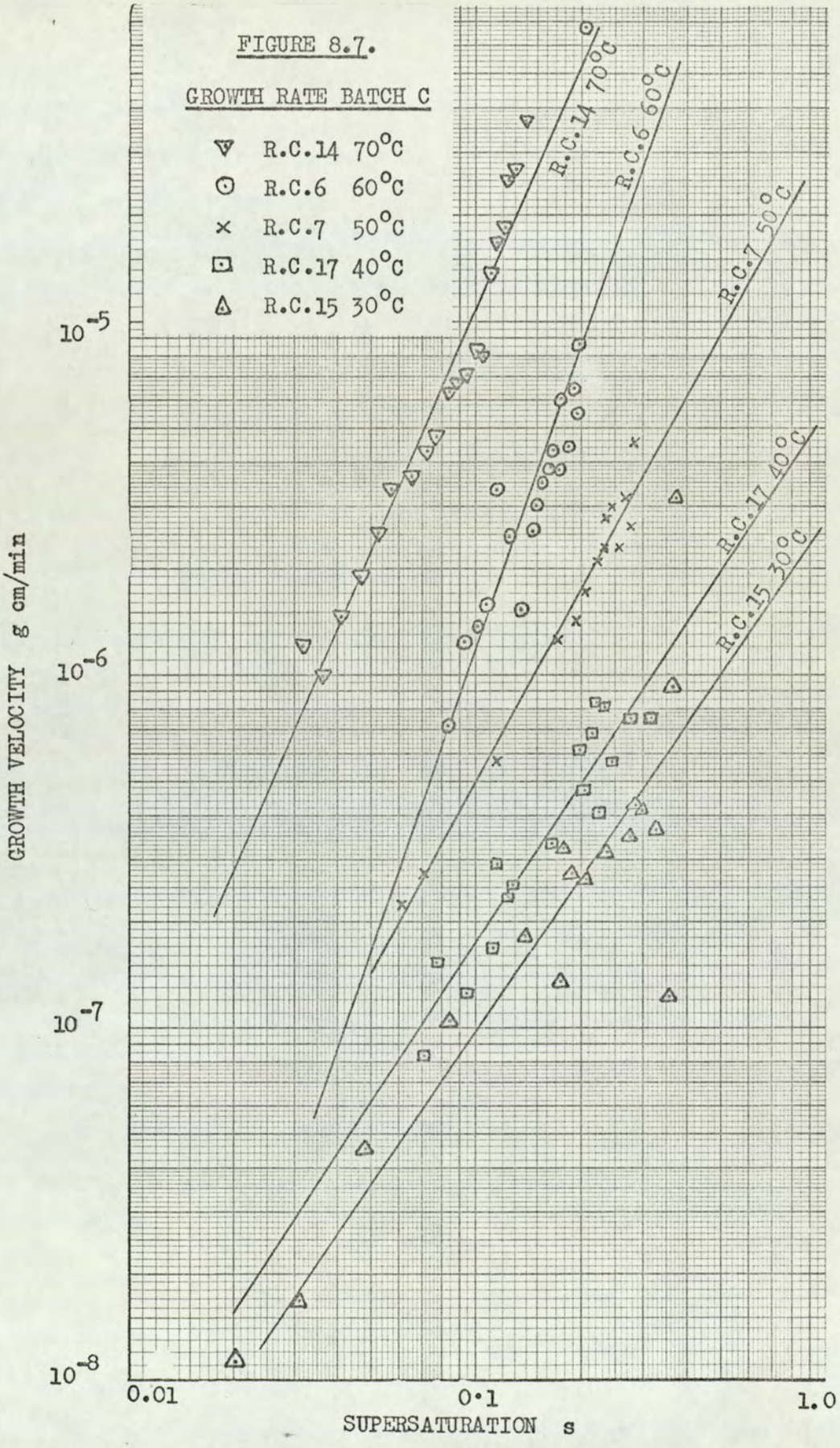


FIGURE 8.7.
GROWTH RATE BATCH C
R.C.14 70°C
R.C.6 60°C
R.C.7 50°C
R.C.17 40°C
R.C.15 30°C
R.C.14 70°C
R.C.6 60°C
R.C.7 50°C
R.C.17 40°C
R.C.15 30°C
GROWTH VELOCITY g cm/min
10-5
10-6
10-7
10-8
0.01
0·1
1.0
SUPERSATURATION s

FIGURE 8.8.

BATCH E GROWTH RATE

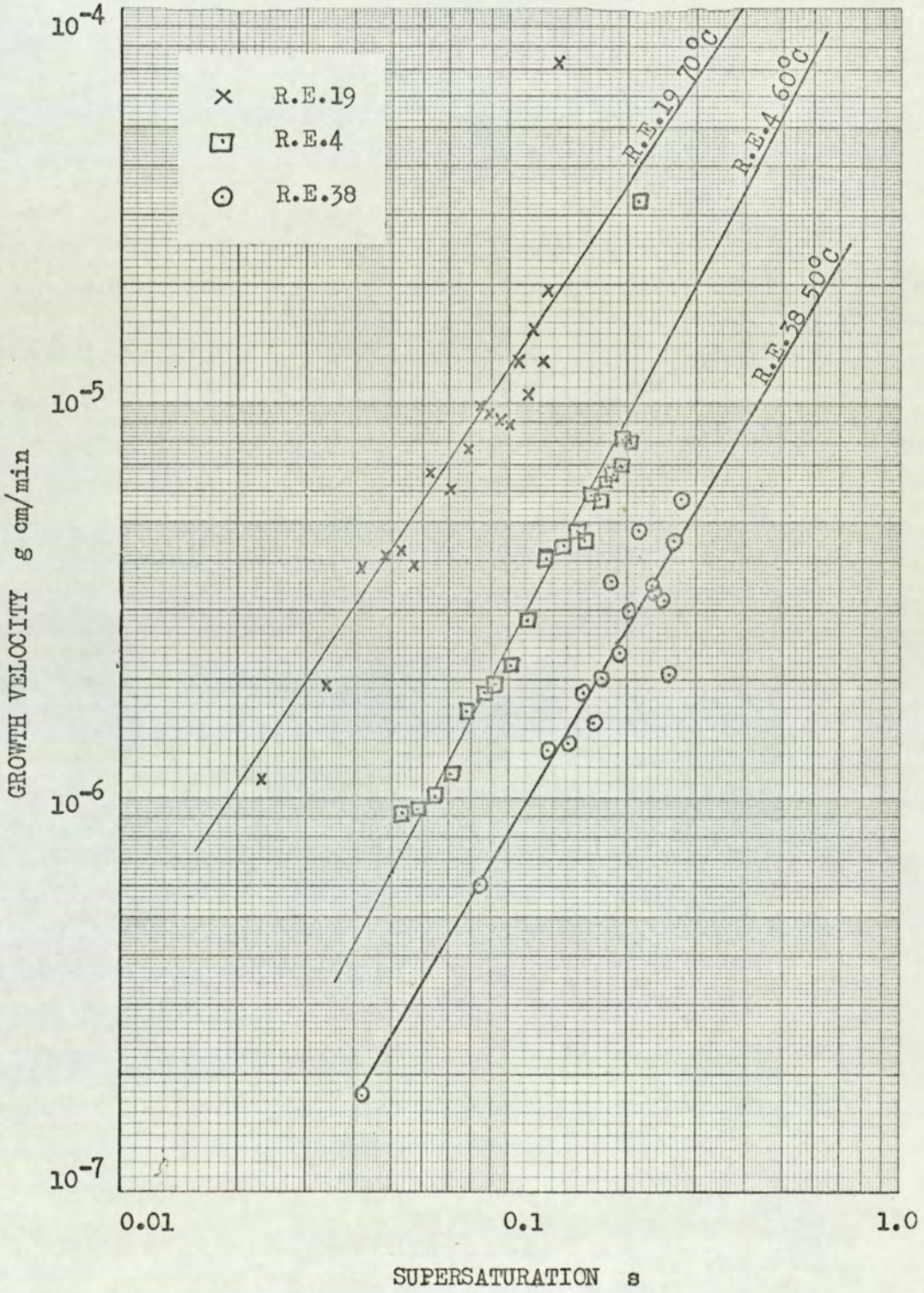
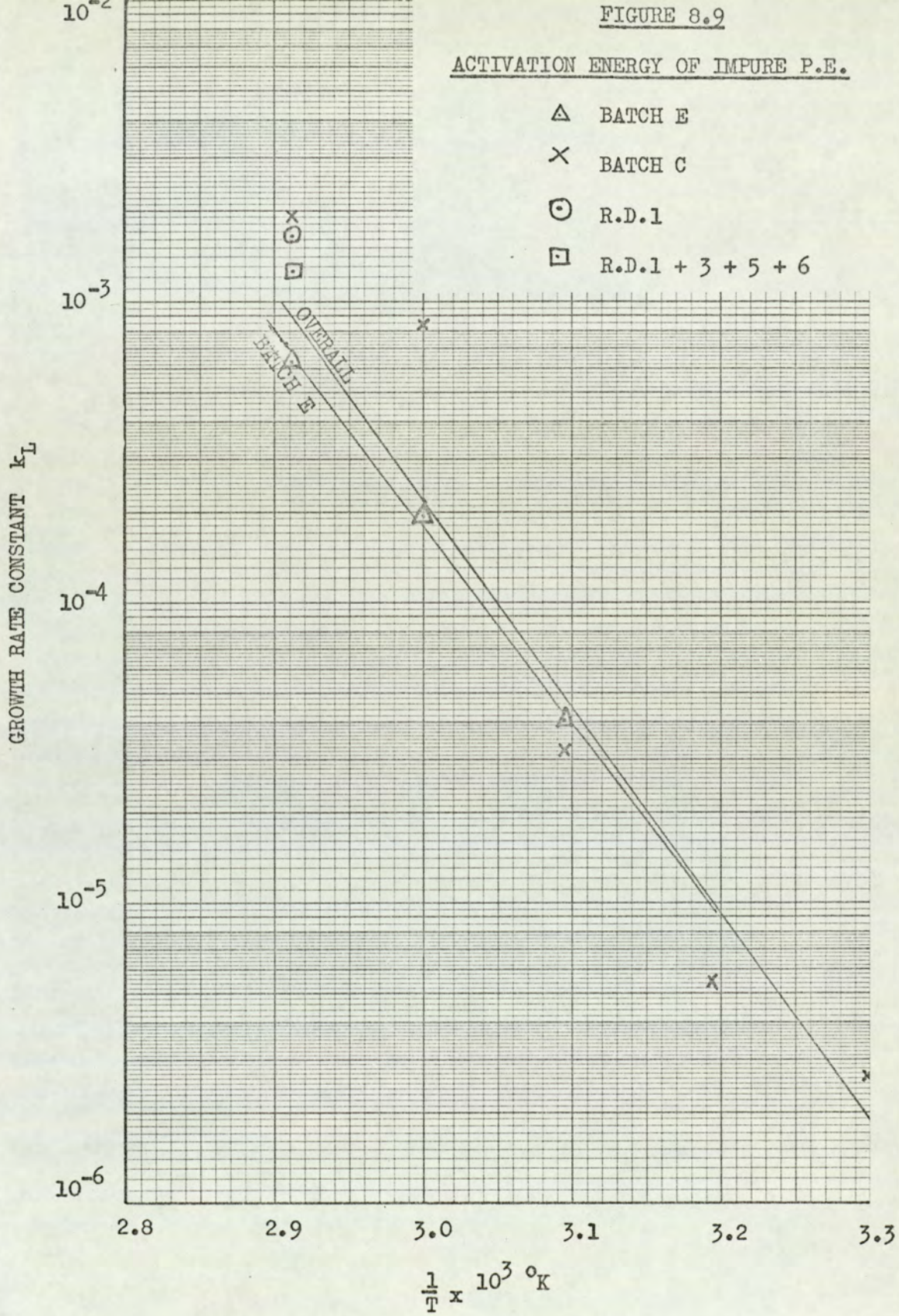


FIGURE 8.9

ACTIVATION ENERGY OF IMPURE P.E.



lowest k_L value, growth rates over the supersaturation range measured were in fact faster than either Batch C or D. k_L values have been plotted vs $\frac{1}{T}$ where T = degrees Kelvin, in figure 8.9. Batch E can be correlated for $50 > T_o > 70^{\circ}C$ by the equation:

$$\log_{10} k_L = 14.07 - \frac{5938}{T}$$

with an Activation Energy of 27.2 Kcal./g.mole., and an overall equation for Batches C. D and E for $30 > T_o > 70^{\circ}C$ is:

$$\log_{10} k_L = 17.04 - \frac{6892}{T}$$

with an Activation Energy of 31.5 Kcal./g.mole.

The deviation of the points from this latter correlation is large, but if a contaminant is present having a changing partition coefficient with temperature this would make the simple Arrhenius type correlation invalid.

8.3.2. 4. Purified Batch E.

Batch E with the highest growth rate of the impure Batches was thought to have the least (if any) contaminant, and crystal growth rates for the purified material were determined. The results obtained were unexpected. Figure 8.10 shows the results of R.P.E.7 $(\Delta c = 2.6\%)$ and R.P.E.20 $(\Delta c = 3.8\%)$ both at 70°C. Both show an apparent break point in the correlation with the growth rates obtained displaced from each other. R.P.E.18 at 75°C (figure 8.11) also suggests this apparent break point, but it is not present at the lower temperatures of 60°C and 50°C. R.P.E.17 and

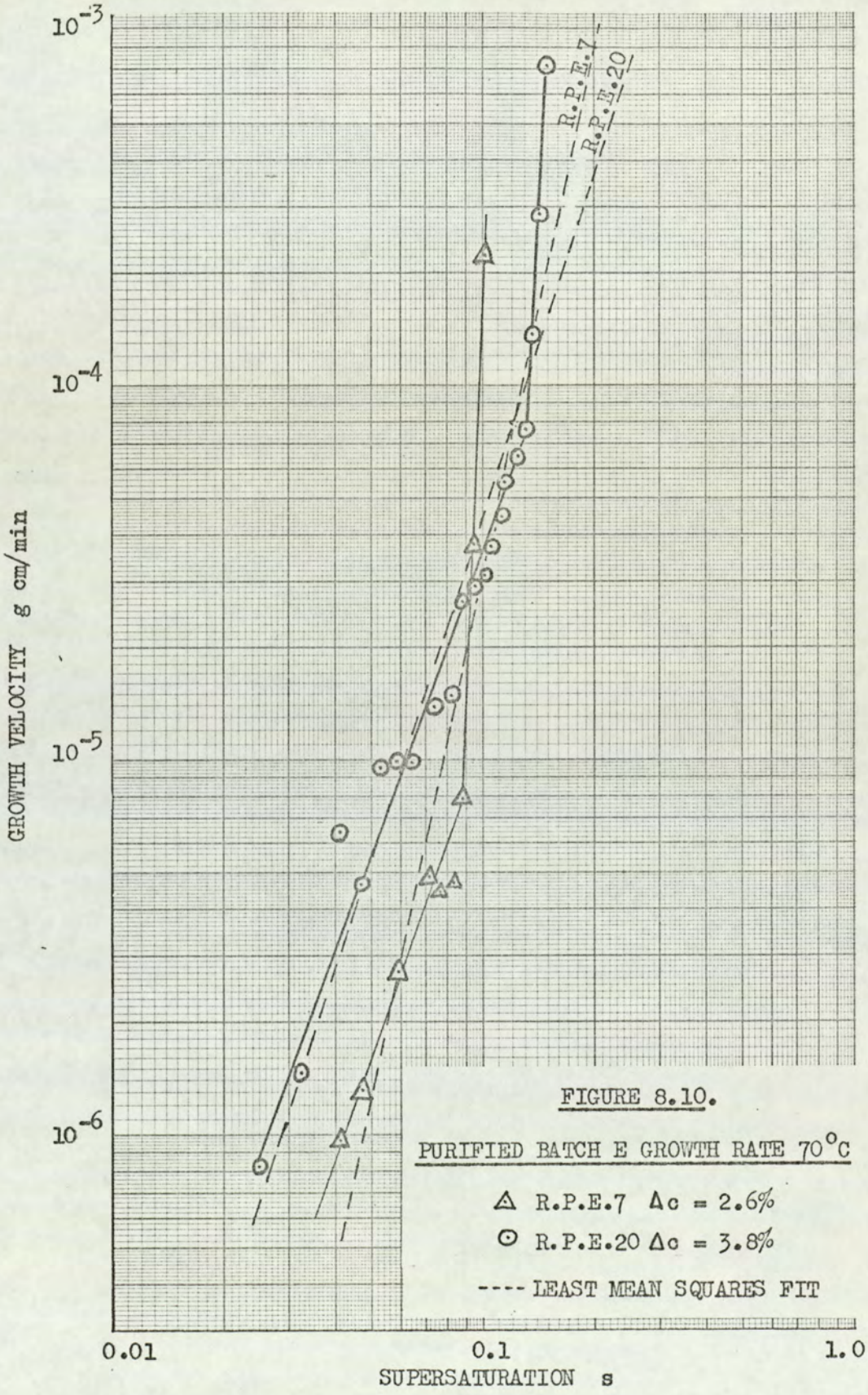


FIGURE 8.10.

PURIFIED BATCH E GROWTH RATE 70°C

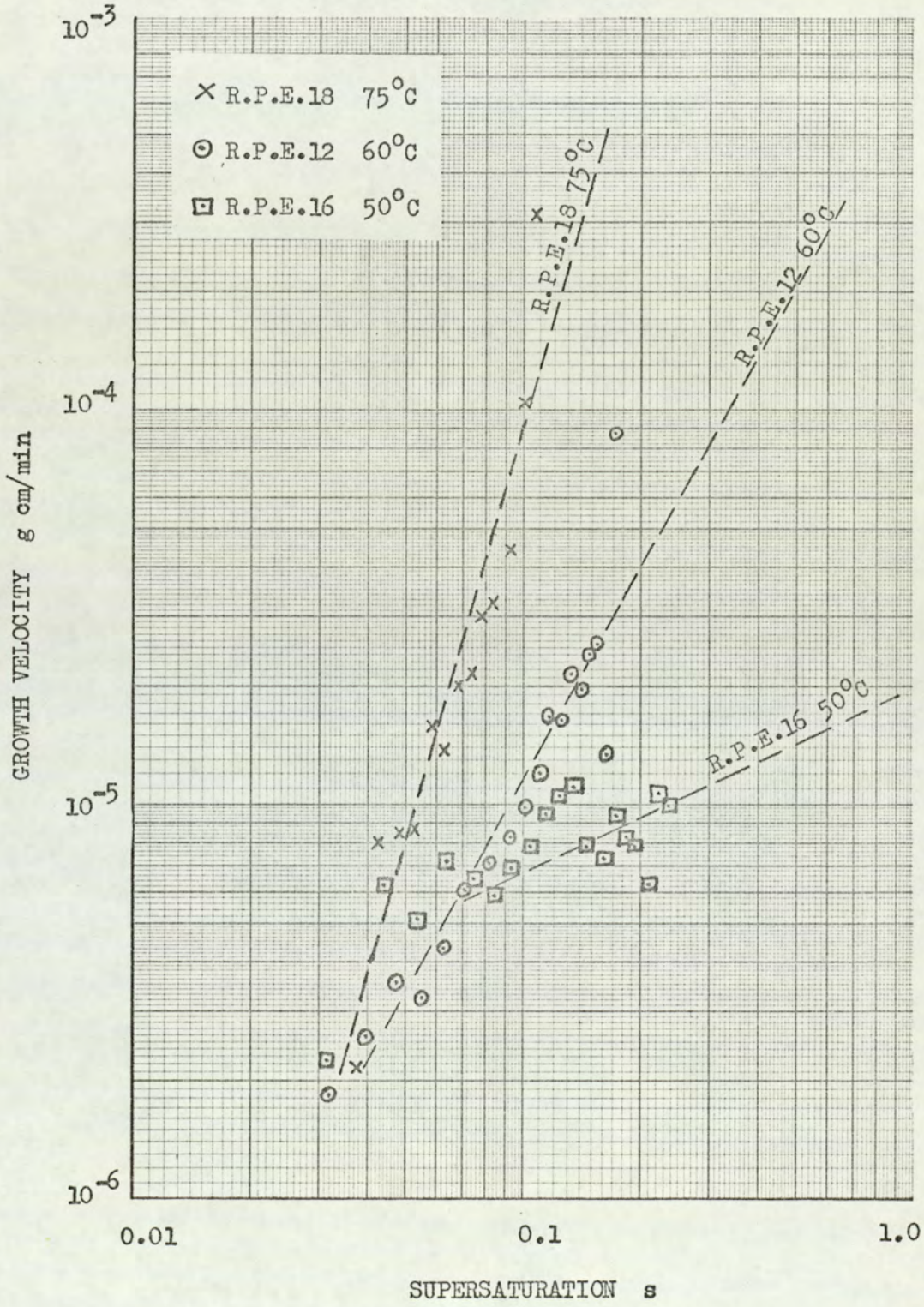
△ R.P.E.7 Δc = 2.6%

⊙ R.P.E.20 Δc = 3.8%

---- LEAST MEAN SQUARES FIT

FIGURE 8.11.

PURIFIED BATCH E GROWTH RATE



the repeat experiment R.P.E.19, at 40°C (figure 8.12) both show an increase in crystal growth rate with decreasing supersaturation to a maximum value (higher than that at 60°C for an equivalent supersaturation) before decreasing. This growth rate after achieving the maximum value then remains higher than equivalent values at 60°C.

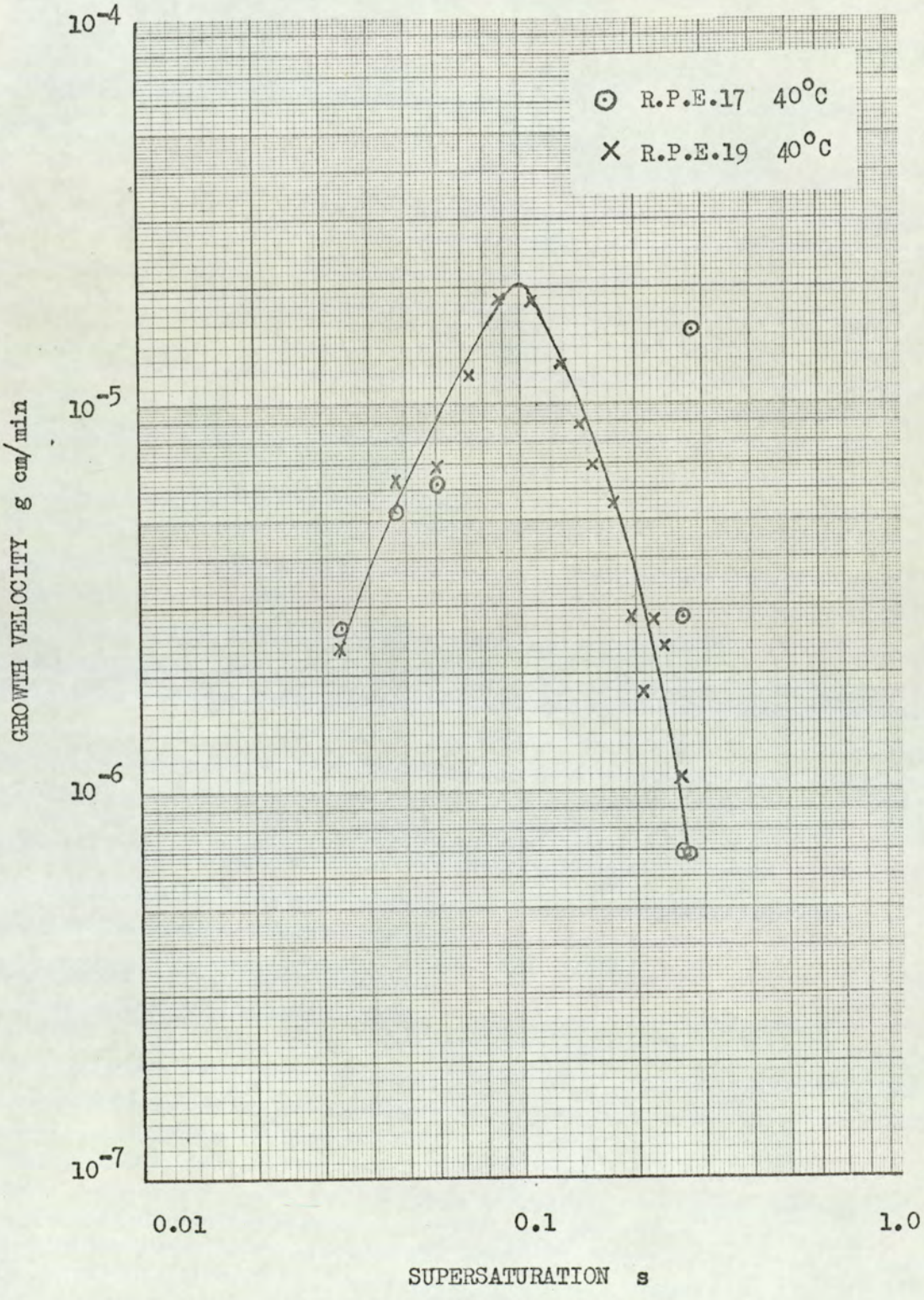
The only explanation thought possible is that of a contaminant effect. Any roughness effect to explain the break point at the higher temperatures (i.e. by achieving a constant roughness) would be expected to be consistent at all temperatures. If it is assumed however that the system contains a contaminant which will achieve an equilibrium partition between the crystals in suspension and the solution, then:

(a) At high temperatures the break point might be explained by the contaminant gradually being adsorbed on the crystal surface and rapidly decreasing the crystal growth rate by blocking adsorption sites until (at the break point) contaminant equilibrium is achieved when the growth will proceed at a slower rate than in the absence of contaminant.

(b) At low temperatures (e.g. 40°C) the maximum value might be explained by assuming a different contaminant partition coefficient whereby all the contaminant was soon adsorbed on the crystal surface almost completely inhibiting growth initially and then as growth proceeded the inhibiting regions gradually became covered by layers of purer crystal. Eventually at the maximum value the solution was purified of contaminant and growth proceeded from pure solution onto pure crystal surface.

FIGURE 8.12.

PURIFIED BATCH E GROWTH RATE AT 40°C



It is thought that the reason this contaminant effect was not so peculiar for the impure solution (Batch E) was because of the greater relative number of adsorption sites. It can be seen from examination of the product crystals figure 8.3b and d that the surfaces of pure crystals are far smoother than those of impure crystals. It was therefore thought that although the same contaminant partition coefficient exists, there were many more adsorption sites with impure P.E. crystals and the percentage effected by contaminant was less and therefore did not have such a pronounced effect on the growth rate.

The results of k_L for $75 > T_o \geq 50^{\circ}C$ obtained from all the data points for each experiment are shown below:

Run No.	T_o °C	Corrected Temp. °C.	k_L	b
R.P.E. 7	70.0	70.2	1.066	4.528
R.P.E.12	60.0	60.1	0.000674	1.774
R.P.E.16	50.0	50.1	0.0000195	0.449
R.P.E.18	75.0	75.3	0.300	3.530
R.P.E.20	70.0	70.2	0.0457	3.002

Although the contaminant markedly effects the above correlations these overall k_L values have been plotted vs $\frac{1}{T}$ (figure 8.13) to give an indication of the activation energy of the pure P.E. in the presence of this trace amount of contaminant. The results can be correlated by:

$$\log_{10} k_L = 54.0 - \frac{19030}{T}$$

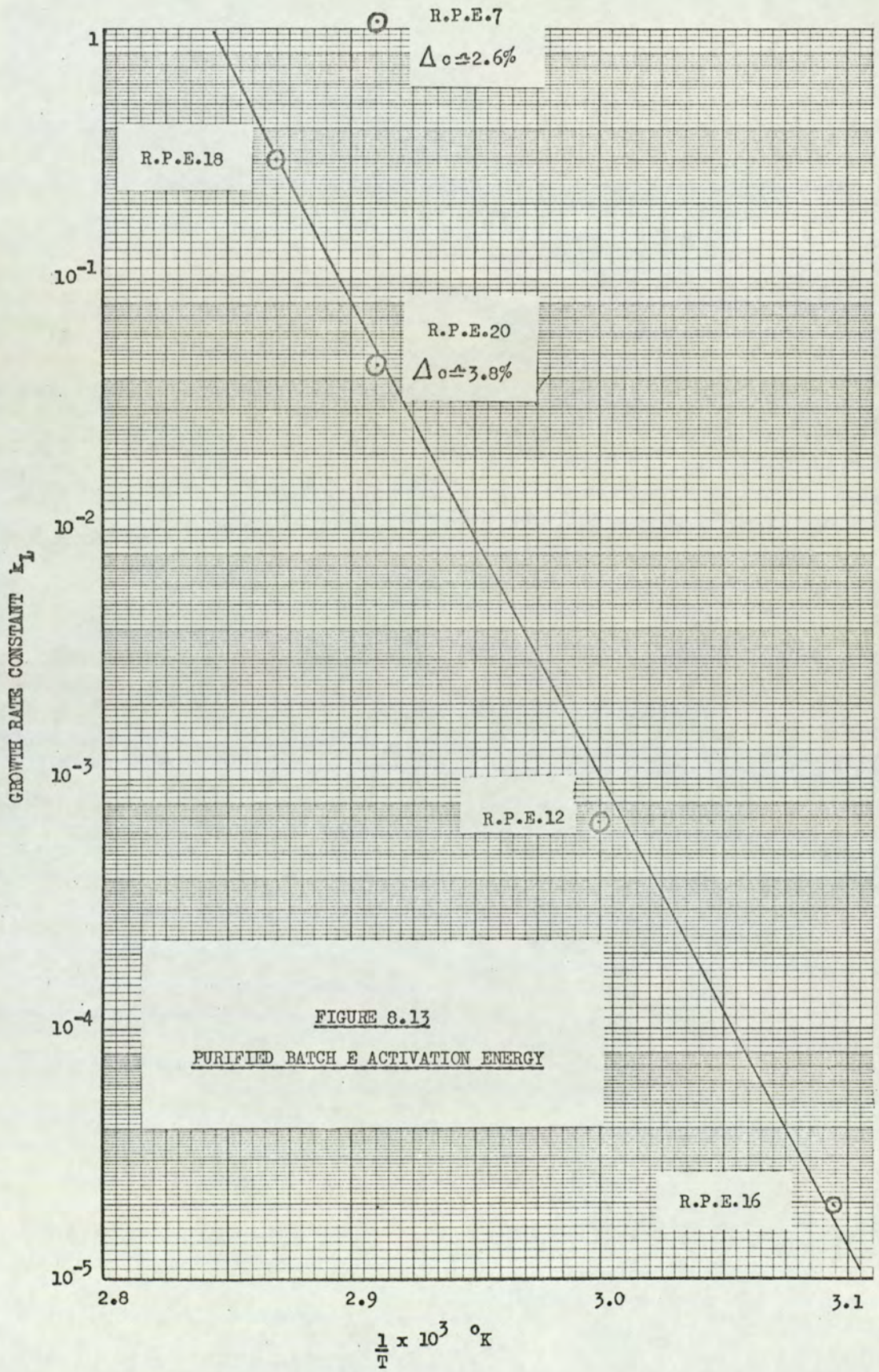


FIGURE 8.13

PURIFIED BATCH E ACTIVATION ENERGY

indicating an activation energy for growth of 87.0 Kcal./g.mole.

8.4. Suggestions for Future Work.

Because of the overlap phenomenon observed with the dissolution experiments it would be interesting to investigate further the cause of this initially enhanced concentration. It has been suggested that it might be due to the method of preparation of the crystals used in the test, producing a non-homogeneous impurity distribution. The effect of isothermal batch preparations of crystals at different temperatures before dissolution could be investigated to find if there is a partition coefficient effect. It would also be interesting to check the effect after annealing the crystals to induce diffusion of the impurity within the crystal lattice and so create homogeneous composition.

The effect of temperature on the partition coefficient of the known impurities Formal and Di-P.E. should be studied, particularly below 50°C, to see if this could explain the enhanced solubility of the impure material at the low temperatures above that suggested by the correlation (for above 50°C). Equilibrium should be approached from dissolution with only a small excess of solute present.

In order to obtain further information on the growth rate of P.E. and to correlate the data obtained in this work which is at present only related to the particular Batches of material with suspected unknown quantities of an unknown contaminant, this unknown contaminant should be identified, analysed and removed in order that

realistic growth rates might be obtained. Although extraction with molecular sieve type 13X was found effective for higher contaminant concentrations, it had no apparent effect with the extremely small concentrations thought to be influencing the crystal growth of the "purest" material (i.e. P.E.). Purification might possibly be achieved by repeated extraction with redistilled Analar benzene. The only method for the determination of the presence of the contaminant resulting from this work is by comparison of the crystal growth rates. As it appears that the effective contaminant concentration is $<$ 1 p.p.m. quantitative analysis might prove difficult. However as it has such a pronounced effect on crystal growth it is possible that the effect on the surface energy of the system would be large. It is therefore possible that a simple quick test might be devised studying the effect on the surface energy by measuring the contact angle of the solution/crystal interface.

Although a preliminary investigation with Batch E at 60°C to find the safe working limits showed that one condition necessary to avoid attrition was an initial supersaturation of $(c - c_\infty) < 4\%$ with 2 g of seed, this should be studied in more detail. Each material with associated impurities ought to be studied individually, as the impurities could be expected to effect the brittleness of the crystal. Each system could then be examined to find the effect of time, supersaturation and the amount of seed on the attrition occuring during the process. If carried out at near room temperature samples of suspension could be taken periodically and analysed with the Coulter

Counter. Any deviations from the mathematical model used in this work, assuming simply growth of the seed according to the ΔL law could then be predicted under any growth rate conditions. Another parameter which appears to effect crystal strength is the amount of heterogeneous material present. It was found in this work that crystals grown from a solution free of heterogeneous particles $> 0.45\ \mu$ were abnormally brittle. Although the safe working limits to avoid attrition are known and were observed for these batch crystallisations, the strength or hardness characteristics of crystals grown under any conditions should be known before operating conditions of a crystalliser can be specified.

It has also been shown that heterogeneous particles enhance crystal growth, possibly by creating dislocations in the crystal surface. The effect of the number and size of heterogeneous particles on the degree of enhancement of the growth rate should be studied in more detail.

If the contaminant can be removed the original problem may be approached, that is the effect of the two individual main impurities on the growth rate of P.E., the evaluation of the optimum crystalliser conditions and the testing of these conditions on a pilot plant crystalliser.

CONCLUSIONS

1. The immersion refractometer used with scale S was correlated for impure P.E. solution (concentration c % m/v) in the metastable zone by the equation:

$$S = 20.365 + 3.5622\,c + 0.0002313\,c^2 - 0.16974\,T_o - 0.0035888\,T_o^2$$

where T_o is the observed temperature of a partially immersed thermometer ^{o}C.

2. The isothermal correlations for pure and impure solutions were well represented by the equation $c = F + BS + GS^2$ with an average standard deviation of ca. $\pm$ 0.04(% m/v) and are shown in tables 3 and 2 respectively, appendix A.

3. The equilibrium of Pure P.E. in aqueous solution between $30^{o}C$ and $75^{o}C$ is given by the equation:

$$\log_{10} x = 4.980 - \frac{1242}{T}$$

where x = P.E. mass %; T = degrees Kelvin.

4. The equilibrium of impure P.E. (with ca. 5% Formal) in aqueous solution $\geq 50^{o}C$ with solids present equivalent to a supersaturation of about $\left(c - c_{\infty}\right) \simeq 4\%$ is given by the equation:

$$\log_{10} x = 5.073 - \frac{1265}{T}$$

5. Below 50°C the solubility of impure P.E. is greater than that expected from the above correlation.

6. The presence of the Formal impurity in P.E. enhances the solubility.

7. Di-P.E. does not appear to affect the equilibrium value at the concentrations used but with the materials used it accelerated the rate of attainment of equilibrium from dissolution.

8. A correction was necessary to the Coulter Counter Theory to allow for particle shape and size of a P.E. crystal during size analysis. This correction amounted to about 5% of the equivalent spherical volume diameter at the recommended 40% particle/orifice diameter ratio limit with a 280 μ orifice tube.

9. Computer program 1 may be used to predict the product size analysis of crystals of a known seed size distribution, by measuring the mass increase when grown at constant supersaturation and when suface integration rate controls.

10. Computer program 2 may be used to calculate the crystal growth rate of seed crystals of known size distribution, grown in a batch crystalliser, by following the decrease in solution concentration with time.

11. The surface integration rate was crystal growth rate controlling for the conditions studied (i.e. $T_o < 70°C$).

12. Heterogeneous particles enhanced crystal growth rate.

13. The absence of heterogeneous particles for the crystallisation of P.E. results in brittle crystals.

14. P.E. material Batches A, B, F and G were contaminated with an unknown impurity not analysed on the gas chromatograph, which inhibited crystal growth.

15. This contaminant could be partially extracted using Molecular Sieve Type 13X. The total concentration of contaminant in Batch G was estimated to be 0.26%.

16. Correlations of the type $g = k_L s^b$ were fitted to the results of Batches C, D and E and the exponent b was found to vary according to material and temperature, with an average value of about 2.

17. k_L values for Batch E, $50^\circ C > T_o > 70^\circ C$ were correlated by:

$$\log_{10} k_L = 14.07 - \frac{5938}{T}$$

with an activation energy of 27.2 Kcal./g.mole.

18. The overall correlation for Batches C, D and E $30^\circ C > T_o > 70^\circ C$ was:

$$\log_{10} k_L = 17.04 - \frac{6892}{T}$$

with an activation energy of 31.5 Kcal./g.mole.

19. Purified Batch E, thought to be the purest material used,

showed unusual growth rates with respect to supersaturation. At high temperatures there is an apparent break in the log g vs log s correlation with two apparent straight lines of different slopes b.

At low temperatures the growth rate increased to a maximum for decreasing values of supersaturation before decreasing on further decrease of supersaturation with an enhanced growth rate to that anticipated. These phenomena were attributed to the presence of a trace amount of contaminant (< 0.01 p.p.m.) and a changing contaminant partition coefficient with temperature.

20. An average overall correlation for purified Batch E of the type $g = k_L s^b$ was found at each temperature $\geq 50^{\circ}C$ and the k_L values obtained correlated by the equation

$$\log_{10} k_L = 54.0 - \frac{19030}{T}$$

indicating an activation energy of 87.0 Kcal / g.mole.

APPENDIX A - CALIBRATION AND EQUILIBRIUM

FIGURE 5.1.

TEMPERATURE CORRECTION FOR PARTIALLY IMMERSED THERMOMETERS

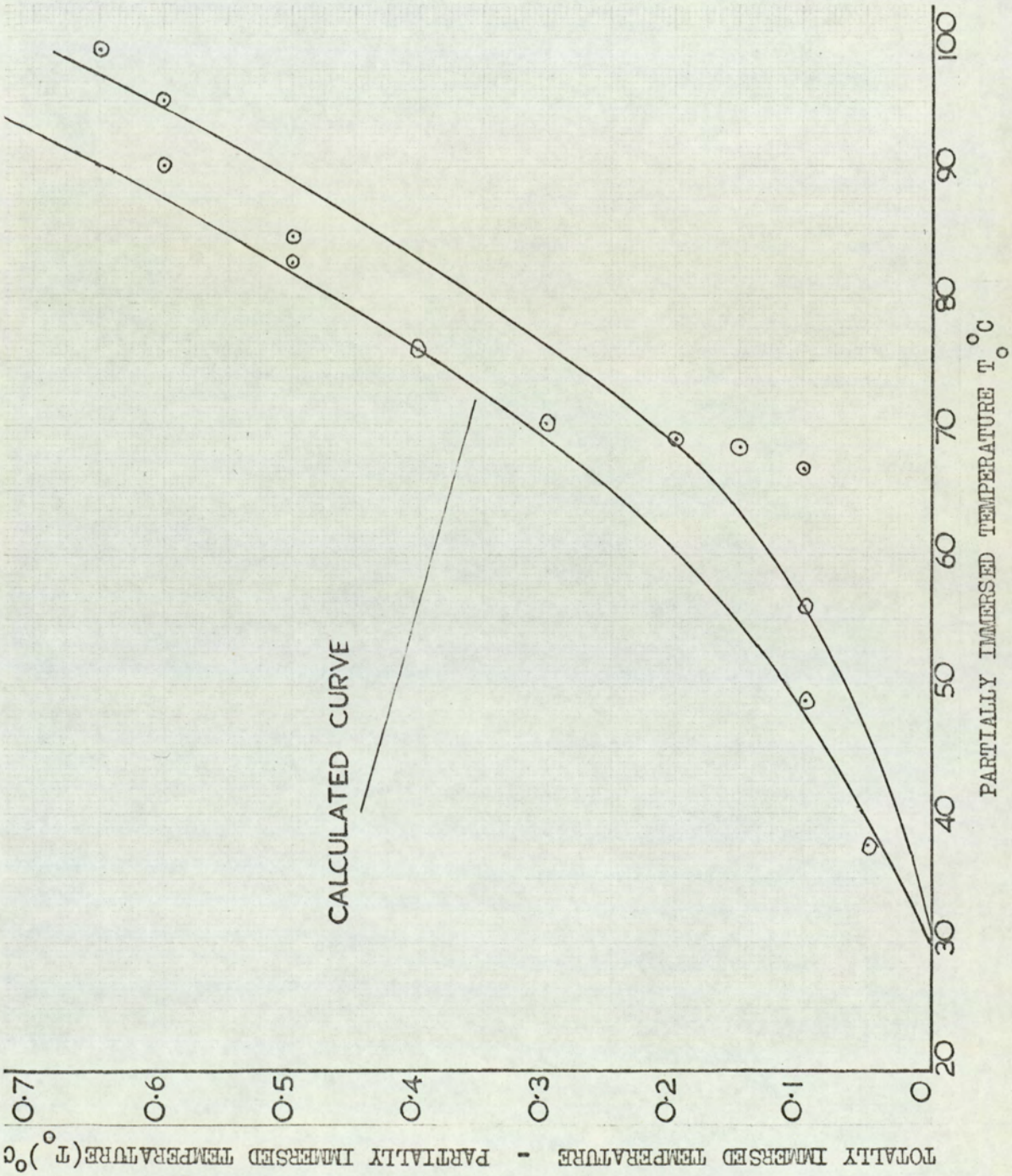


TABLE: 1

MANUFACTURER'S CHROMATOGRAPHIC CHEMICAL ANALYSIS

MATERIAL BATCH	DI-PENTAERYTHRITOL %	FORMAL %*
A	<0.1	4.73
B	<0.1	4.98
C	1.0	4.3
D	1.0	5.5
E	0.9	5.2
F	< 0.1	5.5
G	NONE	5.2
DiPE		4.0
PURIFIED MATERIALS	<0.1	<0.1

* COMPARATIVE ONLY (NOT MASS %)

TABLE: 2

ISOTHERMAL REFRACTOMETER CALIBRATION CORRELATIONS

BATCH D : 1.0% Di-P.E., 5.5% FORMAL

T, °C	LEAST MEAN SQUARES FIT c = % m/v	STANDARD DEVIATION
20.0	$c = -4.194 + 0.2782S$	± 0.044
20.0	$c = -4.124 + 0.2754S + 0.0000247S^2$	± 0.044
25.0	$c = -3.727 + 0.2779S$	± 0.060
25.0	$c = -4.095 + 0.2917S - 0.0001186S^2$	± 0.048
30.0	$c = -3.346 + 0.2800S$	± 0.058
30.0	$c = -3.743 + 0.2951S - 0.0001337S^2$	± 0.042
35.0	$c = -2.876 + 0.2813S$	± 0.055
35.0	$c = -3.208 + 0.2940S - 0.0001070S^2$	± 0.037
40.0	$c = -2.330 + 0.2820S$	± 0.055
40.0	$c = -2.635 + 0.2935S - 0.0000930S^2$	± 0.032
45.0	$c = -1.794 + 0.2836S$	± 0.064
45.0	$c = -2.116 + 0.2962S - 0.0001065S^2$	± 0.037
50.0	$c = -1.168 + 0.2844S$	± 0.069
50.0	$c = -1.479 + 0.2966S - 0.0000990S^2$	± 0.036
55.0	$c = -0.499 + 0.2858S$	± 0.072
55.0	$c = -0.918 + 0.3010S - 0.0001204S^2$	± 0.043
60.0	$c = 0.234 + 0.2865S$	± 0.093
60.0	$c = -0.299 + 0.3058S - 0.0001482S^2$	± 0.036
65.0	$c = 0.983 + 0.2879S$	± 0.099
65.0	$c = 0.473 + 0.3075S - 0.0001576S^2$	± 0.038
70.0	$c = 1.758 + 0.2895S$	± 0.092
70.0	$c = 1.319 + 0.3075S - 0.0001529S^2$	± 0.030
75.0	$c = 2.714 + 0.2889S$	± 0.089
75.0	$c = 2.222 + 0.3068S - 0.0001387S^2$	± 0.041

TABLE:3

ISOTHERMAL REFRACTOMETER CALIBRATION CORRELATIONS

PURE PENTAERYTHRITOL

T_o °C	LEAST MEAN SQUARES FIT	STANDARD DEVIATION
40.0	$c = -2.437 + 0.2852S$	± 0.064
40.0	$c = -2.935 + 0.3072S - 0.0002088S^2$	± 0.864
45.0	$c = -1.930 + 0.2883S$	± 0.063
45.0	$c = -2.343 + 0.3076S - 0.0001918S^2$	± 0.017
50.0	$c = -1.316 + 0.2895S$	± 0.045
50.0	$c = -1.643 + 0.3058S - 0.0001699S^2$	± 0.004
55.0	$c = -0.549 + 0.2890S$	± 0.001
60.0	$c = 0.116 + 0.2907S$	± 0.001
65.0	$c = 0.777 + 0.2932S$	± 0.027
65.0	$c = 0.558 + 0.3019S - 0.0000756S^2$	± 0.016
70.0	$c = 1.692 + 0.2927S$	± 0.028
70.0	$c = 1.820 + 0.2873S + 0.0000502S^2$	± 0.019
75.0	$c = 2.342 + 0.2971S$	± 0.030
75.0	$c = 2.400 + 0.2944S + 0.0000262S^2$	± 0.030

TABLE 4 : EQUIVALENT REFRACTOMETER SCALES

n_D	SCALE ZEROED 1A PRISM	SCALE ZEROED 1B PRISM	CALIBRATION SCALE S UNZEROED 1B (1B+1.20)	n_D	SCALE ZEROED 1A PRISM	SCALE ZEROED 1B PRISM	CALIBRATION SCALE S UNZEROED 1B (1B+1.20)
1.32548	-5.00	-4.65	-3.45	1.33633	23.00	23.25	24.45
1.32587	-4.00	-3.65	-2.45	1.33672	24.00	24.25	25.45
1.32626	-3.00	-2.70	-1.50	1.33710	25.00	25.25	26.45
1.32665	-2.00	-1.70	-0.50	1.33748	26.00	26.25	27.45
1.32704	-1.00	-0.70	0.50	1.33786	27.00	27.25	28.45
1.32744	0.00	0.30	1.50	1.33824	28.00	28.25	29.45
1.32783	1.00	1.30	2.50	1.33862	29.00	29.25	30.45
1.32822	2.00	2.30	3.50	1.33900	30.00	30.20	31.40
1.32861	3.00	3.30	4.50	1.33937	31.00	31.20	32.40
1.32900	4.00	4.30	5.50	1.33975	32.00	32.20	33.40
1.32939	5.00	5.30	6.50	1.34013	33.00	33.20	34.40
1.32978	6.00	6.30	7.50	1.34051	34.00	34.20	35.40
1.33016	7.00	7.25	8.45	1.34089	35.00	35.20	36.40
1.33055	8.00	8.25	9.45	1.34127	36.00	36.20	37.40
1.33094	9.00	9.25	10.45	1.34164	37.00	37.20	38.40
1.33133	10.00	10.25	11.45	1.34202	38.00	38.20	39.40
1.33171	11.00	11.25	12.45	1.34239	39.00	39.15	40.35
1.33210	12.00	12.25	13.45	1.34277	40.00	40.15	41.35
1.33249	13.00	13.35	14.45	1.34315	41.00	41.15	42.35
1.33287	14.00	14.25	15.45	1.34352	42.00	42.15	43.35
1.33326	15.00	15.25	16.45	1.34390	43.00	43.15	44.35
1.33364	16.00	16.25	17.45	1.34427	44.00	44.15	45.35
1.33403	17.00	17.25	18.45	1.34465	45.00	45.15	46.35
1.33441	18.00	18.25	19.45	1.34502	46.00	46.15	47.35
1.33480	19.00	19.25	20.45	1.34539	47.00	47.15	48.35
1.33518	20.00	20.25	21.45	1.34577	48.00	48.15	49.35
1.33556	21.00	21.25	22.45	1.34614	49.00	49.15	50.35
1.33595	22.00	22.25	23.45	1.34651	50.00	50.15	51.35

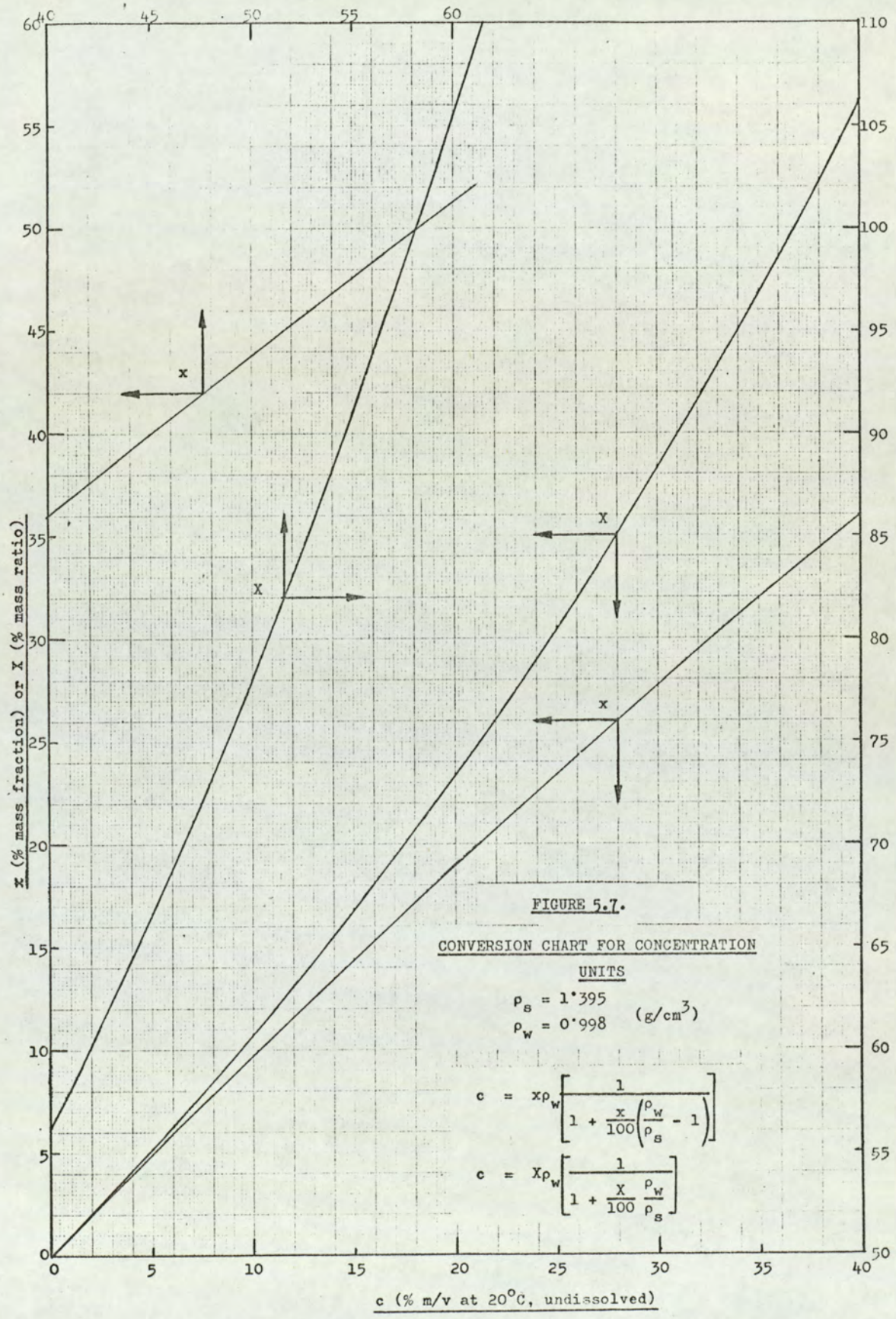


FIGURE 5.7.

CONVERSION CHART FOR CONCENTRATION UNITS

$\rho_s = 1{\cdot}395$

$\rho_w = 0{\cdot}998$ (g/cm^3)

$$c = x\rho_w\left[\frac{1}{1 + \frac{x}{100}\left(\frac{\rho_w}{\rho_s} - 1\right)}\right]$$

$$c = X\rho_w\left[\frac{1}{1 + \frac{X}{100}\,\frac{\rho_w}{\rho_s}}\right]$$

TABLE: 4 (CONTINUED)

n_D	SCALE ZEROED 1A PRISM	SCALE ZEROED 1B PRISM	CALIBRATION SCALE S UNZEROED 1B (1B+1.20)	n_D	SCALE ZEROED 1A PRISM	SCALE ZEROED 1B PRISM	CALIBRATION SCALE S UNZEROED 1B (1B+1.20)
1.34688	51.00	51.15	52.35	1.35712	79.00	79.05	80.25
1.34725	52.00	52.15	53.35	1.35748	80.00	80.05	81.25
1.34763	53.00	53.15	54.35	1.35784	81.00	81.05	82.25
1.34800	54.00	54.15	55.35	1.35820	82.00	82.05	83.25
1.34837	55.00	55.15	56.35	1.35856	83.00	83.05	84.25
1.34874	56.00	56.15	57.35	1.35892	84.00	84.05	85.25
1.34910	57.00	57.15	58.35	1.35928	85.00	85.05	86.25
1.34947	58.00	58.15	59.35	1.35964	86.00	86.05	87.25
1.34984	59.00	59.15	60.35	1.35999	87.00	87.05	88.25
1.35021	60.00	60.15	61.35	1.36035	88.00	88.05	89.25
1.35057	61.00	61.15	62.35	1.36070	89.00	89.05	90.25
1.35094	62.00	62.15	63.35	1.36106	90.00	90.05	91.25
1.35131	63.00	63.15	64.35	1.36141	91.00	91.05	92.25
1.35167	64.00	64.10	65.30	1.36177	92.00	92.05	93.25
1.35204	65.00	65.10	66.30	1.36212	93.00	93.00	94.20
1.35240	66.00	66.10	67.30	1.36248	94.00	94.00	95.20
1.35277	67.00	67.10	68.30	1.36283	95.00	95.00	96.20
1.35313	68.00	68.10	69.30	1.36318	96.00	96.00	97.20
1.35350	69.00	69.10	70.30	1.36353	97.00	97.00	98.20
1.35386	70.00	70.05	71.25	1.36389	98.00	98.00	99.20
1.35422	71.00	71.05	72.25	1.36424	99.00	99.00	100.20
1.35459	72.00	72.05	73.25	1.36459	100.00	100.00	101.20
1.35495	73.00	73.05	74.25	1.36494	101.00	101.00	102.20
1.35532	74.00	74.05	75.25	1.36529	102.00	102.00	103.20
1.35568	75.00	75.05	76.25	1.36565	103.00	103.00	104.20
1.35604	76.00	76.05	77.25	1.36600	104.00	104.00	105.20
1.35640	77.00	77.05	78.25	1.36635	105.00	105.00	
1.35676	78.00	78.05	79.25				

TABLE: 5

BATCHES D AND F DISSOLUTION TESTS

	BATCH D							BATCH F	
DISSOLUTION TEST No.:	D.D.4.	D.D.5.	D.D.6.	D.D.7.	D.D.14	D.D.9.	D.D.13	D.F.2.	D.F.3.
T_o (°C)	60.0	60.0	60.0	60.0	55.0	50.0	40.0	60.0	60.0
EQUIVALENT RUN INITIAL CONCN. c_o (%m/v)	27.50	24.5	26.0	23.0	21.5	24.0	15.25	24.0	27.5
P.E. = (c_o x 2.5 + SEED)g	70.75	63.25	67.0	59.5	55.75	62.0	40.12	62.0	70.75
$H_2O = 250 - \frac{2.50c_o}{1.396}$ cm^3	201	206	203.5	209	211.5	207.5	223	207	201
TOTAL MASS % (fraction)	26.05	23.5	24.75	22.15	20.9	23.0	15.3	23.0	26.05
MAXIMUM VALUE (SCALE S)	69.9	69.6	69.7	69.8	65.6	63.2	56.2	73.8	74.2
EQUILIBRIUM VALUE (SCALE S)	69.3	68.7	68.9	68.6	62.0	57.6	49.0	68.7	69.3
MAXIMUM CONCN. c(%m/v)	20.352	20.267	20.295	20.324	18.309	16.871	13.566	21.462	21.575
EQUILIBRIUM CONCN. c(%m/v)	20.181	20.010	20.067	19.981	17.281	15.277	11.602	20.010	20.181

APPENDIX B - SIZE ANALYSIS

TABLE: 6

BATCHES D AND F EQUILIBRIUM RESULTS

(OBTAINED FROM DISSOLUTION)

TEST	T_o °C	CORRECTED TEMP. °C	TOTAL P.E. CONCN. EQUIVALENT GROWTH RUN INITIAL CONCN %m/v	+SEED (g)	EQUILIBRIUM a %m/v	x MASS %
D.D.1.	30.0	30.0	12.5	2	9.124	8.911
D.D.10	40.0	40.0	14.0	2	11.552	11.205
D.D.13	40.0	40.0	15.25	2	11.602	11.253
D.D.12	40.0	40.0	17.5	2	11.751	11.393
⋕D.D.11	50.0	50.1	19.5	2	15.106	14.512
D.D.9.	50.0	50.1	24.0	2	15.277	14.668
⋕D.D.14	55.0	55.1	21.5	2	17.281	16.502
⋕D.F.2.	60.0	60.1	24.0	2	20.010	18.969
+	60.0	60.1	26.0	1	20.039	18.994
D.D.6.	60.0	60.1	26.0	2	20.067	19.019
+	60.0	60.1	27.5	0.5	20.096	19.045
+	60.0	60.1	27.5	1	20.153	19.095
D.D.4	60.0	60.1	27.5	2	20.181	19.122
D.F.3. D.D.3.	70.0	70.2	27.5	2	26.211	24.437
⋕D.D.2 D.F.1	70.0	70.2	30.0	2	26.311	24.523

+ INTERPOLATED VALUES FROM FIGURE 5.5.

LEAST MEAN SQUARES CORRELATION OF (⋕) RESULTS (FOR $T \geqslant 50^{\circ}C$ AND $(a-a_{\infty}) \geq 4\%$)

$$\log_{10} x = 5.073 - \frac{1265}{T}$$

TABLE: 7

PURE P.E. EQUILIBRIUM RESULTS

T_o(°C)	CORRECTED TEMP.(°C)	c(%m/v)	x(MASS %)
30.0	30.0	7.904	7.745
40.0	40.0	10.610	10.319
50.0	50.1	14.116	10.597
60.0	60.1	18.634	17.716
70.0	70.2	24.771	23.183
75.0	75.3	28.421	26.345

LEAST MEAN SQUARES CORRELATION:

$$\log_{10} x = 4.980 - \frac{1242}{T}$$

where x = mass % , T = degrees Kelvin

APPENDIX B - COULTER COUNTER

B.1. Operation

It has been shown in section 6.2.1.2. that the response of the Coulter Counter is almost proportional to the particle volume. The deviation from proportionality and the dependence of particle shape on the responce increases with the particle/orifice ratio. For each sample analysis, therefore, a suitable orifice tube was chosen for the Coulter Counter, so that the size range of particles in the sample would be within about $1\frac{1}{2}$ to 40% of the aperture diameter. An aqueous electrolyte was used of 0.9g NaCl/100g H_2O + 8.5g Pure P.E./100g H_2O. This was such that the solution was saturated with P.E. at 25°C and the small degrees of supersaturation involved during an analysis at room temperature would not effect the particle size during the short time required for analysis.

A small amount of sample (about 0.01g) was placed in about 250 cm^3 of electrolyte previously filtered with a 0.45μ porosity membrane filter. A few drops of non-ionic dispersant (NONIDET P40) were added and the suspension stirred at about 1000 r.p.m. for about one minute. The suspension was then analysed by placing the beaker under the orifice tube and keeping it well stirred during an analysis to prevent settling. A controled external vacuum was applied indicating flow from the beaker through the orifice and unbalancing a mercury syphon. Releasing the external vacuum caused a syphoning action of the balancing mercury column continuing the sample flow. The advancing mercury column contacted start and stop probes, at a fixed volume apart, activating the electronic counter. Equal volumes of suspension were passed through the orifice at

preset threshold levels of the electronic counter. After each count the reset switch was depressed which zeroed the counting units and changed the polarity of the electrodes to prevent excess polarization. For each threshold level an average of a number of counts was taken depending on the statistical variation.

B.2. Coincidence

The possibility that two or more particles are in the sensing zone at the same time leads to what is called coincidence error. This can be of two forms, primary and secondary coincidence. Primary coincidence is the loss of count which results from only one pulse being generated for the passage of two or more particles. Secondary coincidence is the counting of a particle whose size is the sum of two or more particles. For secondary coincidence caused by a doublet a narrow size range of both particles is required; i.e. two particles larger than 8 microns diameter are needed to give a count equivalent of 10 microns diameter. Also close proximity of the particles is required. So secondary coincidence is negligible for the low concentration used and primary coincidence correction only was required.

The primary coincidence correction is the addition of a number of n_c'' to the actual count n_c'. If the coincidence level lies between 1% and 10% i.e. $0.01\, n_c' < n_c'' < 0.1\, n_c'$.

Then $$n_c'' = P_c \left(\frac{n_c'}{1000} \right)^2$$

Where the coincidence factor P_c is obtained from the formula:

$$P_c = 2.5 \left(\frac{D'}{100}\right)^3 \left(\frac{500}{V'}\right)$$

where D' is the aperture diameter in microns, and V' is the metering manometer volume in microlitres. The factor 2.5 was obtained experimentally by Coulter Electronics Limited using a 100 micron aperture and a 500 microlitre manometer volume at successive dilutions of counting on a monosized system. In order to avoid exceeding the 10% coincidence level n_c' must be less than $\frac{10^5}{D_c}$.

ORIFICE DIAMETER MICRONS	MANOMETER VOLUME MLS.	COINCIDENCE FACTOR	MAXIMUM COUNT FOR 10% COINCIDENCE
560	2	109.76	910
280	2	13.72	7,288
50	0.5	0.3125	320,000
50	0.05	3.125	32,000

B.3. Calibration

The calibration factor, K_c is used for conversion of threshold settings to particle volumes, or their cube roots to equivalent spherical diameters. The calibration factor is constant for a given aperture diameter and electrolyte resistivity.

A quantity of monosized particles, between 5% and 20% of the orifice diameter, such that the count obtained did not give more than 2% coincidence was dispersed in the electrolyte. The suspension was drawn through the orifice with the threshold dial set on zero and the amplifier gain index

on 3. The aperture current switch was adjusted to a value $I_c^{\ast}$ where the pulses on the oscilloscope occupied about one quarter of the screen height. The threshold dial t_c' was varied until the shadow line coincided with the height of the majority of pulses, and a count taken. Counts were taken at $\frac{1}{2}\ t_c'$ and $1\frac{1}{2}\ t_c'$ and averaged. The threshold value $t_c^{\ast}$ was found by trial and error which corresponded to the average of $\frac{1}{2}\ t_c^{\ast}$ and $1\frac{1}{2}\ t_c^{\ast}$.

The aperture resistance was measured by measuring the voltage, V_c between the outer electrode and earth, and calculating the aperture resistance from $R_c = \frac{r_c \times V_c}{300 - V_c}$ ohms where r_c is the resistance of the aperture current switch in the position used. Values of r_c are:-

Aperture Current Setting	r_c (ohms)
10	65,000
9	115,000
8	215,000
7	415,000
6	815,000

From the Scale Expansion Factor, F_c, tables supplied by Coulter Electronics, F_c was found for this aperture resistance at current setting $I_c^{\ast}$ and on Gain 3.

The Calibration factor K_c was then found from $K_c = D/(t_c^{\ast} F_c)^{\frac{1}{3}}$ where D was the diamter of the monosized particles. The diameter corresponding to any threshold level t_c' can then be calculated from $D_c = K_c(\sqrt[3]{t_c' F_c})$. The appropriate interpolated F_c values for particular current settings on Gain 3 are shown in the following pages, Tables 11, 12 and 13. For each consecutive lower gain index the F_c factor was

multiplied by $\sqrt{2}$. Similarly for each consecutive higher gain index the F_c factor was divided by $\sqrt{2}$.

As the electrolyte resistivity changed with temperature the F_c factors are shown for the aperture resistances encountered and the calibrations were done over the temperature range expected.

B.4. Size Analysis

For each size analysis thereafter the temperature of the electrolyte was taken and so the calibration known. The chart showing the Coulter Counter Data representation is shown on Table 14. The first three columns show the threshold settings. Column four shows the Scale Expansion Factors F_c for the particular gain index and aperture current. The product of this and the threshold setting t_c' gives the relative particle volume t_c, column 12. Then using the calibration factor K_c, the diameter for this threshold is found from $D_c = K_c \sqrt[3]{t_c}$ (column 13) The average of a number of counts, n_c', is taken above this diameter, and is shown for three different samples (columns 5,6 and 7) The average of these reading $\bar{n}_c'$ is taken, (column 8) and the coincidence error n_c'' calculated. The size analysis of particles present in the electrolyte as background count is shown (column 10) and the count $\bar{n}_c'$ then corrected by $n_c = \bar{n}_c' + n_c'' - /$ (column 11) Finally the number percentage greater than D_c is calculated (Column 14).

TABLE: 8 - CALIBRATION 50μ TUBE

ELECTROLYTE: 0.9g NaCl/100g H_2O + 8.5g Pure P.E./100g H_2O

MONOSIZED PARTICLES : PUFF BALL SPORES d = 3.62μ

$I^* = 5$ t = 54 at 15°C to 25°C

Temp. °C	14	15	16	17	18	19	20	21	22	23	24	25
Voltage I_c= 8	53	52	51	50	50	49	48	47	46	46	45	44
Resistance KΩ	46.1	45.1	44.1	43.1	43.1	42.0	40.9	40.0	39.0	39.0	38.0	36.9
$F_{\epsilon 5}$	0.06616	0.06610	0.06604	0.06598	0.06598	0.06592	0.06586	0.06580	0.06574	0.06574	0.06568	0.06562
K_c	2.37	2.37	2.37	2.37	2.37	2.37	2.37	2.37	2.37	2.37	2.37	2.37

TABLE: 9 CALIBRATION 280μ TUBE

ELECTROLYTE: 0.9g NaCl/100g H_2O = 8.5g Pure P.E./100g H_2O

MONOSIZED PARTICLES: LYCOPODIUM POWDER d = 28.0μ

$I^* = 5$ t = 52 at 14°C to 25°C

Temp. °C	14	15	16	17	18	19	20	21	22	23	24	25
Voltage $I_c = 8$	12.7	12.6	12.4	12.3	12.1	12.0	11.8	11.7	11.5	11.4	11.3	11.1
Resistance KΩ	9.53	9.43	9.28	9.20	9.04	8.96	8.78	8.71	8.56	8.48	8.41	8.26
F_5	0.06376	0.06375	0.06374	0.06373	0.06372	0.06371	0.06370	0.06369	0.06368	0.06368	0.06367	0.06365
K_c	18.8	18.8	18.8	18.8	18.8	18.8	18.8	18.8	18.8	18.8	18.8	18.8

TABIE: 10 CALIBRATION 560μ TUBE

ELECTROLYTE: 0.9% NaCl/100g H_2O + 8.5g Pure P.E./100g H_2O

MONOSIZED PARTICLES : LYCOPODIUM POWDER d = 28.0μ

$I^* = 9$ t = 36 at 14°C to 25°C

Temp.°C	14	15	16	17	18	19	20	21	22	23	24	25
Voltage $I_c = 8$	8.8	8.6	8.4	8.3	8.1	8.0	7.8	7.7	7.5	7.4	7.3	7.2
Resistance KΩ	6.50	6.35	6.20	6.13	5.97	5.90	5.74	5.65	5.50	5.44	5.36	5.28
E_9	0.00507	0.00505	0.00504	0.00503	0.00502	0.00501	0.00499	0.00498	0.00497	0.00496	0.00496	0.00495
K_c	49.4	49.5	49.5	49.5	49.5	49.5	49.6	49.6	49.6	49.6	49.6	49.7

TABLE: 11

COULTER COUNTER 'F' SCALE EXPANSION FACTORS - FOR USE WITH 50µ TUBE

Resist-ance KΩ	37	38	39	40	41	42	43	44	45	46	47
F_1	1.00000	1.00000	1.00000	1.00000	1.00000	1.00000	1.00000	1.00000	1.00000	1.00000	1.00000
F_2	0.50200	0.50200	0.50200	0.50200	0.50200	0.50200	0.50200	0.50200	0.50200	0.50200	0.50200
F_3	0.25240	0.25260	0.25280	0.25300	0.25300	0.25300	0.25300	0.25300	0.25300	0.25300	0.25300
F_4	0.12782	0.12788	0.12794	0.12800	0.12806	0.12812	0.12818	0.12824	0.12830	0.12836	0.12842
F_5	0.06562	0.06568	0.06574	0.06580	0.06586	0.06592	0.06598	0.06604	0.06610	0.06616	0.06622
F_6	0.03452	0.03458	0.03464	0.03470	0.03476	0.03482	0.03488	0.03494	0.03500	0.03506	0.03512
F_7	0.01899	0.01906	0.01913	0.01920	0.01927	0.01934	0.01941	0.01948	0.01954	0.01960	0.01966
F_8	0.01139	0.01146	0.01154	0.01161	0.01169	0.01176	0.01183	0.01191	0.01198	0.01205	0.01212
F_9	0.00782	0.00791	0.00800	0.00809	0.00818	0.00827	0.00836	0.00845	0.00854	0.00863	0.00872
F_{10}	0.00655	0.00667	0.00679	0.00691	0.00703	0.00715	0.00727	0.00739	0.00751	0.00763	0.00775

INTERPOLATED FROM COULTER ELECTRONICS LTD. DATA.

TABLE: 12

COULTER COUNTER 'F' SCALE EXPANSION FACTORS - FOR USE WITH 280 µ TUBE

Resistance KΩ	9.5	9.4	9.3	9.2	9.1	9.0	8.9	8.8	8.7	8.6	8.5	8.4	8.3	8.2
F_1	1.00000	1.00000	1.00000	1.00000	1.00000	1.00000	1.00000	1.00000	1.00000	1.00000	1.00000	1.00000	1.00000	1.00000
F_2	0.50090	0.50088	0.50086	0.50084	0.50082	0.50080	0.50078	0.50076	0.50074	0.50072	0.50070	0.50068	0.50066	0.50064
F_3	0.25100	0.25100	0.25100	0.25100	0.25100	0.25100	0.25100	0.25100	0.25100	0.25100	0.25100	0.25100	0.25100	0.25100
F_4	0.12617	0.12616	0.12615	0.12615	0.12614	0.12614	0.12613	0.12612	0.12612	0.12611	0.12611	0.12610	0.12610	0.12609
F_5	0.06376	0.06375	0.06374	0.06373	0.06372	0.06372	0.06371	0.06370	0.06369	0.06368	0.06368	0.06367	0.06366	0.06365
F_6	0.03256	0.03255	0.03254	0.03253	0.03253	0.03252	0.03251	0.03251	0.03250	0.03249	0.03248	0.03247	0.03246	0.03245
F_7	0.01695	0.01694	0.01693	0.01693	0.01692	0.01691	0.01690	0.01689	0.01688	0.01687	0.01687	0.01686	0.01685	0.01684
F_8	0.00916	0.00915	0.00914	0.00913	0.00912	0.00912	0.00911	0.00910	0.00910	0.00909	0.00908	0.00907	0.00906	0.00905
F_9	0.00530	0.00529	0.00528	0.00528	0.00527	0.00526	0.00525	0.00524	0.00524	0.00523	0.00522	0.00521	0.00520	0.00520
F_{10}	0.00345	0.00344	0.00343	0.00342	0.00341	0.00340	0.00339	0.00338	0.00337	0.00336	0.00335	0.00334	0.00333	0.00332

INTERPOLATED FROM COULTER ELECTRONICS LTD. DATA.

TABLE: 13

COULTER COUNTER 'F' SCALE EXPANSION FACTORS - FOR USE WITH 560μ TUBE

Resistance KΩ	5.2	5.3	5.4	5.5	5.6	5.7	5.8	5.9	6.0	6.1	6.2	6.3	6.4	6.5
F_1	1.00000	1.00000	1.00000	1.00000	1.00000	1.00000	1.00000	1.00000	1.00000	1.00000	1.00000	1.00000	1.00000	1.00000
F_2	0.50004	0.50006	0.50008	0.50010	0.50012	0.50014	0.50016	0.50018	0.50020	0.50022	0.50024	0.50026	0.50028	0.50030
F_3	0.25100	0.25100	0.25100	0.25100	0.25100	0.25100	0.25100	0.25100	0.25100	0.25100	0.25100	0.25100	0.25100	0.25100
F_4	0.12591	0.12591	0.12592	0.12592	0.12593	0.12593	0.12594	0.12594	0.12595	0.12595	0.12596	0.12596	0.12597	0.12597
F_5	0.06342	0.06343	0.06344	0.06345	0.06346	0.06347	0.06348	0.06349	0.06350	0.06351	0.06352	0.06353	0.06354	0.06355
F_6	0.03222	0.03223	0.03224	0.03225	0.03226	0.03227	0.03228	0.03229	0.03230	0.03231	0.03232	0.03233	0.03234	0.03235
F_7	0.01661	0.01662	0.01663	0.01664	0.01665	0.01666	0.01667	0.01668	0.01669	0.01670	0.01671	0.01672	0.01673	0.01674
F_8	0.00882	0.00883	0.00884	0.00885	0.00886	0.00887	0.00888	0.00889	0.00890	0.00891	0.00892	0.00893	0.00894	0.00895
F_9	0.00494	0.00495	0.00496	0.00497	0.00498	0.00499	0.00500	0.00501	0.00502	0.00503	0.00504	0.00505	0.00506	0.00507
F_{10}	0.00302	0.00303	0.00304	0.00305	0.00306	0.00307	0.00308	0.00309	0.00310	0.00311	0.00312	0.00313	0.00314	0.00315

INTERPOLATED FROM COULTER ELECTRONICS LTD. DATA

TABLE:14 COULTER COUNTER DATA

SAMPLE: SOURCE: ATTRITION:

APERTURE DIAMETER: MANOMETER VOLUME: COINCIDENCE FACTOR(P): CALIBRATION FACTOR(K):

DISPERSANT: TEMPERATURE: APERTURE RESISTANCE: ELECTROLYTE:

GAIN INDEX	t'	l	F	n_1'	n_2'	n_3'	$\bar{n}'$	$n'' = P\left(\frac{\bar{n}'}{10^0}\right)^2$	$\sqrt{}$	$n = \bar{n}' + n'' - \sqrt{}$	$t = t'(F)$	$d = k\sqrt[3]{t}$	CUM No% OVERSIZE
1	2	3	4	5	6	7	8	9	10	11	12	13	14
AMPLIFIER GAIN SETTING	THRESHOLD DIAL SETTING	APERTURE CURRENT SETTING	SCALE EXPANSION FACTORS	RAW COUNTS SAMPLE 1	RAW COUNTS SAMPLE 2	RAW COUNTS SAMPLE 3	AVERAGE OF RAW COUNTS	COINCIDENCE CORRECTION	BACKGROUND COUNT ON BLANK ELECTROLYTE	CORRECTED COUNT	RELATIVE PARTICLE VOLUME	PARTICLE DIAMETER	CUMULATIVE NUMBER % ABOVE STATED SIZE

TABLE: 15

COULTER COUNTER DIAMETER CORRECTION FOR 280 μ TUBE. ORIENTATION B.

Dcμ	Lμ	Dμ	Dcμ	Lμ	Dμ	Dcμ	Lμ	Dμ
5	5.1851	5.0062	53	54.395	52.518	101	101.20	97.712
6	6.2219	6.0072	54	55.402	53.490	102	102.14	98.615
7	7.2586	7.0081	55	56.408	54.462	103	103.07	99.516
8	8.2848	7.9989	56	57.413	55.432	104	104.00	100.41
9	9.3199	8.9983	57	58.417	56.401	105	104.93	101.31
10	10.355	9.9974	58	59.419	57.369	106	105.86	102.21
11	11.389	10.996	59	60.421	58.336	107	106.79	103.10
12	12.424	11.995	60	61.421	59.301	108	107.71	103.99
13	13.458	12.994	61	62.420	60.266	109	108.63	104.88
14	14.492	13.992	62	63.418	61.229	110	109.55	105.77
15	15.526	14.990	63	64.414	62.191	111	110.47	106.66
16	16.559	15.988	64	65.410	63.153	112	111.39	107.54
17	17.592	16.985	65	66.404	64.112	113	112.30	108.43
18	18.625	17.982	66	67.397	65.071	114	113.22	109.31
19	19.657	18.979	67	68.388	66.028	115	114.13	110.19
20	20.689	19.975	68	69.379	66.984	116	115.04	111.07
21	21.721	20.971	69	70.367	67.939	117	115.93	111.93
22	22.752	21.967	70	71.355	68.892	118	116.84	112.80
23	23.782	22.962	71	72.341	69.845	119	117.74	113.67
24	24.813	23.956	72	73.326	70.795	120	118.64	114.54
25	25.842	24.950	73	74.309	71.745	121	119.53	115.41
26	26.872	25.944	74	75.291	72.693	122	120.43	116.27
27	27.900	26.937	75	76.272	73.640	123	121.32	117.13
28	28.928	27.930	76	77.251	74.585	124	122.21	117.99
29	29.956	28.922	77	78.228	75.529	125	123.10	118.85
30	30.983	29.914	78	79.205	76.471	126	123.98	119.70
31	32.009	30.905	79	80.179	77.412	127	124.87	120.56
32	33.035	31.895	80	81.152	78.352	128	125.75	121.41
33	34.060	32.885	81	82.124	79.290	129	126.63	122.26
34	35.084	33.874	82	83.094	80.226	130	127.50	123.10
35	36.108	34.862	83	84.062	81.161	131	128.38	123.95
36	37.131	35.850	84	85.029	82.094	132	129.25	124.79
37	38.153	36.837	85	85.994	83.026	133	130.12	125.63
38	39.175	37.823	86	86.958	83.957	134	130.99	126.47
39	40.196	38.809	87	87.919	84.885	135	131.85	127.30
40	41.216	39.793	88	88.889	85.821	136	132.72	128.14
41	42.235	40.777	89	89.846	86.746	137	133.58	128.97
42	43.253	41.761	90	90.802	87.669	138	134.43	129.79
43	44.271	42.743	91	91.757	88.590	139	135.29	130.62
44	45.288	43.725	92	92.709	89.510	140	136.14	131.44
45	46.303	44.705	93	93.660	90.428	141	136.99	132.27
46	47.318	45.685	94	94.609	91.344	142	137.84	133.09
47	48.332	46.664	95	95.557	92.259	143	138.69	133.90
48	49.345	47.642	96	96.502	93.172	144	139.53	134.72
49	50.357	48.619	97	97.446	94.083	145	140.37	135.53
50	51.368	49.596	98	98.389	94.993	146	141.21	136.34
51	52.378	50.571	99	99.329	95.901	147	142.05	137.15
52	53.387	51.545	100	100.27	96.807	148	142.88	137.95

TABLE: 16

COULTER COUNTER DIAMETER CORRECTION FOR 50μ TUBE. ORIENTATION B.

Dc μ	L μ	D μ	Dc μ	L μ	D μ	Dc μ	L μ	D μ
1	1.0370	1.0012	11	11.259	10.871	21	20.801	20.083
2	2.0733	2.0018	12	12.253	11.830	22	21.700	20.951
3	3.1083	3.0010	13	13.239	12.782	23	22.589	21.809
4	4.1412	3.9982	14	14.217	13.726	24	23.452	22.643
5	5.1713	4.9929	15	15.186	14.662	25	24.311	23.472
6	6.1981	5.9842	16	16.146	15.589	26	25.157	24.288
7	7.2209	6.9717	17	17.098	16.507	27	25.988	25.091
8	8.2390	7.9547	18	18.039	17.416	28	26.806	25.881
9	9.2519	8.9326	19	18.970	18.315	29	27.609	26.657
10	10.259	9.9048	20	19.891	19.204	30	28.398	27.418

TABLE: 17

COULTER COUNTER DIAMETER CORRECTION FOR 560μ TUBE. ORIENTATION B.

Dc μ	L μ	D μ	Dc μ	L μ	D μ	Dc μ	L μ	D μ
5	5.1859	5.0069	33	34.151	32.973	61	62.992	60.818
6	6.2226	6.0078	34	35.184	33.970	62	64.018	61.809
7	7.2594	7.0089	35	36.217	34.967	63	65.044	62.800
8	8.2863	8.0003	36	37.250	35.964	64	66.070	63.790
9	9.3218	9.0001	37	38.282	36.961	65	67.095	64.780
10	10.358	10.000	38	39.314	37.958	66	68.120	65.769
11	11.393	11.000	39	40.346	38.954	67	69.145	66.758
12	12.428	11.999	40	41.378	39.950	68	70.169	67.747
13	13.464	12.999	41	42.410	40.946	69	71.193	68.736
14	14.499	13.999	42	43.441	41.942	70	72.216	69.724
15	15.534	14.998	43	44.473	42.938	71	73.240	70.712
16	16.570	15.998	44	45.504	43.933	72	74.262	71.700
17	17.605	16.997	45	46.534	44.928	73	75.285	72.687
18	18.640	17.997	46	47.565	45.923	74	76.307	73.674
19	19.675	18.996	47	48.595	46.918	75	77.329	74.660
20	20.710	19.995	48	49.625	47.913	76	78.350	75.646
21	21.744	20.994	49	50.655	48.907	77	79.371	76.632
22	22.779	21.993	50	51.685	49.901	78	80.392	77.617
23	23.813	22.992	51	52.714	50.895	79	81.412	78.602
24	24.848	23.990	52	53.743	51.888	80	82.432	79.587
25	25.882	24.989	53	54.772	52.882	81	83.451	80.571
26	26.916	25.987	54	55.800	53.875	82	84.470	81.555
27	27.950	26.986	55	56.829	54.867	83	85.489	82.538
28	28.984	27.984	56	57.857	55.860	84	86.507	83.521
29	30.018	28.982	57	58.884	56.852	85	87.524	84.504
30	31.051	29.980	58	59.912	57.844	86	88.542	85.486
31	32.085	30.978	59	60.939	58.836	87	89.559	86.468
32	33.118	31.975	60	61.966	59.827	88	90.575	87.449

TABLE: 17 (CONT.)

COULTER COUNTER DIAMETER CORRECTION FOR 560μ TUBE. ORIENTATION B.

Dcμ	Lμ	Dμ	Dcμ	Lμ	Dμ	Dcμ	Lμ	Dμ
89	91.591	88.430	138	140.73	135.88	187	188.27	181.77
90	92.607	89.411	139	141.72	136.83	188	189.22	182.69
91	93.622	90.391	140	142.71	137.78	189	190.17	183.60
92	94.636	91.370	141	143.70	138.74	190	191.11	184.52
93	95.651	92.350	142	144.68	139.69	191	192.06	185.43
94	96.664	93.328	143	145.67	140.64	192	193.00	186.34
95	97.678	94.307	144	146.65	141.59	193	193.95	187.26
96	98.690	95.284	145	147.64	142.54	194	194.89	188.17
97	99.703	96.262	146	148.62	143.49	195	195.84	189.08
98	100.71	97.239	147	149.60	144.44	196	196.78	189.99
99	101.73	98.215	148	150.58	145.39	197	197.72	190.89
100	102.74	99.191	149	151.56	146.33	198	198.66	191.80
101	103.75	100.17	150	152.54	147.28	199	199.60	192.71
102	104.76	101.14	151	153.52	148.22	200	200.54	193.61
103	105.77	102.12	152	154.50	149.17	201	201.47	194.52
104	106.77	103.09	153	155.48	150.11	202	202.41	195.42
105	107.78	104.06	154	156.46	151.06	203	203.34	196.33
106	108.79	105.04	155	157.43	152.00	204	204.28	197.23
107	109.80	106.01	156	158.41	152.94	205	205.21	198.13
108	110.80	106.98	157	159.38	153.88	206	206.15	199.03
109	111.81	107.95	158	160.36	154.82	207	207.08	199.93
110	112.82	108.92	159	161.33	155.76	208	208.00	200.82
111	113.82	109.89	160	162.30	156.70	209	208.94	201.73
112	114.83	110.86	161	163.28	157.64	210	209.87	202.62
113	115.83	111.83	162	164.25	158.58	211	210.80	203.52
114	116.83	112.80	163	165.22	159.52	212	211.72	204.42
115	117.84	113.77	164	166.19	160.45	213	212.65	205.31
116	118.84	114.74	165	167.16	161.39	214	213.57	206.20
117	119.84	115.70	166	168.12	162.32	215	214.50	207.10
118	120.84	116.67	167	169.09	163.26	216	215.42	207.99
119	121.84	117.64	168	170.06	164.19	217	216.35	208.88
120	122.84	118.60	169	171.02	165.12	218	217.27	209.77
121	123.84	119.57	170	171.99	166.05	219	218.19	210.66
122	124.84	120.53	171	172.95	166.98	220	219.11	211.55
123	125.84	121.50	172	173.92	167.91	221	220.03	212.43
124	126.84	122.46	173	174.88	168.84	222	220.94	213.32
125	127.83	123.42	174	175.84	169.77	223	221.86	214.20
126	128.83	124.38	175	176.80	170.70	224	222.78	215.09
127	129.82	125.34	176	177.76	171.62	225	223.69	215.97
128	130.82	126.31	177	178.72	172.55	226	224.61	216.86
129	131.81	127.27	178	179.68	173.48	227	225.51	217.73
130	132.81	128.22	179	180.63	174.40	228	226.42	218.61
131	133.80	129.18	180	181.59	175.32	229	227.33	219.49
132	134.79	130.14	181	182.55	176.25	230	228.24	220.36
133	135.79	131.10	182	183.50	177.17	231	229.15	221.24
134	136.78	132.06	183	184.45	178.09	232	230.05	222.11
135	137.77	133.01	184	185.41	179.01	233	230.96	222.99
136	13.76	133.97	185	186.36	179.93	234	231.86	223.86
137	139.75	134.92	186	187.32	180.86	235	232.77	224.74

TABLE: 18

COMPARISON OF DISPERSING TECHNIQUES WITH BATCH D SEED 44-64 μ

PRIOR TO COULTER COUNTER ANALYSIS

	CUMULATIVE No. % OVERSIZE													
		STIRRING			ULTRASONICS									
AGIT: ATION	NONE	800 RPM	800 RPM	1200 RPM	POWER POSN. 2		POWER POSN. 4		POWER POSN. 6		POWER POSN. 8			
(MIN)		0.5	2.0	2.0	0.5	2.0	0.5	2.0	0.5	2.0	0.17	0.33	0.5	2.0
D_{μ}														
82.3	0	0	0	0	0	0	0	0	0	0	0			
72.2	0.03	0.07	0.06	0.06	0.02	0.02	0.02	0.01	0.01	0.01	0	0	0	0
57.8	1.12	0.79	0.80	0.99	0.40	0.33	0.60	0.10	0.12	0.04	0.10	0.02	0.01	0
46.1	3.63	3.92	3.60	4.11	3.11	1.56	1.82	0.31	0.82	0.06	0.45	0.19	0.05	0.01
36.8	10.7	11.6	10.1	9.65	7.40	5.15	5.48	1.81	2.88	0.10	1.43	0.82	0.23	0.03
29.3	20.9	21.8	20.4	18.8	16.1	11.5	12.3	3.15	6.44	0.46	4.60	2.38	1.13	0.07
23.5	35.5	35.8	33.8	35.9	28.6	21.8	22.0	8.15	13.8	2.08	10.8	6.48	4.44	0.20
18.9	53.2	51.4	51.3	47.8	42.3	31.5	36.3	17.8	25.0	7.18	21.4	14.8	15.8	1.04
15.4	66.3	69.0	63.2	65.5	57.8	47.2	52.1	35.2	41.2	16.7	34.4	27.2	23.7	4.00
12.8	80.4	82.9	79.3	78.9	72.0	62.6	68.4	53.6	54.6	30.8	47.5	41.4	38.7	10.3
11.0	90.9	92.0	92.4	91.0	84.5	71.5	85.6	66.1	66.4	42.3	59.6	50.8	49.2	20.4
8.78	100	100	100	100	96.3	89.5	93.3	76.8	84.5	64.8	77.8	69.0	68.6	40.0
7.79					100	96.0	100	92.8	90.5	75.4	86.2	81.3	79.1	57.9
6.95						100		98.3	100	85.6	94.1	93.0	91.5	75.4
6.20								100		100	100	100	100	100
	AVERAGED							※		※	※	※	※	※

※ LOWER LIMIT OF SIZE DISTRIBUTION NOT ATTAINED

TABLE: 19 COULTER COUNTER DATA

SAMPLE: 1 SOURCE: BATCH E PREPARED SEED 89-105μ ATTRITION: 20 MINS. AT 2000 R.P.M. NEW CELL

APERTURE DIAMETER: 280μ MANOMETER VOLUME: 2mls. COINCIDENCE FACTOR(P): 13.72 CALIBRATION FACTOR(K): 18.8

DISPERSANT: NONIDET P40+STIRRING TEMPERATURE: 21°C APERTURE RESISTANCE: 8.71K ELECTROLYTE: AQUEOUS 0.9% NaCl+8.5%PE

GAIN INDEX	t'	1	F	n'	n'	n'	$\bar{n}'$	$n'' = P\left(\frac{\bar{n}'}{1000}\right)^2$	$\surd$	$n = \bar{n}'+n''-\surd$	$t = t'(F)$	$d = k\sqrt[3]{t}$	CUM No% OVERSIZE
3	300	1	1.00000	1	1	2	1	0	1	0	300	126	0
3	210	1	1.00000	4	5	5	5	0	2	3	210	112	0.28
3	150	1	1.00000	22	23	23	23	0	2	21	150	100	1.96
3	90	1	1.00000	44	44	53	47	0	2	45	90	84.2	4.18
3	60	1	1.00000	80	72	76	76	0	2	74	60	73.5	6.88
3	60	2	0.56074	149	126	130	135	0	2	133	30	58.4	12.40
3	60	3	0.25100	241	198	210	216	1	2	215	15.1	46.4	20.00
3	60	4	0.12612	362	301	318	327	1	5	323	7.57	36.9	30.00
3	60	5	0.06369	496	406	444	449	3	6	446	3.82	29.4	41.50
3	60	6	0.03250	684	552	598	611	5	10	606	1.95	23.5	56.40
3	60	7	0.01688	826	706	772	768	8	14	762	1.012	18.9	70.90
3	60	8	0.00910	959	817	864	880	11	21	870	0.546	15.4	81.10
3	60	9	0.00524	1039	971	988	999	14	48	965	0.315	12.8	89.80
3	60	10	0.00337	1130	1021	1165	1105	17	67	1055	0.202	11.0	98.40
3	30	10	0.00337	1160	1119	1176	1152	18	97	1073	0.101	8.78	100.00
4	30	10	0.00238	1005	1064	1104	1058	15	109	964	0.071	7.79	

TABLE:20 COULTER COUNTER DATA

SAMPLE: 2 SOURCE: BATCH E PREPARED SEED 89-105μ ATTRITION::20 MINS. AT 2000 R.P.M. NEW CELL

APERTURE DIAMETER: 280μ MANOMETER VOLUME: 2mls. COINCIDENCE FACTOR(P): 13.72 CALIBRATION FACTOR(K):18.8

DISPERSANT:NONIDET P40+STIRRING TEMPTRATURE:23°C APERTURE RESISTANCE:8.48KΩ ELECTROLYTE: AQUEOUS 0.9% NaCl+8.5%P.E.

GAIN INDEX	t'	1	F	n'	n'	n'	$\bar{n}'$	$n'' = P\left(\frac{\bar{n}'}{1000}\right)^2$	$\surd$	$n = \bar{n}'+n''-\surd$	t = t'(F)	d = $k\sqrt[3]{t}$	CUM No% OVERSIZE
3	300	1	1.00000	2	2	2	2	0	2	0	300	126	0
3	210	1	1.00000	3	7	6	5	0	2	3	210	112	0.30
3	150	1	1.00000	20	27	19	22	0	2	20	150	100	2.01
3	90	1	1.00000	43	61	42	49	0	2	47	90	84.2	4.72
3	60	1	1.00000	63	88	77	76	0	2	74	60	73.5	7.47
3	60	2	0.50070	103	155	103	120	0	2	118	30	58.4	11.9
3	60	3	0.25100	172	253	191	205	1	2	204	15.1	46.4	20.5
3	60	4	0.12611	265	338	298	300	1	7	294	7.57	36.9	29.5
3	60	5	0.06368	341	473	413	409	2	14	397	3.82	29.4	29.9
3	60	6	0.03248	424	636	528	529	4	33	500	1.95	23.5	50.2
3	60	7	0.01687	533	777	657	656	6	38	624	1.012	18.9	62.7
3	60	8	0.00908	643	862	858	788	8	67	729	0.545	15.4	73.3
3	60	9	0.00522	768	1038	910	905	11	86	830	0.314	12.8	83.4
3	60	10	0.00335	868	1076	1007	984	13	92	905	0.201	11.0	90.0
3	30	10	0.00335	884	1195	1178	1086	16	106	996	0.10.	8.78	100.0
4	30	10	0.00237	918	1127	1162	1069	16	147	938	0.071	7.79	

TABLE:21 COULTER COUNTER DATA

SAMPLE: 3 SOURCE: BATCH E PREPARED SEED 89-105μ ATTRITION: 20 MINS. AT 2000 R.P.M. NEW CELL

APERTURE: 280 MANOMETER VOLUME: 2mls. COINCIDENCE FACTOR(P):13.72 CALIBRATION FACTOR(K): 18.8

DISPERSANT:NONIDET P40+STIRRING TEMPERATURE:22°C APERTURE RESISTANCE:8.48KΩ ELECTROLYTE: AQUEOUS 0.9% NaCl+8.5% P.E.

GAIN INDEX	t'	1	F	n'	n'	n'	$\bar{n}'$	$n'' = P\left(\frac{\bar{n}'}{1000}\right)^2$	$\surd$	$n = \bar{n}'+n''-\surd$	$t = t'(F)$	$d = k\sqrt[3]{t}$	CUM NO% OVERSIZE
3	300	1	1.00000	1	1	1	1	0	1	0	300	126	0
3	210	1	1.00000	4	10	12	9	0	2	7	210	112	0.71
3	150	1	1.00000	33	28	20	27	0	2	25	150	100	2.52
3	90	1	1.00000	64	75	66	68	0	2	66	90	84.2	6.65
3	60	1	1.00000	93	105	99	99	0	2	97	60	73.5	9.79
3	60	2	0.50070	128	177	159	155	0	4	151	30	58.4	15.2
3	60	3	0.25100	233	253	260	249	1	4	246	15.1	46.4	24.8
3	60	4	0.12611	321	388	355	354	2	4	352	7.57	36.9	35.5
3	60	5	0.06368	401	470	486	452	3	9	446	3.82	29.4	45.0
3	60	6	0.03248	516	662	673	617	5	17	605	1.95	23.5	61.1
3	60	7	0.01687	639	745	771	718	7	19	706	1.012	18.9	71.1
3	60	8	0.00908	714	869	909	831	9	26	814	0.545	15.4	82.2
3	60	9	0.00522	833	1039	1069	980	13	33	960	0.314	12.8	96.9
3	60	10	0.00335	898	1097	1075	1087	14	46	991	0.201	11.0	100
3	30	10	0.00335	874	1085	1073	1011	14	53	972	0.101	8.78	

TABLE: 22

BATCH E PREPARED SEED SIEVE FRACTION 89-105μ

	CUMULATIVE No. % OVERSIZE						
COULTER DIAMETER D_o	SAMPLE 1	SAMPLE 2	SAMPLE 3	AVERAGE OF 12&3	CORRECTED D	MEAN D	AVERAGE No% OF MEAN D
126	0	0	0	0	119.7	113.6	0.43
112	0.28	0.30	0.71	0.43	107.5	102.2	1.73
100	1.96	2.01	2.52	2.16	96.8	89.6	3.02
84.2	4.18	4.72	6.65	5.18	82.3	77.3	2.90
73.5	6.88	7.47	9.79	8.05	72.2	65.0	5.15
58.4	12.4	11.9	15.2	13.2	57.8	51.9	8.6
46.4	20.0	20.5	24.8	21.8	46.1	41.5	9.9
36.9	30.0	29.5	35.5	31.7	36.8	33.1	10.4
29.4	41.5	39.9	45.0	42.1	29.3	26.4	13.8
23.5	56.4	50.2	61.1	55.9	23.5	21.2	12.3
18.9	70.9	62.7	71.1	68.2	18.9	17.2	10.7
15.4	81.1	73.3	82.2	78.9	15.4	14.1	11.1
12.8	89.8	83.4	96.9	90.0	12.8	11.9	6.4
11.0	98.4	90.8	100	96.4	11.0	9.89	3.6
8.78	100	100		100	8.78		
ΣNo%	100.0	100.0	100.0	100.0			
ΣNo% xD	3,295	3,155	3,580	3,353			
ΣNo% xD^2	154,789	151,273	182,830	163,117			

TABLE: 23

SIZE ANALYSIS COMPARISON OF BATCH C SEED SIEVE FRACTION 44-64μ AFTER ATTRITION IN CELLS A AND C AT DIFFERENT STIRRER SPEEDS

CELL:			A				C			
STIRRER SPEED			500 R.P.M.		2000 R.P.M.		500 R.P.M.		2000 R.P.M.	
D_c/μ	$D\mu$	MEAN $D\mu$	No% OVER	No%	No% OVER	No%	No% OVER	No%	No% OVER	No%
84.2	82.3		0		0		0		0	
		77.3		1.18		0.26		0.41		1.48
73.5	72.2		1.18		0.26		0.41		1.48	
		65.0		23.3		8.08		10.24		9.52
58.4	57.8		24.5		8.34		10.65		11.0	
		51.9		23.8		11.5		13.45		14.4
46.4	46.1		48.3		19.9		24.1		25.4	
		41.5		11.3		8.5		11.1		11.5
36.9	36.8		59.6		28.4		35.2		36.9	
		33.1		9.4		13.8		9.8		9.5
29.4	29.3		69.0		42.2		45.0		46.4	
		26.4		4.5		11.0		11.3		10.5
23.5	23.5		73.5		53.2		56.3		56.9	
		21.2		12.5		8.9		11.5		10.8
18.9	18.9		85.9		62.1		67.8		67.7	
		17.2		11.1		18.9		14.4		15.2
15.4	15.4		97.0		81.0		82.2		82.9	
		14.1		1.0		11.8		14.3		11.6
12.8	12.8		98.0		92.8		96.5		94.5	
		11.9		2.0		6.3		3.5		5.5
11.0	11.0		100		99.1		100		100	
		9.89				0.9				
8.75	8.75				100					

TABLE: 24

COLLECTED SIZE ANALYSES OF SEED MATERIALS

ATTRITED AT 2000R.P.M. IN STIRRED CELL C

SEED BATCH: SIEVE FRACTION			C 44-64μ		C 75-89μ		D 44-64μ		D 75-89μ		E 89-105μ	
$D_{o}\mu$	$D\mu$	MEAN $D\mu$	No.% OVER	No.%	No.% OVER	No.%	No.% OVER	No.%	No.% OVER	No.%	No.% OVER	No.%
141	132											
		126										
126	120								0		0	
		114								0.50		0.43
112	108				0				0.50		0.43	
		102				0.16				6.35		1.73
100	96.8				0.16				6.85		2.16	
		89.6				0.76				32.8		3.02
84.2	82.3		0		0.92		0		39.6		5.18	
		77.3		1.48		0.75		0.05		18.4		2.90
73.5	72.2		1.48		1.67		0.05		58.0		8.05	
		65.0		9.52		1.04		0.88		3.5		5.15
58.4	57.8		11.0		2.71		0.93		61.5		13.2	
		51.9		14.4		0.86		2.73		7.0		8.6
46.4	46.1		25.4		3.57		3.66		68.5		21.8	
		41.5		11.0		1.81		6.84		6.7		9.9
36.9	36.8		36.4		5.38		10.5		75.2		31.7	
		33.1		10.0		3.25		10.0		7.1		10.4
29.4	29.3		46.4		8.63		20.5		82.3		42.1	
		26.4		10.5		5.57		14.7		12.7		13.8
23.5	23.5		56.9		14.2		35.2		95.0		55.9	
		21.2		10.8		7.2		15.7		5.0		12.3
18.9	18.9		67.7		21.4		50.9		100		68.2	
		17.2		15.2		9.2		15.1				10.7
15.4	15.4		82.9		30.6		66.0				78.9	
		14.1		11.6		10.2		14.4				11.1
12.8	12.8		94.5		40.8		80.4				90.0	
		11.9		5.5		7.9		11.2				6.4
11.0	11.0		100		48.7		91.6				96.4	
		9.89				22.5						3.6
8.78	8.78				71.2		100				100	
		8.29				5.0						
7.79	7.79				76.2							
		7.37				23.8						
6.95	6.95				100							
		6.58										
6.20	6.20											

TABLE: 24 (CONTINUED)

COLLECTED SIZE ANALYSES OF SEED MATERIALS

ATTRITED AT 2000 R.P.M. IN STIRRED CELL C

SEED BATCH: SIEVE FRACTION			F 89-105μ		P.E. 89-105μ		P.G. 64-75μ		P.G. 89-105μ	
$D_o\mu$	$D\mu$	MEAN $D\mu$	No%OVER	No%	No%OVER	No%	No%OVER	No%	No%OVER	No%
141	132				0				0	
		126				4.24				0.39
126	120				4.24		0		0.39	
		114				8.46		0.39		1.83
112	108		0		12.7		0.39		2.22	
		102		0.22		13.9		1.57		2.04
100	96.8		0.22		26.6		1.96		4.26	
		89.6		0.34		4.4		35.4		1.26
84.2	82.3		0.56		31.0		37.4		5.52	
		77.3		0.35		3.4		32.6		0.19
73.5	72.2		0.91		34.4		70.0		5.71	
		65.0		0.22		5.1		14.2		0.78
58.4	57.8		1.13		39.5		84.2		6.49	
		51.9		0.23		13.4		2.3		1.07
46.4	46.1		1.36		52.9		86.5		7.56	
		41.5		0.62		9.2		1.7		0.86
36.9	36.8		1.98		62.1		88.2		8.42	
		33.1		0.73		4.6		3.1		1.07
29.4	29.3		2.71		66.7		91.3		9.49	
		26.4		2.84		5.4		4.9		2.01
23.5	23.5		5.55		72.1		96.2		11.5	
		21.2		1.53		1.7		1.4		3.5
18.9	18.9		7.08		73.8		97.6		15.0	
		17.2		8.22		8.7		2.4		4.9
15.4	15.4		15.3		82.5		100		19.9	
		14.1		7.8		13.5				12.1
12.8	12.8		23.1		96.0				32.0	
		11.9		7.7		2.1				5.6
11.0	11.0		30.8		98.1				37.6	
		9.89		15.8		1.9				17.1
8.78	8.78		46.6		100				54.7	
		8.29		17.2						8.7
7.79	7.79		63.8						63.4	
		7.37		16.4						19.4
6.95	6.95		80.2						82.8	
		6.58		19.8						17.2
6.20	6.20		100						100	

※ LOWER LIMIT OF SIZE DISTRIBUTION NOT ATTAINED.

TABLE: 25

COLLECTED SIZE ANALYSES OF SEED MATERIALS

USED IN FLUIDISED BED EXPERIMENTS

SEED BATCH: SIEVE FRACTION:			C 124-150μ		E 124- 150μ		P.E. 124-150μ	
D_o μ	D μ	MEAN D μ	No% OVER	No%	No% OVER	No%	No% OVER	No%
222	213						0	
		201						1.30
194	188		0		0		1.30	
		170		13.6		6.90		27.3
154	151		13.6		6.90		28.6	
		136		61.4		15.4		21.5
122	121		75.0		22.3		50.1	
		109		10.9		15.3		17.2
97.3	96.6		85.9		37.6		67.3	
		86.9		4.6		13.0		19.7
77.5	77.1		90.5		50.6		87.0	
		69.4		9.5		18.7		8.3
61.8	61.6		100		69.3		95.3	
		55.6				15.1		4.7
49.6	49.5				84.4		100	
		44.9				15.6		
40.2	40.2				100			

TABLE: 26

COULTER COUNTER PRODUCT SIZE ANALYSES

SIZE			CUMULATIVE No. % OVERSIZE	
D_o	$L\mu$	$D\mu$	R.E.15 PRODUCT	R.P.E.12 PRODUCT
332	315	304		0
295	285	275		0.4
264	258	250		1.3
222	221	213	0	3.7
194	195	188	0.6	8.0
154	156	151	4.0	16.6
122	125	121	6.2	19.2
97.3	100	96.5	14.5	37.1
77.5	79.9	77.1	28.8	62.7
61.8	63.8	61.6	51.5	95.0
49.6	51.2	49.5	88.3	100
40.2	41.6	40.2	91.2	
33.1	34.2	33.1	96.7	
28.1	29.1	28.1	100	
19.9	20.6	19.9		

TABLE: 27

IMAGE SHEAR SIZE ANALYSES

SIZE			CUMULATIVE No. % OVERSIZE			
			R.E.15 PRODUCT	R.P.E.12 PRODUCT	R.E.F.4 PRODUCT	ATTRITED BATCH E SEED 89 - 105μ FRACTION
1.154Lμ	Lμ	Dμ				
5	4.33	4.18			99.5	100
10	8.65	8.37		100	94.0	99.0
15	13.0	12.5			89.5	97.5
20	17.3	16.7	100	99.5	83.0	95.5
25	21.6	20.9			76.0	91.0
30	26.0	25.0	98	98.0	69.0	86.5
35	30.4	29.4			59.0	80.5
40	34.7	33.5	96	96.0	54.5	76.0
45	38.9	37.5	94		49.0	66.0
50	43.4	41.9	90	92.0	43.0	63.5
55	47.6	46.0	82			54.5
60	52.0	50.2	72	86.5	41.5	48.5
65	56.3	54.4	69			44.5
70	60.5	58.2	62	75.0	35.0	36.5
75	64.9	62.6	56			30.0
80	69.2	66.8	52	61.5	29.5	23.0
85	73.6	71.1				16.0
90	78.0	75.3	49	46.0	27.0	13.0
95	82.4	79.6				8.5
100	86.6	83.6	40	35.5	21.5	8.0
120	104	100	36	22.0	16.0	2.5
140	122	118	22			0
150	130	126	12	18.0	8.5	
160	139	135	5			
200	173	167	0	7.5	3.5	
250	216	209		1.5	0	
300	260	251		0		

TABLE: 28

IMAGE SHEAR PRODUCT SIZE ANALYSES

BATCH E SEEDED GROWTH EXPERIMENTS AT 60.0°C

			CUMULATIVE No. % OVERSIZE						
RUN No.			R.E.28	R.E.32	R.E.33	R.E.34	R.E.35	R.E.36	R.E.37
INITIAL c % m/v			27.5	27.5	27.5	26.0	26.0	24.5	24.5
SEED MASS g			2.0	1.0	0.5	1.0	0.5	1.0	0.5
CALCULATED $\Delta D\mu$			66	91	124	75	108,	61	88
$1.154L\mu$	$L\mu$	$D\mu$							
5	4.33	4.18	99.0			93.5	99.0		
10	8.65	8.37	93.5	100	100	83.0	87.0	99.0	98.0
15	13.0	12.5	76.0		95.0	64.5	71.5	90.5	
20	17.3	16.7	69.0	89.0	92.0	55.0	62.0	86.5	91.5
25	21.6	20.9			88.0	43.0	52.5	72.5	
30	26.0	25.0	56.0	80.0	86.0	39.5	49.5	62.0	83.0
35	30.4	29.4						53.0	75.0
40	34.7	33.5	51.0	72.0	83.5	31.0	39.0	47.5	66.5
45	38.9	37.5							62.5
50	43.4	41.9	43.5	68.5	82.5	26.0	32.5	43.5	54.0
55	47.6	46.0			76.0				
60	52.0	50.2	33.5	62.5	73.5	22.0	29.0	40.5	49.5
65	56.3	54.4			68.5				
70	60.5	58.2	26.5	56.0	64.5	16.5	26.5	35.5	40.5
75	64.9	62.6			57.0				36.5
80	69.2	66.8	20.0	50.0	48.5	11.0	25.0	27.0	27.5
85	73.6	71.1			43.5				20.0
90	78.0	75.3	16.5	38.0	36.0	8.5	20.0	22.5	15.0
100	86.6	83.6	14.0	33.5	24.5	5.0	14.0	19.5	10.0
110	95.3	92.0		25.0	16.0				
120	104	100		22.0	15.0		9.5	13.0	7.0
130	113	109		20.0	10.0				
150	130	126	6.5	16.0	4.5	1.5	4.5	7.0	3.5
200	173	167	1.0	8.0	2.0	0.5	0.5	1.5	1.5
250	216	209	0	1.5	0.5	0	.0	0	
300	260	251		0.5					0

APPENDIX C - REFERRING TO SECTION SIX

CONVERSION OF K TO K_L AND K_M ASSUMING FIRST ORDER GROWTH RATES

$$\frac{dc}{dt} = - KA (c - c_\infty)$$

$$\text{but } c = \frac{100m}{\frac{m}{\rho} + V} = \frac{100m}{250} \quad g/cm^3$$

$$\therefore \quad dc = \frac{100V\, dm}{\left(\frac{m}{\rho} + V\right)^2}$$

$$\therefore \frac{100V\, dM}{dt \left(\frac{m}{\rho} + V\right)^2} = KA (c - c_\infty)$$

$$\text{But } \frac{dr}{dt} = K_L (c - c_\infty)$$

$$\therefore \quad dM = \frac{KA \left(\frac{m}{\rho} + V\right)^2}{V K_L \times 100} \, dr$$

$$\text{But } \frac{dM}{dr} = \rho_s 4\pi r^2$$

$$\rho_s 4\pi r^2 = \frac{K}{K_L} \frac{4\pi r^2 (250)^2}{\left(250 - \frac{m}{\rho_s}\right) 100}$$

$$K_L = K \frac{250^2}{(250\rho_s - m)\, 100}$$

Similarly, where $\frac{dm}{dt} = - K_m A (c - c_\infty)$

$$K_m = K \frac{250^2}{\left(250 - \frac{m}{\rho_s}\right) 100}$$

PROGRAM 1. ELLIOTT 803 VERSION OF ALGOL 60.

CRYSTALLISATION RATE OF PENTAERYTHRITOL IN A FLUIDISED BED

```
BEGIN INTEGER J,P;                    READ P;
BEGIN REAL ARRAY PERCENTNO,NO,VOL,SIGMA1,SIGMA2,SIGMA3(1:P),
D(1:2,1:P),M(1:2);
REAL SUM,FACTOR,U,R,Y,Q,FUNCTION,DIFFUNCTION,DELTAD,DELTAT,MEANA,VEL;
SWITCH S:=RETURN,CONTINUE;
REAL PROCEDURE AREA(Z);
VALUE Z; INTEGER Z;
BEGIN REAL A;                         A:=0;
FOR  J:=1 STEP 1 UNTIL P DO
A:=A+(22/7)*1.181*NO(J)*D(Z,J)**2;
AREA:=A;                              END;
FOR J:=1 STEP 1 UNTIL P DO READ PERCENTNO(J);
FOR J:=1 STEP 1 UNTIL P DO READ D(1,J);
READ M(1),M(2),DELTAT;
SUM:=0;
FOR J:=1 STEP 1 UNTIL P DO BEGIN
VOL(J):=PERCENTNO(J)*(11/21)*D(1,J)**3;
SUM:=SUM+VOL(J);                      END;
FACTOR:=M(1)/(1.396*SUM);
FOR J:=1 STEP 1 UNTIL P DO
NO(J):=FACTOR*PERCENTNO(J);
U:=0;
FOR J:=1 STEP 1 UNTIL P DO BEGIN
SIGMA1(J):=NO(J);
U:=SIGMA1(J)+U;                       END;
R:=0;
FOR J:=1 STEP 1 UNTIL P DO BEGIN
SIGMA2(J):=3*NO(J)*D(1,J);
R:=R+SIGMA2(J);
END;                                  Y:=0;
FOR J:=1 STEP 1 UNTIL P DO BEGIN
SIGMA3(J):=3*NO(J)*D(1,J)**2;
Y:=Y+SIGMA3(J);                       END;
Q:=(M(2)-M(1))/(1.396*(11/21));
DELTAD:=0.0001;
RETURN:FUNCTION:=U*(DELTAD)**3+R*(DELTAD)**2+Y*DELTAD-Q;
DIFFUNCTION:=(3*U*(DELTAD)**2+2*R*DELTAD+Y);
IF ABS(FUNCTION/DIFFUNCTION) LESS .0000001 THEN GOTO CONTINUE
ELSE BEGIN DELTAD:=DELTAD-((FUNCTION)/(DIFFUNCTION));
GOTO RETURN;                          END;
CONTINUE:FOR J:=1 STEP 1 UNTIL P DO
D(2,J):=D(1,J)+DELTAD;
MEANA:=(AREA(1)+AREA(2))/2;
VEL:=DELTAD/(2*DELTAT);
PRINT FREEPOINT(7),DELTAD,SAMELINE,££S5??,FREEPOINT(5),MEANA,££S??,
SCALED(4),VEL;
END;END;END;
```

PROGRAM 2. ELLIOTT 803 VERSION OF ALGOL 60.

CRYSTALLISATION RATE OF PENTAERYTHRITOL IN A SEEDED CELL'

```
BEGIN INTEGER I,J,P,N'
READ P,N'
BEGIN REAL ARRAY PERCENTNO,NO,VOL,SIGMA1,SIGMA2,SIGMA3(1:P),
TIME,M,V(1:N),
SCALE,C(1:N+1),
R,Y,Q,FUNCTION,DIFFUNCTION,DELTAD,MEANA,DELTAT,K,VEL,SUPERSATURATION(1:N-1),
D(1:N,1:P)'
REAL B,F,G,H,SUM,FACTOR,U,EVAPN'
SWITCH S:=RETURN,CONTINUE'
REAL PROCEDURE AREA(Z)'
VALUE Z' INTEGER Z'
BEGIN REAL A'
A:=0'
FOR J:=1 STEP 1 UNTIL P DO
A:=A+(22/7)*1.181*NO(J)*D(Z,J)**2'
AREA:=A'
END'
FOR J:=1 STEP 1 UNTIL P DO READ PERCENTNO(J)'
FOR I:=1 DO
FOR J:=1 STEP 1 UNTIL P DO READ D(I,J)'
FOR I:=1 STEP 1 UNTIL N+1 DO READ SCALE(I)'
FOR I:=1 STEP 1 UNTIL N DO READ TIME(I)'
READ B,F,G,H,EVAPN'
SUM:=0'
FOR I:=1 DO
FOR J:=1 STEP 1 UNTIL P DO BEGIN
VOL(J):=PERCENTNO(J)*(11/21)*D(I,J)**3'
SUM:=SUM+VOL(J)'
END'
M(1):=H'
FACTOR:=M(1)/(1.396*SUM)'
FOR J:=1 STEP 1 UNTIL P DO
NO(J):=FACTOR*PERCENTNO(J)'
FOR I:=1 STEP 1 UNTIL N+1 DO
C(I):=F+B*SCALE(I)+G*SCALE(I)**2'
FOR I:=1 STEP 1 UNTIL N DO
V(I):=250-1.7908*C(1)-TIME(I)*EVAPN'
PRINT £
```

TABLE: _____ RUN NO.: _____

TEMP: _____ C CELL: _____ STIRRER SPEED: _____ R.P.M.

SEED: BATCH _____ SIEVE FRACTION: _____

PROGRAM 2. (CONT.)

```
 CONCN.  TOTAL  CRYSTAL  INCREASE OF     MEAN    GROWTH RATE  MEAN    GROWTH
  %M/V   TIME    MASS    CRYSTAL EQUIV.  AREA    CONSTANT     SUPER-  RATE
         MINS.  (GRAMS)  DIAMETER(CM)    CM      K(MIN CM )   SATN.   CM/MIN.
-----------------------------------------------------------------------------?
FOR I:=1 STEP 1 UNTIL N-1 DO BEGIN
M(I+1):=M(I)+((C(I)-C(I+1))/(100*V(I+1)))*((C(1)*2.5-M(I)+H)/1.396+V(I+1))**2'
U:=0'
FOR J:=1 STEP 1 UNTIL P DO BEGIN
SIGMA1(J):=NO(J)'
U:=SIGMA1(J)+U'
END'
R(I):=0'
FOR J:=1 STEP 1 UNTIL P DO BEGIN
SIGMA2(J):=3*NO(J)*D(I,J)'
R(I):=R(I)+SIGMA2(J)'
END'
Y(I):=0'
FOR J:=1 STEP 1 UNTIL P DO BEGIN
SIGMA3(J):=3*NO(J)*D(I,J)**2'
Y(I):=Y(I)+SIGMA3(J)'
END'
Q(I):=(M(I+1)-M(I))/(1.396*(11/21))'
DELTAD(I):=0.0001'
RETURN:FUNCTION(I):=U*(DELTAD(I))**3+R(I)*(DELTAD(I))**2+Y(I)*DELTAD(I)-Q(I)'
DIFFUNCTION(I):=(3*U*(DELTAD(I))**2+2*R(I)*DELTAD(I)+Y(I))'
IF ABS(FUNCTION(I)/DIFFUNCTION(I)) LESS .0000001
THEN GOTO CONTINUE
ELSE BEGIN DELTAD(I):=DELTAD(I)-((FUNCTION(I))/(DIFFUNCTION(I)))'
GOTO RETURN'
END'
CONTINUE:FOR J:=1 STEP 1 UNTIL P DO
D(I+1,J):=D(I,J)+DELTAD(I)'
MEANA(I):=(AREA(I+1)+AREA(I))/2'
DELTAT(I):=TIME(I+1)-TIME(I)'
K(I):=(1/(MEANA(I)*DELTAT(I)))*LN((C(I)-C(N+1))/(C(I+1)-C(N+1)))'
VEL(I):=DELTAD(I)/(2*DELTAT(I))'
SUPERSATURATION(I):=((C(I+1)+C(I))/2-C(N+1))/C(N+1)'
PRINT FREEPOINT(5),C(I),SAMELINE,££S??,FREEPOINT(5),TIME(I),££S??,FREEPOINT(4),M(I)
££LS26??,FREEPOINT(7),DELTAD(I),SAMELINE,££S5??,FREEPOINT(5),MEANA(I),££S??,
SCALED(5),K(I),££S2??,FREEPOINT(4),SUPERSATURATION(I),££S2??,SCALED(4),VEL(I)'
END'
PRINT FREEPOINT(5),C(N),SAMELINE,££S??,FREEPOINT(5),TIME(N),££S??,FREEPOINT(4),M(N)
PRINT £
-----------------------------------------------------------------------------?
PRINT££L?EQUILIBRIUM CONCENTRATION = ?,SAMELINE,FREEPOINT(5),C(N+1)'
END'END'END'
END'END'
```

PROGRAM 3: ELLIOTT 803 VERSION OF ALGOL 60.

```
COULTER COUNTER DIAMETER CORRECTION FOR 280μ TUBE ORIENTATION A'
BEGIN INTEGER D'
REAL V,F,B,L,Y,FUNCTION,DIFFUNCTION,E,TRUED'
SWITCH S:=RETURN,CONTINUE'
FOR D:=5 STEP 1 UNTIL 150 DO BEGIN
V:=(11/21)*(D**3)'
F:=V/10809400'
B:=87102/10809400'
L:=D'
RETURN:Y:=F+(B*L)'
FUNCTION:=(1+L/248.2)/(1-L/248.2)-1-Y-(Y**2)/2-(Y**3)/6
-(Y**4)/24-(Y**5)/120-(Y**6)/720-(Y**7)/5040-(Y**8)/40320'
DIFFUNCTION:=(2/248.2)/((1-(1/248.2)*L)**2)-B-(Y*B)-(Y**2)*(B/2)
-(Y**3)*(B/6)-(Y**4)*(B/24)-(Y**5)*(B/120)-(Y**6)*(B/720)-(Y**7)*(B/5040)'
IF ABS(FUNCTION/DIFFUNCTION) LESS .01
 THEN GOTO CONTINUE
ELSE BEGIN L:=L-(FUNCTION/DIFFUNCTION)'
GOTO RETURN'
END'
CONTINUE:E:=0.900*(L**3)'
TRUED:=(E)**(1/3)'
PRINT FREEPOINT(3),D,SAMELINE,ΣΣS??,FREEPOINT(5),L,ΣΣS??,
FREEPOINT(5),TRUED'
END,END,END'
```

PROGRAM 4: ELLIOTT 803 VERSION OF ALGOL 60.

```
COULTER COUNTER DIAMETER CORRECTION FOR 280μ TUBE ORIENTATION B'
BEGIN INTEGER D'
REAL V,G,L,FUNCTION,DIFFUNCTION,E,A,TRUED'
SWITCH S:=RETURN,CONTINUE'
FOR D:=5 STEP 1 UNTIL 150 DO BEGIN
V:=(11/21)*(D**3)'
A:=(22/7)*(140**2)'
L:=D'
RETURN:G:=SQRT((A/(0.707*L**2))-1)'
FUNCTION:=(0.707*V*L*G)/A**2+(((L**2)*0.707*G)/A)-ARCTAN(1/G)'
DIFFUNCTION:=(G/A)*((0.707*V)/A+1.414*L)-(V/(A*L)+2)/(L*G)'
IF ABS(FUNCTION/DIFFUNCTION) LESS .01
THEN GOTO CONTINUE
ELSE BEGIN L:=L-(FUNCTION/DIFFUNCTION)'
GOTO RETURN'
END'
CONTINUE:E:=0.900*(L**3)'
TRUED:=(E)**(1/3)'
PRINT FREEPOINT(3),D,
SAMELINE, ΣΣS??,
   FREEPOINT(5),L,ΣΣS??,
FREEPOINT(5),TRUED'
END,END,END'
```

APPENDIX: C

Evaporation in Stirred Cells

Procedure

Evaporation rates for Cells C and S under experimental conditions were obtained as follows. A solution of a known approximate concentration of Batch C, chosen to be undersaturated under experimental conditions, was accurately made up in a $250cm^3$ pyrex graduated flask and dissolved by heating. The cell was heated to the temperature to be studied and the solution poured in. When the temperature was stable the refractometer scale was read and the concentration obtained from the isothermal calibration correlation. The mass of solute, m, in this $250cm^3$ of solution was therefore found. The solution was stirred at 2000 r.p.m. and the time taken for suitable increases of the solution concentration. The total solution volume, v, for this particular concentration, c, was obtained from $v = \frac{m}{c} \times 100$ where m remains constant throughout. From the decrease in total volume over a period of time the evaporation rate is calculated in cm^3/min. This is checked at different intervals to guard against errors of scale reading, but the greatest concentration difference should be the most accurate. It was usually found to take many days to obtain an appreciable concentration increase.

Example

Cell C $T_o = 70.0^oC$ Stirrer Speed = 2000r.p.m.

Time (mins)	Scale Reading S	o % m/v
0	63.75	20.301
130	64.25	20.445
900	66.80	21.178
1020	67.25	21.307
1260	68.40	21.637

$$m = 2.50 \times 20.301$$

$$= 50.753$$

$$\therefore v_1 = 250$$

$$v_2 = 248.2$$

$$v_3 = 239.6$$

$$v_4 = 238.1$$

$$v_5 = 234.5$$

Evaporation Rates cm^3/min

$$v_1/v_2 = 0.01384$$
$$v_2/v_3 = 0.01116$$
$$v_3/v_4 = 0.01250$$
$$v_4/v_5 = 0.01500$$

$$v_1/v_5 = 0.0123$$

Experimental Results

T_o °C	Evaporation cm^3/min.	
	Cell C	Cell S
45.0	0.0030	0.000238
50.0	0.0063	
55.0	0.0083	
60.0	0.0101	
65.0	0.0123	0.000500
70.0	0.0123	0.000595
75.0	0.0173	

Discussion

On the assumption that the loss of solvent is proportional to the difference in vapour pressure between the cell solution and the ambient, the experimental values were plotted figure 6.4, as log (evaporation) vs $(\frac{1}{T})$ where T is absolute temperature (°K), as is suggested for vapour pressure for pure substances by the Clausius-Clapeyron equation.

It can be seen that the rate of evaporation in the Cell S is far less than that of Cell C under the same conditions. This would be expected as the teflon bush was less worn with Cell S, being high on the stirrer shaft near the motor, whereas the motor was less rigid with Cell C and the bush lower on the shaft which was inclined to whip.

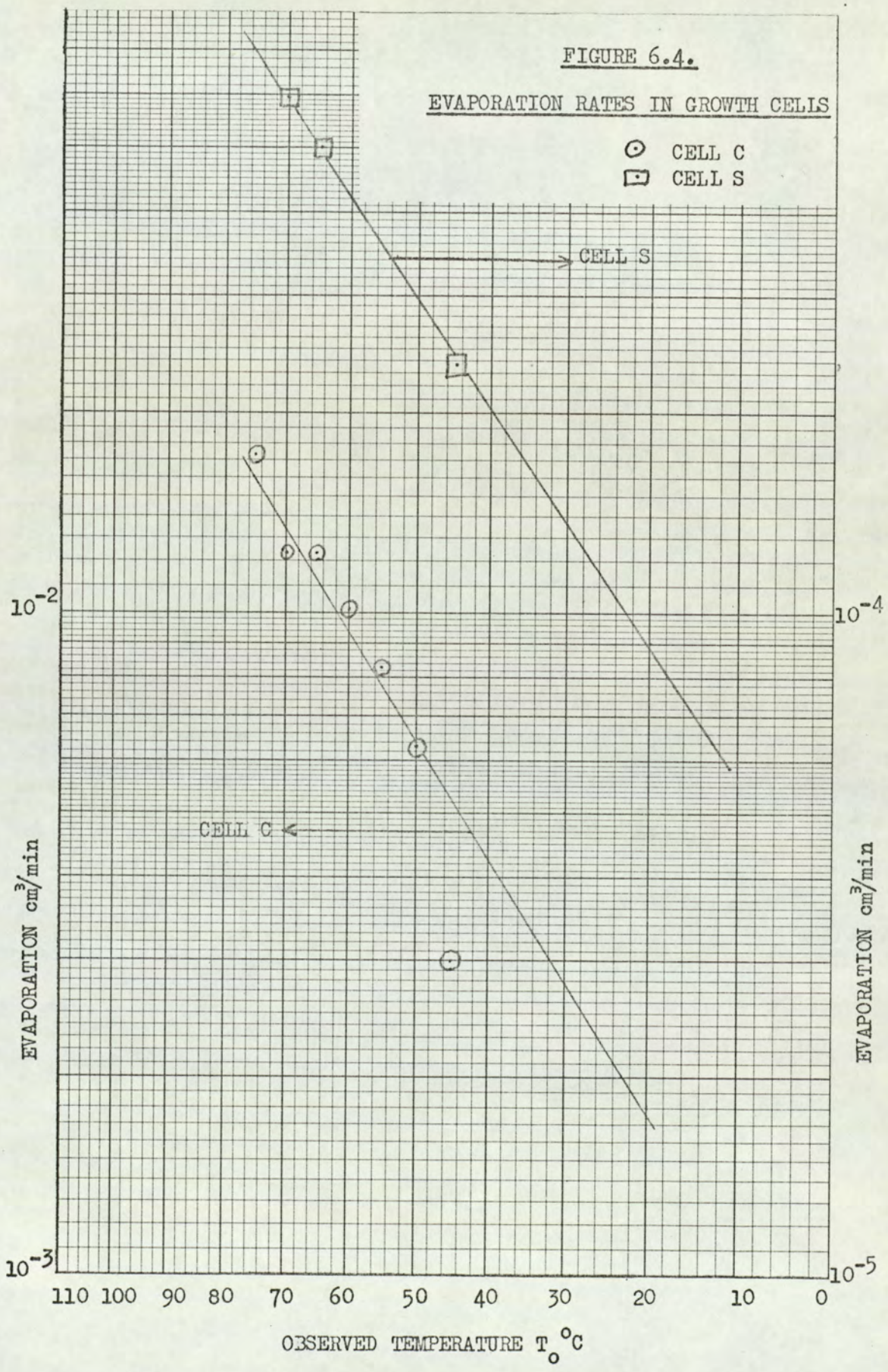


FIGURE 6.4.
EVAPORATION RATES IN GROWTH CELLS
CELL C
CELL S
CELL S
CELL C
10^{-2}
10^{-3}
10^{-4}
10^{-5}
EVAPORATION cm³/min
EVAPORATION cm³/min
110 100 90 80 70 60 50 40 30 20 10 0
OBSERVED TEMPERATURE T_o °C

TABLE: 29

Evaporation values interpolated from figure 6.4.

T_o °C	Evaporation cm^3/min	
	Cell S	Cell C
20	0.00009	0.0018
25	0.00010	0.0022
30	0.00013	0.0028
35	0.00016	0.0035
40	0.00019	0.0043
45	0.00024	0.0053
50	0.00029	0.0064
55	0.00035	0.0078
60	0.00042	0.0094
65	0.00050	0.0112
70	0.00060	0.0135
75	0.00080	0.0160

APPENDIX D - CRYSTAL GROWTH RATE RESULTS

TABLE: 31.

PRELIMINARY FLUIDISED BED RESULTS

RUN No.	TEMP. °C	u cm/s	SEED D μ	PROD. D μ	TIME MIN.	s	GROWTH VELOCITY g x 10⁶	
							DIRECT MEASUREMENT	MASS BASIS
F.C.7	59.5	0.180	94	111	112	0.124	7.60	
F.C.8	56.7	0.175	94	122	313	0.113	4.47	
F.C.9	46.9	0.215	111	137	322	0.220	4.04	
F.C.10	43.4	0.203	111	129	292	0.231	3.08	
F.C.11	39.9	0.165	94	105	296	0.302	1.86	
F.C.12	37.9	0.167	111	121	273	0.430	1.83	
F.C.13	35.6	0.268	132	157	226	0.545	5.50	2.45
F.C.14	51.6	0.149	79	97	50	0.333	18.0	
F.C.15	51.7	0.181	82	102	132	0.262	7.60	
F.C.16	32.7	0.163	111	139	1800	0.558	0.78	
F.C.17	65.5	0.241	132	150	140	0.066	6.42	
F.C.18	68.9	0.210	111	172	132	0.132	23.1	
F.C.19	71.1	0.600	163	217	136	0.101	19.8	19.8
F.C.20	67.0	0.208	110	152	208	0.082	10.1	5.14
F.C.21	63.0	0.195	114	140	254	0.138	5.10	6.11
F.C.22	54.4	0.211	111	144	383	0.184	4.31	2.75
F.C.23	70.2	0.318	159	172	147	0.076	4.42	7.94
F.C.24	58.4	0.210	132	151	122	0.200	7.80	7.86
F.C.25	29.5	0.210	160	161	1144	0.485		1.12
F.C.26	34.0	0.210	122	135	1212	0.326	0.57	
F.C.27	37.6	0.090	94	123	1284	0.372	1.13	2.34

TABLE:32 EXAMPLE FLUIDISED BED DATA WITH MODIFIED APPARATUS

RUN: F.E.1.	TEMP 70.0°C	2 LITRES SOLUTION 30.00% m/v	BATCH E
SEED: 7.96g BATCH E	PREPARED SIEVE FRACTION 124-150μ		
MAGNETIC STIRRER POSN. 2	HEATER POSN. 3.5	T.U.3. 1 = 86°C	T.U.3. 2 = 73°C

TIME MINS	ROTAMETER METRIC NO.7K	CELL TEMP. °C	RESERVOIR TEMP. °C	REFRACTOMETER SCALE	REFRACTOMETER TEMP. °C	HEATING TAPE CONTROL VOLTS	MAKE UP WATER
0	1.2	70.4	80	92.0	72.8	70	-
5	1.2	69.0	80		71.2	80	-
8	1.1	69.5	80		74.2	70	-
10	1.1	70.0	80	90.7	73.6	60	-
13	1.1	70.4	80	90.8	73.7	50	-
15	1.1	70.2	80	92.3	72.0	50	-
19	1.1	70.0	80	92.6	71.5	60	-
24	2.4	70.4	80			40	-
32	3.2	69.4	80	91.9	72.8	50	-
36	3.2	70.2	80	91.9	72.8	50	-
41	2.8	70.0	80		72.3	50	-
45	2.8	70.0	80	92.3	72.0	50	-
55	2.8	70.0				50	-
60	RUN COMPLETED						

WEIGHT OF DRY EMPTY CELL	= 286.88g
WEIGHT OF CELL + PRODUCT CRYSTALS (WET)	= 309.26g
WEIGHT OF CELL + PRODUCT CRYSTALS (DRY)	= 303.13g

TABLE: 35

EXAMPLE EXPERIMENTAL DATA FOR STIRRED CELLS

EXPT. No. 118		RUN No. : R.P.E.16
CELL C	TEMP. 50.0°C	STIRRER SPEED: 2000 R.P.M.
SOLUTION: 17.75%m/v P.E.		SEED: 89-105 μ P.E. PREPARED

DATA	DESCRIPTION
15 20	P = No. of Mean Diameters of size analysis N = No. of readings of time
4.24, 8.46, 13.9, 4.4, 3.4, 5.1, 13.4, 9.2, 4.6, 5.4, 1.7, 8.7, 13.5, 2.1, 1.9	Array of % No. of Mean Diameters of size analysis.
0.0126, 0.0114, 0.0102, 0.00896, 0.00773, 0.0065, 0.00519, 0.00415, 0.00331, 0.00264, 0.00212, 0.00172, 0.00141, 0.00119, 0.000989	Array of Mean diameters of size analysis (cm.).
65.1, 64.5, 64, 63, 62.5, 62, 61.5, 60.5, 60, 59.5, 59, 58.5, 58, 57.5, 57, 56.5, 56, 55.5, 55, 54.3, 53.1	Array of readings of refractometer scale S, + equilibrium scale reading
0, 37, 133, 158, 180, 198, 240, 258, 270, 282, 295, 310, 326, 344, 360, 374, 393, 408, 465	Array of readings of time, t.
0.3058 -1.643 -0.0001699	B, F, G } Calibration coefficients $e = F + B.S. + G.S.^2$
1.969	H Initial seed mass (g).
0.0064	Evaporation rate $cm.^3$/ min.

TABLE: 36

QUALITATIVE RESULTS OF SEEDED BATCH B SOLUTIONS

STIRRED AT 2000 R.P.M. IN CELL C AT 60.0°C

RUN NO:	R.B.3			R.B.4			R.A.6		
SEED:	2g UNCLASSIFIED BATCH B			2g UNCLASSIFIED BATCH A			2g UNCLASSIFIED BATCH A		
	SCALE S	σ %m/v	TIME MIN.	SCALE S	σ %m/v	TIME MIN.	SCALE S	σ %m/v	TIME MIN.
	83.8	24.286	0	87.4	25.296	0	82.9	24.033	0
	83.0	24.061	60	86.0	24.904	32	82.0	23.780	16
	82.5	23.921	160	85.0	24.623	63	81.0	23.498	51
							80.0	23.217	110
							79.0	22.934	155
							78.2	22.708	190
							77.0	22.369	265
							75.8	22.029	320
							75.0	21.802	370

RUN NO:	R.B.5			R.C.2		
SEED:	2g UNCLASSIFIED BATCH B			2g UNCLASSIFIED BATCH C		
	SCALE S	σ %m/v	TIME MIN.	SCALE S	σ %m/v	TIME MIN.
	82.8	24.005	0	82.9	24.033	0
	82.0	23.780	145	82.0	23.78	7
	81.0	23.498	330	81.0	23.498	35
				79.0	22.934	110
				78.0	22.651	145

TABLE: 37

QUALITATIVE RESULTS OF SEEDED SOLUTIONS STIRRED AT 2000 R.P.M. AT 70.0°C

RUN NO:	R.G.1			R.G.2		
SEED:	2g UNCLASSIFIED BATCH G			2g UNCLASSIFIED BATCH G		
CELL	C			S		
	SCALE S	σ %m/v	TIME MIN.	SCALE S	σ %m/v	TIME MIN.
	98.4	30.097	0	97.6	29.875	0
	98.0	29.986	7	96.7	29.624	30
	97.5	29.847	35	96.2	29.485	50
	97.0	29.708	50	95.7	29.346	70
	96.5	29.569	65	93.2	28.650	150
	96.0	29.430	85			
	95.0	29.152	120			

RUN NO:	R.F.2			R.F.5		
SEED:	2g 89-105μ			2g 89-105μ		
CELL	C			C		
	SCALE S	σ %m/v	TIME MIN.	SCALE S	σ %m/v	TIME MIN.
	97.0	29.708	0	97.3	29.791	0
	96.5	29.569	40	97.0	29.708	14
	96.0	29.430	68	96.5	29.569	42
	95.1	29.179	120	96.0	29.430	65
				95.5	29.291	90
				95.0	29.152	120

TABLE: 38 RUN NO.: R.C.3

TEMP: 60.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH C PREPARED SIEVE FRACTION: 75-89μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
24.033	.00000	2.000					
			.0005001	2605.0	6.2387@-07	.1947	6.252@-06
23.780	40.000	2.764					
			.0004060	3532.1	3.6637@-07	.1814	3.384@-06
23.498	100.00	3.608					
			.0003215	4427.4	2.9284@-07	.1673	2.473@-06
23.217	165.00	4.448					
			.0002691	5254.8	2.9223@-07	.1532	2.242@-06
22.934	225.00	5.282					
			.0002330	6028.6	2.1068@-07	.1391	1.456@-06
22.652	305.00	6.112					

EQUILIBRIUM CONCENTRATION = 20.010 INITIAL SEED AREA = 2145

TABLE: 39 RUN NO.: R.C.4

TEMP: 60.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH C PREPARED SIEVE FRACTION: 75-89μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
24.033	.00000	2.000					
			.0005003	2605.1	8.3179@-07	.1947	8.338@-06
23.780	30.000	2.764					
			.0004060	3532.3	3.1401@-07	.1814	2.900@-06
23.498	100.00	3.608					
			.0003215	4427.6	2.9283@-07	.1673	2.473@-06
23.217	165.00	4.448					
			.0002690	5255.0	2.6974@-07	.1532	2.070@-06
22.934	230.00	5.282					
			.0002330	6028.8	2.5929@-07	.1391	1.793@-06
22.652	295.00	6.112					

EQUILIBRIUM CONCENTRATION = 20.010 INITIAL SEED AREA = 2145

TABLE: 40 RUN NO.: R.C.5

TEMP: 60.0°C CELL: A STIRRER SPEED: 2000 R.P.M.

SEED: BATCH C PREPARED SIEVE FRACTION: 44-64μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
24.216	.00000	2.000					
			.0005036	2291.9	3.4256@-07	.2046	3.597@-06
23.991	70.000	2.680					
			.0002652	2706.2	2.6587@-07	.1954	2.652@-06
23.850	120.00	3.104					
			.0002383	3005.0	4.9700@-07	.1884	4.766@-06
23.710	145.00	3.527					
			.0002171	3291.6	4.7158@-07	.1814	4.342@-06
23.569	170.00	3.949					
			.0001999	3567.5	3.2352@-07	.1743	2.856@-06
23.428	205.00	4.371					

EQUILIBRIUM CONCENTRATION = 20.010 INITIAL SEED AREA = 2030

TABLE: 41 RUN NO.: R.C.6

TEMP: 60.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH C PREPARED SIEVE FRACTION: 44-64μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
24.146	.00000	2.000					
			.0002025	2131.1	6.4459@-06	.2046	6.751@-05
24.061	1.5000	2.255					
			.0003004	2392.2	8.4393@-07	.1990	8.582@-06
23.921	19.000	2.679					
			.0001096	2614.5	5.5438@-07	.1940	5.480@-06
23.865	29.000	2.849					
			.0001552	2766.9	6.6707@-07	.1905	6.465@-06
23.780	41.000	3.102					
			.0002379	3003.4	4.6932@-07	.1849	4.405@-06
23.639	68.000	3.524					
			.0002166	3289.2	6.6859@-07	.1779	6.018@-06
23.498	86.000	3.945					
			.0001215	3510.9	4.3678@-07	.1722	3.798@-06
23.414	102.00	4.197					
			.0000779	3646.0	5.0901@-07	.1687	4.330@-06
23.358	111.00	4.365					
			.0001850	3830.0	4.6818@-07	.1638	3.855@-06
23.217	135.00	4.784					
			.0001729	4087.2	4.4036@-07	.1567	3.459@-06
23.075	160.00	5.201					
			.0001625	4336.8	4.0259@-07	.1497	3.010@-06
22.934	187.00	5.618					
			.0001535	4579.5	3.6030@-07	.1426	2.558@-06
22.793	217.00	6.033					
			.0001454	4815.9	2.2541@-07	.1356	1.515@-06
22.652	265.00	6.447					
			.0002708	5158.1	3.9915@-07	.1250	2.461@-06
22.369	320.00	7.272					
			.0001265	5492.8	5.6321@-07	.1144	3.162@-06
22.227	340.00	7.683					
			.0001214	5708.7	3.0419@-07	.1073	1.597@-06
22.086	378.00	8.092					
			.0001167	5920.4	2.8429@-07	.1002	1.389@-06
21.944	420.00	8.500					
			.0001123	6128.0	2.7601@-07	.0931	1.248@-06
21.802	465.00	8.907					
			.0001504	6371.4	1.7552@-07	.0846	7.164@-07
21.604	570.00	9.473					

EQUILIBRIUM CONCENTRATION = 20.010 INITIAL SEED AREA = 2030

TABLE: 42 RUN NO.: R.C.7

TEMP: 50.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH C PREPARED SIEVE FRACTION: 44-64 μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
19.462	.00000	2.000					
			.0002266	2143.5	4.2815@-07	.2851	4.532@-06
19.363	25.000	2.287					
			.0002872	2410.6	2.5438@-07	.2772	2.611@-06
19.222	80.000	2.696					
			.0002545	2711.5	3.2193@-07	.2678	3.181@-06
19.081	120.00	3.103					
			.0002294	2998.8	2.4139@-07	.2585	2.294@-06
18.939	170.00	3.510					
			.0002095	3274.4	3.2779@-07	.2491	2.993@-06
18.798	205.00	3.915					
			.0001932	3539.8	3.1516@-07	.2398	2.760@-06
18.657	240.00	4.319					
			.0001796	3796.3	2.6768@-07	.2304	2.246@-06
18.515	280.00	4.722					
			.0001681	4044.7	2.6198@-07	.2210	2.101@-06
18.374	320.00	5.124					
			.0003079	4402.0	2.2874@-07	.2070	1.711@-06
18.090	410.00	5.924					
			.0001419	4749.8	2.0471@-07	.1929	1.419@-06
17.949	460.00	6.323					
			.0003600	5249.4	2.0936@-07	.1713	1.278@-06
17.438	640.00	7.748					
			.0008364	6439.2	1.5034@-07	.1149	5.786@-07
16.245	1380.0	11.00					
			.0008174	7330.3	1.1370@-07	.0707	2.731@-07
16.103	1540.0	11.38					
			.0000848	7504.9	1.0798@-07	.0613	2.232@-07
15.961	1730.0	11.75					

EQUILIBRIUM CONCENTRATION = 15.106 INITIAL SEED AREA = 2030

TABLE: 43. RUN NO.: R.C.8

TEMP: 40.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH C PREPARED SIEVE FRACTION: 44-64μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
17.154	.00000	2.000					
			.0008259	2478.9	6.1854@-07	.4675	8.259@-06
17.033	50.000	3.202					
			.0002199	3063.8	6.7632@-07	.4436	8.458@-06
16.893	63.000	3.600					
			.0002018	3331.6	5.9371@-07	.4316	7.206@-06
16.753	77.000	3.997					
			.0001868	3590.1	6.1046@-07	.4197	7.186@-06
16.612	90.000	4.393					
			.0001743	3840.3	5.4564@-07	.4077	6.224@-06
16.471	104.00	4.789					
			.0001635	4083.1	5.6957@-07	.3958	6.290@-06
16.331	117.00	5.183					
			.0001543	4319.3	5.5540@-07	.3838	5.933@-06
16.190	130.00	5.577					
			.0001462	4549.4	6.4346@-07	.3718	6.643@-06
16.050	141.00	5.970					
			.0001390	4773.9	2.9050@-07	.3599	2.895@-06
15.909	165.00	6.362					
			.0001326	4993.3	5.3057@-07	.3479	5.098@-06
15.768	178.00	6.753					
			.0002487	5312.3	3.6985@-07	.3299	3.360@-06
15.486	215.00	7.534					
			.0001169	5625.0	2.7343@-07	.3119	2.338@-06
15.345	240.00	7.922					
			.0001126	5827.4	4.2903@-07	.2999	3.519@-06
15.205	256.00	8.310					
			.0001086	6026.1	3.1441@-07	.2879	2.469@-06
15.064	278.00	8.697					
			.0001050	6221.5	2.4977@-07	.2759	1.874@-06
14.922	306.00	9.083					
			.0002002	6507.5	2.4693@-07	.2579	1.725@-06
14.640	364.00	9.852					
			.0002429	6934.2	2.0417@-07	.2303	1.265@-06
14.273	460.00	10.85					
			.0009646	8200.9	1.5783@-07	.1400	5.242@-07
12.517	1380.0	15.51					
			.0000659	9305.5	3.6651@-08	.0592	5.495@-08
12.376	1980.0	15.87					
			.0000891	9483.7	4.4845@-08	.0447	4.949@-08
12.177	2880.0	16.37					

EQUILIBRIUM CONCENTRATION = 11.751 INITIAL SEED AREA = 2030

TABLE: 44 RUN NO.: R.C.9

TEMP: 60.0°C CELL: A STIRRER SPEED: 500 R.P.M.

SEED: BATCH C PREPARED SIEVE FRACTION: 44-64μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
24.188	.00000	2.000					
			.0001371	2097.9	9.2251@-07	.2074	9.796@-06
24.132	7.0000	2.170					
			.0003091	2327.7	8.2835@-07	.2025	8.586@-06
23.991	25.000	2.595					
			.0002715	2644.2	5.4420@-07	.1954	5.429@-06
23.850	50.000	3.019					
			.0002432	2945.8	3.6214@-07	.1884	3.474@-06
23.710	85.000	3.442					
			.0002210	3234.6	3.9990@-07	.1814	3.683@-06
23.569	115.00	3.864					
			.0002031	3512.5	2.8751@-07	.1743	2.539@-06
23.428	155.00	4.286					

EQUILIBRIUM CONCENTRATION = 20.010 INITIAL SEED AREA = 2030

TABLE: 45 RUN NO.: R.C.10

TEMP: 60.0°C CELL: C STIRRER SPEED: 500 R.P.M.

SEED: BATCH C PREPARED SIEVE FRACTION: 44-64μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
24.146	.00000	2.000					
			.0002025	2131.1	1.9338@-06	.2046	2.025@-05
24.061	5.0000	2.255					
			.0003003	2392.1	7.3845@-07	.1990	7.508@-06
23.921	25.000	2.679					
			.0002648	2705.0	6.1574@-07	.1919	6.019@-06
23.780	47.000	3.102					
			.0002378	3003.3	5.0687@-07	.1849	4.757@-06
23.639	72.000	3.524					
			.0002166	3289.2	6.0174@-07	.1779	5.415@-06
23.498	92.000	3.945					
			.0001994	3564.2	5.0305@-07	.1708	4.334@-06
23.358	115.00	4.365					
			.0001849	3829.7	2.8093@-07	.1638	2.312@-06
23.217	155.00	4.783					
			.0001728	4086.7	3.1458@-07	.1567	2.468@-06
23.075	190.00	5.201					
			.0001623	4336.0	2.1744@-07	.1497	1.623@-06
22.934	240.00	5.617					
			.0001532	4578.3	2.7030@-07	.1426	1.915@-06
22.793	280.00	6.031					
			.0001452	4814.3	2.1647@-07	.1356	1.452@-06
22.652	330.00	6.444					
			.0001380	5044.4	1.6775@-07	.1285	1.062@-06
22.510	395.00	6.856					
			.0000534	5202.7	1.4663@-07	.1235	8.895@-07
22.454	425.00	7.020					

EQUILIBRIUM CONCENTRATION = + 20.010 INITIAL SEED AREA = 2030

TABLE: 46 RUN NO.: R.C.11

TEMP: 40.0°C CELL: C STIRRER SPEED: 500 R.P.M.

SEED: BATCH C PREPARED SIEVE FRACTION: 44-64μ

CONCN. ℳ/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
13.877	.00000	2.000					
			.0000639	2061.5	1.0993@-08	.2001	5.918@-08
13.849	540.00	2.078					
			.0001208	2153.1	1.2868@-08	.1964	6.709@-08
13.792	1440.0	2.231					
			.0002183	2328.2	1.6131@-08	.1891	7.909@-08
13.679	2820.0	2.532					

EQUILIBRIUM CONCENTRATION = 11.552 INITIAL SEED AREA = 2030

TABLE: 47 RUN NO.: R.C.12

TEMP: 70.0°C CELL: A STIRRER SPEED: 500 R.P.M.

SEED: BATCH C PREPARED SIEVE FRACTION: 44-64 μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
29.986	.00000	2.000					
			.0002102	2135.0	3.5774@-06	.1381	3.504@-05
29.902	3.0000	2.265					
			.0003107	2406.2	4.6801@-06	.1339	4.439@-05
29.763	6.5000	2.707					
			.0002731	2730.9	2.3128@-06	.1286	2.101@-05
29.624	13.000	3.147					
			.0002448	3040.3	1.7612@-06	.1234	1.530@-05
29.485	21.000	3.587					
			.0002226	3336.5	1.5789@-06	.1181	1.309@-05
29.346	29.500	4.026					
			.0002047	3621.5	1.9926@-06	.1128	1.574@-05
29.207	36.000	4.463					
			.0001898	3896.6	1.1488@-06	.1075	8.628@-06
29.068	47.000	4.901					
			.0001601	4150.1	7.0053@-07	.1025	5.002@-06
28.943	63.000	5.293					
			.0001676	4396.1	5.8911@-07	.0974	3.991@-06
28.803	84.000	5.729					
			.0001582	4648.4	4.7615@-07	.0921	3.042@-06
28.664	110.00	6.163					
			.0001500	4894.2	4.6235@-07	.0868	2.777@-06
28.524	137.00	6.597					
			.0001427	5134.1	4.2273@-07	.0815	2.378@-06
28.385	167.00	7.030					
			.0001362	5368.6	4.1872@-07	.0762	2.196@-06
28.245	198.00	7.462					
			.0001303	5598.1	2.8486@-07	.0709	1.386@-06
28.105	245.00	7.893					
			.0001251	5823.1	2.1420@-07	.0656	9.626@-07
27.965	310.00	8.323					
			.0001203	6043.7	2.4345@-07	.0603	1.003@-06
27.825	370.00	8.753					
			.0001159	6260.3	1.9346@-07	.0550	7.243@-07
27.686	450.00	9.181					

EQUILIBRIUM CONCENTRATION = 26.310 INITIAL SEED AREA = 2030

TABLE: 48 RUN NO.: R.C.13

TEMP: 70.0°C CELL: C STIRRER SPEED: 500 R.P.M.

SEED: BATCH C PREPARED SIEVE FRACTION: 44-64μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
29.930	.00000	2.000					
			.0002101	2135.0	4.3606@-06	.1360	4.203@-05
29.847	2.5000	2.265					
			.0003105	2406.0	2.3782@-06	.1318	2.218@-05
29.708	9.5000	2.706					
			.0002729	2730.5	2.7800@-06	.1265	2.481@-05
29.569	15.000	3.146					
			.0002446	3039.6	2.3903@-06	.1212	2.038@-05
29.430	21.000	3.585					
			.0002224	3335.5	1.9533@-06	.1160	1.588@-05
29.291	28.000	4.024					
			.0002044	3620.1	2.6416@-06	.1107	2.044@-05
29.152	33.000	4.461					
			.0001895	3894.8	1.4333@-06	.1054	1.053@-05
29.012	42.000	4.897					
			.0001770	4160.7	1.4135@-06	.1001	9.834@-06
28.873	51.000	5.332					
			.0001663	4418.7	1.2656@-06	.0948	8.313@-06
28.734	61.000	5.766					
			.0001569	4669.7	1.0577@-06	.0895	6.539@-06
28.594	73.000	6.200					
			.0001487	4914.1	6.7518@-07	.0842	3.914@-06
28.454	92.000	6.631					
			.0001414	5152.5	4.5054@-07	.0789	2.438@-06
28.315	121.00	7.062					
			.0001348	5385.2	3.9445@-07	.0736	1.983@-06
28.175	155.00	7.491					
			.0001289	5612.7	3.4695@-07	.0683	1.611@-06
28.035	195.00	7.919					
			.0001235	5835.2	2.6337@-07	.0629	1.123@-06
27.895	250.00	8.344					
			.0001186	6053.1	3.2474@-07	.0576	1.262@-06
27.756	297.00	8.768					
			.0001140	6266.5	2.2266@-07	.0523	7.805@-07
27.615	370.00	9.190					
			.0001519	6515.7	1.3148@-07	.0459	3.997@-07
27.419	560.00	9.775					

EQUILIBRIUM CONCENTRATION = 26.310 INITIAL SEED AREA = 2030.5

TABLE: 49 RUN NO.: R.C.14

TEMP: 70.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH C PREPARED SIEVE FRACTION: 44-64μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
30.097	.00000	2.000					
			.0005789	2334.3	3.6541@-06	.1392	3.618@-05
29.847	8.0000	2.796					
			.0002668	2794.7	2.8664@-06	.1318	2.668@-05
29.708	13.000	3.236					
			.0002399	3101.2	2.6925@-06	.1265	2.399@-05
29.569	18.000	3.676					
			.0002186	3395.0	2.1401@-06	.1212	1.822@-05
29.430	24.000	4.114					
			.0002013	3677.8	2.0668@-06	.1160	1.678@-05
29.291	30.000	4.552					
			.0001870	3950.8	1.7289@-06	.1107	1.336@-05
29.152	37.000	4.988					
			.0001748	4215.3	1.0836@-06	.1054	7.946@-06
29.012	48.000	5.424					
			.0001644	4472.1	1.1836@-06	.1001	8.219@-06
28.873	58.000	5.858					
			.0001553	4721.9	1.0766@-06	.0948	7.059@-06
28.734	69.000	6.291					
			.0001473	4965.3	1.0851@-06	.0895	6.697@-06
28.594	80.000	6.724					
			.0001403	5202.9	1.1015@-06	.0842	6.375@-06
28.454	91.000	7.155					
			.0001339	5435.2	8.8472@-07	.0789	4.783@-06
28.315	105.00	7.585					
			.0001282	5662.4	8.5033@-07	.0736	4.274@-06
28.175	120.00	8.014					
			.0002416	5993.2	8.2122@-07	.0656	3.660@-06
27.895	153.00	8.870					
			.0001140	6317.6	8.6021@-07	.0576	3.354@-06
27.756	170.00	9.295					
			.0001100	6527.9	7.0925@-07	.0523	2.501@-06
27.615	192.00	9.720					
			.0001063	6734.4	6.0185@-07	.0470	1.898@-06
27.475	220.00	10.14					
			.0001028	6937.3	5.2762@-07	.0416	1.469@-06
27.335	255.00	10.56					
			.0000996	7136.8	4.1210@-07	.0363	9.956@-07
27.195	305.00	10.98					
			.0000965	7333.0	5.8834@-07	.0310	1.207@-06
27.055	345.00	11.40					

EQUILIBRIUM CONCENTRATION = 26.310 INITIAL SEED AREA = 2030

TABLE: 50 RUN NO.: R.C.15

TEMP: 30.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH C PREPARED SIEVE FRACTION: 44-64μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
12.531	.00000	2.000					
			.0000631	2061.2	4.0033@-07	.3719	3.156@-06
12.503	10.000	2.077					
			.0002901	2240.6	1.1805@-07	.3627	9.065@-07
12.363	170.00	2.461					
			.0000535	2418.2	1.6328@-08	.3535	1.216@-07
12.335	390.00	2.537					
			.0007164	2873.2	5.2900@-08	.3274	3.618@-07
11.887	1380.0	3.748					
			.0001131	3373.6	6.7953@-08	.2982	4.188@-07
11.803	1515.0	3.974					
			.0001947	3580.0	7.3715@-08	.2851	4.327@-07
11.648	1740.0	4.385					
			.0000838	3770.8	6.2348@-08	.2728	3.491@-07
11.578	1860.0	4.572					
			.0005870	4269.1	6.5555@-08	.2381	3.156@-07
11.016	2790.0	6.050					
			.0001034	4791.5	6.5682@-08	.2011	2.651@-07
10.903	2985.0	6.343					
			.0000996	4955.4	7.3330@-08	.1888	2.768@-07
10.790	3165.0	6.635					
			.0000485	5076.6	9.0326@-08	.1795	3.234@-07
10.734	3240.0	6.780					
			.0000476	5156.4	3.8381@-08	.1734	1.323@-07
10.678	3420.0	6.925					
			.0004354	5574.3	6.7428@-08	.1394	1.814@-07
10.114	4620.0	8.359					
			.0002862	6216.4	6.5151@-08	.0852	1.037@-07
9.6901	6000.0	9.410					
			.0001569	6630.0	5.1343@-08	.0481	4.470@-08
9.4355	7755.0	10.02					
			.0000334	6812.2	2.9297@-08	.0310	1.663@-08
9.3790	8760.0	10.16					
			.0000245	6868.4	2.0102@-08	.0256	9.265@-09
9.3365	10080	10.26					
			.0000318	6923.2	3.1101@-08	.0202	1.104@-08
9.2799	11520	10.39					

EQUILIBRIUM CONCENTRATION = 9.1242 INITIAL SEED AREA = 2030

TABLE: 51 RUN NO.: R.C.16

TEMP: 70.0°C CELL: C STIRRER SPEED: 1000 R.P.M.

SEED: BATCH C PREPARED SIEVE FRACTION: 44-64μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
30.152	.00000	2.000					
			.0004023	2236.3	4.4010@-06	.1429	4.470@-05
29.986	4.5000	2.531					
			.0002869	2603.6	2.6880@-06	.1371	2.608@-05
29.847	10.000	2.972					
			.0002553	2918.8	2.4951@-06	.1318	2.321@-05
29.708	15.500	3.412					
			.0002309	3220.1	2.5931@-06	.1265	2.309@-05
29.569	20.500	3.851					
			.0002113	3509.3	2.0704@-06	.1212	1.761@-05
29.430	26.500	4.289					
			.0001953	3788.2	2.4078@-06	.1160	1.953@-05
29.291	31.500	4.727					
			.0001819	4057.8	1.8128@-06	.1107	1.400@-05
29.152	38.000	5.163					
			.0001705	4319.2	1.2925@-06	.1054	9.474@-06
29.012	47.000	5.598					
			.0001607	4573.2	1.1574@-06	.1001	8.033@-06
28.873	57.000	6.032					
			.0001521	4820.5	1.4501@-06	.0948	9.504@-06
28.734	65.000	6.465					
			.0001445	5061.7	1.1709@-06	.0895	7.224@-06
28.594	75.000	6.898					
			.0001377	5297.3	1.0819@-06	.0842	6.260@-06
28.454	86.000	7.329					
			.0001316	5527.6	8.6993@-07	.0789	4.702@-06
28.315	100.00	7.759					
			.0001261	5753.0	7.3846@-07	.0736	3.710@-06
28.175	117.00	8.188					
			.0001211	5973.9	3.9511@-07	.0683	1.836@-06
28.035	150.00	8.615					
			.0001164	6190.4	3.0342@-07	.0629	1.294@-06
27.895	195.00	9.041					
			.0001122	6402.7	4.8098@-07	.0576	1.870@-06
27.756	225.00	9.465					
			.0001082	6610.9	2.8014@-07	.0523	9.835@-07
27.615	280.00	9.888					

EQUILIBRIUM CONCENTRATION = 26.310 INITIAL SEED AREA = 2030

TABLE: 52 RUN NO.: R.C.17

TEMP: 40.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH C PREPARED SIEVE FRACTION: 44-64 μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
15.389	.00000	2.000					
			.0004193	2245.5	8.5628@-08	.3179	7.487@-07
15.190	280.00	2.555					
			.0011035	3150.8	9.8757@-08	.2775	7.456@-07
14.452	1020.0	4.591					
			.0000673	3888.9	8.6303@-08	.2433	5.609@-07
14.395	1080.0	4.745					
			.0001296	4031.0	1.2882@-07	.2359	8.102@-07
14.282	1160.0	5.054					
			.0000931	4194.4	6.7041@-08	.2273	4.049@-07
14.196	1275.0	5.285					
			.0000752	4320.3	1.4287@-07	.2206	8.355@-07
14.125	1320.0	5.477					
			.0001019	4455.4	1.2041@-07	.2132	6.791@-07
14.026	1395.0	5.745					
			.0000844	4599.7	8.6563@-08	.2053	4.688@-07
13.940	1485.0	5.975					
			.0000548	4709.1	1.1622@-07	.1991	6.090@-07
13.884	1530.0	6.127					
			.0006270	5280.9	7.8589@-08	.1648	3.318@-07
13.143	2475.0	8.081					
			.0000536	5857.2	7.6944@-08	.1298	2.551@-07
13.072	2580.0	8.266					
			.0000421	5942.9	7.3933@-08	.1243	2.341@-07
13.015	2670.0	8.414					
			.0000518	6027.9	9.5379@-08	.1188	2.879@-07
12.944	2760.0	8.599					
			.0000508	6121.4	5.9439@-08	.1126	1.695@-07
12.872	2910.0	8.783					
			.0002395	6392.1	5.3671@-08	.0942	1.248@-07
12.516	3870.0	9.687					
			.0000366	6651.2	8.0779@-08	.0763	1.523@-07
12.459	3990.0	9.831					
			.0000449	6729.4	4.7844@-08	.0708	8.320@-08
12.387	4260.0	10.01					

EQUILIBRIUM CONCENTRATION = 11.602 INITIAL SEED AREA = 2030

TABLE: 53 RUN NO.: R.D.1

TEMP: 70.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH D PREPARED SIEVE FRACTION: 44-64μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
29.986	.00000	1.968					
			.0002333	3209.1	3.9981@-06	.1371	3.888@-05
29.847	3.0000	2.410					
			.0002014	3707.2	1.5435@-06	.1318	1.439@-05
29.708	10.000	2.851					
			.0001783	4177.4	1.1105@-06	.1265	9.903@-06
29.569	19.000	3.291					
			.0001606	4624.7	1.3466@-06	.1212	1.147@-05
29.430	26.000	3.729					
			.0001467	5052.8	9.0259@-07	.1160	7.333@-06
29.291	36.000	4.167					
			.0001353	5464.4	8.7499@-07	.1107	6.764@-06
29.152	46.000	4.604					
			.0002438	6051.1	6.5579@-07	.1027	4.688@-06
28.873	72.000	5.475					
			.0001108	6619.6	5.6320@-07	.0948	3.695@-06
28.734	87.000	5.909					
			.0002045	7157.0	8.5447@-07	.0868	5.113@-06
28.454	107.00	6.774					
			.0000948	7681.0	6.7420@-07	.0789	3.647@-06
28.315	120.00	7.204					
			.0000906	8017.8	6.0052@-07	.0736	3.020@-06
28.175	135.00	7.633					
			.0000868	8347.4	6.2209@-07	.0683	2.894@-06
28.035	150.00	8.061					
			.0001636	8826.9	4.4538@-07	.0603	1.818@-06
27.756	195.00	8.914					
			.0000772	9297.0	3.9128@-07	.0523	1.379@-06
27.615	223.00	9.339					
			.0000745	9601.3	3.1946@-07	.0470	1.007@-06
27.475	260.00	9.762					
			.0000720	9900.0	3.2351@-07	.0416	9.000@-07
27.335	300.00	10.18					
			.0000697	10193	3.2058@-07	.0363	7.740@-07
27.195	345.00	10.60					
			.0000407	10425	2.1299@-07	.0321	4.523@-07
27.111	390.00	10.85					

EQUILIBRIUM CONCENTRATION = 26.310 INITIAL SEED AREA = 2952

TABLE: 54 RUN NO.: R.D.2

TEMP: 70.0°C CELL: A STIRRER SPEED: 2000 R.P.M.

SEED: BATCH D PREPARED SIEVE FRACTION: 44-64 μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
28.748	.00000	2.000					
			.0001413	3155.5	2.2138@-06	.0911	1.413@-05
28.664	5.0000	2.263					
			.0002089	3556.5	6.8715@-07	.0868	4.177@-06
28.524	30.000	2.702					
			.0001837	4035.2	5.3785@-07	.0815	3.061@-06
28.385	60.000	3.140					
			.0001647	4489.9	3.1041@-07	.0762	1.647@-06
28.245	110.00	3.577					
			.0001499	4924.4	2.1743@-07	.0709	1.071@-06
28.105	180.00	4.013					
			.0001379	5341.8	2.0236@-07	.0656	9.192@-07
27.965	255.00	4.448					
			.0001280	5744.3	2.7942@-07	.0603	1.163@-06
27.825	310.00	4.882					

EQUILIBRIUM CONCENTRATION = 26.310 INITIAL SEED AREA = 3000

TABLE: 55 RUN NO.: R.D.3

TEMP: 70.0°C CELL: A STIRRER SPEED: 2000 R.P.M.

SEED: BATCH D PREPARED SIEVE FRACTION: 44-64 μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
29.930	.00000	1.967					
			.0002760	3257.0	2.4108@-06	.1344	2.300@-05
29.763	6.0000	2.497					
			.0001963	3801.9	1.7997@-06	.1286	1.636@-05
29.624	12.000	2.938					
			.0001745	4267.3	1.0038@-06	.1234	8.725@-06
29.485	22.000	3.378					
			.0003025	4919.6	7.4547@-07	.1154	6.051@-06
29.207	47.000	4.256					
			.0001334	5545.9	5.9193@-07	.1075	4.448@-06
29.068	62.000	4.693					
			.0001123	5921.9	8.7277@-07	.1025	6.238@-06
28.943	71.000	5.086					
			.0001174	6286.2	7.8651@-07	.0974	5.334@-06
28.803	82.000	5.522					
			.0001105	6659.1	7.2015@-07	.0921	4.605@-06
28.664	94.000	5.957					
			.0001046	7021.9	7.9099@-07	.0868	4.753@-06
28.524	105.00	6.391					
			.0000994	7375.4	5.8853@-07	.0815	3.312@-06
28.385	120.00	6.824					
			.0001855	7888.0	1.2224@-06	.0736	6.185@-06
28.105	135.00	7.688					
			.0000869	8389.5	9.6637@-07	.0656	4.344@-06
27.965	145.00	8.119					
			.0000835	8713.4	4.6053@-07	.0603	1.898@-06
27.825	167.00	8.549					
			.0001581	9186.0	3.5293@-07	.0523	1.254@-06
27.545	230.00	9.406					
			.0001476	9799.9	6.5635@-07	.0416	1.845@-06
27.265	270.00	10.26					

EQUILIBRIUM CONCENTRATION = 26.310 INITIAL SEED AREA = 2951

TABLE: 56 RUN NO.: R.D.4

TEMP: 70.0°C CELL: S STIRRER SPEED: 2000 R.P.M.

SEED: BATCH C PREPARED SIEVE FRACTION: 75-89μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
30.235	.00000	1.955					
			.0004266	2472.4	3.4192@-06	.1455	3.555@-05
30.041	6.0000	2.575					
			.0002412	3100.9	2.0370@-06	.1392	2.010@-05
29.902	12.000	3.016					
			.0002076	3593.5	1.3710@-06	.1339	1.297@-05
29.763	20.000	3.456					
			.0001831	4063.3	1.4434@-06	.1286	1.308@-05
29.624	27.000	3.896					
			.0001645	4513.3	1.0546@-06	.1234	9.140@-06
29.485	36.000	4.334					
			.0001498	4946.0	1.0060@-06	.1181	8.323@-06
29.346	45.000	4.772					
			.0001379	5363.5	8.7454@-07	.1128	6.893@-06
29.207	55.000	5.209					
			.0003608	6149.9	9.3798@-07	.1022	6.681@-06
28.789	82.000	6.518					
			.0001061	6914.4	6.4395@-07	.0916	4.080@-06
28.650	95.000	6.951					
			.0003766	7792.6	6.3630@-07	.0783	3.424@-06
28.091	150.00	8.683					
			.0000840	8652.2	5.5573@-07	.0651	2.470@-06
27.951	167.00	9.112					
			.0000808	8978.4	4.3134@-07	.0597	1.756@-06
27.811	190.00	9.540					
			.0001530	9454.9	8.7310@-07	.0518	3.061@-06
27.531	215.00	10.39					
			.0000726	9923.2	4.5472@-07	.0438	1.344@-06
27.391	242.00	10.82					
			.0000703	10228	4.1137@-07	.0384	1.065@-06
27.251	275.00	11.25					
			.0001344	10674	3.9016@-07	.0305	7.909@-07
26.970	360.00	12.09					

EQUILIBRIUM CONCENTRATION = 26.310 INITIAL SEED AREA = 2096

TABLE: 57 RUN NO.: R.D.5

TEMP: 70.0°C CELL: S STIRRER SPEED: 2000 R.P.M.

SEED: BATCH D PREPARED SIEVE FRACTION: 75-89μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
30.069	.00000	1.945					
			.0006938	1295.7	6.9933@-06	.1397	6.938@-05
29.902	5.0000	2.476					
			.0004994	1495.4	3.7653@-06	.1339	3.567@-05
29.763	12.000	2.917					
			.0004472	1666.3	4.1063@-06	.1286	3.727@-05
29.624	18.000	3.357					
			.0004065	1829.4	3.3450@-06	.1234	2.903@-05
29.485	25.000	3.796					
			.0007210	2060.2	2.6178@-06	.1154	2.121@-05
29.207	42.000	4.673					
			.0003237	2282.9	2.6963@-06	.1075	2.023@-05
29.068	50.000	5.110					
			.0003042	2424.2	1.7814@-06	.1022	1.268@-05
28.929	62.000	5.546					
			.0002873	2561.5	1.9408@-06	.0969	1.306@-05
28.789	73.000	5.981					
			.0002725	2695.2	1.9524@-06	.0916	1.239@-05
28.650	84.000	6.415					
			.0002594	2825.7	1.9778@-06	.0863	1.179@-05
28.510	95.000	6.848					
			.0002477	2953.1	1.5853@-06	.0810	8.848@-06
28.371	109.00	7.280					
			.0002372	3077.7	1.9002@-06	.0757	9.885@-06
28.231	121.00	7.712					
			.0002277	3199.8	1.8155@-06	.0704	8.759@-06
28.091	134.00	8.143					
			.0002191	3319.5	1.2960@-06	.0651	5.765@-06
27.951	153.00	8.572					
			.0002112	3436.9	1.2341@-06	.0597	5.028@-06
27.811	174.00	9.001					
			.0002039	3552.2	1.3117@-06	.0544	4.855@-06
27.672	195.00	9.429					
			.0001972	3665.5	1.1843@-06	.0491	3.944@-06
27.531	220.00	9.856					
			.0001910	3777.0	5.8647@-07	.0438	1.736@-06
27.391	275.00	10.28					
			.0001852	3886.7	7.1444@-07	.0384	1.852@-06
27.251	325.00	10.71					
			.0001798	3994.7	8.0791@-07	.0331	1.798@-06
27.111	375.00	11.13					

EQUILIBRIUM CONCENTRATION = 26.310 INITIAL SEED AREA = 1184

TABLE: 58 RUN NO.: R.D.6

TEMP: 70.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH C PREPARED SIEVE FRACTION: 75-89μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
30.263	.00000	1.970					
			.0003152	2383.7	4.9924@-06	.1476	5.253@-05
30.124	3.0000	2.413					
			.0002570	2912.8	1.8168@-06	.1424	1.836@-05
29.986	10.000	2.854					
			.0002185	3415.5	1.6099@-06	.1371	1.561@-05
29.847	17.000	3.295					
			.0001911	3894.3	1.4693@-06	.1318	1.365@-05
29.708	24.000	3.734					
			.0001706	4352.1	1.0659@-06	.1265	9.476@-06
29.569	33.000	4.173					
			.0001546	4791.5	1.1373@-06	.1212	9.659@-06
29.430	41.000	4.610					
			.0002730	5416.6	9.0774@-07	.1133	7.184@-06
29.152	60.000	5.483					
			.0001221	6021.3	5.9601@-07	.1054	4.360@-06
29.012	74.000	5.917					
			.0001145	6406.3	8.2624@-07	.1001	5.726@-06
28.873	84.000	6.351					
			.0002102	6961.5	7.8794@-07	.0921	5.005@-06
28.594	105.00	7.215					
			.0001897	7675.5	7.3844@-07	.0815	4.124@-06
28.315	128.00	8.075					
			.0001733	8358.1	5.6126@-07	.0709	2.707@-06
28.035	160.00	8.931					
			.0000814	8854.3	4.7730@-07	.0629	2.036@-06
27.895	180.00	9.356					
			.0000783	9175.5	5.0344@-07	.0576	1.957@-06
27.756	200.00	9.781					
			.0000754	9490.6	3.0664@-07	.0523	1.077@-06
27.615	235.00	10.20					
			.0001432	9950.8	3.7352@-07	.0443	1.102@-06
27.335	300.00	11.05					
			.0000681	10403	3.5338@-07	.0363	8.512@-07
27.195	340.00	11.46					
			.0000658	10697	1.6982@-07	.0310	3.465@-07
27.055	435.00	11.88					

EQUILIBRIUM CONCENTRATION = 26.310 INITIAL SEED AREA = 2112

TABLE: 59 RUN NO.: R.D.7

TEMP: 70.0°C CELL: S STIRRER SPEED: 2000 R.P.M.

SEED: BATCH P.G. PREPARED SIEVE FRACTION: 64-75μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
29.902	.00000	1.955					
			.0005745	1302.4	1.5131@-05	.1339	1.436@-04
29.763	2.0000	2.397					
			.0005044	1480.0	9.2466@-06	.1286	8.406@-05
29.624	5.0000	2.838					
			.0004520	1647.7	6.4996@-06	.1234	5.651@-05
29.485	9.0000	3.278					
			.0007905	1882.4	2.5635@-06	.1154	2.080@-05
29.207	28.000	4.156					
			.0003512	2107.7	2.3363@-06	.1075	1.756@-05
29.068	38.000	4.593					
			.0003283	2249.7	1.2797@-06	.1022	9.120@-06
28.929	56.000	5.030					
			.0006014	2453.3	3.5296@-06	.0943	2.313@-05
28.650	69.000	5.901					
			.0002769	2651.2	2.3187@-06	.0863	1.385@-05
28.510	79.000	6.335					
			.0002638	2778.0	2.1448@-06	.0810	1.199@-05
28.371	90.000	6.768					
			.0002520	2901.7	2.0154@-06	.0757	1.050@-05
28.231	102.00	7.200					
			.0002414	3022.8	1.2492@-06	.0704	6.036@-06
28.091	122.00	7.632					
			.0002319	3141.2	1.8587@-06	.0651	8.281@-06
27.951	136.00	8.062					
			.0002232	3257.3	2.2788@-06	.0597	9.298@-06
27.811	148.00	8.492					
			.0002152	3371.3	1.3192@-06	.0544	4.891@-06
27.672	170.00	8.921					
			.0002079	3483.1	1.2464@-06	.0491	4.157@-06
27.531	195.00	9.348					
			.0003964	3646.7	8.9351@-07	.0411	2.477@-06
27.251	275.00	10.20					

EQUILIBRIUM CONCENTRATION = 26.310 INITIAL SEED AREA = 1211

TABLE: 60 RUN NO.: R.P.G.3

TEMP: 70.0°C CELL: S STIRRER SPEED 2000 R.P.M.

SEED: BATCH P.G.PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
31.319	.00000	2.000					
			.0008206	1399.1	9.2362@-06	.2602	1.641@-04
31.111	2.5000	2.671					
			.0004464	1813.9	8.7201@-06	.2530	1.488@-04
30.963	4.0000	3.148					
			.0003749	2151.3	5.6476@-06	.2470	9.374@-05
30.814	6.0000	3.624					
			.0003246	2476.1	6.7039@-06	.2410	1.082@-04
30.665	7.5000	4.099					
			.0002873	2788.6	4.5779@-06	.2350	7.183@-05
30.517	9.5000	4.572					
			.0002585	3089.4	4.2396@-06	.2290	6.463@-05
30.368	11.500	5.044					
			.0002356	3379.7	3.9790@-06	.2230	5.890@-05
30.220	13.500	5.514					
			.0002169	3660.6	3.0197@-06	.2170	4.338@-05
30.071	16.000	5.983					
			.0002013	3932.9	2.8900@-06	.2110	4.025@-05
29.923	18.500	6.451					
			.0001880	4197.3	2.7866@-06	.2050	3.761@-05
29.774	21.000	6.917					
			.0003439	4578.9	2.6716@-06	.1960	3.439@-05
29.477	26.000	7.847					
			.0003087	5069.6	2.5693@-06	.1840	3.087@-05
29.180	31.000	8.772					
			.0004132	5650.5	2.3529@-06	.1690	2.583@-05
28.736	39.000	10.15					
			.0001265	6101.1	1.5617@-06	.1571	1.581@-05
28.587	43.000	10.61					
			.0001218	6317.7	1.5676@-06	.1511	1.522@-05
28.439	47.000	11.06					
			.0001175	6530.5	1.2631@-06	.1451	1.175@-05
28.291	52.000	11.51					
			.0002235	6841.9	1.8363@-06	.1361	1.597@-05
27.994	59.000	12.42					
			.0001064	7148.3	1.3162@-06	.1272	1.064@-05
27.846	64.000	12.87					

TABLE : 60 (CONTD.) RUN NO. : R.P.G3 (CONTD.)

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
27.846	64.000	12.87					
			.0002036	7445.5	1.3601@-06	.1182	1.018@-05
27.550	74.000	13.76					
			.0000974	7738.4	1.1790@-06	.1092	8.118@-06
27.402	80.000	14.21					
			.0000948	7929.4	1.0432@-06	.1032	6.772@-06
27.254	87.000	14.65					
			.0000923	8117.8	1.5141@-06	.0973	9.235@-06
27.106	92.000	15.10					
			.0000900	8303.8	8.7601@-07	.0913	5.002@-06
26.958	101.00	15.54					
			.0000878	8487.4	9.1696@-07	.0853	4.880@-06
26.810	110.00	15.98					

EQUILIBRIUM CONCENTRATION = 24.771 INITIAL SEED AREA = 1156

TABLE: 61 RUN NO.: R.E.1

TEMP: 70.0° C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
29.986	.00000	1.982					
			.0004307	1739.2	1.1066@-05	.1371	1.077@-04
29.847	2.0000	2.424					
			.0003672	2034.2	2.8129@-06	.1318	2.623@-05
29.708	9.0000	2.865					
			.0003218	2314.6	2.0042@-06	.1265	1.788@-05
29.569	18.000	3.305					
			.0002877	2582.5	1.6881@-06	.1212	1.438@-05
29.430	28.000	3.744					
			.0002610	2839.4	1.7847@-06	.1160	1.450@-05
29.291	37.000	4.181					
			.0002395	3086.9	1.9361@-06	.1107	1.497@-05
29.152	45.000	4.618					
			.0002218	3326.0	1.8883@-06	.1054	1.386@-05
29.012	53.000	5.054					
			.0002068	3557.6	1.4878@-06	.1001	1.034@-05
28.873	63.000	5.489					
			.0001941	3782.5	1.6427@-06	.0948	1.078@-05
28.734	72.000	5.922					
			.0001830	4001.3	1.3466@-06	.0895	8.318@-06
28.594	83.000	6.355					
			.0001733	4214.6	1.4957@-06	.0842	8.666@-06
28.454	93.000	6.787					
			.0001647	4422.8	1.1709@-06	.0789	6.336@-06
28.315	106.00	7.217					
			.0001571	4626.2	9.7573@-07	.0736	4.909@-06
28.175	122.00	7.647					
			.0002944	4921.9	8.6839@-07	.0656	3.874@-06
27.895	160.00	8.503					
			.0002716	5304.6	8.7186@-07	.0550	3.233@-06
27.615	202.00	9.354					
			.0001283	5583.4	8.8373@-07	.0470	2.790@-06
27.475	225.00	9.778					
			.0001239	5764.1	4.4451@-07	.0416	1.239@-06
27.335	275.00	10.20					
			.0001197	5941.6	4.9499@-07	.0363	1.197@-06
27.195	325.00	10.62					
			.0002273	6200.6	3.8443@-07	.0283	7.103@-07
26.914	485.00	11.45					

EQUILIBRIUM CONCENTRATION = 26.310 INITIAL SEED AREA = 1588

TABLE: 62 RUN NO.: R.E.2

TEMP: 70.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
27.419	.00000	1.986					
			.0011184	2023.2	1.7797@-07	.0375	4.335@-07
26.970	1290.0	3.310					
			.0000557	2480.1	1.0130@-07	.0284	1.856@-07
26.942	1440.0	3.391					
			.0000540	2529.1	6.4525@-08	.0274	1.125@-07
26.914	1680.0	3.472					

EQUILIBRIUM CONCENTRATION = 26.211 INITIAL SEED AREA = 1591

TABLE: 63 RUN NO.: R.E.3

TEMP: 60.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105 μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
27.276	.00000	1.985					
			.0007818	1879.1	1.4179@-05	.3447	2.606@-04
26.998	1.5000	2.849					
			.0003163	2302.0	5.9700@-06	.3343	1.054@-04
26.859	3.0000	3.279					
			.0002833	2564.6	8.2116@-06	.3274	1.416@-04
26.719	4.0000	3.708					
			.0002574	2816.8	7.6414@-06	.3205	1.287@-04
26.580	5.0000	4.136					
			.0002364	3059.9	7.1930@-06	.3136	1.182@-04
26.441	6.0000	4.563					
			.0002191	3294.9	5.4672@-06	.3067	8.765@-05
26.302	7.2500	4.990					
			.0002045	3522.7	5.2342@-06	.2998	8.181@-05
26.162	8.5000	5.416					
			.0001920	3744.1	4.2030@-06	.2929	6.402@-05
26.023	10.000	5.841					
			.0001812	3959.6	4.8870@-06	.2860	7.249@-05
25.883	11.250	6.265					
			.0001718	4169.8	4.7583@-06	.2791	6.870@-05
25.743	12.500	6.688					
			.0001634	4375.0	3.8774@-06	.2721	5.446@-05
25.604	14.000	7.110					
			.0001559	4575.8	3.4187@-06	.2652	4.668@-05
25.464	15.670	7.532					
			.0001492	4772.3	3.3880@-06	.2583	4.493@-05
25.324	17.330	7.953					
			.0001431	4965.0	2.7789@-06	.2514	3.578@-05
25.184	19.330	8.373					
			.0001376	5154.0	3.2988@-06	.2444	4.119@-05
25.044	21.000	8.792					
			.0001325	5339.5	3.1291@-06	.2375	3.787@-05
24.904	22.750	9.210					
			.0001280	5522.0	2.8424@-06	.2305	3.334@-05
24.764	24.670	9.627					
			.0002436	5789.1	2.2600@-06	.2201	2.522@-05
24.483	29.500	10.46					
			.0001160	6052.0	2.7418@-06	.2097	2.901@-05
24.343	31.500	10.88					
			.0001126	6223.2	2.7594@-06	.2027	2.815@-05
24.202	33.500	11.29					
			.0001094	6391.9	2.4743@-06	.1958	2.432@-05
24.061	35.750	11.70					

TABLE :63 (CONTD.) RUN NO. :R.E.3 (CONTD.)

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT K($MIN^{-1}CM^{-2}$)	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
24.061	35.750	11.70					
			.0001064	6558.2	2.5019@-06	.1888	2.365@-05
23.921	38.000	12.12					
			.0001036	6722.3	2.0747@-06	.1818	1.884@-05
23.780	40.750	12.53					
			.0001010	6884.2	1.9322@-06	.1748	1.683@-05
23.639	43.750	12.94					
			.0000985	7044.1	1.8165@-06	.1679	1.515@-05
23.498	47.000	13.35					
			.0001903	7279.7	1.7430@-06	.1574	1.359@-05
23.217	54.000	14.17					
			.0000918	7512.6	2.1118@-06	.1469	1.530@-05
23.075	57.000	14.57					
			.0000898	7665.0	1.7395@-06	.1399	1.197@-05
22.934	60.750	14.98					
			.0000879	7815.7	1.5854@-06	.1329	1.034@-05
22.793	65.000	15.39					
			.0000861	7964.8	1.0743@-06	.1259	6.621@-06
22.652	71.500	15.79					
			.0000843	8112.3	9.6854@-07	.1189	5.622@-06
22.510	79.000	16.20					
			.0000827	8258.2	1.0116@-06	.1119	5.511@-06
22.369	86.500	16.60					
			.0000811	8402.7	1.2245@-06	.1049	6.237@-06
22.227	93.000	17.00					
			.0000796	8545.7	1.2910@-06	.0979	6.120@-06
22.086	99.500	17.40					
			.0000781	8687.2	8.0889@-07	.0909	3.550@-06
21.944	110.50	17.80					
			.0000767	8827.5	1.1171@-06	.0838	4.512@-06
21.802	119.00	18.20					
			.0000753	8966.3	7.2929@-07	.0768	2.691@-06
21.661	133.00	18.60					
			.0000740	9103.9	7.3841@-07	.0698	2.468@-06
21.519	148.00	19.00					
			.0001443	9307.3	5.5755@-07	.0592	1.568@-06
21.235	194.00	19.79					
			.0000703	9508.8	5.8618@-07	.0487	1.353@-06
21.093	220.00	20.19					
			.0000692	9641.3	4.3988@-07	.0416	8.645@-07
20.950	260.00	20.58					
			.0000948	9798.3	4.3702@-07	.0332	6.768@-07
20.751	330.00	21.13					
			.0000399	9927.9	2.7257@-07	.0261	3.324@-07
20.666	390.00	21.37					

EQUILIBRIUM CONCENTRATION = 20.181 INITIAL SEED AREA = 1590

TABLE: 64 RUN NO.: R.E.4

TEMP: 60.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105 μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
24.371	.00000	1.985					
			.0004906	1764.4	2.9794@-06	.2137	3.271@-05
24.202	7.5000	2.495					
			.0003460	2075.8	7.6419@-07	.2060	8.048@-06
24.061	29.000	2.920					
			.0003056	2344.1	7.9323@-07	.1990	8.042@-06
23.921	48.000	3.343					
			.0002747	2600.9	7.0443@-07	.1919	6.867@-06
23.780	68.000	3.765					
			.0002502	2847.6	7.0340@-07	.1849	6.585@-06
23.639	87.000	4.186					
			.0002303	3085.6	7.1271@-07	.1779	6.398@-06
23.498	105.00	4.606					
			.0002138	3315.7	6.5459@-07	.1708	5.626@-06
23.358	124.00	5.024					
			.0001998	3538.9	7.1534@-07	.1638	5.877@-06
23.217	141.00	5.442					
			.0001878	3755.8	5.7050@-07	.1567	4.472@-06
23.075	162.00	5.859					
			.0001774	3966.9	6.2546@-07	.1497	4.667@-06
22.934	181.00	6.275					
			.0003285	4272.3	6.2586@-07	.1391	4.323@-06
22.652	219.00	7.104					
			.0002993	4666.2	6.5587@-07	.1250	4.044@-06
22.369	256.00	7.929					
			.0002754	5044.0	5.1732@-07	.1108	2.810@-06
22.086	305.00	8.750					
			.0001299	5319.1	4.4300@-07	.1002	2.165@-06
21.944	335.00	9.158					
			.0001254	5497.4	4.3266@-07	.0931	1.959@-06
21.802	367.00	9.565					
			.0001211	5672.7	4.4034@-07	.0860	1.835@-06
21.661	400.00	9.971					
			.0001172	5845.0	4.3940@-07	.0789	1.674@-06
21.519	435.00	10.38					
			.0001135	6014.5	3.2864@-07	.0718	1.135@-06
21.377	485.00	10.78					
			.0001100	6181.2	3.2280@-07	.0648	1.000@-06
21.235	540.00	11.18					
			.0000751	6321.1	3.3506@-07	.0587	9.391@-07
21.135	580.00	11.46					
			.0000736	6434.8	3.5976@-07	.0537	9.202@-07
21.036	620.00	11.74					

EQUILIBRIUM CONCENTRATION = 20.010 INITIAL SEED AREA = 1590

TABLE: 65 RUN NO.: R.E.5

TEMP: 60.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105 μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
25.827	.00000	1.986					
			.0002591	1680.1	5.8162@-06	.2850	8.636@-05
25.743	1.5000	2.243					
			.0003793	1911.6	2.6081@-06	.2794	3.793@-05
25.604	6.5000	2.671					
			.0003303	2189.6	2.1240@-06	.2724	3.003@-05
25.464	12.000	3.098					
			.0002939	2455.1	3.0566@-06	.2654	4.199@-05
25.324	15.500	3.524					
			.0002657	2709.6	2.2133@-06	.2585	2.952@-05
25.184	20.000	3.950					
			.0002432	2954.6	2.0872@-06	.2515	2.702@-05
25.044	24.500	4.374					
			.0002246	3191.2	2.2373@-06	.2445	2.808@-05
24.904	28.500	4.798					
			.0002092	3420.4	1.9110@-06	.2375	2.324@-05
24.764	33.000	5.220					
			.0001960	3642.9	1.8497@-06	.2305	2.177@-05
24.623	37.500	5.642					
			.0001846	3859.3	1.8015@-06	.2236	2.051@-05
24.483	42.000	6.063					
			.0001747	4070.2	1.5878@-06	.2166	1.747@-05
24.343	47.000	6.483					
			.0001659	4276.1	1.9534@-06	.2096	2.074@-05
24.202	51.000	6.903					
			.0001581	4477.4	1.9311@-06	.2026	1.977@-05
24.061	55.000	7.321					
			.0001512	4674.3	1.7040@-06	.1955	1.680@-05
23.921	59.500	7.739					
			.0001449	4867.3	1.6982@-06	.1885	1.610@-05
23.780	64.000	8.155					
			.0001392	5056.5	1.3899@-06	.1815	1.265@-05
23.639	69.500	8.571					
			.0001339	5242.2	1.2790@-06	.1745	1.116@-05
23.498	75.500	8.986					
			.0001292	5424.7	1.1894@-06	.1675	9.936@-06
23.358	82.000	9.400					
			.0001248	5604.1	1.2024@-06	.1605	9.604@-06
23.217	88.500	9.814					
			.0001208	5780.5	9.3274@-07	.1534	7.103@-06
23.075	97.000	10.23					
			.0001169	5954.2	7.3374@-07	.1464	5.316@-06
22.934	108.000	10.64					

TABLE:65 (CONTD.) RUN NO.: R.E.5 (CONTD.)

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
22.934	108.00	10.64					
			.0001134	6125.2	1.0308@-06	.1394	7.089@-06
22.793	116.00	11.05					
			.0001101	6293.7	7.6885@-07	.1323	5.006@-06
22.652	127.00	11.46					
			.0001071	6459.7	7.9164@-07	.1253	4.866@-06
22.510	138.00	11.87					
			.0001042	6623.5	9.0040@-07	.1182	5.209@-06
22.369	148.00	12.27					
			.0002005	6864.3	5.9751@-07	.1077	3.133@-06
22.086	180.00	13.09					
			.0001908	7179.3	6.0207@-07	.0935	2.725@-06
21.802	215.00	13.90					
			.0000920	7411.0	5.7524@-07	.0829	2.299@-06
21.661	235.00	14.30					
			.0000899	7562.6	6.1662@-07	.0759	2.248@-06
21.519	255.00	14.70					
			.0001740	7786.0	3.9949@-07	.0653	1.243@-06
21.235	325.00	15.50					
			.0000842	8006.8	5.4034@-07	.0546	1.403@-06
21.093	355.00	15.90					
			.0000824	8151.4	3.3295@-07	.0476	7.490@-07
20.950	410.00	16.30					
			.0000485	8266.0	1.8915@-07	.0419	3.733@-07
20.865	475.00	16.53					

EQUILIBRIUM CONCENTRATION = 20.067 INITIAL SEED AREA = 1591

TABLE: 66 RUN NO.: R.E.6

TEMP: 50.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
23.855	.00000	1.990					
			.0010748	2008.0	1.9950@-05	.5478	4.299@-04
23.436	1.2500	3.253					
			.0002780	2547.5	5.2169@-06	.5295	1.069@-04
23.296	2.5500	3.672					
			.0002529	2794.3	1.0151@-05	.5204	2.039@-04
23.157	3.1700	4.089					
			.0002325	3032.3	7.1155@-06	.5112	1.401@-04
23.017	4.0000	4.506					
			.0002157	3262.5	1.1183@-05	.5021	2.157@-04
22.877	4.5000	4.922					
			.0002014	3485.7	1.0665@-05	.4929	2.014@-04
22.737	5.0000	5.336					
			.0001892	3702.7	7.6371@-06	.4838	1.412@-04
22.597	5.6700	5.751					
			.0001787	3913.9	7.4787@-06	.4746	1.353@-04
22.457	6.3300	6.164					
			.0001694	4119.9	7.1390@-06	.4654	1.264@-04
22.317	7.0000	6.576					
			.0001611	4321.2	6.2050@-06	.4562	1.074@-04
22.177	7.7500	6.988					
			.0001538	4518.0	6.7820@-06	.4471	1.148@-04
22.036	8.4200	7.398					
			.0001472	4710.8	5.9346@-06	.4379	9.814@-05
21.896	9.1700	7.808					
			.0002774	4991.8	5.4932@-06	.4241	8.778@-05
21.615	10.750	8.626					
			.0002574	5356.5	5.3550@-06	.4057	8.146@-05
21.335	12.330	9.441					
			.0002405	5708.8	4.3341@-06	.3873	6.264@-05
21.053	14.250	10.25					
			.0002260	6050.2	4.1248@-06	.3689	5.651@-05
20.772	16.250	11.06					
			.0003162	6462.0	3.4559@-06	.3459	4.416@-05
20.350	19.830	12.27					
			.0000997	6784.6	2.2554@-06	.3275	2.710@-05
20.209	21.670	12.67					

TABLE : 66 (CONTD.) RUN NO. : R.E.6 (CONTD.)

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
20.209	21.670	12.67					
20.040	23.330	13.15	.0001165	6957.0	3.0204@-06	.3173	3.509@-05
19.927	25.170	13.46	.0000758	7112.0	1.8308@-06	.3081	2.061@-05
19.786	27.170	13.86	.0000928	7249.6	2.1234@-06	.2998	2.319@-05
19.645	29.170	14.26	.0000907	7400.8	2.1469@-06	.2906	2.267@-05
19.504	31.330	14.65	.0000887	7550.4	2.0131@-06	.2813	2.053@-05
19.363	33.830	15.05	.0000868	7698.3	1.7645@-06	.2721	1.737@-05
19.222	37.000	15.44	.0000850	7844.5	1.4141@-06	.2629	1.341@-05
19.081	40.830	15.84	.0000833	7989.2	1.1915@-06	.2536	1.088@-05
18.939	44.670	16.23	.0000817	8132.5	1.2121@-06	.2444	1.064@-05
18.798	48.500	16.62	.0000801	8274.2	1.2419@-06	.2351	1.046@-05
18.657	53.330	17.01	.0000786	8414.6	1.0084@-06	.2259	8.141@-06
18.515	56.670	17.40	.0000772	8553.6	1.4964@-06	.2166	1.156@-05
18.374	61.000	17.79	.0000758	8691.2	1.1872@-06	.2073	8.757@-06
18.232	66.170	18.18	.0000745	8827.6	1.0251@-06	.1981	7.207@-06
18.090	71.170	18.57	.0000732	8962.7	1.0956@-06	.1888	7.325@-06
17.807	86.000	19.34	.0001429	9162.7	7.8108@-07	.1749	4.816@-06
17.524	100.00	20.11	.0001383	9425.7	9.0058@-07	.1564	4.938@-06
17.382	110.00	20.49	.0000674	9620.4	6.7782@-07	.1424	3.372@-06
17.240	122.00	20.88	.0000664	9748.4	5.9655@-07	.1331	2.767@-06
17.098	136.00	21.26	.0000654	9875.3	5.4284@-07	.1239	2.336@-06
16.956	139.00	21.64	.0000644	10001	2.7054@-06	.1146	1.074@-05

TABLE : 66 (CONTD.) RUN NO. : R.E.6 (CONTD.)

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
16.956	139.00	21.64					
			.0000635	10126	3.7948@-07	.1053	1.380@-06
16.814	162.00	22.02					
			.0000626	10250	4.5060@-07	.0960	1.490@-06
16.672	183.00	22.40					
			.0000617	10373	4.5042@-07	.0867	1.341@-06
16.530	206.00	22.78					
			.0000608	10494	4.7825@-07	.0774	1.267@-06
16.387	230.00	23.15					
			.0000599	10615	3.2271@-07	.0681	7.492@-07
16.245	270.00	23.53					
			.0000591	10735	4.2289@-07	.0587	8.443@-07
16.103	305.00	23.91					
			.0000583	10854	2.9032@-07	.0494	4.855@-07
15.961	365.00	24.28					

EQUILIBRIUM CONCENTRATION = 15.277 INITIAL SEED AREA = 1594

TABLE: 67 RUN NO.: R.E.7

TEMP: 70.0°C CELL: S STIRRER SPEED: 2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
30.041	.00000	1.937					
			.0004400	1703.1	4.4505@-06	.1392	4.400@-05
29.902	5.0000	2.379					
			.0003741	1997.9	3.5869@-06	.1339	3.401@-05
29.763	10.500	2.820					
			.0003273	2278.0	2.4030@-06	.1286	2.182@-05
29.624	18.000	3.261					
			.0002923	2545.3	1.5300@-06	.1234	1.328@-05
29.485	29.000	3.700					
			.0002649	2801.8	1.4530@-06	.1181	1.204@-05
29.346	40.000	4.139					
			.0002430	3048.7	1.5385@-06	.1128	1.215@-05
29.207	50.000	4.576					
			.0004351	3401.3	1.5639@-06	.1048	1.145@-05
28.929	69.000	5.450					
			.0001967	3743.3	1.3281@-06	.0969	8.940@-06
28.789	80.000	5.885					
			.0003616	4067.0	1.5446@-06	.0890	9.515@-06
28.510	99.000	6.754					
			.0001670	4382.8	1.3595@-06	.0810	7.590@-06
28.371	110.00	7.186					
			.0001593	4585.9	1.3912@-06	.0757	7.239@-06
28.231	121.00	7.618					
			.0001523	4784.8	9.8648@-07	.0704	4.760@-06
28.091	137.00	8.048					
			.0001461	4979.6	1.1725@-06	.0651	5.217@-06
27.951	151.00	8.478					
			.0001404	5170.7	1.0766@-06	.0597	4.387@-06
27.811	167.00	8.907					
			.0001352	5358.4	9.1302@-07	.0544	3.380@-06
27.672	187.00	9.336					
			.0001305	5542.7	1.0878@-06	.0491	3.624@-06
27.531	205.00	9.763					
			.0001262	5724.0	9.2540@-07	.0438	2.742@-06
27.391	228.00	10.19					
			.0001221	5902.4	9.4091@-07	.0384	2.441@-06
27.251	253.00	10.61					

TABLE:67 (CONTD.) RUN NO.: R.E.7 (CONTD.)

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
27.251	253.00	10.61					
			.0000477	6025.8	5.9986@-07	.0347	1.403@-06
27.195	270.00	10.78					
			.0000354	6086.9	5.3348@-07	.0329	1.180@-06
27.153	285.00	10.91					
			.0001499	6224.8	7.8346@-07	.0286	1.499@-06
26.970	335.00	11.46					
			.0000449	6370.6	3.4863@-07	.0240	5.617@-07
26.914	375.00	11.63					
			.0000665	6455.1	6.6446@-07	.0214	9.495@-07
26.830	410.00	11.89					
			.0000654	6555.8	3.3704@-07	.0182	4.085@-07
26.746	490.00	12.14					
			.0000430	6639.1	3.4653@-07	.0155	3.581@-07
26.689	550.00	12.31					

EQUILIBRIUM CONCENTRATION = 26.310 INITIAL SEED AREA = 1552

TABLE: 68 RUN NO.: R.P.G.4

TEMP: 70.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH P.G. PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
30.101	.00000	2.000					
			.0003831	1257.7	6.7017@-06	.2134	9.578@-05
30.012	2.0000	2.284					
			.0005238	1532.0	3.1267@-06	.2086	4.365@-05
29.863	8.0000	2.756					
			.0004260	1872.7	5.6422@-07	.2026	7.607@-06
29.715	36.000	3.227					
			.0003606	2203.3	1.1529@-06	.1966	1.502@-05
29.566	48.000	3.696					
			.0005930	2671.9	6.6418@-07	.1876	8.237@-06
29.270	84.000	4.629					
			.0006940	3398.7	6.0088@-07	.1726	6.804@-06
28.825	135.00	6.019					

EQUILIBRIUM CONCENTRATION = 24.771 INITIAL SEED AREA = 1156

TABLE:69 RUN NO.: R.P.G.5

TEMP:70.0°C CELL: C STIRRER SPEED:2000 R.P.M.

SEED: BATCH P.G. PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
29.953	.00000	2.000					
			.0003825	1257.6	2.7581@-06	.2074	3.825@-05
29.863	5.0000	2.283					
			.0009492	1699.7	8.8353@-07	.1996	1.187@-05
29.567	45.000	3.225					
			.0003600	2201.9	5.7105@-07	.1906	7.199@-06
29.418	70.000	3.693					
			.0036669	5041.8	3.8874@-07	.1200	2.821@-06
26.071	720.00	13.94					

EQUILIBRIUM CONCENTRATION = 24.771 INITIAL SEED AREA = 1156

TABLE: 70 RUN NO.: R.P.G.6

TEMP:70.0°C CELL: S STIRRER SPEED:2000 R.P.M.

SEED: BATCH P.G.PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
30.338	.00000	2.000					
			.0003840	1258.0	6.4137@-07	.2230	9.600@-06
30.249	20.000	2.285					
			.0031495	2972.9	1.8189@-07	.1888	2.461@-06
28.647	660.00	7.390					

EQUILIBRIUM CONCENTRATION = 24.771 INITIAL SEED AREA = 1156

TABLE: 71 RUN NO.: R.P.D.1

TEMP: 70.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH P.G. PREPARED SIEVE FRACTION: 89-105 μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
30.041	.00000	2.000					
			.0021878	2012.6	7.8677@-05	.1972	1.094@-03
29.270	1.0000	4.459					
			.0005000	3158.7	1.4399@-05	.1756	1.667@-04
28.973	2.5000	5.390					
			.0011205	4223.1	6.6257@-06	.1517	6.591@-05
28.083	11.000	8.168					
			.0005652	5455.4	5.1600@-06	.1218	4.037@-05
27.491	18.000	9.987					
			.0002484	6127.8	2.3508@-06	.1038	1.553@-05
27.195	26.000	10.89					
			.0002310	6550.5	2.2089@-06	.0919	1.283@-05
26.899	35.000	11.78					
			.0003196	7057.8	1.9492@-06	.0770	9.399@-06
26.455	52.000	13.11					
			.0003848	7737.0	1.2999@-06	.0561	4.474@-06
25.864	95.000	14.87					
			.0003116	8439.2	4.7885@-07	.0334	9.444@-07
25.332	260.00	16.43					

EQUILIBRIUM CONCENTRATION = 24.771 INITIAL SEED AREA = 1156

TABLE: 72 RUN NO.: R.E.8

TEMP: 70.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
29.819	.00000	2.000					
			.0003471	1723.9	5.3322@-06	.1313	4.958@-05
29.708	3.5000	2.353					
			.0003751	1990.6	3.2267@-06	.1265	2.885@-05
29.569	10.000	2.794					
			.0003274	2273.7	1.9173@-06	.1212	1.637@-05
29.430	20.000	3.234					
			.0002919	2544.0	1.9919@-06	.1160	1.622@-05
29.291	29.000	3.672					

EQUILIBRIUM CONCENTRATION = 26.310 INITIAL SEED AREA = 1602

TABLE: 73 RUN NO.: R.F.3

TEMP: 70.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH F PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
30.235	.00000	2.000					
			.0020588	6990.3	2.0761@-07	.1026	1.525@-06
27.784	675.00	9.618					
			.0000707	11871	7.5524@-08	.0528	2.618@-07
27.615	810.00	10.11					

EQUILIBRIUM CONCENTRATION = 26.310 INITIAL SEED AREA = 2347.2

TABLE: 74 RUN NO.: R.F.4

TEMP: 70.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH F PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
29.902	.00000	1.985					
			.0001205	2479.2	1.3963@-07	.1355	1.339@-06
29.847	45.000	2.161					
			.0002501	2988.4	3.8296@-07	.1318	3.573@-06
29.708	80.000	2.601					
			.0000645	3450.5	8.9412@-08	.1284	8.062@-07
29.666	120.00	2.733					
			.0001908	3883.1	1.9800@-07	.1249	1.735@-06
29.527	175.00	3.170					
			.0002656	4733.7	4.6067@-07	.1178	3.794@-06
29.291	210.00	3.911					
			.0001325	5541.5	3.0815@-07	.1107	2.366@-06
29.152	238.00	4.345					
			.0001199	6103.5	2.2248@-07	.1054	1.620@-06
29.012	275.00	4.777					
			.0001098	6643.3	1.9919@-07	.1001	1.372@-06
28.873	315.00	5.208					
			.0001014	7163.6	1.7348@-07	.0948	1.126@-06
28.734	360.00	5.637					
			.0000944	7666.3	1.7180@-07	.0895	1.049@-06
28.594	405.00	6.064					
			.0000884	8153.7	1.9329@-07	.0842	1.105@-06
28.454	445.00	6.490					
			.0001619	8853.7	1.4328@-07	.0762	7.357@-07
28.175	555.00	7.337					
			.0003955	10551	8.2778@-08	.0549	2.887@-07
27.335	1240.0	9.797					
			.0000453	11940	3.7320@-08	.0369	8.719@-08
27.223	1500.0	10.12					

EQUILIBRIUM CONCENTRATION = 26.310 INITIAL SEED AREA = 2329

TABLE: 75 RUN NO.: R.E.9

TEMP: 70.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
29.763	.00000	2.000					
			.0001796	1663.7	4.8740@-06	.1302	4.490@-05
29.708	2.0000	2.177					
			.0003989	1873.2	3.1840@-06	.1265	2.849@-05
29.569	9.0000	2.617					
			.0006503	2296.6	3.2366@-06	.1186	2.709@-05
29.291	21.000	3.497					
			.0007568	2945.8	1.5532@-06	.1054	1.147@-05
28.873	54.000	4.811					

EQUILIBRIUM CONCENTRATION = 26.310 INITIAL SEED AREA = 1602

TABLE: 76 RUN NO.: R.P.G.7

TEMP: 70.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH P.G. PREPARED SIEVE FRACTION: 89-105μ

CONCN. M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
30.041	.00000	1.990					
			.0007146	1355.6	1.2685@-05	.2092	1.787@-04
29.863	2.0000	2.558					
			.0004618	1731.5	4.8819@-06	.2026	6.598@-05
29.715	5.5000	3.029					
			.0003853	2066.7	2.4582@-06	.1966	3.211@-05
29.566	11.500	3.499					
			.0003319	2390.1	1.7537@-06	.1906	2.213@-05
29.418	19.000	3.967					
			.0002926	2700.9	1.0922@-06	.1846	1.330@-05
29.270	30.000	4.434					
			.0002625	3000.1	1.2419@-06	.1786	1.459@-05
29.121	39.000	4.899					
			.0002387	3288.7	1.2410@-06	.1726	1.404@-05
28.973	47.500	5.363					
			.0002193	3567.7	9.5915@-07	.1666	1.044@-05
28.825	58.000	5.825					
			.0002031	3838.0	6.2640@-07	.1607	6.552@-06
28.676	73.500	6.286					
			.0001895	4100.3	6.9913@-07	.1547	7.018@-06
28.528	87.000	6.745					
			.0005050	4598.4	6.3694@-07	.1427	5.872@-06
28.083	130.00	8.115					
			.0002947	5198.8	7.2090@-07	.1278	5.894@-06
27.787	155.00	9.021					
			.0001373	5542.3	4.5420@-07	.1188	3.432@-06
27.639	175.00	9.470					
			.0001316	5764.1	7.0634@-07	.1128	5.482@-06
27.491	187.00	9.918					

EQUILIBRIUM CONCENTRATION = 24.771 INITIAL SEED AREA = 1150

TABLE: 77 RUN NO.: R.G.3

TEMP: 70.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH P.G. PREPARED SIEVE FRACTION: 89-105μ

CONCN. ΔM/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
29.819	.00000	1.975					
			.0004715	1268.1	2.8190@-07	.1313	2.620@-06
29.708	90.000	2.327					
			.0004787	1554.6	4.8827@-07	.1265	4.352@-06
29.569	145.00	2.765					
			.0010328	2166.8	6.0221@-07	.1160	4.918@-06
29.152	250.00	4.073					
			.0002652	2759.0	3.6421@-07	.1054	2.652@-06
29.012	300.00	4.505					
			.0002403	3033.8	4.3618@-07	.1001	3.003@-06
28.873	340.00	4.936					
			.0002201	3299.5	4.2371@-07	.0948	2.751@-06
28.734	380.00	5.365					
			.0002033	3556.8	3.0297@-07	.0895	1.848@-06
28.594	435.00	5.792					
			.0012324	4572.7	1.8276@-07	.0656	7.655@-07
27.475	1240.0	9.097					
			.0001221	5561.5	8.8597@-08	.0416	2.349@-07
27.335	1500.0	9.499					
			.0000699	5718.8	3.8382@-08	.0374	8.956@-08
27.251	1890.0	9.735					

EQUILIBRIUM CONCENTRATION = 26.310 INITIAL SEED AREA = 1141

TABLE: 78 RUN NO.: R.G.4

TEMP: 70.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
29.791	.00000	1.970					
			.0002680	1669.7	2.4184@-07	.1307	2.233@-06
29.708	60.000	2.234					
			.0003898	1906.9	2.9193@-07	.1265	2.598@-06
29.569	135.00	2.673					
			.0006374	2323.1	4.2664@-07	.1186	3.541@-06
29.291	225.00	3.545					
			.0002699	2720.2	4.3943@-07	.1107	3.374@-06
29.152	265.00	3.979					
			.0004739	3086.4	4.1786@-07	.1027	2.962@-06
28.873	345.00	4.843					
			.0002111	3439.9	3.6126@-07	.0948	2.345@-06
28.734	390.00	5.272					
			.0013030	4353.7	1.9731@-07	.0709	9.048@-07
27.615	1110.0	8.603					
			.0001299	5242.6	6.0130@-08	.0470	1.805@-07
27.475	1470.0	9.005					

EQUILIBRIUM CONCENTRATION = 26.310 INITIAL SEED AREA = 1578

TABLE: 79

RUN NO.: R.P.E.1

TEMP: 70.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH P.G. PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
30.012	.00000	1.980					
			.0038201	3055.3	1.5015@-04	.1727	1.910@-03
28.083	1.0000	8.121					
			.0001516	5083.6	1.8001@-05	.1307	1.516@-04
27.935	1.5000	8.576					
			.0001445	5315.7	9.0187@-06	.1248	7.226@-05
27.787	2.5000	9.030					
			.0002710	5653.7	7.3124@-06	.1158	5.420@-05
27.491	5.0000	9.935					
			.0002501	6091.6	5.4051@-06	.1038	3.572@-05
27.195	8.5000	10.84					
			.0002325	6513.9	3.3319@-06	.0919	1.937@-05
26.899	14.500	11.73					
			.0003216	7020.6	2.6649@-06	.0770	1.286@-05
26.455	27.000	13.06					
			.0001985	7511.8	1.8359@-06	.0620	7.091@-06
26.159	41.000	13.95					
			.0001878	7890.2	1.6856@-06	.0501	5.217@-06
25.864	59.000	14.82					

EQUILIBRIUM CONCENTRATION = 24.771 INITIAL SEED AREA = 1144

TABLE: 80 RUN NO.: R.P.G.T.1

TEMP: 70.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH P.G. PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
30.012	.00000	1.980					
			.0006123	1315.2	1.0926@-05	.2086	1.531@-04
29.863	2.0000	2.453					
			.0004828	1657.2	2.9755@-06	.2026	4.023@-05
29.715	8.0000	2.924					
			.0003994	1994.2	1.2738@-06	.1966	1.664@-05
29.566	20.000	3.394					
			.0003418	2319.6	6.1600@-07	.1906	7.768@-06
29.418	42.000	3.862					
			.0008128	2922.9	6.3783@-07	.1786	7.526@-06
28.973	96.000	5.259					
			.0004294	3638.0	5.9175@-07	.1637	6.314@-06
28.676	130.00	6.181					
			.0003720	4164.8	3.7913@-07	.1517	3.720@-06
28.380	180.00	7.096					
			.0003295	4662.7	3.3415@-07	.1397	2.996@-06
28.083	235.00	8.003					
			.0018409	6534.4	1.6748@-07	.0848	7.639@-07
25.657	1440.0	15.03					

EQUILIBRIUM CONCENTRATION = 24.771 INITIAL SEED AREA = 1144

TABLE: 81 RUN NO.: R.P.E.2

TEMP: 70.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH P.G.PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT K($MIN^{-1}CM^{-2}$)	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
29.774	.00000	1.990					
			.0016326	1723.4	1.8692@-04	.1918	2.474@-03
29.270	.33000	3.593					
			.0006087	2604.9	5.2380@-05	.1756	6.087@-04
28.973	.83000	4.528					
			.0004919	3199.9	2.2874@-05	.1637	2.460@-04
28.676	1.8300	5.456					
			.0004165	3755.6	1.2013@-05	.1517	1.190@-04
28.380	3.5800	6.380					
			.0003633	4278.8	6.8586@-06	.1397	6.221@-05
28.083	6.5000	7.298					
			.0003236	4774.9	6.5409@-06	.1278	5.393@-05
27.787	9.5000	8.210					
			.0002927	5248.0	4.9235@-06	.1158	3.658@-05
27.491	13.500	9.118					
			.0002678	5701.2	3.6751@-06	.1038	2.435@-05
27.195	19.000	10.02					
			.0002473	6137.0	3.0313@-06	.0919	1.766@-05
26.899	26.000	10.92					
			.0002300	6557.4	2.2830@-06	.0799	1.150@-05
26.603	36.000	11.81					
			.0002153	6963.8	1.9448@-06	.0680	8.280@-06
26.307	49.000	12.69					
			.0002024	7357.6	1.3834@-06	.0561	4.820@-06
26.012	70.000	13.57					
			.0000968	7646.1	1.5080@-06	.0471	4.401@-06
25.864	81.000	14.01					

EQUILIBRIUM CONCENTRATION = 24.771 INITIAL SEED AREA = 1150

TABLE: 82 RUN NO.: R.P.E.3

TEMP: 70.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH P.G. PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
27.343	.00000	1.983					
			.0005971	1312.3	2.2584@-05	.1008	1.493@-04
27.195	2.0000	2.443					
			.0004733	1645.2	7.6583@-06	.0949	4.733@-05
27.047	7.0000	2.902					
			.0003927	1973.9	2.7246@-06	.0889	1.571@-05
26.899	19.500	3.359					
			.0003368	2291.5	1.3381@-06	.0829	7.165@-06
26.751	43.000	3.815					
			.0002957	2596.9	7.4745@-07	.0770	3.697@-06
26.603	83.000	4.268					
			.0002644	2890.7	6.9326@-07	.0710	3.148@-06
26.455	125.00	4.720					
			.0002397	3173.7	7.2386@-07	.0650	2.996@-06
26.307	165.00	5.169					
			.0002195	3446.9	4.8924@-07	.0590	1.829@-06
26.159	225.00	5.616					
			.0002028	3711.0	5.0553@-07	.0531	1.690@-06
26.012	285.00	6.061					
			.0001885	3966.8	3.7617@-07	.0471	1.109@-06
25.864	370.00	6.503					
			.0001765	4214.9	5.7442@-07	.0411	1.470@-06
25.716	430.00	6.942					
			.0001655	4455.7	2.9346@-07	.0352	6.366@-07
25.568	560.00	7.378					

EQUILIBRIUM CONCENTRATION = 24.771 INITIAL SEED AREA = 1146

TABLE: 83 RUN NO.: R.P.E.4

TEMP: 70.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH F PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
27.343	.00000	1.987					
			.0002882	2715.7	1.4551@-05	.1008	9.607@-05
27.195	1.5000	2.447					
			.0002249	3459.0	1.4570@-06	.0949	8.998@-06
27.047	14.000	2.906					
			.0001863	4154.8	5.3933@-07	.0889	3.105@-06
26.899	44.000	3.363					
			.0001601	4809.3	3.6544@-07	.0829	1.953@-06
26.751	85.000	3.817					
			.0001412	5429.2	4.0860@-07	.0770	2.017@-06
26.603	120.00	4.270					
			.0001268	6019.9	4.6605@-07	.0710	2.114@-06
26.455	150.00	4.721					
			.0001155	6586.0	4.3602@-07	.0650	1.805@-06
26.307	182.00	5.170					
			.0001061	7130.1	2.8382@-07	.0590	1.061@-06
26.159	232.00	5.617					
			.0000984	7654.5	3.4199@-07	.0531	1.144@-06
26.012	275.00	6.062					
			.0001777	8402.8	2.3987@-07	.0441	6.583@-07
25.716	410.00	6.944					
			.0000490	9034.5	8.3859@-08	.0364	1.385@-07
25.627	540.00	7.206					

EQUILIBRIUM CONCENTRATION = 24.771 INITIAL SEED AREA = 2331

TABLE: 84 RUN NO.: R.P.G.K.1

TEMP: 70.0°C CELL: S STIRRER SPEED: 2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
27.461	.00000	1.928					
			.0001939	1608.7	2.7672@-06	.1074	1.939@-05
27.402	5.0000	2.112					
			.0000914	1704.2	4.1501@-08	.1056	2.856@-07
27.372	165.00	2.204					
			.0003353	1858.0	3.3431@-07	.1026	2.235@-06
27.254	240.00	2.572					
			.0003654	2126.1	2.4088@-07	.0973	1.523@-06
27.106	360.00	3.031					

EQUILIBRIUM CONCENTRATION = 24.771 INITIAL SEED AREA = 1545

TABLE: 85

RUN NO.: R.P.G.B.1

TEMP: 70.0°C CELL: S STIRRER SPEED: 2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105μ

CONCN. ΔM/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
27.491	.00000	1.960					
			.0002811	1665.9	3.9863@-06	.1080	2.811@-05
27.402	5.0000	2.237					
			.0004074	1913.9	5.3078@-07	.1032	3.573@-06
27.254	62.000	2.697					
			.0003514	2210.8	1.5443@-06	.0973	9.762@-06
27.106	80.000	3.155					
			.0003107	2493.2	1.3821@-06	.0913	8.175@-06
26.958	99.000	3.613					
			.0002795	2763.0	1.0563@-06	.0853	5.822@-06
26.810	123.00	4.069					
			.0002548	3022.2	1.1327@-06	.0793	5.790@-06
26.662	145.00	4.524					
			.0006577	3506.7	1.2295@-06	.0674	5.304@-06
26.218	207.00	5.884					
			.0001914	3976.1	7.1292@-07	.0555	2.518@-06
26.071	245.00	6.334					
			.0001808	4197.1	6.3913@-07	.0495	2.009@-06
25.923	290.00	6.782					
			.0001715	4412.6	4.7865@-07	.0435	1.319@-06
25.775	355.00	7.230					
			.0001632	4622.9	4.4709@-07	.0376	1.060@-06
25.627	432.00	7.675					

EQUILIBRIUM CONCENTRATION = 24.771 INITIAL SEED AREA = 1570

TABLE: 86 RUN NO.: R.P.D.2

TEMP: 70.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105 μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
27.076	.00000	1.975					
			.0005279	1769.7	3.8707@-05	.0895	2.256@-04
26.899	1.1700	2.526					
			.0003679	2104.1	1.3259@-06	.0829	7.121@-06
26.751	27.000	2.983					
			.0003222	2390.6	7.9216@-07	.0770	3.929@-06
26.603	68.000	3.438					
			.0005481	2791.3	5.2562@-07	.0680	2.284@-06
26.307	188.00	4.340					
			.0002382	3175.1	2.9782@-07	.0590	1.113@-06
26.159	295.00	4.787					
			.0002200	3416.1	4.3934@-07	.0531	1.467@-06
26.012	370.00	5.231					
			.0005617	3858.3	1.3095@-07	.0411	3.210@-07
25.568	1245.0	6.510					

EQUILIBRIUM CONCENTRATION = 24.771 INITIAL SEED AREA = 1582

TABLE: 87

RUN NO.: R.P.G.M.1

TEMP: 70.0°C CELL: S STIRRER SPEED: 2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105 μ

CONCN. ΔM/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
27.461	.00000	1.964					
			.0001905	1637.5	2.7184@-06	.1074	1.905@-05
27.402	5.0000	2.148					
			.0004204	1855.7	1.2481@-06	.1032	8.407@-06
27.254	30.000	2.609					
			.0003606	2155.8	1.7817@-06	.0973	1.127@-05
27.106	46.000	3.067					
			.0003174	2440.9	1.7881@-06	.0913	1.058@-05
26.958	61.000	3.525					
			.0002847	2713.1	1.9123@-06	.0853	1.055@-05
26.810	74.500	3.981					
			.0002589	2974.3	1.7462@-06	.0793	8.929@-06
26.662	89.000	4.436					
			.0002381	3225.9	1.9418@-06	.0734	9.156@-06
26.514	102.00	4.890					
			.0002208	3468.9	2.3229@-06	.0674	1.003@-05
26.366	113.00	5.343					
			.0002061	3704.3	1.3817@-06	.0614	5.424@-06
26.218	132.00	5.794					
			.0001936	3932.9	1.3042@-06	.0555	4.609@-06
26.071	153.00	6.244					
			.0003563	4262.5	1.0259@-06	.0465	3.020@-06
25.775	212.00	7.141					
			.0001647	4583.9	1.3353@-06	.0376	3.167@-06
25.627	238.00	7.587					
			.0001571	4790.6	2.7831@-07	.0316	5.532@-07
25.480	380.00	8.032					

EQUILIBRIUM CONCENTRATION = 24.771 INITIAL SEED AREA = 1573

TABLE: 88 RUN NO.: R.P.E.5

TEMP: 70.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
27.402	.00000	1.987					
			.0006046	1810.1	6.0428@-05	.1020	4.031@-04
27.195	.75000	2.632					
			.0003572	2174.5	5.5185@-06	.0949	3.402@-05
27.047	6.0000	3.090					
			.0003146	2459.6	1.6077@-06	.0889	9.252@-06
26.899	23.000	3.547					
			.0002821	2731.8	1.0551@-06	.0829	5.642@-06
26.751	48.000	4.002					
			.0002564	2992.7	9.2657@-07	.0770	4.579@-06
26.603	76.000	4.456					
			.0004539	3362.9	6.2326@-07	.0680	2.702@-06
26.307	160.00	5.357					
			.0003944	3832.4	6.1971@-07	.0561	2.191@-06
26.012	250.00	6.249					
			.0001797	4168.1	4.3472@-07	.0471	1.283@-06
25.864	320.00	6.692					
			.0001697	4382.3	3.4894@-07	.0411	8.932@-07
25.716	415.00	7.132					

EQUILIBRIUM CONCENTRATION = 24.771 INITIAL SEED AREA = 1592

TABLE: 89 RUN NO.: R.P.E.6

TEMP: 70.0°C CELL: S STIRRER SPEED: 2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
27.461	.00000	1.975					
			.0006082	1800.6	4.4520@-05	.1044	3.041@-04
27.254	1.0000	2.620					
			.0003590	2165.0	5.4066@-06	.0973	3.420@-05
27.106	6.2500	3.079					
			.0003162	2450.3	3.9583@-06	.0913	2.343@-05
26.958	13.000	3.537					
			.0002837	2722.8	5.1450@-06	.0853	2.837@-05
26.810	18.000	3.993					
			.0002581	2984.2	6.3089@-06	.0793	3.226@-05
26.662	22.000	4.448					
			.0002373	3236.0	6.2909@-06	.0734	2.967@-05
26.514	26.000	4.902					
			.0002201	3479.3	6.3689@-06	.0674	2.752@-05
26.366	30.000	5.354					
			.0002056	3715.1	5.2354@-06	.0614	2.056@-05
26.218	35.000	5.806					
			.0001553	3921.6	4.3465@-06	.0561	1.553@-05
26.100	40.000	6.166					
			.0002200	4144.3	5.3161@-06	.0501	1.692@-05
25.923	46.500	6.705					
			.0001728	4383.6	4.8181@-06	.0435	1.329@-05
25.775	53.000	7.152					
			.0001643	4595.5	5.3280@-06	.0376	1.264@-05
25.627	59.500	7.598					
			.0001568	4802.5	3.7545@-06	.0316	7.467@-06
25.480	70.000	8.043					
			.0001500	5005.1	3.1128@-06	.0256	5.001@-06
25.332	85.000	8.487					
			.0001439	5203.5	2.9352@-06	.0197	3.598@-06
25.184	105.00	8.930					

EQUILIBRIUM CONCENTRATION = 24.771 INITIAL SEED AREA 1582

TABLE: 90 RUN NO.: R.P.E.7

TEMP: 70.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105 μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
27.343	.00000	1.993					
			.0004447	1754.1	3.3791@-05	.1008	2.224@-04
27.195	1.0000	2.453					
			.0003772	2060.7	6.1143@-06	.0949	3.772@-05
27.047	6.0000	2.912					
			.0003292	2351.4	1.3614@-06	.0889	7.838@-06
26.899	27.000	3.369					
			.0002932	2628.4	8.8438@-07	.0829	4.730@-06
26.751	58.000	3.824					
			.0002653	2893.4	8.9449@-07	.0770	4.421@-06
26.603	88.000	4.278					
			.0002428	3148.1	1.0695@-06	.0710	4.857@-06
26.455	113.00	4.729					
			.0006289	3623.8	7.2086@-07	.0590	2.687@-06
26.012	230.00	6.074					
			.0001836	4084.5	4.4361@-07	.0471	1.311@-06
25.864	300.00	6.517					
			.0001732	4301.1	3.7527@-07	.0411	9.622@-07
25.716	390.00	6.957					

EQUILIBRIUM CONCENTRATION = 24.771 INITIAL SEED AREA = 1597

TABLE: 91 RUN NO.: R.P.E.8

TEMP 70.0° C CELL: S STIRRER SPEED 2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105 μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
27.402	.00000	1.955					
			.0004529	1723.7	5.0138@-05	.1032	3.380@-04
27.254	.67000	2.415					
			.0003832	2030.2	7.9034@-06	.0973	5.002@-05
27.106	4.5000	2.875					
			.0003341	2320.7	5.6420@-06	.0913	3.341@-05
26.958	9.5000	3.333					
			.0002976	2597.6	-3.3706@-06	.0853	-1.860@-05
26.810	1.5000	3.789					
			.0002692	2862.8	1.4220@-06	.0793	7.276@-06
26.662	20.000	4.245					
			.0002465	3117.7	5.2237@-06	.0734	2.465@-05
26.514	25.000	4.699					
			.0002278	3363.7	6.5878@-06	.0674	2.848@-05
26.366	29.000	5.151					
			.0002122	3601.8	5.4001@-06	.0614	2.122@-05
26.218	34.000	5.603					
			.0001988	3832.7	5.6209@-06	.0555	1.988@-05
26.071	39.000	6.053					
			.0001873	4057.2	4.2504@-06	.0495	1.338@-05
25.923	46.000	6.502					
			.0006615	4586.4	3.4102@-06	.0346	7.191@-06
25.332	92.000	8.293					

EQUILIBRIUM CONCENTRATION = 24.771 INITIAL SEED AREA = 1566

TABLE: 92 RUN NO.: R.P.D.3

TEMP: 70.0°C CELL: S STIRRER SPEED: 2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
27.402	.00000	1.935					
			.0004572	1707.7	1.2330@-05	.1032	8.312@-05
27.254	2.7500	2.395					
			.0003863	2014.0	7.6284@-06	.0973	4.829@-05
27.106	6.7500	2.855					
			.0003365	2304.2	5.9815@-06	.0913	3.542@-05
26.958	11.500	3.313					
			.0002995	2580.7	5.4282@-06	.0853	2.995@-05
26.810	16.500	3.769					
			.0002709	2845.5	5.8814@-06	.0793	3.010@-05
26.662	21.000	4.224					
			.0002479	3100.0	5.2537@-06	.0734	2.479@-05
26.514	26.000	4.679					
			.0002291	3345.5	5.8878@-06	.0674	2.545@-05
26.366	30.500	5.131					
			.0002133	3583.0	6.7854@-06	.0614	2.666@-05
26.218	34.500	5.583					
			.0001998	3813.4	6.2769@-06	.0555	2.220@-05
26.071	39.000	6.033					
			.0001882	4037.4	3.7373@-06	.0495	1.176@-05
25.923	47.000	6.482					
			.0001780	4255.5	5.3768@-06	.0435	1.484@-05
25.775	53.000	6.930					
			.0001691	4468.2	5.4797@-06	.0376	1.301@-05
25.627	59.500	7.376					
			.0001611	4676.0	3.8561@-06	.0316	7.673@-06
25.480	70.000	7.822					

EQUILIBRIUM CONCENTRATION = 24.771 INITIAL SEED AREA = 1550

TABLE: 93 RUN NO.: R.E.10

TEMP: 70.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105 μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
29.708	.00000	1.988					
			.0004289	1743.8	6.8405@-06	.1265	6.127@-05
29.569	3.5000	2.429					
			.0003659	2038.4	3.0551@-06	.1212	2.613@-05
29.430	10.500	2.869					
			.0003207	2318.5	1.3566@-06	.1160	1.106@-05
29.291	25.000	3.309					
			.0002868	2586.0	1.5407@-06	.1107	1.195@-05
29.152	37.000	3.747					
			.0002602	2842.7	1.6068@-06	.1054	1.183@-05
29.012	48.000	4.184					
			.0012201	3648.2	1.4516@-06	.0868	8.715@-06
28.175	118.00	6.797					
			.0001640	4430.5	9.7672@-07	.0683	4.556@-06
28.035	136.00	7.226					
			.0001564	4633.5	1.1401@-06	.0629	4.888@-06
27.895	152.00	7.654					
			.0001496	4832.2	1.4707@-06	.0576	5.754@-06
27.756	165.00	8.082					
			.0001434	5026.8	1.1919@-06	.0523	4.218@-06
27.615	182.00	8.508					
			.0001378	5217.5	1.2084@-06	.0470	3.828@-06
27.475	200.00	8.933					
			.0002607	5495.9	1.0013@-06	.0390	2.607@-06
27.195	250.00	9.779					
			.0002426	5857.0	6.5118@-07	.0283	1.213@-06
26.914	350.00	10.62					
			.0001150	6120.2	3.3234@-07	.0203	4.422@-07
26.774	480.00	11.03					
			.0001111	6290.6	5.2133@-07	.0150	5.050@-07
26.633	590.00	11.45					

EQUILIBRIUM CONCENTRATION = 26.310 INITIAL SEED AREA = 1593

TABLE: 94 RUN NO.: R.E.11

TEMP: 70.0°C CELL: S STIRRER SPEED: 2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
29.875	.00000	1.958					
			.0003542	1690.3	1.2486@-05	.1334	1.181@-04
29.763	1.5000	2.311					
			.0003818	1956.8	5.9943@-06	.1286	5.454@-05
29.624	5.0000	2.752					
			.0003328	2239.7	6.3754@-06	.1234	5.546@-05
29.485	8.0000	3.192					
			.0002963	2509.6	5.0981@-06	.1181	4.233@-05
29.346	11.500	3.632					
			.0002680	2768.3	4.8410@-06	.1128	3.829@-05
29.207	15.000	4.070					
			.0002454	3017.3	5.4399@-06	.1075	4.090@-05
29.068	18.000	4.508					
			.0002268	3257.8	6.3629@-06	.1022	4.537@-05
28.929	20.500	4.944					
			.0002113	3490.6	6.2666@-06	.0969	4.225@-05
28.789	23.000	5.380					
			.0001980	3716.7	6.2295@-06	.0916	3.960@-05
28.650	25.500	5.815					
			.0001866	3936.6	5.2055@-06	.0863	3.110@-05
28.510	28.500	6.249					
			.0001766	4150.9	4.5113@-06	.0810	2.523@-05
28.371	32.000	6.682					
			.0003281	4461.5	4.0822@-06	.0730	2.051@-05
28.091	40.000	7.547					
			.0001529	4765.2	3.8119@-06	.0651	1.699@-05
27.951	44.500	7.978					
			.0001466	4961.3	3.9897@-06	.0597	1.629@-05
27.811	49.000	8.407					
			.0004082	5339.7	4.5530@-06	.0491	1.512@-05
27.391	62.500	9.695					
			.0001264	5711.5	3.2412@-06	.0384	8.427@-06
27.251	70.000	10.12					
			.0001223	5891.0	4.5654@-06	.0331	1.019@-05
27.111	76.000	10.55					
			.0002336	6154.0	2.6974@-06	.0251	4.493@-06
26.830	102.00	11.40					
			.0001116	6413.2	2.7271@-06	.0171	3.101@-06
26.689	120.00	11.82					

EQUILIBRIUM CONCENTRATION = 26.310 INITIAL SEED AREA = 1569

TABLE: 95 RUN NO.: R.P.E.M.1

TEMP: 70.0°C CELL: S STIRRER SPEED: 2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105 μ

CONCN. ΔM/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
27.432	.00000	1.942					
			.0005378	1743.8	5.2831@-05	.1038	3.585@-04
27.254	.75000	2.495					
			.0003740	2079.2	6.9546@-06	.0973	4.400@-05
27.106	5.0000	2.954					
			.0003275	2366.7	5.5325@-06	.0913	3.275@-05
26.958	10.000	3.412					
			.0002926	2640.9	5.3046@-06	.0853	2.926@-05
26.810	15.000	3.868					
			.0002654	2903.7	5.1871@-06	.0793	2.654@-05
26.662	20.000	4.323					
			.0002434	3156.5	5.1596@-06	.0734	2.434@-05
26.514	25.000	4.777					
			.0002253	3400.6	5.2131@-06	.0674	2.253@-05
26.366	30.000	5.230					
			.0002100	3636.9	5.3479@-06	.0614	2.100@-05
26.218	35.000	5.681					
			.0001970	3866.2	5.5722@-06	.0555	1.970@-05
26.071	40.000	6.131					
			.0001858	4089.1	4.9201@-06	.0495	1.548@-05
25.923	46.000	6.580					
			.0003435	4411.3	4.7999@-06	.0405	1.227@-05
25.627	60.000	7.475					
			.0003122	4826.3	3.5060@-06	.0286	6.244@-06
25.332	85.000	8.366					
			.0001461	5127.3	3.3098@-06	.0197	4.058@-06
25.184	103.00	8.808					

EQUILIBRIUM CONCENTRATION = 24.771 INITIAL SEED AREA = 1556

TABLE: 96 RUN NO.: R.P.D.4

TEMP: 70.0°C CELL: A STIRRER SPEED: 2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
27.446	.00000	1.946					
			.0005368	1747.0	9.8309@-06	.1044	6.710@-05
27.269	4.0000	2.499					
			.0003735	2082.5	7.1537@-07	.0979	4.555@-06
27.121	45.000	2.958					
			.0003271	2370.1	6.0985@-07	.0919	3.635@-06
26.973	90.000	3.416					
			.0002923	2644.4	4.7824@-07	.0859	2.657@-06
26.825	145.00	3.872					
			.0005088	3031.8	3.7997@-07	.0770	1.885@-06
26.529	280.00	4.783					
			.0002250	3405.1	5.1605@-07	.0680	2.250@-06
26.381	330.00	5.236					
			.0002098	3641.5	8.8161@-07	.0620	3.497@-06
26.233	360.00	5.687					

EQUILIBRIUM CONCENTRATION = 24.771 INITIAL SEED AREA = 1559

TABLE: 97 RUN NO.: R.P.D.5

TEMP: 70.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105 μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
27.461	.00000	1.987					
			.0003634	1718.7	1.7463@-05	.1062	1.211@-04
27.343	1.5000	2.356					
			.0003900	1996.3	2.9692@-06	.1008	1.950@-05
27.195	11.500	2.815					
			.0003385	2290.1	9.6525@-07	.0949	5.938@-06
27.047	40.000	3.273					
			.0003004	2569.6	1.0900@-06	.0889	6.258@-06
26.899	64.000	3.729					
			.0002709	2836.9	7.0556@-07	.0829	3.762@-06
26.751	100.00	4.183					
			.0002473	3093.5	7.1711@-07	.0770	3.533@-06
26.603	135.00	4.635					
			.0002281	3340.6	7.8735@-07	.0710	3.564@-06
26.455	167.00	5.085					
			.0002119	3579.4	6.7559@-07	.0650	2.788@-06
26.307	205.00	5.533					
			.0003845	3921.2	6.0566@-07	.0561	2.136@-06
26.012	295.00	6.424					
			.0001757	4253.1	4.9704@-07	.0471	1.464@-06
25.864	355.00	6.865					
			.0001664	4465.1	4.3379@-07	.0411	1.109@-06
25.716	430.00	7.304					
			.0000958	4631.0	4.2535@-07	.0364	9.577@-07
25.627	480.00	7.567					

EQUILIBRIUM CONCENTRATION = 24.771 INITIAL SEED AREA = 1592

TABLE: 98 RUN NO.: R.P.D.6

TEMP: 70.0°C CELL: S STIRRER SPEED: 2000R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
27.609	.00000	1.910					
			.0006273	1748.6	1.4456@-05	.1104	1.046@-04
27.402	3.0000	2.556					
			.0003685	2112.8	1.8271@-06	.1032	1.228@-05
27.254	18.000	3.016					
			.0003237	2397.4	1.2817@-06	.0973	8.092@-06
27.106	38.000	3.474					
			.0002899	2668.9	1.7521@-06	.0913	1.035@-05
26.958	52.000	3.931					
			.0002633	2929.3	1.1386@-06	.0853	6.270@-06
26.810	73.000	4.387					
			.0002419	3179.9	1.2465@-06	.0793	6.365@-06
26.662	92.000	4.841					
			.0008176	3760.0	9.9742@-07	.0644	4.088@-06
26.071	192.00	6.653					
			.0003425	4427.7	1.0046@-06	.0465	2.953@-06
25.775	250.00	7.549					
			.0001590	4739.7	6.7156@-07	.0376	1.590@-06
25.627	300.00	7.994					
			.0001521	4940.6	3.4837@-07	.0316	6.914@-07
25.480	410.00	8.438					

EQUILIBRIUM CONCENTRATION = 24.771 INITIAL SEED AREA = 1530

TABLE: 99 RUN NO.: R.E.12

TEMP: 70.0°C CELL: S STIRRER SPEED: 2000R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
30.235	.00000	1.923					
			.0006009	1750.4	7.2444@-06	.1455	7.511@-05
30.041	4.0000	2.543					
			.0003558	2100.9	4.0088@-06	.1392	3.953@-05
29.902	8.5000	2.984					
			.0003139	2375.7	1.9519@-06	.1339	1.846@-05
29.763	17.000	3.424					
			.0005394	2762.5	3.0368@-06	.1260	2.697@-05
29.485	27.000	4.303					
			.0002364	3134.7	1.9047@-06	.1181	1.576@-05
29.346	34.500	4.741					
			.0002194	3370.0	1.4651@-06	.1128	1.155@-05
29.207	44.000	5.178					
			.0002051	3598.2	1.5206@-06	.1075	1.139@-05
29.068	53.000	5.614					
			.0001928	3819.9	1.6958@-06	.1022	1.205@-05
28.929	61.000	6.049					
			.0001821	4035.8	1.9357@-06	.0969	1.301@-05
28.789	68.000	6.484					
			.0001727	4246.4	1.3631@-06	.0916	8.636@-06
28.650	78.000	6.917					
			.0001644	4452.1	9.8630@-07	.0863	5.872@-06
28.510	92.000	7.350					
			.0004527	4847.1	1.0629@-06	.0757	5.520@-06
28.091	133.00	8.645					
			.0005282	5507.7	1.0221@-06	.0571	3.942@-06
27.531	200.00	10.36					

EQUILIBRIUM CONCENTRATION = 26.310 INITIAL SEED AREA = 1541

TABLE: 100 RUN NO.:R.E.13

TEMP:70.0°C CELL:C STIRRER SPEED:2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION:89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
29.902	.00000	1.971					
			.0005868	1788.7	5.9243@-06	.1329	5.589@-05
29.708	5.2500	2.589					
			.0003486	2139.0	2.0019@-06	.1265	1.788@-05
29.569	15.000	3.030					
			.0003081	2413.8	2.0067@-06	.1212	1.712@-05
29.430	24.000	3.469					
			.0002771	2676.8	1.7038@-06	.1160	1.386@-05
29.291	34.000	3.907					
			.0002526	2929.5	1.8135@-06	.1107	1.403@-05
29.152	43.000	4.344					
			.0002326	3173.1	1.5834@-06	.1054	1.163@-05
29.012	53.000	4.780					
			.0002160	3408.7	1.2940@-06	.1001	9.000@-06
28.873	65.000	5.215					
			.0002019	3637.1	1.2813@-06	.0948	8.414@-06
28.734	77.000	5.649					
			.0003596	3966.0	1.4685@-06	.0868	8.800@-06
28.454	98.000	6.515					
			.0003323	4388.4	1.4454@-06	.0762	7.553@-06
28.175	120.00	7.377					
			.0001545	4694.0	1.0371@-06	.0683	4.829@-06
28.035	136.00	7.805					
			.0001479	4890.9	9.0957@-07	.0629	3.892@-06
27.895	155.00	8.233					
			.0001419	5083.9	1.2115@-06	.0576	4.731@-06
27.756	170.00	8.659					
			.0003952	5455.9	8.9929@-07	.0470	2.823@-06
27.335	240.00	9.933					
			.0001225	5821.3	9.7159@-07	.0363	2.357@-06
27.195	266.00	10.35					
			.0001185	5997.4	6.8510@-07	.0310	1.410@-06
27.055	308.00	10.77					
			.0001147	6170.6	6.5079@-07	.0256	1.102@-06
26.914	360.00	11.19					
			.0000669	6307.0	2.6446@-07	.0214	3.716@-07
26.830	450.00	11.44					

EQUILIBRIUM CONCENTRATION = 26.310 INITIAL SEED AREA = 1579

TABLE: 101 RUN NO.: R.E.14

TEMP: 70.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
30.041	.00000	1.960					
			.0005901	1780.0	5.0073@-06	.1381	4.918@-05
29.847	6.0000	2.579					
			.0003503	2130.4	3.1336@-06	.1318	2.919@-05
29.708	12.000	3.019					
			.0003095	2405.3	2.4796@-06	.1265	2.210@-05
29.569	19.000	3.459					
			.0002782	2668.3	1.6338@-06	.1212	1.391@-05
29.430	29.000	3.898					
			.0002536	2920.9	2.6023@-06	.1160	2.113@-05
29.291	35.000	4.335					
			.0002335	3164.6	1.8886@-06	.1107	1.459@-05
29.152	43.000	4.772					
			.0002168	3400.2	1.8471@-06	.1054	1.355@-05
29.012	51.000	5.208					
			.0002027	3628.6	1.6208@-06	.1001	1.126@-05
28.873	60.000	5.642					
			.0001905	3850.6	1.4523@-06	.0948	9.527@-06
28.734	70.000	6.076					
			.0001800	4066.7	1.4574@-06	.0895	8.999@-06
28.594	80.000	6.508					
			.0001707	4277.4	1.6375@-06	.0842	9.484@-06
28.454	89.000	6.940					
			.0001625	4483.3	1.5016@-06	.0789	8.124@-06
28.315	99.000	7.370					
			.0001551	4684.5	1.4016@-06	.0736	7.051@-06
28.175	110.00	7.800					
			.0001485	4881.6	1.7729@-06	.0683	8.251@-06
28.035	119.00	8.228					
			.0004122	5261.0	1.1039@-06	.0576	4.294@-06
27.615	167.00	9.509					
			.0001275	5633.5	1.6787@-06	.0470	5.311@-06
27.475	179.00	9.933					
			.0001231	5813.0	9.5818@-07	.0416	2.677@-06
27.335	202.00	10.36					
			.0001191	5989.6	1.1691@-06	.0363	2.837@-06
27.195	223.00	10.78					
			.0001154	6163.2	9.6553@-07	.0310	1.990@-06
27.055	252.00	11.20					
			.0001119	6334.0	7.6671@-07	.0256	1.301@-06
26.914	295.00	11.62					
			.0002138	6584.0	8.4029@-07	.0176	9.460@-07
26.633	408.00	12.45					

EQUILIBRIUM CONCENTRATION = 26.310 INITIAL SEED AREA = 1570

TABLE: 102 RUN NO.: R.E.15

TEMP: 70.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
29.847	.00000	1.970					
			.0004328	1729.5	7.1262@-06	.1318	6.658@-05
29.708	3.2500	2.412					
			.0003687	2024.1	1.9187@-06	.1265	1.715@-05
29.569	14.000	2.852					
			.0003230	2304.2	1.8919@-06	.1212	1.615@-05
29.430	24.000	3.292					
			.0002886	2571.6	1.7735@-06	.1160	1.443@-05
29.291	34.000	3.730					
			.0002618	2828.1	1.7796@-06	.1107	1.378@-05
29.152	43.500	4.167					
			.0002402	3075.1	1.6339@-06	.1054	1.201@-05
29.012	53.500	4.604					
			.0002223	3313.7	1.5213@-06	.1001	1.059@-05
28.873	64.000	5.039					
			.0002073	3544.8	1.2135@-06	.0948	7.973@-06
28.734	77.000	5.473					
			.0001945	3769.2	1.5725@-06	.0895	9.724@-06
28.594	87.000	5.906					
			.0003574	4092.8	1.4478@-06	.0815	8.123@-06
28.315	109.00	6.770					
			.0001651	4408.3	1.2603@-06	.0736	6.348@-06
28.175	122.00	7.200					
			.0001574	4611.3	1.1261@-06	.0683	5.246@-06
28.055	137.00	7.629					
			.0004340	5000.9	1.0323@-06	.0576	4.019@-06
27.615	191.00	8.911					
			.0002620	5473.8	7.7432@-07	.0443	2.298@-06
27.335	248.00	9.759					
			.0001242	5746.8	1.1631@-06	.0363	2.822@-06
27.195	270.00	10.18					
			.0001200	5924.1	8.3230@-07	.0310	1.715@-06
27.055	305.00	10.60					
			.0001161	6098.2	5.7071@-07	.0256	9.672@-07
26.914	365.00	11.02					
			.0000676	6235.4	2.0935@-07	.0214	2.939@-07
26.830	480.00	11.27					

EQUILIBRIUM CONCENTRATION = 26.310 INITIAL SEED AREA = 1578

TABLE: 103 RUN NO.: R.E.16

TEMP: 70.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
29.875	.00000	1.974					
			.0005102	1762.0	1.0868@-05	.1323	1.020@-04
29.708	2.5000	2.504					
			.0003579	2084.7	2.6702@-06	.1265	2.386@-05
29.569	10.000	2.944					
			.0003149	2362.2	1.5379@-06	.1212	1.312@-05
29.430	22.000	3.384					
			.0005399	2752.6	2.4242@-06	.1133	1.928@-05
29.152	36.000	4.261					
			.0004556	3245.1	1.6734@-06	.1027	1.199@-05
28.873	55.000	5.133					
			.0002044	3595.8	1.5552@-06	.0948	1.022@-05
28.734	65.000	5.568					
			.0001920	3819.1	1.5519@-06	.0895	9.599@-06
28.594	75.000	6.001					
			.0001812	4036.5	1.7353@-06	.0842	1.007@-05
28.454	84.000	6.433					
			.0003355	4350.9	1.6036@-06	.0762	8.388@-06
28.175	104.00	7.295					
			.0001559	4658.1	1.6722@-06	.0683	7.795@-06
28.035	114.00	7.725					
			.0001492	4856.1	1.7406@-06	.0629	7.460@-06
27.895	124.00	8.153					
			.0001431	5050.1	1.2196@-06	.0576	4.770@-06
27.756	139.00	8.580					
			.0001375	5240.3	1.2958@-06	.0523	4.585@-06
27.615	154.00	9.006					
			.0001324	5427.0	8.7132@-07	.0470	2.759@-06
27.475	178.00	9.430					
			.0002513	5699.7	1.1494@-06	.0390	2.992@-06
27.195	220.00	10.28					
			.0001194	5967.7	7.8155@-07	.0310	1.613@-06
27.055	257.00	10.70					
			.0001156	6141.8	9.7142@-07	.0256	1.651@-06
26.914	292.00	11.12					
			.0002208	6396.8	1.2530@-06	.0176	1.415@-06
26.633	370.00	11.95					

EQUILIBRIUM CONCENTRATION = 26.310 INITIAL SEED AREA = 1581

TABLE: 104 RUN NO.: R.E.17

TEMP: 70.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105 μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
29.930	.00000	1.970					
			.0006609	1816.6	5.3632@-06	.1334	5.084@-05
29.708	6.5000	2.677					
			.0003397	2194.4	2.2383@-06	.1265	1.998@-05
29.569	15.000	3.117					
			.0003014	2466.8	2.2091@-06	.1212	1.884@-05
29.430	23.000	3.556					
			.0002719	2727.6	1.7600@-06	.1160	1.431@-05
29.291	32.500	3.994					
			.0002484	2978.4	2.4697@-06	.1107	1.911@-05
29.152	39.000	4.431					
			.0002292	3220.4	1.5601@-06	.1054	1.146@-05
29.012	49.000	4.867					
			.0002131	3454.6	1.5322@-06	.1001	1.066@-05
28.873	59.000	5.302					
			.0001995	3681.8	1.5189@-06	.0948	9.975@-06
28.734	69.000	5.737					
			.0001877	3902.6	1.2656@-06	.0895	7.822@-06
28.594	81.000	6.169					
			.0001775	4117.7	1.7011@-06	.0842	9.862@-06
28.454	90.000	6.601					
			.0003293	4429.0	1.5753@-06	.0762	8.233@-06
28.175	110.00	7.463					
			.0001533	4733.4	1.0285@-06	.0683	4.790@-06
28.035	126.00	7.892					
			.0001468	4929.6	1.0086@-06	.0629	4.317@-06
27.895	143.00	8.319					
			.0001409	5122.0	1.3875@-06	.0576	5.420@-06
27.756	156.00	8.746					
			.0001355	5310.6	1.1987@-06	.0523	4.236@-06
27.615	172.00	9.171					
			.0003789	5674.9	8.7802@-07	.0416	2.429@-06
27.195	250.00	10.44					
			.0001179	6032.8	1.0216@-06	.0310	2.105@-06
27.055	278.00	10.86					
			.0002246	6289.8	6.4307@-07	.0230	9.598@-07
26.774	395.00	11.70					
			.0001071	6542.3	5.8042@-07	.0150	5.635@-07
26.633	490.00	12.11					

EQUILIBRIUM CONCENTRATION = 26.310 INITIAL SEED AREA = 1578

TABLE: 105 RUN NO.: R.E.19

TEMP: 70.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
29.902	.00000	1.980					
			.0005845	1795.9	7.7442@-06	.1329	7.306@-05
29.708	4.0000	2.598					
			.0003474	2146.3	2.1613@-06	.1265	1.930@-05
29.569	13.000	3.039					
			.0003071	2421.3	1.5003@-06	.1212	1.280@-05
29.430	25.000	3.478					
			.0002763	2684.4	1.8877@-06	.1160	1.535@-05
29.291	34.000	3.916					
			.0002519	2937.3	1.3565@-06	.1107	1.050@-05
29.152	46.000	4.353					
			.0002320	3181.1	1.7549@-06	.1054	1.289@-05
29.012	55.000	4.789					
			.0002155	3416.9	1.2909@-06	.1001	8.978@-06
28.873	67.000	5.224					
			.0002015	3645.5	1.3945@-06	.0948	9.157@-06
28.734	78.000	5.658					
			.0001894	3867.7	1.5324@-06	.0895	9.471@-06
28.594	88.000	6.091					
			.0001790	4084.0	1.7151@06	.0842	9.943@-06
28.454	97.000	6.523					
			.0001698	4295.0	1.4249@-06	.0789	7.716@-06
28.315	108.00	6.954					
			.0003162	4600.9	1.2549@-06	.0709	6.080@-06
28.035	134.00	7.813					
			.0001477	4900.2	1.5681@-06	.0629	6.713@-06
27.895	145.00	8.241					
			.0001417	5093.5	1.0077@-06	.0576	3.936@-06
27.756	163.00	8.668					
			.0001363	5283.1	1.2050@-06	.0523	4.258@-06
27.615	179.00	9.093					
			.0001313	5469.1	1.2969@-06	.0470	4.102@-06
27.475	195.00	9.517					
			.0001268	5651.9	1.3333@-06	.0416	3.729@-06
27.335	212.00	9.941					
			.0002409	5919.2	8.5711@-07	.0337	1.912@-06
27.055	275.00	10.78					
			.0002257	6266.5	7.5519@-07	.0230	1.128@-06
26.774	375.00	11.62					

EQUILIBRIUM CONCENTRATION = 26.310 INITIAL SEED AREA = 1586

TABLE: 106 RUN NO.: R.E.20

TEMP: 70.0°C CELL: C STIRRER SPEED: 2500 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105 μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
29.902	.00000	1.967					
			.0005879	1785.5	6.2317@-06	.1329	5.879@-05
29.708	5.0000	2.585					
			.0003492	2135.7	3.2581@-06	.1265	2.910@-05
29.569	11.000	3.026					
			.0003086	2410.6	2.5835@-06	.1212	2.204@-05
29.430	18.000	3.465					
			.0002775	2673.5	1.8954@-06	.1160	1.542@-05
29.291	27.000	3.903					
			.0009076	3276.1	3.7151@-06	.1027	2.669@-05
28.734	44.000	5.653					
			.0001901	3859.5	3.0713@-06	.0895	1.901@-05
28.594	49.000	6.087					
			.0011016	4673.6	3.3239@-06	.0682	1.530@-05
27.615	85.000	9.117					
			.0005022	5738.9	1.7780@-06	.0390	4.566@-06
27.055	140.00	10.82					
			.0002925	6321.3	1.0658@-06	.0214	1.463@-06
26.689	240.00	11.91					
			.0000429	6576.1	4.0610@-07	.0134	3.574@-07
26.633	300.00	12.08					

EQUILIBRIUM CONCENTRATION = 26.310 INITIAL SEED AREA = 1576

TABLE: 107 RUN NO.: R.E.21

TEMP: 70.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT K($MIN^{-1}CM^{-2}$)	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
29.902	.00000	1.938					
			.0005957	1762.1	4.5102@-06	.1329	4.255@-05
29.708	7.0000	2.556					
			.0003531	2112.0	2.4710@-06	.1265	2.207@-05
29.569	15.000	2.997					
			.0003116	2386.4	2.0297@-06	.1212	1.731@-05
29.430	24.000	3.436					
			.0005357	2772.7	2.1058@-06	.1133	1.674@-05
29.152	40.000	4.312					
			.0002348	3144.3	2.2827@-06	.1054	1.677@-05
29.012	47.000	4.748					
			.0002180	3379.2	1.9580@-06	.1001	1.362@-05
28.873	55.000	5.184					
			.0002037	3607.0	1.9380@-06	.0948	1.273@-05
28.734	63.000	5.618					
			.0003728	3934.8	1.9427@-06	.0868	1.165@-05
28.454	79.000	6.484					
			.0001715	4254.2	1.5824@-06	.0789	8.576@-06
28.315	89.000	6.916					
			.0001632	4459.4	1.6196@-06	.0736	8.162@-06
28.175	99.000	7.346					
			.0001558	4660.0	1.3929@-06	.0683	6.493@-06
28.035	111.00	7.775					
			.0002925	4951.7	1.1525@-06	.0603	4.717@-06
27.756	142.00	8.631					
			.0001375	5237.7	1.0804@-06	.0523	3.821@-06
27.615	160.00	9.056					
			.0001324	5422.7	7.2166@-07	.0470	2.283@-06
27.475	189.00	9.481					
			.0001278	5604.5	6.7231@-07	.0416	1.879@-06
27.335	223.00	9.904					
			.0001233	5782.9	5.4104@-07	.0363	1.312@-06
27.195	270.00	10.32					
			.0001191	5958.2	4.8273@-07	.0310	9.929@-07
27.055	330.00	10.74					

EQUILIBRIUM CONCENTRATION = 26.310 INITIAL SEED AREA = 1553

TABLE: 108 RUN NO.: R.E.23

TEMP: 70.0°C CELL: G STIRRER SPEED: 2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
29.611	.00000	1.930					
			.0021360	2453.1	2.1530@-06	.1075	1.669@-05
28.664	64.000	4.934					
			.0005993	3693.5	2.6519@-06	.0815	1.498@-05
28.245	84.000	6.240					
			.0001771	4132.2	2.0153@-06	.0709	9.841@-06
28.105	93.000	6.673					
			.0000343	4258.2	2.1680@-07	.0677	1.009@-06
28.077	110.00	6.759					
			.0001340	4360.9	7.4960@-07	.0651	3.349@-06
27.965	130.00	7.104					
			.0001605	4543.5	9.7151@-07	.0603	4.012@-06
27.825	150.00	7.535					
			.0004429	4934.4	2.6314@-06	.0496	8.859@-06
27.405	175.00	8.826					
			.0002676	5409.4	1.6566@-06	.0363	4.054@-06
27.125	208.00	9.681					

EQUILIBRIUM CONCENTRATION = 26.310 INITIAL SEED AREA = 1546

TABLE : 109 RUN NO. : R.E.24

TEMP : 70.0°C CELL : G STIRRER SPEED : 2000 R.P.M.

SEED : BATCH E PREPARED SIEVE FRACTION : 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
29.958	.00000	1.935					
			.0005968	1759.8	6.2223@-06	.1350	5.968@-05
29.763	5.0000	2.554					
			.0003537	2109.9	3.8916@-06	.1286	3.537@-05
29.624	10.000	2.994					
			.0003123	2384.5	2.5664@-06	.1234	2.230@-05
29.485	17.000	3.434					
			.0002807	2647.2	2.1145@-06	.1181	1.754@-05
29.346	25.000	3.873					
			.0002557	2899.6	1.7974@-06	.1128	1.421@-05
29.207	34.000	4.311					
			.0002354	3142.9	1.5668@-06	.1075	1.177@-05
29.068	44.000	4.748					
			.0004035	3479.8	1.8123@-06	.0998	1.261@-05
28.803	60.000	5.577					
			.0001932	3806.8	1.6796@-06	.0921	1.073@-05
28.664	69.000	6.012					
			.0003559	4128.0	1.3897@-06	.0842	8.089@-06
28.385	91.000	6.880					
			.0003222	4541.4	1.2249@-06	.0736	6.196@-06
28.105	117.00	7.744					
			.0004347	5031.9	6.3749@-07	.0603	2.619@-06
27.686	200.00	9.036					
			.0008560	5941.7	3.0074@-06	.0336	6.114@-06
26.703	270.00	12.04					

EQUILIBRIUM CONCENTRATION = 26.310 INITIAL SEED AREA = 1550

TABLE: 110 RUN NO.: R.E.25

TEMP: 70.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCHE PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
29.763	.00000	1.962					
			.0005888	1781.3	4.6490@-06	.1276	4.206@-05
29.569	7.0000	2.580					
			.0003496	2131.2	2.0455@-06	.1212	1.748@-05
29.430	17.000	3.020					
			.0003089	2405.7	2.1064@-06	.1160	1.716@-05
29.291	26.000	3.459					
			.0002778	2668.4	1.7918@-06	.1107	1.389@-05
29.152	36.000	3.897					
			.0002531	2920.7	1.7202@-06	.1054	1.266@-05
29.012	46.000	4.333					
			.0002331	3163.9	1.5209@-06	.1001	1.060@-05
28.873	57.000	4.769					
			.0002164	3399.2	1.6451@-06	.0948	1.082@-05
28.734	67.000	5.204					
			.0002023	3627.3	1.4854@-06	.0895	9.197@-06
28.594	78.000	5.637					
			.0001902	3848.9	1.4890@-06	.0842	8.646@-06
28.454	89.000	6.070					
			.0001797	4064.7	1.5057@-06	.0789	8.167@-06
28.315	100.00	6.502					
			.0001704	4275.1	1.2067@-06	.0736	6.085@-06
28.175	114.00	6.932					
			.0001622	4480.5	1.2418@-06	.0683	5.791@-06
28.035	128.00	7.362					
			.0001547	4681.4	7.5231@-07	.0629	3.224@-06
27.895	152.00	7.790					
			.0001481	4877.9	9.0189@-07	.0576	3.526@-06
27.756	173.00	8.217					
			.0001421	5070.4	1.1817@-06	.0523	4.179@-06
27.615	190.00	8.642					
			.0001366	5259.2	1.1988@-06	.0470	3.795@-06
27.475	208.00	9.067					
			.0001315	5444.5	7.3531@-07	.0416	2.055@-06
27.335	240.00	9.490					
			.0001268	5626.3	4.3561@-07	.0363	1.057@-06
27.195	300.00	9.911					

EQUILIBRIUM CONCENTRATION = 26.310 INITIAL SEED AREA = 1572

TABLE: 111 RUN NO.: R.E.26

TEMP: 70.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105 μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
29.708	.00000	1.953					
			.0004359	1715.7	6.0835@-06	.1265	5.449@-05
29.569	4.000C	2.394					
			.0003710	2010.0	2.4098@-06	.1212	2.061@-05
29.430	13.000	2.834					
			.0003248	2289.7	1.9918@-06	.1160	1.624@-05
29.291	23.000	3.274					
			.0005536	2682.4	1.7407@-06	.1080	1.318@-05
29.012	44.000	4.150					
			.0002412	3059.7	1.9222@-06	.1001	1.340@-05
28.873	53.000	4.586					
			.0004319	3411.5	1.6079@-06	.0921	1.028@-05
28.594	74.000	5.456					
			.0013193	4460.4	1.3711@-06	.0656	5.997@-06
27.475	184.00	8.912					
			.0001336	5370.7	9.9389@-07	.0416	2.782@-06
27.335	208.00	9.335					
			.0001287	5553.6	8.2746@-07	.0363	2.011@-06
27.195	240.00	9.758					
			.0001243	5733.2	8.6000@-07	.0310	1.776@-06
27.055	275.00	10.18					
			.0001200	5909.7	5.4362@-07	.0256	9.234@-07
26.914	340.00	10.60					
			.0001160	6082.9	5.7960@-07	.0203	7.734@-07
26.774	415.00	11.02					

EQUILIBRIUM CONCENTRATION = 26.310 INITIAL SEED AREA = 1565

TABLE: 112 RUN NO.: R.E.27

TEMP: 70.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105 μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
33.294	.00000	1.960					
			.0049731	4405.8	6.9080@-06	.1973	1.051@-04
29.708	23.670	13.72					
			.0001946	7401.6	3.2208@-06	.1239	2.718@-05
29.430	27.250	14.57					
			.0005370	8023.2	2.3200@-06	.1027	1.603@-05
28.594	44.000	17.12					
			.0000828	8558.2	4.2092@-06	.0842	2.365@-05
28.454	45.750	17.54					
			.0000812	8704.7	1.4731@-06	.0789	7.733@-06
28.315	51.000	17.95					
			.0000797	8849.7	1.6322@-06	.0736	7.970@-06
28.175	56.000	18.37					
			.0000783	8993.3	1.5747@-06	.0683	7.115@-06
28.035	61.500	18.79					
			.0001525	9205.8	1.6014@-06	.0603	6.354@-06
27.756	73.500	19.62					
			.0000743	9416.2	1.6642@-06	.0523	5.713@-06
27.615	80.000	20.03					
			.0000730	9554.6	1.3197@-06	.0470	4.057@-06
27.475	89.000	20.44					
			.0000718	9691.7	1.1015@-06	.0416	2.993@-06
27.335	101.00	20.85					
			.0000707	9827.5	1.1510@-06	.0363	2.718@-06
27.195	114.00	21.26					
			.0000695	9962.1	8.2489@-07	.0310	1.655@-06
27.055	135.00	21.67					
			.0000684	10095	8.2738@-07	.0256	1.368@-06
26.914	160.00	22.08					

EQUILIBRIUM CONCENTRATION = 26.310 INITIAL SEED AREA = 1570

TABLE: 113 RUN NO.: R.E.28

TEMP: 60.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105 μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
27.623	.00000	1.960					
			.0021265	2486.2	1.2522@-05	.3446	2.363@-04
26.650	4.5000	4.992					
			.0003988	3623.2	4.8603@-06	.3136	7.976@-05
26.371	7.0000	5.845					
			.0003549	4054.2	4.5486@-06	.2998	7.099@-05
26.092	9.5000	6.695					
			.0006163	4657.9	4.2631@-06	.2791	6.163@-05
25.534	14.500	8.390					
			.0002715	5237.0	2.9291@-06	+ .2583	3.879@-05
25.254	18.000	9.230					
			.0001284	5512.4	2.5387@-06	.2479	3.211@-05
25.114	20.000	9.648					
			.0001242	5691.3	2.0248@-06	.2409	2.484@-05
24.974	22.500	10.07					
			.0001202	5867.4	1.8395@-06	.2340	2.186@-05
24.834	25.250	10.48					
			.0001165	6040.8	2.2517@-06	.2271	2.590@-05
24.693	27.500	10.90					
			.0002233	6295.2	2.2671@-06	.2166	2.481@-05
24.413	32.000	11.73					
			.0002112	6627.6	2.0734@-06	.2027	2.112@-05
24.132	37.000	12.56					
			.0001014	6871.8	1.3184@-06	.1923	1.268@-05
23.991	41.000	12.97					
			.0000990	7031.3	1.7835@-06	.1853	1.649@-05
23.850	44.000	13.38					
			.0001912	7266.5	1.3735@-06	.1748	1.195@-05
23.569	52.000	14.20					
			.0000923	7498.9	9.4415@-07	.1644	7.688@-06
23.428	58.000	14.61					
			.0000903	7651.0	1.1603@-06	.1574	9.026@-06
23.287	63.000	15.02					
			.0000884	7801.5	1.3239@-06	.1504	9.817@-06
23.146	67.500	15.42					
			.0000865	7950.3	1.7526@-06	.1434	1.236@-05
23.005	71.000	15.83					
			.0000848	8097.6	1.4077@-06	.1364	9.423@-06
22.864	75.500	16.24					

TABLE :113 (CONTD.) RUN NO. : R.E.28 (CONTD.)

CONCN. GM/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
22.864	75.500	16.24					
			.0000832	8243.4	1.0097@-06	.1294	6.397@-06
22.722	82.000	16.64					
			.0000816	8387.7	1.1371@-06	.1224	6.798@-06
22.581	88.000	17.05					
			.0000801	8530.6	1.0171@-06	.1154	5.719@-06
22.440	95.000	17.45					
			.0000786	8672.1	8.7769@-07	.1084	4.625@-06
22.298	103.50	17.85					
			.0000772	8812.3	6.0415@-07	.1014	2.970@-06
22.157	116.50	18.25					
			.0000759	8951.2	8.7493@-07	.0944	3.994@-06
22.015	126.00	18.66					
			.0000746	9088.9	8.0446@-07	.0874	3.391@-06
21.873	137.00	19.06					
			.0001456	9292.7	7.5942@-07	.0768	2.800@-06
21.590	163.00	19.86					
			.0000710	9494.7	5.0853@-07	.0663	1.614@-06
21.448	185.00	20.25					
			.0000699	9627.6	4.1176@-07	.0592	1.166@-06
21.306	215.00	20.65					
			.0000689	9759.5	5.5364@-07	.0522	1.377@-06
21.164	240.00	21.05					
			.0000677	9890.2	5.2681@-07	.0452	1.129@-06
21.021	270.00	21.44					

EQUILIBRIUM CONCENTRATION = 20.181 INITIAL SEED AREA = 1570

TABLE: 114 RUN NO.: R.E.29

TEMP: 60.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
27.331	.00000	1.984					
			.0005717	1794.6	1.5367@-05	.3495	2.859@-04
27.137	1.0000	2.589					
			.0003412	2137.7	1.2592@-05	.3412	2.274@-04
26.998	1.7500	3.019					
			.0003022	2407.4	8.5631@-06	.3343	1.511@-04
26.859	2.7500	3.449					
			.0002724	2665.7	8.5873@-06	.3274	1.480@-04
26.719	3.6700	3.878					
			.0002486	2914.2	8.8990@-06	.3205	1.498@-04
26.580	4.5000	4.306					
			.0002293	3153.9	6.9785@-06	.3136	1.146@-04
26.441	5.5000	4.733					
			.0002131	3386.0	6.6500@-06	.3067	1.066@-04
26.302	6.5000	5.160					
			.0001994	3611.2	5.4550@-06	.2998	8.522@-05
26.162	7.6700	5.585					
			.0001876	3830.2	5.7062@-06	.2929	8.687@-05
26.023	8.7500	6.010					
			.0001774	4043.6	5.5388@-06	.2860	8.212@-05
25.883	9.8300	6.434					
			.0001684	4251.8	4.1079@-06	.2791	5.928@-05
25.743	11.250	6.857					
			.0001604	4455.2	4.5692@-06	.2721	6.414@-05
25.604	12.500	7.279					
			.0001532	4654.3	3.2074@-06	.2652	4.377@-05
25.464	14.250	7.700					
			.0001467	4849.3	3.8977@-06	.2583	5.167@-05
25.324	15.670	8.121					
			.0001409	5040.5	3.4650@-06	.2514	4.459@-05
25.184	17.250	8.541					
			.0001356	5228.1	3.1033@-06	.2444	3.874@-05
25.044	19.000	8.959					
			.0001307	5412.4	2.7011@-06	.2375	3.268@-05
24.904	21.000	9.378					
			.0001263	5593.6	2.6937@-06	.2305	3.158@-05
24.764	23.000	9.795					
			.0001221	5771.9	2.6931@-06	.2236	3.053@-05
24.623	25.000	10.21					
			.0002332	6033.2	2.8858@-06	.2132	3.109@-05
24.343	28.750	11.04					
			.0001114	6290.6	3.6397@-06	.2027	3.712@-05
24.202	30.250	11.46					
			.0001082	6458.4	3.1484@-06	.1958	3.093@-05
24.061	32.000	11.87					

TABLE 114 (CONTD.) RUN NO.: R.E.29 (CONTD.)

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
24.061	32.000	11.87					
			.0001053	6623.9	1.8578@-06	.1888	1.755@-05
23.921	35.000	12.28					
			.0001026	6787.1	1.8836@-06	.1818	1.710@-05
23.780	38.000	12.69					
			.0001000	6948.3	2.2972@-06	.1748	2.000@-05
23.639	40.500	13.10					
			.0000976	7107.4	1.4628@-06	.1679	1.220@-05
23.498	44.500	13.51					
			.0000953	7264.5	1.7075@-06	.1609	1.361@-05
23.358	48.000	13.92					
			.0001843	7496.3	1.4596@-06	.1504	1.084@-05
23.075	56.500	14.74					
			.0000891	7725.4	1.4383@-06	.1399	9.895@-06
22.934	61.000	15.15					
			.0000872	7875.4	8.9157@-07	.1329	5.812@-06
22.793	68.500	15.55					
			.0000854	8023.9	1.9805@-06	.1259	1.220@-05
22.652	72.000	15.96					
			.0000837	8170.8	1.2020@-06	.1189	6.974@-06
22.510	78.000	16.36					
			.0000821	8316.1	8.8634@-07	.1119	4.827@-06
22.369	86.500	16.76					
			.0000805	8460.0	1.1293@-06	.1049	5.750@-06
22.227	93.500	17.17					
			.0000790	8602.4	1.2825@-06	.0979	6.077@-06
22.086	100.00	17.57					
			.0001538	8813.1	9.1410@-07	.0874	3.845@-06
21.802	120.00	18.37					
			.0000748	9021.7	7.2482@-07	.0768	2.673@-06
21.661	134.00	18.77					
			.0000736	9158.7	6.8811@-07	.0698	2.299@-06
21.519	150.00	19.17					
			.0000723	9294.5	6.0368@-07	.0628	1.808@-06
21.377	170.00	19.56					
			.0000711	9429.0	6.0978@-07	.0557	1.616@-06
21.235	192.00	19.96					
			.0000699	9562.3	5.5893@-07	.0487	1.520@-06
21.093	215.00	20.35					
			.0000688	9694.4	6.9995@-07	.0416	1.376@-06
20.950	240.00	20.75					
			.0000676	9825.1	4.6261@-07	.0346	7.506@-07
20.808	285.00	21.14					
			.0000664	9954.5	3.6964@-07	.0275	4.743@-07
20.666	355.00	21.53					

EQUILIBRIUM CONCENTRATION = 20.181 INITIAL SEED AREA = 1589

TABLE: 115 RUN NO.: R.E.30

TEMP: 60.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
27.414	.00000	1.972					
			.0006483	1129.0	3.4337@-05	.3550	6.483@-04
27.276	.50000	2.404					
			.0005619	1299.4	1.3009@-05	.3481	2.401@-04
27.137	1.6700	2.835					
			.0004982	1462.3	1.0380@-05	.3412	1.873@-04
26.998	3.0000	3.265					
			.0004491	1618.5	8.4913@-06	.3343	1.497@-04
26.859	4.5000	3.695					
			.0007892	1840.5	1.3221@-05	.3240	2.255@-04
26.580	6.2500	4.552					
			.0003515	2055.2	1.0709@-05	.3136	1.758@-04
26.441	7.2500	4.979					
			.0003290	2191.8	5.8706@-06	.3067	9.399@-05
26.302	9.0000	5.405					
			.0003095	2324.6	7.9319@-06	.2998	1.238@-04
26.162	10.250	5.830					
			.0002926	2454.1	7.6948@-06	.2929	1.170@-04
26.023	11.500	6.255					
			.0010404	2760.9	6.9377@-06	.2756	9.909@-05
25.464	16.750	7.950					
			.0004563	3117.8	5.8197@-06	.2548	7.605@-05
25.184	19.750	8.790					
			.0004240	3342.2	5.3058@-06	.2409	6.522@-05
24.904	23.000	9.628					
			.0003965	3559.1	4.9149@-06	.2271	5.665@-05
24.623	26.500	10.46					
			.0005521	3820.2	4.1729@-06	.2097	4.417@-05
24.202	32.750	11.71					
			.0001735	4024.6	4.4208@-06	.1958	4.339@-05
24.061	34.750	12.12					
			.0001690	4123.9	3.9787@-06	.1888	3.756@-05
23.921	37.000	12.53					
			.0001648	4221.8	2.5956@-06	.1818	2.354@-05
23.780	40.500	12.94					
			.0001608	4318.6	6.1602@-06	.1748	5.359@-05
23.639	42.000	13.35					
			.0001570	4414.1	3.1403@-06	.1679	5.616@-05
23.498	45.000	13.76					

TABLE : 11(CONTD.) RUN NO. : R.E.30(CONTD.)

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
23.498	45.000	13.76					
			.0001534	4508.6	2.7512@-06	.1609	2.191@-05
23.358	48.500	14.17					
			.0001500	4601.9	2.8193@-06	.1539	2.142@-05
23.217	52.000	14.58					
			.0002907	4739.7	2.4225@-06	.1434	1.710@-05
22.934	60.500	15.39					
			.0001407	4875.9	2.4001@-06	.1329	1.563@-05
22.793	65.000	15.80					
			.0001380	4965.3	2.6357@-06	.1259	1.623@-05
22.652	69.250	16.20					
			.0002680	5097.3	1.7347@-06	.1154	9.747@-06
22.369	83.000	17.01					
			.0001302	5228.0	2.1320@-06	.1049	1.085@-05
22.227	89.000	17.41					
			.0001278	5313.9	1.4995@-06	.0979	7.099@-06
22.086	98.000	17.81					
			.0001255	5398.8	1.3016@-06	.0909	5.703@-06
21.944	109.00	18.22					
			.0001233	5483.0	1.5288@-06	.0838	6.163@-06
21.802	119.00	18.61					
			.0002404	5607.5	1.2253@-06	.0733	4.293@-06
21.519	147.00	19.41					
			.0002324	5771.3	1.2534@-06	.0592	3.522@-06
21.235	180.00	20.20					
			.0001133	5892.4	1.8919@-06	.0487	4.359@-06
21.093	193.00	20.60					
			.0001115	5972.1	8.8767@-07	.0416	1.742@-06
20.950	225.00	20.99					
			.0001097	6051.0	9.6577@-07	.0346	1.567@-06
20.808	260.00	21.38					
			.0001080	6129.1	1.0506@-06	.0275	1.349@-06
20.666	300.00	21.78					
			.0001059	6206.4	4.3137@-07	.0205	4.075@-07
20.523	430.00	22.16					

EQUILIBRIUM CONCENTRATION = 20.181 INITIAL SEED AREA = 1042

TABLE: 116 RUN NO.:R.P.E.9

TEMP:70.0°C CELL:C STIRRER SPEED:2000 R.P.M.

SEED: BATCH P.E.PREPARED SIEVE FRACTION:89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
27.461	.00000	1.957					
			.0005633	1108.6	2.7074@-05	.1062	1.878@-04
27.343	1.5000	2.326					
			.0006118	1272.3	7.1674@-06	.1008	4.706@-05
27.195	8.0000	2.785					
			.0005363	1446.3	3.1114@-06	.0949	1.915@-05
27.047	22.000	3.243					
			.0004793	1612.4	2.7794@-06	.0889	1.598@-05
26.899	37.000	3.700					
			.0004347	1771.8	3.6972@-06	.0829	1.976@-05
26.751	48.000	4.155					
			.0011130	2069.0	2.4524@-06	.0710	1.113@-05
26.307	98.000	5.513					
			.0003222	2356.7	2.6834@-06	.0590	1.007@-05
26.159	114.00	5.961					
			.0003036	2491.9	2.1510@-06	.0531	7.228@-06
26.012	135.00	6.408					
			.0002870	2623.5	1.2086@-06	.0471	3.588@-06
25.864	175.00	6.853					
			.0002724	2751.8	1.3197@-06	.0411	3.405@-06
25.716	215.00	7.296					
			.0002594	2877.0	1.4771@-06	.0352	3.243@-06
25.568	255.00	7.737					
			.0002477	2999.2	1.5180@-06	.0292	2.752@-06
25.420	300.00	8.176					
			.0002359	3118.5	5.1691@-07	.0232	7.372@-07
25.273	460.00	8.611					

EQUILIBRIUM CONCENTRATION = 24.771 INITIAL SEED AREA = 1034

TABLE: 117 RUN NO.: R.E.31

TEMP: 70.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
29.708	.00000	1.952					
			.0004361	1714.9	6.0863@-06	.1265	5.452@-05
29.569	4.0000	2.393					
			.0003711	2009.2	1.9724@-06	.1212	1.687@-05
29.430	15.000	2.833					
			.0003249	2288.8	1.6605@-06	.1160	1.354@-05
29.291	27.000	3.273					
			.0002902	2555.7	1.7007@-06	.1107	1.319@-05
29.152	38.000	3.711					
			.0002631	2811.7	1.7869@-06	.1054	1.315@-05
29.012	48.000	4.148					
			.0002413	3058.1	1.5735@-06	.1001	1.097@-05
28.873	59.000	4.583					
			.0004320	3409.9	1.8767@-06	.0921	1.200@-05
28.594	77.000	5.453					
			.0001953	3751.0	1.6806@-06	.0842	9.764@-06
28.454	87.000	5.886					
			.0001841	3968.8	1.5420@-06	.0789	8.369@-06
28.315	98.000	6.318					
			.0001743	4181.0	1.4395@-06	.0736	7.264@-06
28.175	110.00	6.749					
			.0001657	4388.0	1.3655@-06	.0683	6.373@-06
28.035	123.00	7.178					
			.0001580	4590.4	9.6912@-07	.0629	4.157@-06
27.895	142.00	7.607					
			.0001510	4788.3	9.6471@-07	.0576	3.775@-06
27.756	162.00	8.034					
			.0001448	4982.2	1.7037@-06	.0523	6.032@-06
27.615	174.00	8.460					
			.0001391	5172.2	1.2190@-06	.0470	3.863@-06
27.475	192.00	8.886					
			.0001339	5358.7	1.3282@-06	.0416	3.719@-06
27.335	210.00	9.309					
			.0001290	5541.7	8.8452@-07	.0363	2.150@-06
27.195	240.00	9.732					
			.0001246	5721.5	7.5405@-07	.0310	1.557@-06
27.055	280.00	10.15					
			.0001203	5898.1	5.9008@-07	.0256	1.002@-06
26.914	340.00	10.57					

EQUILIBRIUM CONCENTRATION = 26.310 INITIAL SEED AREA = 1564

TABLE: 118 RUN NO.: R.P.E.10

TEMP: 30.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH P.E.PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
12.251	.00000	1.991					
			.0030836	1555.4	1.7010@-04	.4861	1.542@-03
11.241	1.0000	4.760					
			.0003028	2119.9	2.257@-06	.4133	1.682@-05
11.100	10.000	5.140					
			.0002860	2240.4	4.0190@-06	.3955	2.860@-05
10.959	15.000	5.518					
			.0002712	2358.0	8.0009@-06	.3777	5.424@-05
10.819	17.500	5.896					
			.0002581	2473.1	8.0103@-06	.3598	5.163@-05
10.678	20.000	6.274					
			.0004830	2640.4	2.0292@-05	.3331	1.207@-04
10.396	22.000	7.027					
			.0004447	2857.3	3.2348@-05	.2974	1.711@-04
10.114	23.300	7.778					
			.0008007	3166.4	1.3324@-04	.2438	5.719@-04
9.5487	24.000	9.275					
			.0003629	3465.8	1.0893@-04	.1902	3.629@-04
9.2658	24.500	10.02					
			.0005079	3702.8	1.5539@-04	.1454	3.907@-04
8.8409	25.150	11.13					
			.0004703	3978.1	1.7905@-04	.0916	2.767@-04
8.4154	26.000	12.23					
			.0002953	4200.6	1.4842@-04	.0467	1.136@-04
8.1314	27.300	12.97					
			.0001427	4330.8	1.8868@-04	.0198	5.945@-05
7.9892	28.500	13.33					

EQUILIBRIUM CONCENTRATION = 7.9040 INITIAL SEED AREA = 1052

TABLE: 119 RUN NO.:R.E.32

TEMP:60.0°C CELL: C STIRRER SPEED:2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
27.248	.00000	.9920					
			.0006434	911.32	1.7326@-05	.3493	3.217@-04
27.137	1.0000	1.337					
			.0006293	1161.8	1.1536@-05	.3431	2.098@-04
26.998	2.5000	1.768					
			.0005134	1419.5	8.2634@-06	.3362	1.467@-04
26.859	4.2500	2.198					
			.0008237	1769.2	7.9882@-06	.3258	1.373@-04
26.580	7.2500	3.057					
			.0003447	2099.7	6.9570@-06	.3155	1.149@-04
26.441	8.7500	3.484					
			.0003134	2304.3	6.4846@-06	.3086	1.045@-04
26.302	10.250	3.911					
			.0002882	2500.9	5.2417@-06	.3017	8.233@-05
26.162	12.000	4.337					
			.0002673	2690.3	5.8212@-06	.2947	8.911@-05
26.023	13.500	4.762					
			.0002498	2873.6	5.5840@-06	.2878	8.326@-05
25.883	15.000	5.186					
			.0004570	3136.8	3.9842@-06	.2774	5.713@-05
25.604	19.000	6.033					
			.0002103	3392.9	5.1059@-06	.2670	7.009@-05
25.464	20.500	6.454					
			.0002002	3557.2	5.0028@-06	.2601	6.672@-05
25.324	22.000	6.875					
			.0001912	3717.6	5.9049@-06	.2531	7.646@-05
25.184	23.250	7.295					
			.0001830	3874.6	4.1633@-06	.2462	5.230@-05
25.044	25.000	7.714					
			.0001757	4028.4	3.6076@-06	.2392	4.393@-05
24.904	27.000	8.132					
			.0001690	4179.2	3.5835@-06	.2323	4.226@-05
24.764	29.000	8.550					
			.0001629	4327.2	3.1731@-06	.2253	3.621@-05
24.623	31.250	8.967					
			.0001573	4472.5	3.1695@-06	.2184	3.496@-05
24.483	33.500	9.382					
			.0001521	4615.4	3.1743@-06	.2114	3.381@-05
24.343	35.750	9.797					
			.0001474	4755.9	3.1872@-06	.2044	3.275@-05
24.202	38.000	10.21					
			.0001429	4894.2	3.6093@-06	.1974	3.573@-05
24.061	40.000	10.62					
			.0001388	5030.5	2.6488@-06	.1905	2.523@-05
23.921	42.750	11.04					

TABLE :119(CONTD.) RUN NO. : R.E.32(CONTD.)

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT K($MIN^{-1}CM^{-2}$)	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
23.921	42.750	11.04					
			.0001349	5164.7	2.6795@-06	.1835	2.452@-05
23.780	45.500	11.45					
			.0001313	5297.1	3.3213@-06	.1765	2.919@-05
23.639	47.750	11.86					
			.0001279	5427.7	2.3378@-06	.1695	1.968@-05
23.498	51.000	12.27					
			.0001247	5556.5	1.9363@-06	.1625	1.558@-05
23.358	55.000	12.68					
			.0001216	5683.7	2.2619@-06	.1555	1.738@-05
23.217	58.500	13.09					
			.0001187	5809.2	1.9094@-06	.1485	1.397@-05
23.075	62.750	13.50					
			.0001160	5933.2	2.2248@-06	.1415	1.547@-05
22.934	66.500	13.90					
			.0001134	6055.8	1.7209@-06	.1345	1.134@-05
22.793	71.500	14.31					
			.0001110	6176.8	1.7809@-06	.1275	1.110@-05
22.652	76.500	14.71					
			.0001086	6296.6	1.6816@-06	.1205	9.876@-06
22.510	82.000	15.12					
			.0001064	6414.9	1.2056@-06	.1135	6.650@-06
22.369	90.000	15.52					
			.0001043	6532.0	1.6837@-06	.1065	8.690@-06
22.227	96.000	15.92					
			.0001022	6647.9	8.5069@-07	.0994	4.089@-06
22.086	108.50	16.33					
			.0001003	6762.5	1.1847@-06	.0924	5.277@-06
21.944	118.00	16.73					
			.0000984	6875.9	1.2619@-06	.0854	5.179@-06
21.802	127.50	17.13					
			.0000966	6988.2	1.0291@-06	.0783	3.864@-06
21.661	140.00	17.52					
			.0000948	7099.4	6.9605@-07	.0713	2.371@-06
21.519	160.00	17.92					
			.0000932	7209.4	7.6109@-07	.0643	2.329@-06
21.377	180.00	18.32					
			.0000915	7318.4	8.4277@-07	.0572	2.289@-06
21.235	200.00	18.72					
			.0001785	7479.5	7.1508@-07	.0466	1.566@-06
20.950	257.00	19.50					
			.0000869	7639.1	6.7675@-07	.0361	1.144@-06
20.808	295.00	19.90					

TABLE 119 (CONTD.) RUN NO. R.E.32 (CONTD.)

CONCN. GM/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
20.808	295.00	19.90					
			.0000855	7743.9	5.7491@-07	.0290	7.768@-07
20.666	350.00	20.29					
			.0000839	7847.5	3.7668@-07	.0219	3.812@-07
20.523	460.00	20.68					

EQUILIBRIUM CONCENTRATION = 20.153 INITIAL SEED AREA = 794.7

TABLE: 120 RUN NO.:R.E.33

TEMP:60.0°C CELL:C STIRRER SPEED:2000 R.P.M.

SEED: BATCHE PREPARED SIEVE FRACTION:89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
27.276	.00000	.4920					
			.0013932	532.22	1.6317@-05	.3538	3.096@-04
27.137	2.2500	.9236					
			.0027663	1085.9	8.6645@-06	.3365	1.581@-04
26.580	11.000	2.648					
			.0004556	1588.1	6.8372@-06	.3192	1.139@-04
26.441	13.000	3.075					
			.0004109	1757.0	1.0113@-05	.3123	1.644@-04
26.302	14.250	3.502					
			.0003755	1918.3	6.7699@-06	.3054	1.073@-04
26.162	16.000	3.927					
			.0003468	2073.2	6.7204@-06	.2984	1.038@-04
26.023	17.670	4.352					
			.0003228	2222.6	6.7873@-06	.2915	1.022@-04
25.883	19.250	4.776					
			.0003025	2367.1	6.8806@-06	.2845	1.008@-04
25.743	20.750	5.199					
			.0002850	2507.2	5.7105@-06	.2776	8.144@-05
25.604	22.500	5.621					
			.0002698	2643.4	6.4848@-06	.2706	8.993@-05
25.464	24.000	6.043					
			.0002564	2776.1	6.3412@-06	.2636	8.546@-05
25.324	25.500	6.464					
			.0004789	2968.3	6.1814@-06	.2532	7.981@-05
25.044	28.500	7.303					
			.0004401	3216.4	3.6262@-06	.2393	4.401@-05
24.764	33.500	8.140					
			.0002075	3396.7	2.7637@-06	.2288	3.192@-05
24.623	36.750	8.556					
			.0002002	3513.4	7.1695@-06	.2218	8.007@-05
24.483	38.000	8.972					
			.0001935	3628.1	5.9771@-06	.2148	6.449@-05
24.343	39.500	9.387					
			.0001873	3740.8	3.3681@-06	.2078	3.507@-05
24.202	42.170	9.801					
			.0001815	3851.6	2.7156@-06	.2008	2.725@-05
24.061	45.500	10.21					
			.0001762	3960.7	4.0516@-06	.1938	3.915@-05
23.921	47.750	10.63					
			.0001712	4068.2	5.2643@-06	.1868	4.891@-05
23.780	49.500	11.04					

TABLE : 120 (CONTD.) RUN NO. : R.E.33 (CONTD.)

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
23.780	49.500	11.04					
			.0001665	4174.1	2.8719@-06	.1798	2.561@-05
23.639	52.750	11.45					
			.0001621	4278.5	2.2306@-06	.1728	1.907@-05
23.498	57.000	11.86					
			.0001580	4381.5	3.8616@-06	.1658	3.159@-05
23.358	59.500	12.27					
			.0003048	4533.0	2.8505@-06	.1553	2.177@-05
23.075	66.500	13.08					
			.0001469	4682.6	1.7273@-06	.1448	1.224@-05
22.934	72.500	13.49					
			.0001436	4780.5	2.1350@-06	.1377	1.436@-05
22.793	77.500	13.90					
			.0001405	4877.1	1.8387@-06	.1307	1.171@-05
22.652	83.500	14.30					
			.0001376	4972.7	2.1794@-06	.1237	1.310@-05
22.510	88.750	14.71					
			.0001347	5067.2	1.9060@-06	.1166	1.078@-05
22.369	95.000	15.11					
			.0001320	5160.7	1.7793@-06	.1096	9.428@-06
22.227	102.00	15.51					
			.0001294	5253.1	1.0903@-06	.1026	5.390@-06
22.086	114.00	15.91					
			.0002515	5389.6	1.3574@-06	.0920	5.988@-06
21.802	135.00	16.71					
			.0001221	5524.6	1.1216@-06	.0814	4.362@-06
21.661	149.00	17.11					
			.0001199	5613.2	1.3025@-06	.0743	4.613@-06
21.519	162.00	17.51					
			.0002337	5744.2	1.0199@-06	.0637	3.075@-06
21.235	200.00	18.30					
			.0001137	5873.9	7.5629@-07	.0531	1.895@-06
21.093	230.00	18.70					
			.0001117	5959.0	7.3785@-07	.0461	1.596@-06
20.950	265.00	19.09					
			.0001098	6043.3	7.5316@-07	.0390	1.373@-06
20.808	305.00	19.48					
			.0001079	6126.6	6.6144@-07	.0319	9.811@-07
20.666	360.00	19.87					
			.0001050	6208.7	1.2182@-07	.0248	1.381@-07
20.523	740.00	20.26					

EQUILIBRIUM CONCENTRATION = 20.096 INITIAL SEED AREA = 394.2

TABLE: 121 RUN NO.:R.E.34

TEMP:60.0°C CELL:C STIRRER SPEED:2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION:89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
25.743	.00000	.9950					
			.0007747	940.45	5.2744@-06	.2812	7.747@-05
25.604	5.0000	1.423					
			.0005968	1214.6	2.7934@-06	.2742	3.979@-05
25.464	12.500	1.851					
			.0004927	1466.7	2.5446@-06	.2672	3.519@-05
25.324	19.500	2.277					
			.0004235	1701.9	2.1027@-06	.2603	2.823@-05
25.184	27.000	2.703					
			.0003738	1923.8	3.5859@-06	.2533	4.672@-05
25.044	31.000	3.127					
			.0003361	2134.8	2.2167@-06	.2463	2.800@-05
24.904	37.000	3.551					
			.0003063	2336.6	1.7876@-06	.2393	2.188@-05
24.764	44.000	3.974					
			.0002822	2530.5	1.9848@-06	.2323	2.352@-05
24.623	50.000	4.396					
			.0002623	2717.6	2.5421@-06	.2253	2.914@-05
24.483	54.500	4.817					
			.0002454	2898.7	2.4611@-06	.2183	2.726@-05
24.343	59.000	5.238					
			.0002308	3074.5	1.7990@-06	.2113	1.924@-05
24.202	65.000	5.657					
			.0002182	3245.4	2.1165@-06	.2043	2.182@-05
24.061	70.000	6.076					
			.0002072	3411.9	2.0859@-06	.1972	2.072@-05
23.921	75.000	6.493					
			.0001973	3574.4	1.8779@-06	.1902	1.794@-05
23.780	80.500	6.910					
			.0001885	3733.2	1.5805@-06	.1832	1.450@-05
23.639	87.000	7.326					
			.0003544	3964.2	1.3705@-06	.1727	1.181@-05
23.358	102.00	8.156					
			.0001669	4190.5	1.7267@-06	.1621	1.391@-05
23.217	108.00	8.569					
			.0001609	4337.0	1.1021@-06	.1551	8.468@-06
23.075	117.50	8.982					

TABLE 121 (CONTD.) RUN NO.: RE.34 (CONTD.)

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
23.075	117.50	8.982					
			.0001554	4480.9	1.4162@-06	.1480	1.036@-05
22.934	125.00	9.393					
			.0001503	4622.4	9.8337@-07	.1410	6.832@-06
22.793	136.00	9.804					
			.0001456	4761.5	1.1060@-06	.1339	7.279@-06
22.652	146.00	10.21					
			.0001412	4898.4	1.0322@-06	.1269	6.418@-06
22.510	157.00	10.62					
			.0001371	5033.3	8.3626@-07	.1198	4.896@-06
22.369	171.00	11.03					
			.0001333	5166.1	8.6626@-07	.1128	4.759@-06
22.227	185.00	11.44					
			.0001297	5297.1	1.0521@-06	.1057	5.406@-06
22.086	197.00	11.84					
			.0001263	5426.3	7.7741@-07	.0986	3.716@-06
21.944	214.00	12.25					
			.0002433	5616.1	6.6718@-07	.0880	2.829@-06
21.661	257.00	13.06					
			.0001172	5803.5	6.8585@-07	.0774	2.547@-06
21.519	280.00	13.46					
			.0001144	5925.9	6.3022@-07	.0703	2.119@-06
21.377	307.00	13.86					
			.0001118	6046.8	5.6237@-07	.0632	1.694@-06
21.235	340.00	14.26					
			.0001093	6166.2	5.8610@-07	.0561	1.561@-06
21.093	375.00	14.66					
			.0001069	6284.1	5.7654@-07	.0490	1.336@-06
20.950	415.00	15.05					
			.0001045	6400.5	3.7863@-07	.0420	7.466@-07
20.808	485.00	15.45					

EQUILIBRIUM CONCENTRATION = 20.039 INITIAL SEED AREA = 797.1

TABLE: 122 RUN NO.:R.E.35

TEMP:60.0°C CELL:C STIRRER SPEED:2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION:89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
25.687	.00000	.4900					
			.0009165	477.65	4.8172@-06	.2798	7.050@-05
25.604	6.5000	.7469					
			.0010601	686.86	2.3901@-06	.2742	3.420@-05
25.464	22.000	1.174					
			.0007860	920.75	2.9866@-06	.2672	4.137@-05
25.324	31.500	1.601					
			.0006378	1130.7	3.1649@-06	.2603	4.252@-05
25.184	39.000	2.026					
			.0010210	1412.1	3.6044@-06	.2498	4.641@-05
24.904	50.000	2.876					
			.0008162	1756.4	2.4145@-06	.2358	2.915@-05
24.623	64.000	3.722					
			.0003571	1996.1	3.1149@-06	.2253	3.571@-05
24.483	69.000	4.143					
			.0003314	2146.5	2.9913@-06	.2183	3.314@-05
24.343	74.000	4.563					
			.0003097	2291.7	2.8962@-06	.2113	3.097@-05
24.202	79.000	4.983					
			.0002912	2432.4	2.5671@-06	.2043	2.647@-05
24.061	84.500	5.401					
			.0005368	2634.9	2.8965@-06	.1937	2.825@-05
23.780	94.000	6.237					
			.0004865	2894.8	2.2528@-06	.1797	2.027@-05
23.498	106.00	7.069					
			.0004462	3143.0	2.2537@-06	.1656	1.859@-05
23.217	118.00	7.898					
			.0002100	3323.2	2.4844@-06	.1551	1.909@-05
23.075	123.50	8.310					
			.0002025	3439.8	1.8449@-06	.1480	1.350@-05
22.934	131.00	8.722					
			.0001955	3554.2	1.2789@-06	.1410	8.888@-06
22.793	142.00	9.132					
			.0003727	3721.6	1.4548@-06	.1304	9.317@-06
22.510	162.00	9.952					
			.0001776	3886.2	1.1664@-06	.1198	6.832@-06
22.369	175.00	10.36					

TABLE :122 (CONTD.) RUN NO. :R.E.35 (CONTD.)

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
2.369	175.00	10.36					
			.0003404	4045.7	1.2314@-06	.1092	6.547@-06
22.086	201.00	11.17					
			.0003222	4253.7	8.9822@-07	.0951	4.130@-06
21.802	240.00	11.98					
			.0003060	4455.9	9.8392@-07	.0809	3.825@-06
21.519	280.00	12.79					
			.0001473	4604.4	7.2999@-07	.0703	2.455@-06
21.377	310.00	13.19					
			.0001438	4701.4	5.9672@-07	.0632	1.798@-06
21.235	350.00	13.59					
			.0001405	4797.1	5.8596@-07	.0561	1.561@-06
21.093	395.00	13.99					
			.0001374	4891.6	6.5837@-07	.0490	1.527@-06
20.950	440.00	14.38					

EQUILIBRIUM CONCENTRATION = 20.039 INITIAL SEED AREA = 392.5

TABLE: 123 RUN NO.: R.E.36

TEMP: 60.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
24.258	.00000	.9920					
			.0003375	853.31	1.4189@-06	.2109	1.534@-05
24.202	11.000	1.162					
			.0006872	1048.7	9.8552@-07	.2060	1.041@-05
24.061	44.000	1.586					
			.0005472	1311.1	1.0778@-06	.1990	1.094@-05
23.921	69.000	2.010					
			.0004604	1554.0	2.6199@-06	.1919	2.558@-05
23.780	78.000	2.432					
			.0004003	1781.7	7.9111@-07	.1849	7.412@-06
23.639	105.00	2.853					
			.0003562	1997.2	1.5246@-06	.1779	1.370@-05
23.498	118.00	3.273					
			.0003221	2202.5	1.1702@-06	.1708	1.007@-05
23.358	134.00	3.692					
			.0002950	2399.3	1.2812@-06	.1638	1.054@-05
23.217	148.00	4.111					
			.0002728	2588.7	1.2415@-06	.1567	9.742@-06
23.075	162.00	4.528+					
			.0002541	2771.7	1.0630@-06	.1497	7.942@-06
22.934	178.00	4.944					
			.0002383	2948.9	1.1191@-06	.1426	7.943@-06
22.793	193.00	5.359					
			.0002246	3121.0	7.2590@-07	.1356	4.882@-06
22.652	216.00	5.773					
			.0004150	3369.1	8.6181@-07	.1250	5.321@-06
22.369	255.00	6.599					
			.0001926	3611.1	5.9081@-07	.1144	3.320@-06
22.227	284.00	7.010					
			.0001841	3766.7	9.2204@-07	.1073	4.846@-06
22.086	303.00	7.420					
			.0003462	3992.9	5.4874@-07	.0967	2.584@-06
21.802	370.00	8.237					
			.0001631	4214.6	5.7526@-07	.0860	2.399@-06
21.661	404.00	8.643					
			.0001572	4358.0	4.4841@-07	.0789	1.709@-06
21.519	450.00	9.048					
			.0001517	4498.7	3.6615@-07	.0718	1.264@-06
21.377	510.00	9.452					
			.0001467	4636.8	4.7335@-07	.0648	1.467@-06
21.235	560.00	9.854					

EQUILIBRIUM CONCENTRATION = 20.010 INITIAL SEED AREA = 794.7

TABLE: 124 RUN NO.: R.E.37

TEMP 60.0°C CELL: C STIRRER SPEED 2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
24.343	.00000	.4900					
			.0013799	528.52	1.0753@-06	.2130	1.190@-05
24.202	58.000	.9147					
			.0009212	781.23	1.3643@-06	.2060	1.439@-05
24.061	90.000	1.338					
			.0007142	1002.8	1.7616@-06	.1990	1.785@-05
23.921	110.00	1.761					
			.0005928	1204.1	1.5217@-06	.1919	1.482@-05
23.780	130.00	2.182					
			.0009659	1476.2	1.7532@-06	.1814	1.610@-05
23.498	160.00	3.022					
			.0004086	1733.0	1.8305@-06	.1708	1.572@-05
23.358	173.00	3.441					
			.0003734	1892.0	1.7497@-06	.1638	1.436@-05
23.217	186.00	3.858					
			.0006661	2116.9	1.4508@-06	.1532	1.110@-05
22.934	216.00	4.691					
			.0005840	2402.1	1.0317@-06	.1291	7.121@-06
22.652	257.00	5.519					
			.0002677	2606.2	1.2414@-06	.1285	7.874@-06
22.510	274.00	5.932					
			.0007295	2861.1	9.1610@-07	.1143	5.137@-06
22.086	345.00	7.164					
			.0002219	3110.1	7.5766@-07	.1002	3.699@-06
21.944	375.00	7.572					
			.0002132	3228.9	7.8573@-07	.0931	3.553@-06
21.802	405.00	7.979					
			.0002052	3345.4	8.2134@-07	.0860	3.419@-06
21.661	435.00	8.385					
			.0001978	3459.6	6.4957@-07	.0789	2.472@-06
21.519	475.00	8.789					
			.0001909	3571.7	6.9176@-07	.0718	2.387@-06
21.377	515.00	9.192					
			.0001846	3681.7	5.9614@-07	.0648	1.846@-06
21.235	565.00	9.594					

EQUILIBRIUM CONCENTRATION = 20.010 INITIAL SEED AREA = 392.5

TABLE: 125 RUN NO.: R.C.F.1

TEMP: 60.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
27.276	.00000	1.992					
			.0004196	1743.9	1.1341@-05	.3481	2.098@-04
27.137	1.0000	2.424					
			.0006753	2166.9	9.4147@-06	.3378	1.688@-04
26.859	3.0000	3.285					
			.0002826	2570.5	8.1927@-06	.3274	1.413@-04
26.719	4.0000	3.715					
			.0002568	2822.9	6.5171@-06	.3205	1.097@-04
26.580	5.1700	4.143					
			.0002359	3066.1	4.7857@-06	.3136	7.865@-05
26.441	6.6700	4.570					
			.0002187	3301.2	4.3170@-06	.3067	6.920@-05
26.302	8.2500	4.997					
			.0003963	3638.7	4.1628@-06	.2964	6.434@-05
26.023	11.330	5.849					
			.0003527	4071.1	3.2789@-06	.2825	4.806@-05
25.743	15.000	6.697					
			.0001631	4383.2	3.3174@-06	.2721	4.659@-05
25.604	16.750	7.119					
			.0001556	4584.0	2.2089@-06	.2652	3.015@-05
25.464	19.330	7.541					
			.0004301	4970.5	2.4362@-06	.2514	3.144@-05
25.044	26.170	8.803					
			.0001323	5349.6	2.1184@-06	.2375	2.563@-05
24.904	28.750	9.221					
			.0001277	5532.2	2.2510@-06	.2305	2.639@-05
24.764	31.170	9.639					
			.0001235	5711.7	1.4211@-06	.2236	1.612@-05
24.623	35.000	10.06					
			.0002355	5974.7	1.8213@-06	.2132	1.962@-05
24.343	41.000	10.89					
			.0001124	6233.7	1.5741@-06	.2027	1.606@-05
24.202	44.500	11.30					
			.0001092	6402.5	1.5880@-06	.1958	1.560@-05
24.061	48.000	11.71					
			.0001062	6568.9	1.1240@-06	.1888	1.062@-05
23.921	53.000	12.13					
			.0001034	6733.1	1.0357@-06	.1818	9.400@-06
23.780	58.500	12.54					

TABLE : 12 (CONTD.) RUN NO. : R.C.F1 (CONTD.)

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
23.780	58.500	12.54					
			.0002956	7053.6	1.4166@-06	.1679	1.182@-05
23.358	71.000	13.77					
			.0000937	7370.1	1.1736@-06	.1539	8.923@-06
23.217	76.250	14.18					
			.0001813	7600.2	1.0929@-06	.1434	7.717@-06
22.934	88.000	14.99					
			.0000877	7827.7	1.1213@-06	.1329	7.306@-06
22.793	94.000	15.40					
			.0000859	7976.8	9.9609@-07	.1259	6.133@-06
22.652	101.00	15.80					
			.0000841	8124.3	1.2089@-06	.1189	7.011@-06
22.510	107.00	16.21					
			.0002431	8413.6	8.5425@-07	.1049	4.341@-06
22.086	135.00	17.42					
			.0000779	8700.0	8.0771@-07	.0909	3.541@-06
21.944	146.00	17.82					
			.0000765	8840.1	9.4820@-07	.0838	3.825@-06
21.802	156.00	18.21					
			.0001490	9047.4	7.0879@-07	.0733	2.483@-06
21.519	186.00	19.01					
			.0001438	9319.9	4.7431@-07	.0592	1.332@-06
21.235	240.00	19.80					
			.0000701	9521.3	6.0883@-07	.0487	1.403@-06
21.093	265.00	20.20					
			.0000690	9653.7	5.0207@-07	.0416	9.355@-07
20.950	300.00	20.59					
			.0000677	9784.7	4.6452@-07	.0346	7.526@-07
20.808	345.00	20.98					
			.0000665	9914.3	2.7347@-07	.0275	3.501@-07
20.666	440.00	21.37					

EQUILIBRIUM CONCENTRATION = 20.181 INITIAL SEED AREA = 1596

TABLE: 126 RUN NO.: R.C.18

TEMP: 60.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105 μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
27.248	.00000	1.945					
			.0003487	1677.1	1.2604@-05	.3474	2.325@-04
27.137	.75000	2.290					
			.0003766	1937.7	8.3347@-06	.3412	1.506@-04
26.998	2.0000	2.721					
			.0003286	2214.3	7.4478@-06	.3343	1.315@-04
26.859	3.2500	3.151					
			.0002928	2478.3	5.6652@-06	.3274	9.762@-05
26.719	4.7500	3.579					
			.0002650	2731.4	4.5031@-06	.3205	7.572@-05
26.580	6.5000	4.007					
			.0002427	2974.9	3.2882@-06	.3136	5.393@-05
26.441	8.7500	4.433					
			.0002243	3210.0	2.5508@-06	.3067	4.078@-05
26.302	11.500	4.859					
			.0002089	3437.6	3.8313@-06	.2998	5.969@-05
26.162	13.250	5.283					
			.0001958	3658.5	4.3014@-06	.2929	6.527@-05
26.023	14.750	5.707					
			.0001844	3873.3	2.2709@-06	.2860	3.353@-05
25.883	17.500	6.129					
			.0003406	4183.6	2.8260@-06	.2756	4.007@-05
25.604	21.750	6.971					
			.0001580	4486.6	1.9474@-06	.2652	2.641@-05
25.464	24.740	7.389					
			.0004351	4869.4	1.9417@-06	.2514	2.484@-05
25.044	33.500	8.641					
			.0001335	5244.5	1.1737@-06	.2375	1.406@-05
24.904	38.250	9.055					
			.0001287	5424.7	1.3071@-06	.2305	1.514@-05
24.764	42.500	9.467					
			.0001243	5601.7	1.7077@-06	.2236	1.913@-05
24.623	45.750	9.879					
			.0001202	5775.7	1.4822@-06	.2166	1.603@-05
24.483	49.500	10.29					
			.0001164	5946.8	1.8602@-06	.2097	1.940@-05
24.343	52.500	10.70					

TABLE : 126 (CONTD.) RUN NO. : R.C.18 (CONTD.)

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
24.343	52.500	10.70					
			.0002222	6197.3	9.8118@-07	.1992	9.660@-06
24.061	64.000	11.51					
			.0005979	6832.7	1.2394@-06	.1713	1.031@-05
23.217	93.000	13.92					
			.0000908	7376.7	7.1689@-07	.1469	5.043@-06
23.075	102.00	14.32					
			.0000887	7524.4	1.1075@-06	.1399	7.388@-06
22.934	108.00	14.71					
			.0004123	7949.1	7.1789@-07	.1189	3.964@-06
22.227	160.00	16.65					
			.0001526	8434.6	7.6811@-07	.0944	3.317@-06
21.944	183.00	17.41					
			.0000738	8634.5	5.1094@-07	.0838	1.942@-06
21.802	202.00	17.79					
			.0000721	8765.0	5.8026@-07	.0768	2.004@-06
21.661	220.00	18.16					
			.0000704	8893.4	4.9297@-07	.0698	1.531@-06
21.519	243.00	18.53					
			.0000687	9019.6	4.6079@-07	.0628	1.271@-06
21.377	270.00	18.89					
			.0002478	9311.8	3.6489@-07	.0451	6.520@-07
20.808	460.00	20.26					

EQUILIBRIUM CONCENTRATION = 20.181 INITIAL SEED AREA = 1558

TABLE: 127 RUN NO.: R.P.E.11

TEMP: 60.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH P.E. PREPARED SIEVE FRACTION: 89-105 μ

CONCN. ΔM/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
24.012	.00000	1.972					
			.0054592	2069.6	4.1068@-04	.2324	4.706@-03
21.919	.58000	8.292					
			.0012552	3427.8	7.2011@-05	.1529	5.021@-04
21.047	1.8300	10.83					
			.0003654	3861.1	4.9627@-05	.1217	2.727@-04
20.756	2.5000	11.66					
			.0003454	4063.8	3.0985@-05	.1061	1.476@-04
20.465	3.6700	12.49					
			.0009420	4449.8	1.3115@-05	.0749	4.251@-05
19.593	14.750	14.97					
			.0005594	4915.2	5.5336@-06	.0359	8.166@-06
19.012	49.000	16.59					
			.0001324	5137.9	3.6345@-06	.0164	2.546@-06
18.866	75.000	17.00					

EQUILIBRIUM CONCENTRATION = 18.634 INITIAL SEED AREA = 1042

TABLE: 128 RUN NO.: R.P.E.12

TEMP: 60.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH P.E. PREPARED SIEVE FRACTION: 89-105 μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
21.890	.00000	1.962					
			.0005277	1106.5	1.0956@-05	.1716	8.795@-05
21.773	3.0000	2.307					
			.0005778	1260.0	1.7100@-06	.1646	1.313@-05
21.628	25.000	2.736					
			.0005097	1423.6	3.4954@-06	.1568	2.549@-05
21.483	35.000	3.165					
			.0004578	1580.3	3.4881@-06	.1490	2.410@-05
21.337	44.500	3.592					
			.0004168	1731.0	3.0405@-06	.1412	1.985@-05
21.192	55.000	4.019					
			.0003834	1876.3	3.4640@-06	.1334	2.130@-05
21.047	64.000	4.444					
			.0003557	2016.9	2.8005@-06	.1256	1.617@-05
20.901	75.000	4.868					
			.0003322	2153.2	3.0768@-06	.1178	1.661@-05
20.756	85.000	5.290					
			.0003121	2285.6	2.3879@-06	.1100	1.200@-05
20.611	98.000	5.712					
			.0002945	2414.5	2.1087@-06	.1022	9.817@-06
20.465	113.00	6.132					
			.0002791	2540.2	1.9149@-06	.0944	8.209@-06
20.320	130.00	6.551					
			.0005191	2722.3	1.8788@-06	.0827	7.016@-06
20.029	167.00	7.386					
			.0002422	2900.5	1.8963@-06	.0710	6.055@-06
19.884	187.00	7.801					
			.0002322	3015.4	1.5183@-06	.0632	4.300@-06
19.739	214.00	8.215					
			.0002231	3128.1	1.3265@-06	.0554	3.280@-06
19.593	248.00	8.627					
			.0002147	3238.6	1.6911@-06	.0476	3.579@-06
19.448	278.00	9.038					
			.0002070	3347.0	1.4693@-06	.0398	2.587@-06
19.303	318.00	9.448					
			.0001998	3453.5	1.2905@-06	.0320	1.816@-06
19.157	373.00	9.855					

EQUILIBRIUM CONCENTRATION = 18.634 INITIAL SEED AREA = 1037

TABLE: 129 RUN NO.: R.P.E.14

TEMP: 60.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH P.E.PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
21.890	.00000	.9650					
			.0005359	544.76	8.2690@-06	.1732	6.699@-05
21.832	4.0000	1.137					
			.0004740	613.24	3.1168@-07	.1700	2.469@-06
21.773	100.00	1.309					
			.0010006	725.61	1.1263@-06	.1646	8.626@-06
21.628	158.00	1.737					
			.0015294	946.11	1.1128@-06	.1529	7.884@-06
21.337	255.00	2.588					
			.0006214	1152.6	1.0654@-06	.1412	6.904@-06
21.192	300.00	3.011					
			.0010663	1338.6	1.5019@-06	.1295	8.886@-06
20.901	360.00	3.854					
			.0004672	1516.8	1.4559@-06	.1178	7.786@-06
20.756	390.00	4.272					
			.0004335	1628.8	1.4520@-06	.1100	7.226@-06
20.611	420.00	4.690					
			.0004051	1737.1	1.2562@-06	.1022	5.787@-06
20.465	455.00	5.105					
			.0003807	1842.0	1.4964@-06	.0944	6.344@-06
20.320	485.00	5.520					
			.0003594	1944.0	1.4492@-06	.0866	5.616@-06
20.175	517.00	5.933					
			.0003409	2043.2	1.8653@-06	.0788	6.555@-06
20.029	543.00	6.344					
			.0001963	2121.0	9.5090@-07	.0725	3.067@-06
19.942	575.00	6.590					

EQUILIBRIUM CONCENTRATION = 18.634 INITIAL SEED AREA = 509.8

TABLE: 130 RUN NO.: R.P.E.16

TEMP: 50.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH P.E. PREPARED SIEVE FRACTION: 89-105 μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
17.545	.00000	1.969					
			.0007244	1138.1	1.2096@-06	.2369	9.789@-06
17.374	37.000	2.455					
			.0005212	1313.7	1.3565@-06	.2258	1.042@-05
17.232	62.000	2.860					
			.0008887	1537.9	8.7586@-07	.2107	6.258@-06
16.948	133.00	3.666					
			.0003874	1753.6	1.1753@-06	.1956	7.747@-06
16.806	158.00	4.067					
			.0003586	1890.2	1.3073@-06	.1855	8.149@-06
16.664	180.00	4.468					
			.0003344	2022.6	1.5800@-06	.1754	9.288@-06
16.521	198.00	4.867					
			.0006100	2213.2	1.3572@-06	.1603	7.262@-06
16.236	240.00	5.665					
			.0002800	2399.0	1.6132@-06	.1451	7.779@-06
16.093	258.00	6.062					
			.0002662	2518.3	2.4794@-06	.1350	1.109@-05
15.951	270.00	6.458					
			.0002539	2635.1	2.5631@-06	.1249	1.058@-05
15.808	282.00	6.853					
			.0002429	2749.4	2.4693@-06	.1148	9.342@-06
15.665	295.00	7.248					
			.0002329	2861.5	2.2568@-06	.1047	7.763@-06
15.522	310.00	7.642					
			.0002238	2971.4	2.2576@-06	.0945	6.994@-06
15.379	326.00	8.035					
			.0002155	3079.4	2.1709@-06	.0844	5.986@-06
15.236	344.00	8.427					
			.0002079	3185.5	2.6861@-06	.0742	6.496@-06
15.092	360.00	8.818					
			.0002009	3289.8	3.4472@-06	.0641	7.174@-06
14.949	374.00	9.209					
			.0001944	3392.5	2.9314@-06	.0539	5.115@-06
14.806	393.00	9.599					
			.0001884	3493.5	4.4524@-06	.0438	6.279@-06
14.662	408.00	9.988					
			.0002543	3612.6	2.2296@-06	.0316	2.231@-06
14.461	465.00	10.53					

EQUILIBRIUM CONCENTRATION = 14.116 INITIAL SEED AREA = 1040

TABLE: 131 RUN NO.: R.P.E.17

TEMP: 40.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH P.E. PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
13.613	.00000	1.983					
			.0002462	1079.8	2.2093@-06	.2804	1.539@-05
13.557	8.0000	2.140					
			.0003313	1157.5	1.0262@-07	.2737	6.706@-07
13.471	255.00	2.367					
			.0003872	1259.2	1.1294@-07	.2643	6.794@-07
13.358	540.00	2.655					
			.0038345	2054.5	9.3195@-07	.1634	2.739@-06
11.330	1240.0	7.186					
			.0001845	2837.5	5.2342@-06	.0610	6.151@-06
11.186	1255.0	7.496					
			.0001782	2924.5	5.7792@-06	.0475	5.240@-06
11.042	1272.0	7.803					
			.0001717	3009.8	4.0786@-06	.0339	2.602@-06
10.898	1305.0	8.109					

EQUILIBRIUM CONCENTRATION = 10.610 INITIAL SEED AREA = 1048

TABLE: 132 RUN NO.: R.P.E.18

TEMP: 75.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH P.E.PREPARED SIEVE FRACTION: 89-105 μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
31.578	.00000	1.919					
			.0010642	1157.9	3.8169@-05	.1071	3.186@-04
31.353	1.6700	2.645					
			.0005861	1392.0	1.3305@-05	.1006	1.036@-04
31.204	4.5000	3.126					
			.0009866	1650.0	6.2724@-06	.0927	4.485@-05
30.904	15.500	4.087					
			.0004257	1897.3	5.0415@-06	.0848	3.275@-05
30.754	22.000	4.564					
			.0003922	2052.8	4.9683@-06	.0795	3.017@-05
30.605	28.500	5.040					
			.0003643	2203.1	3.7912@-06	.0742	2.143@-05
30.455	37.000	5.514					
			.0003405	2348.7	3.8277@-06	.0689	2.003@-05
30.305	45.500	5.986					
			.0003201	2490.2	2.8891@-06	.0637	1.392@-05
30.156	57.000	6.457					
			.0003024	2627.8	3.6130@-06	.0584	1.591@-05
30.006	66.500	6.927					
			.0002866	2761.9	2.1754@-06	.0532	8.686@-06
29.856	83.000	7.395					
			.0002727	2892.7	2.3777@-06	.0479	8.521@-06
29.707	99.000	7.861					
			.0002602	3020.5	2.5587@-06	.0426	8.131@-06
29.557	115.00	8.325					
			.0002485	3145.4	7.8713@-07	.0374	2.180@-06
29.408	172.00	8.787					

EQUILIBRIUM CONCENTRATION = 28.421 INITIAL SEED AREA = 1014

TABLE: 133 RUN NO.: R.P.E.19

TEMP: 40.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH P.E.PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
13.585	.00000	1.987					
			.0014704	1262.7	1.6720@-07	.2616	1.081@-06
13.187	680.00	3.077					
			.0004237	1545.9	4.0819@-07	.2362	2.354@-06
13.045	770.00	3.464					
			.0003878	1684.3	5.1112@-07	.2227	2.77@-06
12.903	840.00	3.850					
			.0003581	1818.0	3.5299@-07	.2093	1.791@-06
12.760	940.00	4.235					
			.0003335	1947.6	5.8729@-07	.1959	2.779@-06
12.617	1000.0	4.619					
			.0006077	2134.2	1.3070@-06	.1757	5.525@-06
12.332	1055.0	5.384					
			.0002788	2316.1	1.8706@-06	.1555	6.969@-06
12.189	1075.0	5.766					
			.0002649	2433.0	2.6017@-06	.1421	8.830@-06
12.046	1090.0	6.147					
			.0002526	2547.3	4.1219@-06	.1286	1.263@-05
11.903	1100.0	6.527					
			.0004736	2713.8	7.1029@-06	.1083	1.822@-05
11.616	1113.0	7.287					
			.0002225	2877.2	8.9085@-06	.0881	1.854@-05
11.473	1119.0	7.665					
			.0002143	2983.0	6.7775@-06	.0745	1.190@-05
11.330	1128.0	8.043					
			.0002067	3087.1	4.8110@-06	.0610	6.890@-06
11.186	1143.0	8.420					
			.0001997	3189.5	5.6303@-06	.0475	6.241@-06
11.042	1159.0	8.796					
			.0001932	3290.2	3.0031@-06	.0339	2.356@-06
10.898	1200.0	9.172					

EQUILIBRIUM CONCENTRATION = 10.610 INITIAL SEED AREA = 1050

TABLE: 134 RUN NO.: R.P.E.20

TEMP: 70.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH P.E.PREPARED SIEVE FRACTION: 89-105 μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
28.587	.00000	1.950					
			.0009541	1160.3	7.1917@-05	.1499	7.120@-04
28.380	.67000	2.602					
			.0005705	1377.9	3.0432@-05	.1427	2.853@-04
28.232	1.6700	3.067					
			.0005059	1548.9	1.5439@-05	.1367	1.382@-04
28.083	3.5000	3.529					
			.0004561	1712.6	8.9057@-06	.1307	7.602@-05
27.935	6.5000	3.991					
			.0004164	1869.9	7.8885@-06	.1248	6.407@-05
27.787	9.7500	4.451					
			.0003840	2021.7	7.1151@-06	.1188	5.486@-05
27.639	13.250	4.910					
			.0003569	2168.6	6.1109@-06	.1128	4.461@-05
27.491	17.250	5.367					
			.0003339	2310.9	5.3817@-06	.1068	3.710@-05
27.343	21.750	5.823					
			.0003141	2449.3	4.8401@-06	.1008	3.141@-05
27.195	26.750	6.277					
			.0002968	2584.0	4.8761@-06	.0949	2.968@-05
27.047	31.750	6.731					
			.0002816	2715.3	4.7157@-06	.0889	2.682@-05
26.899	37.000	7.183					
			.0002680	2843.6	2.8157@-06	.0829	1.489@-05
26.751	46.000	7.633					
			.0005014	3029.7	2.9671@-06	.0740	1.393@-05
26.455	64.000	8.531					
			.0002350	3212.0	2.3841@-06	.0650	9.792@-06
26.307	76.000	8.977					
			.0002259	3329.8	2.6423@-06	.0590	9.824@-06
26.159	87.500	9.421					
			.0002177	3445.4	2.8409@-06	.0531	9.463@-06
26.012	99.000	9.864					
			.0002099	3558.8	1.5496@-06	.0471	4.563@-06
25.864	122.00	10.31					
			.0002028	3670.3	2.4737@-06	.0411	6.339@-06
25.716	138.00	10.75					
			.0003852	3832.9	7.4091@-07	.0322	1.459@-06
25.420	270.00	11.62					
			.0000740	3961.3	5.3486@-07	.0250	8.226@-07
25.361	315.00	11.79					

EQUILIBRIUM CONCENTRATION = 24.771 INITIAL SEED AREA = 1030

TABLE: 135 RUN NO.: R.E.38

TEMP: 50.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105 μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
19.307	.00000	1.973					
			.0002497	1665.8	5.5595@-07	.2753	5.675@-06
19.222	22.000	2.219					
			.0003669	1887.2	4.5127@-07	.2678	4.474@-06
19.081	63.000	2.627					
			.0003202	2153.2	2.1830@-07	.2585	2.079@-06
18.939	140.00	3.035					
			.0002855	2407.3	3.4678@-07	.2491	3.172@-06
18.798	185.00	3.440					
			.0002585	2650.9	3.6823@-07	.2398	3.231@-06
18.657	225.00	3.845					
			.0002368	2885.5	4.0248@-07	.2304	3.383@-06
18.515	260.00	4.249					
			.0004234	3220.5	5.9800@-07	.2163	4.705@-06
18.232	305.00	5.054					
			.0001912	3545.2	4.0852@-07	.2023	2.988@-06
18.090	337.00	5.455					
			.0001801	3752.5	3.3219@-07	.1929	2.309@-06
17.949	376.00	5.854					
			.0001705	3954.5	5.3862@-07	.1835	3.552@-06
17.807	400.00	6.253					
			.0001619	4151.6	3.2453@-07	.1742	2.024@-06
17.665	440.00	6.650					
			.0001543	4344.1	2.6235@-07	.1648	1.543@-06
17.524	490.00	7.046					
			.0001474	4532.5	3.3343@-07	.1554	1.843@-06
17.382	530.00	7.441					
			.0002770	4806.4	2.7710@-07	.1413	1.385@-06
17.098	630.00	8.227					
			.0001305	5075.0	2.9137@-07	.1272	1.305@-06
16.956	680.00	8.619					
			.0008971	5811.8	2.2036@-07	.0848	6.021@-07
15.818	1425.0	11.69					
			.0000972	6535.9	1.2405@-07	.0425	1.767@-07
15.676	1700.0	12.07					

EQUILIBRIUM CONCENTRATION = 15.106 INITIAL SEED AREA = 1581

TABLE: 136 RUN NO.: R.P.E.F.2

TEMP: 70.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH P.E. PREPARED SIEVE FRACTION: 89-105 μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
28.602	.00000	1.944					
			.0010177	1166.1	1.0254@-04	.1502	1.018@-03
28.380	.50000	2.643					
			.0005646	1392.2	4.0158@-05	.1427	3.764@-04
28.232	1.2500	3.107					
			.0005015	1562.4	2.8008@-05	.1367	2.507@-04
28.083	2.2500	3.570					
			.0004527	1725.4	1.7679@-05	.1307	1.509@-04
27.935	3.7500	4.031					
			.0004138	1882.1	1.4555@-05	.1248	1.182@-04
27.787	5.5000	4.491					
			.0003818	2033.4	1.1005@-05	.1188	8.485@-05
27.639	7.7500	4.950					
			.0003551	2179.7	9.7274@-06	.1128	7.103@-05
27.491	10.250	5.408					
			.0003324	2321.6	8.7658@-06	.1068	6.044@-05
27.343	13.000	5.864					
			.0006094	2526.1	8.4181@-06	.0979	5.299@-05
27.047	18.750	6.773					
			.0002807	2725.3	7.0477@-06	.0889	4.010@-05
26.899	22.250	7.225					
			.0002673	2853.2	4.5918@-06	.0829	2.430@-05
26.751	27.750	7.676					
			.0002553	2978.3	4.5338@-06	.0770	2.220@-05
26.603	33.500	8.125					
			.0002445	3100.7	4.5241@-06	.0710	2.037@-05
26.455	39.500	8.573					
			.0002346	3220.7	3.8043@-06	.0650	1.564@-05
26.307	47.000	9.020					
			.0002256	3338.3	3.3677@-06	.0590	1.254@-05
26.159	56.000	9.465					
			.0004276	3510.0	3.0313@-06	.0501	9.503@-06
25.864	78.500	10.35					
			.0002027	3678.7	2.4680@-06	.0411	6.335@-06
25.716	94.500	10.79					
			.0001962	3788.2	3.0946@-06	.0352	6.765@-06
25.568	109.00	11.23					
			.0001147	3874.6	1.1264@-06	.0304	2.123@-06
25.480	136.00	11.49					

EQUILIBRIUM CONCENTRATION = 24.771 INITIAL SEED AREA = 1027

TABLE: 137 RUN NO.: R.E.39

TEMP: 60.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH D PREPARED SIEVE FRACTION: 75-89μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
27.303	.00000	1.968					
			.0006715	1307.2	1.8119@-05	.3488	3.357@-04
27.137	1.0000	2.486					
			.0009220	1584.5	1.5420@-05	.3378	2.760@-04
26.859	2.6700	3.348					
			.0003967	1830.5	9.9180@-06	.3274	1.710@-04
26.719	3.8300	3.777					
			.0003652	1984.2	1.1791@-05	.3205	1.985@-04
26.580	4.7500	4.205					
			.0006572	2203.0	8.9830@-06	.3102	1.460@-04
26.302	7.0000	5.060					
			.0002983	2415.0	7.6351@-06	.2998	1.193@-04
26.162	8.2500	5.485					
			.0002819	2550.1	7.4051@-06	.2929	1.128@-04
26.023	9.5000	5.910					
			.0005230	2745.3	7.1380@-06	.2825	1.046@-04
25.743	12.000	6.758					
			.0004771	2996.8	5.7362@-06	.2687	7.952@-05
25.464	15.000	7.603					
			.0002239	3179.2	5.6281@-06	.2583	7.463@-05
25.324	16.500	8.024					
			.0002154	3297.1	4.5734@-06	.2514	5.886@-05
25.184	18.330	8.444					
			.0002077	3412.8	6.2086@-06	.2444	7.751@-05
25.044	19.670	8.863					
			.0002006	3526.5	3.9861@-06	.2375	4.823@-05
24.904	21.750	9.281					
			.0005658	3746.8	3.8321@-06	.2236	4.352@-05
24.483	28.250	10.53					
			.0001771	3963.6	5.5818@-06	.2097	5.903@-05
24.343	29.750	10.95					
			.0001722	4068.6	4.8235@-06	.2027	4.920@-05
24.202	31.500	11.36					
			.0001676	4172.1	3.4117@-06	.1958	3.352@-05
24.061	34.000	11.78					
			.0001633	4274.2	3.8388@-06	.1888	3.628@-05
23.921	36.250	12.19					

TABLE :137(CONTD.) RUN NO. :R.E.39(CONTD.)

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
23.921	36.250	12.19					
			.0001592	4374.9	4.3834@-06	.1818	3.980@-05
23.780	38.250	12.60					
			.0003074	4523.2	4.5040@-06	.1714	3.843@-05
23.498	42.250	13.42					
			.0001482	4669.7	2.8606@-06	.1609	2.281@-05
23.358	45.500	13.83					
			.0001450	4765.5	2.1175@-06	.1539	1.611@-05
23.217	50.000	14.24					
			.0005521	4999.1	2.9416@-06	.1364	1.972@-05
22.652	64.000	15.87					
			.0002593	5274.8	1.7730@-06	.1154	9.974@-06
22.369	77.000	16.68					
			.0002498	5452.6	2.1181@-06	.1014	1.041@-05
22.086	89.000	17.48					
			.0001215	5583.9	1.2584@-06	.0909	5.521@-06
21.944	100.00	17.88					
			.0001193	5670.1	1.4783@-06	.0838	5.967@-06
21.802	110.00	18.28					
			.0001173	5755.5	1.4460@-06	.0768	5.332@-06
21.661	121.00	18.68					
			.0003409	5923.3	1.2196@-06	.0628	3.627@-06
21.235	168.00	19.88					
			.0001098	6089.6	1.1332@-06	.0487	2.614@-06
21.093	189.00	20.27					
			.0001080	6171.1	8.8675@-07	.0416	1.742@-06
20.950	220.00	20.66					

EQUILIBRIUM CONCENTRATION = 20.181 INITIAL SEED AREA = 1198

TABLE: 138 RUN NO.: R.A.7

TEMP: 70.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH P.E.PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
29.736	.00000	1.920					
			.0008007	1120.5	1.8552@-06	.1271	1.668@-05
29.569	24.000	2.449					
			.0005679	1310.5	1.1088@-06	.1212	9.464@-06
29.430	54.000	2.889					
			.0005031	1474.4	1.3449@-06	.1160	1.094@-05
29.291	77.000	3.327					
			.0004534	1631.3	1.6283@-06	.1107	1.259@-05
29.152	95.000	3.764					
			.0004138	1782.3	1.5661@-06	.1054	1.15@-05
29.012	113.00	4.199					
			.0003815	1927.9	1.3727@-06	.1001	9.537@-06
28.873	133.00	4.634					
			.0003544	2068.9	1.2286@-06	.0948	8.056@-06
28.734	155.00	5.067					
			.0003315	2205.6	1.2796@-06	.0895	7.893@-06
28.594	176.00	5.499					
			.0003118	2338.4	1.4189@-06	.0842	8.206@-06
28.454	195.00	5.930					
			.0002946	2467.8	1.2990@-06	.0789	7.015@-06
28.315	216.00	6.360					
			.0002794	2593.9	1.0709@-06	.0736	5.374@-06
28.175	242.00	6.788					
			.0002659	2717.1	8.6871@-07	.0683	4.029@-06
28.035	275.00	7.215					
			.0002536	2837.4	6.6198@-07	.0629	2.818@-06
27.895	320.00	7.640					
			.0002426	2955.1	6.9474@-07	.0576	2.695@-06
27.756	365.00	8.064					

EQUILIBRIUM CONCENTRATION = 26.310 INITIAL SEED AREA = 1014

TABLE: 139 RUN NO.: R.E.F.1

TEMP: 70.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH D PREPARED SIEVE FRACTION: 75-89μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
29.833	.00000	1.948					
			.0005299	1270.2	1.2639@-05	.1315	1.177@-04
29.708	2.2500	2.345					
			.0005168	1443.8	4.6266@-06	.1265	4.135@-05
29.569	8.5000	2.786					
			.0004602	1617.1	2.9952@-06	.1212	2.557@-05
29.430	17.500	3.226					
			.0004166	1782.2	2.8433@-06	.1160	2.314@-05
29.291	26.500	3.664					
			.0003817	1940.3	2.8991@-06	.1107	2.245@-05
29.152	35.000	4.102					
			.0003531	2092.4	2.6680@-06	.1054	1.962@-05
29.012	44.000	4.538					
			.0006387	2309.4	2.6185@-06	.0974	1.774@-05
28.734	62.000	5.409					
			.0002911	2519.9	2.7671@-06	.0895	1.712@-05
28.594	70.500	5.843					
			.0002757	2654.2	2.7942@-06	.0842	1.622@-05
28.454	79.000	6.275					
			.0005127	2848.5	2.3328@-06	.0762	1.221@-05
28.175	100.00	7.138					
			.0002392	3038.2	2.8486@-06	.0683	1.329@-05
28.035	109.00	7.568					
			.0002293	3160.4	1.3372@-06	.0629	5.732@-06
27.895	129.00	7.996					
			.0002204	3280.2	2.1666@-06	.0576	8.475@-06
27.756	142.00	8.423					
			.0002122	3397.6	2.4983@-06	.0523	8.842@-06
27.615	154.00	8.849					
			.0004029	3569.0	2.1154@-06	.0443	6.295@-06
27.335	186.00	9.699					
			.0001913	3737.4	1.3568@-06	.0363	3.298@-06
27.195	215.00	10.12					
			.0001853	3846.7	1.4954@-06	.0310	3.088@-06
27.055	245.00	10.54					
			.0001795	3954.1	1.1736@-06	.0256	1.995@-06
26.914	290.00	10.96					

EQUILIBRIUM CONCENTRATION = 26.310 INITIAL SEED AREA = 1185

TABLE: 140 RUN NO.: R.E.F.2

TEMP: 60.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
27.192	.00000	1.970					
			.0009189	1921.4	3.3838@-05	.3391	6.126@-04
26.859	.75000	3.006					
			.0005775	2522.8	1.6880@-05	.3240	2.887@-04
26.580	1.7500	3.865					
			.0004801	3020.6	1.9655@-05	.3102	3.201@-04
26.302	2.5000	4.721-					
			.0006032	3593.6	1.3142@-05	.2929	2.011@-04
25.883	4.0000	6.000					
			.0003470	4134.9	9.7217@-06	.2756	1.388@-04
25.604	5.2500	6.847					
			.0001607	4443.1	1.0137@-05	.2652	1.385@-04
25.464	5.8300	7.269					
			.0003010	4738.2	9.8191@-06	.2548	1.286@-04
25.184	7.0000	8.112					
			.0001412	5027.4	5.6477@-06	.2444	7.060@-05
25.044	8.0000	8.531					
			.0001359	5214.6	5.6071@-06	.2375	6.793@-05
24.904	9.0000	8.950					
			.0002578	5488.2	4.9580@-06	.2271	5.728@-05
24.623	11.250	9.786					
			.0002411	5844.3	4.9651@-06	.2132	5.358@-05
24.343	13.500	10.62					
			.0002268	6189.5	3.7660@-06	.1992	3.780@-05
24.061	16.500	11.45					
			.0002143	6524.7	3.8454@-06	.1853	3.571@-05
23.780	19.500	12.27					
			.0001028	6770.8	2.9468@-06	.1748	2.570@-05
23.639	21.500	12.69					
			.0001983	7010.6	4.0415@-06	.1644	3.304@-05
23.358	24.500	13.51					
			.0000955	7247.5	3.1327@-06	.1539	2.387@-05
23.217	26.500	13.92					
			.0001847	7478.7	3.0705@-06	.1434	2.173@-05
22.934	30.750	14.73					
			.0001768	7782.0	2.2263@-06	.1294	1.415@-05
22.652	37.000	15.55					
			.0000856	8005.5	2.4536@-06	.1189	1.427@-05
22.510	40.000	15.95					
			.0000839	8152.1	2.5618@-06	.1119	1.398@-05
22.369	43.000	16.36					

TABLE :140 (CONTD.) RUN NO. :R.E.F2(CONTD.)

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
22.369	43.000	16.36					
			.0000823	8297.2	2.3030@-06	.1049	1.175@-05
22.227	46.500	16.76					
			.0000807	8440.8	2.8320@-06	.0979	1.345@-05
22.086	49.500	17.16					
			.0001571	8653.2	2.4826@-06	.0874	1.048@-05
21.802	57.000	17.96					
			.0001516	8932.4	1.9580@-06	.0733	6.890@-06
21.519	68.000	18.76					
			.0001465	9206.4	2.3571@-06	.0592	6.658@-06
21.235	79.000	19.56					
			.0000714	9409.1	1.1848@-06	.0487	2.747@-06
21.093	92.000	19.96					
			.0001394	9608.2	2.2908@-06	.0381	4.100@-06
20.808	109.00	20.75					
			.0000681	9805.6	2.3880@-06	.0275	3.096@-06
20.666	120.00	21.14					
			.0001591	10025	2.4402@-06	.0155	1.591@-06
20.324	170.00	22.09					
			.0000516	10230	1.5731@-06	.0042	2.578@-07
20.210	270.00	22.40					

EQUILIBRIUM CONCENTRATION = 20.181 INITIAL SEED AREA = 1578

TABLE: 141 RUN NO.:R.E.F.4

TEMP:60.0°C CELL: C STIRRER SPEED:2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION:89-105 μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
24.483	.00000	1.988					
			.0004150	1738.6	2.6207@-06	.2200	2.964@-05
24.343	7.0000	2.414					
			.0003555	2023.3	9.5837@-07	.2130	1.045@-05
24.202	24.000	2.838					
			.0003127	2294.3	2.1236@-06	.2060	2.233@-05
24.061	31.000	3.262					
			.0002802	2553.5	8.6471@-07	.1990	8.756@-06
23.921	47.000	3.685					
			.0002547	2802.5	1.3075@-06	.1919	1.274@-05
23.780	57.000	4.106					
			.0002341	3042.5	1.0424@-06	.1849	9.753@-06
23.639	69.000	4.527					
			.0002170	3274.5	1.0990@-06	.1779	9.863@-06
23.498	80.000	4.947					
			.0002026	3499.5	9.8202@-07	.1708	8.440@-06
23.358	92.000	5.366					
			.0001902	3718.0	1.0523@-06	.1638	8.647@-06
23.217	103.00	5.784					
			.0001795	3930.7	9.5392@-07	.1567	7.480@-06
23.075	115.00	6.201					
			.0001701	4138.2	8.7629@-07	.1497	6.543@-06
22.934	128.00	6.617					
			.0001618	4340.7	9.5032@-07	.1426	6.742@-06
22.793	140.00	7.032					
			.0001544	4538.8	8.2003@-074	.1356	5.514@-06
22.652	154.00	7.446					
			.0001477	4732.6	9.6848@-07	.1285	6.155@-06
22.510	166.00	7.859					
			.0001417	4922.6	9.1001@-07	.1214	5.450@-06
22.369	179.00	8.271					
			.0001362	5109.0	7.1237@-07	.1144	4.006@-06
22.227	196.00	8.683					

TABLE :141 (CONTD.) RUN NO. R.E.F4 (CONTD.)

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
22.227	196.00	8.683					
			.0001312	5291.9	7.7936@-07	.1073	4.099@-06
22.086	212.00	9.093					
			.0002491	5559.3	6.7710@-07	.0967	3.193@-06
21.802	251.00	9.911					
			.0002332	5907.1	6.6300@-07	.0825	2.650@-06
21.519	295.00	10.73					
			.0001112	6161.8	7.2906@-07	.0718	2.528@-06
21.377	317.00	11.13					
			.0002131	6408.7	5.5956@-07	.0612	1.639@-06
21.093	382.00	11.94					
			.0001021	6652.1	6.4149@-07	.0505	1.547@-06
20.950	415.00	12.34					

EQUILIBRIUM CONCENTRATION = 20.010 INITIAL SEED AREA = 1593

TABLE: 142 RUN NO.: R.E.F.5

TEMP: 50.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH D PREPARED SIEVE FRACTION: 75-89μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
19.278	.00000	1.985					
			.0003251	1703.0	1.3432@-07	.2725	1.355@-06
19.165	120.00	2.312					
			.0002182	1897.3	3.7066@-07	.2660	3.637@-06
19.081	150.00	2.556					
			.0003264	2109.7	2.9579@-07	.2585	2.814@-06
18.939	208.00	2.963					
			.0002902	2365.7	4.0716@-07	.2491	3.720@-06
18.798	247.00	3.369					
			.0002622	2611.2	4.5313@-07	.2398	3.972@-06
18.657	280.00	3.773					
			.0002397	2847.5	4.7583@-07	.2304	3.996@-06
18.515	310.00	4.176					
			.0002213	3075.6	3.7247@-07	.2210	2.991@-06
18.374	347.00	4.579					
			.0002060	3296.4	5.3728@-07	.2117	4.119@-06
18.232	372.00	4.980					
			.0001928	3510.7	3.3003@-07	.2023	2.410@-06
18.090	412.00	5.380					
			.0001815	3719.1	3.9613@-07	.1929	2.750@-06
17.949	445.00	5.779					
			.0017972	5015.8	3.4464@-07	.1252	1.404@-06
16.046	1085.0	11.05					
			.0000619	6256.5	2.7667@-07	.0594	5.628@-07
15.961	1140.0	11.28					
			.0000608	6349.7	2.2102@-07	.0538	4.054@-07
15.875	1215.0	11.51					
			.0001535	6514.8	4.5482@-08	.0434	6.477@-08
15.647	2400.0	12.10					

EQUILIBRIUM CONCENTRATION = 15.106 INITIAL SEED AREA = 1590

TABLE: 143 RUN NO.:R.P.E.F.4

TEMP:70.0°C CELL:C STIRRER SPEED:2000 R.P.M.

SEED: BATCH P.E.PREPARED SIEVE FRACTION:89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
27.491	.00000	1.970					
			.0006886	1133.6	3.6570@-06	.1068	2.550@-05
27.343	13.500	2.431					
			.0005913	1314.6	1.9186@-06	.1008	1.258@-05
27.195	37.000	2.890					
			.0005200	1486.8	7.9947@-07	.0949	4.906@-06
27.047	90.000	3.346					
			.0004659	1651.2	8.4818@-07	.0889	4.853@-06
26.899	138.00	3.801					
			.0004232	1808.8	7.6612@-07	.0829	4.069@-06
26.751	190.00	4.253					
			.0003885	1960.4	7.9213@-07	.0770	3.885@-06
26.603	240.00	4.703					
			.0003588	2106.5	3.4745@-07	.0710	1.560@-06
26.455	355.00	5.149					
			.0003342	2247.6	5.4514@-07	.0650	2.228@-06
26.307	430.00	5.593					
			.0002212	2364.2	4.9163@-07	.0599	1.843@-06
26.204	490.00	5.902					

EQUILIBRIUM CONCENTRATION = 24.771 INITIAL SEED AREA = 1041

TABLE: 144 RUN NO.: R.P.E.F.5

TEMP: 60.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH P.G. PREPARED SIEVE FRACTION: 89-105 μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
21.744	.00000	1.995					
			.0004580	1276.8	8.5255@-06	.1638	6.543@-05
21.628	3.5000	2.339					
			.0004679	1557.3	1.4862@-06	.1568	1.088@-05
21.482	25.000	2.769					
			.0003884	1867.0	7.0122@-07	.1490	4.854@-06
21.337	65.000	3.196					
			.0003328	2168.0	4.6346@-07	.1412	3.025@-06
21.192	120.00	3.622					
			.0002917	2458.2	2.2664@-07	.1334	1.389@-06
21.046	225.00	4.046					
			.0002605	2737.5	2.6702@-07	.1256	1.533@-06
20.901	310.00	4.467					
			.0002360	3006.9	2.7541@-07	.1178	1.475@-06
20.756	390.00	4.886					
			.0003377	3341.9	2.8943@-07	.1076	1.407@-06
20.523	510.00	5.552					
			.0011312	4339.4	2.6384@-07	.0737	8.197@-07
19.491	1200.0	8.434					
			.0001156	5225.2	1.7598@-07	.0425	3.210@-07
19.360	1380.0	8.790					

EQUILIBRIUM CONCENTRATION = 18.634 INITIAL SEED AREA = 1153

TABLE: 145 RUN NO.:R.P.E.F.6

TEMP:70.0°C CELL:C STIRRER SPEED:2000 R.P.M.

SEED: BATCH P.E.PREPARED SIEVE FRACTION:89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT K($MIN^{-1}CM^{-2}$)	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
27.580	.00000	1.944					
			.0004324	1083.3	2.9667@-06	.1116	2.162@-05
27.491	10.000	2.221					
			.0006320	1229.4	5.0582@-07	.1068	3.511@-06
27.343	100.00	2.679					
			.0010420	1486.2	1.7140@-06	.0979	1.085@-05
27.047	148.00	3.592					
			.0008491	1807.2	1.3521@-06	.0859	7.448@-06
26.751	205.00	4.498					
			.0010490	2173.1	1.1119@-06	.0710	4.995@-06
26.307	310.00	5.843					
			.0003061	2451.6	6.8787@-07	.0590	2.551@-06
26.160	370.00	6.286					
			.0006686	2670.3	7.3651@-07	.0489	2.229@-06
25.805	520.00	7.340					
			.0003049	2895.3	7.2208@-07	.0382	1.694@-06
25.627	610.00	7.862					
			.0005083	3093.8	2.6589@-07	.0280	4.382@-07
25.302	1190.0	8.791					

EQUILIBRIUM CONCENTRATION = 24.771 INITIAL SEED AREA = 1027

TABLE: 146 RUN NO.:R.P.E.D.1

TEMP:70.0°C CELL:C STIRRER SPEED:2000 R.P.M.

SEED: BATCH P.E.PREPARED SIEVE FRACTION:89-105 μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
28.528	.00000	1.964					
			.0006976	1131.4	2.8460@-05	.1487	2.790@-04
28.380	1.2500	2.430					
			.0005985	1314.4	1.8230@-05	.1427	1.710@-04
28.232	3.0000	2.894					
			.0005267	1488.6	1.1759@-05	.1367	1.053@-04
28.084	5.5000	3.357					
			.0004722	1655.1	6.5046@-06	.1307	5.555@-05
27.935	9.7500	3.819					
			.0004292	1815.0	4.8026@-06	.1248	3.902@-05
27.787	15.250	4.279					
			.0007606	2042.3	3.0213@-06	.1158	2.271@-05
27.491	32.000	5.196					
			.0006620	2331.4	3.4089@-06	.1038	2.283@-05
27.195	46.500	6.108					
			.0003020	2538.7	2.6122@-06	.0949	1.589@-05
27.047	56.000	6.561					
			.0002861	2671.3	2.7961@-06	.0889	1.589@-05
26.899	65.000	7.012					
			.0002719	2800.7	1.7152@-06	.0829	9.063@-06
26.751	80.000	7.462					
			.0002593	2927.2	2.4114@-06	.0770	1.179@-05
26.603	91.000	7.911					
			.0004860	3110.9	1.6646@-06	.0680	7.147@-06
26.307	125.00	8.804					
			.0002282	3290.8	1.8086@-06	.0590	6.713@-06
26.160	142.00	9.248					
			.0002196	3407.1	1.4364@-06	.0531	4.774@-06
26.012	165.00	9.690					
			.0002114	3521.2	7.2043@-07	.0471	2.114@-06
25.864	215.00	10.13					
			.0002038	3633.0	6.1516@-07	.0411	1.568@-06
25.716	280.00	10.57					
			.0001151	3720.1	3.4384@-08	.0364	7.477@-08
25.627	1050.0	10.82					
			.0001096	3782.2	4.1863@-08	.0328	7.941@-08
25.539	1740.0	11.07					

EQUILIBRIUM CONCENTRATION = 24.771 INITIAL SEED AREA = 1038

TABLE: 147 RUN NO. R.P.E.D.2

TEMP: 50.0°C CELL: C STIRRER SPEED: 2000R.P.M.

SEED: BATCH P.G. PREPARED SIEVE FRACTION: 89-105 μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
17.516	.00000	1.994					
			.0005301	1298.4	3.2832@-07	.2359	2.651@-06
17.374	100.00	2.399					
			.0004298	1591.3	2.7999@-07	.2258	2.149@-06
17.232	200.00	2.802					
			.0003616	1881.8	2.0663@-07	.2157	1.506@-06
17.090	320.00	3.203					
			.0002535	2136.7	2.0272@-07	.2067	1.408@-06
16.977	410.00	3.523					
			.0025852	3894.3	3.1173@-07	.1288	1.202@-06
+ 14.892	1485.0	9.237					
			.0001135	5634.0	1.6132@-07	.0499	2.522@-07
14.748	1710.0	9.615					

EQUILIBRIUM CONCENTRATION = 14.116 INITIAL SEED AREA = 1152

TABLE: 148 RUN NO.: R.E.40

TEMP: 70.0°C CELL: C STIRRER SPEED: 2000 R.P.M.

SEED: BATCH E PREPARED SIEVE FRACTION: 89-105μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
29.708	.00000	1.976					
			.0004312	1734.2	4.8150@-06	.1265	4.312@-05
29.569	5.0000	2.417					
			.0003676	2028.7	3.0698@-06	.1212	2.626@-05
29.430	12.000	2.857					
			.0003222	2308.6	2.6340@-06	.1160	2.148@-05
29.291	19.500	3.297					
			.0002880	2576.0	2.1836@-06	.1107	1.694@-05
29.152	28.000	3.735					
			.0005014	2954.1	1.8382@-06	.1027	1.320@-05
28.873	47.000	4.609					
			.0004293	3432.9	1.8642@-06	.0921	1.193@-05
28.594	65.000	5.479					
			.0010276	4292.2	1.6394@-06	.0709	7.904@-06
27.756	130.00	8.076					
			.0001439	5016.5	1.4503@-06	.0523	5.141@-06
27.615	144.00	8.502					
			.0001383	5207.2	1.3621@-06	.0470	4.322@-06
27.475	160.00	8.928					
			.0003861	5575.3	1.2355@-06	.0363	2.970@-06
27.055	225.00	10.20					
			.0001199	5937.0	1.3027@-06	.0256	2.221@-06
26.914	252.00	10.62					
			.0001161	6111.5	1.3111@-06	.0203	1.759@-06
26.774	285.00	11.04					
			.0001124	6283.1	1.0439@-06	.0150	1.022@-06
26.633	340.00	11.46					

EQUILIBRIUM CONCENTRATION = 26.310 INITIAL SEED AREA = 1583

APPENDIX E - ANALYSIS OF PREVIOUS WORK (1)

TABLE: 149

SUMMARY OF PREVIOUS WORK (1) ON SEEDED CELLS STIRRED AT 500 R.P.M.

OLD RUN NO:	R.14			R.20			R.21		
NEW RUN NO:	R.A.1			R.A.2			R.A.3		
T_o °C	60.0			70.0			55.0		
SEED	44-53μA*			44-53μA*			44-53μA*		
c%m/v	TIME MINS	Kx10^7 $min^{-1}cm^{-2}$	c%m/v	TIME MINS	Kx10 7 $min^{-1}cm^{-2}$	c%m/v	TIME MINS	Kx10 7 $min^{-1}cm^{-2}$
24.00	0	13.95	30.00	0	2.97	21.50	0	126
23.88	9	1.71	29.86	5	1.475	21.41	10	77.5
23.77	80	2.25	29.73	14	1.175	21.29	25	12.9
23.65	130	1.69	29.60	25	1.100	21.78	110	7.37
23.54	195	2.335	29.47	37	1.40	21.06	255	8.02
23.43	240	1.70	29.35	46	1.19	20.26	1110	8.01
23.31	300	2.05	29.22	60	1.59	20.15	1230	4.09
23.08	400	1.17	28.96	75	1.097	20.03	1470	5.00
21.83	1440	2.51	28.84	86	1.242	19.46	2550	4.98
21.71	1500	0.552	28.71	96	0.428	19.35	2790	4.28
21.60	1800		28.58	125	0.395	19.23	3090	
			28.45	158	0.359			
			28.33	195	0.275			
			28.07	300	0.313			
			27.94	350	0.169			
			27.18	1230	0.147			
			27.05	1560				

* SPECIALLY PREPARED SEED

TABLE: 149 (CONT.)

OLD RUN NO:	R.23			R.27			R.29		
NEW RUN NO:	R.A.4			R.P.A.1			R.P.A.2		
T_o °C	80.0			60.0			70.0		
SEED	44-53μA*			44-53μP.A.*			44-53μP.A.*		
	c/m/v	TIME MINS	Kx10⁷ $min^{-1}cm^{-2}$	c/m/v	TIME MINS	Kx10⁷ $min^{-1}cm^{-2}$	c/m/v	TIME MINS	Kx10⁷ $min^{-1}cm^{-2}$
	37.00	0		24.00	0		29.00	0	
			7.4			15.3			49.0
	36.81	2.5		23.88	7		28.70	3	
			7.79			5.08			2.94
	36.68	4.5		23.77	26		28.64	95	
			4.22			0.78			12.18
	36.56	8		23.65	140		28.39	115	
			2.15			6.79			16.05
	36.43	15		23.45	165		27.75	150	
			1.65			6.47			9.44
	36.31	24		23.20	190		27.62	163	
			1.55			4.79			10.3
	35.68	74		22.63	270		27.50	175	
			1.07						10.5
	35.57	90					27.24	200	
			1.225						9.31
	35.45	105					27.11	215	
			0.94						9.82
	35.20	150					26.99	230	
			0.895						9.01
	34.95	210					26.48	310	
			1.06						15.55
	34.82	243					26.35	325	
									5.46
							26.22	375	
									6.70
							26.09	425	
									7.93
							25.97	480	

* SPECIALLY PREPARED SEED

TABLE: 149 (CONT.)

OLD RUN NO:	R.30			R.31			R.32		
NEW RUN NO:	R.P.A.3			R.C.1			R.P.A.4		
T_o °C	80.0			60.0			50.0		
SEED	44-53 μ P.A.*			44-53 μ P.A.*			44-53 μ P.A.*		
	c%m/v	TIME MINS	Kx10^{-7} min^{-1}cm^{-2}	c%m/v	TIME MINS	Kx10^{-7} min^{-1}cm^{-2}	c%m/v	TIME MINS	Kx10^{-7} min^{-1}cm^{-2}
	36.00	0		24.00	0		18.50	0	
			2.82			5.70			4.82
	35.19	26		23.88	25		18.18	25	
			1.53			5.17			3.38
	34.94	45		23.77	40		18.06	60	
			1.17			10.5			1.78
	34.56	85		23.20	100		17.95	120	
			1.42			5.63			0.860
	34.44	97		22.97	145		17.84	300	
			1.635			5.20			
	34.19	120		22.85	170				
			1.13			5.31			
	33.94	160		22.63	220				
			2.75			4.78			
	33.82	170		22.51	250				
			1.62			7.44			
	33.57	215		22.28	290				
			0.983			6.52			
	33.44	270		22.17	315				

* SPECIALLY PREPARED SEED

TABLE: 149 (CONT.)

OLD RUN NO:	R.33			R.34		
NEW RUN NO:	R.A.5			R.P.A.5		
T_o °C	50.0			80.0		
SEED	44-53μ A^{36}			44-53μ A^{36}		
	o%m/v	TIME MINS	Kx10^7 $min^{-1}cm^{-2}$	o%m/v	TIME MINS	Kx10^7 $min^{-1}cm^{-2}$
	19.50	0		36.00	0	
			4.22			8.4
	19.08	960		35.56	4	
			5.16			4.21
	18.96	1140		35.44	7.5	
			3.74			4.19
	18.85	1380		35.31	11	
						1.60
				35.19	20	
						1.835
				35.06	28	
						1.837
				34.94	36	
						3.41
				34.69	45	
						2.315
				34.56	52	
						2.44
				34.44	59	
						2.01
				34.32	68	
						0.723
				34.19	95	
						1.125
				34.07	114	
						1.21
				33.94	124	

COMPUTED PREVIOUS RESULTS(1) WITH PREVIOUS SIZE ANALYSES(D_o) AND PREVIOUS EQUILIBRIUM VALUES

TABLE: 150 RUN NO.: R.P.A.2

TEMP: 60.0° C CELL: A STIRRER SPEED: 500 R.P.M.

SEED: BATCH P.A. PREPARED SIEVE FRACTION: 44-53 μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
24.021	.00000	2.000					
			.0002117	3242.9	1.2334@-06	.2491	1.512@-05
23.887	7.0000	2.404					
			.0001739	4014.9	3.8595@-07	.2420	4.576@-06
23.750	26.000	2.815					
			.0001430	4761.1	5.4683@-08	.2350	6.273@-07
23.616	140.00	3.217					
			.0002346	5803.4	4.2830@-07	.2245	4.692@-06
23.349	165.00	4.019					
			.0001901	7116.2	3.7244@-07	.2106	3.802@-06
23.082	190.00	4.817					
			.0003653	9164.2	2.5378@-07	.1863	2.283@-06
22.419	270.00	6.787					

EQUILIBRIUM CONCENTRATION = 19.177 INITIAL SEED AREA = 2856

TABLE: 151 RUN NO.: R.A.5

TEMP: 50.0°C CELL: A STIRRER SPEED: 500 R.P.M.

SEED: BATCH A PREPARED SIEVE FRACTION: 44-53 μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
19.380	.00000	2.000					
			.0006010	4225.6	3.2251@-08	.2501	3.130@-07
18.880	960.00	3.452					
			.0001052	5896.2	3.4119@-08	.2296	2.922@-07
18.753	1140.0	3.818					
			.0000928	6523.2	2.3463@-08	.2214	1.933@-07
18.628	1380.0	4.175					

EQUILIBRIUM CONCENTRATION = 15.302 INITIAL SEED AREA = 2877

COMPUTED PREVIOUS RESULTS(1) WITH PREVIOUS SIZE ANAIYSES(D_c)

BUT CORRECTED EQUILIBRIUM VALUES

TABLE: 152 RUN NO.: R.P.A.1

TEMP: 60.0°C CELL: A STIRRER SPEED: 500 R.P.M.

SEED: BATCH P.A.PREPARED SIEVE FRACTION: 44-53μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
24.021	.00000	2.000					
			.0002117	3242.9	1.1074@-06	.2855	1.512@-05
23.887	7.0000	2.404					
			.0001739	4014.9	3.4548@-07	.2782	4.576@-06
23.750	26.000	2.815					
			.0001430	4761.1	4.8795@-08	.2710	6.273@-07
23.616	140.00	3.217					
			.0002346	5803.4	3.8026@-07	.2602	4.692@-06
23.349	165.00	4.019					
			.0001901	7116.2	3.2822@-07	.2459	3.802@-06
23.082	190.00	4.817					
			.0003653	9164.2	2.2010@-07	.2209	2.283@-06
22.419	270.00	6.787					

EQUILIBRIUM CONCENTRATION = 18.634 INITIAL SEED AREA = 2856

TABLE: 153 RUN NO.: R.A.5

TEMP: 50.0°C CELL: A STIRRER SPEED: 500 R.P.M.

SEED: BATCH A PREPARED SIEVE FRACTION: 44-53μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
19.380	.00000	2.000					
			.0006010	4225.6	3.2036@-08	.2522	3.130@-07
18.880	960.00	3.452					
			.0001052	5896.3	3.3871@-08	.2317	2.922@-07
18.753	1140.0	3.818					
			.0000928	6523.2	2.3286@-08	.2235	1.933@-07
18.628	1380.0	4.175					

EQUILIBRIUM CONCENTRATION = 15.277 INITIAL SEED AREA = 2877

COMPUTED PREVIOUS RESULTS(1) WITH CORRECTED SIZE ANALYSES(D)

AND EQUILIBRIUM VALUES

TABLE: 154 RUN NO.: R.A.5

TEMP: 50.0C CELL: A STIRRER SPEED: 500 R.P.M.

SEED: BATCH A PRERARED SIEVE FRACTION: 44-53μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
19.380	.00000	2.000					
			.0005954	4264.9	3.1740@-08	.2522	3.101@-07
18.880	960.00	3.452					
			.0001043	5946.4	3.3585@-08	.2317	2.897@-07
18.753	1140.0	3.818					
			.0000920	6577.1	2.3095@-08	.2235	1.918@-07
18.628	1380.0	4.175					

EQUILIBRIUM CONCENTRATION = 15.277 INITIAL SEED AREA = 2907

TABLE: 155 RUN NO.: R.A.1

TEMP 60.0° C CELL: A STIRRER SPEED: 500 R.P.M.

SEED: BATCH A PREPARED SIEVE FRACTION: 44-53 μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
23.845	.00000	2.000					
			.0002027	3275.6	1.1652@-06	.1884	1.126@-05
23.716	9.0000	2.390					
			.0001680	4022.6	1.2737@-07	.1819	1.183@-06
23.583	80.000	2.789					
			.0001384	4763.1	1.5515@-07	.1753	1.384@-06
23.454	130.00	3.178					
			.0001201	5475.7	1.0785@-07	.1688	9.240@-07
23.324	195.00	3.566					
			.0001065	6165.6	1.4395@-07	.1624	1.183@-06
23.194	240.00	3.954					
			.0000959	6833.2	1.0152@-07	.1559	7.990@-07
23.064	300.00	4.341					
			.0001698	7796.1	1.1533@-07	.1461	8.490@-07
22.802	400.00	5.122					
			.0006098	11270	5.6616@-08	.1057	2.932@-07
21.448	1440.0	9.131					
			.0000407	14329	1.0074@-07	.0689	3.396@-07
21.329	1500.0	9.476					
			.0000395	14768	2.1415@-08	.0629	6.579@-08
21.209	1800.0	9.821					

EQUILIBRIUM CONCENTRATION = 20.010 INITIAL SEED AREA = 2907

TABLE: 156 RUN NO.: R.A.2

TEMP: 70.0°C CELL: A STIRRER SPEED: 500 R.P.M.

SEED: BATCH A PREPARED SIEVE FRACTION: 44-53μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
29.424	.00000	2.000					
			.0002137	3298.4	2.6006@-06	.1159	2.137@-05
29.294	5.0000	2.414					
			.0001719	4082.2	1.2203@-06	.1109	9.548@-06
29.163	14.000	2.828					
			.0001440	4857.3	8.7898@-07	.1060	6.545@-06
29.032	25.000	3.240					
			.0001269	5617.3	7.4742@-07	.1009	5.286@-06
28.898	37.000	3.661					
			.0001007	6321.5	8.3348@-07	.0961	5.597@-06
28.778	46.000	4.037					
			.0000913	6965.0	5.1077@-07	.0916	3.261@-06
28.658	60.000	4.413					
			.0001611	7888.4	9.1115@-07	.0847	5.369@-06
28.418	75.000	5.163					
			.0000720	8785.9	6.0682@-07	.0779	3.271@-06
28.298	86.000	5.536					
			.0000674	9360.3	6.6592@-07	.0733	3.372@-06
28.178	96.000	5.909					
			.0000635	9920.7	2.3117@-07	.0687	1.095@-06
28.058	125.00	6.281					
			.0000709	10518	2.4526@-07	.0637	1.075@-06
27.915	158.00	6.722					
			.0000668	11151	2.2568@-07	.0583	9.023@-07
27.772	195.00	7.162					
			.0001232	12068	1.7151@-07	.0502	5.868@-07
27.487	300.00	8.041					
			.0000571	12964	1.9980@-07	.0420	5.709@-07
27.344	350.00	8.478					
			.0002781	14789	1.1249@-07	.0242	1.580@-07
26.549	1230.0	10.90					
			.0000398	16555	1.4256@-07	.0066	6.029@-08
26.419	1560.0	11.29					

EQUILIBRIUM CONCENTRATION = 26.310 INITIAL SEED AREA = 2907

TABLE: 157 RUN NO. R.A.3

TEMP 55.0°C CELL: A STIRRER SPEED: 500 R.P.M.

SEED: BATCH A PREPARED SIEVE FRACTION: 44-53 μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
21.358	.00000	2.000					
			.0002035	3277.3	1.0108@-06	.2321	1.017@-05
21.225	10.000	2.392					
			.0001523	3988.1	5.2341@-07	.2247	5.076@-06
21.104	25.000	2.750					
			.0001271	4655.0	7.9790@-08	.2178	7.476@-07
20.985	110.00	3.099					
			.0001139	5305.9	4.3416@-08	.2108	3.928@-07
20.863	255.00	3.456					
			.0005511	7632.7	4.0719@-08	.1831	3.223@-07
20.027	1110.0	5.903					
			.0000629	9909.1	4.0043@-08	.1552	2.623@-07
19.900	1230.0	6.272					
			.0000596	10451	1.9940@-08	.1478	1.241@-07
19.772	1470.0	6.640					
			.0002579	11977	2.2701@-08	.1258	1.194@-07
19.138	2550.0	8.461					
			.0000437	13463	2.1093@-08	.1039	9.110@-08
19.016	2790.0	8.809					
			.0000422	13916	1.7526@-08	.0969	7.038@-08
18.893	3090.0	9.156					

EQUILIBRIUM CONCENTRATION = 17.281 INITIAL SEED AREA = 2907

TABLE: 158 RUN NO.:R.P.A.1

TEMP:60.0°C CELL:A STIRRER SPEED:500 R.P.M.

SEED: BATCH P.A.PREPARED SIEVE FRACTION:44-53μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
24.021	.00000	2.000					
			.0002088	3288.0	1.0922@-06	.2855	1.491@-05
23.887	7.0000	2.404					
			.0001717	4065.9	3.4115@-07	.2782	4.519@-06
23.750	26.000	2.815					
			.0001413	4817.6	4.8222@-08	.2710	6.199@-07
23.616	140.00	3.217					
			.0002320	5867.7	3.7609@-07	.2602	4.640@-06
23.349	165.00	4.019					
			.0001882	7190.3	3.2484@-07	.2459	3.763@-06
23.082	190.00	4.817					
			.0003618	9254.1	2.1797@-07	.2209	2.261@-06
22.419	270.00	6.787					

EQUILIBRIUM CONCENTRATION = 18.634 INITIAL SEED AREA = 2898

TABLE: 159 RUN NO.:R.P.A.4

TEMP:50.0°C CELL:A STIRRER SPEED:500 R.P.M.

SEED: BATCH P.A.PREPARED SIEVE FRACTION:44-53μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
18.151	.00000	2.000					
			.0001797	3228.3	3.7052@-07	.2816	3.593@-06
18.032	25.000	2.342					
			.0001489	3882.7	2.2694@-07	.2732	2.128@-06
17.913	60.000	2.683					
			.0001276	4520.1	1.1739@-07	.2648	1.064@-06
17.794	120.00	3.023					
			.0001096	5129.5	3.4772@-08	.2564	3.046@-07
17.678	300.00	3.355					

EQUILIBRIUM CONCENTRATION = 14.116 INITIAL SEED AREA = 2898

TABLE: 160 RUN NO.: R.P.A.2

TEMP: 70.0°C CELL: A STIRRER SPEED: 500 R.P.M.

SEED: BATCH P.A. PREPARED SIEVE FRACTION: 44-53μ

CONCN. %M/V	TOTAL TIME MINS.	CRYSTAL MASS (GRAMS)	DIAMETER INCREASE (CM)	MEAN AREA CM^2	GROWTH RATE CONSTANT $K(MIN^{-1}CM^{-2})$	MEAN SUPER-SATN.	GROWTH RATE CM/MIN.
28.848	.00000	2.000					
			.0002079	3286.1	3.2231@-06	.1620	3.464@-05
28.721	3.0000	2.402					
			.0003094	4415.9	1.6426@-07	.1543	1.682@-06
28.466	95.000	3.204					
			.0002580	5924.6	6.8366@-07	.1434	6.451@-06
28.178	115.00	4.103					
			.0004686	8432.4	8.3412@-07	.1226	6.695@-06
27.435	150.00	6.418					
			.0000691	10467	3.9430@-07	.1047	2.658@-06
27.295	163.00	6.846					
			.0000653	11049	4.2754@-07	.0991	2.721@-06
27.156	175.00	7.272					
			.0001198	11886	4.1259@-07	.0908	2.396@-06
26.881	200.00	8.113					
			.0000563	12704	3.5775@-07	.0824	1.878@-06
26.742	215.00	8.537					
			.0000494	13213	3.3659@-07	.0770	1.648@-06
26.615	230.00	8.923					
			.0001803	14376	2.7873@-07	.0642	1.127@-06
26.109	310.00	10.45					
			.0000416	15519	4.2877@-07	.0515	1.386@-06
25.982	325.00	10.84					
			.0000394	15954	1.3557@-07	.0464	3.942@-07
25.858	375.00	11.21					
			.0000437	16408	1.7046@-07	.0410	4.370@-07
25.716	425.00	11.63					
			.0000406	16875	1.6729@-07	.0354	3.692@-07
25.580	480.00	12.04					

EQUILIBRIUM CONCENTRATION = 24.771 INITIAL SEED AREA = 2898

APPENDIX F - IMPURITY EXTRACTION WITH MOLECULAR SIEVE

APPENDIX F

Impurity extraction with Molecular Sieve (Type 13X)

Extraction Procedure:

1 litre of 10.00% mass fraction purified Batch G (P.G.) solution was made up and held at 40°C in a flask, heated with an isomantle, controlled with a voltage regulator. 10.00g of molecular sieve (type 13X) 1/16in pellets were added and stirred for 2 hours. The solution was filtered through a No. 1. Whatman paper and a sample of the wet sieve weighed, dried at 100°C, reweighed and then calcined in an open muffle furnace maintained between 700°C and 900°C for 3 days. The resulting sieve was again weighed enabling the amount of vaporised material to be calculated. The solution was filtered through a 0.45μ millepore filter to remove attrited sieve and placed in the flask at 40°C for the second extraction.

Example Calculation: 1st Extraction

Readings:

1) Weight of empty, dry crucible = 26.0189g.

2) Weight of crucible + wet sieve = 38.4380g.

3) Weight of crucible + dry sieve = 34.38 52g.

4) Weight of crucible + calcined sieve = 32.5320g. after 3 days heating

= 32.5324g. after 6 days heating

Blank Test on Dry Molecular sieve

	Loss of weight per 10.000g. sieve
1st Test	0.690g.
2nd Test	0.680g.
3rd Test	0.672g.
Average:	0.682g./ 10.000g.sieve

Calculation:

Weight of water in sieve sample = Weight (2) - Weight (3) = 4.0528g.

10.0% P.E. mass fraction in solution = 11.1% mass ratio

∴ Weight of attached P.E. in sieve from dried mother liquor

= 4.0528 x 0.111 = 0.4499g

Total Weight of vapourised adsorbent = Weight (3) - Weight (4)

= 1.8532g.

Weight of dry sieve sample = (Weight(4) - Weight(1)) + $\frac{0.682}{10}$(Wt.(4)-Wt(1))

= 6.9 571g.

Weight of adsorbed contamirant = 1.8532-0.4499-0.0682x6.9571

= 0.9285g.

Extract = 0.9285 x $\frac{10.00}{6.9571}$

= 1.33g./10.00g sieve ie per 100.0gP.G.

Summary of Results

Sample	Extract (g / 100 g. solute)
1st Extraction of Purified Batch G.	1.33
2nd " " " " "	1.15
3rd " " " " "	1.11
4th " " " " "	1.13
5th " " " " "	1.11
6th " " " " "	1.13
1st Extraction of Purified Batch D	1.11
2nd " " " " "	1.12
Extractionof Batch E	1.09

It is apperent from the results that a datum exists at about 1.11g. extract / 100g. solute even for P.D. with little or no expected contaminant. As the growth studies showed no effect of molecular sieve extraction on purified Batch D, this is assumed to be an absorption of Pentaerythritol molecules in the sieve. The disappointing scatter of results about this datum of 1.11g. / 100g. solute is attributed to the difficulty of weighing a wet sample in an evaporating crucible with any accuracy.

The contaminant extractions are assumed to be above this datum for P.G. material and indicate 0.22g. contaminant / 100g. P.G. removed

with the first extraction, 0.04g contaminant / 100g P.G. removed with the second extraction but an immeasurable effect with each successive extraction. This indicates a total of about 0.26g. contaminant per 100g. P.G. extracted.

Estimation of remaining contaminant after 6 successive extractions of P.G.

Assume a partition coefficient (K') exists of the contaminant with x_w mass fraction in solution and x_s mass fraction in the molecular sieve such that: $K' = \frac{x_w}{x_s}$

If the mass of solution is W, the mass of molecular sieve S', and the ratio $\frac{S'}{W} = \gamma$

Then on the 1st extraction $Wx_{w_0} = Wx_{w_1} + Sx_{s_1}$

where Wx_{w_0} is the original mass of contaminant.

$$\therefore \quad Wx_{w_0} = Wx_{w_1} + \frac{S'}{K'}x_{w_1} \qquad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots (1)$$

$$= Wx_{w_1}\left(1 + \frac{\gamma}{K'}\right)$$

$\therefore$ Contaminant left in solution, $Wx_{w_1} = \frac{Wx_{w_0}}{\left(1 + \frac{\gamma}{K'}\right)}$

On 2nd extraction $Wx_{w_1} = Wx_{w_2} + S'x_{s_2} \qquad \ldots\ldots (2)$

$$= Wx_{w_2} + \frac{S'}{K'}x_{w_2}$$

$$= Wx_{w_2}\left(1 + \frac{\gamma}{K'}\right)$$

$\therefore$ Contaminant left in solution, $Wx_{w_2} = \frac{Wx_{w_1}}{\left(1 + \frac{\gamma}{K'}\right)} = \frac{Wx_{w_0}}{\left(1 + \frac{\gamma}{K'}\right)^2}$

Hence, Contaminant left in solution after 'n' extractions:

$$Wx_{wn} = \frac{Wx_{wo}}{(1 + \frac{\gamma}{K'})^n} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots(3)$$

From (1) $$x_{w_1} = \frac{Wx_{wo} - S'x_{s_1}}{W} = x_{wo} - \gamma x_{s_1}$$

$$\therefore \quad K_1' = \frac{x_{w_1}}{x_{s_1}} - = \frac{x_{wo}}{x_{s_1}} - \gamma \quad \ldots\ldots\ldots\ldots\ldots\ldots(4)$$

From (2) $$x_{w2} = \frac{Wx_{w_1} - S'x_{s2}}{W} = x_{w_1} - \gamma x_{s2}$$

$$\therefore \quad K_2' = \frac{x_{w2}}{x_{s2}} = \frac{x_{w_1}}{x_{s2}} - \gamma$$

but $$x_{w_1} = \frac{x_{wo}}{(1+\frac{\gamma}{K'})}$$

$$\therefore \quad K_2' = \frac{x_{wo}}{x_{s2}(1+\frac{\gamma}{K'})} - \gamma \quad \ldots\ldots\ldots\ldots\ldots\ldots(5)$$

For each extraction S = 10.0g and W = 1000g

Contaminant 1st extraction = 0.22g / 100g P.G. / 10g sieve

$$\therefore \quad x_{s_1} = 0.22$$

Contaminant 2nd extraction = 0.04g / 100g P.G. / 10g sieve

$$\therefore \quad x_{s_2} = 0.004$$

Now $Wx_{wo} \simeq Sxs_1 + Sxs_2 + Sxs_3 + \cdots$

$$x_{wo} \simeq \frac{S}{W}(0.022 + 0.004)$$

$$= 0.00026$$

$$Y = \frac{S}{W} = 0.01$$

$$\therefore \quad K'_1 = \frac{0.00026}{0.022} - 0.01 \quad \text{.....from (4)}$$

$$= 0.0118 - 0.01$$

$$= 0.0018$$

$$K'_2 = \frac{0.00026}{0.004\left(1+\frac{0.01}{0.0018}\right)} - 0.01 \quad \text{.....from (5)}$$

$$= 0.0093 - 0.01$$

This gives a negative K'_2 which is impossible, however if x_{s_2} is taken to be 0.003, which is within experimental error.

Then $$K'_2 = \frac{0.00025}{0.003\left(1 + \frac{0.01}{0.0018}\right)} - 0.01$$

$$= 0.0127 - 0.01$$

$$= 0.0027$$

However K'_1 should equal K'_2, and K'_2 is of the expected order with the accuracy of this experiment and the value of 0.00018 will be used in thecalculations. Hence in (3):

After 6 extractions $x_{w_6} = \frac{0.00026}{\left(1 + \frac{0.01}{0.0018}\right)^6}$

$$= \frac{0.00026}{79000}$$

$$= 0.00000000329 \text{ g / g solution}$$

$\therefore$ Contaminant in P.G. = 0.0000000329 g / g P.G.

or $\underline{\underline{0.0329 \text{ p.p.m. P.G.}}}$

after 6 successive extractions.

NOMENCLATURE

SYMBOL	MEANING	UNITS
A	Total surface area of crystals.	cm^2
$\bar{A}$	Mean surface area of crystals over a time interval.	cm^2
A'	Arrhenius equation constant.	
A_c	The Coulter Counter orifice area normal to the flow axis.	cm^2
a	The projected area, parallel to the orifice axis, of the particle as it is orientated in passing through the Coulter Counter orifice.	cm^2
a_p	The surface area of a crystal.	cm^2
a'	Molecular spacing of the adsorbed layer on the crystal surface.	
B	Calibration constant in $c = F + B.S. + G.S.^2$ or Constant $= \frac{2}{\sqrt{A_c}}$	
b	Index in equation $g = k_L s^b$	
C	A general constant.	
c	Concentration % Mass/Volume (fraction) (= m/v)	$g/(100cm)^3$
c_∞	Concentration % Mass/Volume (fraction) at Equilibrium.	$g/(100cm)^3$
Δc	$c = c - c_\infty$	
D	Equivalent Spherical Diameter of Crystal.	micron or cm
D'	The Coulter Counter orifice diameter	micron
D_c	Indicated Coulter Counter Equivalent spherical diameter.	micron
D_L	Diffusion coefficient.	cm^2/s
D_m	Molar Diffusivity.	mole/cm.s
d	Distance between layers of a double layer condenser.	

SYMBOL	MEANING	UNITS
E	Activation energy for growth.	Kcal/g mol
e	Crystalliser bed voidage.	
e'	Apparent expansion of Hg in glass = 0.000156	
F	Calibration constant in $c = F + B.S. + G.S.^2$ or Constant = $\frac{v_c}{0.707 A_c^{\frac{2}{2}}}$	
F_c	Coulter Counter Scale expansion factor.	
G	Calibration constant in $c = F + B.S. + G.S.^2$	
ΔG	Excess free energy between a particle and the solute in solution.	
ΔG^{*}	Free energy of formation of a nucleus of the critical size r^{*}	
$\Delta G^{*'}$	Free energy of formation of the critical nucleus for heterogeneous nucleation.	
ΔG_d	Free energy of activation of diffusion.	
ΔG_S	Surface excess free energy between the surface of the particle and the bulk of the particle.	
ΔG_V	Excess free energy per unit volume between a very large particle and the solute in solution.	
g	Crystal growth velocity $(= \frac{dr}{dt})$	cm/min
H	Initial crystal seed mass.	g
H'	Heat of adsorption	cal
h	The side of a particle element, (Coulter Counter Theory)	cm
I	Integer counter for the time dimension (in Computer Program 2).	
I_c	The Coulter Counter aperture current setting.	

SYMBOL	MEANING	UNITS
I_c^*	The Coulter Counter aperture current setting used in calibration.	
J	Integer Counter for size distribution dimension. in Computer Programs 1 and 2.	
J'	Growth flux.	$mol/cm^2 s$
j	Rate of nucleation.	$1/cm^3 s$
K	Growth rate constant in equation $\frac{dc}{dt} = -KA(c - c_\infty)$	$1/cm^2.min$
K'	Contaminant Partition coefficient $(= \frac{x_w}{x_s})$	
$\bar{K}$	Average value of K.	$1/cm^2.min$
K_c	The Coulter Counter calibration factor.	
K_d	Coefficient of Mass transfer by diffusion.	
K_L	Growth rate constant (linear basis) where $\frac{dr}{dt} = K_L(c - c_\infty)$	$cm/min(g/100cm^3)$
K_M	Growth rate constant (mass basis) where $\frac{dm}{dt} = -K_M A(c - c_\infty)$	$g/min cm^2 (g/100cm^3)$
K_r	A rate constant for the surface integration step.	
k_B	Boltzman constant = 1.3805×10^{-16}	ergs/degK mol
k_L	Growth rate constant where $g = k_L s^b$	
L	Characteristic crystal dimension of the P.E. pyramid base.	cm or micron
L'	Length of emergent Hg column of thermometer expressed in degrees.	
l	Half the particle length as it is orientated in the axis of the Coulter Counter orifice.	cm
M	Crystal mass.	g
M_m	Mean molecular weight of the solution.	
m	Mass of solute in solution.	g

SYMBOL	MEANING	UNITS
$No.$	Number of crystals of a particular size D.	
N	Number of time readings of Experimental Data.	
n_c	The Coulter Counter corrected count.	
n_c'	The average of counts of a particular sample using the Coulter Counter.	
$\bar{n}_c'$	The average number of counts on the Coulter Counter, n_c' of each sample.	
n_c''	The Coulter Counter coincidence correction.	
n_D	Refractive Index for mean sodium D lines.	
$n_{D\infty}$	Refractive Index for mean sodium D lines at equilibrium.	
P	Number of size readings of size distribution.	
ΔP	Pressure difference in a droplet.	
P_c	The Coulter Counter coincidence factor.	
Q	Defined by:- $Q = \frac{M(I+1) - M(I)}{1.396 \times (\frac{11}{21})}$ (Computer Program 2).	cm^3
q	The electric charge on a particle.	
R	Defined by:- $R = \sum 3 \times (No.) \times D$ at any particular time.	
R'	Universal Gas constant.	$cal/(g.mole)^oK$
R_o	The Coulter Counter aperture resistance.	ohms
ΔR_o	The charge in the Coulter Counter aperture resistance produced by the particle.	ohms
Re	Reynold's Number.	
r	Equivalent spherical volume radius.	cm

SYMBOL	MEANING	UNITS
r_c	The resistance of the Coulter Counter aperture current switch in the position used.	ohms
S	Refractometer scale used in calibration (Equivalent to zeroed prism 1B + 1.20)	
S'	Mass of Molecular Sieve.	g
s	Supersaturation $\frac{c - c_\infty}{c_\infty}$	
T	Absolute temperature.	^oK
T_o	Observed temperature	^oC
T_s	Mean temperature of thermometer emergent stem.	^oC
t	Time	min
t_c	The Coulter Counter relative particle volume ($=t_c'F_c$)	
t_o'	The Coulter Counter threshold level.	
t_c^*	The Coulter Counter threshold value found for the monosized particles used in the calibration.	
U	Σ(No.) at any particular time.	
u	Relative crystal / solution velocity.	
u_t	Terminal Falling Velocity.	cm/s
V	Volume of solvent.	cm^3
V'	The Coulter Counter metering manometer volume.	$10^{-6} dm^3$
V_c	The voltage between the outer electrode of the Coulter Counter and the earth, when immersed in the electrolyte.	Volts
V_m	Molar Volume of Solute.	
v	Total volume of Solution.	cm^3
v_c	Indicated Coulter Counter Particle Volume.	cm^3
v_p	Volume of particle.	cm^3

SYMBOL	MEANING	UNITS
W	Mass of solution	g
w	Work required to form a droplet from its vapour.	
X	% Mass Ratio.	g/100g
x	% Mass Fraction.	g/100g
x_s	Mass fraction of contaminant in sieve.	
x_w	Mass fraction of contaminant in solution.	
Y	Defined by;- $Y = \Sigma 3 \times (\text{No.}) D^2$ at any particular time (Computer Program 2)	cm^2
Y	Defined by:- $Y = \ln \frac{1 + \frac{L}{\sqrt{A_o}}}{1 - \frac{L}{\sqrt{A_o}}}$ in Computer Program 3	cm
y	Correction to be added to T_o to correct partially immersed thermometer.	^oC
y_o	Distance apart of ledges on crystal surface.	
Z	Dummy integer (Computer Programs 1 and 2).	
z	The height of the crystal pyramid apex above the projected side of an element.	
z_s	Average diffusion distance of adsorbed molecules.	
$\surd$	Background count of blank electrolyte.	
α	Angle of contact between crystalline deposit and a foreign solid surface.	
β	The factor by which G^{II} for homogeneous nucleation is greater than that for heterogeneous nucleation.	
β'	A reflection coefficient.	
γ	Ratio of Molecular Sieve to Solution Masses $(\frac{S'}{W})$	

SYMBOL	MEANING	UNITS
δ	Film thickness surrounding a crystal.	cm.
θ	Area Shape Factor ($= \frac{a_p}{\pi L^2}$)	
μ	Solution viscosity.	Poise
∇	The energy of attachment of a molecule.	
ρ	Density.	g/cm^3
ρ_s	Density of Solute.	g/cm^3
ρ_w	Density of Solvent.	g/cm^3
σ	Surface energy per unit area of crystal.	
ϕ	Volume shape factor ($= \frac{6v_p}{\pi L^3}$)	
ψ	Dielectric constant.	
Ω	The particle resistivity.	$microhms/cm^3$
Ω_o	The electrolyte resistivity used for the Coulter Counter.	$microhms/cm^3$

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