DROPLET BREAK-UP AND COALESCENCE IN ROTARY AGITATED COLUMN EXTRACTORS.

by

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June, 1970.

SUMMARY

The systematic design of agitated continuous countercurrent extractors requires an understanding of the influence of contactor variables, viz. rotor speed, geometry and volumetric throughput, on droplet size distributions, residence times and interactions. Therefore the literature pertaining to droplet break-up and coalescence, droplet phenomena in agitated systems and the design, characteristics and uses of agitated columns has been critically examined and the important phenomena studied experimentally.

The most common commercial columns are the Rotating Disc Contactor, the Scheibel Column and the Oldshue-Rushton Column. A column section having interchangeable internal components was constructed and used to study droplet hydrodynamics in these three designs. Systems with a range of interfacial tension from 9.7 to 55 dynes per cm. were employed in the absence of mass transfer. High and normal speed cine, high speed still photography and the injection of organic soluble dye were among the techniques used.

The manner in which capacity and hold-up vary with operating parameters was confirmed. Interdroplet coalescence was shown to be largely absent in the R.D.C. A critical speed was established and consideration was given to the applicability of drop size correlations above and below this speed. At low speeds an approximate form of correlation is,

$$d_{12} = k \left(\frac{\sigma}{\Delta\rho_{\cdot} g}\right)^{0.5}$$

where k is approximately 0.55.

Mathematical models have been proposed for break-up in the latter case and for the case of a dispersed phase wetted disc.

Interdroplet coalescence has been found to predominate in the Scheibel column but, due to globule formation from the packing, mean interfacial area is unpredictable.

Above a definable critical speed, a coalescence-redispersion mechanism has been found in the Oldshue-Rushton column; solute transfer rates should therefore approximate to those for fully circulating droplets. At reduced speeds, operation was acollisive and the form of drop size correlation approximates to that for the R.D.C. with k approximately equal to 0.34 or, at very low speeds, 0.57. For normal operation, the proposed form is,

$$d_{vs} = d_{vs}^{o} + mX$$

Effects arising from dispersed phase wetting of column rotors, stators or packing were determined. Reliable estimation of efficiency or capacity was found to be impracticable.

Conclusions have been drawn regarding the potential uses and practical limitations of the designs and current design procedures. Two illustrative cine films have been compiled and further work has been initiated in several of the areas recommended.

ACKNOWLEDGEMENTS.

The author wishes to thank the following:-

Professor G. V. Jeffreys,
for his encouragement and suggestions while supervising this work.

Technical Staff in the Department of Chemical Engineering,
for their assistance in fabricating equipment and developing films.

Mr. A. Jones, Visual Aids Department,
for his skill in printing all the photographs referred to.

Dr. S. Z. Sterbaceck,
for translation of reference 46 whilst Visiting Professor in the Department.

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1. INTRODUCTION

In recent years advances in equipment design have facilitated the increased commercial application of liquid-liquid extraction. The characteristics, applications and developments in the design of the various newer types of contactor have been considered by the author elsewhere (1). This paper is presented in Appendix 1.1.

Continuous countercurrent extractors in which the concentration changes continuously along the length are widely used. In this type of extractor phase flows are assisted by gravity. In extractors of the spray or packed tower design, large H.E.T.S. or H.T.U. values are associated with systems of high interfacial tension or with viscous phases. Therefore, rotary agitated columns are favoured.

Operation comprises the dispersion of one liquid as droplets into a continuous liquid phase, solute transfer to or from the droplets and finally separation of the dispersed phase by flocculation and coalescence. Droplet size is an important factor affecting the mode of mass transfer and, in association with dispersed phase hold-up, determines the interfacial area available. Volumetric capacity is also dependant on the droplet size distribution. Therefore fundamental design should be based on droplet considerations.

In unagitated columns at low hold-up the behaviour of swarms of drops can be represented by the behaviour of single droplets in discrete droplet contacting devices. Spray and sieve plate columns have been designed on this basis (2). For convenience allowance may be made for the effects of axial mixing by means of a single eddy diffusivity (3). The assumptions made are,

a. Uniform spherical drop size

b. Negligible drop interaction.

In rotary agitated columns these assumptions may be invalidated. In addition if droplet coalescence and redispersion occurs the application of empirical formulae to allow for axial mixing is complicated. These phenomena are dependant on the column design and operating conditions. The significance of observed droplet phenomena and the limitations imposed on established design methods and analysis have been reviewed in a paper which forms Appendix 1.2. (4).

Systematic design cannot be effected until more information on the influence of contactor variables on drop size distribution, residence times, interactions and distortions is available for analysis, with reference to their effect on solute transfer efficiency and volumetric capacity. Therefore in this work a research study into droplet characteristics in a pilot scale section of a rotary agitated extractor having variable internal components is described. Experiments were carried out in saturated two component systems where no mass transfer occurred, that is under conditions excluding the special interfacial effects which arise from the presence of an unequally distributed solute (5,6). Some results of this investigation have been given in a recent paper which forms Appendix 1.3. (7).

2. COLUMN DESIGNS

The following rotary agitated columns have acheived widespread commercial application,

(1), 1. The Scheibel column

2. The Rotating Disc Contactor

3. The Oldshue-Rushton column.

2.1. ORIGINAL DESIGNS

2.1.1. The Scheibel Column (8)

The Scheibel Column comprises an alternate sequence of mixing sections and packed sections in a vertical column. Each mixing compartment is agitated by a paddle, or turbine, supported on a central shaft. The packed sections contain open-knitted mesh packings. A typical design is shown in Figure 1.

The column has been described in several publications as a mixer-settler which is incorrect since droplets persist throughout the packed sections. The primary function of the packing is to isolate the mixing

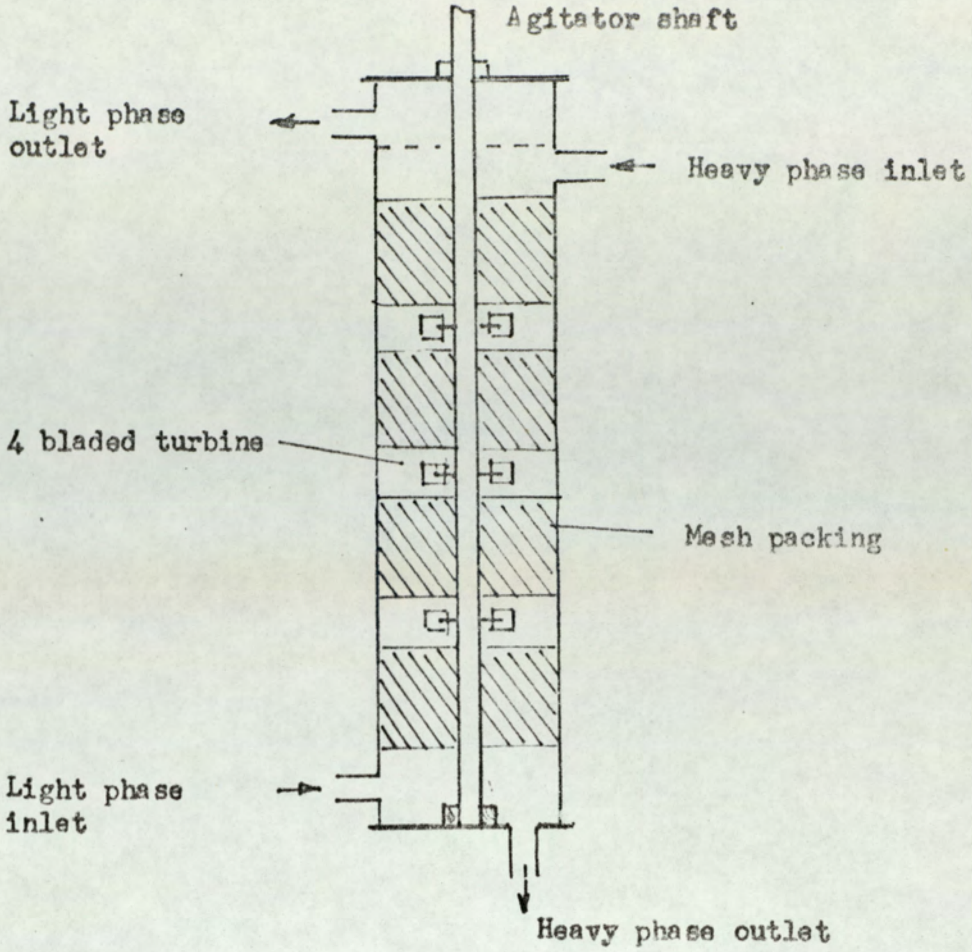


FIGURE 1. ORIGINAL SCHEIBEL COLUMN (8)

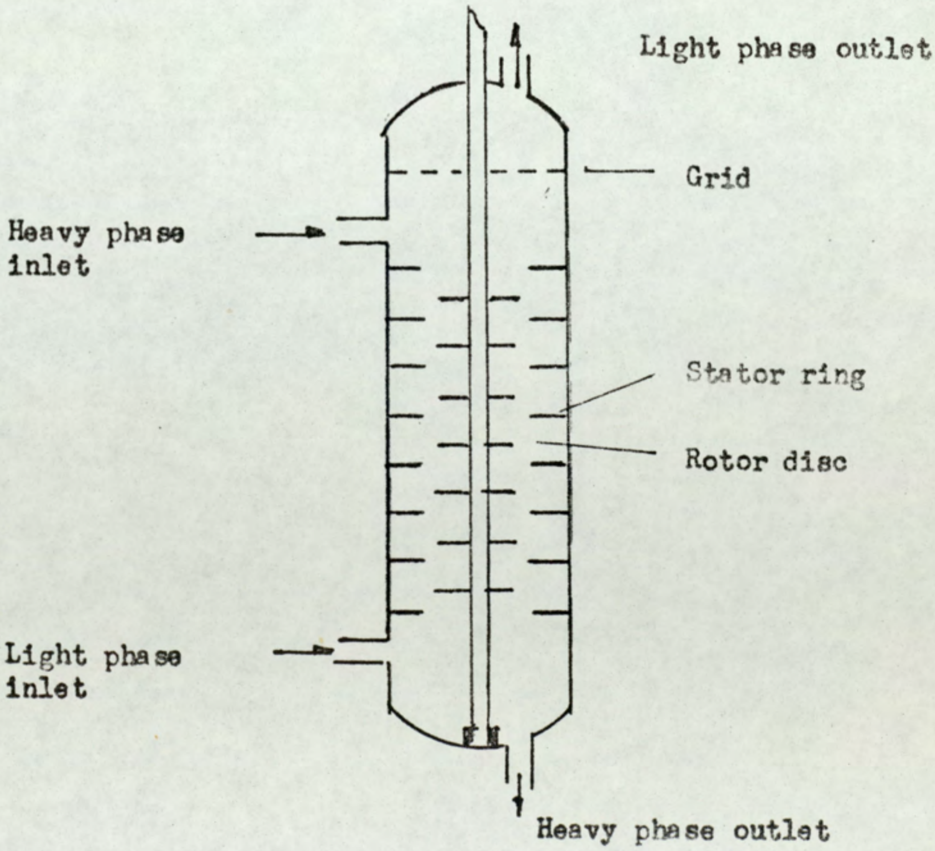


FIGURE 2. ROTATING DISC CONTACTOR (13)

sections not to act as a coalescence chamber (9). Therefore the packing material is selected so as to be preferentially wetted by the continuous phase; for example, an early polythene column had silver packing (10).

The relative heights of packed and mixing sections may be varied to suit prevailing circumstances (11). The standard range of pilot scale contactors have packing and mixing section heights which increase on a pro-rata basis with column diameter (12).

2.1.2. The Rotating Disc Contactor (13,14)

The contactor consists of a vertical cylindrical shell divided into a number of compartments by a series of stator rings. A rotating disc, supported on a rotating shaft is centred in each compartment. Adjacent to the top and bottom compartments are feed ports which are arranged tangentially in the direction of rotation. The dense phase is introduced into the top of the column and the light phase into the bottom; countercurrent flow of the phases is thus assisted by gravity. One of the phases is dispersed by the action of the rotating discs. A typical design is shown in Figure 2.

The contactor is in fact an improved form of an extractor described in a notable patent by Coahran over 20 years earlier (15).

Flat rotor discs and stator rings without any protrusion are used to create uniform shearing conditions and hence obtain as small a spread in droplet sizes as possible. Reman (16) suggested that the flow pattern consists of a rotation of the whole mass of liquid and that superimposed on this rotation there is a slower movement of liquid from the shaft towards the wall of the contactor in the vicinity of the discs and from the wall back towards the shaft in the vicinity of the stator rings. The resulting flow of liquid is thus, in theory, toroidal as illustrated in Figure 3.

Reference has been made elsewhere (1) to the extensive commercial application of this design.

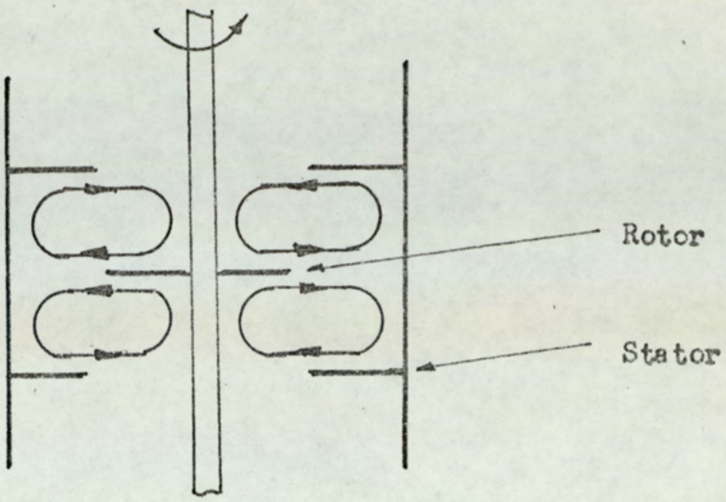


FIGURE 3. IDEALISED FLOW PATTERN IN A ROTATING DISC CONTACTOR (16)

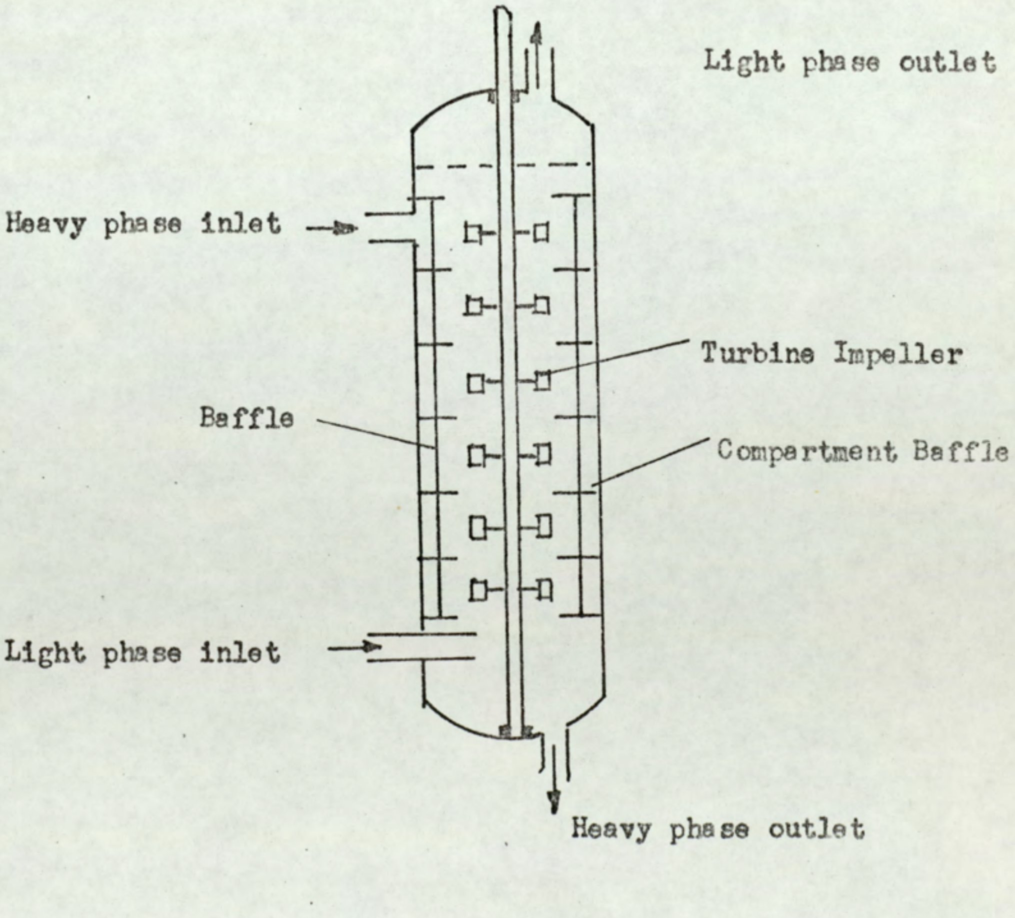


FIGURE 4. OLDSHUE-RUSHTON EXTRACTOR (17)

2.1.3. The Oldshue-Rushton Extractor

The Oldshue-Rushton extractor (17,18) is a natural development from the use of a mixing vessel as an extractor. It consists of a vertical column divided into compartments by horizontal stator rings. Each compartment has four vertical baffles arranged radially in the manner used in mixing vessels. Agitation is provided by a turbine impeller driven by a central shaft. A typical design is shown in Figure 4.

2.2. MODIFIED DESIGNS

Numerous modified designs have been proposed. The aim of the more noteworthy modifications, summarised below, is to reduce the extent of axial mixing and so give an increased driving potential for mass transfer and hence a smaller contactor for a given mass transfer coefficent. Future developments will follow a similar line (19).

2.2.1. The Scheibel Column

A later design of the Scheibel column has become established for sizes greater than 1 ft. diameter (20,12). As shown in Figure 5 this design has ring shaped baffles supported immediately above and below the impeller and stator rings attached to the column shell. For column diametersbelow 3 ft. it is commercial practice with low interfacial tension and low viscosity systems to alternate the agitated sections with the usual woven mesh sections (11). For systems of high interfacial tension and viscosity the mesh sections may be omitted.

The purpose of the internal baffling is to provide uniform mixing of the phases and thus eliminate the tendency for H.E.T.S. to increase with increasing column diameter.

With this later design Scheibel (20) used knitted mesh sections inserted between the inner annular plates to remove the rotational energy previously dissipated in the mesh sections. This served also to improve phase contacting and in fact correlated with 'fully baffled' conditions. A similar effect could be acheived by installing vertical baffles around the column wall or between the two inner annular baffles at the discharge

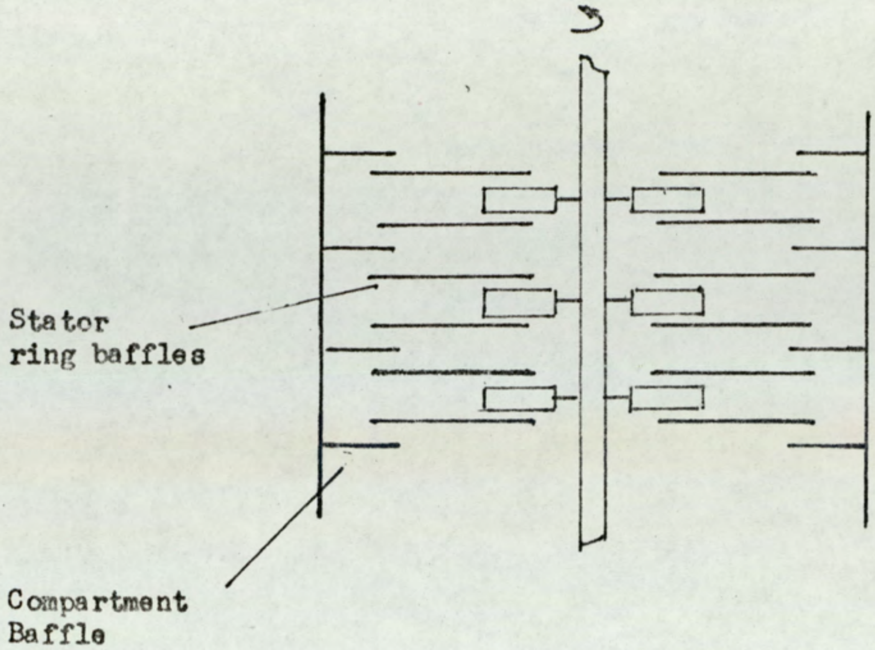


FIGURE 5. MODIFIED SCHEIBEL DESIGN (20)

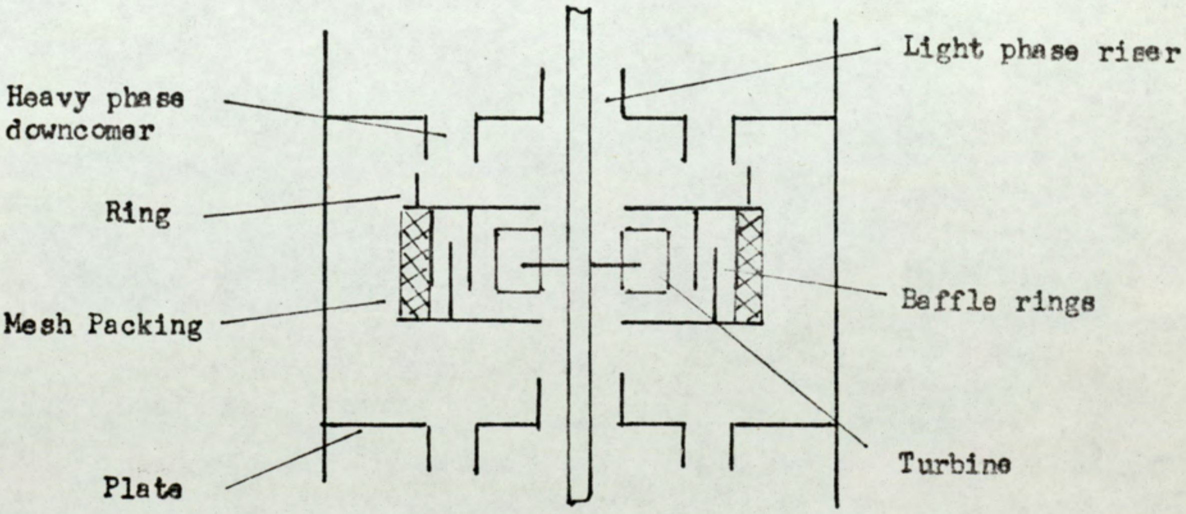


FIGURE 6. ONE STAGE IN A WIRZ EXTRACTOR (21)

of the impeller.

A similar design has been described by Leisibach (21). As shown in Figure 6, the calming baffles enclosing the impeller each have short connections for conveying the phases from one zone to the next. The heavy phase flows through the downcomers to the zone beneath whilst the light phase flows upward through the annular connection at the points where the agitator shaft passes through the baffles. For the system propionic acid/toluene/water in 105 mm, 285 mm. and 500 mm. diameter columns Leisibach obtained H.E.T.S. values of 0.1 to 0.2 metres. These were less dependent on column diameter than in conventional designs. The internal design is however complex and therefore expensive (22) and the column has yet to achieve commercial application.

A variation of the original Scheibel design in which the packed sections were replaced by baffled calming sections has been tested by Tudose (23). This is similar to an earlier design in which the packed sections were replaced by perforated plates (24). The Tudose design is shown in Figure 7. The mixing sections have distribution discs occupying the column cross-section. These are pierced to form orifices and are fitted with blades bent alternately up and down in the direction of rotation so that as the disc rotates it forces liquid either up or down through the orifices. The regions between the discs are provided with substantial vertical baffles which approach closely to the rotating shaft. From experiments in a 3 inch column only, an improved efficiency has been claimed over an R.D.C.

2.2.2. The R.D.C.

A modification of the R.D.C. by Misek (25,26) has the agitated compartments placed off-centre. Next to the agitated zone is a transfer section separated from the agitated zone by a baffle that permits flow between the zones only through passages near the periphery. A set of horizontal settling-zone baffles may be positioned at any height with respect to the firm position of the set of mixing zone baffles (27).

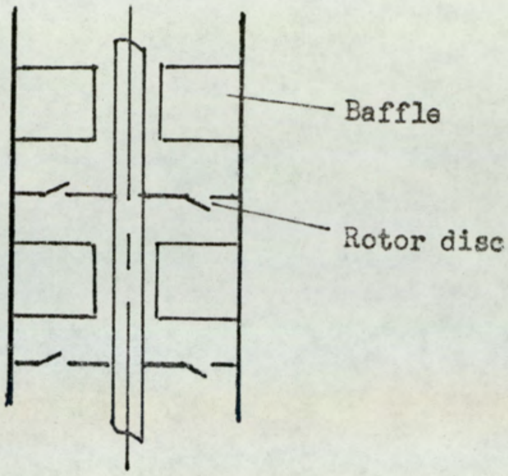


FIGURE 7. TUDOSE COLUMN (23)

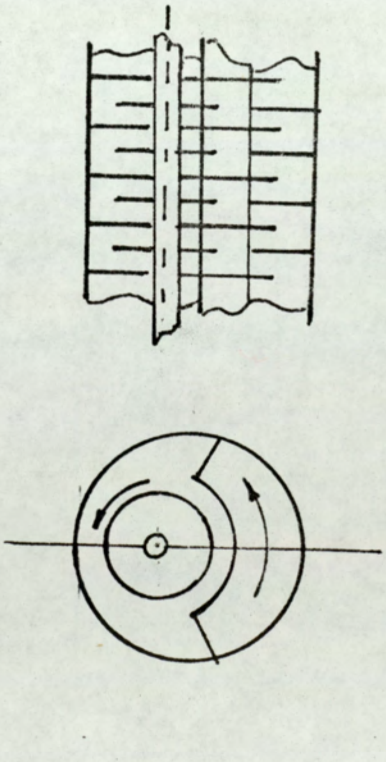


FIGURE 8. ASSYMETRIC R.D.C. (25)

It is claimed to inhibit unfavourable flow behaviour such as axial mixing. Nevertheless in scaling up from 1 m. diameter to 2 m. diameter it is necessary to allow for an increase in the 'nominal H.E.T.S.' from 1.2 m. to 1.75 m. Some loss in efficiency probably occurs due to phase entrainment in the settling zones. The design appears therefore to offer no practical advantage over the conventional R.D.C. considering the increased column diameter required. An approximate relation for the estimation of extractor capacity has been published (27). One known application is in caprolactam extraction but several are claimed to be in operation in typical R.D.C. applications (26).

A modified design patented by Reman (28) has two or more rotors in the column shell. This is claimed to minimise instability of the vortex flow pattern in large diameter columns. There is no record of its industrial use nor of a design having ring shaped baffles above and below the rotor discs after the style of the Scheibel section shown in Figure 5. (29).

A design employing perforated disc rotors has been tested on a pilot scale (30). Experiments with one system gave improved mass transfer efficiencies, as shown by a reduction of $(H.T.U)_{OR}$ values by 20 - 30%, compared with a conventional R.D.C. Column capacity was however significantly reduced. These results are in accordance with the smaller average drop sizes observed. Under the specific conditions of operation the increased interfacial area presumably outweighed the effects of back-mixing. No data has yet been obtained on the back-mixing characteristics of this design (31) but these could be critical. Similar results might be expected from using one of the new serrated edge discs (32).

Sokov et. al. (33) have employed a modified R.D.C. in which the stator rings were omitted and the discs were of two sizes. The larger discs were perforated. For the extraction of aromatics, this design is claimed to give both a fourfold increase in capacity and improved efficiencies at low rotor speeds compared with the R.D.C.

A design has been tested in which the discs were replaced by rotors comprising 4, 60 arcs (34). A higher mass transfer coefficient was obtained in experiments with the system benzene-acetone-water. Finally a patent has been issued for a design in which annular ring stators are alternated with centrally located stator discs and the rotor comprises a rotating cylinder (35). This is, in effect, a combination of the older laboratory rotary annular extractor and a disc-and-doughnut plate column (11).

2.2.3. Oldshue-Rushton Column

A form of the Oldshue-Rushton column has been described in which the impeller is arranged eccentrically and small pipes are provided for the interstage flow (36). As would be expected, this design significantly reduced the amount of back-mixing but at the expense of volumetric capacity.

A recent investigation has been reported in which a central draft tube was introduced in an attempt to reduce back-mixing. Though satisfactory for single phase studies this modification offered no improvement in practice (37).

Finally a contactor has been patented wherein agitation is provided by a perforated vertical baffle oscillating about a vertical axis. With one system, carbon tetrachloride - acetic acid - water, H.T.U.'s obtained with this design were claimed to compare favourably with those in an R.D.C. (38).

3. HYDRODYNAMICS OF DROPLET BREAK-UP

Mass transfer in extraction columns occurs between a dispersion of one liquid as droplets and a partially miscible continuous phase. Therefore prediction of performance necessitates an understanding of the mechanism of droplet break-up and coalescence.

3.1. FUNDAMENTAL THEORY

Globules in a continuous phase may split up due to hydrodynamic forces in any of the following ways,

Type 1. Lenticular deformation. The globule is flattened into an oblate ellipsoid and finally reduced to a large number of fine droplets.

Type 2. Cigar shaped deformation. The globule extends into a prolate ellipsoid and then into a long cylindrical thread which ultimately breaks up into droplets.

Type 3. Bulgy-break-up. The surface of the globule is deformed in various places and droplets separate from the protruberances created.

Hinze identified the various local flow patterns producing these deformations, assuming they are large compared with the globule size, as reproduced in Table 1. (39). This differentiates between deformation by external pressures and that by viscous stresses.

Break-up occurs when there is a sufficient degree of deformation and if the flow pattern persists for a sufficient length of time. Naturally it is dependant on the physical properties of the system, viz interfacial tension, viscosity and density.

In liquid-liquid dispersions the most probable condition causing break-up under turbulent conditions is A.2d but A.1c A.2b and A.3f can occur. In viscous flows condition B.2d predominates but B.2b is also possible.

TABLE 1. POSSIBLE CONDITIONS FOR GLOBULE BREAK-UP. (39)

Type of Deformation	A. Dynamic pressures	B. Viscous stresses
	Flow pattern around the globule	
1. Lenticular	(a) Parallel	(a) Parallel
	(c) Axisymmetric hyperbolic	(c) Axisymmetric hyperbolic
	(e) Rotating	(e) Rotating
2. Cigar-shaped	(b) Plane hyperbolic	(b) Plane hyperbolic
	(d) Couette	(d) Couette
3. Bulgy	(f) irregular	

Hinze considered an isolated globule being deformed by the action of external forces. The force per unit area τ may be a viscous stress or a dynamic pressure in the surrounding continuous phase. τ is a function of time and will vary along the surface. Deformation is counteracted by a surface force arising from interfacial tension σ; this force is of magnitude σ/d. Dynamic pressure τ will create flow velocities of magnitude $(\tau/\rho_D)^{\frac{1}{2}}$ causing a viscous stress $\frac{\mu_D}{d}(\tau/\rho_D)^{\frac{1}{2}}$.

Therefore the forces controlling droplet break-up are,

$$\tau, \quad \frac{\sigma}{d} \quad \text{and} \quad \frac{\mu_D}{d}(\tau/\rho_D)^{\frac{1}{2}} .$$

From the ratios between each two of these Hinze selected two dimensionless groups:-

$$N_{vi} = \frac{\mu_D}{\sqrt{\rho_D \sigma . d}} \quad , \quad \text{a viscosity group}$$

$$N_{We} = \frac{\tau . d .}{\sigma} \quad , \quad \text{a generalised Weber number}$$

(c.f. the accepted definition $\frac{\rho v^2 d}{\sigma}$). Clearly deformation increases with increasing N_{we} and at a critical value $(N_{we})_{crit.}$ break-up occurs.

3.2. BREAK-UP IN AGITATED SYSTEMS

Hinze made a theoretical study of droplet break-up in dilute liquid-liquid systems, i.e. a non-coalescing environment (39).

For droplet break-up to result from viscous shearing action the deformed drop must be small compared with the local region of viscous flow; Hinze considered that the maximum drop sizes observed in liquid-liquid dispersions invalidated this. Hence the size of the largest drops is determined by the dynamic pressure of the turbulence. These forces are caused by changes in velocity over distances equal at the most to the drop diameter. Assuming these fluctuations cause break-up,

$$(N_{we})_{crit.} = \frac{\rho . \overline{V}^2 . d_{max}}{\sigma} \qquad 1.$$

where $\overline{V}^2$ is the average value across the whole flow field of the squares of the velocity differences over a distance d_{max}.

Hinze considered the case of a homogeneous isotropic turbulence after Kolmogoroff (40). This condition has been summarised as follows (41). In a turbulence, instability of the main flow amplifies existing disturbances and produces primary eddies of similar wavelength to the main flow. These primary eddies disintegrate into successively smaller eddies until all their energy is dissipated by viscous flow. At high Reynold's numbers for the main flow the large eddies contain nearly all the kinetic energy but the majority of dissipation occurs in the smallest eddies. If the scale of the main flow is large relative to that of the energy dissipating eddies a wide spectrum of intermediate eddies exist; these contain and therefore dissipate little of the total energy. Such eddies transfer kinetic energy from the large to small eddies and in so doing the directional information of the large eddies is lost. Kolmogoroff concluded that the main flow is independant of the statistical properties of any oscillation on a smaller scale. Therefore the statistical properties of small eddies can be characterised by the local energy dissipation rate per unit mass E.

Isotropic turbulence is defined by,

$$\bar{v}^2 = \bar{u}^2 = \bar{w}^2$$

where $\bar{v}^2$, $\bar{u}^2$ and $\bar{w}^2$ are the root mean squares of the velocity vectors.

Although most practical mixing applications are unisotropic Kolmogoroff assumed local isotropy if the volume under consideration is small compared to the scale of the main flow L. Then if r_1, r_2 are two points in this small volume and r is the radius vector $r_1 r_2$, a relative velocity u(r) may be defined by,

$$\overline{u(r)^2} = \overline{u(r_1)^2 - u(r_2)^2} \qquad 2.$$

$\overline{u^2 r}$ is determined mainly by small eddies which are independant of the main flow. Therefore for $r \ll L$, $\overline{u^2(r)}$ is mainly a function of E and γ the kinematic viscosity of the main fluid which is independant of the main flow; $\overline{u^2(r)}$ is also independant of the direction of the radius vector r for small values of r only.

When $r \gg \eta$, the microscale of the turbulence, $\overline{u^2 r}$ is independant of the viscosity γ and is a function of E only. Kolmogoroff employed dimensional analysis to obtain the relationships,

$$L \gg r \gg \eta \; ; \quad \overline{u^2(r)} = C_1 E^{\frac{2}{3}} r^{\frac{2}{3}} \qquad 3.$$

$$r \ll \eta \; ; \quad \overline{u^2 r} = C_2 \frac{E}{\gamma} . r^2 \qquad 4.$$

(where no longer independant of γ)

C_1 and C_2 are measurable, universal constants.

Hinze therefore by assuming that the fluctuations are in the region of wavelengths where the Kolmogoroff energy distribution law is valid obtained from equations 1 and 4.

$$\frac{\rho_c . d_{max}}{\sigma} . C_1 \ (E . d_{max})^{\frac{2}{3}} = \text{const.} \qquad 5.$$

or

$$d_{max} \left(\frac{\rho_c}{\sigma} \right)^{\frac{3}{5}} . E^{\frac{2}{5}} = C_1 \qquad 6.$$

Practical application of Kolmogoroff's theory and hence Equation 6 presupposes.

(a) A high Reynolds number for the main flow,
$Re \longrightarrow \infty$ or a least $> 50,000$.

(b) $L \gg \eta$ where L is related to the width of the agitator blades.

Both conditions apply frequently to processes in agitated dispersion vessels (41). Based on an analysis of the rotating cylinder data of Clay (42), Hinze has reported a value for C of approximately 0.72.

Shinnar and Church derived a similar equation to Hinze for break-up by turbulent pressure fluctuations (41). Break-up was found to be dependant on Weber number by assuming that for nearly equal densities and viscosities the droplet oscillates with the surrounding fluid. An oscillating drop becomes unstable if the kinetic energy E_k is sufficient to make up the difference in surface energy between itself and the two smaller ones formed by break-up.

$$\frac{E_k}{\sigma d^2} \doteq 0.26 \qquad 7.$$

In agitated dispersions where local isotropy prevails the kinetic energy of a single oscillating drop is expressed by

$$E_k \propto \rho \overline{u^2 (d)}^{\swarrow \text{ND}} . d^3 \qquad 8.$$

Hence from Equations 7 and 8, [illegible]

$$\frac{\rho\, \overline{u^2(d)}.\, d}{\sigma} = W_e \doteq 0.26 \qquad 9.$$

For fully baffled conditions the average energy consumption per unit mass by a turbine agitator is given by (43),

$$E = K.\, N^3.\, R^2$$

Substituting this into Equation 3 gives,

$$\overline{u^2(r)} = K_1\, N^2\, R^{\frac{4}{3}} d^{\frac{2}{3}} \quad \text{for } d \gg \eta \qquad 10.$$

or into Equation 4 gives,

$$\overline{u^2(r)} = \frac{K_2\, N^3\, R^2\, d^2}{\gamma} \quad \text{for } d \ll \eta \qquad 11.$$

Hence for the conditions to which Equation 10 is applicable,

$$W_e = \frac{K_1\, N^2\, R^{\frac{4}{3}} d^{\frac{5}{3}} .\rho .}{\sigma} \qquad 12.$$

The average droplet size has been measured in two geometrically similar tanks with paddle agitators by Vermeulen (44).
The results for various liquids were correlated by,

$$\frac{\rho\, N^2\, R^{\frac{4}{3}} d^{\frac{5}{3}}}{\sigma\, (f_x)^{\frac{5}{3}}} = 0.016 \qquad 13.$$

giving good agreement with the derived equation 12. f_x is an empirically determined function of dispersed phase hold-up allowing for coalescence.

Following Hinze, Misek derived a relationship for the break-up of drops within the turbulent mixing region by a rotating disc (45). It was intended to study R.D.C. hydrodynamics; the influence of the vertical velocities of both liquids on droplet splitting was considered to be negligible.

The additional assumptions made were:

i. That the highest dynamic pressure is exerted near the column wall and is proportional to the square of the continuous phase velocity at the particular point.

ii. The continuous phase liquid velocity in the vicinity of the rotor is proportional to the peripheral speed and diminishes exponentially towards the wall.

Hence,

$$\text{Velocity } V = \frac{V_o}{\exp\,(K_1\ \Delta R)} \qquad 14.$$

Substituting in Equation 1.,

$$W_e = \frac{\rho\, V_o^2 \,.\, d_h}{\sigma\ \exp\ (K_2 \Delta R)} = \frac{\rho\, N^2 R^2\ d_h}{\sigma\ \exp\ (K_2\ \Delta\ R)} \qquad 15.$$

Assuming further that the effect of compartment height can be expressed as a power function,

$$W_e = K_3 \left(\frac{H}{D}\right)^a \qquad 16.$$

Misek evaluated the exponents experimentally and proposed the expression,

$$\frac{d_h\ N^2\ R^2\ \rho c.}{\sigma\ \exp\ (0.0887\ \Delta\ R)} = 16.3 \left(\frac{H}{D}\right)^{0.46} \qquad 17.$$

On diminishing the mixing intensity below a critical value of the Reynold's number,

$$(Re)_{crit} = \left[\frac{R^2\ N\ \rho c}{\mu_c}\right]_{crit} \doteq 6 \times 10^4 \qquad 18.$$

splitting of the drops was deemed to occur in a transitive region. The dependence of drag force on Reynold's number is significant as the viscosity of the continuous phase exerts an influence on the break-up mechanism. Misek claims that the expression,

$$\frac{d_h\ N^2\ R^2\ \rho c}{\sigma\ \exp\ (0.0887\ \Delta\ R)} = 1.345 \times 10^{-6} \left[\frac{R^2\ N\ \rho c}{\mu_c}\right]^{1.42} \qquad 19.$$

is valid in this region (46) which is within the practicable operating range of an R.D.C.

Misek states that a further decrease in mixing intensity into the laminar region leads to a different mechanism (45). The vertical velocity of the drops becomes significant and drops split mainly by successive impacts on the stators and discs. By analogy with the break-up mechanism in packings the drop diameter is determined by the ratio of buoyancy and surface forces. From very limited experimental work, Misek proposed the expression,

$$d = 0.38\left(\frac{\sigma}{\Delta\rho . g.}\right)^{0.5} \qquad 20.$$

for values of Re $\ll 10^4$ (46). In this region mixing intensity is claimed to have a negligible effect on drop size.

3.3. BREAK-UP IN PACKINGS.

Since some agitated columns contain packed sections droplet hydrodynamics in packings are of interest. Studies of the mechanism of droplet breakdown are of additional relevance since under certain conditions analogies may be drawn with break-down in agitated sections(46).

For a given system with packing above a critical size $d_{f.c.}$, this being the size in which the mean void size is equal to the mean droplet size, droplets rise freely within the interstices and are brought to rest periodically by random collisions with packing elements. The energy dissipated may result in droplet breakdown. Therefore droplet movement through the packing results in a gradual reduction in d_{vs} with height (5, 47).

Ramshaw and Thornton (48) have found that droplets at their terminal velocity are broken down by collision with a packing element provided they are above a critical size $d_{cr.}$ This size is defined by the condition that the net gain in surface energy of the system after breakdown is just equal to the sum of the kinetic and potential energies of the parent drop. The approximation of an initially spherical droplet forming two equal sized drops yielded the expression, neglecting friction, :-

$$1.8 \; d_{cr}^{2} \, \Delta\rho . g \; + \; d_{cr} \, V_t^{2} . \, \rho_d \; = \; 3.12 \; \sigma \qquad 21.$$

V_t is the terminal droplet velocity in an infinite medium from drag coefficient measurements. In practice the impact velocity V is a function of hold-up and continuous phase superficial velocity

$$V \; = \; \phi \, V_t \; - \; \frac{V_c}{\varepsilon \, (1 - X)} \qquad 22.$$

where ϕ is a correction factor for a droplet moving in a restricted medium. Values of d_{cr} were dalculated from Equation 21 using impact velocities given by Equation 22. For the system toluene-water in the absence of mass transfer the equilibrium droplet size (d_{vs}) eq. was related to the critical value by

$$(d_{vs})_{eq} \; = \; 0.85 \; d_{cr} \qquad 23.$$

Clearly the numerical constant is specific to the system and the analysis applies to a droplet striking a baffle symmetrically, since when the droplet is offset two unequally sized droplets are produced.

In a bed of randomly orientated packing elements therefore the droplet size distribution is dependant on:

(a) Initial size distribution.

(b) The probability of droplet breakdown and split ratios for symmetrically colliding drops.

(c) The probability of breakdown and split ratios for assymmetrically colliding drops.

(d) The rate of droplet coalescence within the interstices of the packing.

Ramshaw studied the size distribution of toluene droplets in a continuous aqueous phase in a 3" column packed with $\frac{3}{4}$" Raschig rings. The equilibrium drop size (dvs)eq. was approached only after several feet of packing. The change in d_{vs} with column height was represented by an exponential expression of the form

$$d_{vs} = (dvs)_{eq} + 0.27 \exp - (0.0157H). \qquad 24.$$

The equilibrium value of the mean droplet size (dvs)eq was 0.45 cm. for this system.

Over a range of flowrates the size distributions were found to be positively skewed and could be represented approximately by means of a log - normal distribution function.

Lewis et. al. observed that packings less than the critical size d_{fc} produced larger droplets (49). Droplets are apparently arrested in the interstices and coalesce before a sufficiently large head is built up to redisperse the coalesced phase as fresh droplets. This is the accepted theory in the design of coalescers but deviations have been reported (50).

4. DROPLET PHENOMENA

4.1. FORMATION AT ORIFICES

In a study of drop formation at a single orifice Hayworth and Treybal (51) found that for most systems at orifice velocities between zero and 0.33 ft. s^{-1} the drops were of substantially uniform size. Above this velocity the drop-size was non-uniform and its mean value decreased with increasing velocity. Controllable drop size is dependant on the orifice having a sharp chamferred end and the orifice plate not being wetted by the dispersed phase (11, 52, 53). Numerous correlations for the prediction of mean drop sizes from a correctly designed orifice have been reviewed by Jeffreys (54) and will not be considered here. It is of interest to note however that in an agitated column, in the absence of interdrop coalescence, the size of drops formed at the dispersed phase inlet constitutes the upper limit of the size distribution generated by droplet break-up.

4.2. COALESCENCE

The tendency, or otherwise, of a dispersed phase to coalesce in a liquid-liquid extraction column has a marked influence on the performance.

In a spray column coalescence of drops, after formation at the distributor, to form large drops leads to (i) a reduction in the interfacial area and (ii) an increase in the rate of drop movement relative to the continuous phase. In agitated columns a more complex situation exists because of the potential enhancement of mass transfer by continued droplet break-up and coalescence.

It is essential for coalescence to occur at the interface of phase separation near the dispersed phase outlet from a column. If dispersed phase droplets do not coalesce readily at this interface the residence time will be increased and result in reduced capacity.

Whilst droplet coalescence at a plane interface is excluded from this study, it has been the subject of much research (55, 56) and enables deductions to be made regarding droplet-droplet coalescence which is inherently difficult to study experimentally.

Droplet-droplet coalescence takes place in two stages:-

(a) Initially liquid must drain from the region between the elements which are to coalesce until a certain critical film thickness is attained.

(b) The critical film ruptures and coalescence occurs.

The speed of coalescence is therefore dependant on those factors which affect the force impelling the elements towards each other, the resistance to drainage of the liquid film, the critical film thickness and the ultimate strength of the critical film. It is known that the presence of impurities such as grease or dirt in liquid-liquid systems, even as trace amounts, markedly decrease the rest time of drops at an interface i.e. tend to aid droplet-droplet coalescence. When the impurity is surface-active the result is invariably a marked decrease in the speed of coalescence. Linton and Sutherland (57) have found that the coalescence of one drop with another on collision was delayed by the air film trapped between the opposing surfaces which deform on approaching each other. The drops were observed to bounce or coalesce after a time proportional to the cube of the diameter of the smaller drop.

In the presence of mass transfer the ease of coalescence may vary considerably depending on the direction of transfer. Numerous investigators have found that mass transfer tends to aid coalescence when the solute is transferred from the dispersed to the continuous phase but to oppose coalescence when it is in the other direction. This is due to the mechanism proposed by Groothuis and Zuiderweg (58) on the basis of the generation of motion in a fluid interface due to variations in surface tnesion associated with concentration gradients i.e. the Marangoni effect. Some attempt has been made to provide a mathematical treatment based on this mechanism (59). This is however an over-simplified mechanism since coalescence is known to be influenced by interfacial turbulence arising from rapid mass transfer. This may cause premature film rupture as could associated heat effects at the interface.

In a turbulent flow field drops undergoing the drainage process stand a greater chance of being separated. Conversely the energy of impact is greater. Howarth (60) has developed an equation for the frequency of coalescence of uniformly sized spherical drops in a homogeneous isotropic turbulent flow suspension of infinite extent. The frequency of coalescence is considered to depend on the frequency of collision and the fraction of collisions which result in coalescence. In an isotropic turbulence the drops are in random motion and frequent collisions occur between them; when two droplets collide they may rebound, cohere or coalesce. Howarth assumes that most collisions result in either cohesion or immediate coalescence. In this case the average number of collisions suffered by each drop in unit time can be shown to be given by,

$$N' = 4\pi\ n'x.\ D'_d \qquad 25.$$

where D'_d is the coefficient of self diffusion of the drops which cohere when their centres approach to a distance x apart.

Assuming further that,

i. The drops are small compared to the smallest wavelength in the turbulence and their motion relative to the main fluid follows Stoke's Law.

ii. During the motion of each drop its neighbourhood is formed by the same liquid molecules.

iii. Any external forces acting on the drops originate from a potential field. The nature of the turbulence is not affected by the presence of the particles i.e. the dispersion is dilute.

iv. The density of the dispersed phase does not differ greatly from that of the continuous phase,

application of Taylor's theory yields an expression for the time dependance of the diffusion coefficient

$$\varepsilon_p = \varepsilon_f = \bar{v}^2 . \theta \qquad 26.$$

where $\bar{v}^2$ is the mean square Langrangian turbulent velocity fluctuation. The time interval for collision is the diffusion time, hence,

$$N' = (4 \pi n' x \bar{v}^2)^{\frac{1}{2}} \qquad 27.$$

Substituting for the number of drops per unit volume $n' = 6X / \pi d^3$

$$N' = \left[\frac{24 \, Xx \, \bar{v}^2}{d^3} \right]^{\frac{1}{2}} \qquad 28.$$

In considering the fraction of collisions resulting in coalescence, Howarth discounts the earlier proposals of Shinnar (61). Shinnar considered that during the film drainage process, since the cohesive forces holding any pair of drops together against the action of the turbulence are dependant on drop diameter, there should exist at a given intensity of turbulence a minimum size of separable drops. If the maximum stable drop size is less than this rapid coalescence should occur. Although in agreement with the observation in an agitated tank that there exists a critical agitator speed below which coalescence will not occur, as Howarth maintains the weaknesses of Shinnar's theory are,

(a) That compared to the other forces in the turbulence interfacial tension is unlikely to provide a significant cohesive force.

(b) That a significant fraction of collisions may result in coalescence immediately.

Howarth assumes an analogy to drops falling onto a plane liquid surface for which a critical approach velocity exists above which coalescence is immediate. It is assumed that at approach velocities less than the critical value, coherence may occur but eventual coalescence is improbable. On this basis the fraction of collisions resulting in coalescence can be derived.

$$f = \exp\left(- 3\, w^{*2} / 4\, \bar{v}^2 \right) \qquad 29.$$

Combination of equations 28 and 29 yields the collision frequency

$$\nu = \left[\frac{24 \; X \; x \; \bar{v}^2}{d^3} \right]^{\frac{1}{2}} \exp\left(- 3\, w^{*2} / 4\, \bar{v}^2 \right) \qquad 30.$$

Clearly the assumptions made in the derivation seriously limit the application of this equation to real systems. For any particular system the critical approach velocity w^* will depend on the interaction of the complex factors mentioned earlier as affecting coalescence phenomena. In any event the size of drops in a turbulent flow field covers a wide range and the turbulence possesses pronounced directional properties. However, Equation 30 gave good agreement with Madden and Damerells observation that $\nu \propto X^{\frac{1}{2}}$ for water drops dispersed in toluene in an agitated tank (62).

Misek (63) has reported work on droplet coalescence in agitated columns. The following simplifying assumptions were made,

i. Every collision leads to a combination of the colliding drops.

ii. The mixing region is turbulent and dispersed phase droplets follow the direction and velocity of the turbulence fluctuations in the continuous phase.

iii. The dispersion can be characterized by a hydraulic mean drop diameter.

Since drop to drop coalescence may either occur in the bulk of the turbulent liquid or at the column wall, Misek employed two previously derived frequency collision relations by Levich (64);

For coalescence in the bulk liquid,

$$N''_t = \frac{K_2\, n'^2\, d^3\, v_0^{\frac{3}{2}}}{\sqrt{\gamma D}} \qquad 31.$$

For coalescence at the column wall,

$$N''_g = \frac{K_1 n'^2 . d^3 v_o^2}{\gamma} . \qquad 32.$$

The drop sizes combining at each collision correspond to the mean diameter after the preceeding collision. The ratio of diameters of the combined and original drops can therefore be written,

$$\frac{d}{d_o} = \exp. K_3 \frac{N''\theta}{n'} \qquad 33.$$

In a turbulent stream, the size of drops formed by break-up is such that the dynamic pressure on the drop equals the surface pressure of the drop, that is (64),

$$v_o = K_4 \sqrt{\frac{\sigma}{d_o \rho}} \qquad 34.$$

If it is further assumed that the time during which drop to drop coalescence occurs is proportional to the ratio of the characteristic dimension of the column and the characteristic velocity of the turbulence fluctuations i.e. D/v_o , combining equations 32, 33 and 34 for coalescence at the vessel wall and noting that $n'd^3 = X$.

$$\mathrm{In}\ \frac{d}{d_o} = K_5 (n'd^3)\ v_o \left(\frac{D}{\gamma}\right) = K_6\ X \left(\frac{\sigma}{d_o \rho}\right)^{0.5} . \left(\frac{D}{\gamma}\right) \qquad 35.$$

$$= Z_1 . X \qquad 36.$$

For coalescence in the bulk liquid.

$$\mathrm{In}\ \frac{d}{d_o} = K_7\ (n'd^3)\ v_o^{\frac{1}{2}} \left(\frac{D}{\gamma}\right)^{\frac{1}{2}} = K_8 . X \left(\frac{\sigma}{d_o \rho}\right)^{0.25} \left(\frac{D}{\gamma}\right)^{0.5} \qquad 37.$$

$$= Z_2\ X. \qquad 38.$$

It is reasonable to assume that coalescence at the vessel wall is not significant compared with that in the bulk liquid. Misek's correlation of

Equations 37 and 38 is shown in Figure 9, for results from several different columns. Coefficient Z_2 was determined indirectly based on phase flowrate measurements and using Equation 78 considered in para. 7.4. Drop size d_o was calculated from the terminal falling velocities of rigid spheres (65). Despite the evident spread of results they were considered to conform with theory for the binary systems studied. A constant K_8 of value 1.59×10^{-2} was claimed to be independant of the type of mixer.

Misek does not differentiate between results for different column designs. It is doubtful however whether coalescence characteristics in columns as different in operation as the R.D.C. and Oldshue-Rushton can properly be characterised by a single equation. Equation 38, makes no allowance for the known variation in ease of coalescence with drop size (57). The mean drop size in an Oldshue-Rushton is relatively small and droplets would therefore be expected to be prone to coalescence even at low hold-ups.

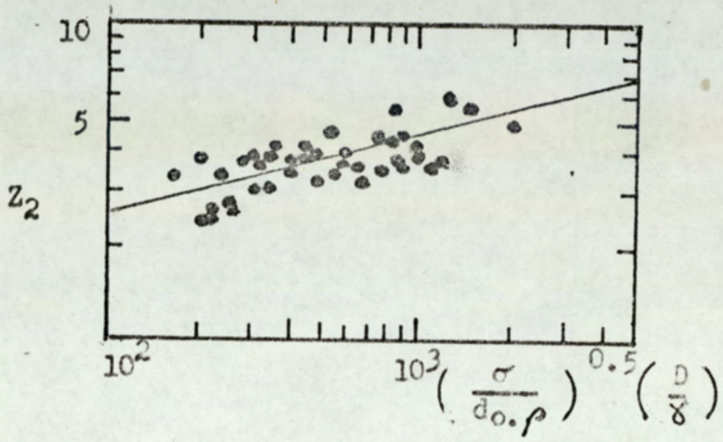


FIGURE 9. EXPERIMENTAL VERIFICATION OF EQUATION 37 (63).

4.3. DROPLET BEHAVIOUR IN PACKINGS

The conventional packings employed in packed towers serve principally to jostle and distort the dispersed phase droplets, so promoting high rates of solute transfer, and to reduce axial mixing. Preferential wetting by the continuous phase is essential. Packings are selected so as to be larger than a critical size d_{fc} given for rings and saddles by (49,66).

$$d_{fc} = 2.42 \left(\frac{\sigma}{\Delta\rho . g}\right)^{0.5} \qquad 39.$$

Under these conditions there is a characteristic mean exit droplet size that is almost independent of packing size and shape. This exit droplet size is independent of the size of droplets entering the packing; larger drops initially present break down whilst smaller drops grow by coalescence albeit more slowly.

The mean drop diameter after 3 ft. of packing has been correlated for a range of aqueous-organic systems, in the absence of mass transfer, by

$$\begin{aligned} d_{vs} = {} & 1.42 \left[\frac{\Delta\rho . \sigma^3}{\mu_c^4 . g}\right]^{0.475} \left[\frac{\mu_c^2}{\Delta\rho . \sigma}\right] \left[\frac{V_o . \varepsilon . X}{V_D}\right] \\ & 0.92 \left[\frac{\sigma}{\Delta\rho . g}\right]^{0.5} \left[\frac{V_o . \varepsilon . X}{V_D}\right] \qquad 40. \end{aligned}$$

At a set value of V_c and at low value of V_D hold-up was found to increase linearly with V_D up to a 'loading-point'; further increase in V_D caused a sharp increase in hold-up and eventually to flooding. Up to the loading point the dispersed phase hold-up was given by

$$\frac{V_D}{X} + \frac{V_c}{1-X} = \varepsilon . V_o (1 - X) \qquad 41.$$

In a Scheibel column smaller packing sizes are employed. One commercial application is known to have used 15 mm. Lessing rings however.

For small packing sizes, below the critical size, larger exit droplet sizes have been observed (49,66). Only a qualitative description of hydrodynamics has been given and no correlation is currently available for volumetric capacity (67). A small amount of reported work on wire-mesh packings is discussed in para. 7.3 and 8.2.

4.4. MOTION OF SINGLE DROPS

The mass transfer characteristics of a single drop are related to the nature of its size and motion. For a given system the terminal velocity of a drop under the influence of gravity varies with drop size as shown in the generalised curve in Figure 10 (68). This curve is applicable to low viscosity systems of high purity.

Small drops in region A are effectively perfect spheres. They do not circulate internally and behave as rigid spheres. Drops of larger diameter are distorted into a generally ellipsoidal shape with the minor axis orientated along the direction of bulk flow - Region B. As the peak drop diameter is approached the horizontal planar symmetry is lost and oscillations set in. Oscillations become more violent as drop size is increased until at some size rupture occurs.

Terminal velocity, drop size, peak size, peak velocity and maximum drop size have been related to the physical properties of the system (68).

The droplets observed in agitated systems are either small spheres or ellipsoids with the minor axis in the direction of bulk flow. By means of a force balance at the steady state terminal velocity, the drag coefficient for a sphere may be defined by the expression,

$$C_D = \frac{4}{3} \cdot \frac{\Delta\rho}{\rho_c} \cdot \frac{g.d}{V^2} \qquad 42.$$

This may be modified for an ellipsoid by employing the minor axis as the length term. Typical plots of C_D vs N_{Re} are given in Figure 11. Such curves cannot yet be predicted for any specific system.

Internal circulation and a mobile interface comprise the main differences between fluid and rigid spheres. For mass transfer from a rigid sphere the interfacial area remains substantially constant. In the case of a circulating fluid sphere however interfacial area is continuously created on the upstream hemisphere and destroyed on the downstream half. The evidence for circulation inside droplets and in particular the early work of Garner et. al. has been well summarised by Kintner (68).

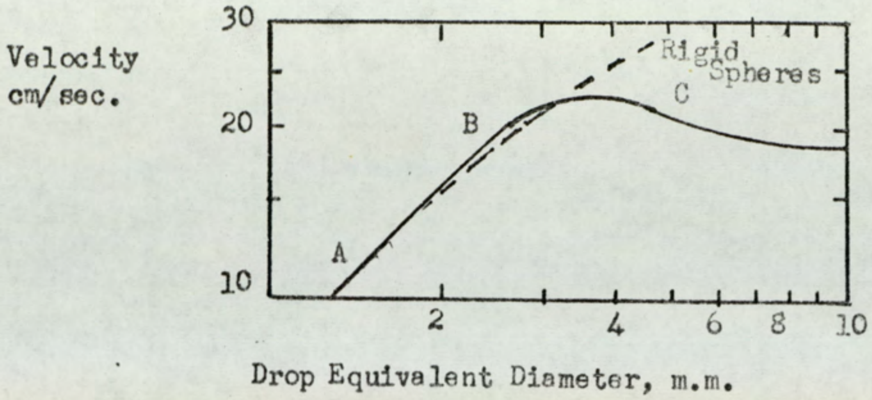


FIGURE 10. GENERALISED TERMINAL - VELOCITY CURVE

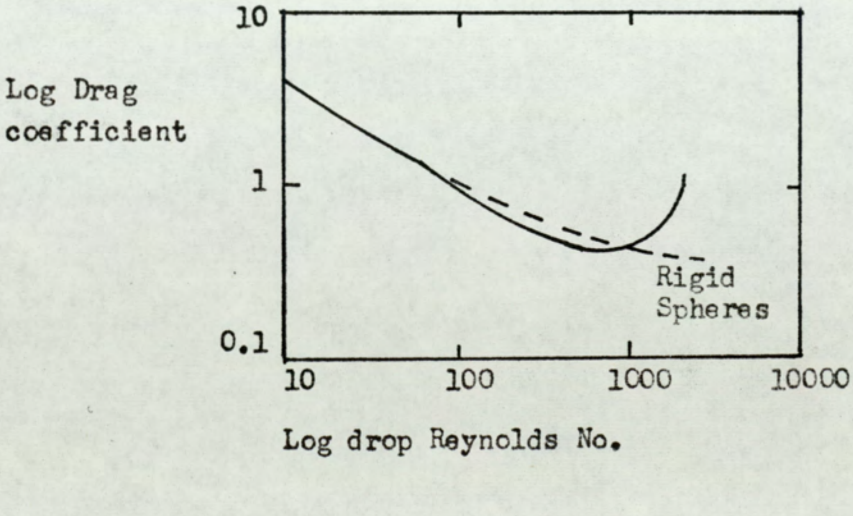


FIGURE 11. TYPICAL DRAG CURVES, liquid drops in liquid media.

5. MASS TRANSFER PHENOMENA

5.1. MASS TRANSFER IN SINGLE DROPS

Prediction of extraction rates requires an understanding of the mechanism of mass transfer within single droplets. Results are available from numerous studies of solute transfer to or from, single droplets (69, 54). Whilst these are unrealistic in terms of the behaviour of the droplet population in a column they are simple to interpret and enable mass transfer rates to be evaluated in terms of a known interfacial area. The rates of diffusion inside, at the interface and outside the droplet determine the mass transfer coefficient.

5.1.1. Mass Transfer within Droplets

The internal mass transfer coefficient is dependant on the rate of circulation of the liquid inside the droplet. This is known to increase with droplet diameter and with the ratio of the viscosity of the continuous phase to that of the dispersed phase.

For stagnant drops, which normally correspond to spheres less than 0.1 to 0.3 cms. diameter, the internal mass transfer coefficient for use with a linear concentration driving force is given by (11),

$$k_d = \frac{2 \pi^2 D'}{3 d.} \qquad 43.$$

For the case of laminar circulation for which $N_{Re} < 1.0$ (11),

$$k_d = \frac{17.9 D'}{d} \qquad 44.$$

Higher N_{Re} numbers are common in practice and once circulation has exceeded a certain minimum the mass transfer rate is apparently independant of the circulation rate. Surface active agents and dirt absorbed on the surface arrest circulation.

Dispersed phase coefficients for turbulent circulation were formerly estimated by the Handlos and Barron correlation (11,70),

$$k_d = \frac{0.00375\ V_t}{1 + \frac{\mu_D}{\mu_c}} \qquad 45.$$

This applies strictly only to drops which oscillate (71) and is based on a model involving turbulence superimposed on the usual circulatory motion inside the drop. This is an unconvincing model and therefore for oscillating drops the correlations of Rose and Kintner (72) or Angelo et.al. (73) , based on the concept of interfacial stretch, have been recommended (54). Oscillation is found to set in at N_{Re} numbers above 200. The interface is assumed to be renewed during each oscillation in which time the interior of the drop is intimately mixed. Rose and Kintner's major assumptions are those of a constant volume for the mass transfer zone, to account for area changes resulting from oscillation, and, local mass transfer coefficients varying in proportion to the thickness of this zone. Their correlation takes the form (72),

$$k_d = 0.45\ (D'_d \cdot w)^{0.5} \qquad 46.$$

where w is the period of oscillation given by,

$$w^2 = \sigma \frac{b}{r^3} \left[\frac{n\ (n-1)\ (n+1)\ (n+2)}{(n-1)\ \rho_d + n\ \rho_c} \right]$$

and the amplitude coefficient b is equivalent to $d_e^{\,0.225}/\ 1.242$.
Experimentally n was found to be 2.
Angelo et. al. proposed alternative equations based upon,

$$k_d = \bar{k} \left[\frac{D'_A}{\pi\ \theta_o} \right]^{0.5} \qquad 47.$$

where $\bar{k}$ is a dimensionless mass transfer coefficient dependant on the type of stretching of the surface. This coefficient has been expressed by equations for the cases of a droplet undergoing formation and for a free-falling oscillating drop (73). More data are necessary but in future these models should prove of value in the analysis of mass transfer in agitated systems in which oscillating drops are produced.

As discussed in para. 4. 2. a regime of continual droplet coalescence and break-up may predominate in agitated extractors. This will have a significant effect on internal mass transfer. Because only a small amount of fresh interface is created by coalescence a negligible contribution is likely to mass transfer; the reduction in area may in fact cause an overall decrease (71). However results on droplet formation reviewed by Jeffreys (54) provide conclusive evidence that appreciable extraction occurs during droplet formation. Precise rates are, as yet, unpredictable but this is a particular case of transfer under conditions of surface stretch. In the event, efficient extraction in an agitated vessel with droplets in a size range normally associated with a molecular diffusion transport process has been attributed to break-up and coalescence (74).

5.1.2. Interfacial Resistance

The interface offers some degree of positive resistance to mass transfer. In practical extraction this is attributable to the presence of surface active agents. Most of the experimental evidence supports a Barrier Layer Theory (75). Surface active agents are adsorbed at the interface and, by either mechanical or chemical interaction, form a barrier to solute diffusion. However, hydrodynamic effects cannot be neglected since a reduction in internal circulation has been observed due to adsorbed surface active molecules. The latter mechanism is given support by the observation that at high concentrations of surfactants mass transfer to drops approximates to that to solid spheres.

In certain systems, turbulence and spontaneous emulsification may occur at the interface. These are due to interfacial tension effects and are dependant on the direction of mass transfer (75). The phenomena are mostly apparent at high transfer rates associated with high concentration gradients and their effect can usually be neglected in practical extractor design. (2).

5.1.3. Mass Transfer outside Droplets

Numerous correlations have been given for the continuous phase mass transfer to rigid spheres (70, 2, 69). For all but the lowest N_{Re} numbers ,

$$Sh = 0.6 \; (N_{Re})^{0.5} \; (Sc)^{0.33} \qquad 48.$$

For circulating droplets the relationship is,

$$Sh = 1.13 \; (N_{Re})^{0.5} \; (Sc)^{0.5} \qquad 49.$$

For oscillating droplets the relationship is . (69),

$$Sh = 50 + 0.0085 \; (N_{Re}) \; (Sc)^{0.7} \qquad 50.$$

5.1.4. Overall Coefficient of Mass Transfer

The overall coefficient is the sum of the coefficients of the individual phases. The resistance of one phase may predominate and design can then be based on that phase. If the physical properties of the system predict a drop size greater than 0.1 to 0.2 cms. liquid inside the drop probably circulates rapidly and the continuous phase coefficient is controlling (2). This can be confirmed by comparing the times for a droplet to attain 60% or 90% solute concentration based upon equations for diffusion in the two phases (71) ; the phase requiring the larger time is controlling. For transfer inside the drop (2),

$$\frac{Q_\theta}{Q_o} = 1 - \exp \left[-2.25 \left(\frac{4 \, D'_d \, \pi^2 \theta}{d^2} \right) \right] \qquad 51.$$

For transfer outside the drop (76),

$$\frac{Q_\theta}{Q_o} = 1 - \text{erf} \frac{x}{\sqrt{4 D'_c \theta}} \qquad 52.$$

In order to apply equation 52, it is necessary to assume that droplets are uniformly dispersed and hence the depth of diffusion is half the distance between adjacent drops (71). Unfortunately this neglects mixing effects.

6. THE EFFECT OF AGITATION UPON MASS TRANSFER BETWEEN IMMISCIBLE LIQUIDS

The primary object of dispersing one phase as droplets is to create a large interfacial area. Any desired dispersion can be achieved by agitation which also serves to distribute droplets uniformly throughout the continuous phase.

A further effect is the reduction of the resistance to diffusion across both phase films as discussed in para. 5.1.

Appreciable extraction may occur during droplet formation and therefore when continual droplet coalescence and break-up predominates in a contactor enhanced mass transfer rates should occur. Sawistowski and Goltz have found that it is difficult to predict the enhancement of extraction rates during drop formation because rapid changes occur in interfacial tension (77). However, they confirmed the results of earlier workers that, in spray columns, about 10% of the extraction occurs during drop formation. As mentioned in para. 5.1.1. it has been suggested that transfer occurs under surface stretch conditions (73). From limited experimental work, Angelo et.al. found that the mass transfer coefficient during drop formation from a sieve plate may be 15 times greater than that predicted from empirical boundary layer correlations for flow past a cylinder.

6.1. Enlargement of Interfacial Area

The hydrodynamics of droplet break-up were considered in para.3. The deformation of the droplets by shear, which is proportional to the intensity of agitation, acts in opposition to the interfacial tension. Rodger et.al. (43) suggested the modification of the Weber number,

$$We = \frac{\rho v^2 . d_o}{\sigma}$$

to directly represent the effect of agitation

$$We = \frac{R^3 N^2 \rho c}{\sigma}$$

Not only droplet break-up but also coalescence results from agitation however. Droplets in the turbulence travel in all directions resulting in collisions. Droplet coalescence phenomena were discussed in para.4.2.

(a) Batch mixing

In batch mixing, under fully baffled conditions, after a period at constant impeller speed, a state of dynamic equilibrium is established between the number of combining and splitting droplets (62). This effect is dependant on agitator speed. The drop size distribution produced is dependant on the dispersed phase hold-up, i.e. the characteristics of coalescence predominates in concentrated dispersions and those of break-up in dilute (44, 78). Since the liquid velocity varies throughout the vessel, being largest near the impeller, a variation of droplet size exists with position in the vessel. Splitting of droplets is more effective the higher the intensity of agitation; drop diameter decreases therefore with increased impeller speed. Several investigations have been made into the dependance of interfacial area on agitation in baffled vessels in the absence of solute transfer (43, 44, 79, 80). Combining the physical constants and specific influence of a given agitator design into a single constant the results take the simplified form,

Agitator Type	Relation for a	Authors
6 blade turbine baffled vessels.	$K_1 R^{1.1} D^{-1.2} N^{0.72}$	Rodger et.al. (43)
Impeller with 4 paddles, baffled vessels.	$K_2 R^{0.8} N^{1.2}$	Vermeulen et.al. (44)
Various agitators, baffled vessels.	$K_3 R^{0.7} N^{1.1}$	Kafarov et.al. (78)

Agitator Type	Relation for a	Authors
Impeller with 4 straight blades, 6 blade turbine, baffled vessels.	$K_4R^{0.8} N^{1.2}$	Calderbank (79)
Various agitator designs	$K_5R^2 D^{-1.7} N^{1.2}$	Rodriguez et.al. (80)

Apart from differences in impeller design and location, the use of different methods for the measurement of interfacial area accounts for some degree of discrepancy.

According to Resnick (81) the variation of average drop size with agitator speed has more recently been found to be expressed by:-

$d_{32} \propto N^{-1.2}$, when inertial break-up forces predominate.

$d_{32} \propto N^{-1.5}$, when break-up due to viscous shear predominates.

$d_{32} \propto N^{-0.75}$, when coalescence effects predominate.

(b) Fully Baffled - Continuous Flow

In a fully baffled continuous flow system the Sauter mean droplet diameter at constant impeller power has been found to increase linearly with hold-up due to coalescence effects (74). Thus the drop size may be expressed by an equation of the form,

$$d_{vs} = d^o_{vs} + m.X. \qquad 53.$$

where d^o_{vs} is the drop size at zero hold-up, i.e. in the absence of coalescence, and is a function of the power dissipation and the physical properties of the system. The term m in equation 53 was found to be constant for a given system regardless of power level in the agitated vessel; this is shown by the typical results reproduced in Figure 12.

Since m corresponds to the rate of change of droplet size with hold-up Thornton considers it to be a measure of the coalescence characteristics of the system. Data obtained for a range of aqueous-

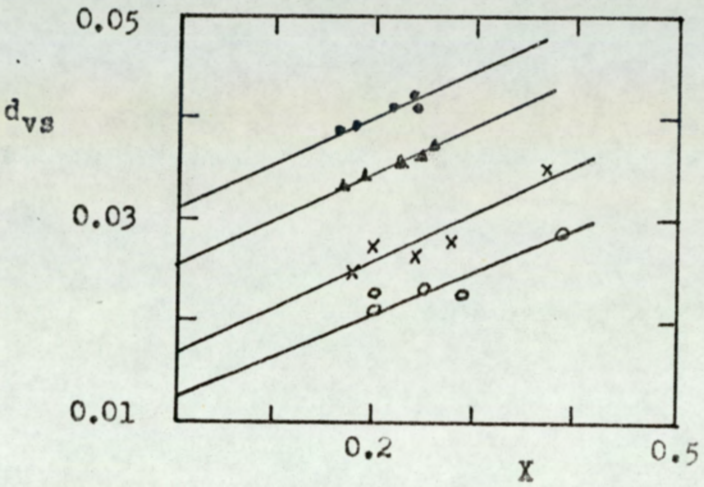


FIGURE 12. DROPLET SIZE vs DISPERSED PHASE HOLD-UP (74)

ethyl acetate - water.

- ● 400 r.p.m.
- ▲ 500 r.p.m.
- × 750 r.p.m.
- ○ 1,000 r.p.m.

organic solvent systems in a 7 inch diameter fully baffled vessel, in the absence of mass transfer, could be correlated by the expression,

$$d_{vs} = d^{o}_{vs} + 1.18 \times \left[\frac{\sigma^{2}}{\mu_c^{2} g}\right] \left[\frac{\Delta\rho.\sigma^{3}}{\mu_c^{4}.g}\right]^{-0.62} \left[\frac{\Delta\rho}{\rho_c}\right]^{0.05} \qquad 54.$$

The droplet size at almost zero hold-up d^{o}_{vs} could be correlated with power dissipation and physical properties by the expression,

$$\frac{d^{o}_{vs}{}^{3}.\rho_c^{2}.g.}{\mu_c^{2}} = 29.0 \left[\frac{P_v^{3}.g_c^{3}}{\rho_c^{2}\mu_c.g^{4}}\right]^{-0.32} \left[\frac{\rho_c.\sigma^{3}}{u_c^{4}.g}\right]^{0.14} \qquad 55.$$

(c) Agitated Column Contactors

In a study of droplet break-up by a rotating disc under acollisive conditions, Misek (45) assumed break-up to be identical with that in a closed system because phase flow-rates are negligible in comparison with the disc peripheral velocity. In practice the hold-up has a finite value and although it is generally less than in agitated vessels conditions are not necessarily acollisive; the equilibrium drop size distribution is then dependant on specific contactor characteristics. These are considered in para. 8.

6.2. UNIFORMITY OF DISPERSION

Extraction efficiency is dependant not only upon the size of the dispersed phase droplets but also upon the gross uniformity of the propertions of dispersed to continuous phase throughout a contactor.

A correlation has been established for batch agitated vessels giving the minimum impeller speed above which separate liquid layers disappear (11) and the application of 250 to 500 ft.lb./min. impeller power / ft^3 of liquid volume has been found to give generally near-uniform dispersions (82).

Relatively little data are available on the existence of a minimum speed for proper functioning of the various columns. Reference is made in para. 8.3. to a minimum tip speed in the R.D.C. claimed to eliminate droplets trapped beneath the rotor and stators (83) and according to Misek (46) the only function of the rotating disc at $R_e \triangleq 10^4$ is to distribute droplets throughout the column cross-section.

6.3. MASS TRANSFER RATES.

Little data are available on individual mass transfer coefficients in agitated systems and these are normally estimated by the equations given in para. 5 (11). Reference has been made to the work of Thornton (74). Coughlin and von Berg have studied mass transfer in a mixer settler and found that with increasing impeller speed, coefficients were in agreement with a transition in droplet behaviour from rigid via circulating to oscillating droplets (84).

7. THE DESIGN AND PERFORMANCE OF ROTARY AGITATED COLUMN EXTRACTORS

7.1. DESIGN PROCEDURE

For any application dispersion of the phase with the larger throughput will yield the maximum interfacial area. An ambivalent region exists, possibly extending over a concentration range of 10% to 90% of one phase (71), within which either phase may be dispersed. This is dependant upon which phase is introduced into the column initially to form the continuous phase. A recent analysis suggests that when the separation coefficient is < 1.0, with agitation of moderate intensity as in the majority of practical examples, a dispersed feed phase is preferable (85).

Design then involves evaluation of,

- i. The number of transfer units for the required degree of extraction.
- ii. The height of a transfer unit.
- iii. The column capacity.

Alternatively design may be based on the height of a theoretical stage or a convenient height may be arbitrarily designated as a real stage and performance characterised in terms of stage efficiency (86).

The number of transfer units, or stages is estimated from mass balances (11). The height of a transfer unit, or stage efficiency, is determined from pilot scale experiments or predicted from experience. Capacity may be predicted from flooding correlations derived by dimensional analysis and aided by empiricisms (16).

A more rigorous design procedure is based on droplet analysis considerations. The rate of mass transfer, dx/dt, is evaluated from the equation,

$$\frac{dx}{dt} = K.\ A.\ \Delta C. \qquad 56.$$

The dependance of the overall mass transfer coefficient on the series resistance to diffusion inside the droplet, at the interface and outside the droplet has been considered in para. 5.1. It is not usual to allow for interfacial resistance in design (70), hence:-

$$\frac{1}{K_d} = \frac{1}{mk_c} + \frac{1}{k_d} \qquad 57.$$

In practice the concentration driving force is reduced to some extent by axial mixing. Axial mixing exists in both the dispersed and continuous phase. It comprises two effects, viz, turbulent diffusion in the axial direction super-imposed upon the non-uniform velocity which occurs across any tower cross-section. In any application it results in a reduction in the solute concentration driving force and hence in the extraction rate compared to that achievable under plug-flow conditions.

Sleicher (3) expressed the effects in terms of an eddy diffusivity E' which was incorporated into a Peclet Number $Pe = \frac{V.H.}{E'}$

Two diffusivities E'_c and E'_D and hence two Peclet numbers Pe_c and Pe_D are involved. For plug flow $E' = 0$ and $Pe = \infty$; the other limit is complete axial mixing for which $Pe = 0$.

Complex equations, on solution by computer, yielded the following empirical equation relating the height of a column required for true counter-current (plug) flow to that required with axial mixing for the same amount of extraction,

$$\frac{H\ plug}{H} = \frac{Pe_D . \ Pe_c}{Pe_D . \ Pe_c + H.Z / H.T.U_{o.c.}} \qquad 58.$$

where

$$Z = a.Pe_c + b.Pe_D + c\,(Pe_D . Pe_c)^{0.5} - d\,(Pe_D + Pe_c)^{0.5} + g\,(Pe_c - Pe_D)\,e^{-hH/\,H.T.U_{o.c.}}$$

The constants a to h are a function of the extraction factor, i.e. $\frac{m.V_c}{V_D}$.

The practical application of equation 58, in agitated column design is dependant on the correlation of E'_C and E'_D with the physical properties of the system, the geometry of the column and the operating parameters i.e. agitator speed and phase flow rates. Measured axial mixing data has been published for the R.D.C. (87 - 91) and the Oldshue - Rushton column (92,93) and this is summarised in paras. 7.4. and 7.5.

The interfacial area is a function of droplet size and dispersed phase hold-up. These may be predicted as follows,

(a) Dispersed Phase Hold-Up

The dispersed phase hold-up can be predicted from the dispersed phase superficial flow-rate and the mean residence time of the droplets in the contactor. The residence time of the droplets is a function of the droplet size, the difference in density between the phases, the density and viscosity of the continuous phase and the distance travelled between consecutive coalescences. The velocity of ascent or descent of the droplet must be predicted by reference to the data summarised in para. 4.3. and 4.4.

It is more convenient to predict the dispersed phase hold-up by means of equations relating fractional hold-up to the superficial velocities of the dispersed and continuous phases and the characteristic velocity of the droplets in the contactor.

Equations take the form,

$$\frac{V_d}{X} + \frac{V_c}{1-X} = V_n (1 - X) \qquad 59.$$

The characteristic droplet velocity V_n depends upon whether or not the contactor is agitated, contactor geometry and the physical properties of the liquids. The equation is applicable providing the mean drop size is independant of phase flow rates and dispersed phase hold-up over the desired range. It makes no allowance for the effects of droplet break-up and recoalescence which influences both droplet velocity and the extent of axial mixing.

(b) Droplet Size

The mean diameter of droplets formed in an agitated column may be predicted from the equations given in para. 3.2. Again no allowance is made for coalescence and redispersion however.

Column capacity may be estimated from the rate of ascent or descent of droplets through the continuous phase.

Knowledge of the characteristics of each contactor is clearly essential for design based on droplet analysis. These are considered in paras. 7.3., 7.4. and 7.5.

7.2. GENERAL DESIGN CONSIDERATIONS.

7.2.1. Wettability of Column Internals.

For optimum performance the column internals should be preferentially wetted by the continuous phase. If this is not achieved, bulk coalescence of the dispersed phase may occur on rotors, stators, walls or packing. This leads to a reduced interfacial area between the phases. The effect has been demonstrated conclusively in the R.D.C. by Davies et.al. (94) using kerozine to extract phenol from a dispersed aqueous phase. A comparison of extraction efficiency was made between operation with naturally water wetted glass and brass surfaces and with surfaces made hydrophobic by the application of trimethylsilane. Their results are summarised in Figures 13 and 14, and show the greater efficiencies achieved with continuous phase wetted, i.e. in this case hydrophobic, internals.

Earlier in the same contactor the effect of a dispersed (kerozine) phase wetting the stator and rotor was recorded photographically (95). A similar effect has been recorded in a 3 inch Scheibel

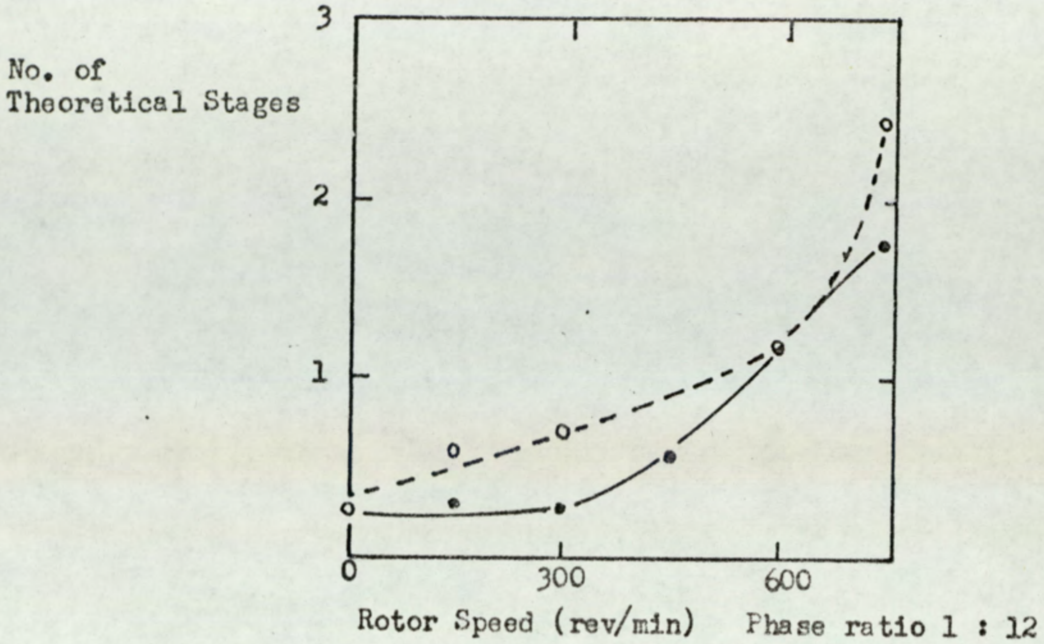


FIGURE 13. R.D.C., EFFECT OF ROTOR SPEED ON NUMBER OF THEORETICAL STAGES (94).

(Open points - hydrophobic surfaces,
Solid points - hydrophilic surfaces)

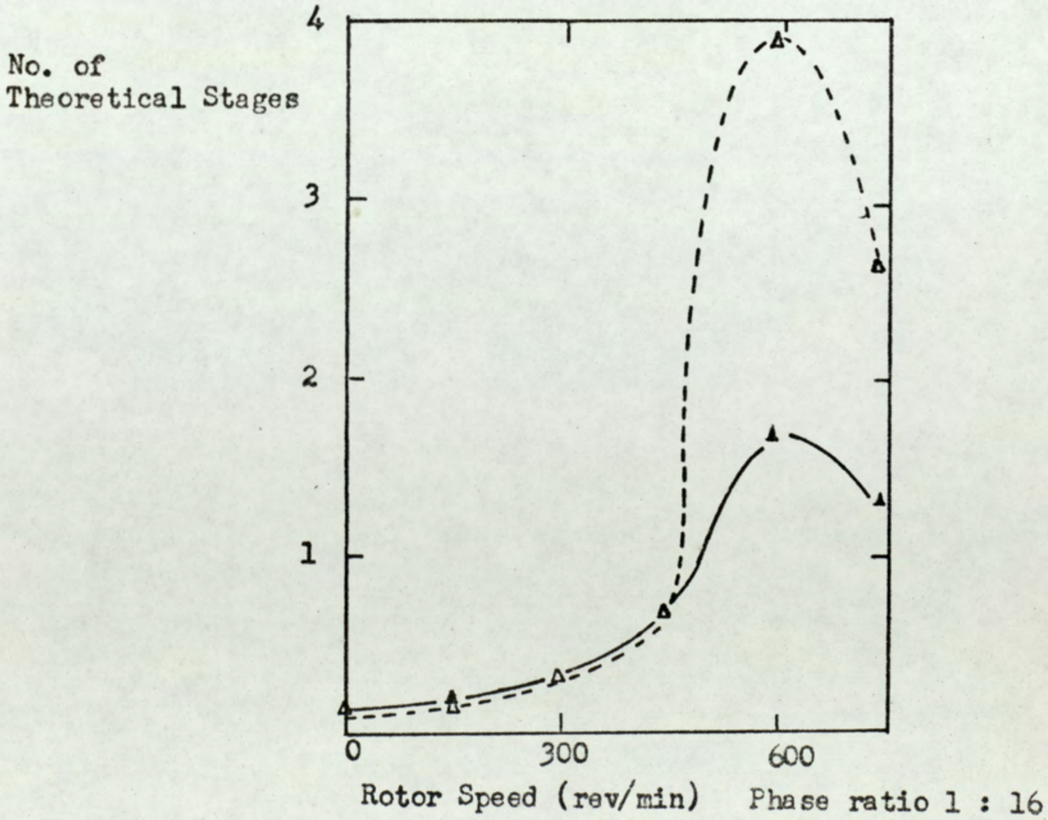


FIGURE 14. R.D.C., EFFECT OF ROTOR SPEED ON NUMBER OF THEORETICAL STAGES (94).

(Open points - hydrophobic surfaces, solid points - hydrophilic column (96); the dispersed (kerozine) phase wetted the S.S. agitator shaft and formed a continuous stream up it due to ineffective cleaning.

Wetting effects can be reduced when an aqueous phase is dispersed by applying a suitable hydrophobic lining to the impeller and stators. Thus Davies suggested nylon (94) and Madden and Damerell (62) relied on polytetrafluoroethylene in their coalescence studies. Most plastics especially polythene, polypropylene and p.t.f.e. are hydrophobic and would be suitable for such an application. For operation with a continuous aqueous phase, most metals would be suitable being neither strongly hydrophobic or hydrophilic with the exception of molybdenum and copper (11).

The packing in a Scheibel extractor may be constructed from metal or plastic depending on the wetting characteristics of the extract and raffinate either of which may be dispersed under the operating conditions in the column. Unless the continuous phase preferentially wets the packing, large scale coalescence of the dispersed phase may occur in the packing. This may result in the formation of a coalesced core throughout the packing with the continuous phase channeling at the vessel walls. This was experienced by Piper (96) due to failure to remove the film of oil normally present on metal wire mesh packing as despatched from the manufacturer (97). Droplet formation of the dispersed phase as it left the packing was then variable between a fast flowing jet giving uniform drops and drip formation.

7.2.2. Direction of Mass Transfer

In the absence of mass transfer, dispersed phase hold-up may be predicted by a form of equation 59, and mean droplet diameter predicted by use of equations considered in para. 5.2. From the consideration of coalescence phenomena in para. 4.2. however, it would be expected that mass transfer from the dispersed to the continuous phase would result in larger irregular droplets. Hence a lower extraction efficiency but increased volumetric capacity should obtain in comparison with operation with transfer in the reverse direction.

This effect is well established in other types of extractor (1). It has been observed in a Scheibel column with the systems M.I.k - acetic acid-water, o-xylene-acetic acid-water and o-xylene-acetone-water by Karr and Scheibel (98) and independantly by Hixson (99). In an R.D.C., Logsdail et.al. observed that the volumetric capacity was increased between 100 - 200% when solute transfer took place from the dispersed to the continuous phase but decreased by 10% in the reverse direction both being compared with the solute free system.

It follows therefore, that hydrodynamic correlations determined in the absence of mass transfer may require some modification for practical extractor design. This is not unfortunately always made clear (46).

7.3. SCHEIBEL COLUMN DESIGN

Mixing Section.

The preferred agitator for droplet break-up and dispersion should be a turbine giving predominantly radial flow. In a Scheibel column dependance is placed on packing, or horizontal stators, to restrict axial flow. In the original designs (8,98) and that of Piper (96) the agitator was referred to as a paddle; this is not substantiated by the dimensions or operating speeds. The agitator was in fact a non-standard turbine; this was modified by Scheibel in the later design (20). Honekamp and Burkhart failed to report agitator details (9).

The effect of vertical baffles has been studied (20). As would be expected the elimination of tangential flow was only achievable at the expense of increased axial mixing thus requiring higher packing heights to isolate the mixing sections.

An optimum agitator speed exists for any duty. The optimum speed is that which produces droplets giving the highest mass transfer rates without presenting hold-up or settling problems. Scheibel reported optimum speeds for the system M.I.k - water-acetic acid in a 4" column as shown in Figure 15 (20). Excessive agitator speeds reduced efficiency due to the formation of finer dispersions which did not coalesce in the calming sections and to the overlapping of the flow patterns in successive mixing stages. Curves of similar shape were found with a higher interfacial tension system (o-xylene-water-acetone) but with a lower maxima due to the absence of any significant mass transfer in the packing (86).

Packed Section.

The extent to which coalescence occurs in the packed sections is dependant on drop size, the physical properties of the system and the material of construction and interstices size of the packing.

A characteristic drop size may exist in mesh packings. Drops above or below this characteristic drop size approach the characteristic size providing sufficient packed height is available. (9).

Little change was observed in average drop size when a dispersion of M.I.K in water was passed through 5 inches of 97% void standard Yorkmesh S.S. packing. From measurements of packing exit drop size as a function of inlet drop size, flow-rate and packed height, Honekamp and Burkhart concluded that the outlet drop size was independant of dispersed and continuous phase flow-rates. The decrease in size of drops greater than the characteristic size occurred rapidly but smaller drops increased in size slowly. From the significant difference found in drops leaving 5 inches as against 15 inches of packing the characteristic size was clearly not attained in 5 inches of packing.

In a similar study with a high interfacial tension system (kerozine - water) in addition to very small drops which pass through the packing unaffected and drops which tended to attain the characteristic size, Piper observed a number of large globules in the packing (96). Globule formation occurred at any point in the packing where a number of drops were held up for a sufficient time to coalesce; with

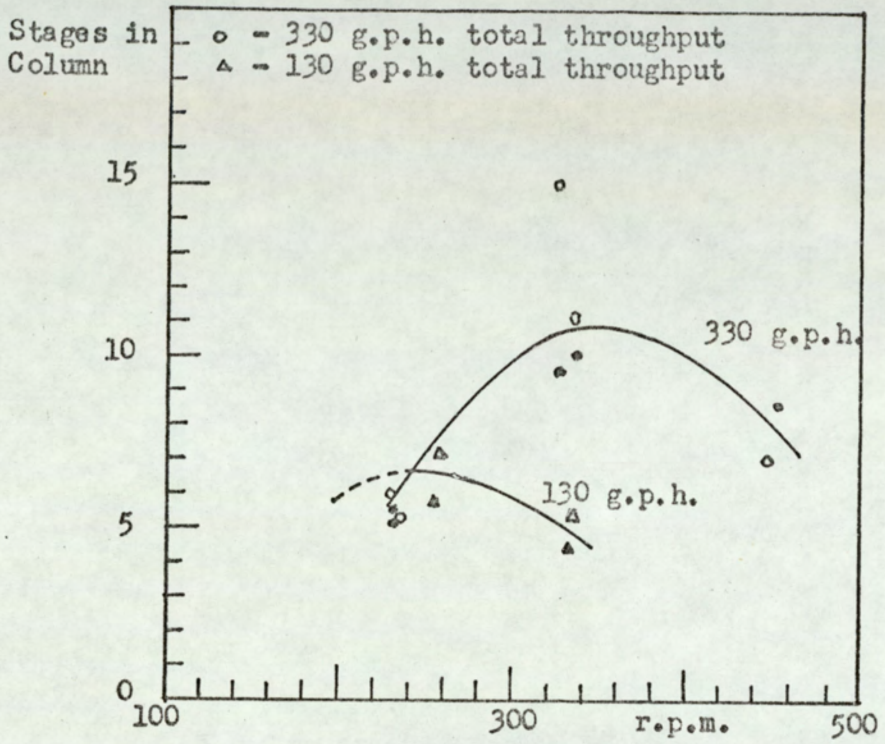


FIGURE 15. EFFICIENCY vs AGITATOR SPEED

4 inch. Scheibel, M.I.K. - acetic acid - water system (20) (Open points, water — ketone; Solid points ketone — water).

the three packings tested a decrease in interstices size resulted in increased globule formation. The dispersed phase comprising the globules left the packing by drip point formation.

The basic purpose of the packing is to isolate the mixing sections. Hence a continuous phase wetted packing with the highest practical voidage is desirable to minimise packed height. Interstices size should not be so small as to place an impractical limit on throughput and assembly should be such as to inhibit globule formation.

Nevertheless with a low interfacial tension system, a significant amount of extraction may occur in the packing. With the system M.I.K. - acetic acid - water, Honekamp and Burkhart confirmed that the percentage of the total mass transfer which occurred in the packing was dependant on agitator speed and flowrates. With the 3 inch column and 5 inches of packing under optimum operating conditions the order was 50% (9). Based on earlier work Scheibel suggested that the optimum packing height was approximately proportional to the square root of the column diameter for columns of 1 and 12 inches diameter (10).

Capacity

The capacity of the column is limited by the permissible flowrate through the packing. As was pointed out earlier the interstices size of the mesh packing is below the critical packing size, calculable for any system, for which capacity correlations have been established (49,100). The hydrodynamics of flow through such packings is as yet little understood (67,101).

Hold-Up

The total hold-up comprises dynamic and static hold-up. The latter is the dispersed phase retained in the column on shut-down.

The variation of total hold-up with agitator speed has been determined by Karr and Scheibel (98) with an M.I.K - acetic acid - water system in a 12 inch diameter column. When the M.I.K extractant (light) phase was dispersed the hold-up was found to increase linearly with agitator speed. Similar results have been reported by Honekamp and Burkhart (9) from work with the same system in a 3 inch column. When the water extractant (heavy) phase was dispersed however, the hold-up was found to pass through a maxima as shown in Figure 16. This phenomena was attributed to centrifugation of the dispersed phase.

Phase Ratios

At phase ratios greatly different from unity, high agitator speeds are apparently required to achieve satisfactory performance (20).

Design Procedure.

No fundamental design procedure has been attempted for the Scheibel column presumably because of the absence of hold-up data.

A stagewise technique has been proposed by Jeffreys using the data of Scheibel and Karr to predict stage efficiency (70,98). Column capacity is not, as yet, calculable.

No measurements have been reported on axial mixing coefficients.

7.4 ROTATING DISC CONTACTOR DESIGN

Contactor Geometry

The dimensions to be selected for a column to perform a given duty include the column diameter D, the rotor diameter R, the diameter of the stator opening S and the compartment height H.

Küng and Beckmann (83) have stated that on the basis of their investigation and those of others (14,102), the optimum operating conditions for R.D.C.'s exist over the range of variables :-
$S/D = 0.66$ to 0.75, $R/D = 0.5$ to 0.66 and $H/D = 0.33$ to 0.5. That these are the preferred range of variables is confirmed in the nomogram by Misek (46) and supported by other published data (103, 87).

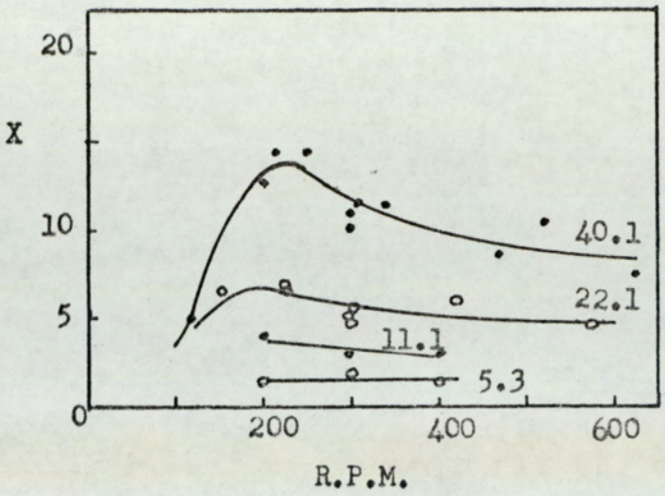


FIGURE 16, DISPERSED PHASE HOLD-UP vs AGITATOR SPEED (98)

12 inch Scheibel, M.I.K. - acetic acid - dispersed water.
Parameter V_d.

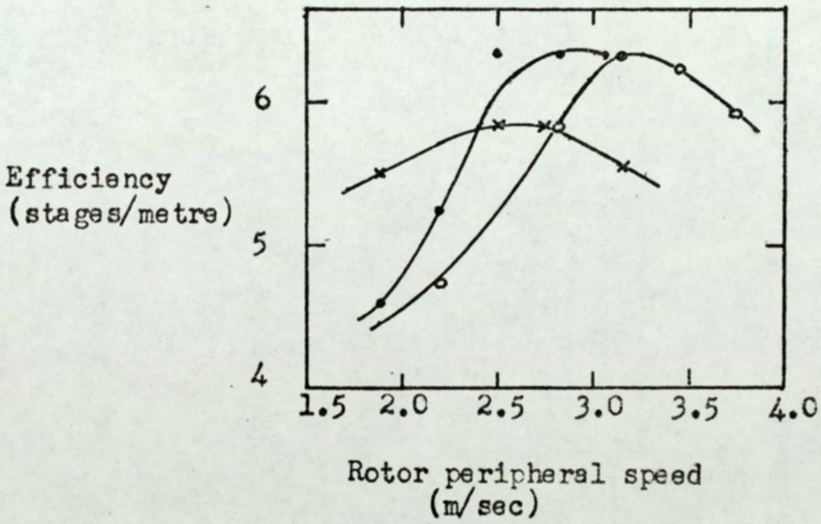


FIGURE 17, EFFICIENCY vs ROTOR SPEED, 200 m.m. R.D.C.

system M.I.B.K. - water - acetic acid,
specific load $V_d + V_c = 11.8 \times 10^{-3}$ cu.m/sq.m.sec. x ,
8.0×10^{-3} cu.m/sq.m.sec. • ,
and 5.0×10^{-3} cu.m/sq.m.sec. ○ .

$V_d / V_c = 1.0$ (16).

Unfortunately some investigators have not conformed to these dimensions and their data are therefore not comparable (90,91,104,105). Below a certain stator-rotor gap the column does not behave as an R.D.C. (83) ; premature flooding would be expected in such cases.

Rotor Speed

The effect of rotor speed on extraction efficiency was determined by Reman (14,16,106) with several systems. The efficiency, expressed as stages per metre of contactor height, was observed to increase with increasing rotor speed. In certain cases however the efficiency passed through a definite maximum as shown in Figure 17. A similar phenomenon was observed by Vermijs and Kramers (103). It arises as axial mixing becomes increasingly effective due to increased turbulence and smaller average drop sizes associated with high rotor speeds.

On one unreported occasion a satisfactory performance was inadvertantly obtained on lubricating oil purification with the rotor stationary (107). This was attributed to the provision made for tangential inlet of the phases.

The effect of rotor speed on capacity is considered below. A minimum rotor speed of 300 ft./min. has been recommended for all systems (83). This is discussed in para. 8.3.

Hold-Up

Logsdail and co-workers (102) concluded from visual observations of a 3 inch dia. R.D.C. that the size of the dispersed phase droplets was independent of the phase flow rates and the dispersed phase hold-up. Their study covered column variables i.e. stator opening at 2.0 and 2.5 inches, compartment height at 0.5, 1.0, 2.0 and 4.0 inches and rotor speeds between 500 and 1600 r.p.m. Water was used throughout as the continuous phase with dispersed phases of toluene, butyl acetate, iso-octane, white spirit or benzene. Under such conditions they postulated that for a specific system at a given column geometry and rotor speed, the phase flow-rates and hold-up could be related by a form of equation 59, viz.

$$\frac{V_d}{X} + \frac{V_c}{(1-X)} = \bar{V}_n \quad (1 - X) \qquad 60.$$

where $\bar{V}_n$ is the mean vertical component of velocity of the droplets with respect to the continuous phase.

This gave good agreement with experimental results. An expression was then developed through dimensional analysis relating $\bar{V}_n$ to the physical properties of the system, the column diameter and the rotor speed, viz.

$$\frac{\bar{V}_n \ \mu_c}{\sigma} = 0.012 \left(\frac{\Delta\rho}{\rho_c}\right)^{0.9} \left(\frac{g}{RN^2}\right)^{1.0} \left(\frac{S}{R}\right)^{2.3} \left(\frac{H}{R}\right)^{0.9} \left(\frac{R}{D}\right)^{2.7} \qquad 61.$$

This gave a correlation for the majority of the data within ± 15%.

Kung and Beckmann (83) determined the effect of column geometry and operating flowrates on Equations 60 and 61. Hold-up was found to increase with decreasing stator opening and compartment height and increasing disc diameter and rotor speed. Hold-up was found to increase linearly with dispersed phase flow-rate up to the loading point and then to increase more sharply as in Figure 18, and as reported earlier by Vermijs and Kramers (103). Equation 60, was found to be applicable to a wide range of column geometries in the form,

$$\frac{V_d}{X} + \frac{K_1 V_c}{1 - X} = \bar{V} \ (1 - X) \qquad 62.$$

where coefficient K_1 had the value 1.0 at $(S - R) / D > 1/24$ or 2.1 at $(S - R) / D \leqslant 1/24$.

The characteristic velocity term $\bar{V}$ was found to remain constant with various rotor speeds less than 300 ft. min.$^{-1}$. At two higher speeds

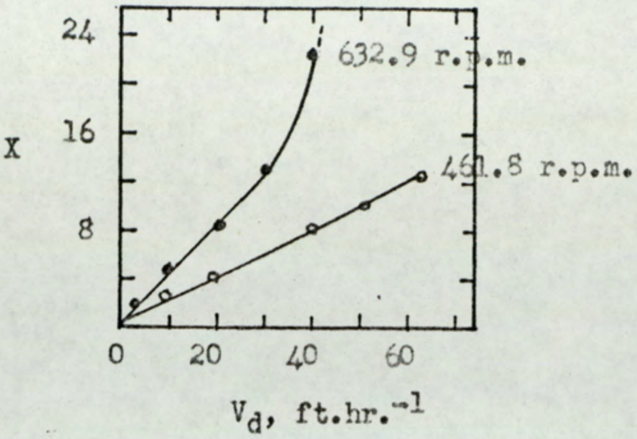
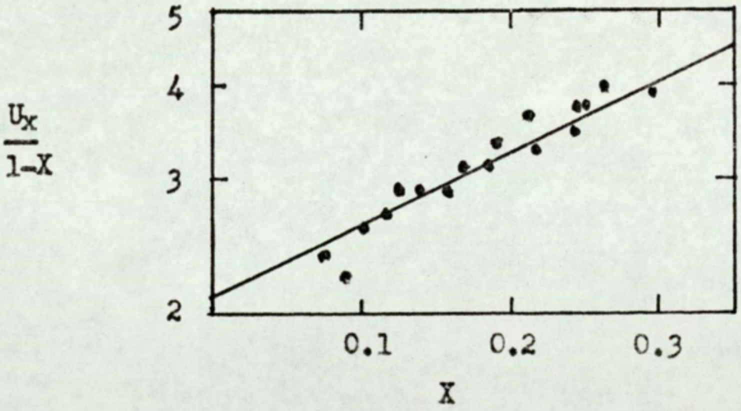
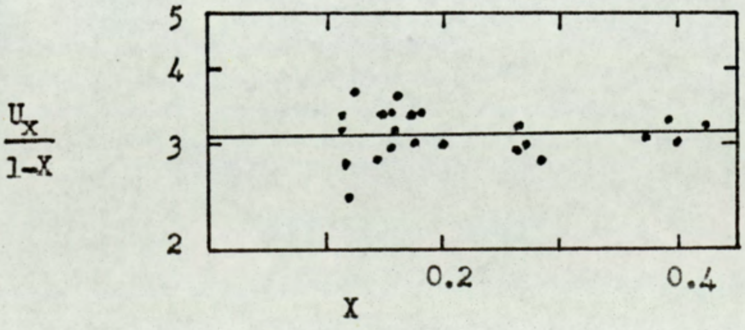


FIGURE 18. DISPERSED PHASE HOLD-UP vs ROTOR SPEED (83)

6 inch R.D.C., system toluene - water



system, water - butyl acetate



system, water - toluene

FIGURE 19. EXPERIMENTAL VERIFICATION OF EQUATION 78 (108).

values of $\bar{V}$ were correlated by a modification of Equation 61, viz.

$$\frac{\bar{V}\mu_c}{\sigma} = K_1\left(\frac{\Delta\rho}{\rho_c}\right)^{0.9}\left(\frac{g}{RN^2}\right)^{1.0}\left(\frac{S}{R}\right)^{2.3}\left(\frac{H}{R}\right)^{0.9}\left(\frac{R}{D}\right)^{2.6} \qquad 63.$$

where coefficient K_1 had the value 0.0225 at $(S - R) / D \leqslant 1/24$ and 0.012 for $(S - R) / D > 1/24$.

In employing equations 60 and 61 to calculate hold-up, no allowance is made for coalescence and break-up of droplets in the contactor. Misek (108) therefore proposed a modified form of Equation 60, the theoretical basis for which is given in Appendix 8.
The modified equation is,

$$\frac{V_d}{X} + \frac{V_c}{1-X} = \bar{V}(1 - X) \text{ exp. } \left[X\left(\frac{Z}{m} - 4.1\right)\right] \qquad 78.$$

where Z is the coalescence factor referred to previously in para. 4.2.

The validity of this equation was investigated by Misek using different systems in a 250 m.m. diameter R.D.C. The results were plotted graphically as ,

$$\log \frac{V_d}{X(1-X)} + \frac{V_c}{(1-X)^2} = f(X) \qquad 79.$$

Some results are shown in Figure 19.

Misek interpreted the results as demonstrating good applicability for the R.D.C. Whereas this is apparently true for systems of low interfacial tension, viz. water - butyl acetate, water - diethyl ether, water - amyl alchol and diethylene glycol-gasoline the results for the water - toluene system having an interfacial tension of 24.48 dynes/cm. show poor agreement. Deviations are particularly noticeable at low hold-ups where coalescence effects would be expected to be least pronounced.

Olney and co-workers have determined not only average values but also radial and axial point values of hold-up in 6 inch and 42 inch diameter R.D.C's. Results for radial profiles in the 6 inch R.D.C. were erratic presumably due to the small dimensions; results for the large column with the system water - toluene are reproduced in Figures 20 and 21. (87).

The hold-up was found to increase initially moving up the column; this presumably represents the finite time for droplet break-up. Hold-up was then substantially constant; the slight reduction observed towards the top of the column was presumably due to the unrestrained discharge of droplets from the top compartment into the settling zone.

The hold-up was found to remain substantially constant with radial position excepting in the vicinity of the rotor shaft as illustrated in Figure 21. This indicates some centrifugal separation of the phases. The characteristic value of X should therefore be taken as that between R/2 and S/2 from the centre; X_{av} has been found not to vary greatly from X_{max} however except at low rotor speeds.

Capacity

i. Empirical Approach

In a series of tests Reman found that there was (13,14,16, 106,109).

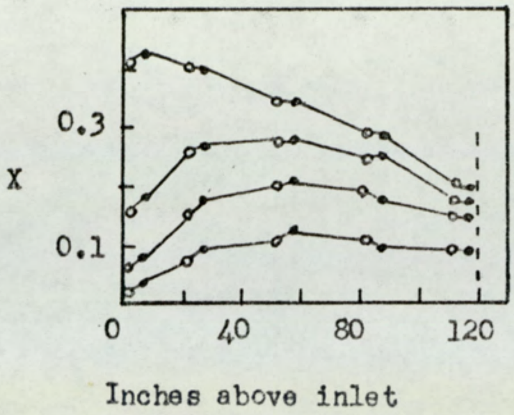
a. A decrease in capacity with increasing rotor speed and increasing diameter of the rotor discs.

b. An increase in capacity with increasing diameter of the opening in the stator rings.

c. An increase in capacity with increasing compartment height.

d. A decrease in capacity with increasing ratio of the flow-rates of the dispersed and continuous phases.

Successful correlation of capacity data was obtained by taking the



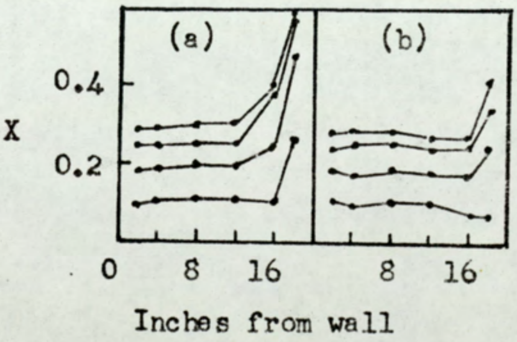
Open points = 2.5 inch above stators

Closed points = 2.5 inch below stators

Rotor speeds, 130, 150, 160 & 170 r.p.m.

FIGURE 20. AXIAL PROFILE OF HOLD-UP (87)

42 inch R.D.C., system water - toluene.



Compartment 9.

(a) 2.5 inch above stator

(b) 2.5 inch below stator

R.P.M. as for Fig. 20.

FIGURE 21. RADIAL PROFILE OF HOLD-UP

42 inch R.D.C., system water - toluene

energy input per unit volume, $\frac{N^3R^5}{HD^2}$ as determining the drop size of the dispersed phase. The effect of restrictions in the compartment caused by stator and rotor discs on settling of the drops and hence capacity was allowed for by simply dividing the observed capacities by a factor C_R. This factor was equal to the ratio of the minimum cross-section to the free cross-section of the contactor, i.e. either S^2 / D^2 or $(D^2 - R^2) / D^2$. Typical results at constant phase ratio are shown in Figure 22.

Whilst successful for specific systems this method, excluding as it does any reference to significant physical properties particularly interfacial tension, is unsuitable for general correlation. Nevertheless a rough guide to power input group operating range has been given of the form shown in Figure 23 (110).

ii. Characteristic Velocity Approach

A more fundamental approach is from the characteristic velocity, Equation 60, discussed earlier. (102). By introducing the condition that at flooding point the flow-rates reach a maximum, by differentiating and setting dV_c/ dX and dV_d / dX equal to zero the following equations result,

$$V_{d\,(f)} = 2\,\overline{V}_n\, X_f^2\,(1 - X_f) \qquad 80.$$

$$V_{c\,(f)} = \overline{V}_n\,(1 - X_f)^2\,(1 - 2\,X_f) \qquad 81.$$

which relate the phase flow-rates at flooding to the corresponding hold-up X_f. By eliminating $\overline{V}_n$ between Equations 80 and 81 a relation between X_f and the flow ratio $\frac{V_{d(f)}}{V_{c(f)}}$ was obtained,

$$X_f = \frac{(L^2 + 8L)^{0.5} - 3L}{4\,(1 - L)} \qquad 82.$$

where $L = \frac{V_{d(f)}}{V_{c(f)}}$.

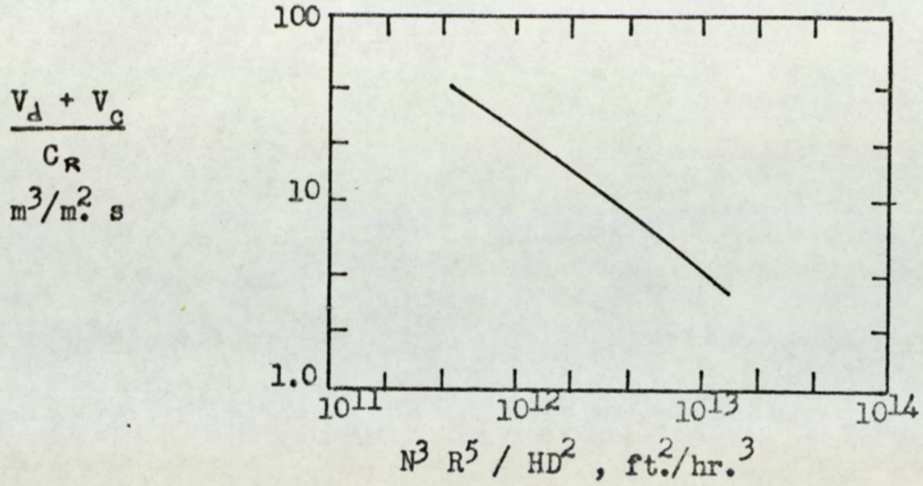


FIGURE 22, CAPACITY DATA, 82 - 200 mm. R.D.C's, system deasphalted residue - furfural (91)

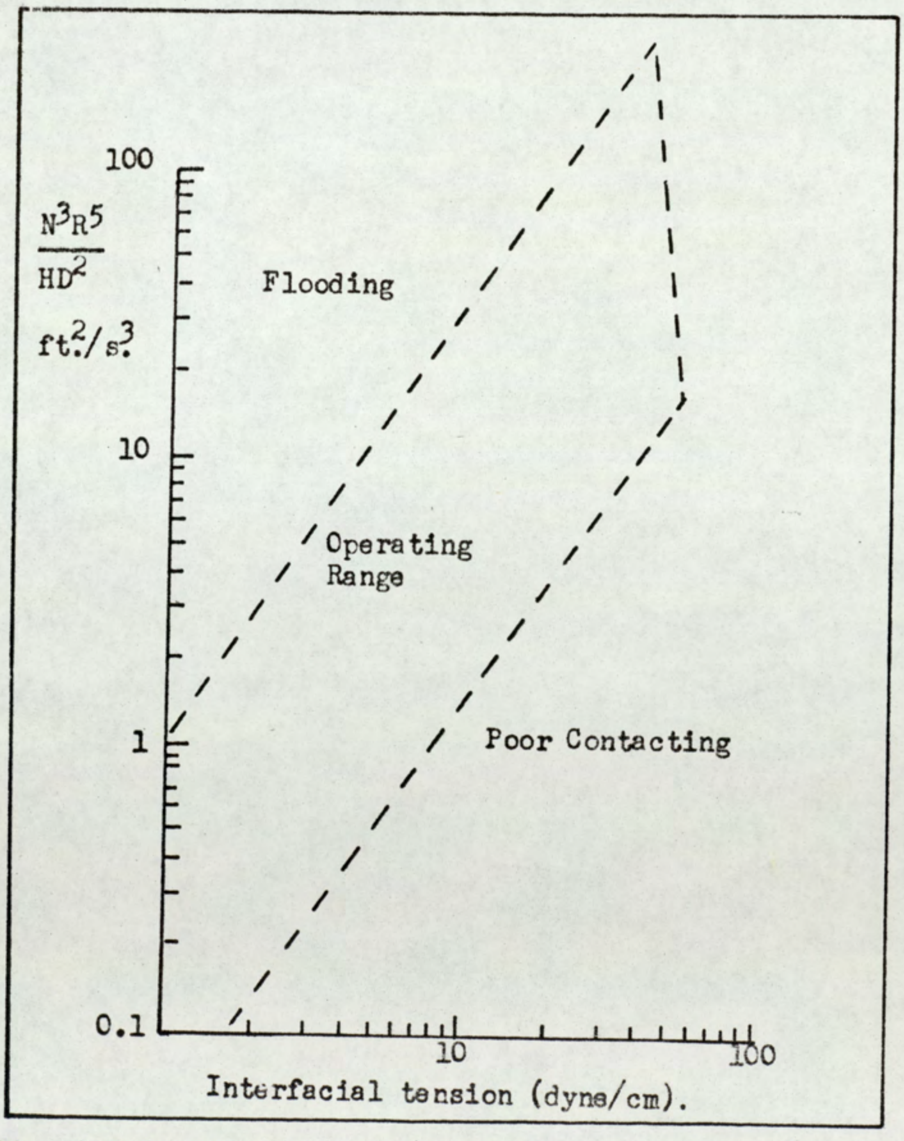


FIGURE 23, R.D.C. POWER INPUT GROUP OPERATING RANGE (110).

Logsdail et.al. (102) evaluated values of $\overline{V}_n$ from their flooding rate data using Equations 80 and 81 and found good agreement with values obtained by inserting experimental X_f values in Equation 60.

Strand et.al. (87) carried out a similar analysis based on Equations 60 and 82 but included the constriction factor C_R. The procedure was found to be valid for columns up to 42 inch diameter. Clearly this an approximation since Equation 60. and this method are applicable only when the mean drop size and hence $\overline{V}_n$ is constant over the entire hold-up region involved. Olney later reported otherwise (111).

Limiting values of hold-up can be similarly calculated using the modified Equation 78 due to Misek (108).

Phase Ratios

Phase ratios, on a continuous : dispersed phase basis, may vary in practice between 0.5 to 1 and 4 to 1.
although Kagan et.al. have employed 5 to 1 (105). In research studies values of 12 to 1 and 16 to 1 (94) and 2 to 1 and 6 to 1 (91) have been employed. Olney employed 1 to 1 (111) as did Reman (16) and the latter recommends 0.3 to 1 as the lower limit for stable operation. Vermijs and Kramers observed that with the system water - acetic acid - M.I.B.K., axial mixing of the continuous phase had a significant effect on efficiency at phase ratios of less than 1 to 1 (103). This effect was independant of rotor speed and hold-up and is significant in that column diameter was only 41 mm.

Axial Mixing

Strand et.al. (87) have measured axial mixing for the continuous phase in columns of 6,25,42 and 85 inches diameter. Within the range $\frac{R.N}{V_c} > 30$ data were correlated by the following equation,

$$\frac{(1 - X)\, E_c'}{V_c\, H} = 0.5 + 0.09\, (1 - X)\, \frac{RN}{V_c}\left(\frac{R}{D}\right)^2 \left[\left(\frac{S}{D}\right)^2 - \left(\frac{R}{D}\right)^2\right] \qquad 83.$$

An analagous form assumed for the dispersed phase was,

$$\frac{X\,E_D'}{V_D\,H} = 0.5 + 0.09\;X\;\frac{R.N}{V_D}\left(\frac{R}{D}\right)^2\left[\left(\frac{S}{D}\right)^2 - \left(\frac{R}{D}\right)^2\right] \qquad 84.$$

The latter gave only fair agreement in a 6 inch R.D.C. using either solid particles in a continuous Kerozine phase or a non-coalescing dispersed Kerozine phase in water.

In later work reported by Stemerding et.al. (112), continuous phase axial diffusivity was correlated for R.D.C.s of 3 inch to 7 ft. diameter by the equation,

$$E_c' = 0.5\;H.V_c + 0.012\;R.N.H\left(\frac{S}{D}\right)^2 \qquad 85.$$

The relation has not been so well established for dispersed phase diffusivity but this apparently varies from one to three times the continuous phase value (90,112).

Design Procedure

Design may be based on predicted mean drop diameters (87,111). An illustration of the design procedure has been given by Jeffreys (70) using data published by Misek (46). Flexibility is then obtained in practice by the provision of a variable speed drive (87,99).

7.5. OLDSHUE-RUSHTON COLUMN DESIGN.

Little data are available on the design of Oldshue-Rushton type columns. With a specific liquid system, at a fixed phase ratio, performance is dependant on,

a. Stator diameter, compartment height and the extent of baffling.

b. The design and location of the agitator.

c. The agitator speed.

Column Geometry

Column diameter is normally 3 times that of the impeller and twice the height of a compartment (113) but a multitude of column geometries have been employed (17,18,92,109). The optimum position for the turbine impeller is midway between the stator rings (17).

The purpose of the stators is to reduce the extent of interstage mixing; thus Gutoff found that decreasing the stator diameter reduced the interstage mixing (92); clearly this can only be achieved at the expense of capacity. A similar effect was obtained by inserting screening across the stator opening, in the fashion of a Scheibel column, or by using alternate mixing and calming sections. In practice the width of each vertical baffle may approximate to 10% of column diameter as in agitated vessels.

Agitator Speed.

Typical efficiency - agitator speed curves showing a definite maximum have been reported for the extraction of acetic acid from M.I.B.K. to water and vice-versa (17). For the extraction of uranium from a continuous nitric acid phase by tributyl phosphate in a 5 inch. column the optimum agitator speed for any given flowrate was found to be 10 to 20 r.p.m. below flooding (18).

The presence of an optimum speed is again attributable to the increased axial mixing at high rotor speeds. Gutoff measured the extent of interstage mixing in a model single phase system (92). A 4 inch column was fed with salt solution at the top and distilled water at the bottom; the dilute salt solution was removed from the top. Mixing between stages was found to be relatively small at low agitator speeds, less than 90 r.p.m., but to increase very rapidly and to a first approximation linearly with speed once the turbulent region was reached. This is supported by the work of Ingham (37).

Hold-up

Little data have been reported on the variation of hold-up with operating parameters in an Oldshue-Rushton column. Some work was done by Misek in a column with stators of similar dimensions to those used in an R.D.C. and agitators consisting of 2 - bladed paddles. Two different compartment heights were examined (108). The results were claimed to be capable of correlation by means of Equation 78.

Reference has been made by Treybal to work with either toluene or kerozine dispersed in water (93). The characteristic velocity term $\bar{V}_n$ in equation 59, was found to be correlated by the expression,

$$\frac{\bar{V}_n \mu_c}{\sigma} = 1.77 \times 10^{-4} \left(\frac{g}{R.N^2}\right) \left(\frac{\Delta\rho}{\rho_c}\right)^{0.9} \qquad 86.$$

Capacity

Oldshue and Rushton observed that flooding in their column was similar to that in other columns. Limiting flow rates were determined over a limited range with one system (17).

Phase Ratios

Phase ratios have been of the order of 0.6 to 1 : 1 (17,18); the limiting phase ratios in practice would be expected to be of a similar order to those given for an R.D.C. earlier.

Axial Mixing

Bibaud and Treybal extended the study of Gutoff to a two phase system and measured the axial mixing in both phases (93). With an aqueous continuous phase and toluene or kerozine dispersed, the continuous phase diffusivity E_c' was correlated by,

$$\frac{X.\ E_c'}{V_c\ H} = -\ 0.14 + 0.0268 \ \frac{R.N.X.}{V_c} \qquad 87.$$

A similar correlation was not found possible for the dispersed phase diffusivity E'_D and the relationship,

$$\frac{R^2 N}{E'_D} = 3.93 \ (10^{-8}) \left(\frac{R^3 N^2 \rho_c}{\sigma}\right)^{1.54} \left(\frac{\rho_c}{\Delta\rho}\right)^{4.18} \left(\frac{R^2 N \rho_c}{\mu_c}\right)^{0.61} \qquad 88.$$

was applied with limited success.

Design Procedure

On the assumption that equations 57 and 83 may be used to predict hold-up, estimation of a mean droplet diameter and a coefficient of mass transfer enables a fundamental design procedure to be employed for the Oldshue-Rushton column. Treybal (93) has interpreted his data as indicating that formulae applicable to rigid spheres can be used to predict k_c and k_D. This would seem to be incorrect and may have resulted because drop sizes were not measured in situ but were estimated from the work of Thornton (74) which is applicable only to agitated vessels.

Current practice is for these columns to be designed using the concept of theoretical stages with stage efficiencies based on experience (113).

7.6. PERFORMANCE COMPARISONS

Performance comparisons can only be drawn for the same liquid-liquid -solute system; thus a number of claims made for 'improved' designs may be misleading (23). Account must be taken of both the efficiency and volumetric capacity. Therefore Reman (13) proposed an index of effectiveness of operation,

$$Ie = \frac{\text{total throughput}}{\text{volume of one theoretical stage}} \qquad 89.$$

Vermijs and Kramers (103) have used the effectiveness index to compare their own data with that of others (16,17,86) for the system water - acetic acid - M.I.B.K. In general for this system, over the range of variables covered, the R.D.C. and Oldshue-Rushton contactors were found to have a similar effectiveness in the range 1.6 to 3.4 min^{-1} . The Scheibel columns showed rather lower figures, 1.4 to 2.2 min^{-1} ; in effect they have relatively small H.E.T.S. values but the capacity is lower than the other two types.

Ponikarov et.al. (114) employed a similar index to compare the performance of spray and packed columns, sieve plate columns, the R.D.C. and pulsed plate columns. For one system they found that the R.D.C. whilst not so effective as the pulsed plate column possessed a lower H.E.T.S. value.

Clearly, from preceeding sections, proper comparisons can be made only if the selection of the dispersed phase, solute concentration, phase ratios and the direction of mass transfer are identical. Even then effectiveness must be expected to vary to different degrees on scaling up the various designs. Nonetheless some workers have drawn comparisons between the efficiencies of rotary agitated columns and other continuous columns. The results of Ziolkowski (115) with two systems confirm that the R.D.C. has an efficiency of similar order to a pulsed plate column.

In practice other factors may govern extractor selection. Thus the R.D.C. is preferred for systems prone to emulsification since it has controllable shear characteristics (99). Similarly, whereas liquids containing suspended solids can be satisfactorily handled in an R.D.C. (116) and presumably in an Oldshue-Rushton column, they may present problems in the Scheibel, or Wirz, columns which cannot be overcome by a simple back-flushing technique (10). Maintenance of moving parts may also provide a problem in the Scheibel column.

7.7. MISCELLANEOUS MASS TRANSFER STUDIES

A number of workers have performed efficiency and capacity studies with specific systems and column designs. The results have been treated empirically, are specific to the systems studied and add little to the fundamental understanding of either the hydrodynamics or solute transfer processes involved. However, for completeness, the relevant papers are summarised in Appendix 2.

7.8. LIMITATIONS OF DESIGN METHODS

It has been shown in para. 7.1. that the assumption of spherical droplets of uniform size enables an interfacial area a, contact time t and transfer coefficients k_c and k_d to be defined for a mean droplet. In practice however wide distributions occur in drop sizes.

The distribution arising from the initial disperser is of minor importance. In the contact zones the distributions arise from the competing effects of droplet break-up and coalescence.
The equilibrium attained depends upon the characteristics of the particular contactor. Two limiting situations exist,

(a) A distribution of drop sizes and residence times, i.e. little droplet coalescence.

(b) A size distribution of droplets all of approximately the same age, i.e. equal probability of repeated coalescence and redispersion among drops of all sizes.

Dependant on the prevailing break-up processes the size distribution is bounded by a maximum stable size and a minimum size. In a non-coalescing environment, case (a), if back-mixing is significant the smallest drops, which remain in the contact zone for longer than average, become highly back-mixed. Large drops pass rapidly through the contact zone and are less affected. The effects of back-mixing cannot therefore be analysed without reference be droplet size distribution. It follows that the assumption that E'_D is constant throughout a column may also be invalidated. In so far as continuous phase back-mixing is due to transportation of continuous phase in the wake of drops then E'_C could also be influenced by droplet characteristics.

It is apparent from single droplet studies referred to in para. 6.1. that the dispersed phase film coefficient is dependant on the droplet size distribution. Thornton (47) has pointed out that in a non-coalescing environment the mass transfer process with small droplets is governed by essentially molecular diffusion; for larger droplets it is independant of molecular diffusivity. Solute transfer therefore occurs simultaneously by both molecular and eddy diffusional mechanisms and the preponderance of either mechanism is dependant on the droplet size distribution.

These conclusions may be invalidated if droplet coalescence-redispersion effects occur to any significant extent in the contactor. Such effects greatly enhance the rate of mass transfer due to the complete remixing which occurs and to the additional turbulence set up within the droplet. This has been demonstrated in a mixer-settler unit (74). The magnitude of such effects is partly dependant on the droplet size distribution. Additionally, centrifugation of the dispersed phase such as occurs in an R.D.C. (87), may cause a shift of smaller droplets, which should coalesce more readily, to the periphery.

It is clear therefore that liquid-liquid extraction occuring in an agitated column contactor can only be described fortuitously by a transfer mechanism based on non-coalescing droplets of mean diameter. The problems are accentuated in those cases where extraction produces a significant change in phase ratios along the column. For these reasons, and to account for any subsequent variations in flowrates, concentrations and physical properties of process streams it is customary for agitated columns to be provided with variable speed agitator drives (113).

8. DROPLET CHARACTERISTICS IN ROTARY AGITATED COLUMN EXTRACTORS.

8.1. GENERAL

8.1.1. Effect of Impurities or Surface Active Agents

It follows from the summary given in para. 4.2. that the presence of impurities or of surface active agents will have a marked effect on droplet characteristics and hence on contactor efficiency and capacity. This has been overlooked in some otherwise impeccable researches. For example, Vermijs and Kramers (103) used natural rubber connectors and neoprene rubber packings with the system water - acetic acid - M.I.K. and commented only that both needed renewal at frequent intervals. Lee and Lewis also risked leaching of impurities from neoprene gaskets (126). Logsdail et.al. used flexible connections of rubber or P.V.C. (102). Similarly Piper experienced leaching of plasticiser from P.V.C. tubing with Kerozine (96). Contamination of this sort has recently been shown to significantly alter coalescence phenomena and lead to unreproducible results (67,127).

Care must be taken to select a solvent not subject to degradation. Otherwise in addition to extraneous impurities, the accumulation of products of solvent degradation at the interface can also effect interfacial tension.

Presence of a surfactant directly influences the interface as well as drop size and movement. The lowered interfacial tension results in smaller droplets and retardation of coalescence has a similar effect (69). In a spray column, hold-up has been found to increase by a factor of 2 to 2.5 for a coalescing system and 1.4 to 1.6 for a non-coalescing system (128). Surfactants retard circulation in droplets as described in para. 5.1. the agent being collected like a cap at the rear of the drop. In practice some increase in mass transfer generally accompanies the addition of surfactants (69).

Davies et.al. (94) have studied the effect of a surface active agent on extraction efficiency in an R.D.C. Teepol was used to obtain an 18% reduction in interfacial tension with the system Kerozine - water - phenol. The equivalent number of stages in the R.D.C. were then compared at different rotor speeds with the number in the absence of Teepol. The results are illustrated in Figures 24 and 25. Smaller droplets, giving a large interfacial area and a considerable increase in extraction efficiency, were observed when Teepol was present. Column capacity was reduced due to the small drop sizes. Recent work has indicated that any sample of industrial chemicals prepared in bulk contains some materials which affect the properties of the interface when the sample is dispersed in an aqueous medium (129). Thus process development is preferably carried out with actual plant feed streams. The use of impure commercial materials for research (44,46) is inadvisable however because of these contaminants and the potential variation in specification.

On a commercial scale surface active contaminants have led to the formation of a suitable dispersion in an R.D.C. at zero rotor speed (99). Reman (109) has reported that when extracting heavy distillates with furfural, emulsification caused by pitch contamination may reduce R.D.C. capacity by a factor of 2.5. The absence of droplet coalescence in R.D.C.'s during the extraction of phenols from a continuous aqueous phase with Fenosolvan or benzene has been attributed to the presence of surface active contaminants (119).

8.1.2. Effect of Mass Transfer

The effects which mass transfer may have on droplet characteristics have been discussed earlier.

To eliminate mass transfer from hydrodynamic studies it is necessary that the phases be mutually saturated before entering the contactor. This can be achieved by pre-contacting the phases and allowing time for separation to occur.

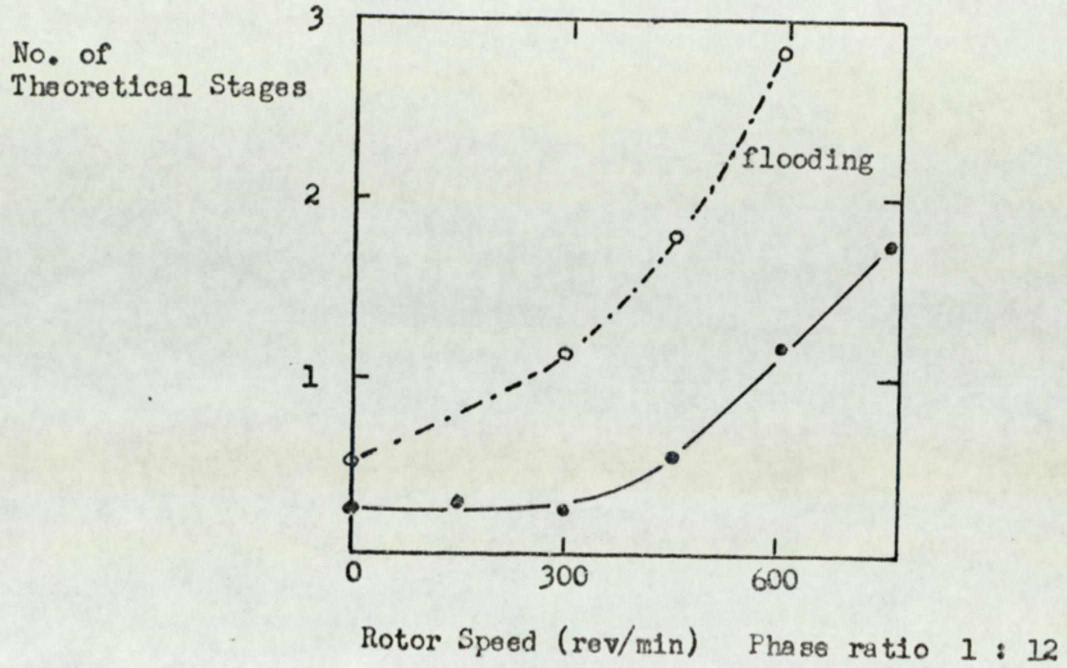


FIGURE 24. R.D.C., HYDROPHOBIC SURFACES.
EFFECT OF SURFACE - ACTIVE AGENT (94)

(Solid points - no additive, open points - Teepol added)

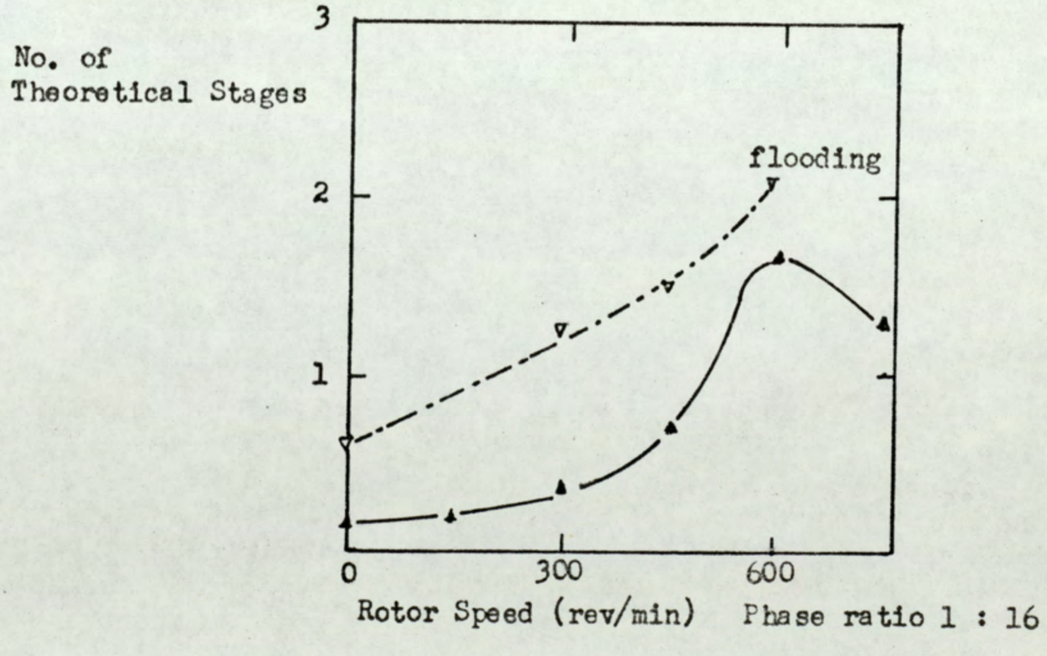


FIGURE 25. R.D.C., HYDROPHILIC SURFACES
EFFECT OF SURFACE - ACTIVE AGENT (94)

(Solid points - no additive, open points - Teepol added)

Some systems are notoriously slow to coalesce, produce a secondary haze or fog on settling and several hours may be necessary before clear phases are obtained. Thus the procedure sometimes adopted (109) of circulating the phases through the column or in a closed circuit prior to use may be open to criticism.

8.2. DROPLET CHARACTERISTICS - SCHEIBEL COLUMN

Using the system M.I.K. - acetic acid - water, Karr and Scheibel observed that when the light organic phase was both the dispersed phase and the extractant a much finer dispersion and hence a higher hold-up was obtained at a given agitator speed than for the case of M.I.K. dispersed - water extractant (98). This yielded a tenfold greater mass transfer coefficient. Higher mass transfer coefficients when solute transfer was from the continuous to the dispersed phase were also obtained with two other systems. These and similar results referred to by Hixson (99) are in agreement with the coalescence phenomena described in para. 4.2.

Droplet characteristics as a significant factor in the selection of packing was mentioned in para. 7.3. Having regard to the drop size distribution generated in the mixing section it appears from the work of Piper (96) and Honekamp and Burkhart (9) that droplet behaviour in the packing comprises three effects, viz,

(a) Very small drops pass through the packing without change.

(b) Drops small compared to the interstices size. These drops attain a predictable 'characteristic' drop size after Pratt (49,66).

(c) Observable with a high interfacial tension system only, large drops compared to the interstices size. These droplets coalesce at the leading edge of the packing to form large globules which move through the packing in stages and finally leave by drip point formation.

The preponderance of any one effect would be expected to depend on the properties of the system and the degree of agitation, i.e. the initial drop size distribution, and on the characteristics and interstices size of the packing.

Some limited data on drop size in mixing sections has been given (96, 9). With the system M.I.B.K. - water, Honekamp and Burkhart reported only the average drop size over six different flow rate combinations as shown in Figure 26.

8.3. DROPLET CHARACTERISTICS - ROTATING DISC CONTACTOR

Davies et.al. concluded from work on a pilot scale R.D.C. that the high rate of mass transfer was due primarily to the maintenance of a large interfacial area assisted by the intense agitation produced at the liquid-liquid interface by droplet spin (94). It follows from para 5.1.4. that with the system studied the continuous phase was controlling. A large concentration driving force was employed and therefor interfacial turbulence effects probably enhanced mass transfer (71). The only evidence for droplet spin is the very limited early work of Howells and Steen (95).

In contrast to their work with the same contactor, Davies found that coalescence and break-up of the dispersed phase was not a significant factor. These results however were obtained with phase ratios of 12 to 16 : 1 which are uncommon in practice; 0.5 to 4 : 1 is the usual operating range (16,99). Clearly such a high proportion of continuous phase would greatly reduce the probability of droplet collision and possible subsequent coalescence.

Logsdail et.al. (102) suggested from visual observation that, in the absence of undistributed solute, coalescence of the droplets was negligible as dispersed phase hold-up was increased progressively to the flood-point. (They also suggested however that the mean droplet size remained substantially constant with increasing dispersed phase hold-up which is now disproved) A similar result was suggested when

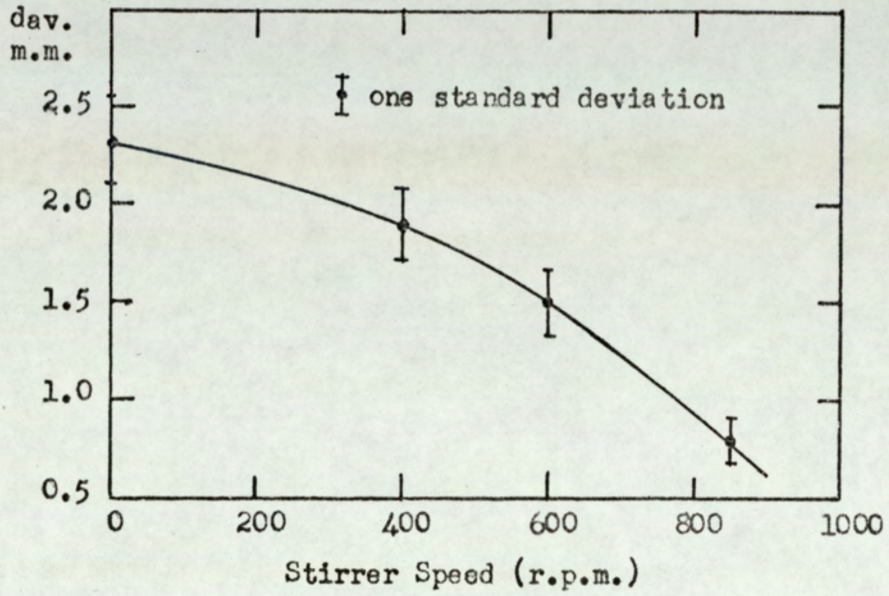


FIGURE 26, AVERAGE DROP DIAMETER IN MIXING SECTION

system M.I.B.K. - water : (9).

solute (acetone) was initially present in the continuous (aqueous) phase. When the solute was present in the dispersed phase however the droplets were noticeably larger; this has been confirmed by Strand et. al. (87) and is in agreement with the coalescence theory summarised in para. 4.2. The effect of varying amount of solute in the dispersed phase on drop diameter was not determined but from visual evidence with acetone or acetic acid as solute it was suggested that the diameter increased rapidly with solute concentrations up to 1 - 2 % and beyond this point the increase became less pronounced.

The actual flow pattern of dispersed phase droplets has been observed to differ from that shown in Figure 3. (83). The two general flow patterns observed are shown in Figures 27 and 28. With the system toluene - water, at peripheral rotor speeds less than 300 $ft.min^{-1}$, Figure 27, the flow of liquid was countercurrent with a rotation of the whole liquid mass around the rotor shaft. Layers of droplets were trapped under the rotor discs and stator rings. The re-circulation loops shown in Figure 3. were not evident and hence back-mixing was less severe. At speeds in excess of 300 $ft.min.^{-1}$, Figure 28. the pattern was similar to Figure 3. but with less difference in quantity between the portion of liquid re-circulated and the main body of flow. At the vicinity of the flood point the flow pattern of the dispersed phase could be represented by Figure 3. Kung and Beckmann recommended this critical peripheral speed as being a minimum for all systems. However, the physical properties of the system and the material and construction of the disc would be expected to be important parameters. In the event its use for systems of low interfacial tension, and in particular systems not including an aqueous phase, has been found to give too fine a dispersion (71).

Droplet characteristics have been studied photographically by Olney in a 6 inch R.D.C. (111). A series of drop size - cumulative volume curves were obtained for five systems of which Figure 29. is typical. The maximum stable drop size, d_m, is shown on the right

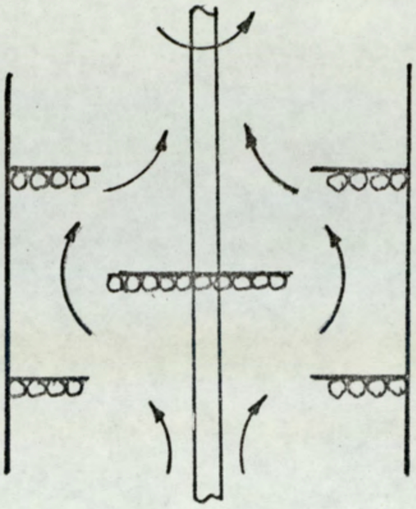


FIGURE 27. DISPERSED PHASE FLOW PATTERN

<300 ft/min. peripheral speed (83)

R.D.C., system toluene - water

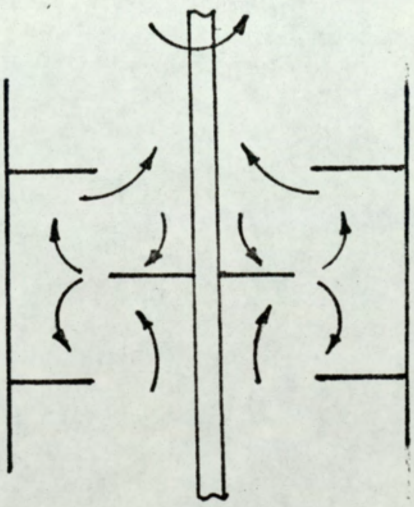


FIGURE 28. DISPERSED PHASE FLOW PATTERN

>300 ft/min. peripheral speed (83)

R.D.C., system toluene - water

hand scale. For all systems, drop sizes were found to increase over the whole spectrum with increased hold-up. Olney considered this to have been due to both higher drop interaction rates, associated with smaller interparticle distances, and the varying dispersion effect of the specific power input, i.e. the power per unit mass of dispersed phase,

$$P_D = \frac{\rho N^3 R^5}{\rho_D X.H.D^2} \qquad 90.$$

Values of d_{32}/d_{50} were found to range from 0.79 to 1.06.

Size distribution data was analyzed and compared with an upper limit distribution defined in terms of maximum drop size d_m. The special upper limit function as applied to log - probability distributions is given by,

$$\frac{dv}{dy} = \frac{\delta}{\sqrt{\pi}} e^{-\delta^2 y^2}$$

where $y = \ln \left(\frac{a.d.}{d_m - d} \right)$

Data from five systems gave straight line plots of $\left(\frac{d}{d_m - d} \right)$ vs v on log-probability paper, from which were obtained,

$$a = \text{skewness parameter} = \frac{d_m - d_{50}}{d_{50}}$$

$$\delta = \text{uniformity parameter} = \frac{0.907}{\ln_e \left[\frac{d_{90}}{d_m - d_{90}} \cdot \frac{d_m - d_{50}}{d_{50}} \right]}$$

The uniformity parameter determines the spread of the distribution, a value > 1 indicating a wider range of drop sizes larger than d_{50} and a smaller value indicating a wider range of drop sizes. For the systems studied, δ was in the range 0.60 to 1.04.

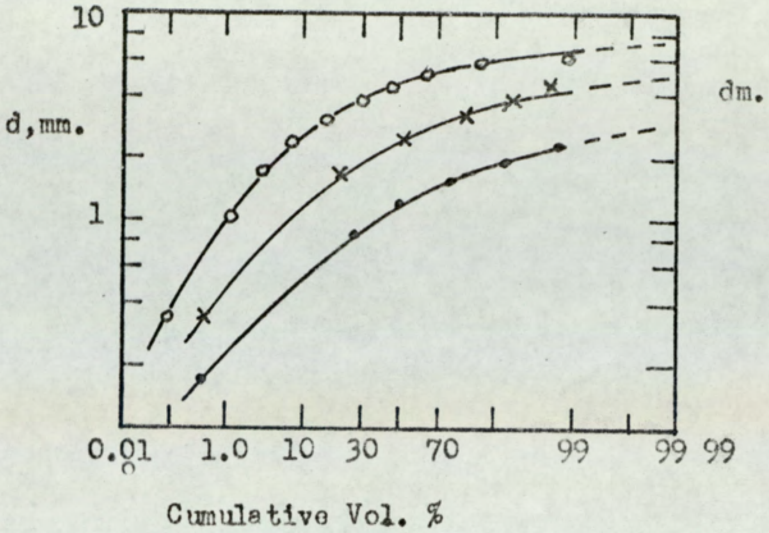


FIGURE 29. DROP SIZE DISTRIBUTIONS,

6 inch R.D.C., system 68% kerozine/ 32% CCl_4 (dispersed) - water ; (111).

r.p.m.	hold-up	symbol
400	11 %	○
550	10 %	×
700	11 %	•

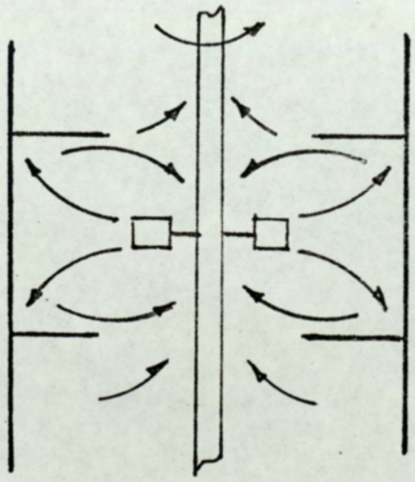


FIGURE 30. IDEALISED FLOW PATTERN IN AN OLDSHUE-RUSHTON COLUMN (17)

In earlier work (87) drop hold-up in the range 5 to 20 % was found to have no significant effect on the maximum stable drop size d_m and results were not reported in detail. Variation of d_m with rotor speed gave straight lines of measurable slopes when plotted on log-log scales. Data for nine systems were compared with the Hinze equation 6. Power/mass was defined as ,

$$E = \frac{4}{\pi} C_2 \frac{N^3 R^5}{H D^2} \qquad 91.$$

where C_2 is the coefficient in the power number,

$$C_2 = \frac{P}{n \rho N^3 R^5} \qquad 92.$$

When the organic phase was dispersed, values of C_2 fell in the range 0.4 to 0.6. The effect of E was greater than given by the exponent $- 2/5$, poorest agreement being with viscous oil dispersion. Agreement was generally poor at low rotor speeds i.e. when drop sizes were large such that conditions of local isotropy were improbable.

8.4. DROPLET CHARACTERISTICS - OLDSHUE-RUSHTON COLUMN

From observations of a coloured acid-base indicator with the system M.I.K. - water - acetic acid, Oldshue and Rushton concluded that each compartment in their 6 inch column had a distinct and uniform concentration i.e. that the column comprised a series of distinct stages (17). Since their approach was unrelated to either hold-up or rotor speed however it is an over-simplification.

Figure 30. shows the idealised radial flow pattern for dispersed phase droplets (17). Clearly in practice this is dependant on impeller speed. There has been no reported work on droplet characteristics or flow patterns in this column.

9. EXPERIMENTAL METHODS FOR DROPLET STUDIES AND THE MEASUREMENT OF INTERFACIAL AREA.

9.1. PHOTOELECTRIC TECHNIQUES

Interfacial area may be related directly to the amount of light transmitted through a dispersion. This technique has been used by a number of investigators with batch agitated vessels (43,44,79,80, 130) and a number of correlations proposed between the ratio of incident to emergent light and the interfacial area. Light transmission readings may be calibrated by high speed photography of the dispersion off which drop counts enable the evaluation of interfacial area per unit volume.

The method necessitates immersion in the extractor of a probe containing a light source and a photo-cell. In column contactors the potential disturbance of the flow patterns may prohibit its use (9). The main disadvantage however is that only the average area/unit volume or mean drop size is determined.

9.2. HIGH SPEED PHOTOGRAPHY

The sizes of dispersed droplets in vessels or glass columns have been measured photographically by a number of workers. The method, although tedious, produces no interference with operation and provides information on drop size distribution, distortions and interactions and on hold-ups.

With glass columns horizontal distortion of the photographed drops may be eliminated by using a square section or alternatively by inserting a round column inside a square glass tank filled with water (131). More often droplets are photographed in a plane just inside a round column; the effects of distortion may be neglected (9,87,96). Exposure times of the order of 0.001 seconds have been found necessary to give good droplet images in agitated columns (9,87,95).
A suitable photographic probe may be employed to photograph dispersions

in situ. (74,130).

9.3. USE OF RADIOISOTOPES

The use of radioactive isotopes has been suggested for the measurement of interfacial area (9). Short range particles emitted by a selected radio-isotope restricted to one phase react with a second phase only in the vicinity of the interface. If the minimum dimensions of the second phase are large compared with the range of the particle the reaction rate is directly proportional to the interfacial area. Reference is made to one experimental system but clearly the problems of stringent design and operating requirements and the need for complete phase immiscibility render this technique of limited application.

9.4. OTHER METHODS

An electronic particle counter has been used to determine the distribution of drop diameters in a dispersion (132). The instrument determined the number and size of droplets suspended in an electrically conductive continuous phase by forcing the dispersion through a small aperture between two electrodes. The resistance between the electrodes changed as a droplet passed through the aperture this change being converted into a pulse which was scaled and counted. Work was limited to distributions in the vicinity of a turbine in the batch mixing of dilute iso-octane and salt water. Clearly the technique could be more widely used within the limitations imposed by a conductive continuous phase.

Resnick has recently described an encapsulation and sampling method (81). Encapsulation was performed by a polymerization reaction between terephthallic acid chloride monomer dissolved in a dispersed carbon tetrachloride phase and piperizine dissolved in continuous aqueous phase in a sampling device. Encapsulated sampled drops were measured directly under a microscope or by photography.

10. DESIGN OF EXPERIMENTS

10.1. PURPOSES OF INVESTIGATION

Models based on the behaviour of single discrete mean droplets do not account adequately for droplet break-up and coalescence phenomena in agitated contactors. The purpose of this investigation therefore was to obtain new evidence of droplet characteristics in the various columns and to assess the practical limitations of the designs and design methods.

It has been found that droplet size distributions are not directly affected by continuous phase flowrates in the R.D.C. (108) or Scheibel columns (9). Therefore, to reduce the number of operating parameters, all drop size measurements in this work were made at zero continuous phase flowrate. Variation of hold-up at any rotor speed was obtained by variations in dispersed phase flowrate.

10.2. SELECTION OF LIQUID-LIQUID SYSTEMS

Three stable systems of widely different interfacial tensions were selected to cover the range over which agitated extractors are best employed. For any specific application tests with in-plant systems would be essential (1).

The systems selected and the reasons for their choice were as follows:-

Toluene - water

a. High interfacial tension, i.e. > 30 dynes per cm.

b. Availability at low cost to a fixed specification. Other workers have used Kerozine which is of variable composition and subject to oxidative degradation (44,96).

c. No known chronic toxicity (133) and a relatively high b.pt.

d. Some results of other workers with the same system facilitated correlation and comparison of results (83,102,108).

Methyl - isobutyl Ketone and water

a. Low interfacial tension, i.e. of the order of 10 dynes per cm.

b. Relatively low toxicity (133) and a high b.pt.

c. Availability at low cost to a fixed specification and stable over a long period (17).

d. Some results with the same system were available for comparison purposes (9,17,98,103)

Iso - octane (2.2.4. Trimethylpentane) - water

a. Very high interfacial tension, i.e. > 50 dynes per cm.

b. Availability to a fixed specification.

c. Low toxicity and a high b.pt.

G.P.R. grade iso-octane was selected since additives present in aviation gasoline affect coalescence phenomena (127).

The specifications to which the organic materials were purchased and their relevant physical properties are given in Appendix 4. Because of their known toxic and lachrymatory effects and flammability a leak proof apparatus and good natural ventilation were necessary.

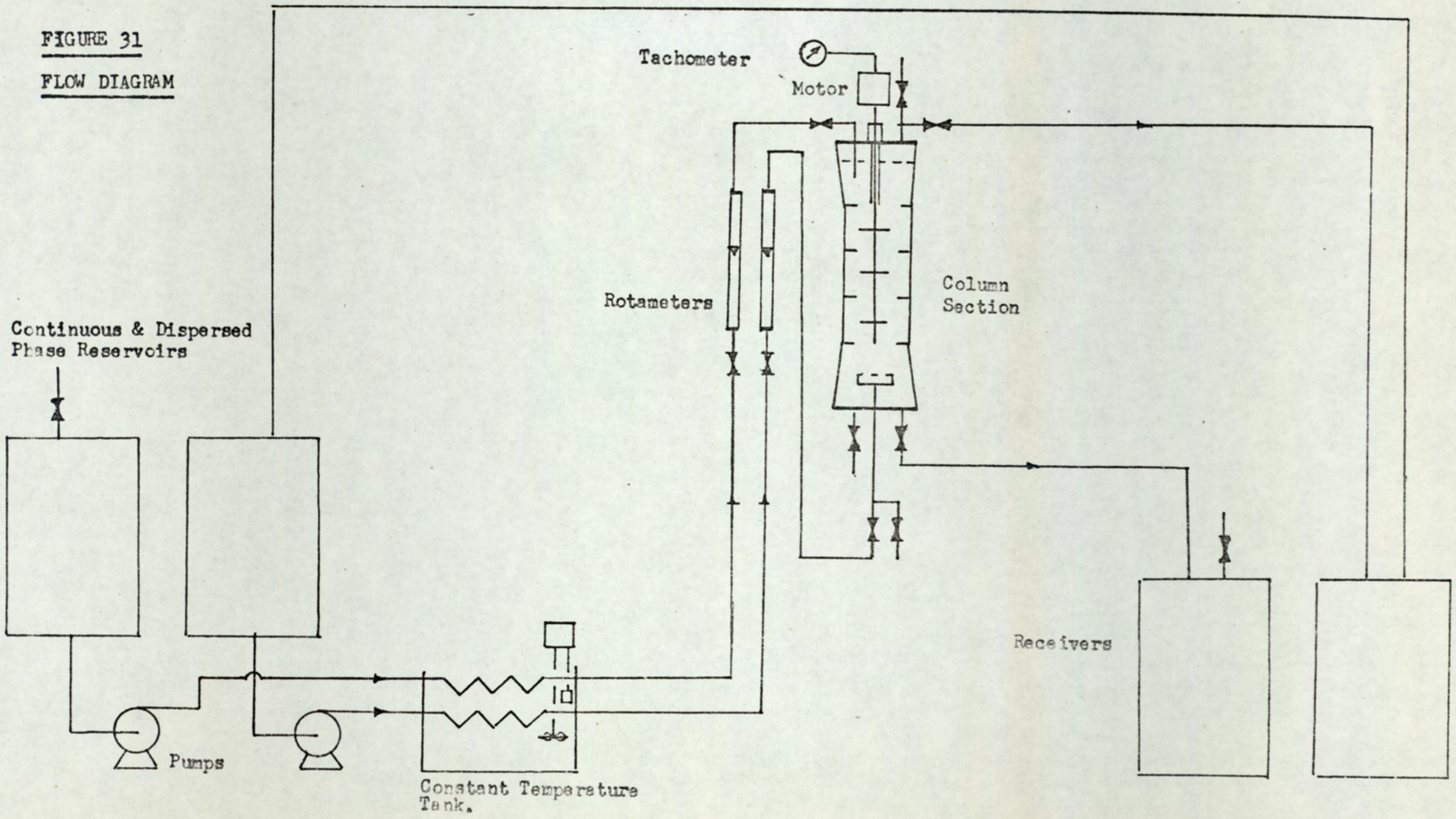
10.3. DESCRIPTION OF EQUIPMENT

A flow diagram of the equipment is shown in Figure 31. The general arrangement was as shown in Figures 32 and 33; this was designed to provide maximum accessibility to the column section for observation, photography and maintenance without restricting the operation of control valves or switches, or the observation of instruments mounted on the control panel.

Feed vessels and receivers comprised 20 litre glass aspirators to facilitate cleaning. Transfer of materials from the feed vessels was by means of two Stuart Turner No.10 centrifugal pumps each capable of pumping 120/140 g.p.h. against 5/20 ft. head of water.

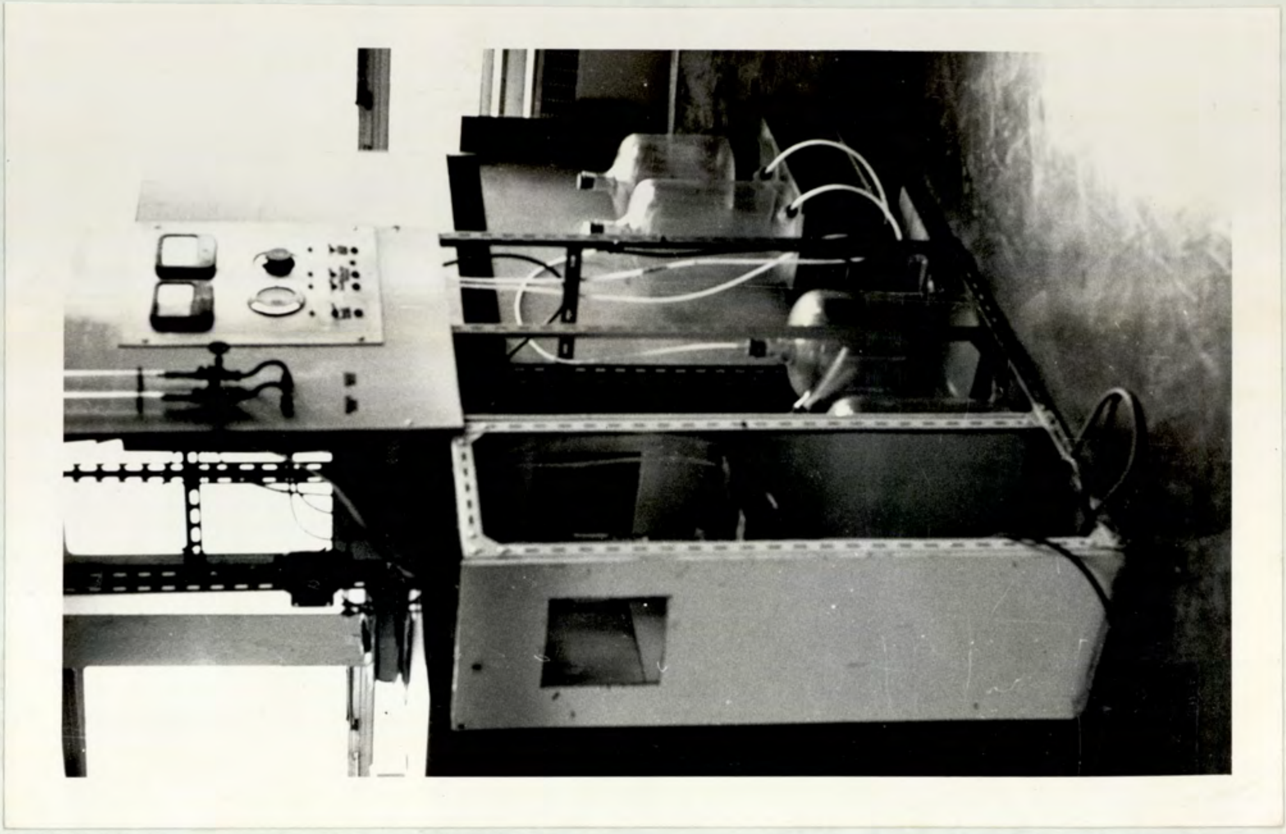
FIGURE 31

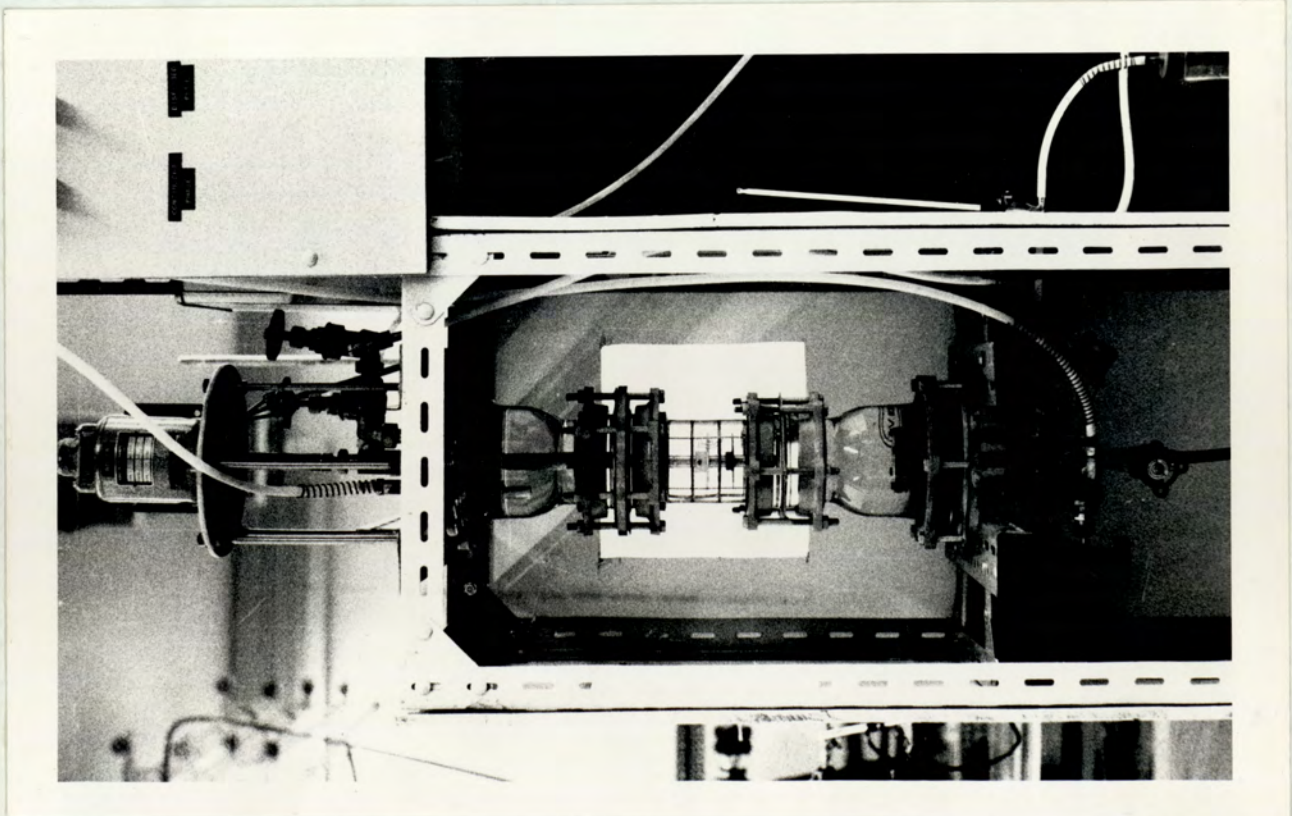
FLOW DIAGRAM



FIGURES 32 and 33.

GENERAL ARRANGEMENT OF EQUIPMENT.





All interconnecting piping consisted of 5/16 inch bore p.t.f.e. tubing so as to be completely inert. Screwed joints, where necessary, were made with p.t.f.e. tape. Neoprene bungs in aspirators were fitted with simple p.t.f.e. sheaths.

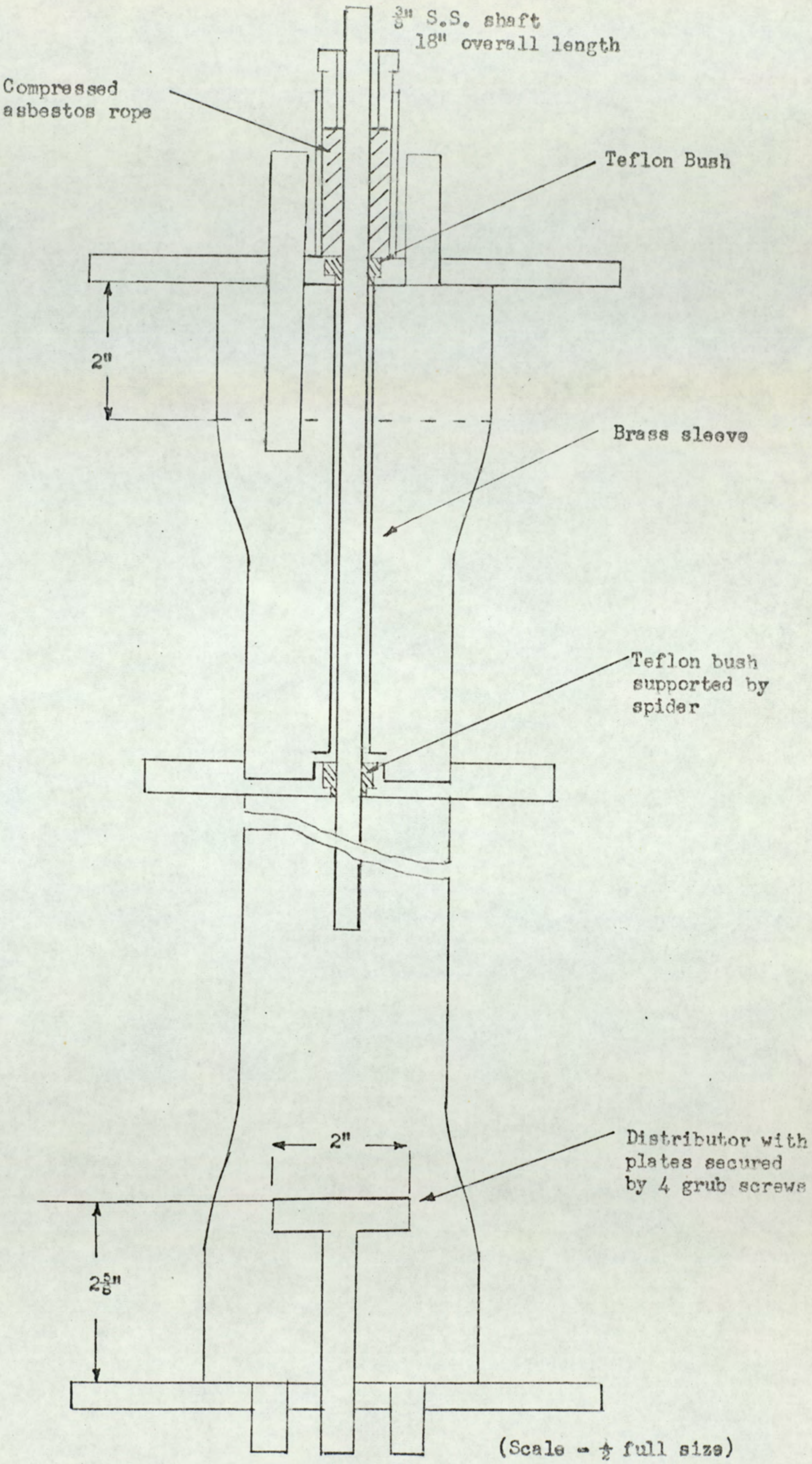
The feed aspirators were mounted on a 1 ft. high plinth to ensure priming of the transfer pumps. Brass valves of the ¼ inch globe type were used for all applications excepting fine control of flowrates. For this duty ¼ inch needle valves were installed. Flowrates were measured by means of independantly calibrated Type 10 S rotameters with stainless steel floats.

Temperature control of dispersed and continuous phase feeds was achieved by passage through copper coils immersed in a water bath fitted with a Tempunit Thermo-regulating unit. Temperature was indicated by $0^\circ - 110^\circ$C mercury in glass thermometers at the outlet from the column. The column section was shielded on 3 sides by blockboard to eliminate extraneous draughts. Control was achieved within ± 1 C with this arrangement.

The column section is detailed in Figure 34 and illustrated in Figure 35. The test section consisted of a 6 inch Q.V.F. glass pipe section of 3 inch nominal bore. This diameter was selected as the minimum suitable without wall effects becoming significant. The upper settling zone comprised a 4 inch to 3 inch Q.V.F. glass pipe reducer. A disc of 12 mesh steel gauze was installed horizontally at approximately 2 inches below the top plate to assist in separating the settling zone from the feed inlet. The continuous phase inlet projected 1.5 inches below this.

The agitator shaft was ⅜ inch stainless steel rod and was flexibly coupled to a 1/30 H.P., 2000 r.p.m. electric motor. The shaft was located in a p.t.f.e. bush in the top plate and in a similar bush supported in a brass holder at the junction of the glass sections. The holder was carefully machined so as to fit flush with the column

FIGURE 34. DETAIL OF COLUMN SECTION.



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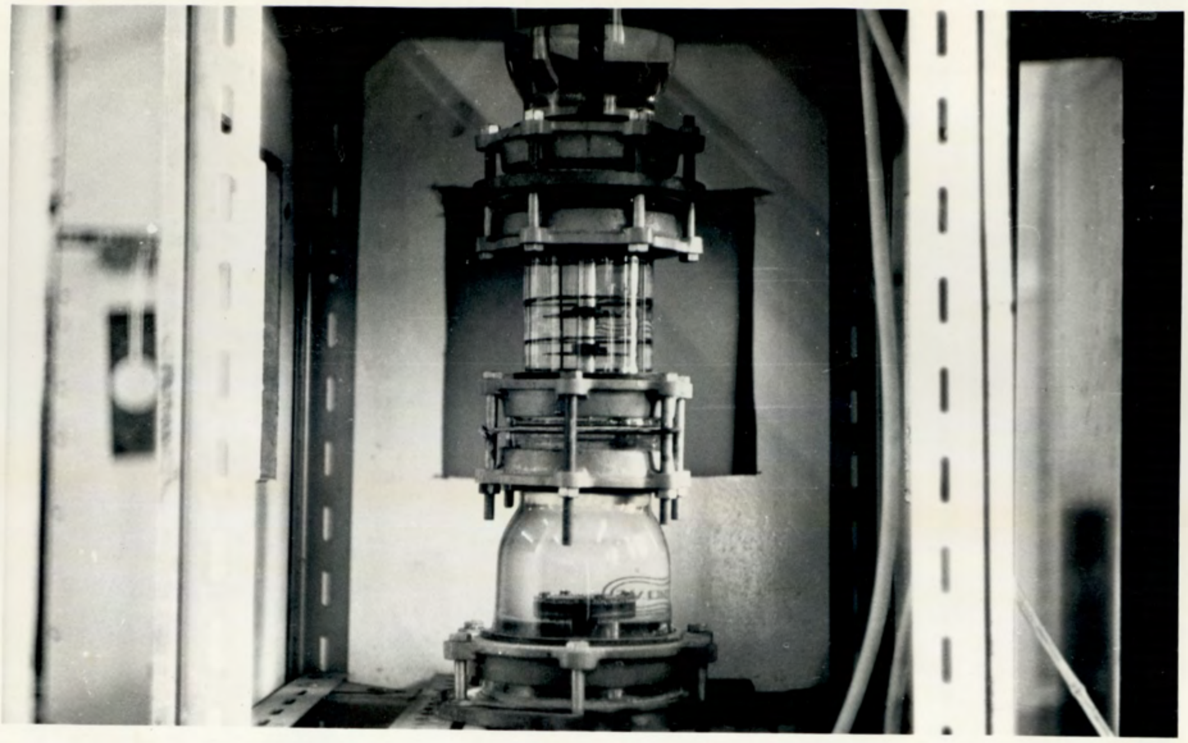


FIGURE 35. ILLUSTRATION OF COLUMN SECTION.

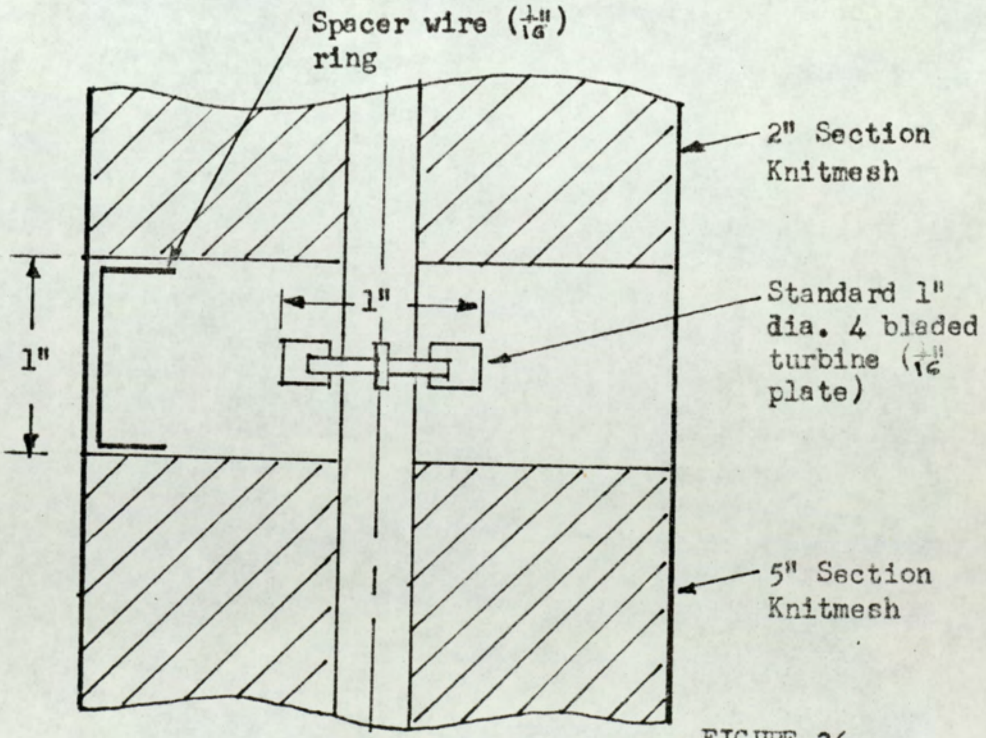


FIGURE 36

SCHEIBEL COLUMN SECTION.

(Scale - full size)

walls. A stationary brass sleeve surrounded the shaft between the lower bush and the top plate to eliminate turbulence in the settling zone. A simple stuffing box assembly containing wound asbestos rope and having a variable compression lock-nut was employed as a shaft seal.

Agitator speed was controlled by means of a Variac voltage regulator. Speed was indicated on a 0 - 1000 r.p.m. tachometer mounted on the control panel and permanently coupled to the top of the shaft via a flexible drive.

The dispersed phase was introduced via a brass disperser located centrally in a brass end plate. The disperser was designed as recommended by Treybal (11); provision was made for changing the perforated plate. The disperser was located in a 4 inch to 3 inch Q.V.F. glass pipe reducer to eliminate any effect of continuous phase flow on droplet formation.

The lower column supports were designed so as to facilitate sliding out the disperser section, bottom plate and fittings as one and the bottom 6 inches of the agitator shaft was made removable. This enabled column internals to be altered without disturbance to the agitator shaft bearings or stuffing box assembly. It was subsequently found convenient, because of the tolerance on internal diameter, to use separate glass sections for each column section.

10.4. CONTACTOR DESIGN

10. 4.1. Scheibel Column

One stage of a conventional Scheibel column was constructed as shown in Figure 36. The agitator was a standard flat-bladed turbine to the dimensions shown in Figure 38; these agreed approximately with those scaled from the later Scheibel design (20).

Open-knitted wire mesh packings of F.S.L. quality, (A.I.S.I.) 304, stainless steel were obtained in 2 inch and 5 inch sections from Knitmesh Limited (97). Two types were selected initally, type 9033 identical with that used by other workers (9) and 9036. Details are given in Appendix 3. Sections were obtained just over 3 inches in diameter so as to fit snugly into the column. Care was taken to use packing without intermediate joins which are known to affect droplet hold-up (96).

10.4.2. Rotating Disc Contactor

The modified designs described in para. 2.2. were considered to offer no significant advantages and therefore a conventional R.D.C. was constructed. This comprised 4 stators and 2 rotors and is illustrated in Figure 37. The optimum dimensions were determined from the nomogram by Misek (46). Based on data considered earlier, preliminary design assumed a phase ratio of 0.5 to 2.0 to 1.

Rotor discs and stators were of brass so as to be preferentially wetted by the aqueous continuous phase. Care was taken to obtain a good fit between the stator rings and the column since the uncharacteristic performance of many laboratory columns could be attributable to phase flows between the periphery of the stator rings and the column wall (107). Stator spacing was by means of 4 equi-spaced pieces of 1/16 inch. brass wire. This was the minimum possible to give rigidity without introducing extraneous baffling effects which are another factor creating uncharacteristic performance in laboratory columns (107).

The rotor discs were installed with the securing bosses on their upper sides, that is effectively in dead zones, so as to cause minimum disturbance to the pattern of agitation. The discs were cylindrical since any narrowing of cross-section towards the edge would have produced greater axial flow (32).

To reduce the total hold-up of dispersed phase in the equipment and to facilitate a rapid approach to equilibrium hydrodynamic conditions in the contactor, the initial stages of droplet break-up were simulated by means of a perforated distributor plate. This was designed, as described in Appendix 6, to give an average drop size just greater than the anticipated maximum stable drop size for each set of hydrodynamic conditions to be investigated. The contactor section was located $4\frac{1}{2}$ inches above the distributor so that initial droplet formation was substantially unaffected by agitation.

10.4.3. Oldshue-Rushton Extractor

A section of a conventional Oldshue-Rushston extractor was constructed. This is illustrated in Figure 38.

Turbine dimensions were geometrically scaled from the data of Oldshue and Rushton (17). The agitator shaft was of stainless steel; turbines, stator rings and baffles were of brass. Turbines were soldered to the shaft to avoid any extraneous agitation effects due to securing bosses.

10.5 EXPERIMENTAL TECHNIQUES

10.5.1. Commissioning - Scheibel Section

With the Scheibel section a small amount of vibration of the packing was observed due to the rotating shaft. This could have been eliminated by a fixed collar but this would have affected droplet behaviour in the vicinity. The vibration was considered to have an insignificant effect on column hydrodynamics; for example, qualitative observations with the agitator alternately on then off showed that vibration had no effect in dislodging droplets from packing elements.

A minor amount of coalescence of dispersed phase occurred on the agitator shaft. Piper has attributed this effect to inefficient cleaning (96) but it was, in fact, unavoidable by reason of the shaft extending through the packed sections.

The cleaning procedure described in 10.5.4. was taken to be adequate since droplet characteristics leaving the packing were similar to those found by Piper after a more complex procedure (96).

Because of the complex droplet hydrodynamics observed, subesquent work; with the exception of wetting phenomena, was restricted

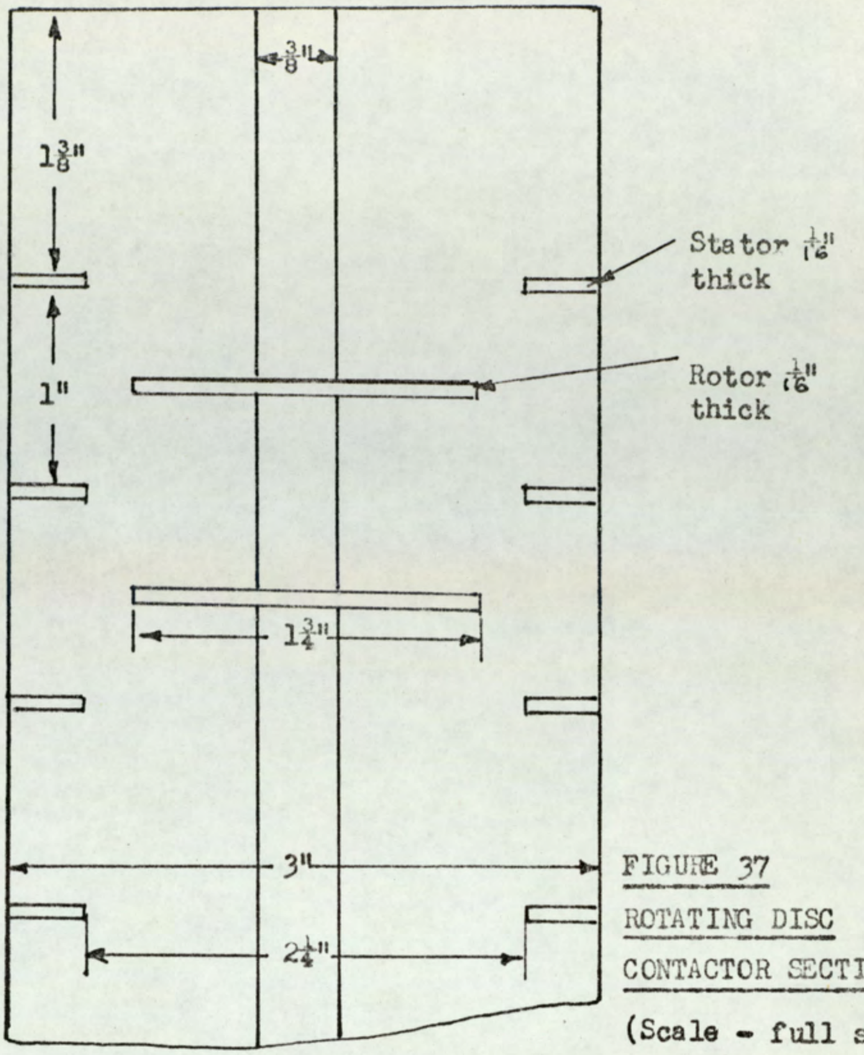


FIGURE 37

ROTATING DISC

CONTACTOR SECTION

(Scale - full size)

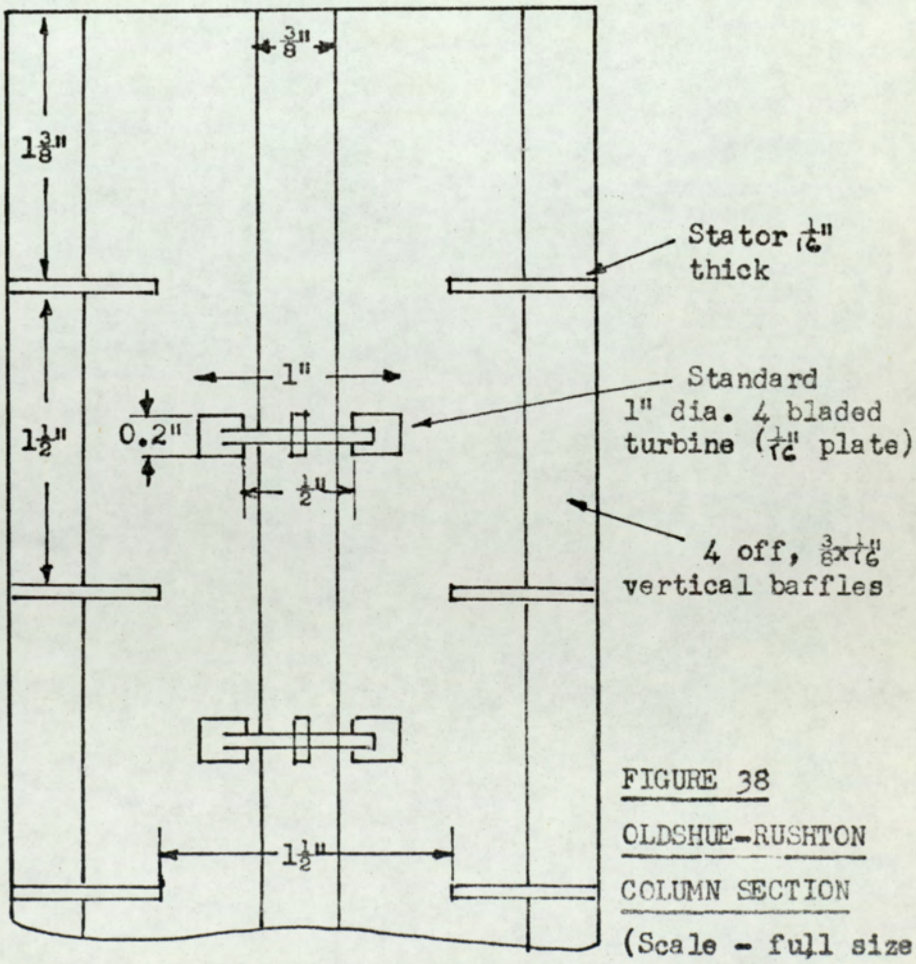


FIGURE 38

OLDSHUE-RUSHTON

COLUMN SECTION

(Scale - full size)

to packing 9036. A separate study of behaviour in packings was initiated (67,134).

10.5.2. Commissioning - R.D.C. Section

The R.D.C. Section was tested for flaws by noting the pumping effect of the rotating discs after the fashion employed commercially (95). The column was filled with water and a manometer fitted to the bottom drain nozzle. The results are given in Table 2. in the Appendix 5. Since the maximum head was only 1 m.m. of water the shaft and rotors were considered to be correctly balanced.

A sparging run was performed with toluene dispersed and a stationary aqueous phase. During this period, prior to cleaning of the column and internals, undesirable gross wetting and coalescence occurred on interior surfaces as has been illustrated (7) - Appendix 1. This was attributed to the effect of oil and dirt retained on the brass during fabrication giving typical dispersed phase wetting effects as described later in para. 11.4.

10.5.3. Commissioning - Oldshue-Rushton Section

In commissioning the Oldshue-Rushton design a degree of whip was observed in the rotor shaft particularly at intermediate speeds. The bottom end of the shaft was not restricted, as it was by packing in the Scheibel section. The installation of an additional steady bearing would have introduced extraneous baffling effects however and therefore some shaft whip had to be tolerated.

As with the other sections, it was found undesirable to admit dispersed phase to the column with the rotor stationary since this allowed larger droplets to travel in uncharacteristic paths and to build-up beneath stators and rotors. In all experimental procedures therefore the rotor speed was adjusted before dispersed phase was admitted to the columns.

10.5.4. Cleaning Procedure

Prior to insertion into the column all internals were cleaned with solvent to remove any grease or oil.

Cleaning of the column section was achieved using an aqueous solution of Decon 75. Solution was added via an inlet at the top and the agitator run for approximately 5 minutes. The equipment was then allowed to stand for a minimum of 12 hours. The column was then drained, refilled with clean water with the agitator running and re-drained several times. Care was taken to thoroughly flush out the dead zones present due to valved connections at the column base. Water was then pumped through the system to drain for approximately 10 minutes.

This cleaning procedure was repeated between every series of runs.

Whilst not as rigorous as the methods (55) used for single droplet coalescence studies the design precluded the use of chromic acid and cleaning involving dismantling was found to be impracticable. In the event, comparison of observed phenomena with those of other workers showed the procedure to be satisfactory (96).

10.5.5. Miscellaneous Techniques

The organic liquids employed in each run were stored in the reservoirs in contact with a layer of aqueous phase for a minimum of 24 hours prior to use to ensure mutual saturation. A similar procedure was followed for the aqueous phase. Care was necessary during operation to maintain the free water level in the organic phase reservoir below the outlet; any failure to do this resulted in the formation of emulsions in the pump and transfer line.

Rotameters were calibrated directly for each liquid used.

10.6. PHOTOGRAPHIC TECHNIQUES

10.6.1. Still Photography

A Miranda Sensorex camera with a 200 m.m. f 4.5 telephoto lens, through the lens metering and extension tubes as necessary were used for still photography. Lighting was provided by 2 or 3 independant 500 watt photoflood lamps positioned at the rear and left hand side of the column section. Clean tracing paper was placed over the apertures in the blockboard draught exclusion cabinet to give diffused lighting.

The camera was focused on the shaft in the centre of the column. Duplicate, or sometimes triplicate, photographs were taken at suitable intervals of each condition to be recorded. These showed excellent reproducibility, for example, Figures 39 and 40 show droplets in the R.D.C. under identical hydrodynamic conditions. This consistency was subsequently confirmed by observation of all the cine film taken.

Kodak Tri - X, 400 A.S.A. and later 32 A.S.A. film was used. Camera aperture was adjusted, dependant on column geometry and anticipated droplet hold-ups, to facilitate a shutter speed of 1/1000 th second. This generally required f numbers of 8 or 11.

10.6.2. Cine Photography

A Wollensak Fastax Type WF 14 T camera fitted with a Fastax - Raptar 50 mm. or 152 mm., f2 lens and extension tubes as required, were used for high speed cine photography. This had a film capacity of 400 feet of 16 mm. film and a framing range of 500 to 6000 pictures per second. The operation of this camera has been fully described by Lawson (55). Kodak Tri X reversal black and white film, A.S.A. 160 was used throughout.

Lighting was provided by independant 500 watt photoflood lamps. The arrangement of these was selected by trial and error to give good

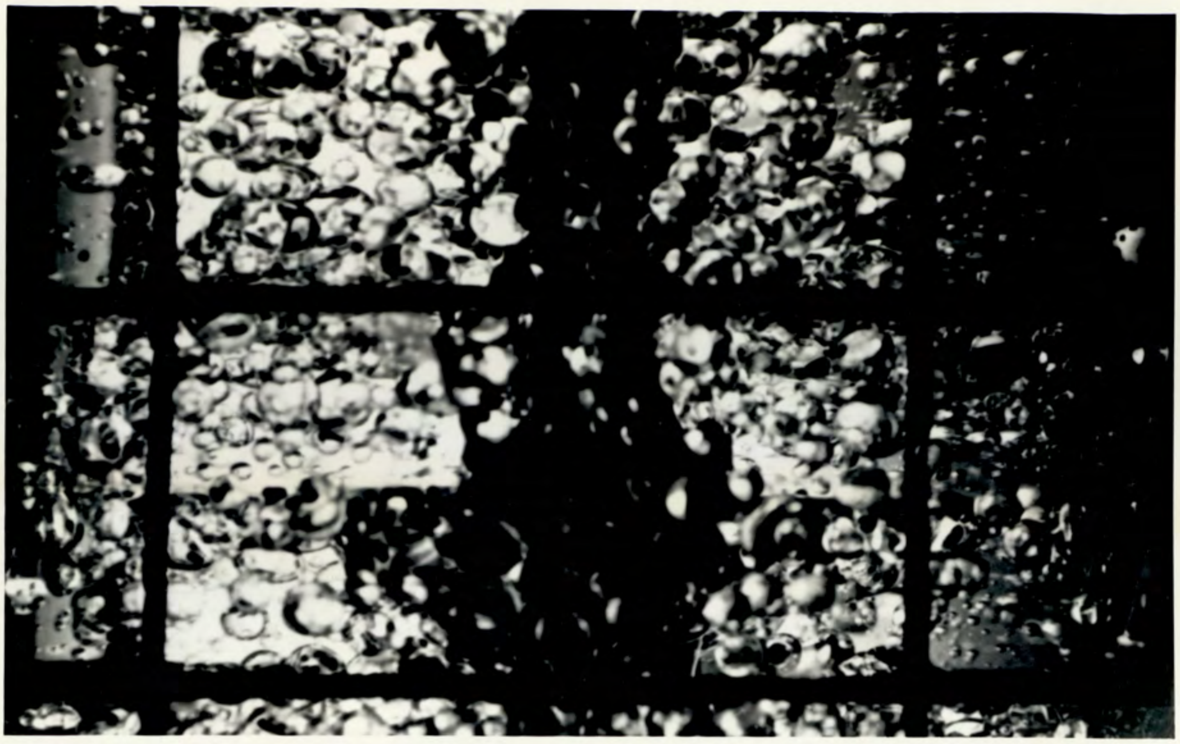


FIGURE 39. DROPLETS IN R.D.C. SECTION UNDER TYPICAL CONDITIONS
(System toluene - water, 850 r.p.m., $X \doteq 0.09$)

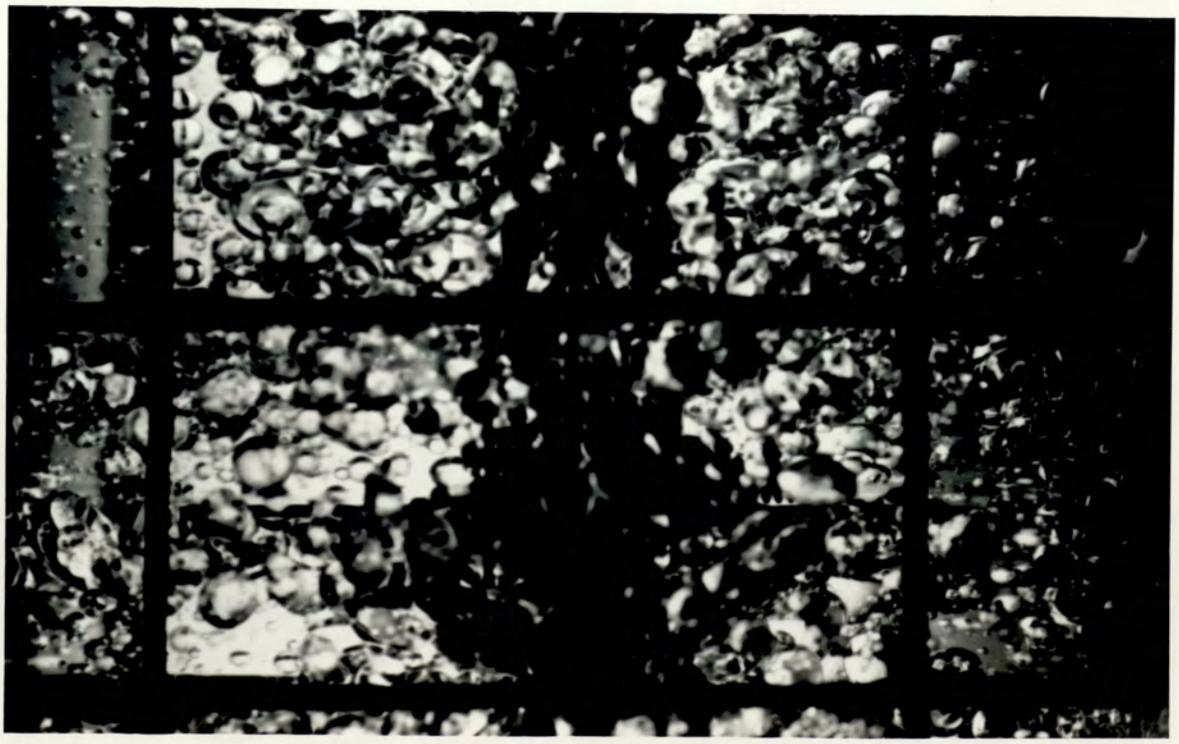


FIGURE 40. DROPLETS IN R.D.C. SECTION UNDER SIMILAR CONDITIONS TO FIGURE 39.

definition of droplets. A typical arrangement is shown in Figure 41. Immediate processing of the first exposed film in any series using a Hadland 16 mm. Negative Processor served as a check on exposure for subsequent film.

The actual speed of any sequence was determinable either by reference to speed/applied voltage calibration curves for the camera or from a spark trace on the edge of the film. The motion analysis projector used had facility for single-shotting and for holding a frame for any desired period so that it was hoped to achieve a frame by frame analysis of all sequences of interest.

In the event, with certain exceptions, it was found that only a qualitative study was possible and mean camera speeds of 4000 or 5000 pictures per second were employed. The magnification in time, that is, the ratio of picture frequency in the camera to picture frequency in the projector, was therefore approximately 250 when projecting normally at 16 p.p.s. or rarely 2000 when projecting at 2 p.p.s.

A Beaulieu camera was used for cine photography at 64 p.p.s. The camera, an R 16. with a P3 type, f 0.75 lense, was located approximately 1 metre from the column. 3 Photoflood lamps were used located generally as in Figure 41, the two side lights being adjusted until glare from the column edge disappeared.

10.6.3. Measurement of Distortion

Preliminary observations and reference to the photographic work of Honekamp and Burkhart (9) suggested that the errors due to magnification and distortion of droplets caused by viewing through the 3 inch diameter glass column would be negligible with an aqueous continuous phase. This was confirmed by photographing a section of 20 x 20 mesh 32 S.W.G. stainless steel gauze placed across a diameter of the column. Figure 42, shows that distortion is insignificant excepting for the outer $\frac{1}{8}$ inch of the column which was in most cases unphotographable.

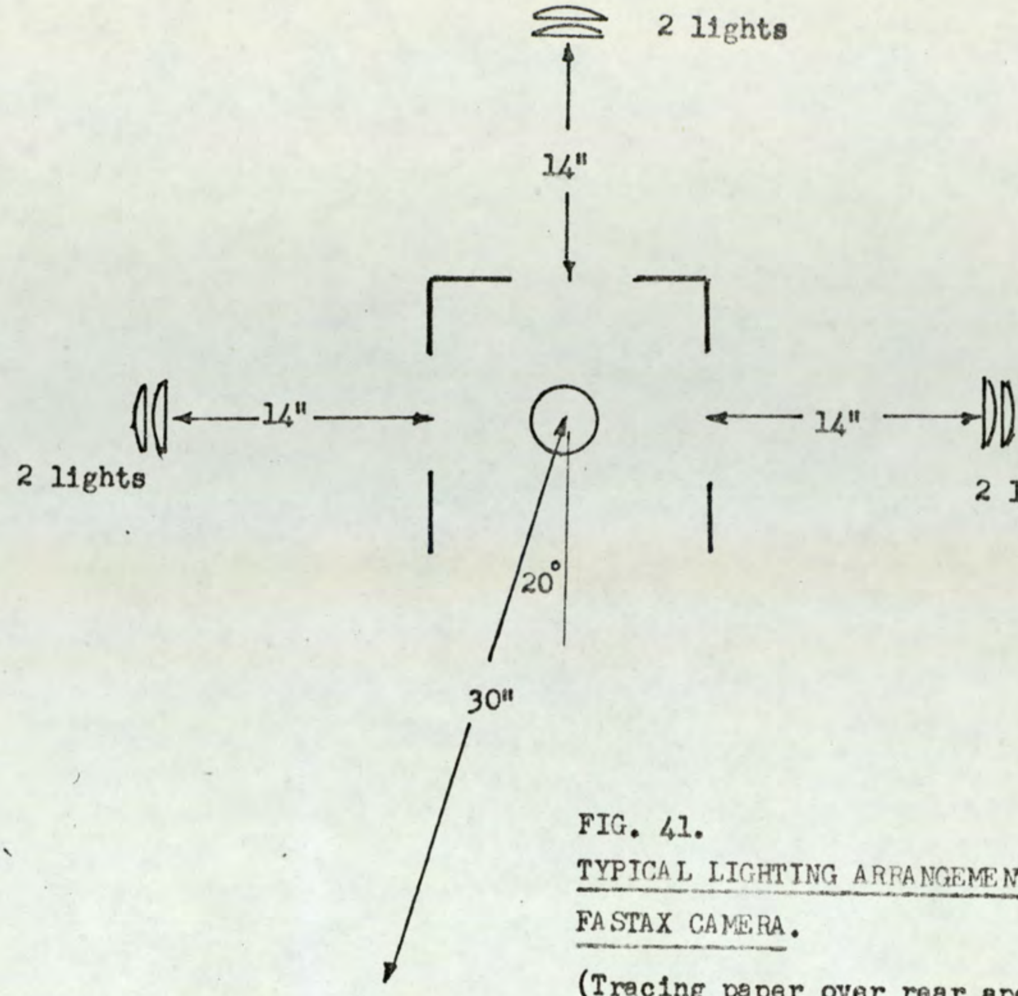


FIG. 41.

TYPICAL LIGHTING ARRANGEMENT FOR FASTAX CAMERA.

(Tracing paper over rear aperture only)

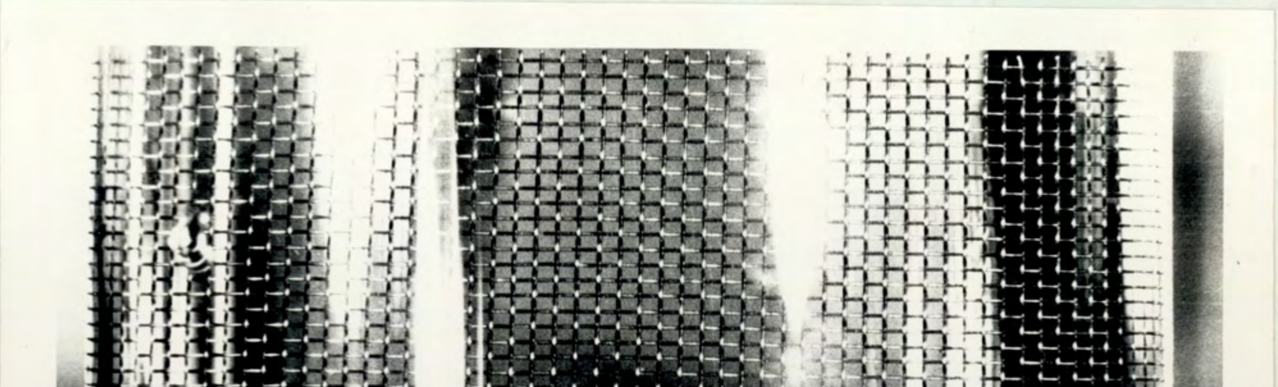


FIGURE 42. 20 x 20 mesh. GAUZE IN AQUEOUS CONTINUOUS PHASE

(Magnification 1.9)

11. EXPERIMENTAL PROCEDURES & RESULTS

11.1. HOLD-UP

Attempts were made initially to determine average values of dispersed phase hold-up under variable hydrodynamic conditions. The method used was based on that of other workers (102,108) and comprised operating the column under the desired conditions but with the upper settling gone incompletely filled, rapidly closing the inlet valves and then measuring the increase in interface height in the 4 inch column section.

This method was found to give poor reproducibility and accuracy. In addition its sensitivity was affected by the inclusion of droplets present in the upper and lower parts of the column which were not subject to the hydrodynamic regime in the agitated section.

A sampling technique was therefore developed to determine point hold-up values. A section of 3/16 inch plastic tubing was inserted through the top of the column and located by a small brass collar. With the R.D.C. section, the determination of radial hold-up profiles was found to be impracticable owing to the relatively small column diameter. This confirmed the experience of Olney who obtained erratic results in a 6 inch column (87). Samples were therefore taken from a point midway between the rotor and upper stator ring and on a vertical centreline of 1 inch from the shaft centre. This corresponded to a position between $R/2 = 0.875$ inch and $S/2 = 1.125$ inch at which hold-up has been found to be characteristic in larger columns (87). Preliminary trials established that reproducible results were dependant upon the withdrawal of samples of not less than 20 to 25 ccs, corresponding to a maximum of 20% of compartment capacity.

The rotor speed was set to the desired value and the dispersed phase flow-rate adjusted. When equilibrium conditions were attained in the column section, usually after 3 minutes, a sample was rapidly drawn up the sampling tube into a glass pipette. A pipette filler was used for this operation.

The volume of the sample was measured in a 25 cc. measuring cylinder; after settling the volume of dispersed phase was measured in a 5 cc. measuring cylinder. The sampling line was blown-out after each sample and the pipette flushed out. Mean values were taken of at least three determinations for each hydrodynamic condition. Poorest reproducibility was obtained at low values of hold-up, < 0.1, and at the onset of flooding.

Results for the R.D.C. section are plotted in Figures 43,44 and 45.

A similar procedure was followed with the Oldshue-Rushton section. Samples were taken from a point midway between the turbine and upper stator and on a vertical centreline of 1 inch from the shaft centre. This was justifiable because of the small column diameter and the rapid circulatory mixing in each compartment.

Results for the Oldshue-Rushton section are plotted in Figures 46,47 and 48. Hold-up determinations at low agitator speeds, of the order of 200 r.p.m. gave poor reproducibility and are not reported.

Point hold-ups in the agitated zone of the Scheibel section could not be determined reproducibly by the method described. Static hold-up in the packing was found to be negligible. A value of approximately 2% has since been found for similar low voidage packings in other work (134).

11.2 FLOODING PHENOMENA

For the determination of flooding rates, the rotor speed and continuous phase flow rate were first adjusted to the desired values. Steady conditions were deemed to be established when there was complete absence of movement of the interface at the top of the column, that is outlet rate = inlet rate. The flow of dispersed phase was then commenced and increased incrementally up to the flood-point. Complete rejection of the dispersed phase as a dense layer of droplets was used

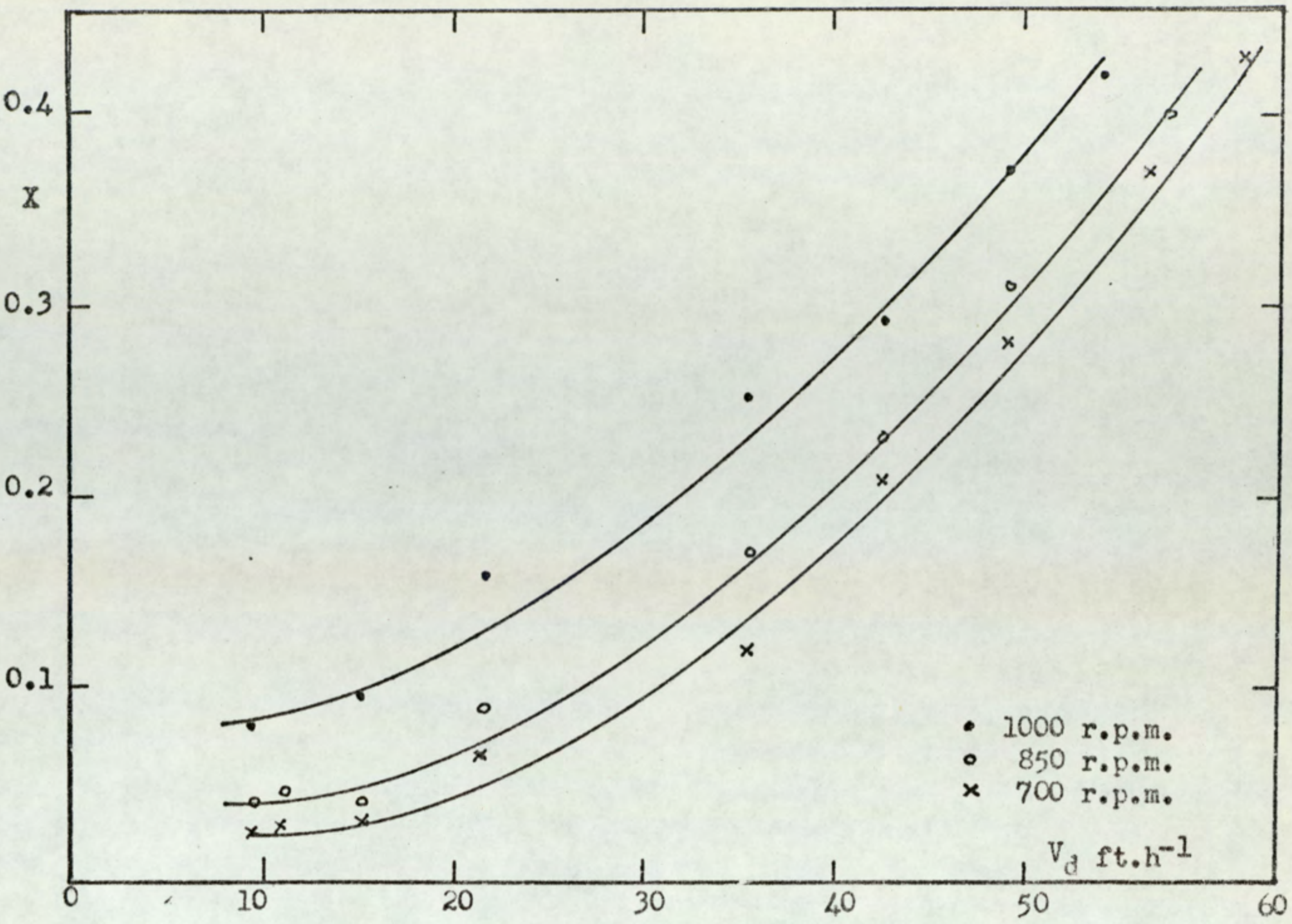


FIGURE 43. R.D.C.; EFFECT OF ROTOR SPEED ON DISPERSED PHASE HOLD-UP
(System : toluene - water)

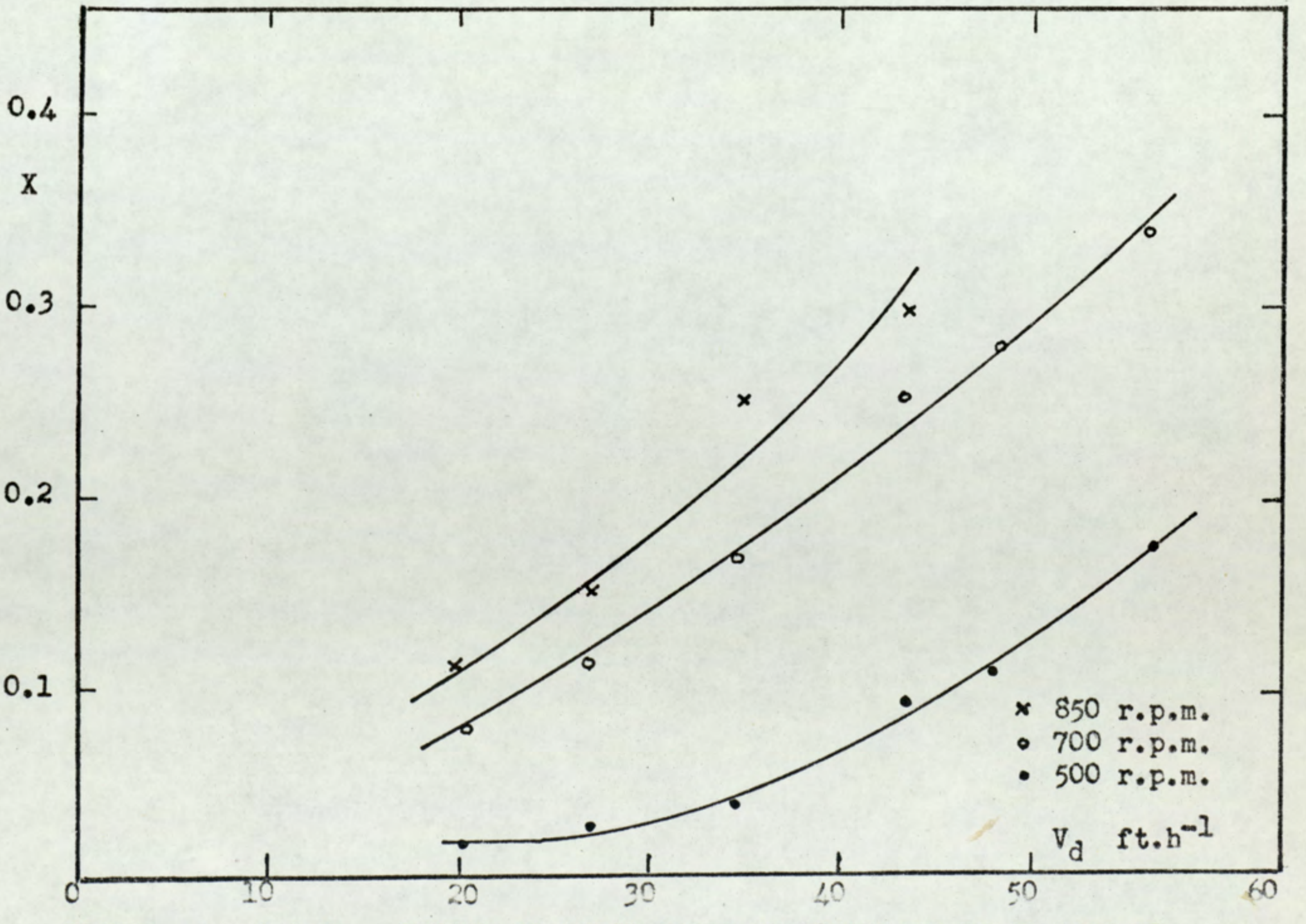


FIGURE 44. R.D.C., EFFECT OF ROTOR SPEED ON DISPERSED PHASE HOLD-UP
(System : M.I.B.K. - water)

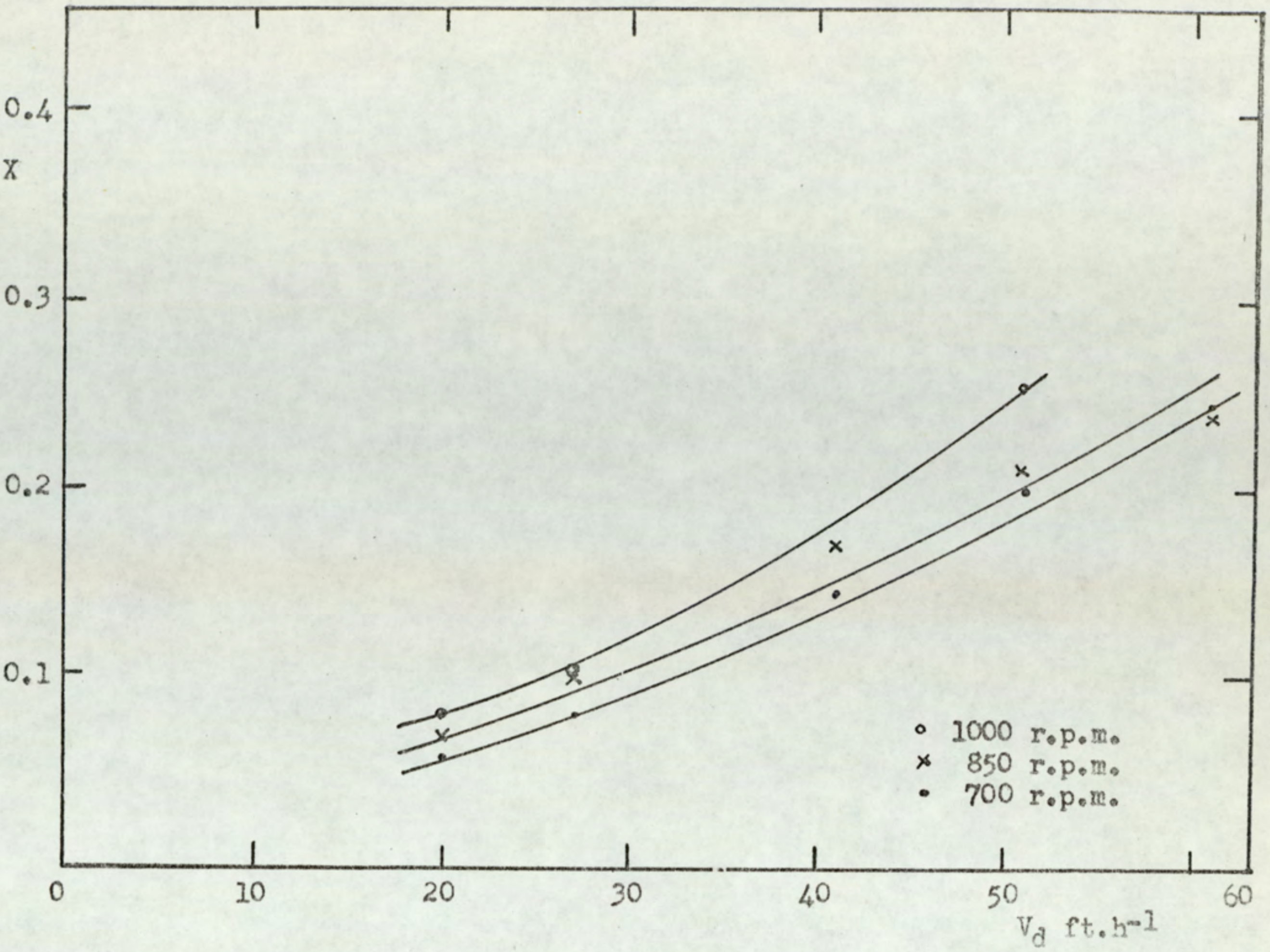


FIGURE 45. R.D.C. ; EFFECT OF ROTOR SPEED ON DISPERSED PHASE HOLD-UP
(System : trimethylpentane - water)

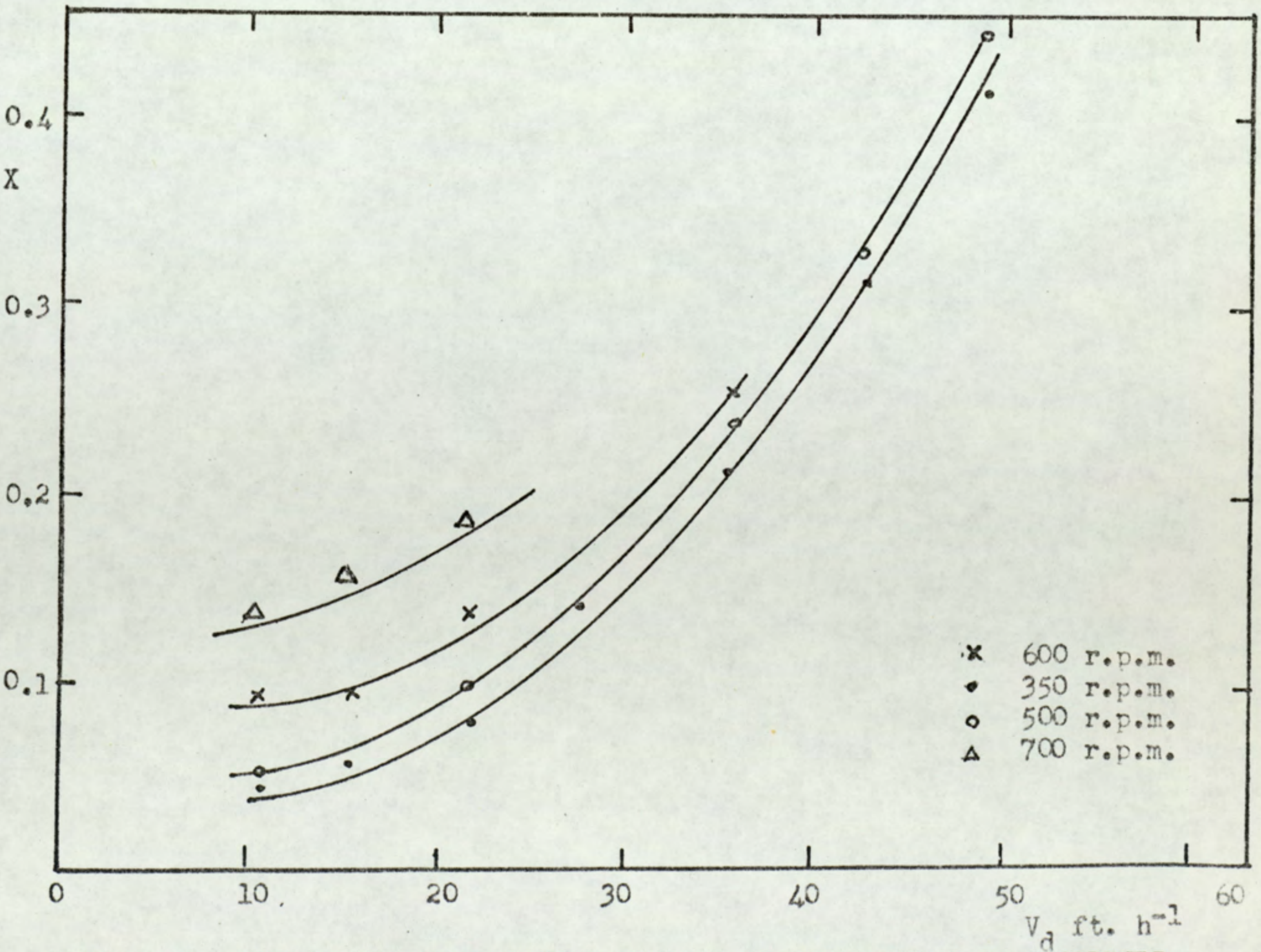


FIGURE 46. OLDSHUE-RUSHTON SECTION; EFFECT OF ROTOR SPEED ON DISPERSED
PHASE HOLD-UP (System : toluene - water)

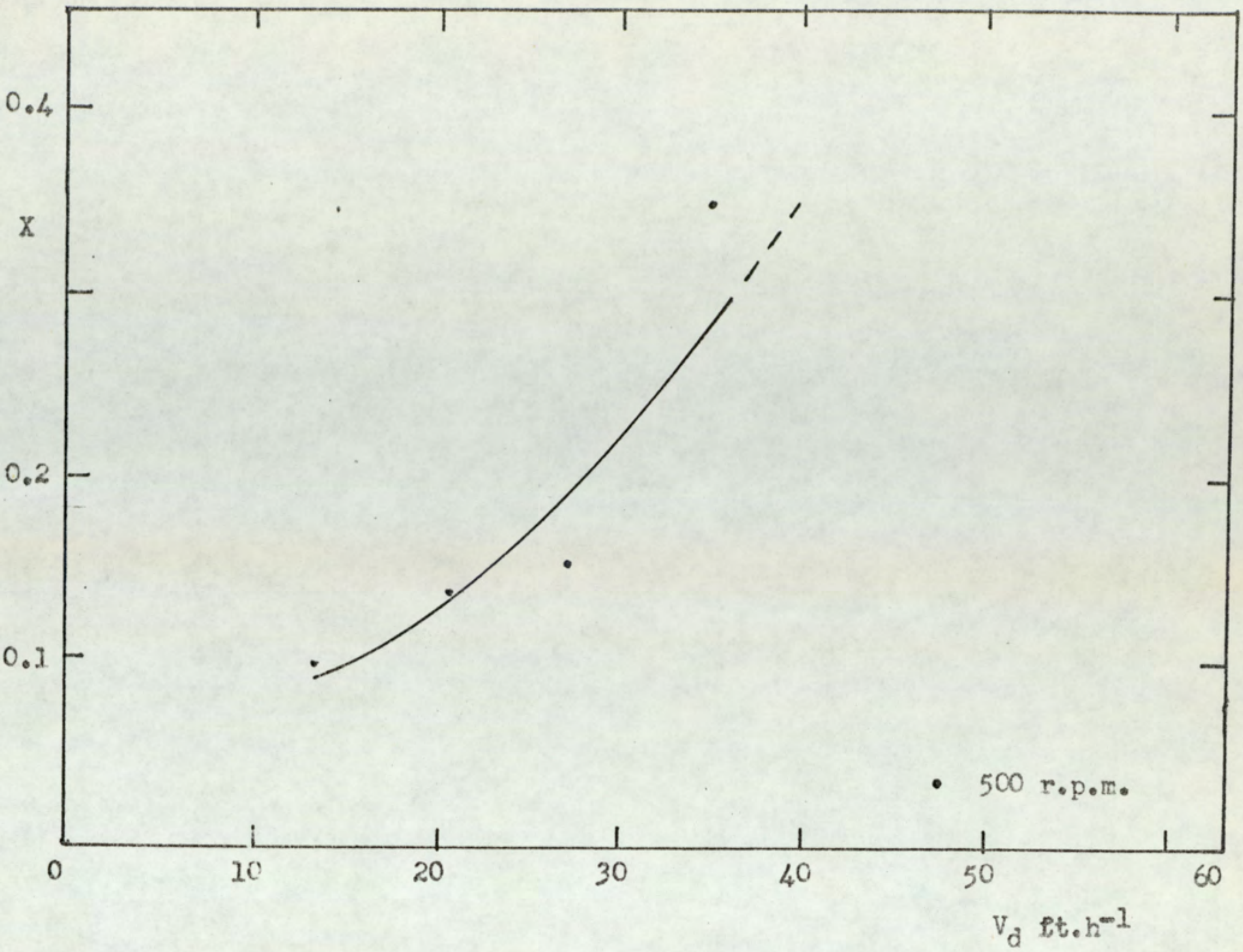


FIGURE 47. OLDSHUE-RUSHTON SECTION; EFFECT OF ROTOR SPEED ON DISPERSED PHASE HOLD-UP (System : M.I.B.K. - water)

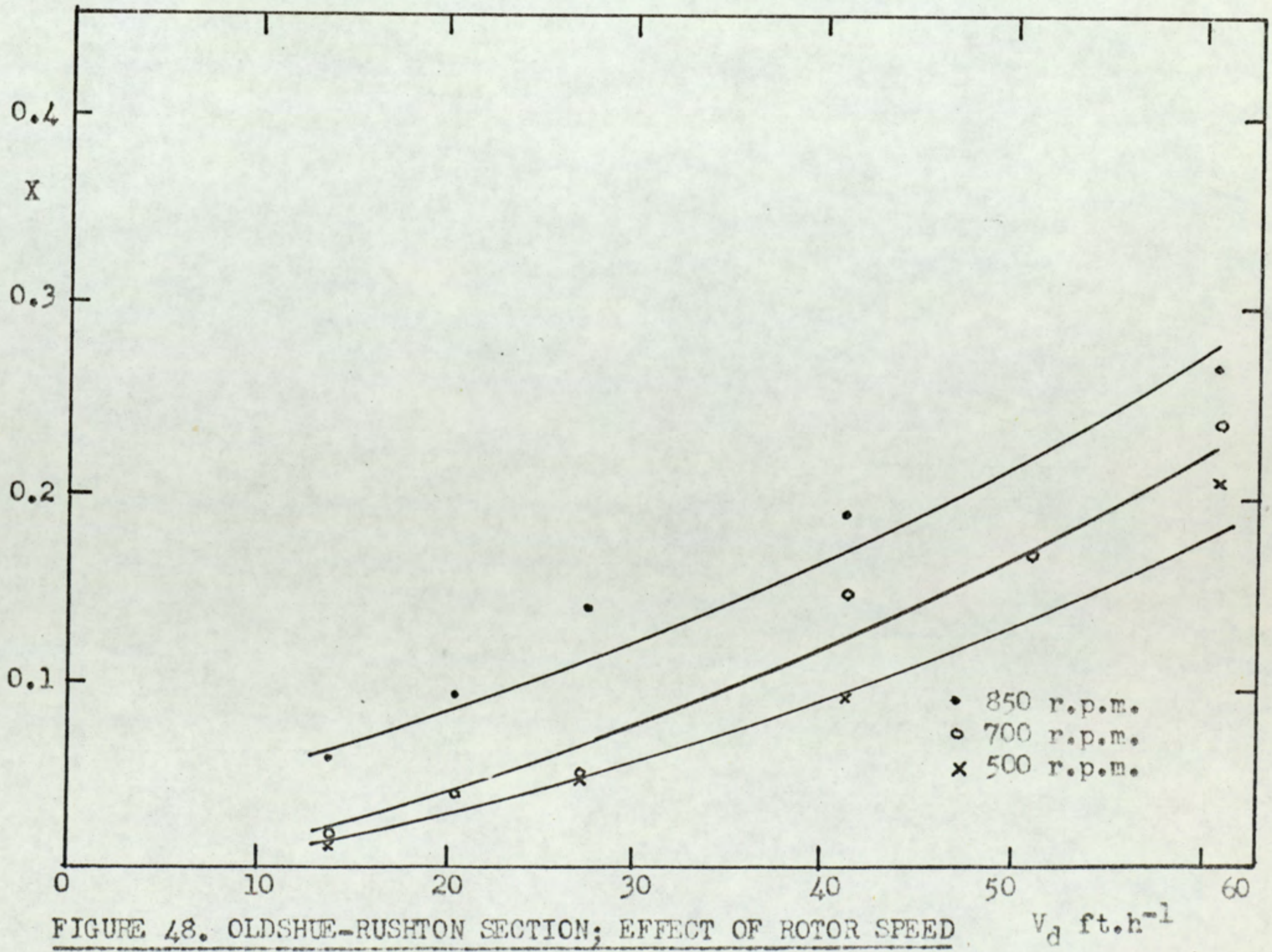


FIGURE 48. OLDSHUE-RUSHTON SECTION; EFFECT OF ROTOR SPEED ON DISPERSED PHASE HOLD-UP (System : trimethylpentane-water)

as a criteria of flooding. As has been reported by others (83,91) intense backmixing was observed in all contactors immediately prior to the onset of flooding.

Results for the R.D.C. section are plotted in Figures 49,50 and 51. These values are the mean of triplicate or, rarely, duplicate readings. The onset of flooding with the system toluene - water at 1000 r.p.m. is recorded in the cine film - Appendix 7.

In the Oldshue-Rushton section the onset of flooding commenced with flocculation of droplets in the plane of the stator ring gradually spreading downwards in a compartment. At the lower agitator speeds flooding was more easily determinable than in the R.D.C. but was more difficult at higher speeds since the circulatory mixing pattern and intense back-mixing gave a premature appearance of flooding. At very low agitator speeds, of the order of 200 r.p.m. with all systems, turbulence was insufficient to prevent droplets being restricted between the stators and vertical baffles. This phenomena, which has also been commented on by Bibaud and Treybal (93), created conditions conducive to premature flooding. Results for the Oldshue-Rushton section are plotted in Figures 52,53 and 54. A few results were also obtained at zero and higher rotor speeds but these are of no practical value and are not reported. At zero rotor speed large droplets were formed due to coalescence on the stator rings and subsequent drip-point breakaway. It was observed that at low rotor speeds, < 100 r.p.m., the column had a slightly greater capacity than at zero rotor speed due to disturbance of droplets flocculating beneath the stators. It follows from the work of Garner et.al. (135) in a perforated plate column that this would also give improved efficiencies. At high rotor speeds difficulty arose in obtaining primary break of the phases and this made them impracticable. (Primary breaks were obtained by the addition of a further quantity of the dispersed phase).

The assessment of flooding points presented greatest difficulty in the Scheibel section; similar difficulties arose in a parallel study (67) . Nevertheless the same criteria was used as above. Piper (96) arbitrarily selected a 1 cm. layer of dispersed phase beneath the packing to characterize flooding in the artificial case of no agitation; recent work has shown that some build up beneath certain packings is normal and does not constitute flooding however (134). The onset of flooding appeared within the section of packing immediately above the agitator; a further small increase in dispersed phase flowrate at constant continuous phase flowrate and impeller speed led to the rejection of large droplets of dispersed phase beneath the packing. This is illustrated in Figure 58. showing the persistence of droplets rather than streamers. The Results are plotted in Figures 55,56, and 57.

11.3 DROPLET PHENOMENA AND DROP SIZE DISTRIBUTIONS

11.3.1. Experimental Procedures

The observation and photography of droplet phenomena and drop size distributions were performed with a stationary continuous phase. As mentioned earlier, distributions have been found not to be directly affected by continuous phase flowrate (9,108). (This was confirmed by a trial with the system trimethylpentane - water in the R.D.C. section the results of which are given in Table 14 in Appendix 9).

After any change in operating parometers it was found necessary to allow about 3 minutes for hydrodynamic equilibrium to be attained. A considerably longer time, of the order of 5 minutes, was necessary on start-up to clear air from the dispersed phase transfer system. If air became trapped beneath a rotor in the R.D.C. it was found to remain there in the form of an inverted cone around the shaft; succesive stopping and starting of the rotor was necessary to remove this. Use was made of the time to reach true conditions to lubricate the stuffing

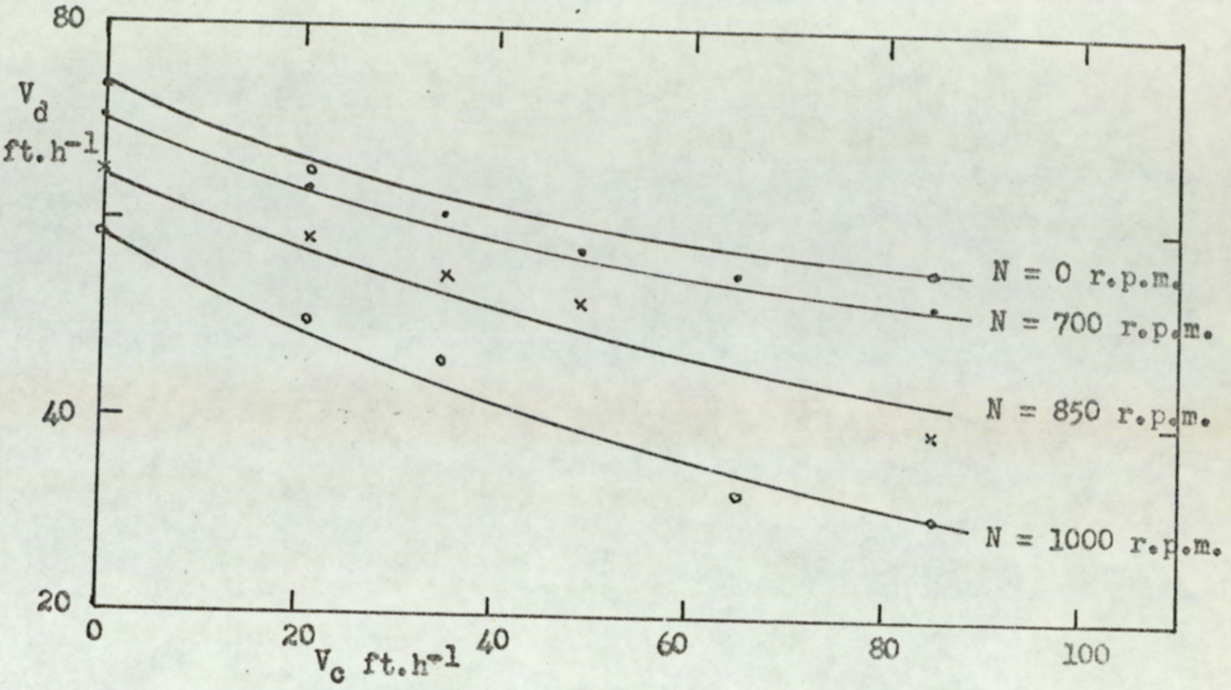


FIGURE 49. R.D.C. - FLOODING CURVES
(System : Toluene - water)

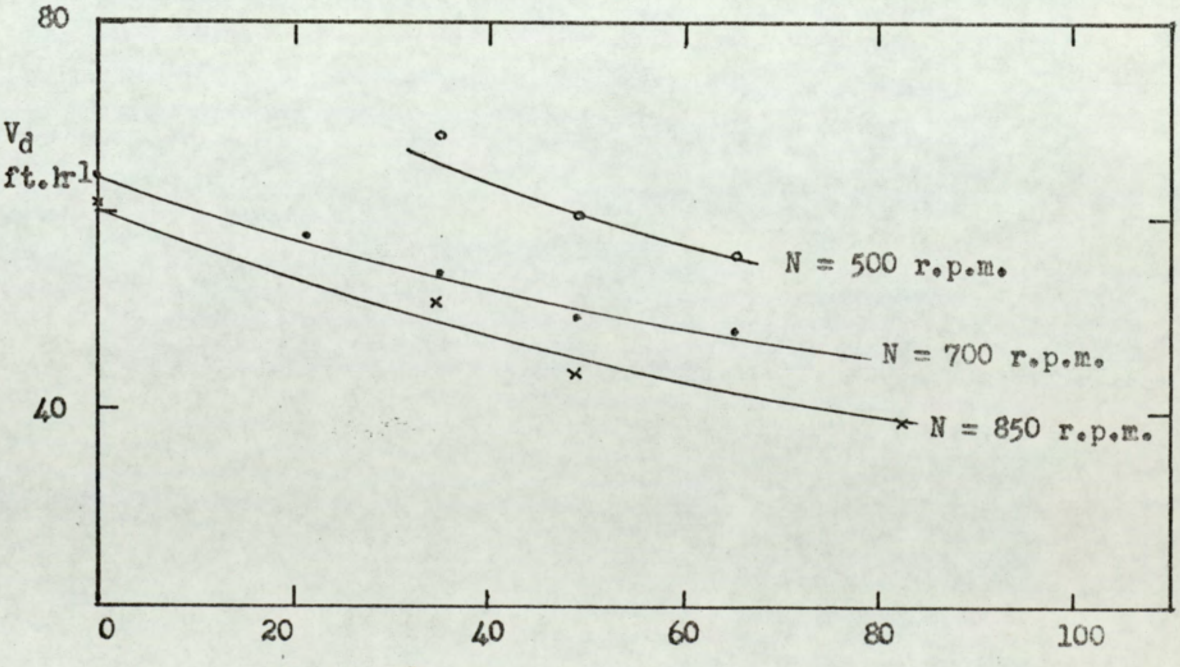


FIGURE 50. R.D.C. - FLOODING CURVES
(System : M.I.B.K. - water)

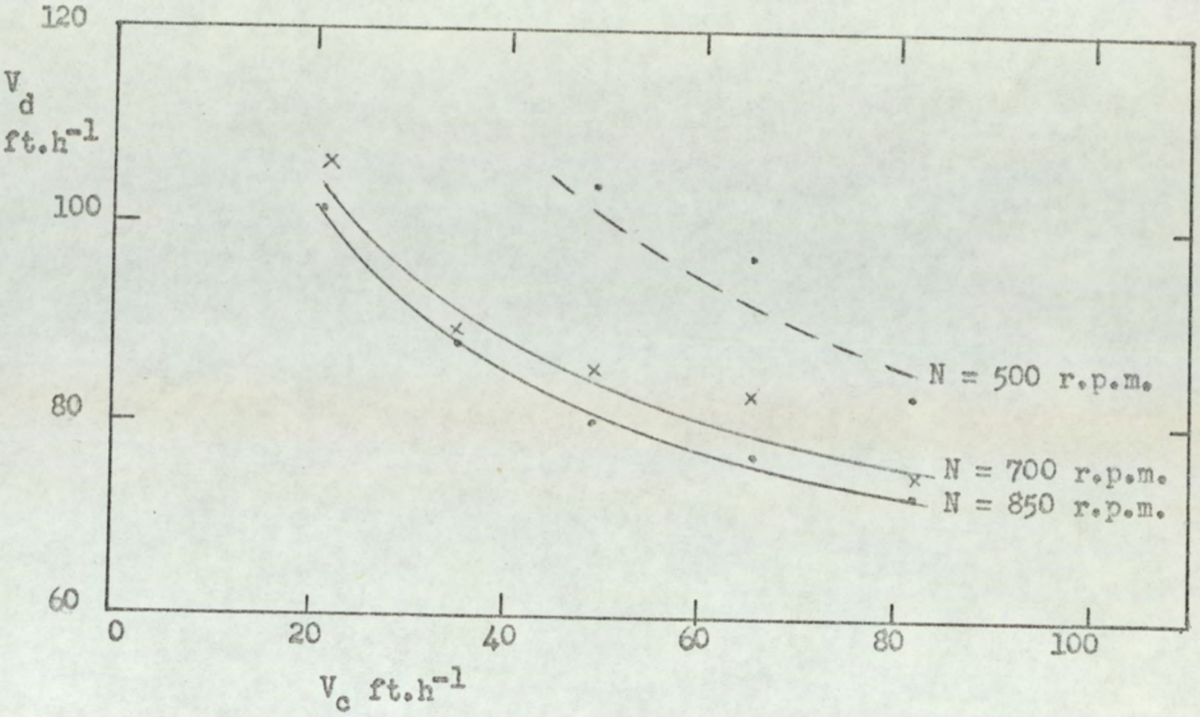


FIGURE 51. R.D.C. - FLOODING CURVES

(System : trimethylpentane - water)

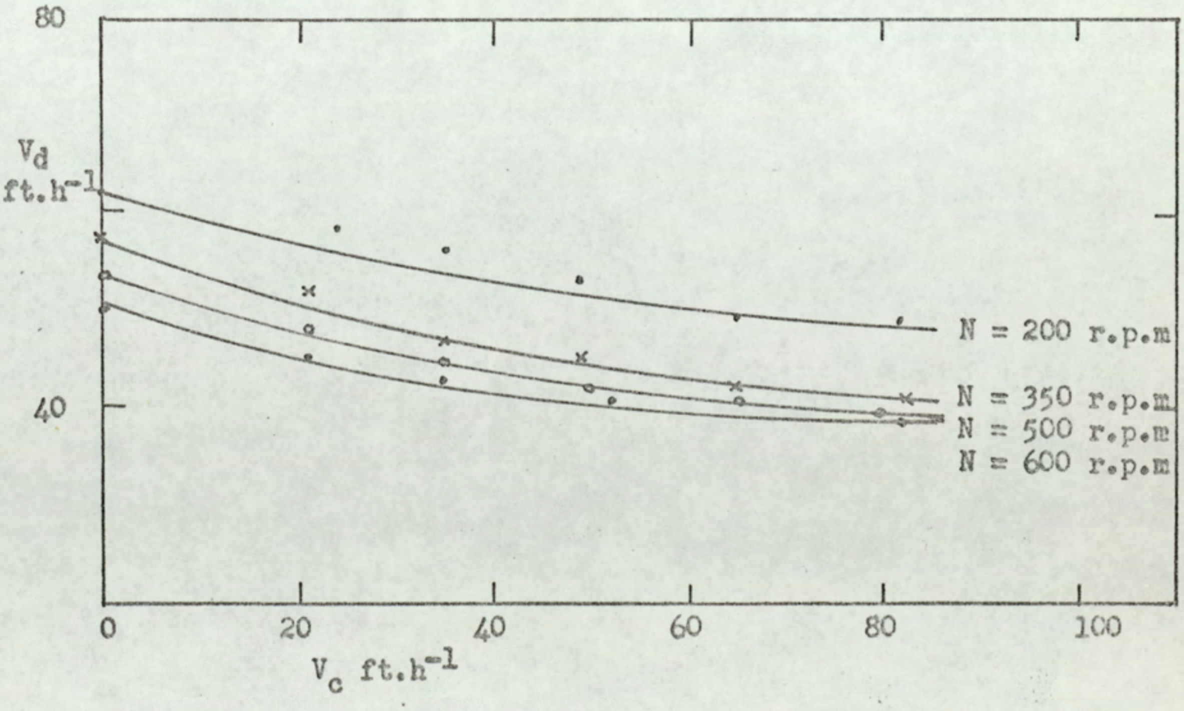


FIGURE 52. OLDSHUE-RUSHTON SECTION - FLOODING CURVES

(System : Toluene -water)

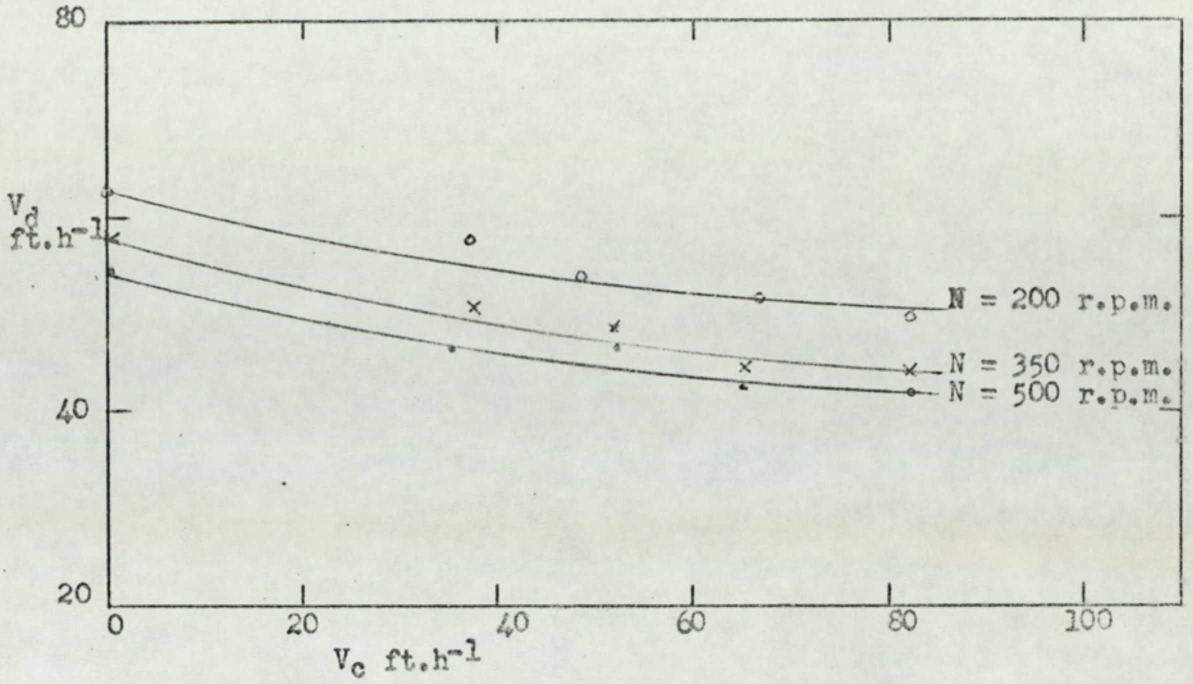


FIGURE 53. OLDSHUE-RUSHTON SECTION - FLOODING CURVES
(System : M.I.B.K. - water)

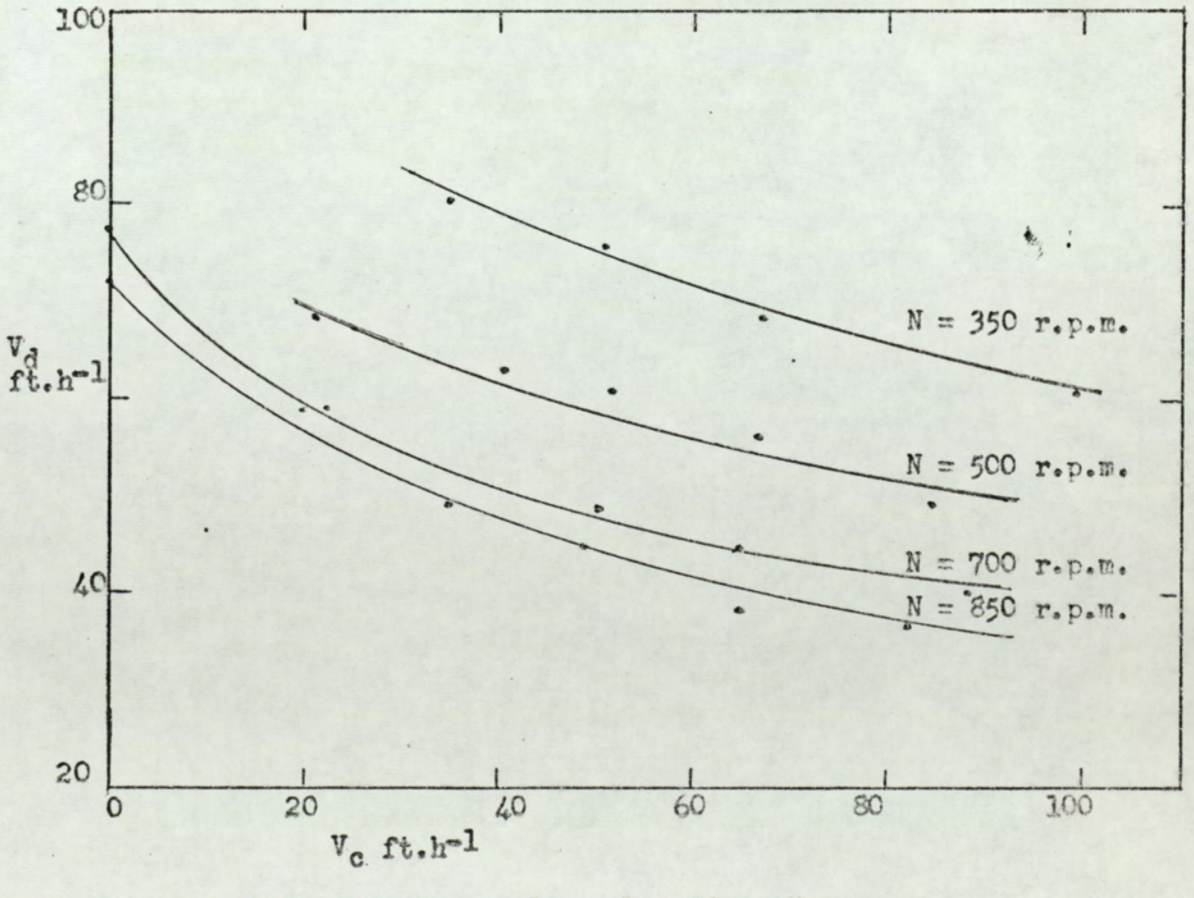


FIGURE 54. OLDSHUE-RUSHTON SECTION - FLOODING CURVES
(System : trimethylpentane-water)

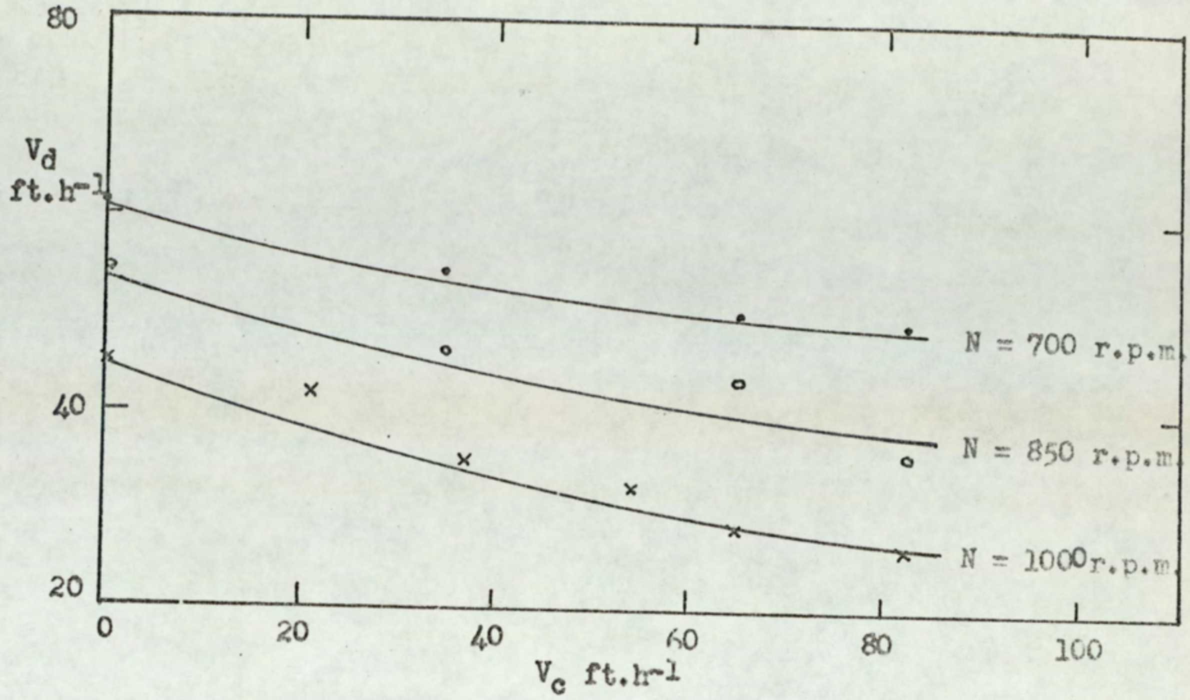


FIGURE 55. SCHEIBEL SECTION - FLOODING CURVES
(System : toluene - water)

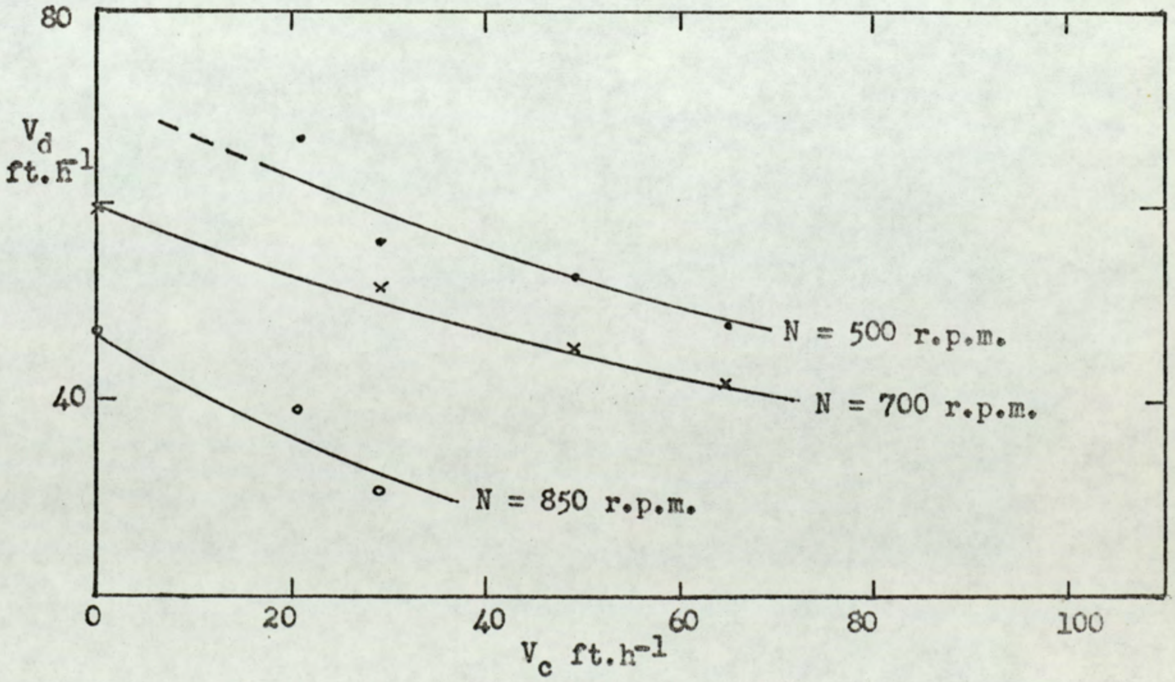


FIGURE 56. SCHEIBEL SECTION - FLOODING CURVES
(System : M.I.B.K. - water)

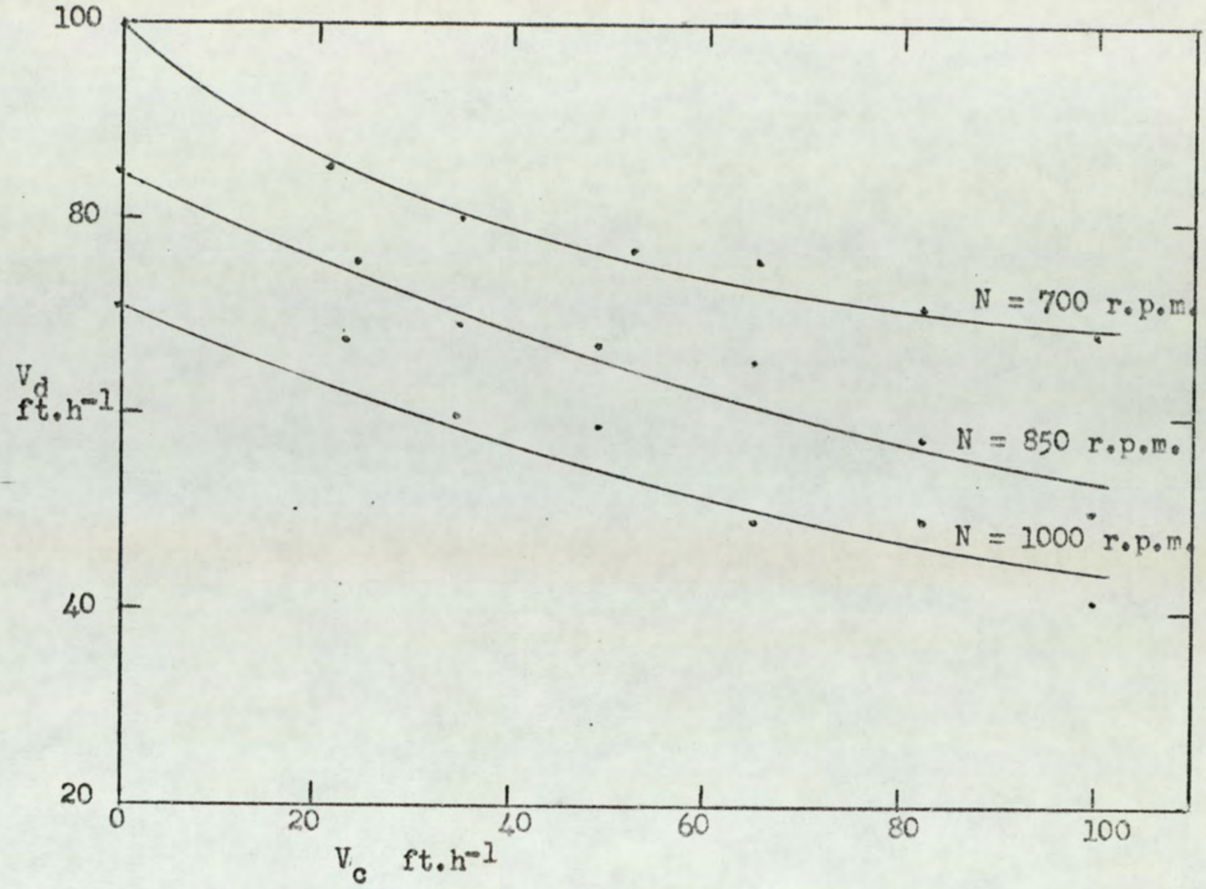


FIGURE 57. SCHEIBEL SECTION - FLOODING CURVES
(System : trimethylpentane - water)

FIGURE 58. SCHEIBEL SECTION - ONSET OF FLOODING
(System: Toluene - water, N = 700 r.p.m.
$V_c = 0$, $V_D = 61$ ft.sec^{-1})

box with water from a syringe thus ensuring even agitator rotation.

Adequate cleaning of the column section was found to be essential between each series of runs. Photographic techniques have been described in para 10.6.

Drop size measurements were taken either from projection or prints. A magnification of up to 12 was obtainable by projection and a few drop counts were made by marking tracing paper placed over a viewer. Counts from prints reproduced professionally were considered to be more reliable since small drops, though in focus, tended not to show up well on projection. A magnification of 5 to 6 times was found preferable for prints. However, because of the large number of prints to be analyzed it was generally necessary to use only a 2 to 2.5 times magnification with a corresponding reduction in accuracy.

Drops were counted by the traditional method using a ruler with m.m. graduations blackened on either side of the characteristic measurement. A dot was marked in coloured ink on the photograph, or on tracing paper placed over the photograph, to indicate drop size and all drops in each size range subsequently counted. An S.P.R.I. Particle Size Analyser Type II subsequently became available (136) and later a Carl Zeiss Particle Size Analyzer TG Z. 3 (137). Only those droplets with a clear outline were counted. Ellipsoids were recorded as spheres of equivalent diameter excepting those with a ratio of major to minor axis greater than 1.4 which were discarded. Only a few droplets were in this category.

Hydraulic mean drop diameter was evaluated from,

$$d_{12} = \frac{\sum nd^2}{\sum nd}$$

and Sauter mean drop diameter from,

$$d_{32} = \frac{\sum nd^3}{\sum nd^2}$$

Typical drop counts and calculations are given in Appendix 9.

11.3.2. Scheibel Column

The function of the packing in the Scheibel section was generally dependant on drop size distribution i.e. agitator speed (96).

At low rotor speeds, when the mean drop size was considerably larger than the interstices size of the packing, the packing acted mainly as a coalescing stage producing large globules which left the packing by drip-point formation. The mechanism of droplet break-up is illustrated in Figure 59, which shows the formation of a satellite drop by shearing.

At increased throughputs of the dispersed phase, jetting occurred from preferential areas on the packing surface. These jets were not effectively broken up leading to numerous large globules. A typical mechanism of jet break-up producing large and small satellite drops is illustrated in Figure 89. Mechanisms of jet formation have since been evaluated by Thomas (134).

Formation of globules was not eliminated by increased rotor speeds. Their presence, as illustrated in the paper - Appendix 1.3 (7), precluded the estimation of meaningfull average drop sizes. Maximum drop sizes are summarised in Table 3 in Appendix 5.

11. 3.3. Rotating Disc Contactor

With all 3 systems, a critical minimum rotor speed was found to exist below which a layer of dispersed phase droplets built up beneath the rotor disc. For the system toluene-water this is illustrated in the paper comprising Appendix 1.3 (7) and in the cine-film - Appendix 7.1. Figures 60 to 62 show the effect with M.I.B.K. - water at various speeds below the critical minimum. The precise value of the critical minimum speed was indeterminable; the values given in Table 4 are the mean of of numerous observations throughout this work.

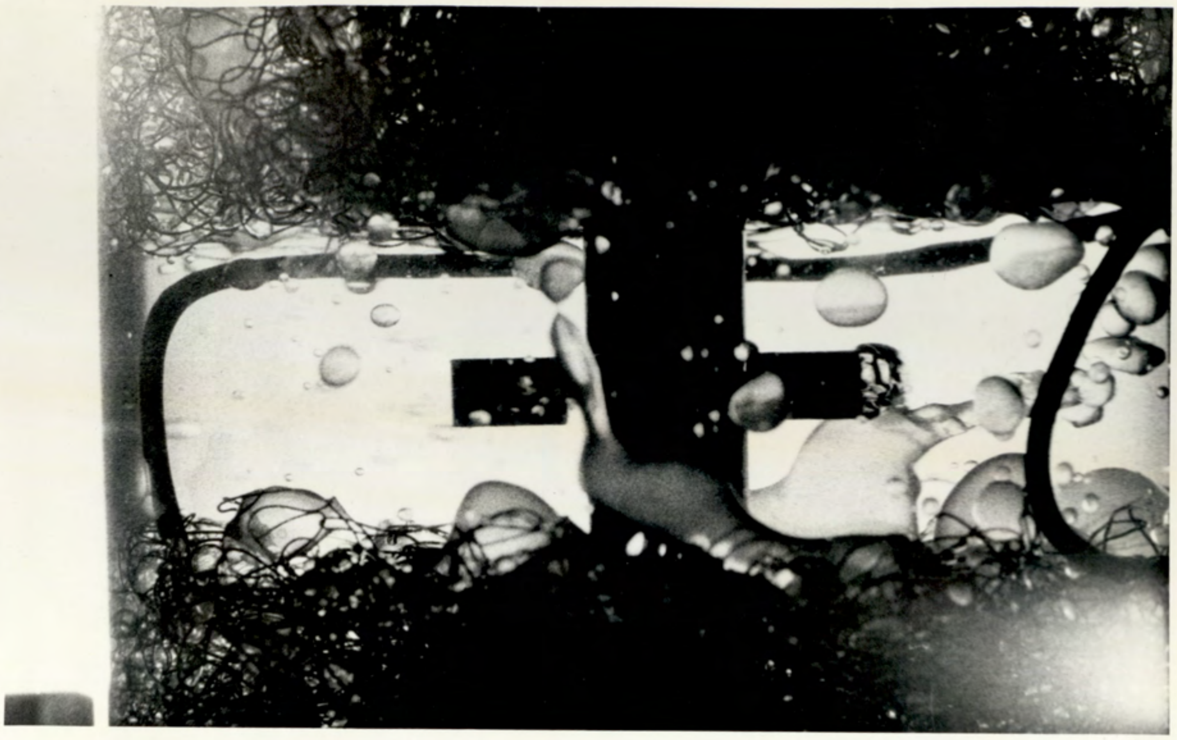


FIGURE 59. DROPLET FORMATION FROM OUTLET OF PACKING IN SCHEIBEL COLUMN.

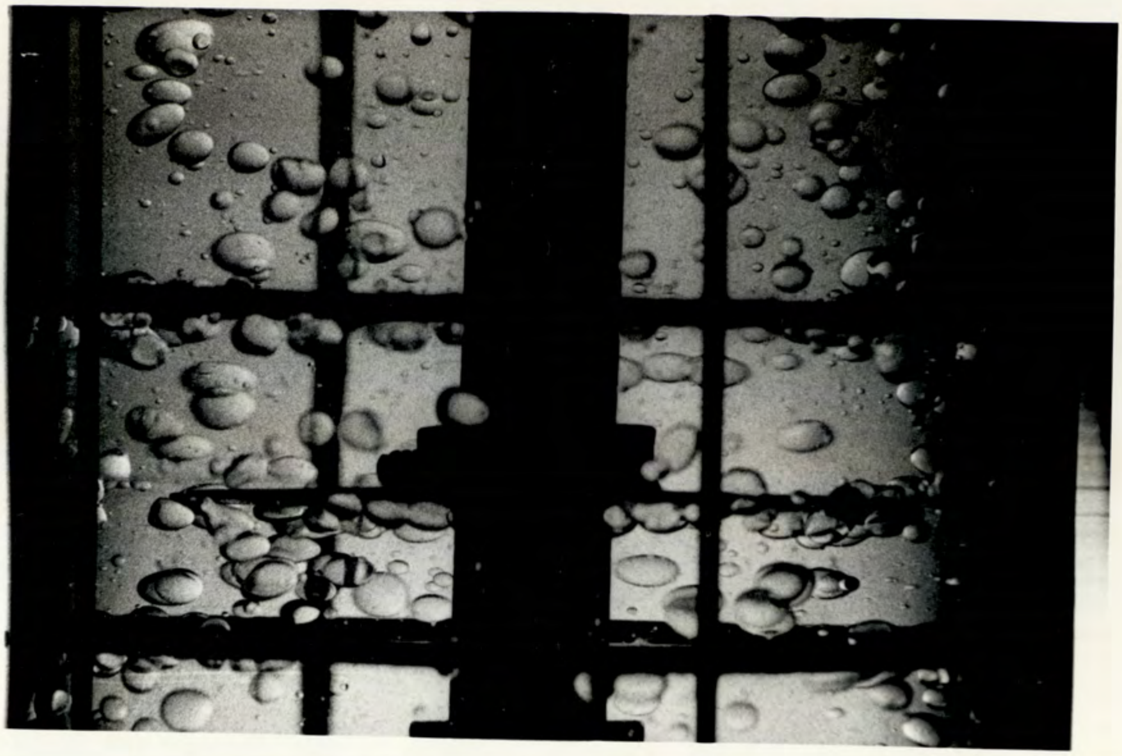


FIGURE 60, R.D.C. SECTION WITH ROTOR SPEED BELOW THE CRITICAL MINIMUM

(System: M.I.B.K. - water, N = 400 r.p.m.)

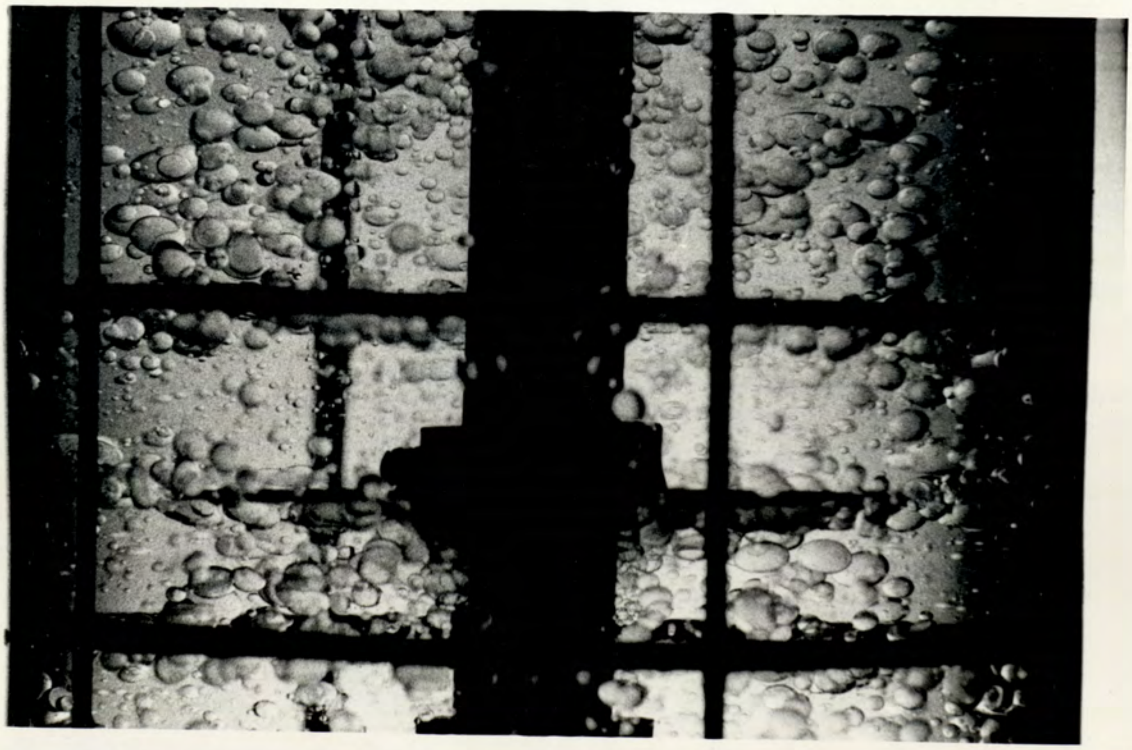


FIGURE 61, AS ABOVE with N = 500 r.p.m.

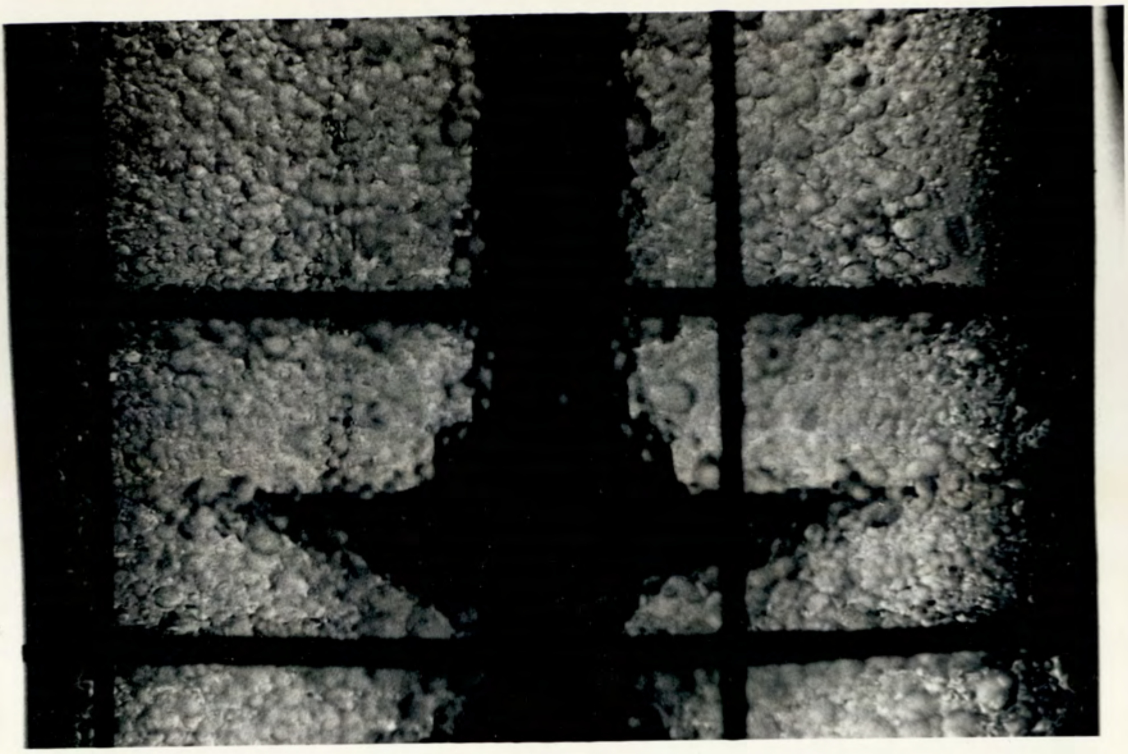


FIGURE 62, AS FIGURES 60 and 61 with N = 600 r.p.m.

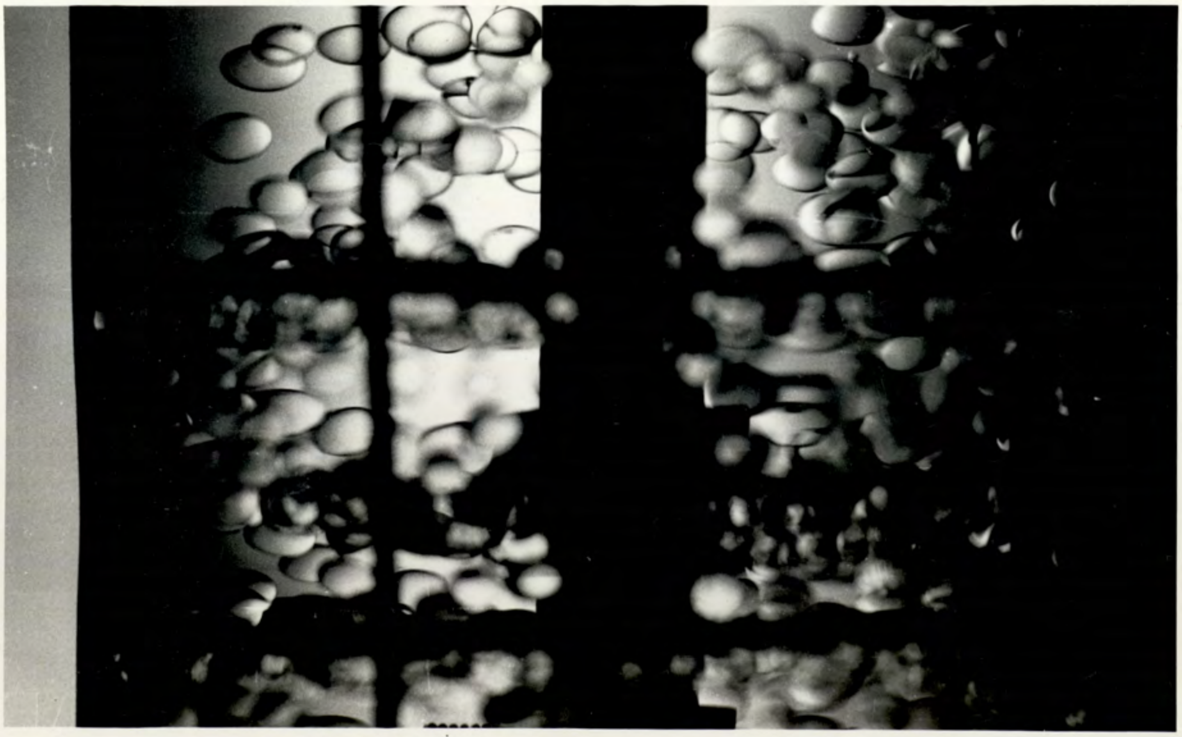


FIGURE 63, R.D.C. SECTION WITH ROTOR SPEED WELL BELOW THE CRITICAL MINIMUM, TRIMETHYPENTANE - WATER SYSTEM ONLY;
Mono-layer of drops beneath stator (N = 200 r.p.m.)

Table 4.

Observed Critical Minimum Speeds - R.D.C.

Toluene - Water	650 r.p.m.
M.I.B.K.- Water	550 - 600 r.p.m.
Trimethylpentane - Water	650 - 700 r.p.m.

Contrary to the observations of Kung and Beckmann (83), droplets did not build up beneath the stators at finite rotor speeds with the exception of a monolayer present in all runs with trimethylpentane at rotor speeds below 200 r.p.m. as shown in Figure 63.

From observations of high speed cine film (Appendix 7) and the use of coloured dispersed droplets, as described in para.11.5, the drops beneath the discs were found to be discrete and to maintain their identities, that is no coalescence occurred. The population changed by drops sliding to the periphery before being thrown off with insufficient momentum to impinge on the column wall. At these low speeds the turbulence existing in the continuous phase was insufficient to cause shearing.

It appeared likely that a small amount of extraneous turbulence in the volume beneath the disc would be sufficient to eject the drops and this was tested. The rotor was inverted so that the retaining collar was on the underside and a grub screw inserted and made to protrude ¼ inch. A significant but irregular decrease in critical speed was achieved.

A summary of drop-size measurements made below the normal critical minimum rotor speed is given in Table 5. in Appendix 5.

Typical droplet behaviour at higher rotor speeds is illustrated in the cine-film - Appendix 7.1. Analysis of high and normal speed cine films confirmed that droplet flow patterns were as described earlier (83) ; axial mixing became increasingly superimposed upon the general toroidal path as impeller speed was increased.

A summary of drop size measurements above the critical minimum rotor speed is given in Table 6. in Appendix 5.
Typical photographs comprise Figures 39 and 40.

11.3.4. Oldshue-Rushton Column

At low agitator speeds with all 3 systems the agitator functioned as a paddle. The characteristic flow pattern was tangential and the main function of the agitator was to disperse droplets throughout the continuous phase. This is illustrated in the cine film - Appendix 7.1. Observation of cine films indicated that break-up of drops in this region was due not to dynamic pressure fluctuations but was caused in the main by impact with the agitator. A summary of drop size measurements is given in Table 7. in Appendix 5.

At verylow agitator speeds, as described earlier, a floculllation zone of droplets was present beneath the stator ring as shown in Figure 64. At such low speeds droplets were not efficiently distributed over the column cross-section.

With the systems studied, the transition from the tangential flow pattern to a radial pattern characteristic of a turbine occurred gradually in the range of speeds 350 to 450 r.p.m. with a mean of 400 r.p.m. At higher speeds within the practicable operating range; the agitator functioned as a turbine generating flow patterns similar to those shown in Figure 30. A typical drop size distribution is shown in Figure 65. Drop size measurements are summarised in Table 8. in Appendix 5.

Reference has been made earlier to the tendency for emulsification at high rotor speeds.

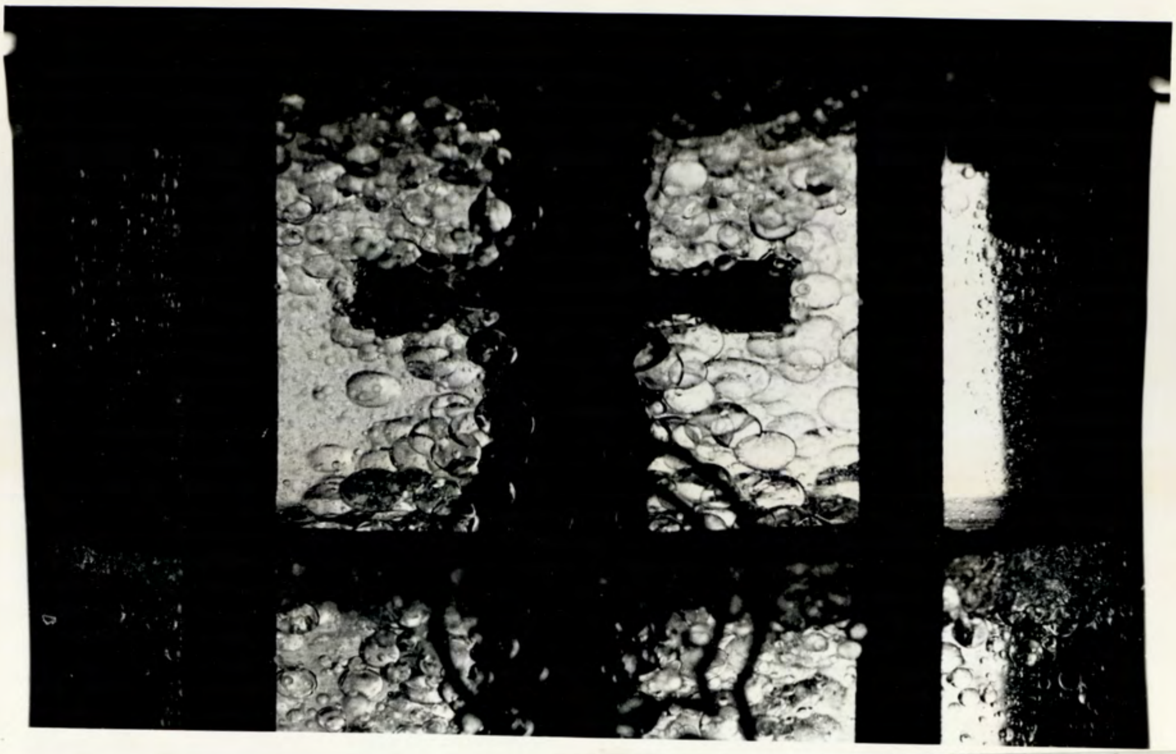


FIGURE 64, OLDSHUE-RUSHTON SECTION WITH AGITATOR SPEED WELL BELOW THE CRITICAL MINIMUM. Flocculation zone beneath stator and uneven distribution over column cross-section (System: M.I.B.K. - water, N = 200 r.p.m.)

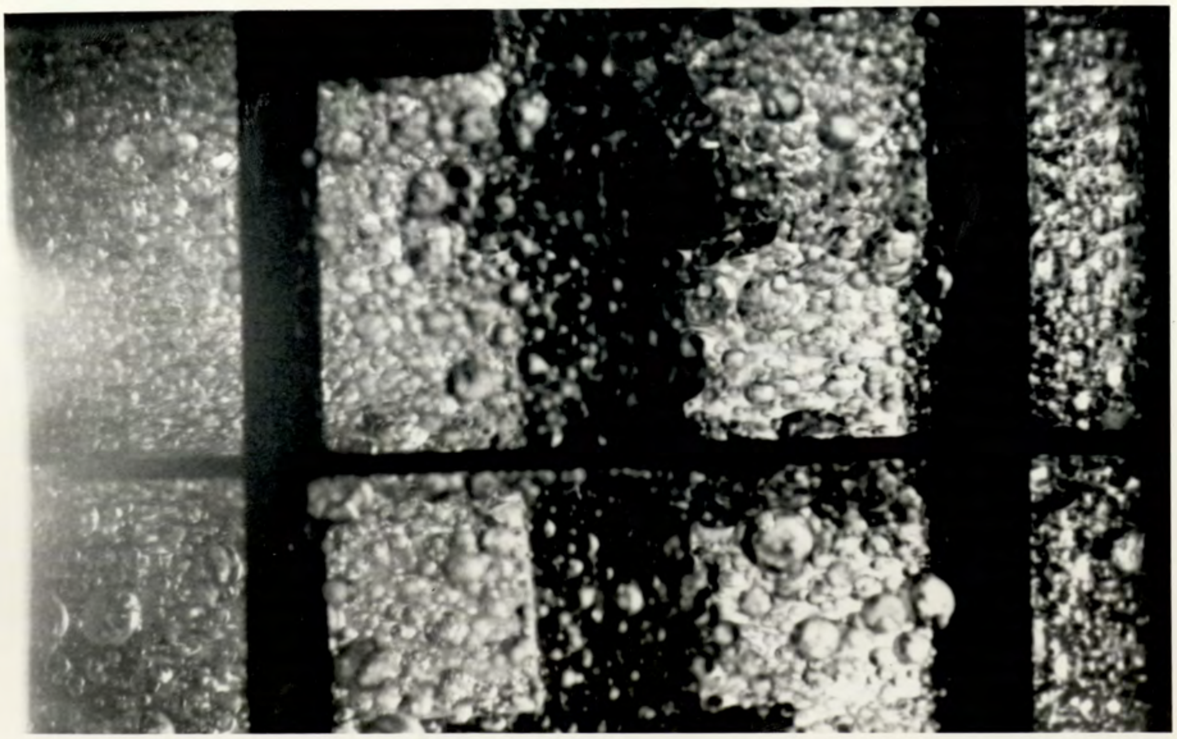


FIGURE 65, OLDSHUE-RUSHTON SECTION - TYPICAL DROPLET SIZE DISTRIBUTION (System : Toluene - water, N = 500 r.p.m.)

11.3.5 Ex. Distributor

In order to check that initial drop sizes into the agitated zones did not vary greatly within the range of flow-rates employed in drop size determinations, droplets from the distributor were photographed and counted in the usual way. The results are summarized in Table 9. in Appendix 5.

At moderate velocities through the orifices, a neck of dispersed liquid was visible as illustrated in Figure 66. The shape of drops at the moment of break away is as described by Null and Johnson (138), that is a truncated cone adhering to the edge of the orifice and joined by another truncated cone attached to a hemisphere. At larger velocities the neck increased in size as illustrated in Figure 67. The extreme of atomisation at the orifice was not reached.

Mean drop sizes are plotted against linear velocity at the orifice in Figure 68. These verify the information given by Treybal for properly designed distributors in that up to 0.33 ft./sec. the variation of drop size is relatively small (11). Hence it has been recommended that practical nozzle velocities should not exceed about 0.5 ft./sec. (54).

In this work drop size measurements in the agitated sections were limited to low hold-ups corresponding to low nozzle velocities.

11.4 WETTING PHENOMENA

Normal experiments indicated that preferential wetting of the internal column surfaces by the continuous phase deteriorated with time. Similar effects have been reported in pulsed plate laboratory columns (139). Tests were performed therefore to investigate what the effect of this, or the selection of dispersed phase wetted materials of construction, would be on column capacity and efficiency.

The packing in the Scheibel column was replaced by two, 2 inch thick sections of polypropylene 9036 mesh packing by Knitmesh Ltd. By the nature of its fabrication this packing is inhomogeneous. Runs

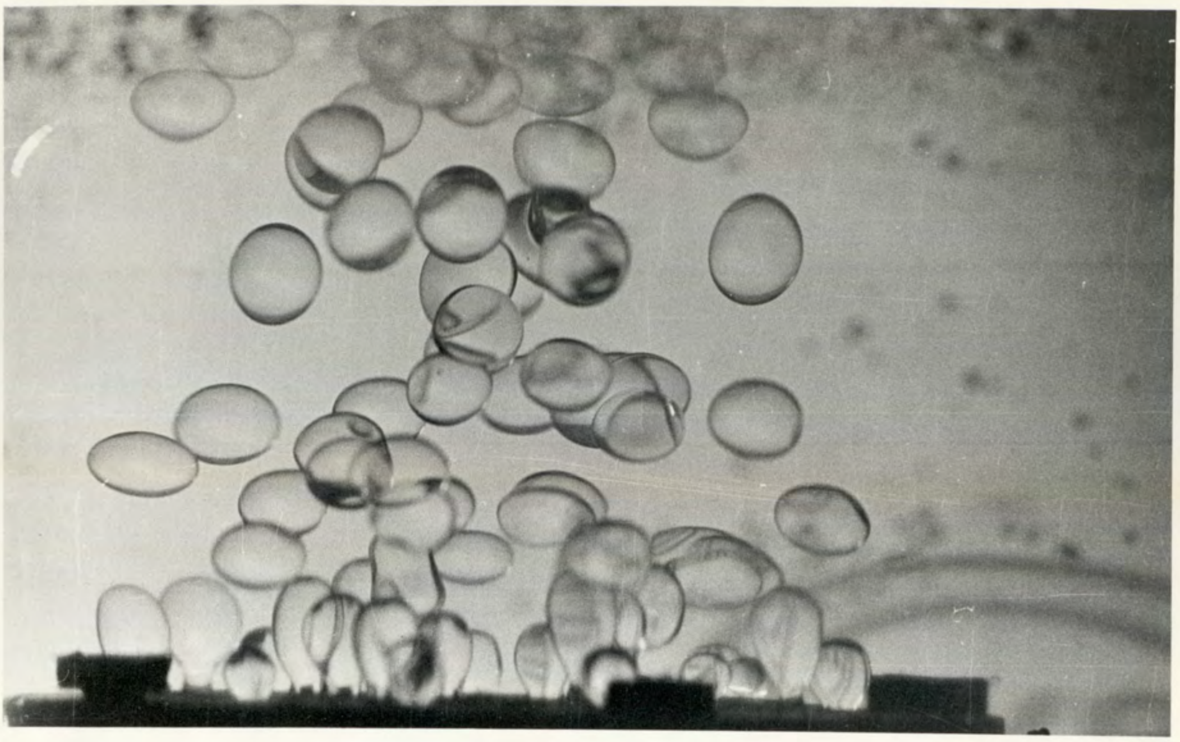


FIGURE 66, DROPLET FORMATION FROM DISTRIBUTOR - MODERATE VELOCITY

(System : Toluene - water, V $\doteqdot$ 0.32 ft.sec.$^{-1}$)

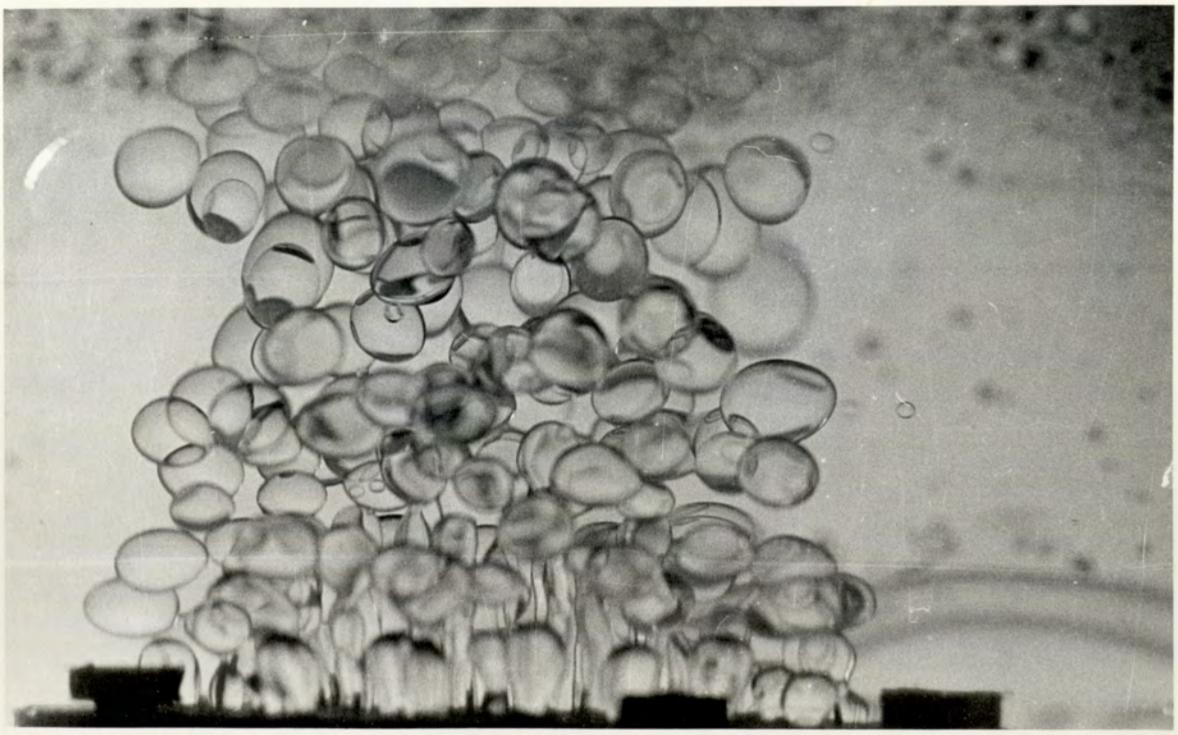


FIGURE 67, DROPLET FORMATION FROM DISTRIBUTOR - HIGH VELOCITY

(System : Toluene - water, V $\doteqdot$ 0.69 ft.sec.$^{-1}$)

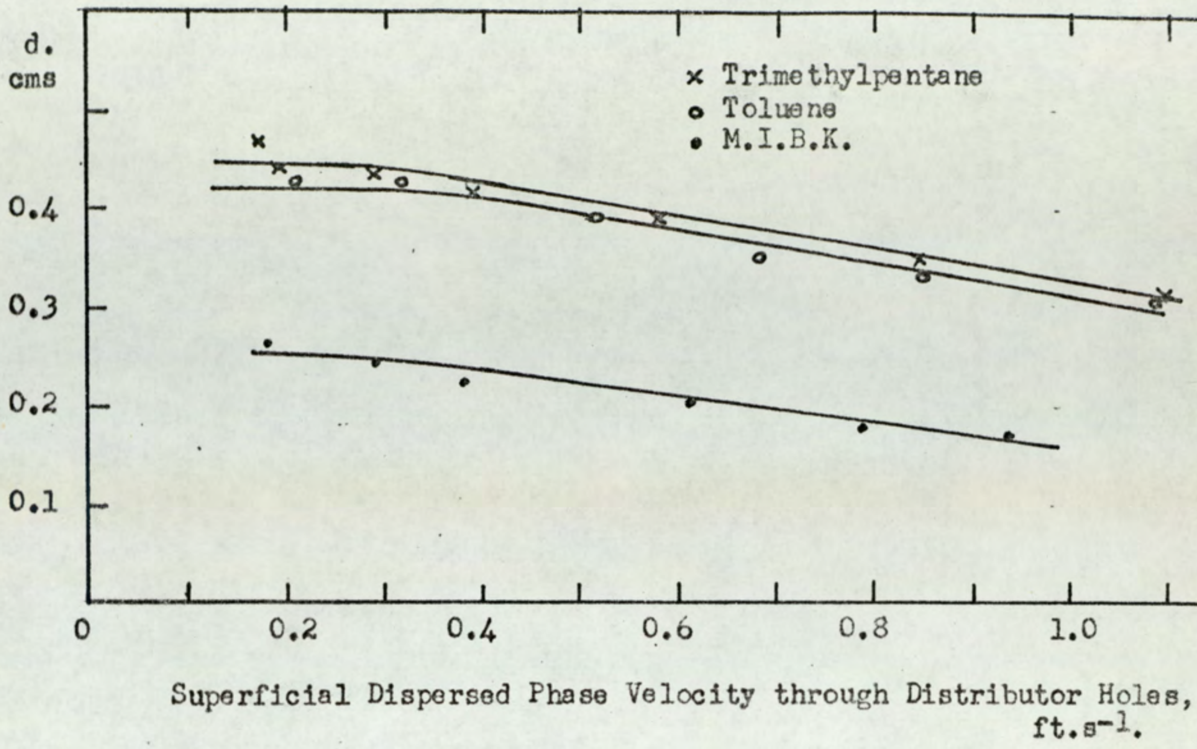


FIGURE 68. MEAN DROP SIZES EX DISTRIBUTOR.

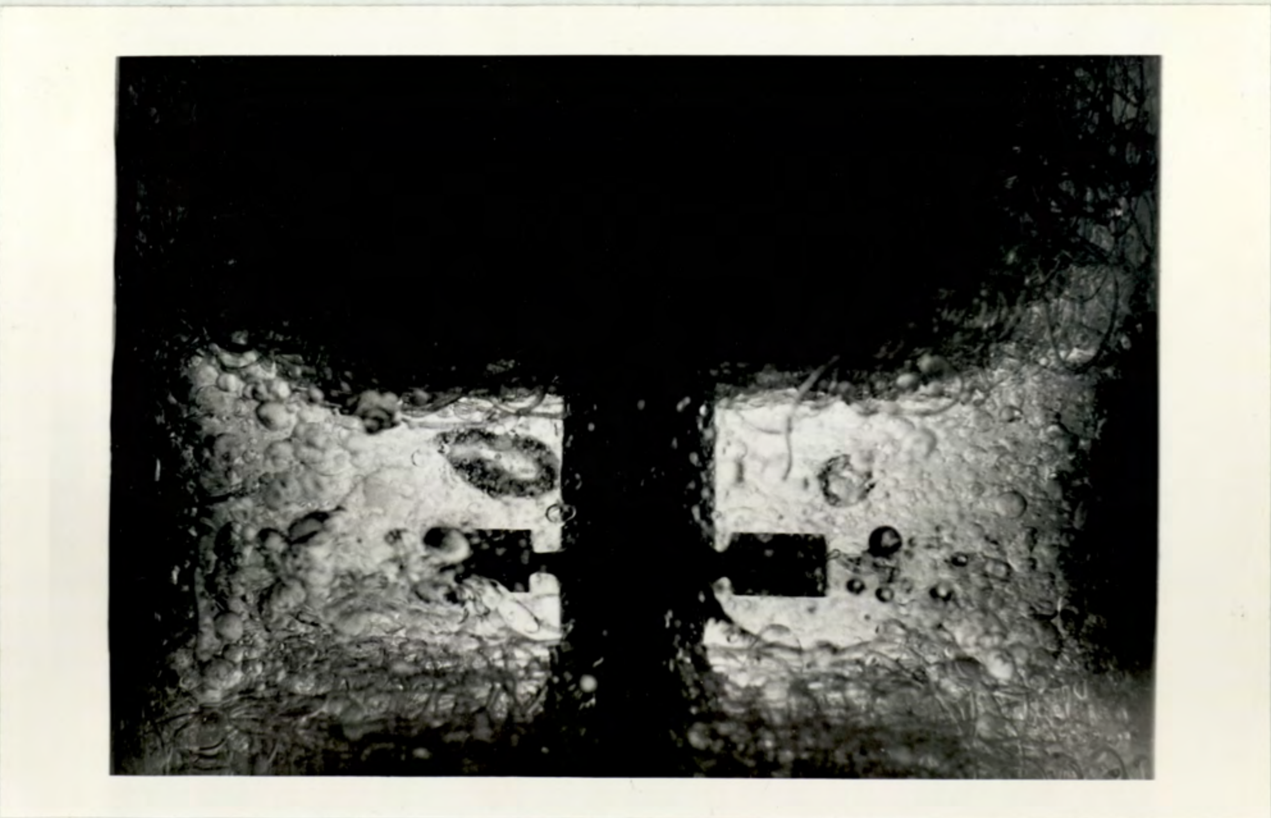


FIGURE 69. SCHEIBEL SECTION WITH POLYPROPYLENE PACKING - NORMAL OPERATION.

(System : M.I.B.K. - water, N = 700 r.p.m. V = 43.4 ft. sec.$^{-1}$)

were performed with the system M.I.B.K. - water. At low throughputs and normal rotor speeds, observations and photographs showed that droplet characteristics were similar to those for steel packing but with an increased tendency for drip-point globule formation; Figure 69 is typical. At zero rotor speed jetting occurred and as throughput was increased numerous jets passed through the agitated section without break-up. As throughput was increased at finite rotor speeds a transition point was reached at which the upper surface of the lower section of packing and the lower surface of the upper section became covered with a pool of dispersed phase. An emulsion of fine droplets was then formed in the agitated zone. This occurred prior to true flooding as characterised by complete rejection of the dispersed phase. Conditions just prior to this transition have been illustrated in the paper forming Appendix 1.3. (7).

Wettability of the turbine agitators, as distinct from the shaft which could provide a by-pass for the dispersed phase, had no significant effect.

In the R.D.C. initial observations suggested that the critical minimum speeds described earlier were dependant on cleanliness of the rotor discs which would deteriorate in practice. Therefore to investigate this, runs were made with the system toluene-water with the disc in one compartment replaced by one fabricated from p.t.f.e. Organic liquids exhibit large contact angles with this plastic.

At all rotor speeds gross coalescence of droplets occurred beneath the rotor disc producing a layer of dispersed phase and a circular element was attached to the tip of the disc. The width of this element varied with rotor speed as shown in Table 10.

Table 10.

Element width & p.t.f.e. rotor speed.

Rotor speed r.p.m.	Approx. width of peripheral element, c.m.
200	> 0.3
400	0.25
850	0.16
1000	< 0.1

Measurement was made difficult at high rotor speeds by the high hold-ups generated. At a critical speed of approximately 850 r.p.m., corresponding to an $Re \doteq 2.3 \times 10^4$, the layer of dispersed phase thinned appreciably and was no longer observable. Similar observations were made with a disc made of polypropylene.

The phenomena is illustrated in the paper comprising Appendix 1.3 (7) ; Figure 70. shows the same effect at a low rotor speed.

So far as could be observed, flooding commenced in the plane of the p.t.f.e. disc. The flooding curves are plotted in Figure 71. together with those for all brass discs for comparison.

Similar tests were made with the system toluene-water in the R.D.C. section using standard brass rotor discs but p.t.f.e. plastic stator rings on threaded brass spacer rods.

At all rotor speeds (up to 1000 r.p.m.) gross coalescence of dispersed phase occurred on the under and upper sides of the stators. Large globules of 10 to 15 m.m. equivalent diameter then left the stators from random positions by drip-point formation. These moved spirally through each compartment before coalescing on the next stator. Hence the mean drop size was greatly increased; the extent could not be measured since drip-point formation varied with time. The size of globules leaving the stators appeared to be largely independant of rotor

FIGURE 70, R.D.C. SECTION WITH P.T.F.E.ROTOR DISC;

Illustration of spread of dispersed phase beneath disc and attachment at periphery
(System : toluene - water, N = 400 r.p.m.)

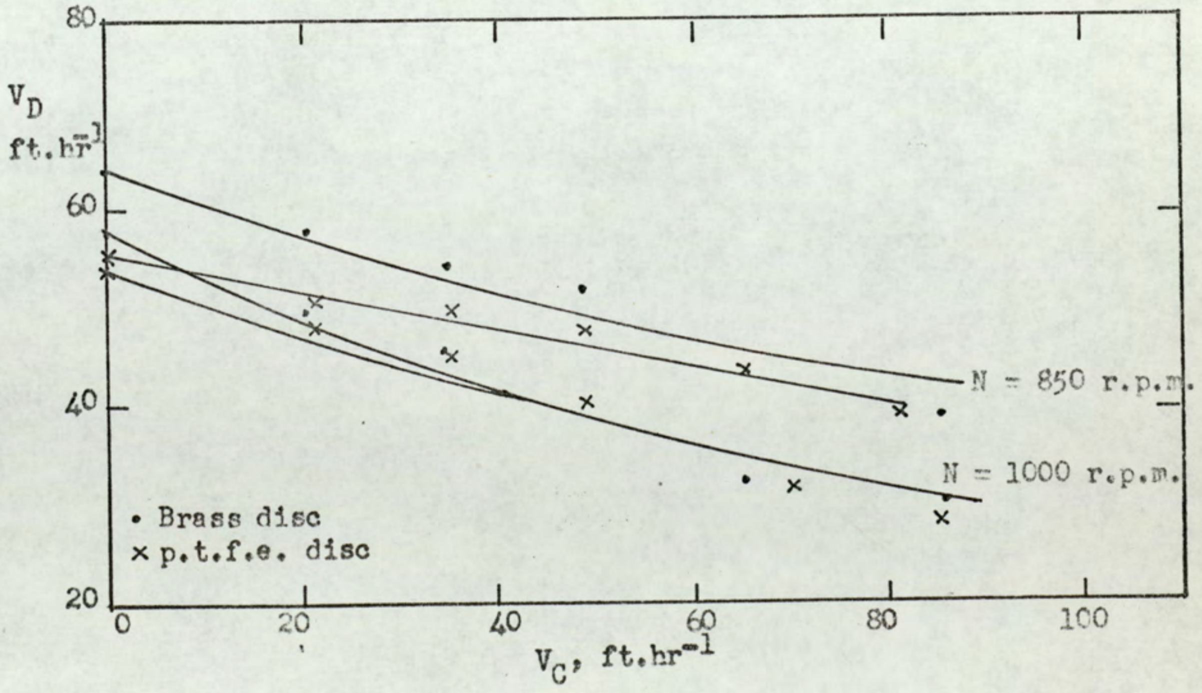


FIGURE 71, R.D.C. SECTION - FLOODING CURVES WITH P.T.F.E. DISC
(System : toluene - water)

speed in the range 500 to 1000 r.p.m. The phenomena is illustrated in Figure 72. At zero rotor speed the mean size of the globules was somewhat greater, providing an illustration of the mode of operation of an inefficient disc and doughnut baffle plate column.

The phenomena of drip point formation of globules from p.t.f.e. stator rings was also observed in the Oldshue-Rushton section as shown in Figure 73. The size of globule produced has been illustrated in the paper (4) - Appendix 1.2. It is of interest that for organic droplets formed from an orifice an increase in size up to 10 fold has been reported for p.t.f.e. compared with metal plates (52).

11.5. DROP-DROP COALESCENCE.

Tests were made to establish the extent of inter-droplet coalescence in the various columns using the 3 different systems. The importance of this in design is discussed in para. 7.8.

Hold-up, drop-size distribution and flow patterns (i.e.residence times) comprise the important parameters. The earlier limited work of Howells and Steen neglected these (94,95).

Coloured droplets of substantially the same size as those from the distributor were introduced into the column via a single sharp-edged orifice from a 100 c.c. hypodermic syringe. The arrangement is shown in Figure 74. The organic soluble dye used was Calco Oil Red N - 1700 ex American Cynamid Co. This dye has been recommended by Kintner because it has no effect on interfacial tension and is devoid of other interfacial activity for organic liquid-water systems (140). A spread of dye amongst a significant number of droplets, giving a pink tinge, was taken as evidence of a coalescence-redispersion mechanism.

In the R.D.C. section, runs were performed at fixed rotor speeds of 500, 700, 850 and 1000 r.p.m. and at various flowrates up to flooding. With the system toluene-water, in no case was inter-droplet coalescence significant until the flowrates approximated to the flood-

FIGURE 72, R.D.C. SECTION WITH P.T.F.E. STATORS ;

Dispersed phase leaving stator by drip-point formation with shearing by disc to produce large globules
(System : toluene - water, N = 850 r.p.m.)

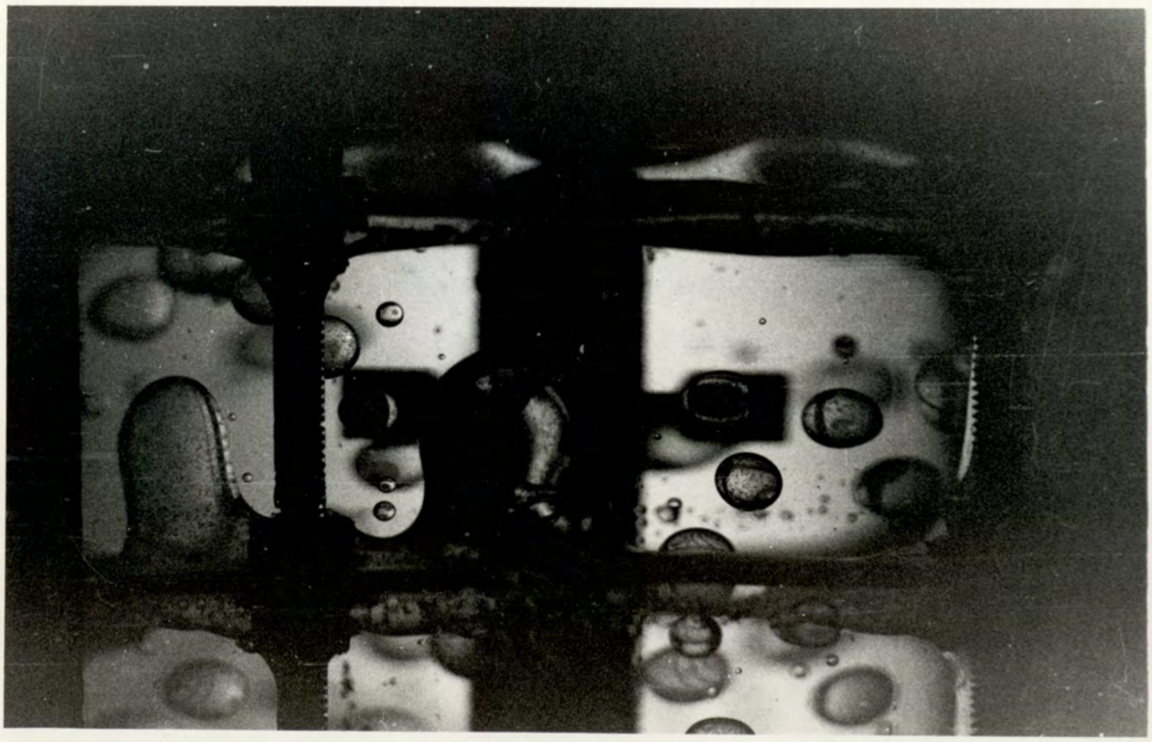


FIGURE 73, OLDSHUE-RUSHTON SECTION WITH P.T.F.E. STATORS ;

Drip point formation from stators
(System : toluene - water, N = 200 r.p.m.)

points determined earlier. This absence of coalescence was borne out by a study of all high speed cine film taken albeit necessarily at low hold-ups. At 500 r.p.m. and low values of hold-up visual observation of injected coloured droplets confirmed the observations from cine-films that droplets trapped beneath the rotor were discrete and constituted a continually changing population. At all rotor speeds with clean, continuous phase wetted internals surface coalescence was insignificant.

With the system M.I.B.K. - water, drop-drop coalescence did not occur until the flood-points were reached corresponding to very high hold-ups and residence times . A similar result was obtained with the system trimethylpentane. With the latter system droplets trapped beneath the stator rings at low rotor speeds e.g. 200 r.p.m. were found to be discrete and continually changing.

In one section of a high speed cine film a droplet was observed to spin and break-up in about 0.02 seconds. This is included in the film - Appendix 7.1. From the forces acting on droplets, discussed later in para. 12.4.1. some rotation would be expected about a vertical axis, and to a lesser extent about a horizontal axis. This could be one factor inhibiting coalescence. An attempt was made to investigate this using black acrylic plastic spheres ($\rho \doteq 1.3$ gm. per c.c.) of 0.318 cm. diameter dispersed in water. One half of each sphere was hand painted white. A small number were introduced into the top of the column and photographed at rotor speeds of 500,850 and 1000 r.p.m. using the Fastax camera with a framing speed of 500 p.p.s. Typical results are shown in the cine film - Appendix 7.2.

Unfortunately reflection of light precluded the measurement of droplet spin rates observed visually. Variation of lighting did not yield better results and since each trial consumed 100 feet of film this work had to be curtailed. The acceleration and subsequent retardation experienced by a 'droplet' as it traversed a compartment (94) was however clearly recorded.

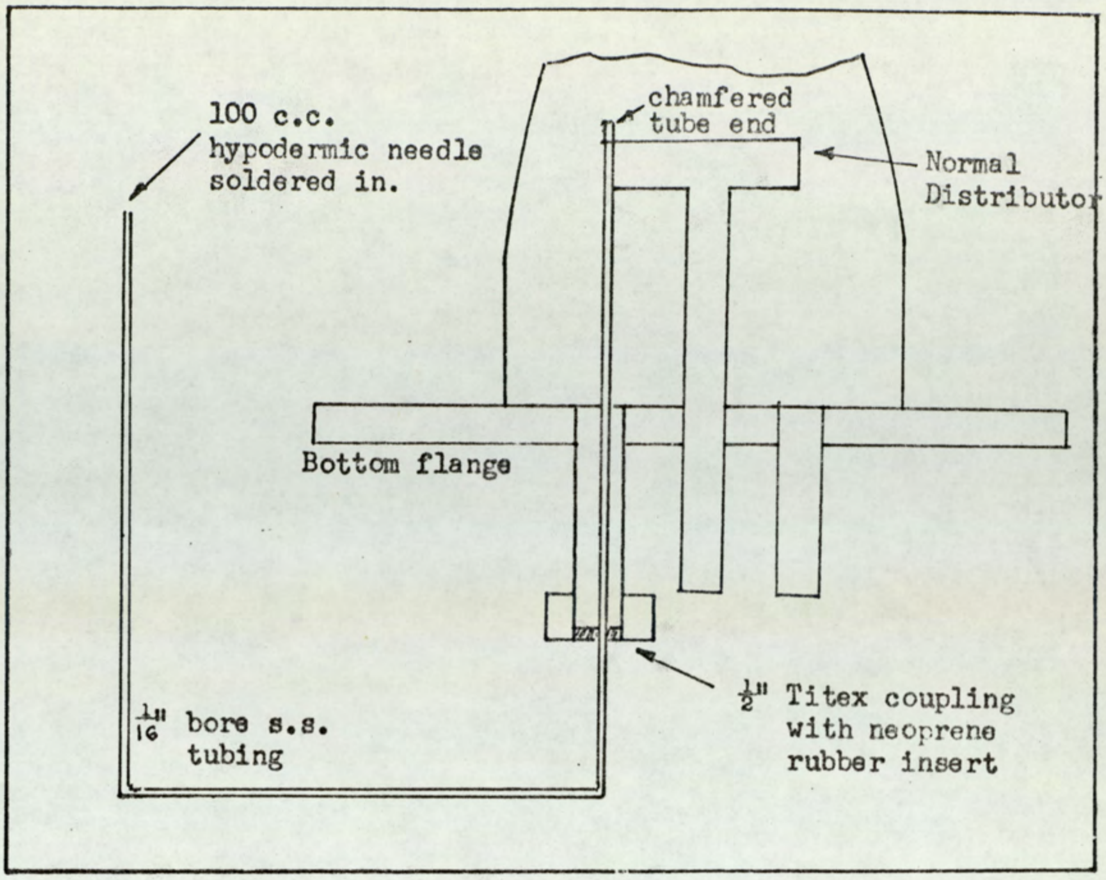


FIGURE 74. ARRANGEMENT OF INJECTOR

for Coloured Droplet studies

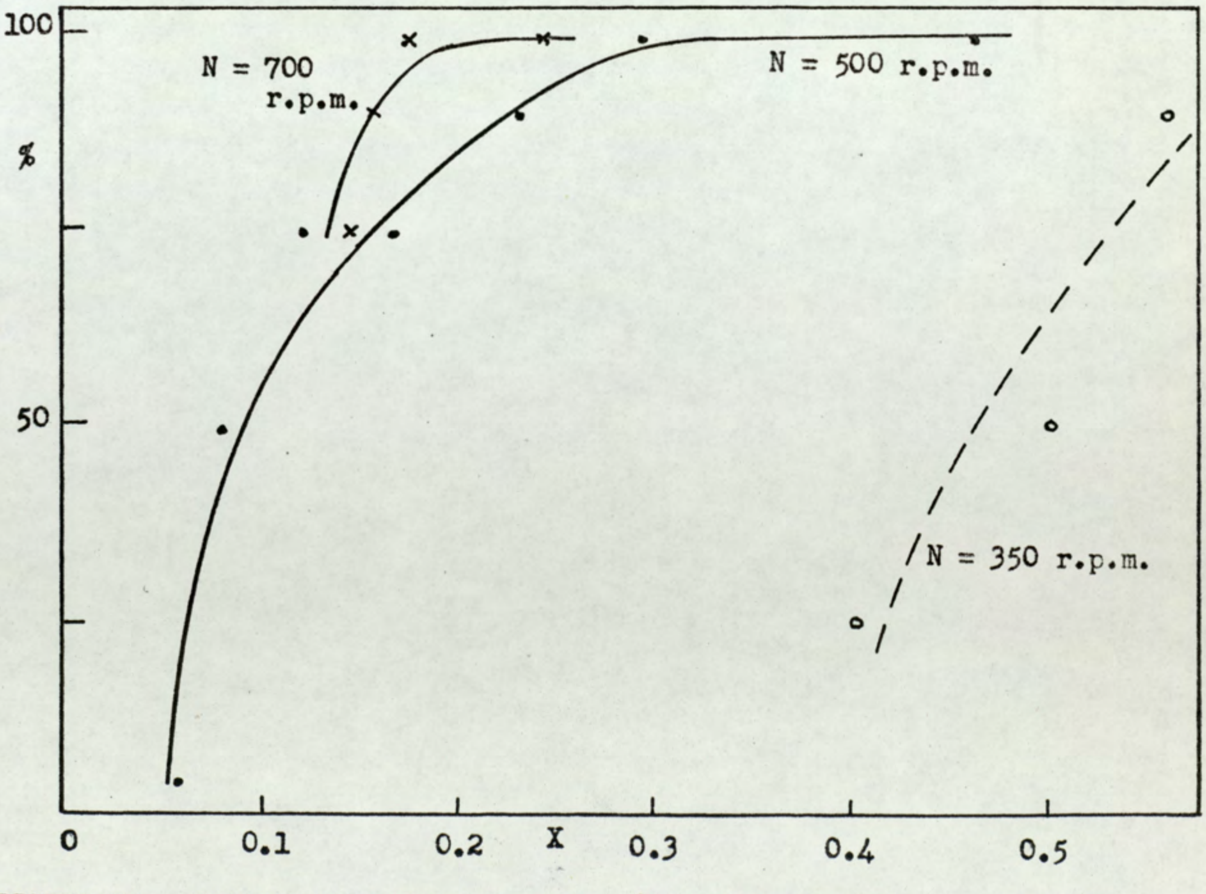


FIGURE 75. PERCENTAGE COLOURED DROPS (VISUAL ESTIMATE) vs HOLD-UP - OLDSHUE-RUSHTON COLUMN (system: toluene - water)

Coloured droplet studies were performed in the Oldshue-Rushton section. With all three systems at practical operating speeds, and at all but the lowest hold-ups, a spread of dye was almost complete throughout the dispersed phase. That is a coalescence-redispersion mechanism normally predominated.

For the system toluene-water, estimates were made of the percentages of coloured drops leaving the agitated zone. These were based on visual not photographic observation. The data for various agitator speeds are plotted against hold-up in Figure 75. A few droplets maintained their identities. Droplet residence time is clearly an important parameter not allowed for here, but in similar work in a 12 stage column with toluene dispersed in water, hold-ups in the range 0.05 to 0.36 and rotor speeds of 150 to 300 r.p.m. persistence of a few discrete drops has been reported (93).

At low operating speeds the onset of drop-drop coalescence was not easily determin able due to bulk coalescence effects in the dynamic flocculation zone created beneath the stator rings as described earlier.

With all 3 systems in the Scheibel column, spread of dye was effectively complete at all but the lowest agitator speeds. This predominance of the coalescence - redispersion mechanism down to very low hold-ups and agitator speeds supports the earlier description of the packing as a coalescing aid under these conditions. The dependance on agitator speed arose only because of the need to adequately distribute the coloured droplets across the column cross-section. At very low agitator speeds drops or globules tended to travel in definite paths through the packed and agitated sections and hence the mechanism was inhibited.

12. DISCUSSION OF RESULTS.

12.1. HYDRODYNAMICS

In all cases in the R.D.C., hold-up increased with increasing dispersed phase flowrate. The increase was sharper beyond a point corresponding to about 40 - 50 % of the flooding point suggesting that some change in flow regime occurred in this region. Similar phenomena have been described in packed columns (11) and related to the loading condition in gas -liquid contact. The shape of the curves and quantitative values of hold-up are in agreement with the results of others (83,102).

As will be shown later (Figure 81), the variation of d_{vs} with X was small at high values of X. Therefore, it would be expected that flooding data could be correlated approximately by the characteristic velocity approach (102), given in para. 7.4. This is illustrated in Figure 76, using the data from Figure 49. Reasonable straight lines capable of extrapolation through the origin are obtained for V_d vs $X_f^2 (1 - X_f)$ (102). The data give capacities about 15% higher than that obtained by Logsdail et.al. after allowance has been made for improved column geometry by means of the restriction factor C_R.

In practice the characteristic drop size is dependant on the mass transfer process. The effect is dependant on solute concentration. Hence to utilize the characteristic velocity design approach, outlined earlier, hold-up measurements should be made in the presence of mass transfer and at several points along the length of the extractor.

For the Oldshue-Rushton column the hold-up data versus dispersed phase flowrate curves are of similar form to those for the R.D.C. with the 'loading point' being at about 50% of the flooding point. Hold-up at comparable rotor speeds was of a significantly higher order however.

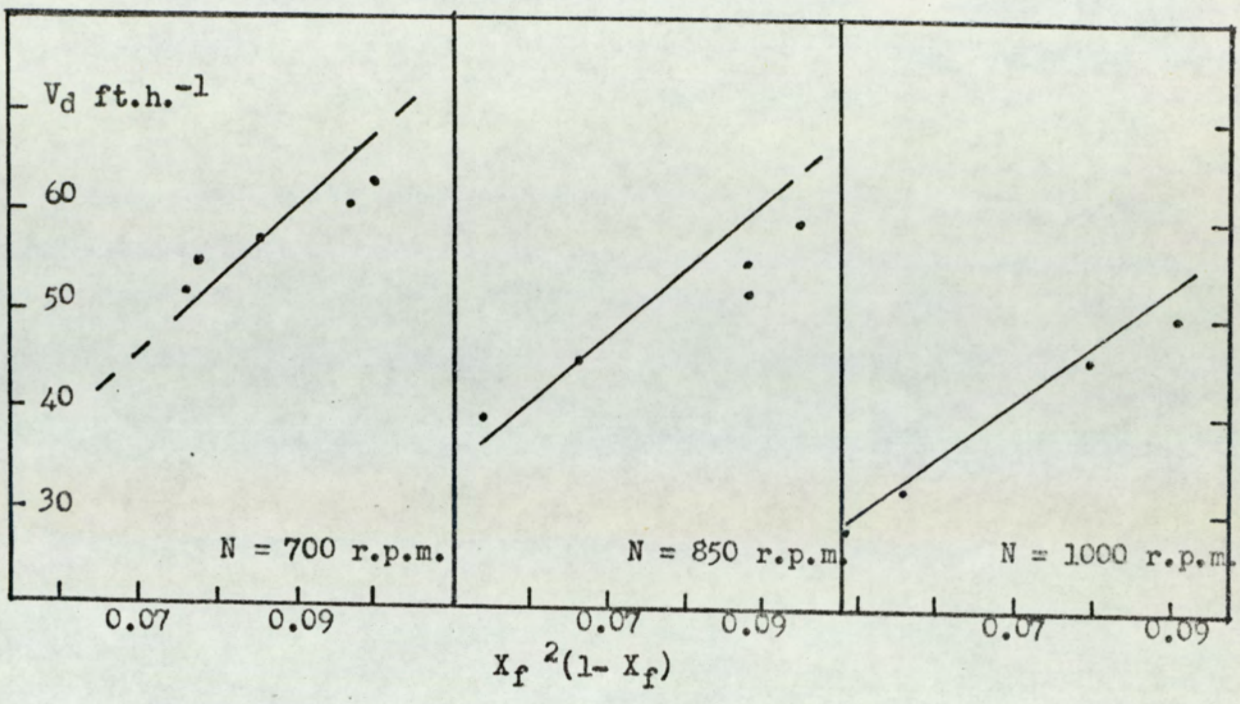


FIGURE 76. CORRELATION OF FLOODING DATA FOR R.D.C. FROM FIGURE 49.

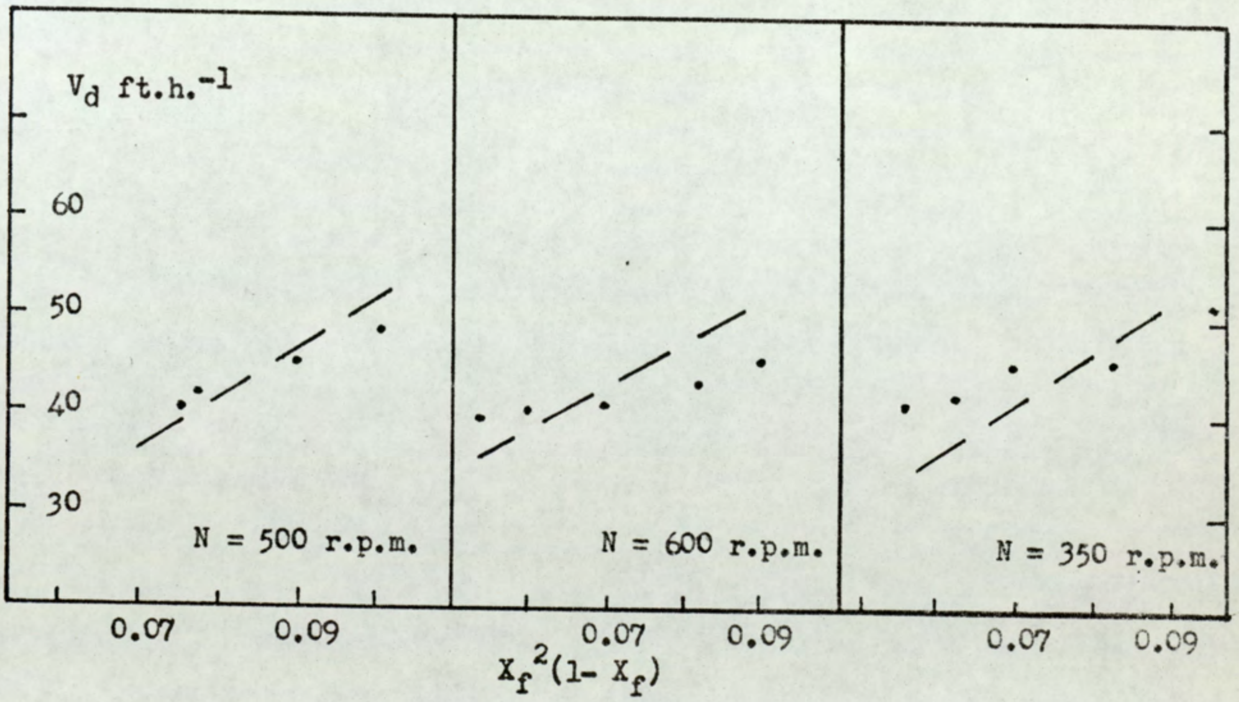


FIGURE 77. ATTEMPTED CORRELATION OF FLOODING DATA FOR OLDSHUE-RUSHTON COLUMN FROM FIGURE 52.

Comparison of flooding data with that for the R.D.C. indicates that the capacities are similar providing both are operated within the practicable operating range. An attempt was made to correlate data for the Oldshue-Rushton column using the characteristic velocity formulae. Results from Figure 52, replotted in Figure 77. suggest that this method cannot be used with confidence. In order to pass through the points plotted, the slope of any curve would need to increase with hold-up. This would be in accordance with $\bar{V}_N$ and hence mean drop diameter increasing with hold-up. Measured values of drop size plotted later in Figure 86, are in agreement with this.

Observations suggest that there should be some advantage, for operation in the low speed range, if stators were fabricated with a gap of approximately $\frac{1}{8}$ to $\frac{3}{16}$ inch in width, between their periphery and the column wall. This would provide an alternative means of escape for flocculating drops and hence inhibit premature flooding.

Since characteristic hold-ups could not be determined for the Scheibel section it would be unrealistic to attempt to correlate capacity in the usual way. The data in Figures 55 to 57, suggest that the capacity, that is $V_c + V_d$, is little different to that of an R.D.C. at comparable rotor speeds.

12.2. DROPLET COALESCENCE AND BREAK-UP

In the Scheibel section, as would be expected, a coalescence-redispersion mechanism predominated with all systems provided the agitator speed was above a minimum necessary to distribute drops across the column cross-section. A mechanism leading to globule formation has been described. Globule formation occurred randomly and therefore both mean drop size and hold-up in the mixing section varied with time. This accounts for the difficulty encountered in measuring hold-ups.

The maximum drop sizes which were observed in the agitated zone are plotted against rotor speed in Figure 78; these are larger

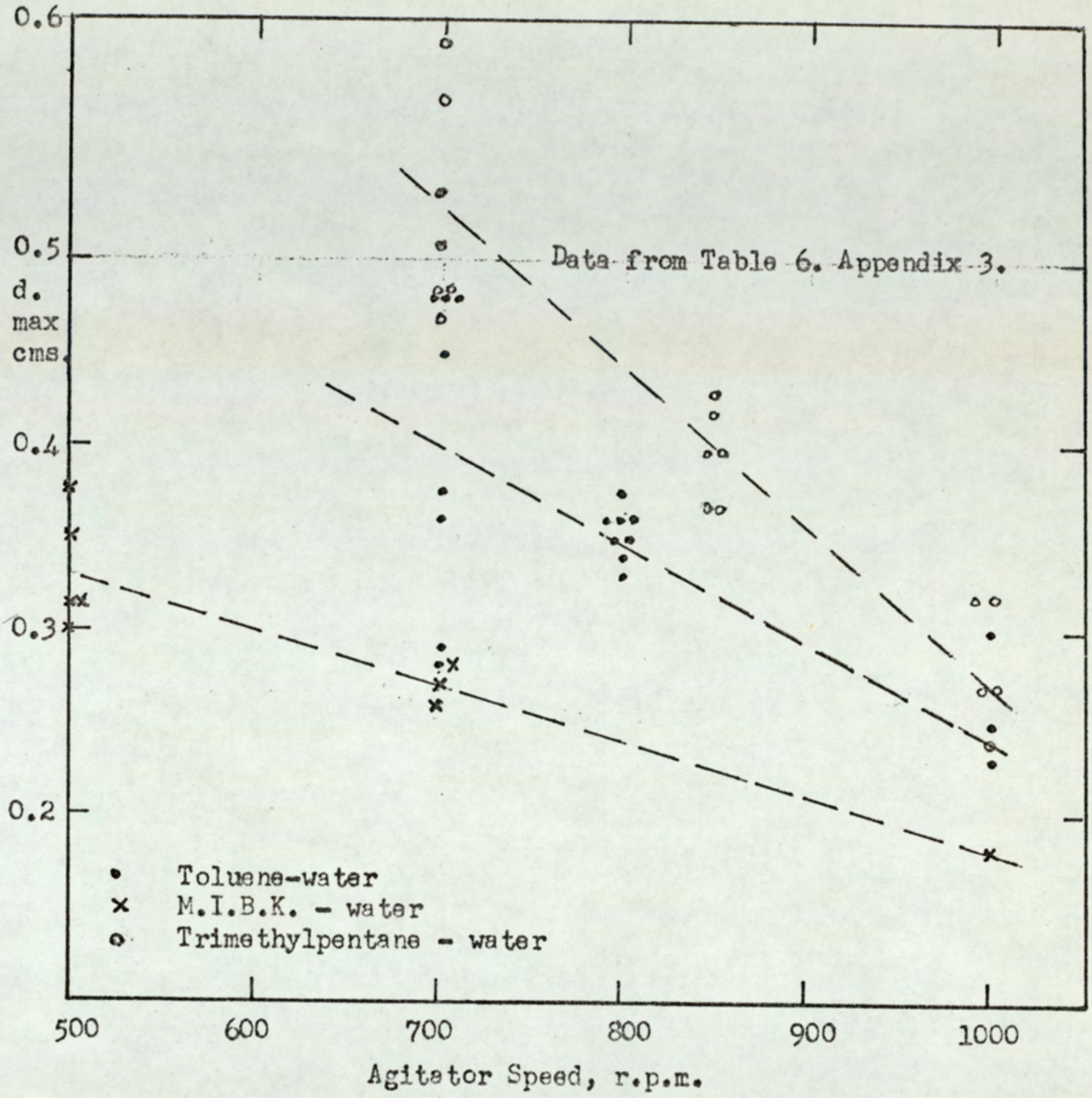


FIGURE 78. SCHEIBEL COLUMN : MAXIMUM OBSERVED DROP SIZES IN AGITATED ZONE.

than those in the Oldshue-Rushton column under comparable conditions. The maximum size is independant of hold-up and decreases with agitator speed. This is in accordance with globules being formed by a shearing mechanism from the packing (although all the measured drops were not necessarily formed in this way). As expected, smallest drop sizes occurred with the system M.I.B.K. - water; this may explain why Honekamp and Burkhart did not observe the mechanism described (9).

The packed sections apparently achieve no purpose which could not be better served by the use of calming zones, either baffled or unbaffled, and could be omitted. No efficiency measurements were made in this work but Scheibel has also proposed replacement of the packings by baffled calming zones for high interfacial tension or high viscosity systems (20) and as described in para. 2.2.1. others have followed suit (23).

Because of globule formation, neither mean drop size or hold-up in the agitated zone are predictable and hence interfacial area cannot be estimated with confidence. A further difficulty arises since, by analogy with gas-liquid behaviour in packed columns (141), the extent of mass transfer which occurs in the packing will be dependant on the proportion of the dispersed phase present as rivulets or droplets. Recent evidence suggests that this could be adjusted, temporarily at least, by pre-treatment of the packing (134).

Drop-drop coalescence has been found to be absent with all 3 systems in the R.D.C. This explains in part the persistence of small drops when present among drops of all sizes. Since practical column operation is normally at less than 80% of flooding rates, then in the absence of any Marangoni effects, the column operates as a true countercurrent, discrete droplet contacting device. Design can therefore be based conveniently on data derived for single droplets of mean diameter. The absence of surface coalescence was as expected but as discussed later the hydrophobic nature of column surfaces was found to be time dependant.

The absence of drop-drop coalescence means that there is no justification for the use of any coalescence factor in R.D.C. analysis and therefore no theoretical basis for its use in the characteristic velocity relationship as given by Misek and included as Appendix 8.

One explanation for the absence of interdroplet coalescence may be the spinning of droplets as described earlier. This would tend to counteract film thinning. The time of contact of any area of colliding droplets would also be extremely short.

With an aqueous continuous phase the critical minimum speed for the rotor in the R.D.C., though slightly dependant on interfacial tension, occurred at $R_e \doteq 1.75 \times 10^4$. Observations confirmed that the mechanism of droplet break-up changed at this point. Below it the mechanism resulted in a smaller spread in drop sizes, as shown in Figure 79.

It is concluded that this Reynolds number delineates the laminar and transition zones (46). Misek did not comment on this phenomena but has given a value of $R_e = 1 \times 10^4$ for the transition point. In the laminar region the mean drop sizes were, for all practical purposes, independant of hold-up. Support for this is provided by the results of Kung and Beckmann described earlier (83) ; they found with the system toluene-water at speeds below a critical value of 300 ft.min.$^{-1}$ the characteristic velocity $\overline{V}$ remained constant. Mean drop sizes gave good agreement with Equation 20 as shown by Figure 80, with the numerical constant having a value of 0.55.

However, visual comparison of drop size distributions with those when the rotor disc was stationary confirmed that break-up was not due solely to impact on the disc and stator ring. For example, with a stationary rotor trimethylpentane drops were observed to bounce 2 or 3 times on impact before either being trapped or flowing away in streamlines. It is of interest to consider the mechanism of droplet break-up and this is discussed in para. 12.4.1.

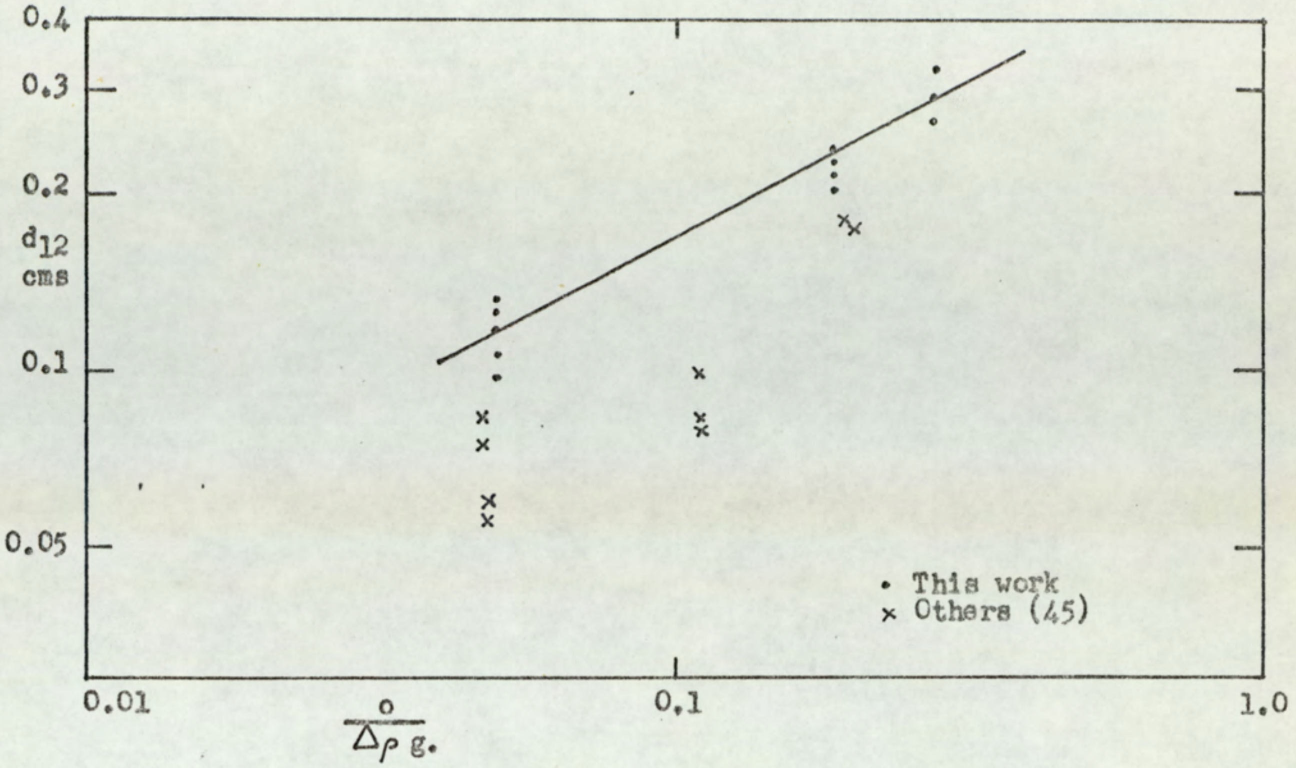


FIGURE 80. CORRELATION OF HYDRAULIC MEAN DROP DIAMETERS – R.D.C. LAMINAR REGION.

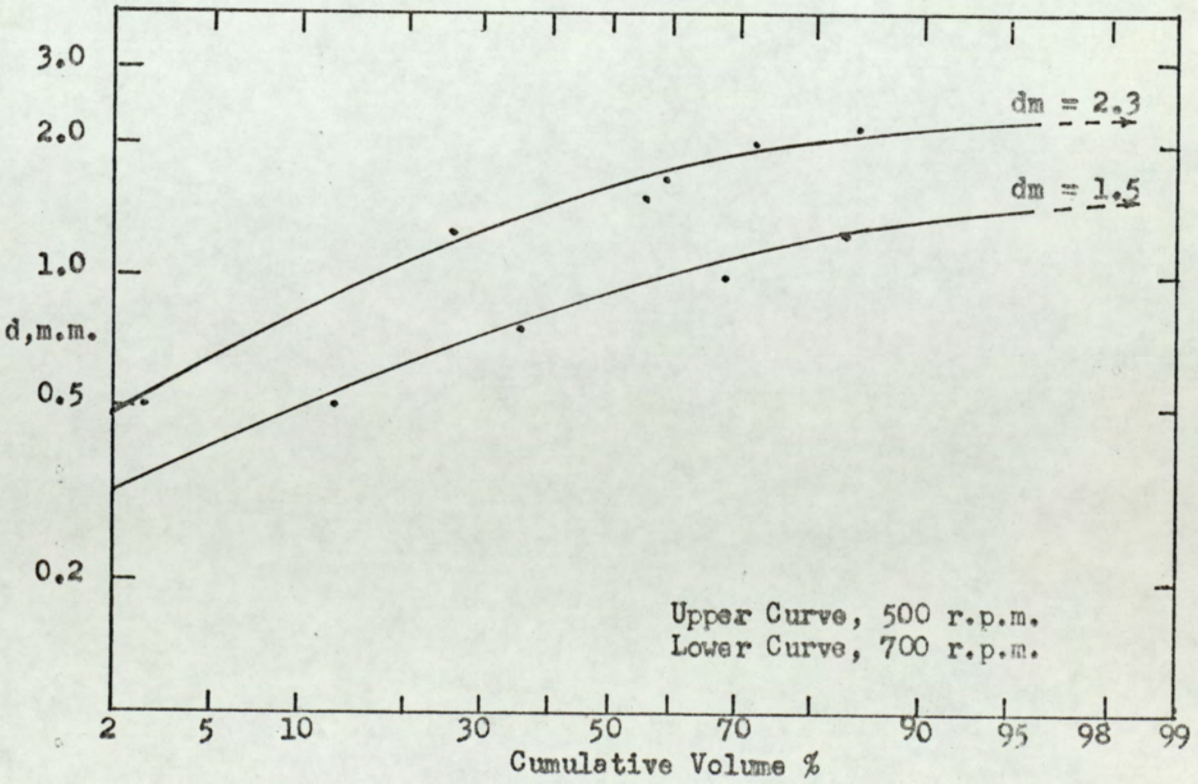


FIGURE 79. DROP SIZE DISTRIBUTIONS ABOVE AND BELOW $Re \doteq 1.75 \times 10^4$ AT CORRESPONDING HOLD-UPS – R.D.C.

(System : M.I.B.K. – water, X = 0.08) Part only.

At higher rotor speeds droplet break-up was limited by equipment size to Misek's transition region (46). Mean drop sizes were found to increase with hold-up over the whole spectrum thus supporting the observations of Olney (111). This is shown in Figure 81. The variation is not attributable to an increase in coalescence frequency therefore the major factor in the dispersion process is the specific power input $\dfrac{\bar{\rho}\ N^3\ R^5}{\rho_D X.H.D^2}$.

Therefore, correlations such as Equation 19 would appear capable at best of predicting only the order of mean drop size. The change in drop size distribution is however small over the range of hold-ups for which measurements were possible. For example, drop size vs cumulative volume for rotor speeds of 700 r.p.m. and 850 r.p.m. with the system toluene-water are plotted in Figures 82 and 83.

The maximum drop sizes observed for each system are plotted against hold-up in Figure 84. Strand et.al. (87) reported that drop hold-up in the range 5 to 20% and drop residence time had no effect on the value of d_m. The results, covering a range from 3 to 40%, confirm Strand's conclusion for the range above 15% but below this hold-up the increase of d_m with X is more significant with the systems toluene-water and trimethylpentane-water. Results for the system M.I.B.K. showed no significant variation with hold-up within the range 0 to 30%.

Because of the analogy between the transition in droplet hydrodynamics occurring in the Oldshue-Rushton and R.D.C. columns, results for the former operating in the 'laminar region' below $Re \doteq 4 \times 10^3$ are plotted in Figure 85 as in Figure 80 and show fair agreement. The numerical constant in this case is 0.34.

Within this region the main function of agitation is probably not that of creating interfacial area since break-up occurs by impact on the agitator. This is illustrated in Figure 88. Energy may however be dissipated as turbulent eddies within droplets hence improving both film coefficients.

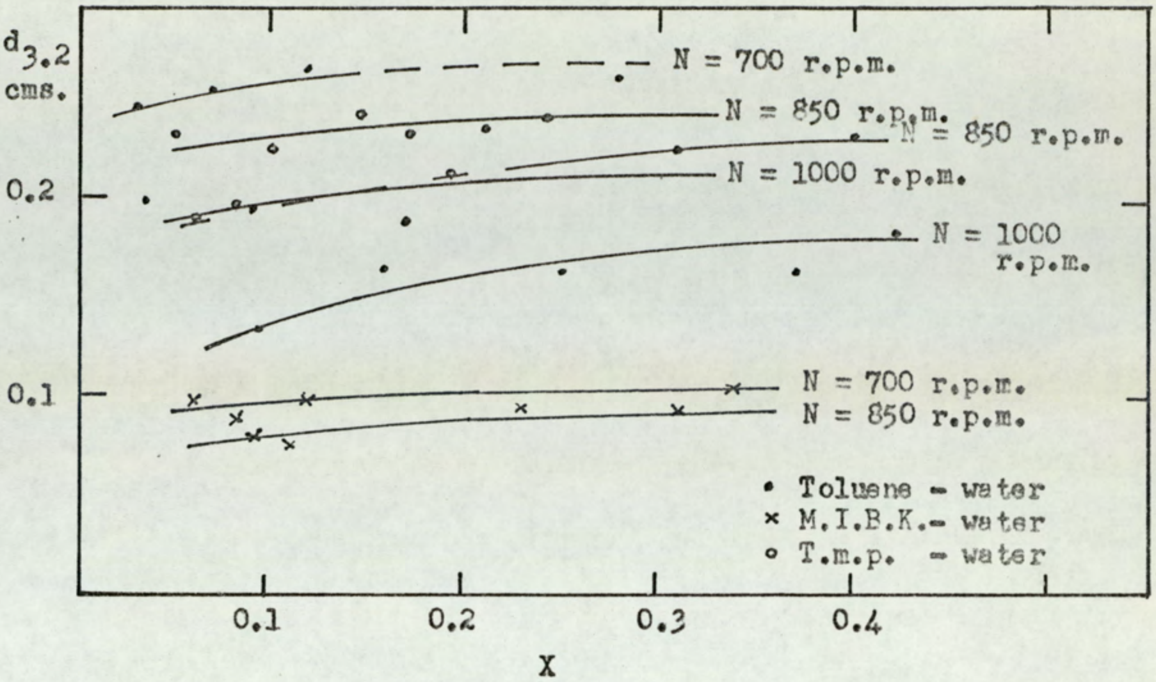


FIGURE 81, VARIATION OF MEAN DROP-SIZE WITH HOLD-UP; R.D.C. TRANSITIVE REGION.

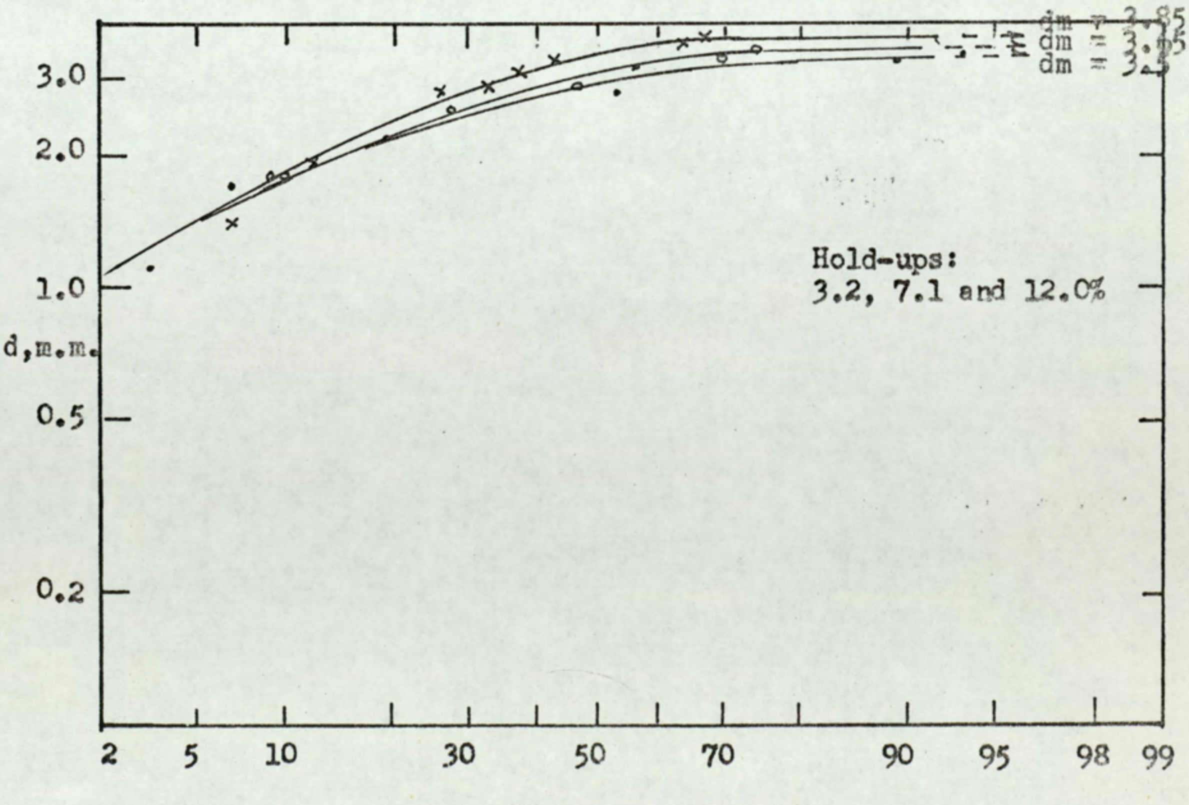


FIGURE 82, DROP SIZE vs % CUMULATIVE VOLUME; (System ; toluene - water, N = 700 r.p.m.) part only.

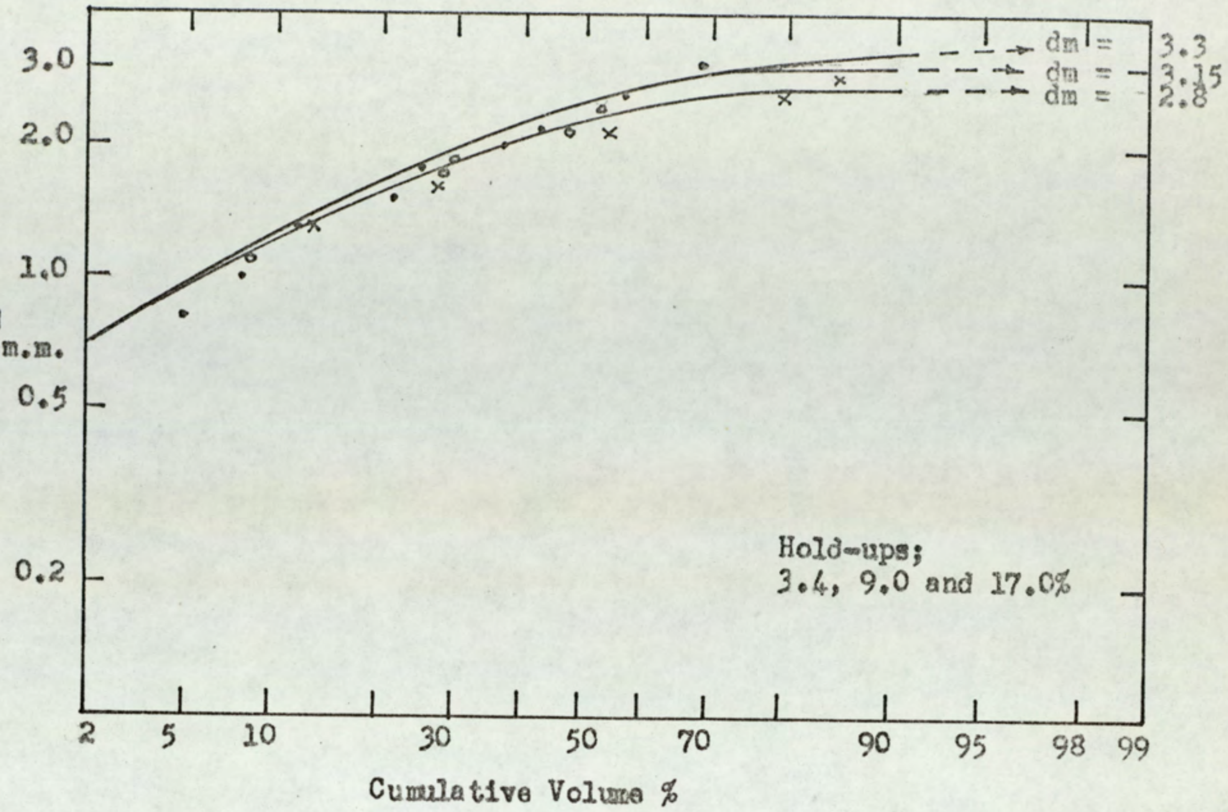


FIGURE 83. DROP SIZE vs % CUMULATIVE VOLUME

(System : toluene - water, N = 850 r.p.m.)
part only.

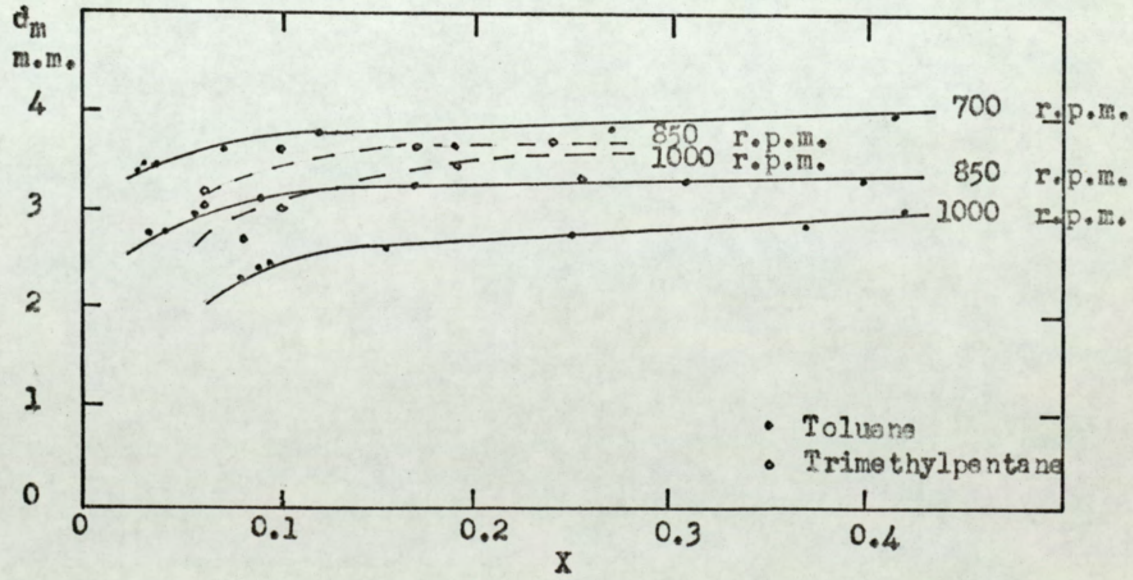


FIGURE 84. MAXIMUM DROP SIZE vs HOLD-UP, R.D.C. TRANSITIVE REGION

At very low values of rotor speed the ability of droplets to leave the flocculation zone beneath a stator ring without being affected by the agitator resulted in a larger mean drop size as in Figure 87. (The numerical constant in Equation 20 is 0.57 for this case). This effect and the uneven dispersion explains the irregular hold-ups determined in this region.

A study of high speed cine film - Appendix 7.1. showed that even at a Reynolds number of 6×10^3, droplet coalescence was localised in the region of the turbines. Although this was necessarily at low hold-ups it does cast doubt both on the theoretical basis for the modified characteristic velocity relationship - Equation 78 (108) and also Resnick's reference to a circulation path involving break-up near the impeller and coalescence along the path in an agitated vessel (81). It is well established however that drop sizes vary throughout agitated vessels.

In the region in which the agitator functioned as a turbine a coalescence-redispersion mechanism predominated down to low hold-ups. Therefore, drop size results are plotted in Figure 86, on the basis of Equation 53. These show good agreement; the mean drop size d_{vs} varies linearly with hold-up and for any one system the slope is independant of power input.

According to Thornton (74), m is a measure of coalescence characteristics. However coalescence frequency must be related to drop size distribution which is itself dependant on agitator speed. Therefore, some variation of m may become apparent over a wider range of operating parameters. In this work the range over which consistent values of d_{vs} were determinable was limited by,

(i) The need to restrict hold-up to obtain photographic clarity for drop counts.

(ii) The avoidance of higher agitator speeds producing a myriad of small drops.

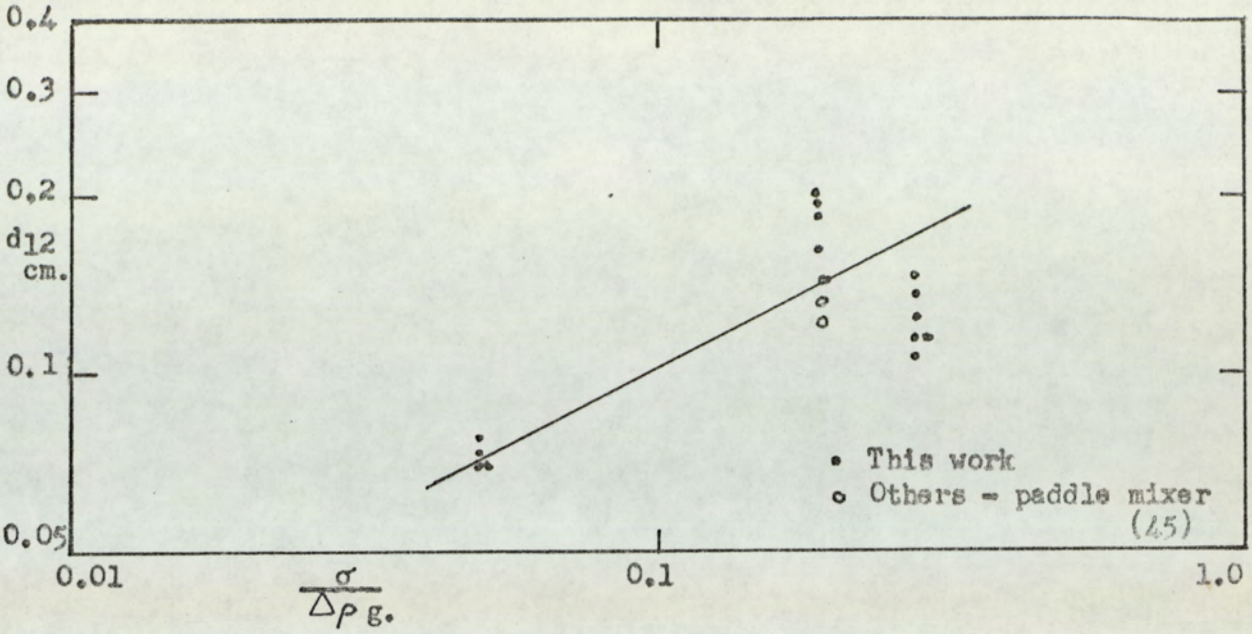


FIGURE 85. CORRELATION OF HYDRAULIC MEAN DROP DIAMETERS - OLDSHUE-RUSHTON COLUMN - LAMINAR REGION.

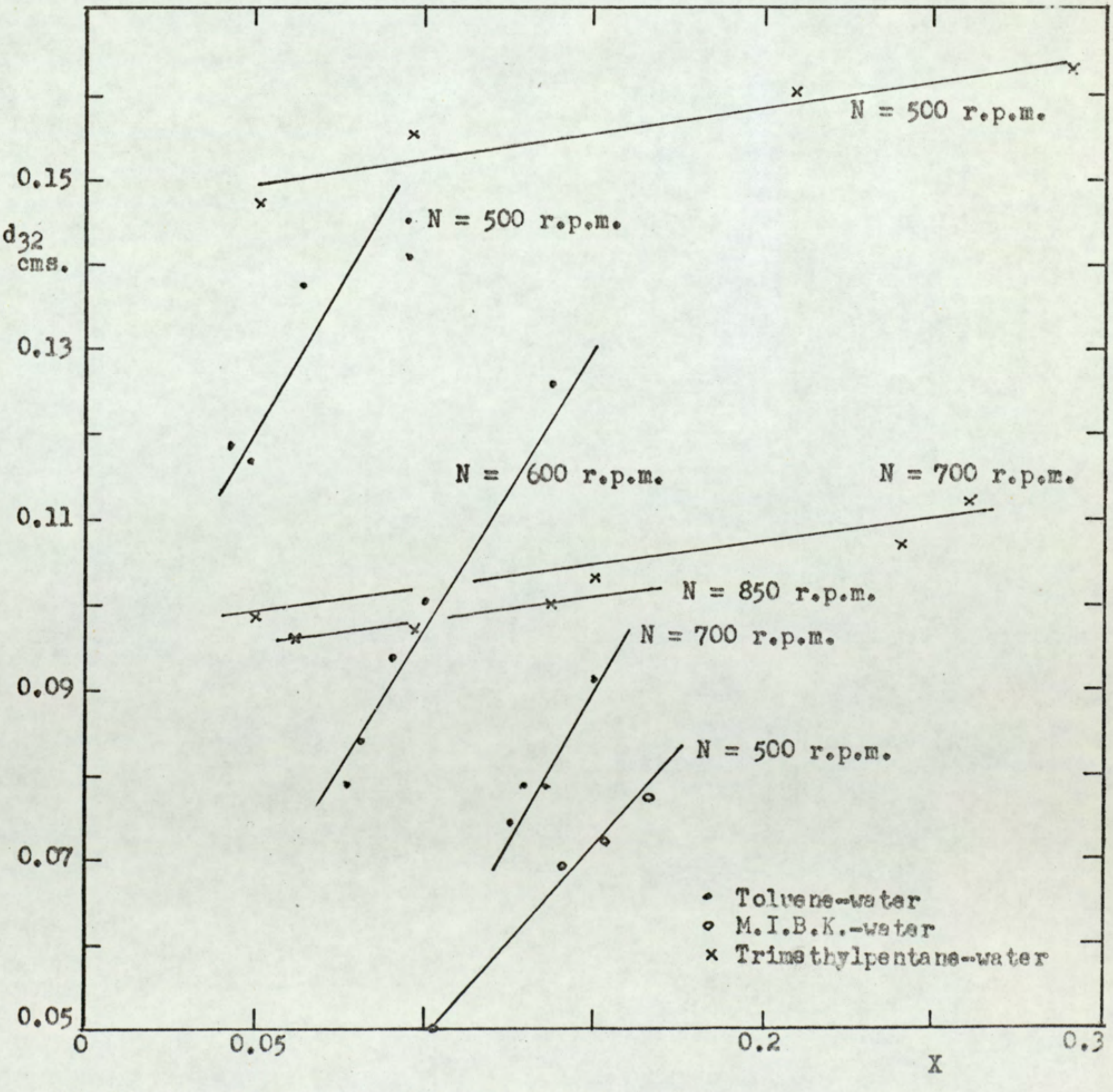


FIGURE 86. OLDSHUE-RUSHTON COLUMN, MEAN DROP SIZE vs HOLD-UP.

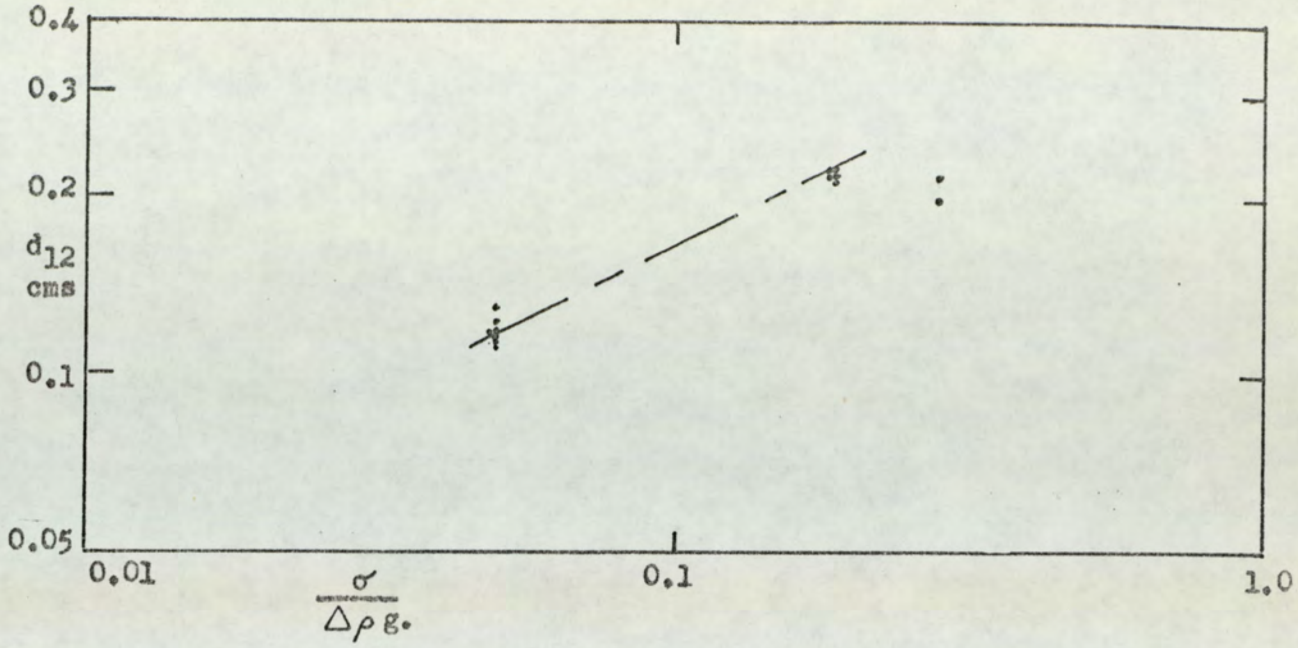


FIGURE 87. HYDRAULIC MEAN DROP DIAMETERS - OLDSHUE-RUSHTON COLUMN : VERY LOW ROTOR SPEED.

FIGURE 88. OLDSHUE-RUSHTON COLUMN - BREAK-UP OF DROPS BY IMPACT ON AGITATOR.

(System : Toluene - water, N = 350 r.p.m.)

Some indication of the conditions under which a coalescence-redispersion mechanism becomes predominant is given by Figure 75. This is only based on visual observations of droplets with curtailed residence times and is therefore of uncertain numerical accuracy. However, it is clear that, as expected, predominance occurs at a certain minimum hold-up. The onset is also dependant on agitator speed.

Because a coalescence - redispersion mechanism usually predominates it would be expected that although at the lower end of the droplet size distribution range single drop data would indicate a molecular transfer process for solute transfer, the rate in practice should be more in accordance with an eddy diffusional mechanism. The recent paper by Mensing et.al. suggests that even a more complex regime may be present (142); they showed that the regime and rate of mass transfer from a single droplet was dependant on its age. In the case under consideration a distribution of both sizes and ages exists.

In practice the circulating mixing in a compartment produces a high average residence time for the dispersed phase droplets and therefore concentration changes between compartments should be approximately stagewise. This facilitates preliminary design (17,113).

12.3 WETTING PHENOMENA

In the Scheibel column with dispersed phase wetted packing there was an increased tendency for drip point formation and jetting. As well as producing large globules of indeterminate size dispersion by the latter mechanism produced small satellite droplets as illustrated in Figure 89. Such small droplets persisted in the column due to their low settling velocities. Under conditions observed following the transition point, that is, when layers of dispersed phase covered the packing adjacent to the agitated zone, the columns mode of operation changed to that of a continuous mixer-settler. Such a design has not formerly been proposed. The capacity of the column section was not determined since phase rejection could not be satisfactorily observed;

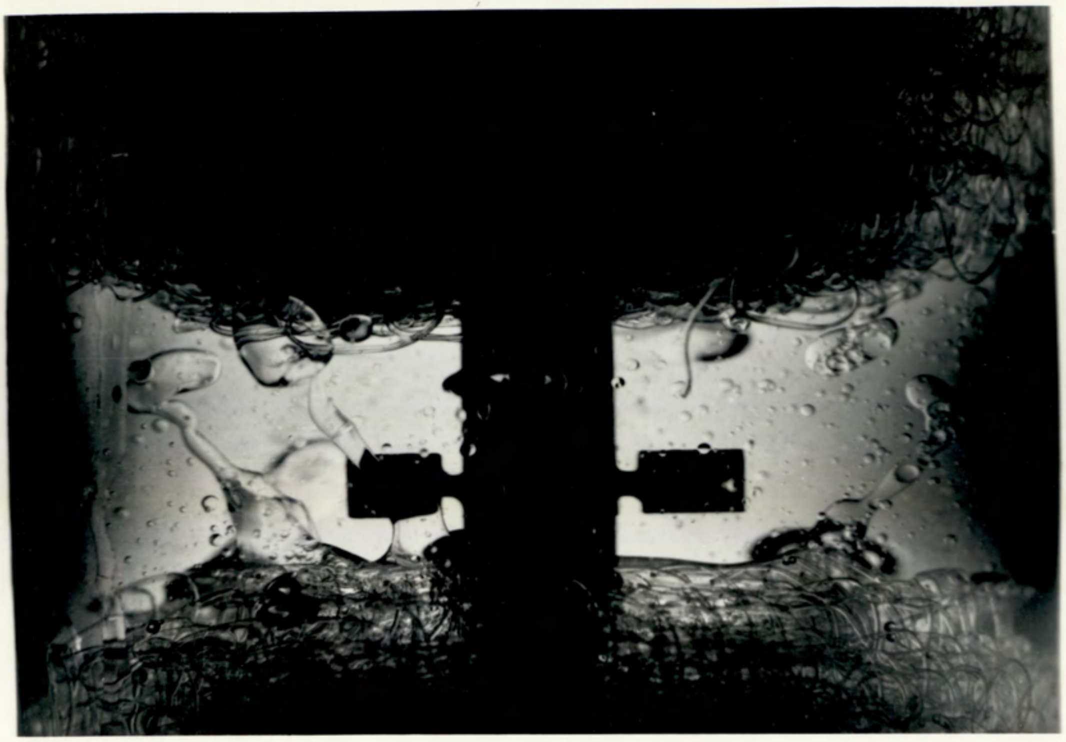


FIGURE 89, BREAK-UP MECHANISM YIELDING SATELLITE DROPS IN SCHEIBEL COLUMN WITH PLASTIC PACKING.

(System : M.I.B.K. - water, N = 350 r.p.m.
V_d = 43.4 ft.sec.$^{-1}$)

capacity would be less than for an equivalent column with steel packing however.

In the R.D.C. with a disc wetted by the dispersed phase, droplets coalesced into a layer beneath the disc and dispersion was by streaming from the periphery. Large globules formed during this process, caused the mean drop size to be larger than with a non-wetted disc under similar conditions. The reduced capacity associated with the wetted disc is shown in Figure 71 ; the maximum reduction observed was approximately 14%. In line with the observation that flooding commenced in the horizontal plane of the disc, some correlation might be expected between the increased constriction factor $\frac{D^2 - (R + 2w)^2}{D^2}$

where w is the element width.

For example, at 850 r.p.m. with a 0.16 cm. element attached C_R = 0.61 c.f. 0.66 with a non-wetted disc. The anticipated decrease in capacity is therefore 8% which was of the order observed.

Analysis of high speed cine film indicates that the mechanism of dispersion is conducive to premature flooding. As illustrated - Appendix 7.2, a high hold-up is produced across the column section in the plane of the disc. The mechanism of dispersion from a wetted disc is considered in para. 12.4.2.

No significant difference in capacities occurred at low dispersed phase flowrates i.e. conditions leading to an incompletely wetted disc.

Large globules were observed in the R.D.C. with dispersed phase wetted stators. Such globules would result in a reduced interfacial area thus providing an explanation for the greatly reduced efficiencies found by Davies under similar conditions (94). A similar consideration applies to the Oldshue-Rushton column. One important conclusion deriving from this is that when an increase in dispersed phase wetting of column stators and walls, together with the rotors as discussed earlier, occurs in practice possibly due to prolonged operation, column efficiency will be reduced.

The mechanism leading to globule formation was analagous to that described by Haynes et.al. in their work on the formation of droplets from a circular orifice at varying contact angles (52). They found that as the contact angle (i.e. the angle between the plate and a tangent to the liquid-liquid surface constructed at the three phase boundary) was increased the dispersed phase tended to spread along the plate surface before growing into sizeable droplets which broke away under the action of gravity. Conversely liquids with small contact angles grew from the orifice without spreading. At large contact angles, droplets were formed from a base of fluid which remained on the plate surface. The magnitude of such wetting effects was found to be large but as in this work the data could not be correlated.

This area of the work was based originally on the concept that wetting of column packing or internals by the dispersed phase might occur through error in materials selection or by displacement during operation. It has recently been suggested however that since contact angle of the dispersed phase is dependant on solute concentration (53) then in the Scheibel column behaviour in the packed sections may vary from top to bottom of the column. Clearly such a consideration should apply equally to the R.D.C. and Oldshue-Rushton columns. The little studied phenomenon of dynamic contact angle is also of relevance.

12.4. DROPLET BREAK-UP MECHANISMS IN A ROTATING DISC CONTACTOR.

The mechanisms of discrete droplet formation from a non-wetted disc in the laminar region and from a dispersed phase wetted disc will now be considered.

12.4.1. Droplet Break-Up below a Critical Re number.

In the operation of the R.D.C. with a non-wetted disc below the critical Reynolds number referred to in para.12.2. the persistance of discrete droplets beneath the disc was independant of flowrate or rotor speed. As discussed in para. 11.3.3. the turbulence existing in the continuous phase was found to be insufficient to cause break-up of drops after leaving the disc. Therefore, conditions near the disc merit consideration.

Following para. 3.2. droplet break-up occurs when the difference in dynamic pressures at different points on the surface of a drop, which arise due to velocity variations within the continuous phase, exceeds the smallest value of surface force at any point.

From Kolmogoroff the difference in dynamic pressure exerted on opposite sides of a drop of radius r is (40)

$$\triangle P_1 = \frac{K_f . \rho_c \ (V_1^2 - V_2^2)}{2} \qquad 93.$$

where K_f is a coefficient and V_1 , V_2 are the velocities of the turbulent fluid at two points separated by a distance d = 2r. Large scale eddies that do not vary over the drops diameter will not produce deformation and break-up. Therefore, only relatively small eddies should be considered.

The change in eddy velocity over a length of d = 2r is,

$$\triangle V_{2r} = (2r\,E)^{\frac{1}{3}} \qquad 94.$$

where $E = \frac{\varepsilon}{\rho}$ the energy dissipated per unit mass of liquid.

From equations 93 and 94 therefore,

$$\Delta P_1 = K_f \rho_c \left(\frac{2r\mathcal{E}}{\rho}\right)^{\frac{2}{3}} \qquad 95.$$

This is balanced by the force due to interfacial tension,

$$P_2 = \frac{\sigma}{d} = \frac{2\sigma}{r}$$

Equating P_1 and P_2

$$K_f \rho_c \left(\frac{2r\mathcal{E}}{\rho}\right)^{\frac{2}{3}} = \frac{2\sigma}{r} \qquad 96.$$

But

$$= \frac{\rho.(\Delta V)^3}{L} \qquad 97.$$

where L is a characteristic dimension of the turbulence. Hence the radius of a stable drop is (40,64),

$$r_{s.d.} = \sqrt{2}\left(\frac{\sigma}{K_f \rho}\right)^{\frac{3}{5}} \frac{L^{\frac{2}{5}}}{V^{\frac{6}{5}}} \qquad 98.$$

which is a form of equation 6.

For a drop of diameter d in the region beneath the disc the difference in velocity across the drops, if axial velocity effects are neglected, results from the difference in the horizontal planes z and z + d. Radial velocity at the disc, i.e. in plane z = 0, is zero and its variation in any other horizontal plane z from radius R' to R' + d is small and will therefore be neglected. Further drops will be assumed spherical; in practice they were slightly elliptical with a ratio of axes up to 1.3 to 1.

The exact solution of the Navier-Stokes Equation for the flow around a flat disc rotating about an axis perpendicular to its plane in a fluid at rest has been evaluated by Cochran (142). The fluid layer immediately adjacent to the disc is carried with it through friction and is ejected due to centrifugal force. This fluid is replaced by further fluid flowing axially towards the disc. The three dimensional flow field is shown in Figure 90. A velocity component w exists in the axial direction z, u in the radial direction r, and v in the circumferential direction ϕ when the uniform angular velocity of the disc is ω.

The Navier - Stokes Equations are,

$$u.\frac{\partial u}{\partial r} - \frac{v^2}{r} + w.\frac{\partial u}{\partial z} = -\frac{1}{\rho}.\frac{\partial p}{\partial r} + v\left[\frac{\partial^2 u}{\partial r^2} + \frac{\partial}{\partial r}\left(\frac{u}{r}\right) + \frac{\partial^2 u}{\partial z^2}\right]$$

$$u.\frac{\partial v}{\partial r} + \frac{uv}{r} + w.\frac{\partial v}{\partial z} = v\left[\frac{\partial^2 v}{\partial r^2} + \frac{\partial}{\partial r}\left(\frac{v}{r}\right) + \frac{\partial^2 v}{\partial^2 z}\right]$$

$$u.\frac{\partial w}{\partial r} + w.\frac{\partial w}{\partial z} = -\frac{1}{\rho}.\frac{\partial p}{\partial z} + v\left[\frac{\partial^2 w}{\partial r^2} + \frac{1}{r}\frac{\partial w}{\partial r} + \frac{\partial^2 w}{\partial z^2}\right]$$

$$\frac{\partial u}{\partial r} + \frac{u}{r} + \frac{\partial w}{\partial z} = 0 \qquad 99.$$

A dimensionless distance from the wall is introduced,

$$\zeta = \frac{z}{\delta}$$

where δ is the thickness of the layer of fluid carried by the disc. The velocity components are expressed as,

$$u = r\omega F\ (\zeta)$$

$$v = r\omega G\ (\zeta)$$

$$\text{and} \quad w = \sqrt{r\omega}H\ (\zeta) \qquad 100.$$

and the functions F, G and H calculated by a method of numerical

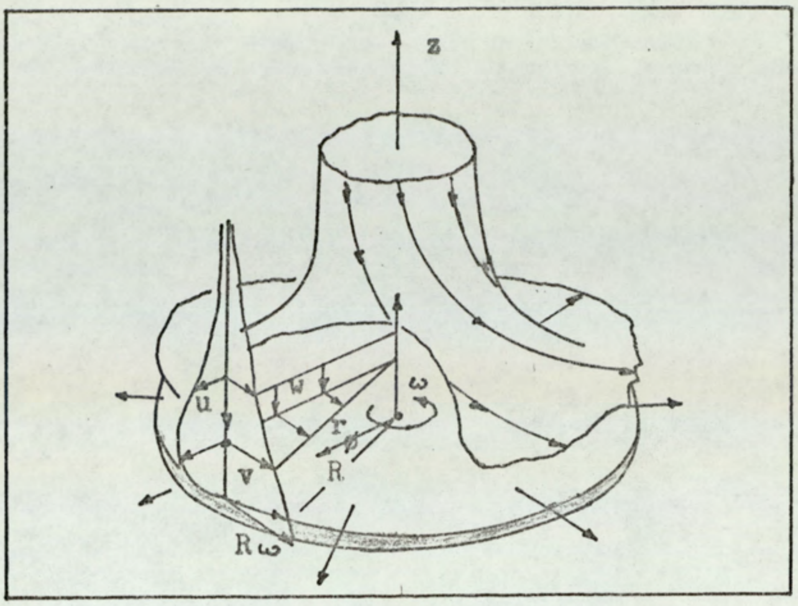


FIGURE 90. FLOW IN THE NEIGHBOURHOOD OF A ROTATING DISC.(after 142, 159) Bulk fluid at rest. A layer is carried by the disc owing to viscous forces Centrifugal forces in the thin layer cause secondary flow directed radially outward.

(Components of velocity are w - axial, u - radial and v - circumferential).

integration (142). These are reproduced in Figure 91, in Appendix 8. It should be noted that the axial velocity w is small, of the order of $\sqrt{\nu\omega}$ and can therefore be neglected as mentioned earlier.

By means of equation 100 and Figure 91, the value of the resultant velocity difference across a drop, at any location beneath the disc, can be found and hence by substitution in equation 98, the stable drop size. If the diameter of the drop d exceeds the stable drop diameter d_{sd} then provided the drop is present in the flow for sufficient time deformation and break-up will occur.

Application of the above theory is complicated since the resultant velocities on either side of a drop will, of course, not be in identical directions. However, in the material range the resultants are largely tangential so that this can be neglected. The theory would be best tested for $d \ll R$ since the derivation is for a disc of infinite extent. However, as an approximation drops for which $d = 0.2$, 0.3 and 0.4 cms. have been considered travelling in vertical planes at distance $r = R'$, the effective radius from the disc centre,

(i) arriving at the disc underside

(ii) for convenience, when they are still one drop diameter away from the disc.

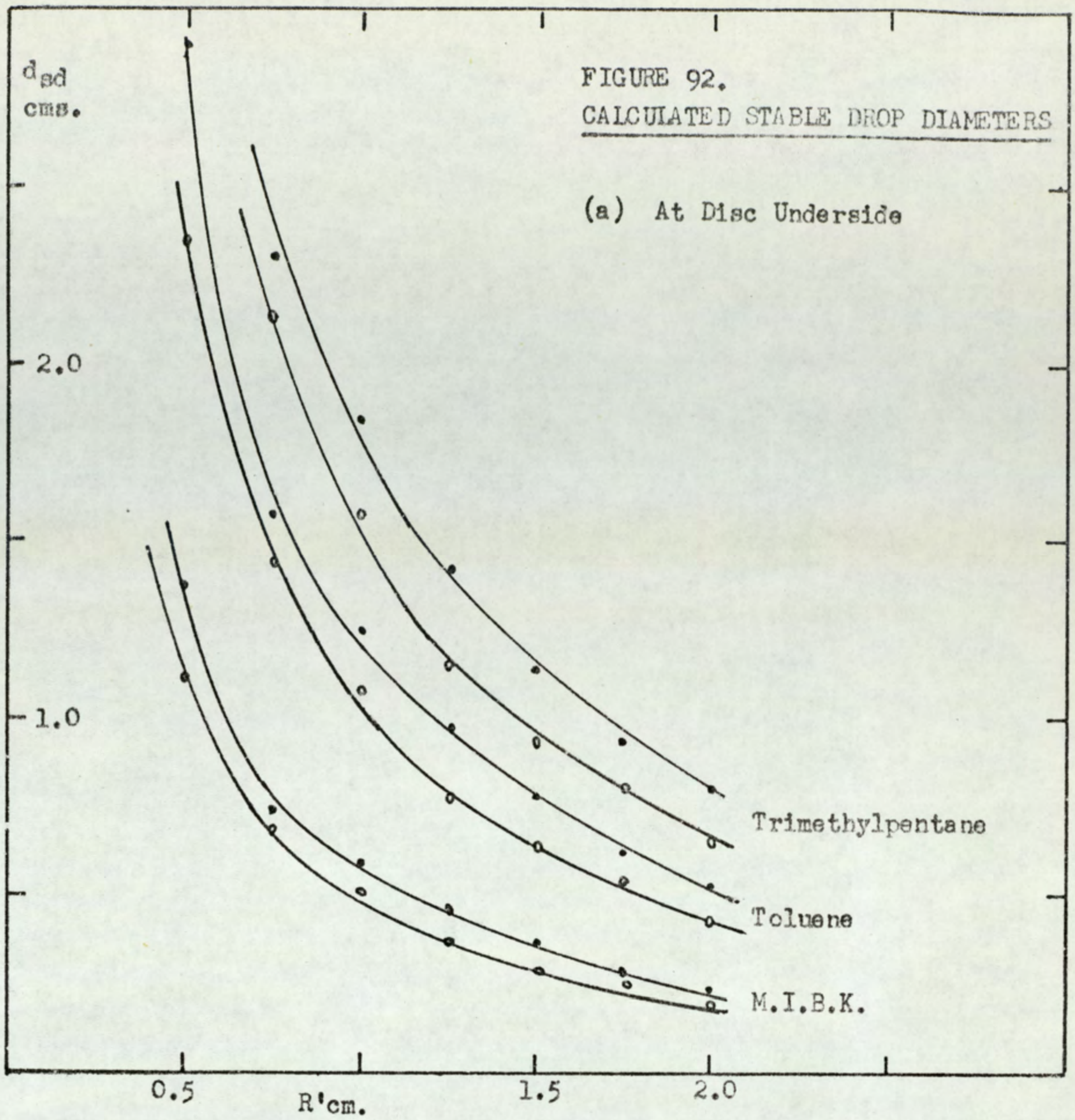
This necessitated the calculation of resultant velocities in three horizontal planes only, viz. $z = 0$, $\zeta = 0$; $z = d$, $\zeta = 1.0$ and $z = 2d$, $\zeta = 2.0$. Calculations were performed for two rotor speeds, 500 r.p.m. and 600 r.p.m. for which the presence of drops beneath the disc was well established. Calculated radial and tangential velocities are given in Table 11 and resultant differences in velocities across drops in Table 12 in Appendix 8. Stable drop diameters were calculated from equation 98, using the physical properties given in Appendix 4, and a value for K_f of 0.5 (64). These are given in Table 13 in Appendix 8, and are plotted in Figure 92.

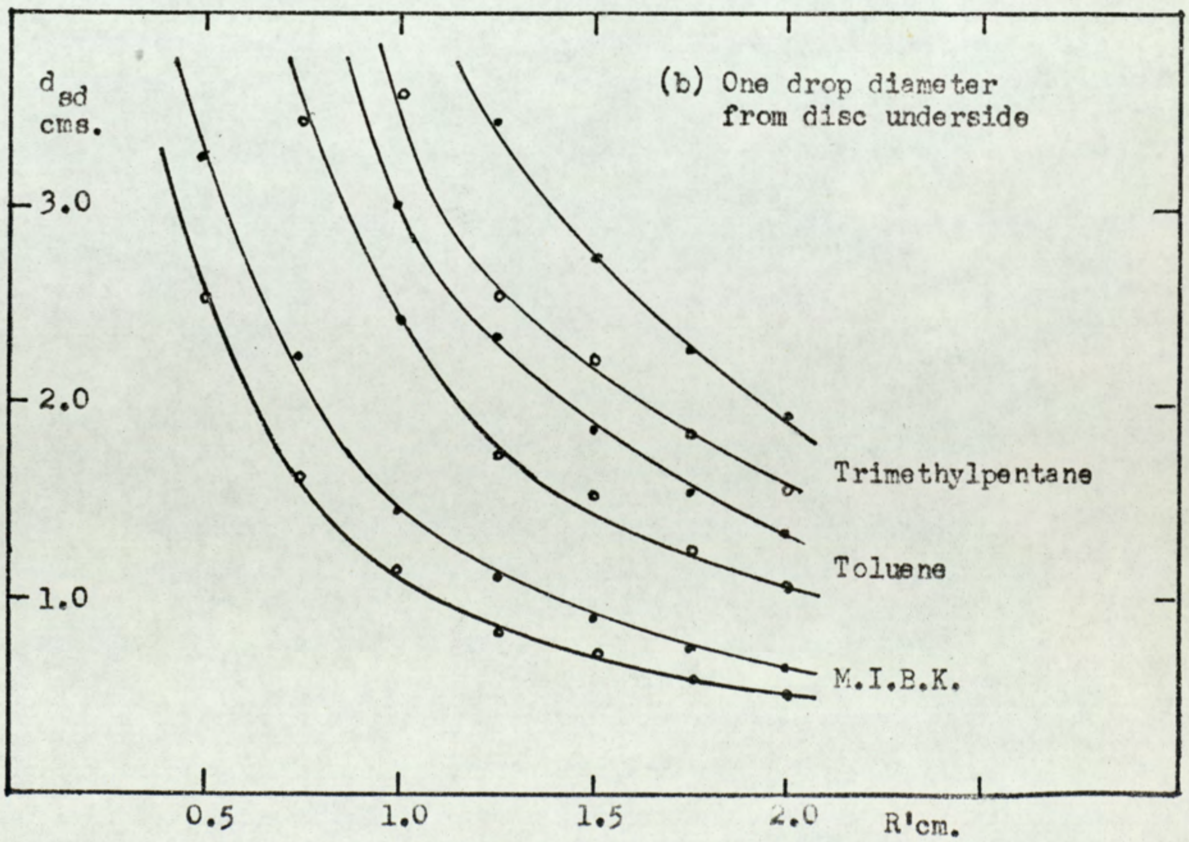
Reference to Figure 92, shows that for drops of 0.2 to 0.4 cm. equivalent diameter at the disc, break-up will occur by the mechanism proposed only in the case of M.I.B.K. in water for which the stable drop-size at $R' > 1.25$ cm. is less than the initial drop diameter. Figures 93 and 94 show trimethylpentane and toluene droplets leaving the disc without break-up.

Droplet break-up at > one diameter from the disc underside is remote. Equipment limitations did not allow droplets much greater than the calculated stable drop size to be generated in this column. Further work is proposed in para. 14.1. Clearly in larger columns, allowing increased values of R', droplets will spend a certain time in an 'unstable region' before being ejected. For a given velocity V of a homogeneous isotropic flow, drops formed in it must be of a single size. For a small disc the variation of V in the unstable region will be relatively small and hence a small spread of drop sizes should exist. Conversely for a large diameter disc a spectrum of stable drop sizes can be produced. Therefore droplet size distribution will be dependant to some extent on disc diameter. Nevertheless the proposed mechanism satisfactorily explains the smaller spread in drop sizes observed for disc speeds below $Re \doteq 1.75 \times 10^4$. Small drops may be formed by impact with the disc tip.

The reason why no interdroplet coalescence occurs beneath the disc, even at very low rotor speeds, is clear from the velocity profiles. Adjacent droplets in any radial plane undergo separation due to the difference in their radial velocities; adjacent droplets at the same radius have exactly equal velocities and hence there is no force promoting film drainage.

FIGURE 92.
CALCULATED STABLE DROP DIAMETERS


(a) At Disc Underside


(b) One drop diameter from disc underside

Note: • 500 r.p.m. o 600 r.p.m.

FIGURE 93. R.D.C. SECTION - INTACT DROPLETS LEAVING DISC PERIPHERY
(System: Trimethylpentane - water, N = 500 r.p.m.
V_d = 13.8 ft.hr.$^{-1}$)

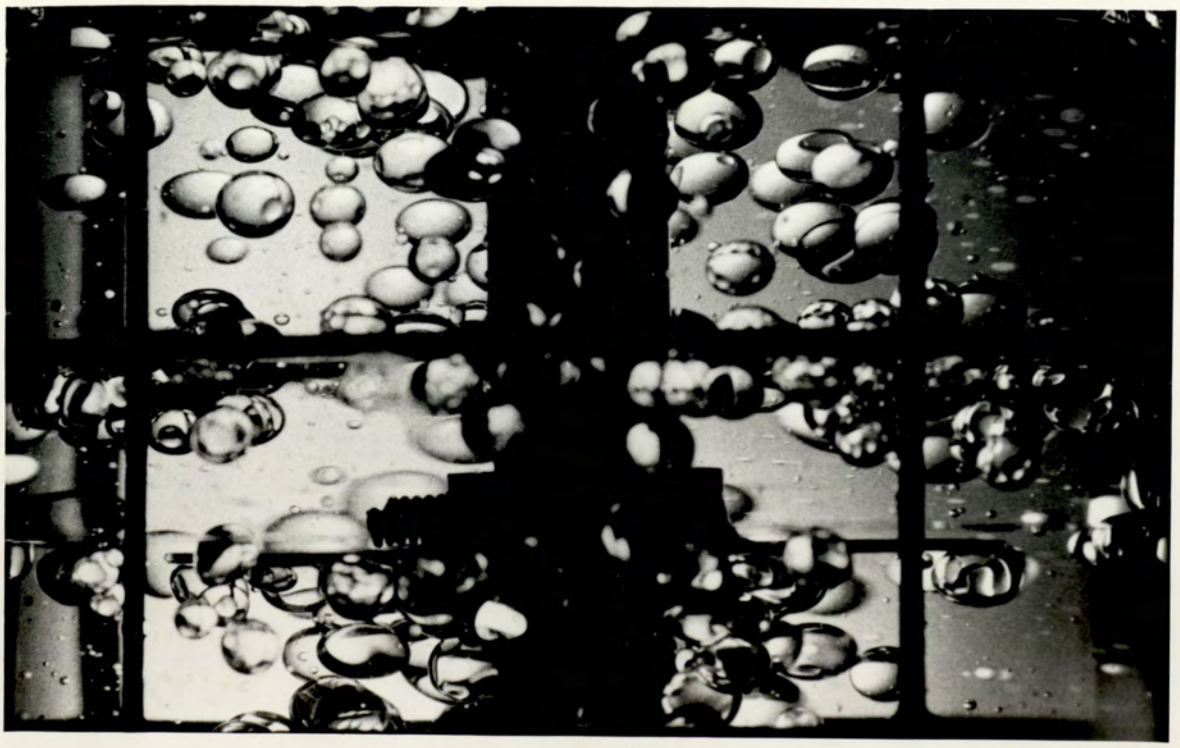


FIGURE 94. R.D.C. SECTION - INTACT DROPLETS LEAVING DISC PERIPHERY
(System: Toluene-water, N = 500 r.p.m.)

12.4.2. Droplet Formation from a Dispersed Phase Wetted Disc.

There is some similarity between drop formation from a disc wetted by the dispersed phase and atomisation from centrifugal discs such as those used in spray driers, humidifying equipment and oil burners (143). In the latter, liquid is discharged as a flat sheet and the manner in which this disintegrates into drops is dependant on the operating conditions. Three regimes have been identified (144),

(i) Rim disintegration. At very low ejection velocities the rims of the liquid sheet contract continually due to surface tension effects. Threads of liquid are pulled out during contraction and produce large droplets on break-up.

(ii) Perforated sheet disintegration. Over a very limited range of conditions the free edge disintegrates through a network of threads due to expanding perforations in an undisturbed sheet.

(iii) Wavy sheet disintegration. This is the most common cause of break-up. Waves are produced on the sheet due to velocity differences between the two phases. These waves grow in amplitude until, at some critical value, the sheet disintegrates into unstable cylindrical ligaments. Drops form by subsequent break-up of the ligaments. Regime iii. is illustrated in Figure 95.

Considering this regime only, the break-up of ligaments, which are essentially jets with a predominantly longitudinal velocity, may occur in any of three ways dependant on ejection velocity. At low ejection velocities the jet is in laminar flow and varicose break-up occurs. In this the jet swells and contracts symmetrically. This varicosity grows in amplitude until it is greater than the jet radius and the jet disintegrates. The phenomena of sinuous break-up (146) or complete

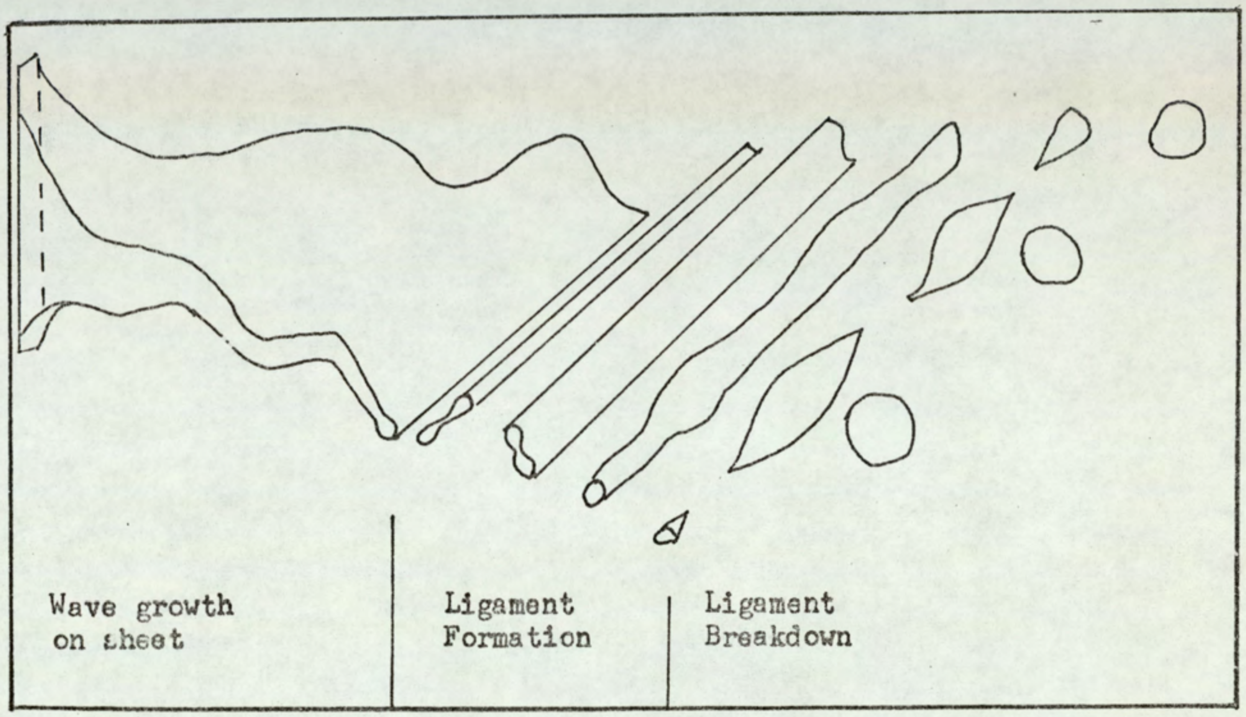


FIGURE 95. WAVY SHEET DISINTERGRATION AND LIGAMENT BREAKDOWN (145).

atomisation, which occur at high velocities are not relevant to this analysis.

In the present case, in the vicinity of the disc, motion of the continuous phase in the axial direction is generally of a much lower order than that of the dispersed phase in the plane of the disc. Therefore, as was done earlier, sheet disintegration may be assumed to occur in a stationary continuous phase. Hence break-up is a special case of that of a liquid sheet having finite viscosity and which decreases in thickness as it moves away from the source. Dombrowski and Johns have studied the growth of waves on a liquid sheet and extended this to consider the size of ligaments produced by wave disintegration of such a sheet attenuating according to $h = F\,\theta^{-1}$ in a stationary gas (145). By a mass balance, the ligament diameter

$$d_L = \left(\frac{4h}{n''} \right)^{\frac{1}{2}} \qquad 101.$$

In order to evaluate this, h the sheet thickness at break-down and n'', the wave number, were expressed in terms of f, the total growth of the wave. The derivation is complex and recourse to empiricisms based on experimental observations is necessary to yield the equation (145),

$$d_L = 0.9614 \left[\frac{B^2 \sigma^2}{\rho_c \rho_D U^4} \right]^{\frac{1}{6}} \left[1 + 2.6\, \mu \sqrt[3]{\frac{B \rho^4 U^7}{72 \rho_D^{\,2} \sigma^5}} \right]^{\frac{1}{5}} \qquad 102.$$

As described above ligaments break down through symmetrical waves. The properties of these waves where surface tension forces predominate has been analysed by Weber (147). Assuming that since the ligaments move transversely through the continuous phase the latter has no effect on the wavelength, the relevant equation is,

$$n'' d_L = \left[\frac{1}{2} + \frac{3\mu}{2(\rho_D \sigma \; d_L)^{\frac{1}{2}}} \right]^{\frac{1}{2}} \qquad 103.$$

The waves grow until they have an amplitude equal to the ligament radius and therefore one drop is produced per wavelength. Thus, by mass balance, the relation between drop size and wave number is (145)

$$d^3 = \frac{2\pi d_L^2}{n''} \qquad 104.$$

Combining equations 103 and 104,

$$d = \left[\frac{3\pi}{\sqrt{2}}\right]^{\frac{1}{3}} d_L \left[1 + \frac{3\mu}{(\rho_D \sigma d_L)^{\frac{1}{2}}}\right]^{\frac{1}{6}} \qquad 105.$$

This equation provides a means of predicting drop sizes using values of d_L calculated from equation 102.

Insufficient results could be obtained to check the validity of the above theory owing to the dispersed phase hold-up associated with increased rotor speeds. This made it impracticable to differentiate between drops formed by ligament break down and those subsequently broken down by dynamic pressure fluctuations in the bulk continuous phase. Reference is made in para. 14.1. to further work, but preliminary conclusions may be drawn from observations given in para. 11.4. For toluene in water at 500 r.p.m. both x and $h \doteq 0.32$ cm. Assuming the velocity is of similar magnitude to that at the disc tip ($\zeta = 1$), Table 11 gives $U \doteq 18.9$ cm.s^{-1}. Substitution of the relevant physical properties in Equation 102 gives $d_L \doteq 0.21$ cm. and hence from Equation 105, $d = 0.43$ cm. Drops larger than this were photographed together with numerous small droplets. The mechanism at this speed is probably that of rim disintegration.
At 800 r.p.m., x and $h \doteq 0.1$ cm. and $U = 30.3$ cm.s^{-1}. Under these conditions $d_L \doteq 0.07$ cm. and $d \doteq 0.13$ cm. Recorded drop sizes were of this order of magnitude but smaller drops were also present.

Assuming wavy sheet disintegration this is to be expected. Ligaments do not break down instantaneously but stretch and thin out due to their transverse velocity component (145) and in this example the tangential velocity is > 2.5 U. Drop size is directly related to ligament diameter, hence smaller drops will be produced than predicted by Equation 105. Droplet formation also differs from the model due to the constraints imposed by the column wall and stators (148).

In preparation for further work (149) break-up from a p.t.f.e. disc was recorded by cine photography at 500 f.p.s. using carbon tetrachloride as a descending dispersed phase in water - Appendix 7.2. Repetition of the above calculation for $N = 800$ r.p.m. gave a calculated $d_L \triangleq 0.07$ cm. and $d \triangleq 0.135$ cm. Most drops produced were in fact of the order of $\leqslant 0.1$ cm. but some as large as 0.16 cm. were also present.

13. CONCLUSIONS.

A wide disparity exists between the quantity and quality of research on extraction in agitated columns and the criteria still employed in many cases for industrial design and operation. For example, one commercial column known to have functioned below design efficiency did so because the extraction produced a reduction in raffinate flow of approximately 25%, and hence a significant difference in phase ratio between the top and bottom, and the effect of this was overlooked. Examples have been given of columns being inadvertantly, yet satisfactorily, operated at zero rotor speed. Modified designs have in most cases been introduced to circumvent patents rather than because they offer advantages per se.

It is hoped that this thesis will make some contribution to rational design. No one column design is best for all applications and claims based on tests with one system are of limited value. Concentration of research on the few established designs, as has been done here, would be of more value. Numerous recommendations for further work in this and other areas of liquid extraction are discussed in paras. 14.1. and 14.2.

The main conclusions to be drawn from this work may be summarised as follows:-

a. For systems having an interfacial tension > 9.7 dynes.cm^{-1} it is desirable to omit the packed sections from the Scheibel column. Within the normal operating range a coalescence-redispersion mechanism predominates in this column but due to globule formation the mean interfacial area in the agitated zone is unpredictable. Therefore a stagewise design procedure is necessary (70). The problem of estimating volumetric capacity remains (134).

b. In the absence of special interfacial effects associated with mass transfer, the R.D.C. is a discrete droplet contacting device and can be designed on this basis. The drop-size produced at $Re < 1.75 \times 10^4$ with an aqueous continuous phase wetted disc is independant of hold-up and may be predicted for practical purposes by the empirical correlation Equation 20, with the numerical constant having a value of 0.55. A mathematical model has been proposed in para. 12.4.1. for drop size prediction from fundamentals. Further work is needed before drop sizes in the transition range can be predicted with confidence by Equation 19, since hold-up has some effect.

c. Under normal operating conditions a coalescence-redispersion mechanism predominates in the Oldshue-Rushton column. Solute transfer rates should therefore be in accordance with those for fully circulating droplets. Drop-size data is represented by forms of Equation 53. For speeds below $Re \triangleq 4 \times 10^3$ conditions are acollisive and mean drop sizes can be predicted by Equation 20, with the numerical constant having a value of 0.34 or at very low speeds 0.57.

d. With dispersed phase wetted internals, such as may obtain after prolonged operation, no reliable estimate can be made of efficiency or capacity. This is one factor dictating the provision of variable speed agitators on commercial columns. A model for disc atomisers in para. 12.4.2. shows promise for calculations with wetted discs in the R.D.C.

Two 16 mm. cine films were compiled to record some of the phenomena observed. The first film was presented with the paper forming Appendix 1.3 (7). Both films, the contents of which are described in Appendix 7., have been deposited in the Chemical Engineering Department, University of Aston in Birmingham.

14. RECOMMENDATIONS FOR FURTHER WORK.

14.1. As an Extension to This Study.

It is recommended that this work be continued in the following ways:-

1. Clearly a limit exists for general purpose liquid extraction beyond which it becomes uneconomic to introduce complex internals into agitated column extractors in order to achieve decreased H.E.T.S. values. This will be partly clarified by research to be undertaken into the performance of a pilot scale Wirz column (21,150).

2. The results of drop size distributions in this work appear both consistent and in general agreement with other published work. However it is a possible limitation of the measurements on the Oldshue-Rushton column, which has been shown to involve a coalescence-redispersion mechanism and for which no other data are available for comparison, that an equilibrium drop size may not have been attained. Initial break-up was by a disperser and photographs were of the second real stage. Oldshue and Rushton reported a decrease in drop size from bottom to top of their multistage column but this was in the presence of solute transfer, in particular transfer from the dispersed to the continuous phase, and was based on visual observation only. (17). In the event, it is proposed to examine the variation of drop-size distribution with column height in a laboratory multi-stage Oldshue-Rushton column of standard dimensions both in the presence and absence of mass transfer. This work will necessitate the use of the S.P.R.I. Particle Size Analyser Type II (136) shown in use in Figure 96. Alternatively, where the requisite degree of magnification is economic, the Zeiss Analyser TG Z 3 (137) will be employed. This is shown in use in Figure 97. Measurements will also be made of column efficiency on

FIGURE 96. S.P.R.I. PARTICLE SIZE ANALYSER TYPE II. (136)
in use for the determination of drop size distribution.

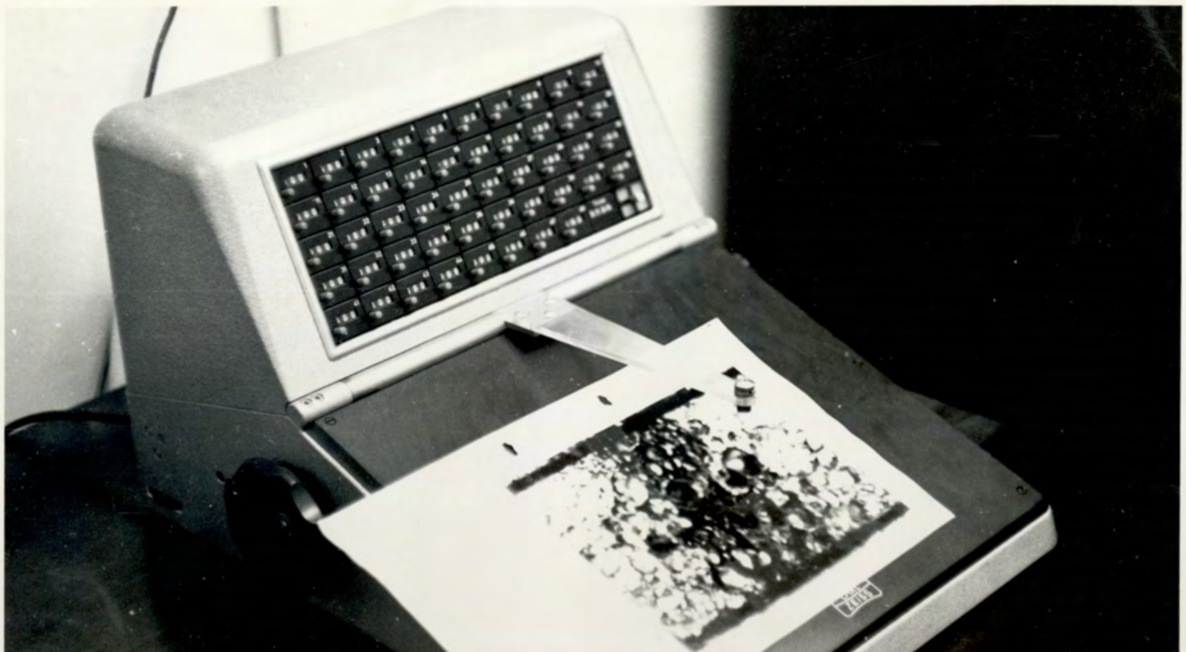


FIGURE 97. ZEISS ANALYSER T.G.2.3. (137)
in use for the determination of drop size distribution.

simple extractions with determinable mean drop sizes (151). In view of the irrefutable evidence of a coalescence-redispersion mechanism in this contactor it is inconceivable that the transfer process is representable by a stagnant drop model as suggested by Bibaud and Treybal (93) and this will be clarified. The correlation of m in Equation 53, with operating parameters will also be undertaken. However it is clear from the work described that a variation in microturbulence exists in any compartment of the Oldshue-Rushton column; this results in greater break-up and coalescence near the turbine than in the bulk continuous phase. Therefore, this correlation, or any other correlation based on agitator Reynolds number, will be specific to the particular geometry studied.

The potential of the column as a heterogeneous reactor, which is referred to in para. 14.2.4., will also be examined.

3. The behaviour of an R.D.C. in which the disc is wetted by the dispersed phase may have practical significance (152). For those systems, admittedly few in number, in which the dispersed phase mass transfer coefficient is controlling, the creation of new surface in the layer of this phase wetting the underside of the disc would be expected to make a significant contribution to mass transfer. Work has been put in hand to determine the magnitude of this effect. (148). It is proposed to study separately the mechanism for droplet break-up from a wetted disc proposed in para. 12.4.2 by means of a single disc in a stationary continuous phase (149).

4. Work is in hand to obtain experimental data regarding stable drop sizes and droplet break-up for a rotating disc below the critical Re number. These will be compared with those predicted in para.12.4.1. (149). In common with all studies of droplet break-up phenomena the main difficulty is to differentiate between the distribution of drops

generated by the disc and that observed in a compartment at any time due to the persistance of small drops with low settling velocities.

14.2. Other Studies.

The following useful areas of work have become apparent from this study,

1. Certain extractors having considerable practical potential have not been examined in terms of droplet hydrodynamics nor indeed have their efficiencies and limitations been established. Such work could yield results of immediate practical value. Equipment has therefore been designed and studies initiated (153) on a Morris Contactor, a novel horizontal extractor in which droplets persist throughout (154).

2. Under certain conditions, e.g. small mean drop diameters and low interfacial tension systems, a limiting factor in extraction column operation is the efficiency of phase separation at the phase boundary. This may result from either predominantly interdroplet coalescence or coalescence of drops at a plane interface. Apart from work with single drops (55) the parameters determining which mechanism prevails, and hence the time needed for separation, have not been investigated. The volume needed for phase separation represents additional height in any column design or alternatively is an indication of the size of any external vertical gravity settler. Work has been initiated in this area (127).

3. Observation of the Scheibel column illustrated the extremely complex phenomena associated with the flow of mixtures of immiscible liquids in small interstices size packings. A comprehensive study of the flow of droplets through woven mesh packings would be of value and could assist in the prediction of coalescer performance. Some results have been obtained from a recent study (67) and other studies

are in progress (101,134).

4. Agitated column contactors have considerable potential as reactors. This is not limited to liquid phase systems and indeed reactors based on the Oldshue-Rushton design are claimed to be in operation (155) but no data are available. The Oldshue-Rushton column is characterized by high hold-ups so that it should be suitable for slow liquid-phase reactions, that is, when a finite time is required to reach equilibrium. Westerterp et.al.(160) have suggested the use of the R.D.C. for homogeneous liquid phase reactions in which residence time could be varied. A study of the potential of agitated columns as continuous reactors would be of practical value. The work could also be extended to consider the different basic mechanisms. For example, with any system in which reaction occurs in the dispersed phase, in discrete droplets in an R.D.C. the mechanism is analagous to a multitude of batch reactors. In an Oldshue-Rushton column with a coalescence-redispersion regime the mechanism is analagous to a continuous reactor (71).

5. Numerous independant investigations have been performed on the various column contactors. Similar studies have not been made of the newer centrifugal extractors which are finding increased industrial application (1). Therefore the proposal, originally made by Thornton (156), for a comparison and experimental investigation into this range of equipment would be of practical value. However, it would probably be uneconomic to obtain a complete range of equipment.

6. Droplet phenomena, and in particular phase separation, are affected by the presence of solids. Therefore a short study was initiated to determine the effect of particulate solid on coalescence times. Coarse solids were found to assist phase separation (157).

There would appear to be some potential here for a new approach to coalescer design.

7. In the past insufficient attention has been given to finding the most economic design for liquid-liquid extraction processes. A critical investigation has been initiated into which optimisation policies are of use (158) and this may be extended to study real processes.

APPENDIX 1.

SUPPORTING PUBLICATIONS.

1. 'Advances in Equipment for Liquid-liquid Extraction'.
Reprinted from Brit. Chem. Eng.13, No.7 (1968), 981.

2. 'Drop Phenomena affecting Agitated Liquid-liquid Contactors'.
Reprinted from Birm. Univ. Chem. Eng.19, 3 (1968), 87.

3. 'Droplet Characteristics in Agitated Extraction Columns'.
Presented at 3rd International Congress on Chemical Engineering, CHISA, Marianske Lazne, Czechoslovakia, September, 1969.

In recent years several new designs of extractor have appeared and offer scope for a wider application of extraction techniques. Here the author examines the later designs and reviews their actual and potential fields of application — among the latter the conducting of chemical reactions. Mixer-settlers, rotary agitated columns, centrifugal extractors and design aspects are among the subjects treated below.

by C. J. MUMFORD*

Advances in equipment for liquid-liquid extraction

HE field of application of liquid-liquid extraction has been eatly increased by advances in equipment design. Until the mid-940's industrial application was generally limited to those pro-sses, such as neutralising and treating, which require only a few ages. Since then many extractor designs with improved efficiencies ve been introduced. These have made commercially viable extrac-on processes requiring many more stages.

Traditional equipment has been reviewed elsewhere (32, 61). A view of the literature concerning equipment developments and iscellaneous mass transfer studies for 1964 and 1965 has been ven by HARRIS, VASHIST, and BECKMANN (12) and for the period eceding this by DE NIE (6) and TREYBAL (62). In this article the aracteristics, applications and developments in the design of the rious newer types of extractor are considered.

quipment classification

iquid-liquid extraction operations may be performed in either:

- (*a*) Equipment in which the liquids are mixed, extracted and separated in discrete stages. This class includes the mixer-settler range of equipment and also the different plate columns.
- (*b*) Equipment in which continuous countercurrent contact is established between the immiscible phases to give the equivalent of any desired number of stages. These may be categorised as follows:
  - A. Gravity operated extractors
    - (1) Non-mechanical dispersion
      - (*a*) Spray columns
      - (*b*) Baffle plate columns
      - (*c*) Packed columns
    - (2) Mechanically agitated extractors
      - (*a*) Pulsed columns
      - (*b*) Rotary agitated columns
  - B. Centrifugal extractors.

election of equipment

he choice between the various types of extractor for any particular plication is based largely on experience. Continuous contactors e in general preferable to mixer-settlers when large throughputs e to be handled since they offer economies in agitation and power uipment cost, floor space and solvent inventory. They operate ith relatively small amounts of hold-up of raffinate and extract hich is especially important when processing radioactive, flam-able or low stability materials. A prime advantage is their flexi-lity of operation which enables extraction to be performed with stems likely to form fine dispersions. In extraction processes it is cessary as a final step, or in multiple contact stagewise equipment intermediate steps, to separate the two phases. Rapid coalescence desirable otherwise an excessive residence time is required. iling this some of the continuous phase will be removed with the ulk' dispersed phase resulting in reduced efficiency, capacity and loss of solvent. This means that the contactor which gives the most rapid solute transfer is not necessarily in practice the most economic. The sporadic formation of fine dispersions is a major problem in industrial liquid extraction processes (14); providing a stable emulsion is not formed, the problem can be alleviated by controlling the degree of dispersion (42, 53).

Continuous columns without mechanical agitation are unsuitable for use with systems of high interfacial tension since adequate dispersion cannot be achieved throughout the continuous phase. This results in large H.E.T.S. or H.T.U. values. This type of column however is simple and inexpensive and is widely used for low interfacial tension systems. When continuous columns are shut down the liquids settle out; on start-up there is thus a lag before steady state conditions and on-specification streams are produced. Mixer-settlers may therefore be preferable for intermittent operation; this depends on the economics of the process.

Mixer-settler equipment is commonly used where only a few contact stages are required although, in theory, any number of stages may be coupled together in a cascade system. For applications requiring up to five or six stages a mixer-settler cascade is generally the most reliable and economic choice. The major advantages in operation are flexibility with regard to both throughput and phase ratio. Phase ratios of 10:1 can be handled in most designs and phase ratios up to 100:1 may be accommodated by recycling the phase with the lower throughput to aid dispersion. Mixer-settlers are able to handle mixtures of liquids containing suspended solids better than most other contactors with the exception of certain rotary agitated types (19). They are the most reliable type to design from laboratory data since a conservative estimate can be made of stage efficiency; the provision of ample volume for phase separation is invariably the governing factor.

Centrifugal extractors have relatively high capital and operating costs and the number of stages which can be accommodated in a single unit is limited. Nevertheless they are superior to all other contactors for processes requiring a low hold-up or low contact time or where there is a low density difference between the phases.

Table I is a useful rule-of-thumb method for a preliminary narrowing of the choice between the various types of extractor (57). The ratings are Unsuitable – 0, Poor – 1, Fair – 2, Adequate – 3, Good – 4, and Outstanding – 5. Not all of the features in the table can be equated and, as will be discussed, special process factors often govern extractor selection. Equipment installed and operating costs are of primary importance. On this basis and dependent on the number of stages for a given application and the ease of phase dispersion/separation an extractor selection chart can be drawn for any given feed-rate range (57).

Mixer-settlers

A wide variety of mixer-settler, stagewise contact units are available in which operation may be batch or continuous countercurrent. The means for dispersion is commonly some form of agitated vessel. One pilot scale design employs pulsing to achieve dispersion. Flow

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niversity of Aston, Gosta Green, Birmingham 4.

or in-line mixers of the nozzle, injector, jet or mechanically agitated type may be used for more easily dispersed systems requiring low hold-up.

The settler may range from a simple vessel, traditionally about 2·5 times the size of the mixing vessel, to a complex vessel containing baffles or packing to assist coalescence. For applications where rapid settling is essential, a hydrocyclone might be used (31). This, in effect, combines the dual functions of separation and mass transfer. In its development for processing radioactive materials it has been suggested that by design improvements to eliminate the non-functional volume, residence times could be reduced to less than 2·5 sec, about one-tenth of that for pulsed columns (70).

Most mixer-settler cascades are built on a single level. Many integrated designs have been introduced to reduce the amount of interstage pumping and piping, floor space or agitation equipment required. For example, Treybal (63) has described the development of a mixer-settler having a single drive shaft. A number of mixing stages are incorporated in an inner column from which the phases flow via suitable connections to outer settling stages.

Some of the wide range of mixer-settler equipment has been described elsewhere (11, 32, 61).

Spray and packed columns

Adequate descriptions have been published of spray, packed and baffle plate columns and their applications (32, 61). No significant innovations leading to their wider applicability have been reported.

Pulsed columns

In pulsed columns the liquid phases are interdispersed by the application of a pulse. The pulse may be produced by means of a diaphragm pump or a valveless piston pump. A large number of reports have appeared of pulsing in laboratory packed and sieve plate columns, notably of late from Eastern Europe. Investigations have been concerned especially with the variation in extraction efficiency with pulse frequency and amplitude for different systems in addition to the usual range of variables, i.e. process conditions and column geometry.

Coggan (4) has recently presented a design procedure embodying a criterion for optimal design which it is claimed will enable production scale pulsed plate columns to be designed from experiments with a ⅞ in. diameter column. This paper includes consequential references to pulsed column studies of the atomic energy authorities.

A major difficulty with pulsed columns is to obtain uniform hydrodynamic conditions across the column cross-section. The original proposals by Van Dijck to pulse the plates themselves have been repeated in an attempt to overcome this. Complex and expensive mechanical design would be required, however, for other than the laboratory scale columns tested.

Commercial application of pulsed columns has, until now, been limited to the extraction of metals from radioactive solutions in the atomic energy field. For these applications the pulsing unit can be located outside the biological shielding thus facilitating routine maintenance. A 4 ft diameter pulsed plate column is, however, in use by Dutch State Mines for a non-radioactive separation. A packaged pilot scale unit with a 4 in. diameter column in industrial glassware containing SS perforated plates and a piston pump pulsing unit is manufactured by A.P.V. Ltd (1).

Power requirements of pulsed columns are relatively high. No information has been published on scaling-up problems but it is open to doubt whether pulsing is either economic or technically worthwhile for larger diameter columns on general duties.

Rotary agitated columns

Within this category the contactors which have achieved commercial application are (*a*) the Scheibel column, (*b*) the Rotary Disc Contactor and (*c*) the Oldshue-Rushton column.

The Scheibel column was introduced in 1948 and was subsequently patented (48). The original design consists of an alternate sequenc of mixing and packed sections in a vertical column. Each mixin compartment is agitated by a turbine impeller supported on central shaft. A typical design is shown in Fig. 1. The packe sections contain open-knitted wire or plastic mesh packings. Th material is dependant on the wetting properties of the extract an raffinate either of which may be the dispersed phase. The colum does not operate as a mixer-settler since droplets persist throughou the packed sections (13, 36). Thus, although some phase separatio does occur in the packing its main function is not to act as coalescence chamber but to isolate the mixing sections. Indeed unless the packing is preferentially wetted by the continuous phase large scale coalescence may result leading to the formation of coalesced core throughout the packing, channelling of the continuous phase at the walls and a reduction in capacity. The relativ height of packed and mixing sections may be varied to suit th particular duty. In the standard range of 1 to 6 in. diameter pilo scale contactors, by York Process Equipment Corp. (67), th packing and mixing section heights are increased on a *pro rat* basis with column diameter.

A modified design of the Scheibel column is now utilised fo sizes greater than 1 ft diameter (50, 67). This design has ring-shape baffles supported immediately above and below the impeller. Compartment baffles are attached to the column shell. This is illustrate in Fig. 2. For column diameters below 3 ft, with systems of lo viscosity and interfacial tension, the agitated sections may b isolated by the usual woven mesh sections. The purpose of th internal baffling is to provide uniform mixing of the phases an thus eliminate the tendency for H.E.T.S. to increase with colum diameter. Scheibel also used knitted mesh sections between th inner baffle plates.

A design somewhat similar to the later Scheibel column has bee described and tested by Leisibach (24). Results given for the syste propionic acid/toluene/water in 105, 285 and 500 mm diamete columns confirm the improved scale-up characteristics of thi design in that the H.E.T.S., 0·1 to 0·2 m, were less dependent o column diameter. The rotor speed for optimum extraction wa correlated by

$$N.D^{0\cdot9} = \text{const.} \qquad \ldots\ldots (1$$

The internal design of this column is fairly complex however and i remains to be seen whether it will be economic on a larger scale Columns to this design are available in 4 in. diameter glass with S internals (39). A variation of the original Scheibel design has bee described by Tudose (60). In this modification the packed section are replaced by baffled calming sections. An improved efficiency i claimed over the rotating disc contactor.

Scheibel columns have been widely used in the U.S.A. i diameters up to 7 ft. Few data are available other than for pil scale columns (48, 49, 50). No details have been published of an commercial application outside North America.

The Rotating Disc Contactor was proposed by Reman in 195 (41). The contactor consists of a vertical cylindrical shell divide into a number of compartments by a series of stator rings. rotating disc supported on a central shaft is located in each compartment. A typical design is shown in Fig. 3. Adjacent to the to and bottom compartments are feed inlets which are arrange tangentially in the direction of rotation. The dense phase is introduced into the top of the column and the light phase into th bottom; countercurrent flow is thus established by gravity. At th ends of the column are settling zones to allow the phases to separat One of the phases is dispersed by the action of the rotating disc Flat rotor discs without any protrusion are used to create unifor shearing conditions and hence obtain as small a spread in drople sizes as possible. There is limited photographic evidence that th dispersed phase droplets spin at fairly high speeds. This is supporte by the extraordinary sphericity of the droplets as illustrated i Fig. 4. At low hold-ups, in the absence of any coalescence redispersion effects, this factor accounts for the high efficienc obtainable (5).

Reman suggested initially that the flow of liquid is toroidal bu

TABLE I—Advantages and disadvantages of various contactors (57)

Type	Capital cost	Operating and maintenance costs	Efficiency	Total capacity	Flexibility	Volumetric efficiency	Space Vertical	Space Floor	Ability to handle systems that emulsify
Spray tower	5	5	1	2	2	1	0	5	3
Baffle plate tower	4	5	2	4	2	3	1	5	3
Packed tower	4	5	2	2	2	2	1	5	3
R.D.C.	3	4	4	3	5	4	3	5	3
Pulsed plate column	3	3	4	3	4	4	3	5	1
Mixer-settler	2	2	3	4	3	3	5	1	0
Centrifugal	1	2	5	3	5	5	5	5	5

Symbols used

C = concentration;
C_R = factor accounting for column restriction in an R.D.C. equal to S^2/D^2 or $(D^2 - R^2)/D^2$;
D = column diameter;
d_{crit} = critical drop diameter;
d_{vs} = average surface-volume drop diameter;
d_{pc} = critical packing diameter;
E_c = axial diffusivity-continuous phase;
g = acceleration due to gravity;
H = compartment height;
h = height of column packing;
H.E.T.S. = height equivalent to a theoretical stage;
H.T.U. = height of a transfer unit;
K = overall mass transfer coefficient;
k = film transfer coefficient;
l = a characteristic length;
m = distribution ratio;
N = rotor speed;
R = rotor diameter;
r_i = interfacial resistance;
S = diameter of stator opening;
t = time;
V = superficial liquid velocity;
ρ = density;
μ = viscosity;
y = interfacial tension.

Subscripts

c = continuous phase;
d = dispersed phase.

nis is an idealised picture which takes no account of the axial nixing which occurs above a certain critical rotor speed (22, 44).

In a design which is claimed to inhibit this unfavourable flow ehaviour Misek (29) has the agitated compartments placed off-entre. Next to the agitated zone is a transfer section separated rom the agitated zone by a baffle that permits flow between the ones only through passages near the periphery. This is illustrated n Fig. 5. Nevertheless, in scaling up from 1 to 2 m column diameter, llowance is made for an increase in nominal H.E.T.S. from 1·2 to ·75 m (54). Considering the increased column diameter required is doubtful whether this 'Asymmetric R.D.C.' will cost less to onstruct than an R.D.C. for an equivalent duty.

A modification patented by Reman involving two or more rotors n the column shell has not been adopted commercially (45). A esign employing perforated discs has been tested on a pilot scale s also has one in which the discs are replaced by rotors comprising our 60° arcs (21, 33). For the specific systems and operating ranges nvestigated both these designs had an extraction efficiency superior o the R.D.C. Smaller average drop sizes would be expected of ourse to lead to increased interfacial area but lower throughputs nd, in different circumstances, more serious axial mixing.

The original use of the R.D.C. by the Shell Companies was in ne furfural extraction of petroleum lubricating oils for which they roduced a saving of about 60% of the capital cost of equivalent acked towers and had a separating power equivalent to a 14-stage nixer-settler system (43). The R.D.C. has now found extensive use n petroleum extraction processes including propane deasphalting 28), SO_2 extraction of Kerosine fractions, Sulpholane extraction nd naphtha sweetening. About 150 units are estimated to be in ommercial operation (46); units 10 ft in diameter and 70 ft high ave been built. Industries as diverse as food, by-product coke, rganic chemical and metal separation have utilised the R.D.C. Descriptions have appeared recently of its use for the separation of xy-compounds from Fischer-Tropsch synthesis oil (2) and in the J.S.S.R. for the extraction of caprolactam (19). For the latter pplication it is claimed to be more economic than centrifugal xtractors and superior in flexibility of operation and in insensitivity o solid phase impurities. The simple basic design has enabled a nit to be fabricated in glass reinforced epoxy resin for the aqueous xtraction of metal chlorides from a hydrocarbon solvent slurry in polyolefin manufacturing process (68).

Westerterp and Landsman (66) have noted the potential of the .D.C. as a continuous reactor for homogeneous liquid phase eactions especially in cases where large heat effects and long olding times are involved. Dependent on the combination of hroughput and rotor speed such a reactor could, in theory, be perated at a desired spread in residence times ranging from that f a perfect mixer to that of a nearly ideal tubular reactor (66).

The Oldshue-Rushton extractor (7, 34) consists of a vertical olumn divided into compartments by horizontal stator rings. Each ompartment has four vertical baffles and is agitated by a turbine npeller driven by a central shaft.

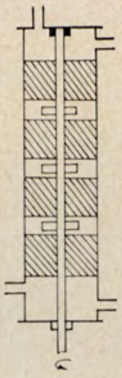


Fig. 1 Original Scheibel column (48).

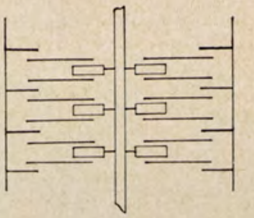


Fig. 2 Modified Scheibel design.

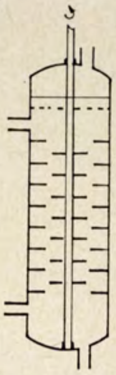


Fig. 3 Rotating disc contactor (41).

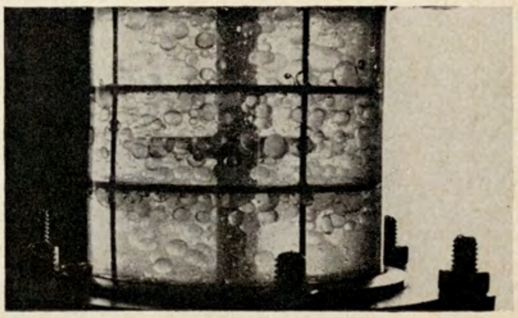
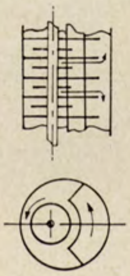


Fig. 4 Droplets in one compartment of a pilot scale rotating disc contactor.



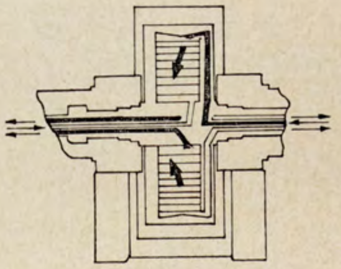


Fig. 5 Asymmetric rotating disc contactor (29)
Fig. 6 Podbielniak centrifugal extractor (37).

The extractor was first introduced as early as 1945 but is only now achieving commercial status outside North America. The design has been adapted for a variety of other unit operations (26) and for chemical reactors.

In the comparison of performance of different contactor designs, account must be taken of both the efficiency and the volumetric flow capacity. Comparisons can only be drawn for the same liquid-liquid-solute system. An index of effectiveness has been defined (41),

$$I_e = \frac{\text{total throughput}}{\text{volume of one theoretical stage}} \quad \dots . (2)$$

Vermijs and Kramers have used this index to compare data for the system water-acetic acid – MIBK (65). Over the range of variables and small column diameters covered, the R.D.C. and Oldshue-Rushton contactors were found to have a similar effectiveness in the range 1·6 to 3·4 min^{-1}. The Scheibel columns have rather lower figures, 1·4 to 2·2 min^{-1}. In effect the Scheibel columns have relatively small H.E.T.S. values but the capacities are also lower, comparable in order to those of packed columns. Using a similar index, Ponikarov, *et al.* (38) compared the performance of spray and packed columns, sieve plate columns, the R.D.C. and pulsed plate columns. For one system they found that the pulsed plate column was most effective but that the R.D.C. had a smaller H.E.T.S. value. Results are only strictly comparable, however, if the selection of the dispersed phase, solute concentration, phase ratios and the direction of solute transfer are identical. Even then consideration must be given to the variations in efficiency of the various contactors with scale-up. In general, the R.D.C. by virtue of its controllable shear characteristics is preferable for systems prone to emulsification. The turbulent agitation produced in the Oldshue-Rushton contactor renders it more suitable for extractions with chemical reaction in which dispersion is rate controlling. It has, for example, been used in the recovery of butadiene from other C_4 hydrocarbons by extraction with cuprammonium acetate solution.

Centrifugal contactors

Countercurrent flow in the contactors discussed above is maintained by the gravitational effect arising from specific gravity differences between the phases. This imposes a limit on liquid velocities and hence on the minimum extractor volume for a given extraction. Centrifugal contactors utilise large centrifugal forces to induce rapid countercurrent flow of the phases. The general theory of centrifugal extraction has been reviewed by Todd and Podbielniak (59).

Foremost in this class are the Podbielniak extractor (37) and the Luwesta extractor. The Podbielniak extractor is shown in Fig. 6. It comprises a horizontal shaft on which is mounted a rotating drum containing perforated concentric cylinders. The liquids enter and leave through mechanical seals at the shaft. The heavy liquid enters the drum near the shaft and the light liquid near the periphery. Radial counterflow and highly efficient contacting/phase separation are produced by rotational speeds in the range 1,400 to 3,000 rpm. Internal construction is designed specifically for the number of stages and operating conditions required. The number of effective contact stages may vary up to 10. Podbielniak units with total flow capacities within the range 5 to 800 U.S. gpm have total hold-ups of only 2 to 180 gal.

The Luwesta extractor, developed in West Germany (8, 9) contains discrete mechanical stages each comprising a countercurrent mixing and settling stage. Intimate mixing is accomplished by flow of the liquids through a number of parallel nozzles mounted in a vertical non-rotating tube. The tube is centrally located in, and interconnected to, a centrifugal separator bowl which serves for separation of the equilibrated dispersions. Three stage extractors are available with total liquid capacities of 130, 530, and 1,850 gph; a two stage extractor is also available with a capacity of 2,650 gph (46).

Centrifugal extractors are necessarily precision built machines and consequently have a high first cost and proportionately high maintenance costs. Against this, however, may be set some quite considerable advantages for certain applications. The high flow capacity with minimal hold-up has led to the extensive use of both the above extractors in the pharmaceutical industry. There are also potential savings in solvent inventory and in off-specification material on start-up. The latter is important when a wide variety of feeds may be handled such as in lubricating oil refining. Centrifugal extractors are able to handle phases with a very small density difference and to operate over a wide range of phase ratios and viscosities. A back-flushing technique may be necessary when handling liquids containing finely divided solids unless solids can be removed with one of the liquids. This is necessary to avoi imbalance of the rotor.

The Podbielniak unit has been used in a range of petroleum refining operations, in vegetable oil refining, in the dephenolisatio of coke oven liquors and in uranium extraction. A description ha been given of its use as an acid sludge separator and for causti phase separation/water washing in packaged acid treating processe for the removal of trace olefins from paraffins or aromatics (58 Although obsolescent in petroleum refining this process is of con siderable general interest; performance in pressure tight centrifuga contactors enables losses due to overreaction and entrainment t be reduced. In recent years the unit has been utilised for the remova of inorganic and other catalysts from sometimes quite viscous solu tions of some of the newer polymers. Very low residual catalys levels, perhaps 10 ppm, are achieved for example in the productio of E.P. rubbers or polybutene by countercurrent aqueous washing

Currently the majority of the existing Luwesta extractors, totallin about 180 machines, contain three stages. Their main use is in th extraction and concentration of penicillin but some are in use i the recovery of hormones, caffein, pyrethrin, ephedrine an quinine. Machines are also in use in the extraction of fruit juice detergents and uranium oxide. The field of general application i limited by the volumetric capacity of the available machines.

A centrifugal contactor is also marketed by Alfa-Laval Ltd. Th unit is vertically mounted and countercurrent flow of liquids occur in a spiral path in a rotating bowl. The liquids are intimately mixe by passage through orifices at intervals along the path. Data o applications and performance have not been published other tha the maximum feed rate of 3,800 gph and three to twenty extrac tion stages suggested by Reman (46) depending upon th system handled.

A Quadronic centrifugal extractor has recently been introduce by Liquid Dynamics (69). This is in general similar to the Pod bielniak unit but has means for internally altering the perforate area of the cylindrical plates.

Design fundamentals

The traditional design procedure for extraction equipment neces sitates the evaluation of

(*a*) The number of stages, or transfer units for the required degre of separation.
(*b*) The stage efficiency or H.T.U. and
(*c*) The equipment volumetric flow capacity.

The number of stages or transfer units are estimated from mathe matical or graphical mass balances (61). For continuous contac equipment the stage efficiency or H.T.U. may be determined b pilot scale experimentation or from previous plant experience; fo mixer-settler units the stage efficiency usually approximates to 7 to 90%. Capacity may be predicted from flooding correlation derived by empirical correlation of dimensionless relationships.

Fundamentally the operation of liquid-liquid extractors depend on the dispersion of one liquid as droplets into another, with whic it is wholly or partially immiscible. This is followed by mass trans fer to or from the droplets and finally separation involvin coalescence of the dispersed phase. Hence droplet size is a facto influencing both the process of mass transfer and also the volumetri capacity. Extractor design ought, therefore, to be from drople analysis considerations.

The rate of mass transfer *dx*/*dt* may be evaluated as the produc of an overall mass transfer coefficient *K*, interfacial area *A* and th concentration driving force ΔC.

$$\frac{dx}{dt} = K.A.\Delta C \quad \dots . (3$$

The interfacial area is a function of droplet size and the disperse phase hold-up in the contactor. Droplets are assumed to be spherica and of uniform size. The overall coefficient is related to th coefficients of the individual phases by the familiar equation,

$$\frac{1}{K_d} = \frac{1}{k_d} + \frac{1}{mk_c} + r_i \quad \dots . (4$$

where k_d is the coefficient for diffusion within the droplets, k_c i the continuous phase coefficient, m the distribution ratio and r_i th interfacial resistance. The last term may be neglected for desig purposes in all but a few cases, such as when solute transfer i accompanied by slow chemical reaction at the interface. Coefficient are taken from single droplet studies.

This is an over-simplified approach which takes no account o the respective roles of molecular and eddy diffusional mechanisms For any system the film coefficients are dependent on the flo patterns set up inside and outside individual droplets. Mass transfe udies with flowing single droplets have confirmed that the magniıde of these flows is governed by droplet size (55). Terminal elocity is a maximum at a critical droplet size corresponding with e commencement of droplet oscillations. THORNTON established e existence of three hydrodynamic regimes; at low Reynolds umbers droplet circulation was virtually absent and mass transfer roceeded by a molecular diffusion mechanism. In the transition egion the film coefficients were governed by the same mechanism. t a size sufficiently larger than the critical size large rapid oscillaons were apparent, and an eddy diffusional mechanism preominated. The importance of considering the distribution of roplet sizes in any contactor when extrapolating from single roplet data is clear; this data is unfortunately seldom available.

The extent of droplet-droplet interaction and the effect of terfacial turbulence in promoting or inhibiting it are also signicant factors. These will be referred to later. The driving force, in rms of concentration difference, may also be significantly reduced practice by the effects of axial mixing dependent on the charactertics and diameter/length ratio of the particular contactor.

The application of the simplified model approach to the design f mixing vessels in mixer-settler units has been covered by EFFREYS (16). There is now evidence that solute transfer in mixing essels is considerably enhanced by the complete remixing and dditional turbulence set up within droplets resulting from a oalescence-redispersion mechanism (56, 64). Several workers have ound that the order of interface generation in a stirred vessel is 00-1,000 ft^2/ft^3. This and the droplet interaction process accounts r the high mass transfer rates achievable. Scale-up of mixing essels based on geometric similarity, constant power per unit olume and the same order of liquids residence time is adequate for ost practical purposes. The capacity of mixer-settlers is therefore etermined by the settler design.

In the simplest form of settler comprising a horizontal tank, the ispersion from the mixer is distributed from the inlet port in the rm of a heterogeneous wedge between the two phases. The wedge esides above or below the interface dependent upon the relative ensities of the dispersed and continuous phases. The settler is ormally operated so that the wedge extends over only 70% of the ngth of the interface. The wedge should not cover the whole area therwise any increase in throughput will increase the wedge thickess which may cause loss of extract through the raffinate port and ffinate through the extract port. This would result in decreased ficiency. The wedge width and length are determined by the ifference between the input rate of the emulsion and the rate of oalescence of droplets of the dispersed phase in the wedge with e interface. JEFFREYS, *et al.* (17) have developed a mathematical odel based on the concept of a 'coalescence stage' in order to redict these dimensions. They evaluated the number of stages for xperiments in a laboratory scale mixer-settler and found good greement between predicted and experimental ratios of final to itial drop size, wedge width and length. This is a first step away om the usual trial and error design procedure.

Numerous types of mechanical aids have been incorporated in ettling vessels in an attempt to promote phase separation. Perorated impingement baffles are commonly used to reduce agitation the settler due to the injection of emulsion; baffles parallel to the irection of flow have also been used. Alternatively, the settler may e filled with some form of packing, such as glass wool or plastic r wire mesh, which is preferentially wetted by the dispersed phase. t present, design involving baffles or packing necessitates a trial nd error procedure. There is a need for fundamental work in this rea. The process of coalescence at a plane interface is already ttracting a considerable amount of study and droplet-droplet oalescence somewhat less because of the experimental difficulties volved (3, 23).

The design of spray and packed columns from droplet analysis onsiderations has been discussed by JEFFREYS and ELLIS (15). The bsence of any internal packing or plates however renders the spray olumn particularly prone to axial mixing. This can lead to an verestimation of column performance by as much as 30%. The eories of axial mixing and specific research results with both pray and packed columns have been reviewed by LI and ZIEGLER 25).

For a packed column it is well established that, in the absence f mass transfer, the mean droplet size at the outlet is independent f the mean droplet size entering from the distributor providing e packing is greater than a limiting size defined by

$$d_{pc} = 2{\cdot}42 \left(\frac{y}{\Delta p.g}\right)^{0.5} \qquad \ldots . (5)$$

where d_{pc} is the critical packing diameter, y the interfacial tension and $\Delta\rho$ the density difference between the phases. Packings larger than d_{pc} should always be used in practice. A useful nomogram has been published for estimating the critical packing size for a given pair of liquids (47). RAMSHAW and THORNTON (40) have made an interesting study of the basic mechanism of breakdown of droplets larger than the critical size by collision with elements of packing. They found that the equilibrium droplet size (d_{vs}) eq. was established after several feet of packing. In the absence of mass transfer, the change in droplet size d_{vs} with height h could be correlated by the expression

$$d_{vs} = 0{\cdot}45 + 0{\cdot}27 \exp - (0{\cdot}0157h) \qquad \ldots . (6)$$

For the case of mass transfer from the continuous to the dispersed phase

$$d_{vs} = 0{\cdot}45 + 0{\cdot}22 \exp - (0{\cdot}0382h) \qquad \ldots . (7)$$

Equilibrium droplet size distribution was in both cases positively skewed and could be roughly represented by a log-normal distribution function. As would be expected, mass transfer in the opposite direction produced large irregular droplets. A critical droplet size d_{crit} was found to exist below which further breakdown did not occur. The relationship between (d_{vs}) eq. and d_{crit} and other factors affecting (d_{vs}) eq. require further study before (d_{vs}) eq. can be predicted from fundamentals.

No fundamental design has been attempted for the Scheibel contactor because of the difficulty of correlating the design variables. The application of a stagewise technique has been discussed by JEFFREYS (16) using published data to predict stage efficiency (49). HONEKAMP and BURKHART found that 25 to 50% of the solute transfer may occur in the packing (13). The capacity of the column is limited by the permissible flowrate through the packing. Since the packing is of small interstices size however the usual flooding correlations are not applicable and unusual hydrodynamic phenomena occur (13, 36). Little fundamental work of consequence has been done in this area.

R.D.C. performance was correlated initially by REMAN and co-workers (41-44) in terms of the power input per unit mass N^3R^5/HD^2 as determining the drop size of the dispersed phase. Good correlation of capacity $(V_C + V_D)/C_R$ against this group was obtained over a range of column sizes.

This has now been superseded by an approach based on 'characteristic velocity' (22, 27, 53). A detailed procedure has been published for the fundamental design of an R.D.C. incorporating factors to make allowance for coalescence and axial mixing (16, 29). The empirical coalescence factor is necessarily based on an over-simplified model in which every collision results in coalescence (30). STERMEDING, *et al.* have found that axial mixing behaviour in an R.D.C. can be correlated with operating and geometric variables (52). In R.D.C.s between 3 in. and 7 ft in diameter the axial mixing, expressed as axial diffusivity, was correlated for the continuous phase by

$$E_C = 0{\cdot}5\ HV_C + 0{\cdot}012\ \text{R.N.H.} \left(\frac{S}{D}\right)^2 \qquad \ldots . (8)$$

Dispersed phase diffusivity apparently varied up to a factor of three times the value for the continuous phase. The results of a series of studies on axial mixing in R.D.C.s have been reviewed by REMAN (46) who confirms that the theoretical concepts are borne out by experimental investigation. Current R.D.C. design and scale-up makes allowance for axial mixing. However, as has been pointed out by OLNEY (35), the solute transfer process from the assemblage of drops produced in an R.D.C. is extremely complex. The distribution of drop sizes in addition to determining the mode of solute transfer, as discussed earlier, also govern the frequency of droplet-droplet coalescence, if present, and also the extent of axial mixing. That is to say the smaller the drop the longer it will persist in the contact zone compared to the average drop and it may be highly backmixed by any gross eddy motions in the continuous phase since it has small inertia. The balance between the optimum droplet size distribution for mass transfer and the tendency for axial mixing to increase with smaller drop sizes are then contributing factors determining the optimum rotor speed which exists at any given throughput for any extraction (42, 44). Similar results apply to other contactors (34, 49, 50, 24). A large number of unconnected mass transfer studies with different systems have been reported for laboratory scale R.D.C.s. For these reference should be made to the general reviews mentioned earlier.

In contrast little data have been published on the Oldshue-Rushton contactor. Recently GUTOFF has measured the extent of interstage mixing in a 4 in. column of this design (10). Mixing between stages was found to be slight at low agitator speeds but to increase very rapidly at higher speeds. The extent of interstage mixing was similar to that reported in the literature for R.D.C.s.

Centrifugal contactors are all of proprietary manufacture. As would be expected, therefore, no design data or extraction studies have been reported other than those referred to earlier.

Other design considerations

There are a number of factors of general importance in the scale-up and design of liquid-liquid extractors.

Dynamic similarity and geometric similarity should be maintained. In fact, dynamic similarity cannot be maintained since equal Reynolds numbers ($lv\rho/\mu$) which control mass transfer and Weber numbers ($lv^2\rho/\gamma$) which control drop sizes and their distribution are obviously impossible on two scales (64). Scale-up by making the model an element of the larger unit is rarely feasible, certainly in rotary agitated columns. Much work remains to be done in this area. In maintaining geometric similarity it is important with the R.D.C. to conform to the established range of variables for optimum performance, i.e. S/D 0·66 to 0·75, R/D 0·5 to 0·66 and H/D 0·33 to 0·5 (22). Below a certain stator-rotor gap the dispersed phase tends to flow in rivulets so that the column no longer operates as an R.D.C. A rotor peripheral speed larger than 300 ft/min. is recommended. Some reported mass transfer studies have unfortunately been in columns of different geometry (18, 20). In the Oldshue-Rushton column, diameter is nominally three times that of the impeller and twice the compartment height.

Wettability of column internals has an important effect on efficiency. For optimum performance any packing, rotors or stators should be preferentially wetted by the continuous phase to prevent reduction in interfacial area due to coalescence or, in the extreme, channelling of the dispersed phase. This has been investigated in an R.D.C. by Davies (5) and the effects noted by others in the Scheibel column (36) and pulsed plate column (4). When an aqueous phase is dispersed wetting effects can be avoided by the application of a suitable hydrophobic lining; most plastics would be suitable. For operation with a continuous aqueous phase most metals are suitable, with the exception of copper and molybdenum, providing the surface is oil free.

With many systems the rate of mass transfer is higher when the direction of transfer is from the continuous phase to the dispersed phase than vice-versa. This is due to the Marangoni effect; the effect of local variations in surface tension in the fluid interface, associated with concentration gradients, which tend to hinder coalescence in the first case and aid it in the second. Results of several workers confirming this phenomena for spray columns have been summarised by Smith (51); data have been reported also for packed columns, mixer-settlers, pulsed columns, the R.D.C. (27) and the Scheibel column (5, 49). Depending on the significance of this effect on mass transfer rates and contactor flooding characteristics it may be equally as important as relative throughputs in the selection of the dispersed phase. It is for this reason that pilot scale studies of contactor hydrodynamics in the absence of mass transfer are carried out with mutually saturated phases. Clearly any flooding correlations or droplet size analyses obtained in the absence of mass transfer may be subject to considerable modification in practice.

The presence of traces of surface active agents has a marked effect on droplet characteristics and hence contactor efficiency and capacity. Davies has reported that the addition of Teepol produced an increase in extractor efficiency but a reduction in capacity in a pilot scale R.D.C. (5). The seriousness of such effects in petroleum refining applications is well known (5, 43). Conversely the presence of grease or dirt tends to aid droplet-droplet coalescence. These are further factors determining the provision of variable speed rotor drives in commercial scale contactors. All commercial grade materials contain varying amounts of extraneous matter and due caution must be applied in scaling-up results obtained by laboratory investigation.

In conclusion, given the present state of knowledge, adequate pilot scale trials are advisable for any new extraction process. These should be under simulated process conditions in an extractor of similar design and materials of construction.

Actual plant feeds should be used whenever possible. Preliminary shake-out tests may be of value as an indication of the time required to reach equilibrium, settling times and the effect of phase ratios (64). Pilot scale studies should cover a wide range of flow-rat and phase ratios and provision should be made for solvent recove and recycle. Repeated contact with a feed and the process recovery from the extract can alter solvent properties and sign ficantly affect contactor performance. Columns of 3 or 4 in. di meter are generally considered a minimum to avoid excessive wa effects. Packed towers may need to be larger to utilise the san packing as on the larger scale without detriment to liquid distrib tion; a minimum column diameter is eight times the nominal packir diameter. Allowance must be made for end effects.

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Drop Phenomena affecting Agitated Liquid-liquid Contactors

by

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Foremost among recent advances in quipment for liquid-liquid contacting e numerous designs in which mechanical ergy is applied to achieve controlled ispersion and high mass transfer rates. dustrial applications and developments the design of such equipment for quid extraction have been discussed sewhere (15).

Equipment is generally classified cording to its mode of operation, *i.e.*,

) Stagewise contact; batch or continuous, *e.g.*, mixer-settlers.

) Continuous countercurrent columns with phase flows established by gravity, *e.g.*, pulsed columns; rotary agitated columns.

his excludes a special group, centri-gal extractors, in which phase separa-on is also mechanically aided).

In either case the basis of operation is spersion of one liquid as droplets into continuous liquid phase followed by ass transfer to or from the droplets (and ssibly chemical reaction) and finally paration of the dispersed phase involv-g coalescence. The distribution of drop es and the extent of droplet interaction the contactor are the major factors ecting mass transfer efficiency and also, continuous columns, the volumetric pacity. This paper considers the nificance of some droplet phenomena ich occur under practical operating nditions in agitated contactors.

ESIGN PROCEDURE

The significance of the various enomena to be discussed will be clear m a summary of the classical design cedure. This assumes spherical drops uniform size. The rate of solute nsfer dx/dt is then evaluated as the product of an overall coefficient K, interfacial area A and concentration driving force ΔC.

$$\frac{dx}{dt} = K.A.\Delta C. \qquad \text{............(1)}$$

The overall coefficient is dependant on the series resistance to diffusion inside the drop, outside the drop and at the interface (if any). Prediction of these from discrete single droplet studies has been fully treated by Jeffreys and Ellis (6) and by Kintner (9). For a specific system, drop diameter and velocity relative to the continuous phase are the governing factors. The initiation of internal circulation is desirable to establish solute transfer by eddy diffusion within the droplet and to enhance continuous phase transfer. More recently the production of oscillating droplets, in which internal circulation may be deformed into random mixing, has been shown to result in higher internal mass transfer rates. Correlations have been proposed for this case on the basis of the new surface created.

Equations also exist for the estimation of the enhanced interfacial area of oscillating droplets.

The interfacial area is a function of droplet size and dispersed phase hold-up. In batch operation the hold-up is solely determined by the phase ratios. For continuous flow operation, hold-up is conveniently predicted by means of equations relating it to the superficial velocities of the dispersed and continuous phases and the characteristic velocity of the droplets in the contactor. These take the form

$$\frac{V_C}{1-X} + \frac{V_d}{X} = V_N(1-X) \qquad \text{........(2)}$$

The characteristic velocity V_N depends on the geometry of the contactor and the physical properties of the liquids.

The maximum diameter of droplets formed in an agitated contactor may be predicted from the equation derived by Hinze for a homogeneous isotropic turbulence after Kolmogoroff and where the microscale of the turbulence is smaller than the drop size (5), *i.e.*,

$$d_{max}\left(\frac{\rho C}{\sigma}\right)^{3/5} E^{2/5} = C_1 \quad \text{........(3)}$$

where E is the energy dissipation rate per unit mass and C_1 is a constant dependent on agitator design. Splitting of drops is more effective the higher the intensity of agitation; drop diameter decreases therefore with increased impeller speed. Alternative equations have been derived using a similar approach by Shinnar and Church (23) and for a rotating disc by Misek (14). The mean drop diameter may be estimated from d_{max} using empirical relationships (8).

For continuous flow operation, capacity may be predicted from the rate of ascent or descent of droplets of mean diameter through the continuous phase.

BATCH OPERATION

For batch operated mixer-settler units in which the mixer is simply an agitated vessel this simplified model approach gives at best a conservative estimate only (8). Agitation causes not only droplet break-up; in the turbulence droplets travel in all directions resulting in collisions and coalescence. Thus under fully baffled conditions at constant impeller speed a state of dynamic equilibrium is established between the number of coalescing and splitting droplets. The drop size distribution is then dependent on hold-up as well as the intensity and nature of agitation. Numerous studies have correlated the dependance of interfacial area on agitation in baffled vessels. These have been tabulated in the form

$$A = k_1 R^a D^b N^c \quad \text{............(4)}$$

where k_1 is a constant combining the physical constants and specific influenc of agitator design (24). Values of th exponents lie within the range a=0.8 t 2.0, b=0 to –1.2 and c=0.7 to 1.2. Som degree of discrepancy inevitably arise from the different methods employed fo the measurement of the interfacial area High speed photography and photo electric techniques, relating interfaci area directly to the amount of light transmitted through a dispersion, are mo common.

In any event it is sufficient to recor here that the order of interface generation is generally of high order, e.g 500–1,000 ft.^{2}/ft.^{3} The coalescence – redispersion mechanism leads to a reduction of the resistance to diffusion acro both phase films. At each instance th concentrations in the main phases ar adjacent to each other, thus producin extremely high concentration gradient Therefore, high mass transfer rates occ due to interface renewal and also t stretching of the surface of the dro during oscillations about the spheric norm.

For these reasons traditional metho based on geometrical similarity, equiv lent liquid residence times and consta power per unit volume are usual sufficient for the design of batch operat contactors. Indeed, in many instances is the provision of too much agitatio perhaps in conjunction with materi having surfactant properties, whi creates problems due to the formation fine dispersions which necessitate excessive residence time in the settl Some of the fundamental and practic aspects of settler design have be referred to elsewhere (15).

CONTINUOUSLY OPERATED MIXER-SETTLERS

In a continuously operated fully-baffl mixer, the Sauter mean droplet diamet at constant impeller speed has be reported to increase linearly with hold- due to coalescence effects (26), *i.e.*,

$$d_{vs} = d_{vs}^{o} + mX \quad \text{............}$$

where d_{vs}^{o} is the drop size at zero hol o and therefore a function of the power ssipation and the physical properties of e system. The term m was found to be onstant for a given system regardless of ower level and may be considered a easure of the coalescence characteristics f the system. Thornton believes that ese preliminary results provide scope r a unified treatment of droplet size-old-up data in mixers albeit in the sence of mass transfer.

GITATED COLUMNS

There exists a multiplicity of designs r agitated liquid-liquid column contacrs. Either a rotating agitator or a ulsing device may be employed. Since e technical and economic limitations of ulsed columns has precluded their use on rge scale general duties (15) they will t be considered here. Nor as yet have y systematic studies been reported on e Morris Contactor, a novel horizontal ntactor in which droplets persist roughout (16). This leaves the range rotary agitated columns.

Column geometries and internals have een varied usually with the expressed tention of improving the scale-up aracteristics, *i.e.*, reducing the tendency r H.T.U. to increase with column ameter due to axial mixing effects. stablished designs are the Rotating Disc ontactor (19, 20, 25) and its modificaon the Assymetric R.D.C. (11), the cheibel Column (22) and the Oldshue-ushton Column (17). The basic design fferences are summarised in Fig. 1. The .D.C. comprises a column with equiaced horizontal stator rings each mpartment being agitated by a centrally cated flat rotor disc. In the A.R.D.C. which there has been little published ork, the agitated compartments are offntre and countercurrent phase flow curs only via a special transfer section. he Scheibel Column has agitated comrtments alternated with sections conining open-knitted mesh packings. The ldshue-Rushton Column with horizonl stator rings and four vertical baffles is early a development from the use of agitated vessels in liquid-liquid contacting and indeed has some similar characteristics.

Certain droplet phenomena are common and the columns may be considered together. It is however worthwhile to summarise what is established regarding the operating characteristics of the individual columns.

(a) Rotating Disc Contactor

The optimum operating conditions exist over a fixed range of column internal geometry (10, 13). Within this range, extraction efficiency increases with increasing rotor speed; a maximum may be reached however as axial mixing becomes increasingly effective due to the increased turbulence and smaller mean drop sizes associated with higher speeds. Capacity decreases with increasing rotor speed (19). Hold-up increases with dispersed phase flow-rate up to the loading point and then increases more sharply (10, 27). The hold-up is related to phase flow-rates by a modified form of equation (2) for which V_N may be predicted from correlations relating it for any system to column diameter and rotor speed (10). In order to apply such correlations Misek (13) has suggested the necessity of introducing a coalescence factor. Operating phase ratios, on a continuous dispersed phase basis generally lie between 0.5 to 4:1.

(b) Scheibel Column

Consideration will be limited here to the earlier design employing woven mesh packed sections (22) since omission of these or their replacement by baffled calming zones (21) is associated with droplet phenomena not unlike those in the Oldshue-Rushton Column. The preferred agitator is a turbine giving predominantly radial flow since vertical baffles are not recommended (21). As for the R.D.C., an optimum agitator speed exists for any given duty. Total hold-up comprises dynamic and static hold-up and increases linearly with agitator speed. The contactor capacity is determined by the limiting flow-rate through the packing.

(c) Oldshue-Rushton Column

Little data are available on the performance of these columns. Efficiency-agitator speed curves are of typical form (17). Hold-up has been related to phase flows by Misek using a modified form of equation (2) (12). Limiting phase ratios and flooding phenomena are similar to those in the R.D.C.

The extent to which droplet interaction occurs in any column is of prime importance. Such effects enhance mass transfer and also affect the drop size distribution and hence mean droplet velocity.

As mentioned earlier, Misek introduced a coalescence factor into procedures for the design and analysis of R.D.C.'s and has more recently attributed the lack of inter-droplet coalescence in practice to the presence of surfactants. However, Davies, in contrast to earlier workers with the same contactor, established that in the absence of mass transfer coalescence did not occur to any significant extent, albeit at high phase ratios of 12 to 16:1 (3). More recent work has reinforced this conclusion at low phase ratios with low and medium interfacial tension systems below the flooding point (7). It is conceivable that in the R.D.C. droplet spin is a factor not only in achieving high mass transfer rates but also in stabilising the drops against coalescence. In contrast in a Scheibel Column the nature of the agitation and the significant proportion of solute transfer which occurs in the packing would suggest that a coalescence-redispersion mechanism predominates and this has been confirmed experimentally. Within the normal range of operating parameters the same is true for an Oldshue-Rushton Column. This does not preclude a certain proportion of drops maintaining their identity however (1.7). Thus in terms of extraction efficiency only, the Scheibel would be expected to perform better than the R.D.C.; this is borne out by limit published data (28).

It would seem reasonable to analy R.D.C. column performance in terms discrete drops of predicted mean diamete However, two important factors mu not be overlooked,

(a) It is well established that coalescen is aided by special interfacial effec associated with solute transfer (This occurs for example in the case the transfer of solute which reduc interfacial tension from the disperse to the continuous phase. Larg droplets are then observed compare with those when the feeds consist mutually saturated liquids as customary in hydrodynamic studie The effect is usually a function solute concentration so that hold-u may vary along the contactor.

(b) Wide distributions occur in dro sizes. This is illustrated in an R.D. in Fig. 2, which also incidental shows the tendency for centrifug tion of small drops to the periphe in this type of column.

These phenomena are common to other contactors.

These wide distributions which occ in drop sizes may significantly affect t dispersed phase film coefficient sin solute transfer occurs by both molecul diffusion (*i.e.*, with small non-circulati drops) and by eddy diffusion simultan ously. The preponderance of either dependent on the form of the distrib tion. In practice, it may be possible neglect the enhancement of diffusivi due to internal circulation within drople in an R.D.C. (25) and especially so trace material, which would inhib circulation, may be present.

Axial mixing, which results in reduced solute concentration differen between the contacted phases, is partic larly important in agitated columns. exists in both continuous and dispers phases. At any rotor speed with a giv system the extent of axial mixing greatly dependent on the design an

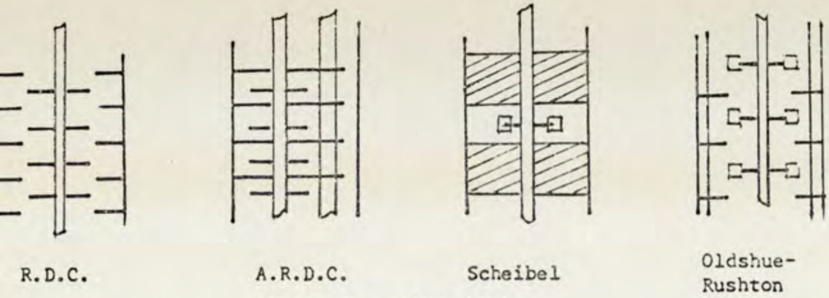

Fig. 1. *Rotary Agitated Columns.*

ocation of internal stators (10, 17). Design methods are now available which make allowance for axial mixing but elatively little experimental data have been published (1, 25). Our concern here s with drop phenomena and hence the validity of dispersed phase axial mixing models based on volume-surface mean diameter drops. High speed cine-photog-aphy serves to illustrate the complexity of the problem (7). Superimposed upon the normal distribution of drop rise times are the effects of eddy diffusion which are in themselves dependent on drop size distribution. For example, drops which are smaller than the average and which persist in any column section for long periods have small inertia and will therefore be highly back-mixed by gross continuous phase flow patterns. Larger drops will pass more rapidly through the ection. The only fundamental analysis of such departures from ideal flow behaviour in agitated columns has been that of Olney et al (18, 25).

The optimum agitator speed in any contactor is dependant on the effects of axial mixing. A minimum speed also exists for any duty. For example, in the R.D.C. a minimum disc tip speed of 300 ft./min. has been recommended to avoid drops becoming trapped beneath the rotors and stators (10). This was based on observations with the toluene-water system and is excessive for systems of lower interfacial tension, *e.g.*, systems excluding an aqueous phase, since it produces fine dispersions. In the Oldshue-Rushton Column there exists a minimum speed for transition from a tangential to the desired radial flow pattern. There is also a tendency at low speeds for flocculating droplets to become trapped in a dead space between stator, vertical baffle and column wall which may lead to premature flooding (1, 7).

Another general limitation is that hydrodynamic and laboratory mass transfer studies are invariably performed in columns with ultra-clean internal surfaces. These should be preferentially wetted by the continuous phase. In practice, since most metals are only weakly hydrophobic, wetting of column internals or woven mesh packings by an organic dispersed phase in any aqueous continuous phase increases with exposure time in the contactor. Thus fall off in efficiency, due mainly to decreased interfacial area, can occur with time as in conventional packed columns. For example, Fig. 3 shows globules much greater than the predictable maximum drop size being sheared off from an organic dispersed phase coalescing on a horizontal stator in an agitated column section (7). Such drip point phenomena occur randomly as regards both time and position. Similar effects arise from column walls and certain rotor designs and have also been reported in pulsed plate laboratory columns (2).

Certain droplet phenomena arise specifically in the Scheibel Column due to the presence of the small interstices size packed sections. Although the basic purpose of the packing, which is preferentially wetted by the continuous phase,

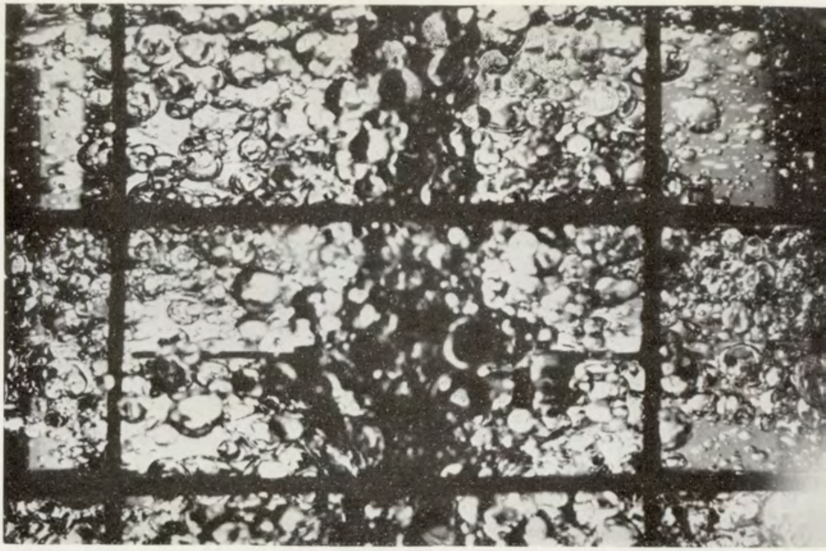

Fig. 2. Drop Size Distribution in an R.D.C. (*System; toluene-water*, $N=850$ *r.p.m.*, $X=0.1$).

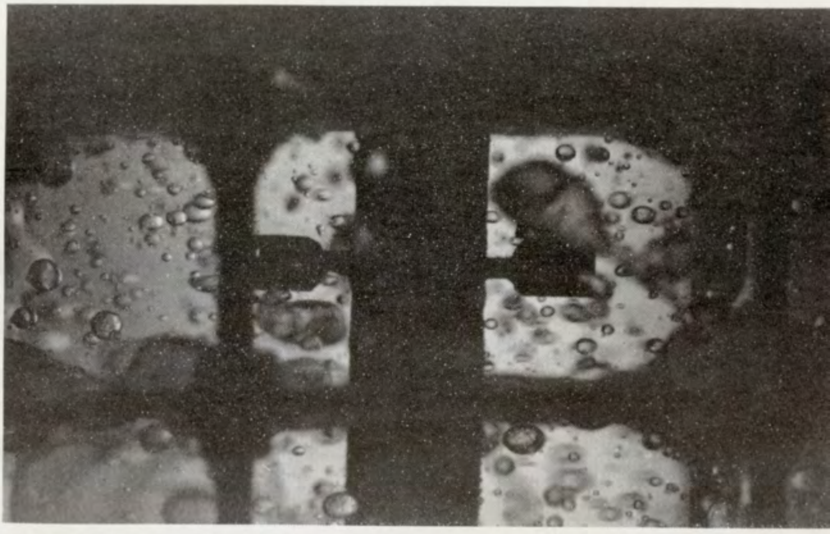

Fig. 3. Globules from Dispersed Phase Wetted Stator (*System; toluene-water*, $N=500$ *r.p.m.*, $X=0.23$

to isolate the flow patterns in successive ixing stages some coalescence does ccur. The extent is dependant on drop ze, the physical properties of the system specially interfacial tension) and the terstices size of the packing. Whereas roplets much smaller than the interices size may pass through the packing nchanged, larger drops may obtain a haracteristic size. However, large lobules have been observed to form and ow through the packing with both high d low interfacial tension systems. imilar phenomena presumably occur in e Wirz Column in which the central rbines are enclosed by such packing 5). A study is being made of hydroynamics in packings of this type for both ntinuous phase and dispersed phase etted packings but droplet behaviour is idently complex. Universal correlaons such as those well established for nventional packings above a certain itical size are unlikely.

Other facets of droplet phenomena of actical importance are those associated ith separation at the dispersed-continuus phase boundary. Rapid coalescence then desirable so that the height of lumn allocated to this duty can be inimised (and to avoid the need for ternal settlers). Alternatively, if paration is incomplete a proportion of e continuous phase will be removed ith the 'bulk' dispersed phase: this will sult in reduced efficiency, capacity and ss of solvent. Under certain conditions, ., small mean drop diameters which ise with low interfacial tension systems d turbulent agitation such as in an ldshue-Rushton Column, phase separan may constitute the limiting factor on pacity. This is analogous to what was d earlier regarding mixer-settlers. pparently phase separation may result edominantly from either interdroplet alescence or coalescence of drops at a ne interface. Some work is in progress establish how the various parameters termine the mechanism and therefore time needed for separation in an unagitated column. The situation in agitated columns is more complex however due to the wider drop size distribution and because it is impracticable to completely isolate the settling zone from extraneous agitation effects even when calming grids are installed. In practice, separation efficiency is profoundly influenced by the purity of the dispersed phase, in particular the presence of any surfactants, by the presence of suspended solids and also by the direction of any residual solute transfer.

Probably the most serious problem in the analysis and design of contactors is that it is never possible to fully account for the significant effect which the presence of trace impurities can have on droplet characteristics. Trace materials with surfactant properties result in smaller droplets giving a large interfacial area and, in some cases at least (3), a considerable increase in extraction efficiency. However, this is accompanied by a serious reduction in capacity. For example, Reman has described a 2.5 times lower capacity encountered in a furfural extraction process due to pitch contamination. Clearly it is important to incorporate flexibility into contactor design by having a variable speed impeller drive. Indeed in another case a suitable dispersion was apparently produced in an R.D.C. at zero rotor speed (3).

In view of the incomplete state of knowledge concerning the droplet coalescence, break-up and mass transfer phenomena which have been briefly described above it is necessary to supplement the design procedure by various empiricisms (13, 27). The situation is indeed so complex that this may always be the case. Nevertheless it is from the classical studies of discrete droplet behaviour, and their more recent extension to the behaviour of swarms of croplets, that a proper understanding of operating parameters in agitated contactors will be gained.

In addition to those already mentioned, several useful areas of work are apparent.

Further study of the rates of interaction between drops of unequal size, for example, would assist in explaining the wide distribution of drop sizes found in an agitated-coalescing environment. This would also complement studies of coalescence at a plane interface in defining the mechanism of phase separation and hence aid the systematic design of settler sections. Further work is also needed to elucidate axial mixing (18) and mass transfer processes for drop populations in continuous columns.

SYMBOLS USED

A = interfacial area
C = concentration
D = column diameter
d = drop diameter
d_{vs} = average surface — volume drop diameter
E = rate of energy dissipation
H.T.U. = height of a transfer unit
K = overall mass transfer coefficient
N = impeller speed
R = impeller diameter
V = superficial liquid velocity
V_N = characteristic drop velocity
X = fractional hold-up of dispersed phase
ρ = density
σ = interfacial tension.

Subscripts

c = continuous phase
d = dispersed phase.

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Paper to be presented to the 3rd CHISA Congress,

Marianske Lazne, September 1969

Droplet Characteristics in Agitated Extraction Columns

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The wide field of application, characteristics and developments in the design of agitated extraction columns have recently been reviewed (1). In these contactors, continuous countercurrent flow between the phases is established by gravity but mechanical energy is applied to achieve controlled dispersion and high mass transfer rates. Either a pulsing device or a rotating agitator may be employed. Because of the technical and economic limitations of pulsed columns only established rotary agitated designs will be considered in this paper. These are the Rotating Disc Contactor (2,3) and its modification the Assymetric R.D.C. (5,26), the Scheibel Column (6,7) and the Oldshue-Rushton Column (8).

The basic differences in design between these columns are illustrated in Figure 1. The R.D.C. comprises a column with equispaced horizontal stator rings, each compartment being agitated by a centrally located flat disc. In the A.R.D.C. the agitated compartments are off-centre and phase flows occur only via a special transfer section. The Scheibel Column has agitated compartments alternated with sections containing open-knitted mesh packings. Under certain circumstances these packings may be omitted. The column is then in many respects similar to the Oldshue-Rushton Column. Turbine agitators are employed in both designs.

For any column the solute transfer process and volumetric capacity are primarily dependent on the size distribution of the dispersed phase droplets and the extent of droplet interaction. It is implicit in rigorous design procedures that droplet interaction is negligible and a uniform spherical drop size may be defined. Dependent on column geometry and operating conditions either or both of these assumptions may be invalidated. A wide distribution of drop sizes occurs in agitated extractors and, therefore, in a non-coalescing environment, mass transfer takes place simultaneously by molecular diffusion and for larger droplets, eddy diffusion mechanisms. Droplet coalescence redispersion effects may be of sufficient magnitude to greatly enhance the rate of mass transfer (13). Therefore, serious errors may be introduced into the design procedure, and the application of empirical formulae to allow for the effects of axial mixing is also complicated (4).

The significance of some observed drop phenomena (10) and recent concepts affecting design and analysis (11) have been discussed elsewhere. More information is required however, on the influence of contactor variables on drop size distribution, residence times, interactions and distortions. This paper summarises some new evidence regarding droplet break-up and coalescence phenomena in agitated columns.

Droplet Break-up

In a non-coalescing environment Hinze derived an expression for the maximum droplet diameter which could exist in a homogeneous isotropic turbulence after Kolmogoroff, (28),

$$d_{max}\left(\frac{\rho_C}{\sigma}\right)^{3/5} E^{2/5} = C_1 \qquad (1)$$

Practical application of this equation presupposes a high Reynolds number for the main flow and that its scale L, which is related to the width of the agitator blades, is very much greater than the microscale of the turbulence, i.e. conditions which frequently apply in agitated columns. Only fair correlation was obtained for drop-size measurements in an R.D.C., however. (4)

A similar expression relating break-up to the Weber number has been derived (29).

$$We = \frac{k_1 N^2 R^{4/3} d^{5/3} \rho}{\sigma} \qquad (2)$$

Misek has studied break-up of drops by a rotating disc in the turbulent mixing region (27). With exponents evaluated experimentally the final expression takes the form,

$$\frac{d_{1.2} N^2 R^2 \rho_C}{\sigma \exp(0.0887 \Delta R)} = 16.3\left(\frac{H}{D}\right)^{0.46} \qquad (3)$$

On diminishing the mixing intensity below a critical value of the Reynolds number, $(Re)_{crit} \simeq 6 \times 10^4$, splitting of drops is deemed to occur in a transitive region in which viscous shear forces have some effect. The expression then valid is (17),

$$\frac{d_{1.2} N^2 R^2 \rho_C}{\sigma \exp(0.0887 \Delta R)} = 1.345 \times 10^{-6} \left(\frac{R^2 N \rho_C}{\mu_C}\right)^{1.42} \qquad (4)$$

Further decrease in mixing intensity into the laminar region results in the vertical droplet velocity becoming significant in that break-up occurs mainly by successive impacts on the stators and discs. By analogy with break-up in packings, therefore, for Reynolds numbers below 10^4 (17),

$$d = 0.38 \left(\frac{\sigma}{\Delta\rho . g}\right)^{0.5} \qquad (5)$$

These equations are claimed to enable prediction of drop sizes with a fair degree of success, albeit under acollosive conditions. Under actual column operating conditions, however, droplets which travel in all directions may collide and coalesce.

For any application, an optimum agitator speed, corresponding to maximum extraction efficiency, exists at any given capacity. This results from a balance between the optimum droplet size for the mass transfer process and the tendency for axial mixing to increase with smaller drop sizes and increased turbulence in the continuous phase. The presence of an optimum speed is well established in the various contactors (3,14,8,7).

Relatively little data are available on the existence of a minimum speed for proper functioning of the various columns. A minimum disc tip speed of 300 ft/min has been recommended in an R.D.C. however to avoid droplets being trapped beneath the rotors and stators (15).

Droplet Coalescence

Phenomena affecting droplet coalescence, and hence phase separation, at the dispersed-continuous phase interface are outside the scope of this paper (18). However, the extent to which interdroplet coalescence occurs in a column is of fundamental importance since, in conjunction with subsequent break-up by turbulence, it may lead to enhancement of mass transfer rates and affect the drop size distribution and hence mean droplet velocity.

By analogy with droplet coalescence at a plane interface, droplet-droplet coalescence can be deduced to occur in two stages, viz. a drainage of continuous phase from between elements until a critical film thickness is attained and b. rupture of the critical film. Therefore, the speed of coalescence is dependent on the forces impelling droplets towards each other, the resistance to liquid film drainage and the thickness and ultimate strength of the critical film. Common experience is that the presence of traces of grease or dirt aid droplet interaction whereas surfactants profoundly decrease the tendency for coalescence. In systems in which mass transfer is taking place, coalescence is favoured when the direction of transfer is from the dispersed to the continuous phase and hindered when it is in the opposite direction (4,9,19,22). Therefore, hydrodynamic correlations which are normally determined in the absence of mass transfer may require considerable modification in practice. Further, since the effect is usually a function of solute concentration, mean drop size d_{vs} should be expected to vary with contactor height.

In a turbulent flow field, drops undergoing the drainage process stand a greater chance of being separated; conversely, the energy of impact is high. Howarth has derived an equation giving the frequency of coalescence of uniformly sized spherical drops in an infinite homogeneous isotropic turbulence (21). This gave good agreement with experimental data for water dispersed in toluene in a stirred tank, Despite this, the large number of assumptions and variables involved limit its application to real systems.

There are little data on coalescence in agitated columns. In a pilot-scale R.D.C. with the system kerozine-water, Davies et al (9) found that drop-drop coalescence was not significant. However, this was with phase ratios of 12 to 16:1 compared with the 0.5 to 4:1 used in practice. However, Misek assumed that a dispersion can be characterised by a hydraulic mean drop diameter, that these drops exactly followed the turbulent fluctuations in the continuous phase and that every collision of droplets resulted in coalescence (20). Then for coalescence in the bulk liquid,

$$\ln \frac{d}{d_o} = K.X.\left(\frac{\sigma}{d_o.\rho}\right)^{0.25} \left(\frac{D}{Y}\right)^{0.5} = Z.X \qquad (6)$$

Coefficient Z was determined indirectly based on phase flow-rate measurements using equation (11) below. Drop size d_o was calculated from the terminal falling velocities of rigid spheres.

Fair agreement was obtained with this equation for a number of binary systems in various column designs; K was found to have the value of 1.59×10^{-2} independent of the type of agitator.

It is questionable, however, whether coalescence characteristics in columns as different in operation as those considered here can, in fact, be described by a single equation. For example, equation 6 makes no allowance for the known variation in ease of coalescence with drop size.

In a fully baffled, turbine agitated continuous flow system the mean droplet size d_{vs} at constant impeller power has been shown to increase linearly with hold-up due to coalescence effects (24), i.e.

$$d_{vs} = d_{vs}^{o} + mx \qquad (7)$$

Over a range of aqueous-organic solvent systems the term m was constant for a given system, regardless of power level, and could be correlated with physical properties by the expression

$$m = 1.18 \left[\left(\frac{\sigma^2}{\mu_C^{2} \cdot g}\right) \left(\frac{\Delta\rho\,\sigma^3}{\mu_C^{4} \cdot g}\right)^{-0.62} \left(\frac{\Delta\rho}{\rho_C}\right)^{0.05} \right] \qquad (8)$$

An expression was also obtained for the correlation of the droplet size at zero hold-up d_{vs}^{o},

$$\left(\frac{d_{vs}^{o\,3}\rho_C^{\,2}g}{\mu_C^{\,2}}\right) = 29.0\left(\frac{P_v^{\,3}g_C^{\,3}}{\rho_C^{\,2}\mu_C g^4}\right)^{-0.32}\left(\frac{\rho_C\sigma^3}{\mu_C^{\,4}\cdot g}\right)^{0.14} \qquad (9)$$

These equations may offer a means of predicting the mean drop-size in an Oldshue-Rushton Column (23).

It is recognised that any packing rotors or stators should be constructed from materials preferentially wetted by the continuous phase. However, in practice, a film of dispersed phase often builds up with time on internal surfaces. For example, since most metals are not strongly hydrophilic build-up occurs in any application with an organic dispersed phase and aqueous continuous phase in a steel column. Such effects can arise, or be aggravated, due to the presence of oil or dirt deposits. Coalescence effects arising in this way are then superimposed upon any droplet-droplet coalescence in the bulk continuous phase.

The effect is especially important in the Scheibel Column containing small interstices size packed sections. The extent to which coalescence occurs in these sections is dependent on drop size, the physical properties of the system and the material of construction and interstices size of the packing.

Hydrodynamics

For any system at a given column geometry and rotor speed the phase flowrates and hold-up can be correlated by an equation of the form

$$\frac{V_d}{X} + \frac{V_C}{(1-x)} = \overline{V}_N\,(1 - X) \qquad (10)$$

where $\overline{V}_N$, the mean vertical component of velocity of the droplets with respect to the continuous phase can be correlated in terms of the system properties and operating parameters (22,15,23).

In order to allow for coalescence and subsequent break-up of droplets in the contactor, Misek proposed a modified form of this equation (30).

$$\frac{V_d}{X} + \frac{V_C}{1-X} = \overline{V}\,(1 - X)\,\exp\left(X(\frac{Z}{M} - 4.1)\right) \qquad (11)$$

where Z is the coalescence factor referred to previously. This was claimed to give a good correlation of results for the R.D.C. (17).

In the present work, drop phenomena have been studied in mutually saturated systems in the absence of mass transfer. Toluene, methyl isobutyl ketone, or 2.2.4 trimethylpentane constituted the dispersed phase with water as the continuous phase in all cases.

Experimental Study

Equipment

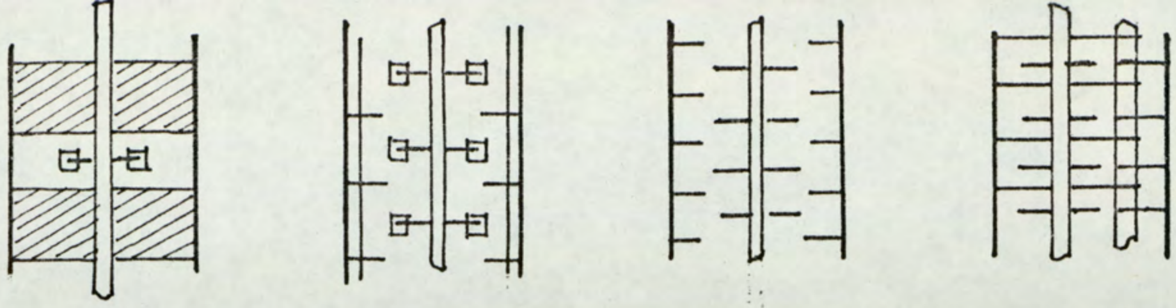
The test section consisted of a 6 inch length of 3 inch nominal bore glass pipe with a settling zone comprising a 4 inch to 3 inch reducer and containing a disc of 12 mesh steel gauze. The dispersed phase was introduced via a disperser located centrally in a similar reducer. This disperser had interchangeable perforated plates and facilitated the injection of presized dispersed phase droplets into the contactor section. The agitator shaft was $\frac{3}{8}$ inch stainless steel rod flexibly coupled to a 1/30 h.p. electric motor. Speed was controlled by a voltage regulator and indicated on a tachometer permanently coupled to the shaft.

Temperature control of both feeds was achieved by passage through coils immersed in a thermostatically controlled water bath. The column section was shielded on three sides to eliminate extraneous draughts. Control was generally within ± 1°C at 20°C.

20 litre glass aspirators were employed as phase reservoirs and receivers. Transfer was by means of two stainless steel Stuart No. 10 centrifugal pumps. Flow was indicated on independently calibrated rotameters. All interconnecting piping consisted of 5/16 inch inert p.t.f.e. tubing.

The R.D.C. was of standard geometry and design (15,17) dimensions of the other sections were scaled geometrically from the original designs, (6,8). The equipment is shown in Figure 2. All column internals were of brass. Packing for the Scheibel section was stainless steel type 9036 supplied by Knitmesh Ltd.

As part of the study and to determine whether each column section functioned in the



Scheibel

Oldshue-Rushton

R.D.C.

A.R.D.C.

Figure 1. Agitated Column Contactors

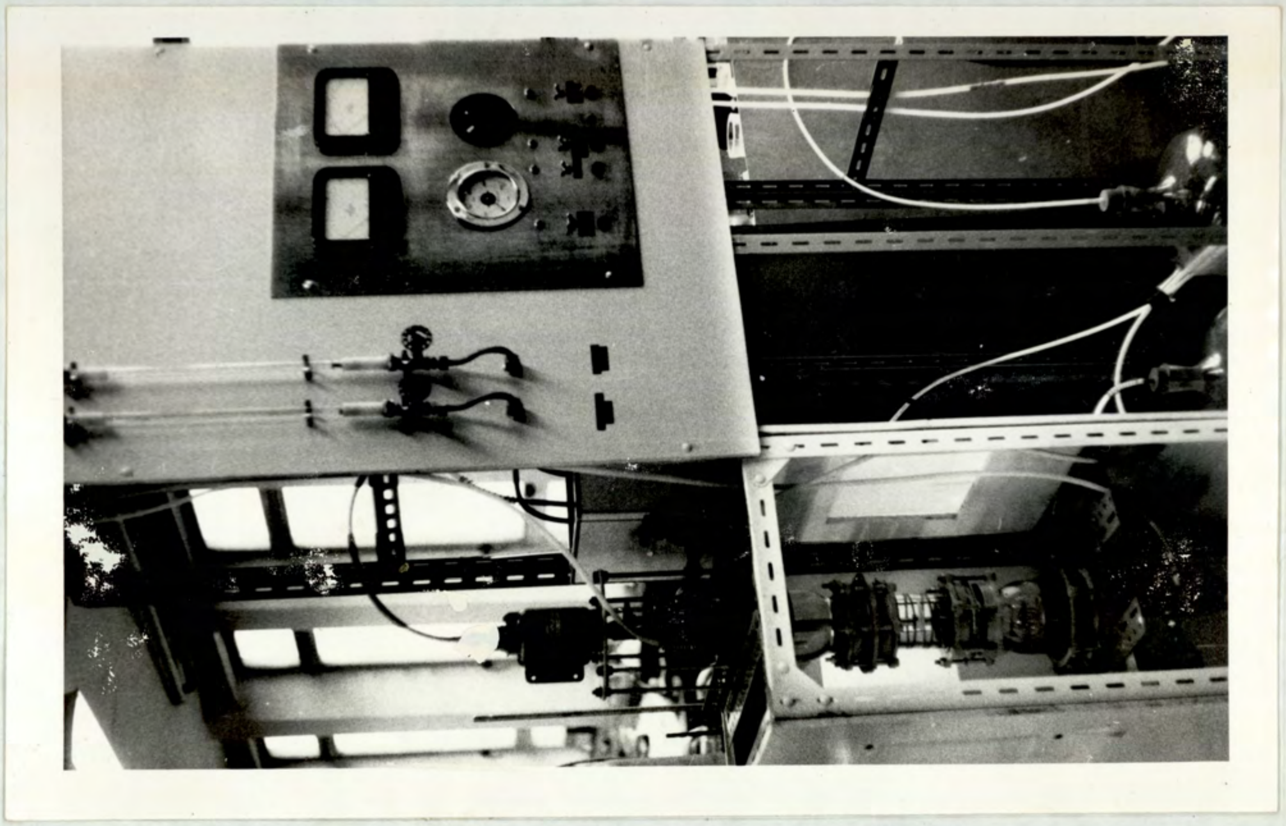


Figure 2. Experimental Equipment

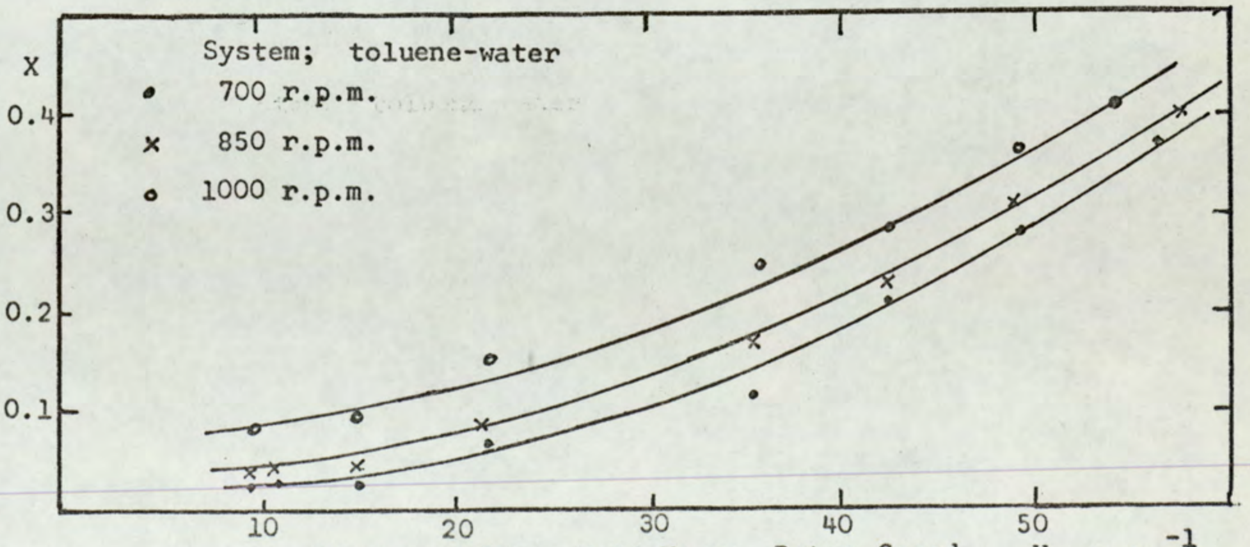


Figure 3. Dispersed Phase Hold-Up vs. Rotor Speed, R.D.C. section.

accepted manner, a series of dispersed phase hold-up, volumetric capacity and drop-size determinations were made and compared with the results of other workers.

(a) Hold-up

The effect of rotor speed on point hold-up values was determined over a range of flow rates by the rapid withdrawal of triplicate 25 c.c. samples via a 3/16 inch polythene sampling tube. In the R.D.C. samples were taken from a position corresponding to midway between the rotor and upper stator ring and between S/2 and R/2 from the centre and hence could be taken as characteristic (4). Samples were taken from a similar position in the Oldshue-Rushton column. It was impractible to determine a characteristic hold-up for the agitated Scheibel compartment due to extraneous droplet break-up effects described later.

Typical results are shown in Figure 3. With the R.D.C. the rate of increase of hold-up with dispersed phase flow rate altered at a loading point corresponding to about 40-50% of the flooding point. This and quantitative values of hold-up were in general agreement with the results of other workers (22,15). For the Oldshue-Rushton column graphs of dispersed phase hold-up against throughput were of similar form to Figure 3 but, as would be expected, hold-ups were of a higher order of magnitude.

(b) Volumetric Capacity

Capacities were determined by establishing steady conditions with flow of both phases at a fixed rotor speed and then incrementally increasing the dispersed phase flowrate up to the floodpoint. Triplicate observations were made for each condition. Typical results are shown in figure 4.

For the system toluene-water in the R.D.C., after allowance for improved column geometry by means of the restriction factor C_R, capacity at 1000 r.p.m. was 15% higher than that obtained by Logsdail et. al. (22). This may be explained by the different criteria taken to characterise true flooding. Throughout this work, complete rejection of the dispersed phase as a dense layer of droplets was the criteria. It was confirmed that in all columns this was proceeded by intense back-mixing (15).

In general, the Oldshue-Rushton column was found to have a similar order of capacity to the R.D.C. within its practical range of operation.

Droplet Size Distribution

Drop phenomena and size distribution were recorded photographically. Errors due to magnification and distortion caused by viewing through the glass column were found to be negligible except at the extreme edges of the column.

Still photographs were taken using a Miranda Sensorex camera with a telephoto lens. Diffused rear lighting was provided by two 500 watt photoflood lamps. Kodak TRI-X film was used throughout with 0.001 seconds exposure time. Triplicate or duplicate photographs of each event were taken at intervals and confirmed the attainment of hydrodynamic equilibrium. Drop size measurements were taken either from projection or from prints reproduced professionally so as to show all droplets in focus. A Beaulieu camera was used for cine photography at 64 frames per second. This speed was suitable for recording general hydrodynamic phenomena only. A Wollensak Fastax WF 14T camera with a Fastax- Raptor 152 mm. f2.7 telephoto lens and extension tubes as required was used for high speed cine photography at 4000 frames per second. This was necessary since the time taken for drops in the size range studied to coalesce is of the order of 1 millisecond. Lighting was supplemented by a further four photoflood lamps. Details of the techniques used with this camera have been given by Lawson (18).

Only a selection of results can be reported here; a full analysis of the data will be published elsewhere. Most difficulty was encountered with the system M.I.B. k-water due to the myriad of small drops.

In the R.D.C. droplet break-up was limited to the transitive and laminar regions by virtue of equipment size. Over the range of interfacial tensions studied a critical minimum speed was found to exist below which a layer of dispersed phase droplets built up beneath the rotor disc as shown in Figure 5. Contrary to previous reports (15) droplets did not build up beneath the stators at finite rotor speeds with the exception of a monolayer observed in runs with the trimethylpentane-water system below $Re \doteq 6 \times 10^3$. Specification of a minimum tip speed for one rotor diameter

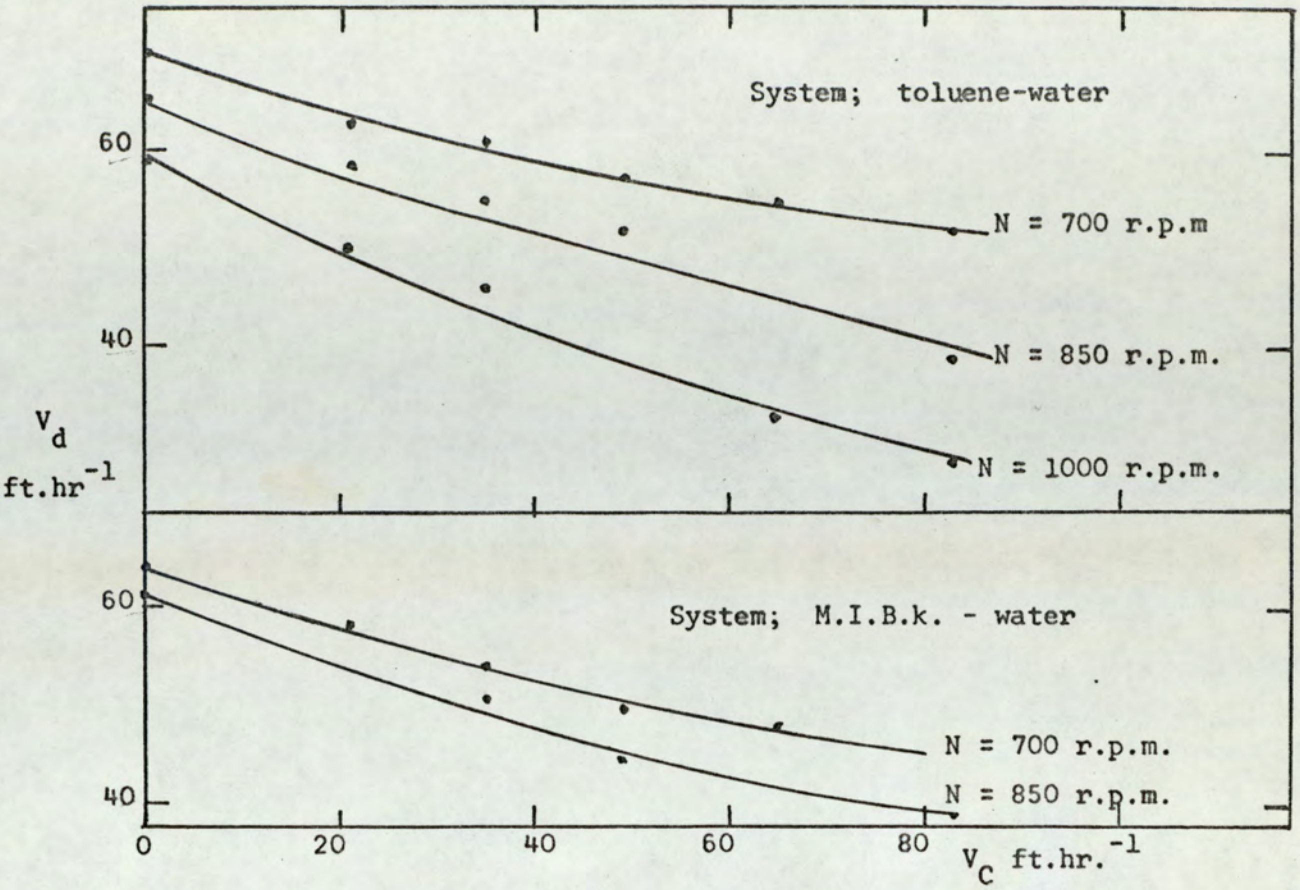


Figure 4. Flooding curves, R.D.C. section.

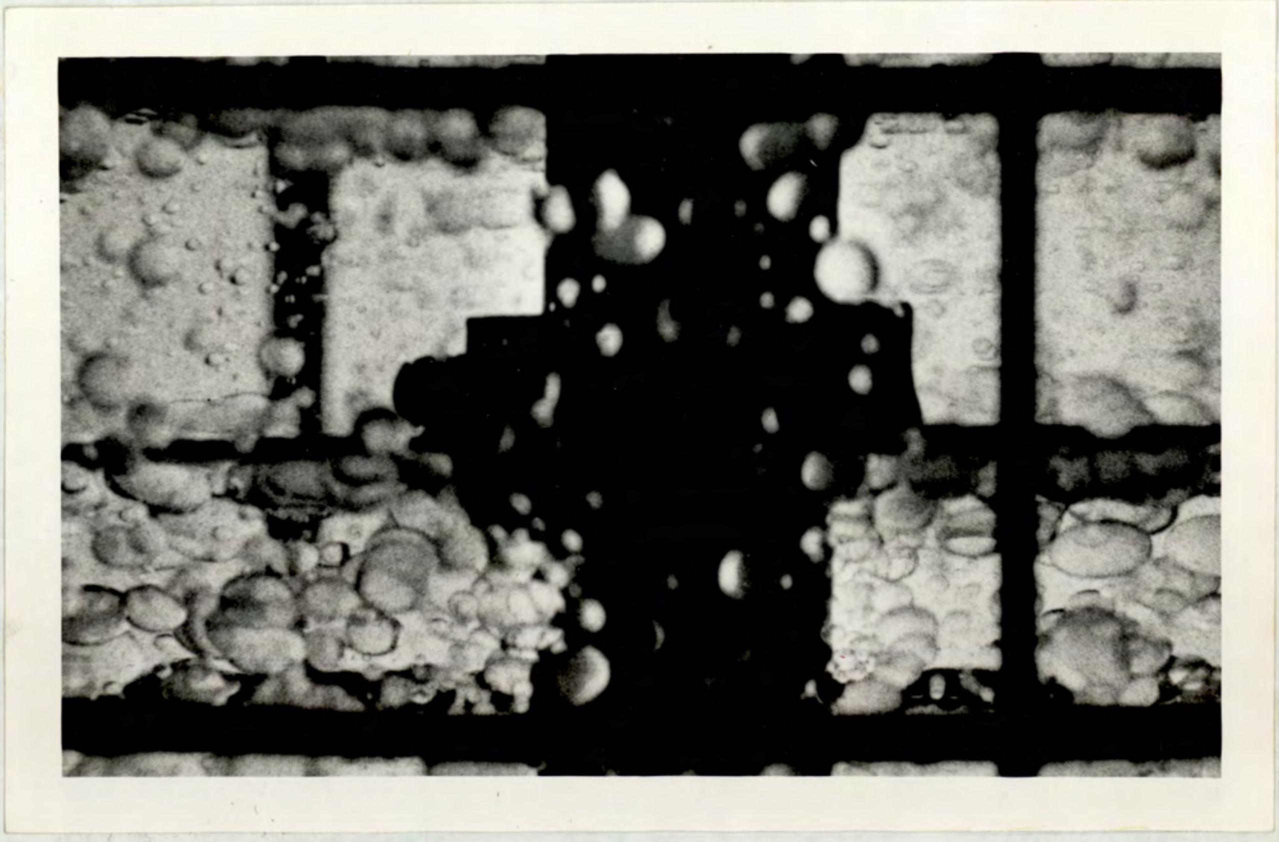


Figure 5. Build-up of droplets beneath the disc in an R.D.C. below the critical minimum speed.

does not serve any purpose. In this work with an aqueous continuous phase the phenomena occurred at a Reynolds number, $\frac{R^2 N \rho_C}{\mu_C} \approx 1.75 \times 10^4$ independently of hold-up, but as would be expected was slightly dependent on interfacial tension. This delineates the change in break-up mechanism in the R.D.C. which according to Misek occurs in the region Re $\approx 10^4$ (15). In this region the mean drop sizes were for all practical purposes independent of hold-up and were in agreement with the correlation proposed by Misek as shown in Figure 6. However, visual comparison of drop size distributions with those when the rotor disc was stationery confirmed that break-up was not due solely to impact on the disc and stator ring. From high speed cine film and observations using a coloured dispersed phase droplets were found to maintain their identities. The population beneath the disc changed by droplets sliding to the periphery. A significant proportion of the droplet population spent some time beneath the rotor disc before being thrown off with insufficient momentum to impinge on the column wall. At these low speeds, the turbulence existing in the continuous phase was insufficient to cause shearing and a smaller spread in drop size resulted. The distribution of drop sizes below the critical speed and at some higher speed at corresponding hold-ups with, for example, the M.I.B.K.-water system are shown in Figure 7. The point of onset of this phenomena could be significantly reduced by the introduction of extraneous turbulence beneath the disc using a small protrusion from the shaft.

At increased speeds, the overall tendency was for drop sizes to increase over the whole spectrum with hold-up as found by Olney (31). As will be discussed later, acollisive conditions were found to exist in the R.D.C. from which it follows that the specific power input $\frac{\bar{\rho} N^3 R^5}{\rho_D . X . H . D^2}$ is the major factor in the dispersion process. Therefore correlations such as Equation 4 would appear capable at best of predicting only the order of mean drop size. The variation with hold-up of some of the d_{vs} values determined in this work are represented in Figure 8.

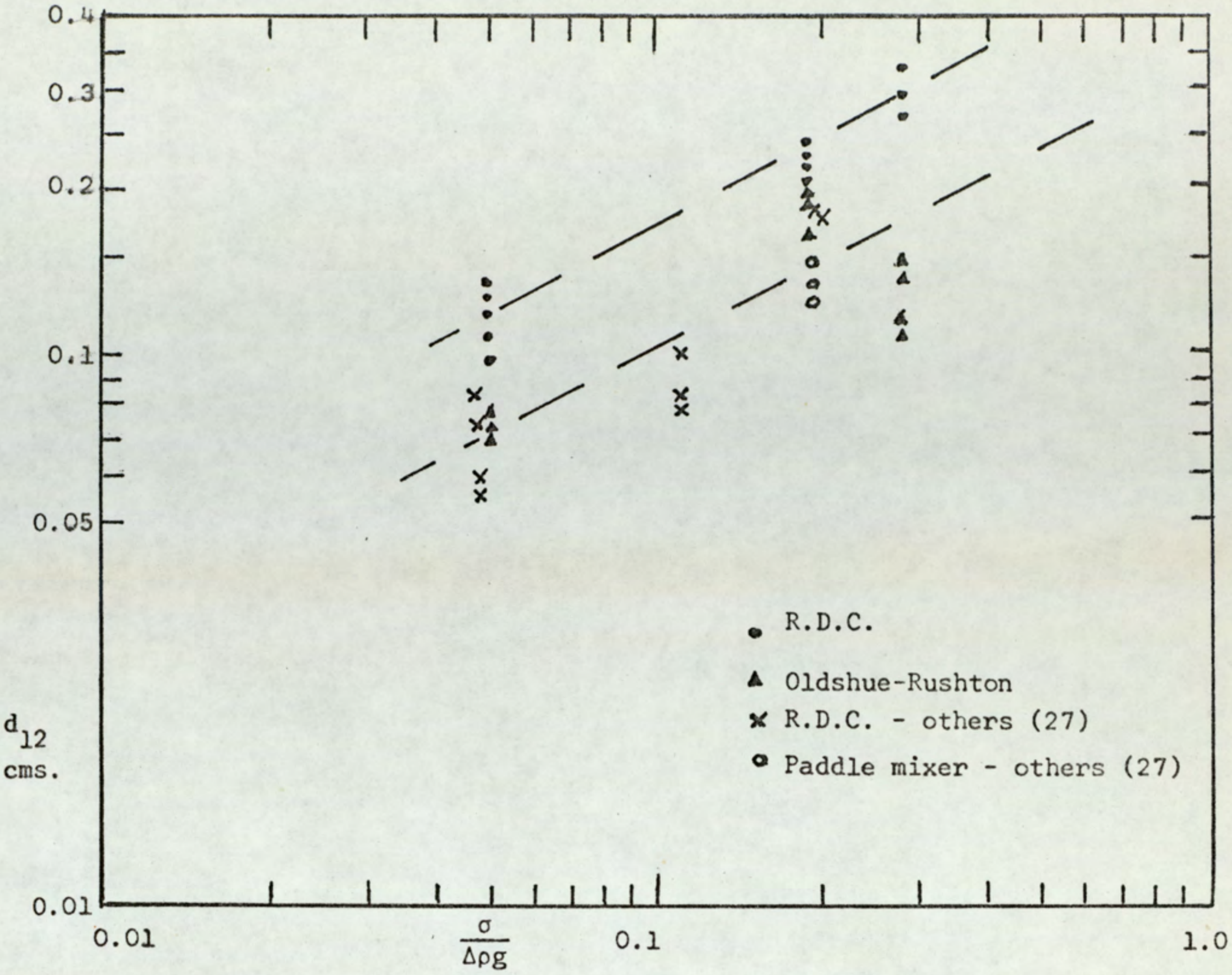


Figure 6. Correlation of hydraulic mean drop diameters - Laminar Regions.

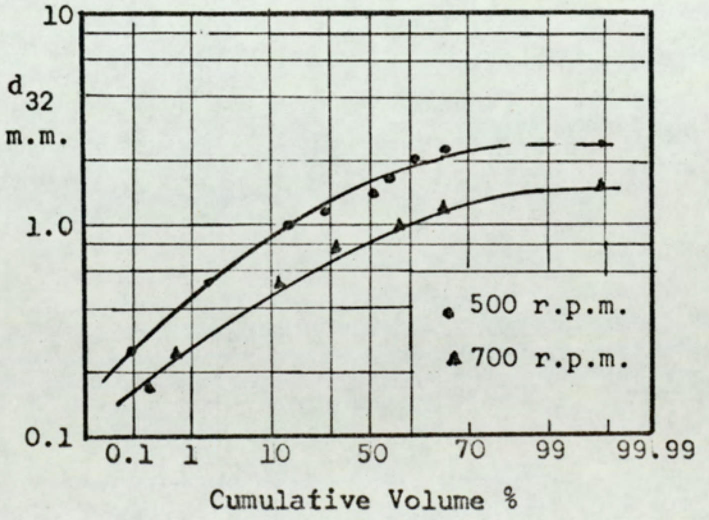


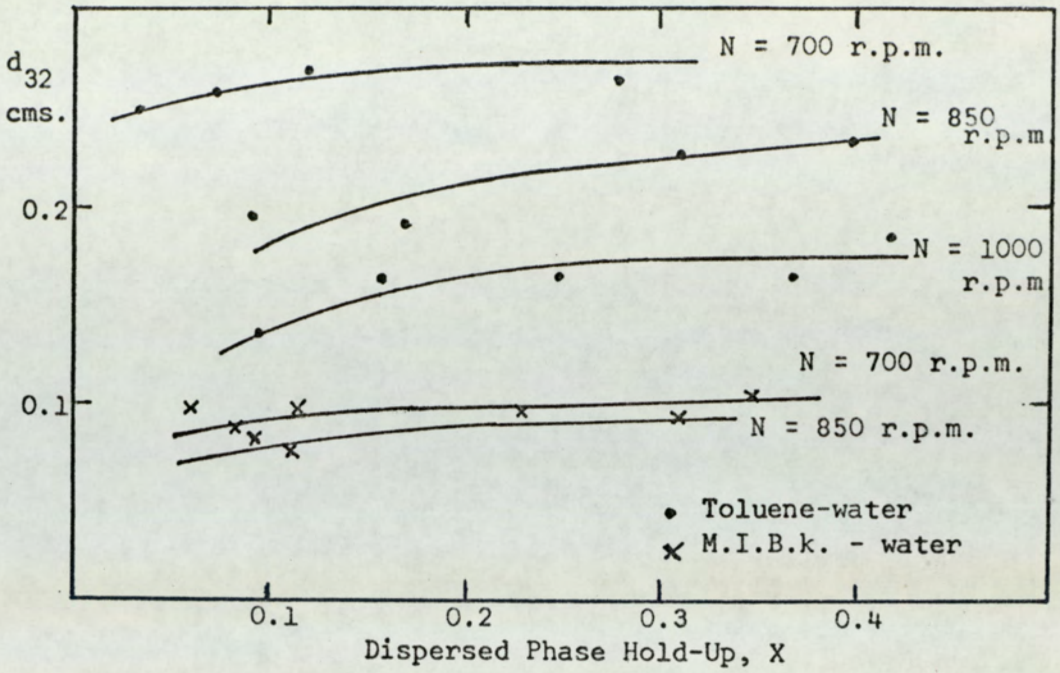
Figure 7. Drop size distributions above and below $Re \approx 1.75 \times 10^4$ at corresponding hold-ups, R.D.C. (M.I.B.k - water, $X \geq 0.08$).

Observations on the Oldshue-Rushton section described later showed that at practicable operating speeds, such that the agitator functioned as a turbine, a coalescence - breakup mechanism predominated down to low hold-ups. Therefore, drop size measurements were analysed on the basis of the correlation by Thornton (24). Some results are plotted in Figure 9.

Since at low agitator speeds and hold-ups conditions were accollisive and drop sizes were largely independent of hold-up it was assumed that break-up was not due to dynamic pressure fluctuations. This was subsequently confirmed by cine photography. Therefore, drop sizes in this region were compared with those predicted by Equation 5; some results are shown in Figure 6. This suggests that the transition observed at $Re \approx 3.8 \times 10^3$ from the tangential flow pattern of a paddle to the radial flow pattern of a turbine is analagous to the transition reported for break-up in the R.D.C. At agitator speeds much below this value turbulence was insufficient to completely remove droplets from between the stators and vertical baffles. This has also been observ by Bibaud and Treybal (23). In the present work this phenomena was found to preclude the measurement of a characteristic dispersed phase hold-up and to create a condition conducive to premature flooding. It was observed that the column had a slightly greater capacity at small finite rotor speeds than at zero rotor speed, the effect of agitation being to thin the zone of flocculation beneath the stators.

Observation of the Scheibel section with all 3 systems confirmed that the function of the packing was dependent on drop size distribution, i.e. agitator speed (32). At low speeds, when the mean drop size was considerably larger than the interstices size of the packing, the packing acted mainly as a coalescing stage to produce globules which left the packing by drip-point formation. At higher throughputs jetting occurred from preferential areas on the packing surface. These jets were not effectively broken up. At high rotor speeds a proportion of the drop population was still observed to form by shearing off of globules as shown in Figure 10.

This occurred randomly and therefore both mean drop diameter and hold-up in the mixing section varied with time. The maximum drop sizes observed, which were



igure 8. Variation of Sauter mean drop sizes with hold-up; R.D.C. - Transitive region

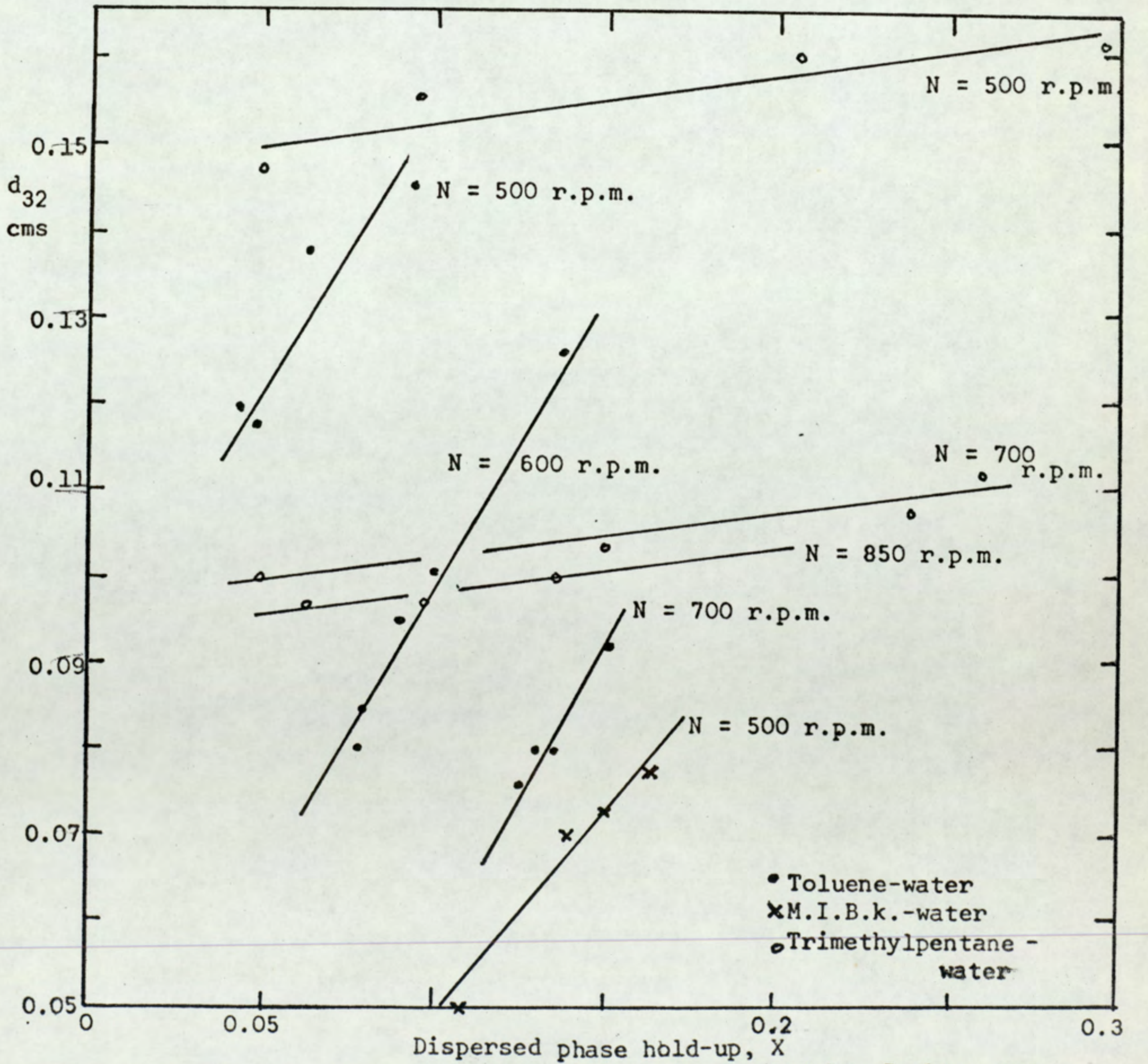


Figure 9. Mean drop size vs. dispersed phase hold-up in the Oldshue-Rushton Column,

considerably larger than in the Oldshue-Rushton section under comparable conditions are plotted against rotor speed in Figure 11. This phenomena was not reported by Honekamp and Burkhart (33).

Wettability of Column Internals

The manner in which wetting effects may cause a reduction in efficiency and capacity was investigated in the three designs.

In the R.D.C. volumetric capacity was compared using alternatively discs of brass and p.t.f.e. with which organic liquids exhibit large contact angles. Some results with the system toluene (dispersed) - water are shown in Figure 12.

The p.t.f.e. discs caused a reduction in capacity of up to 14% which is much less than previously supposed (17). At all rotor speeds a circular element of dispersed phase was attached to the tip of the p.t.f.e. rotor as illustrated in Figure 13. Element width varied with rotor speed and the fact that, at high dispersed phase flows, reduced capacities correlated approximately with the constriction factor $(D^2 - R^2)/D^2$ supported the observation that flooding commenced in the plane of the rotor disc.

A critical minimum speed existed, again not entirely independent of interfacial tension, at which the layer of dispersed phase created by coalescence of droplets beneath the rotor disc thinned so as to be no longer observable. This occurred at a Reynolds number $\simeq 2.3 \times 10^4$; below this globules leaving the disc caused the mean drop size to be larger than predicted by means of Equation 5.

In the Scheibel and Oldshue-Rushton sections it was found that wettability of the turbine agitators, as distinct from the shaft, which could provide a by-pass for the dispersed phase, was not important.

Observations were made on the R.D.C. section with stator rings of p.t.f.e. These provided an explanation for the greatly reduced efficiencies reported for similar conditions by Davies (9). At all rotor speeds gross coalescence of the dispersed toluene phase occurred on the upper and lower surfaces of the stator rings. Large globules of 10 to 15 mm. equivalent diameter left the base of fluid from random positions

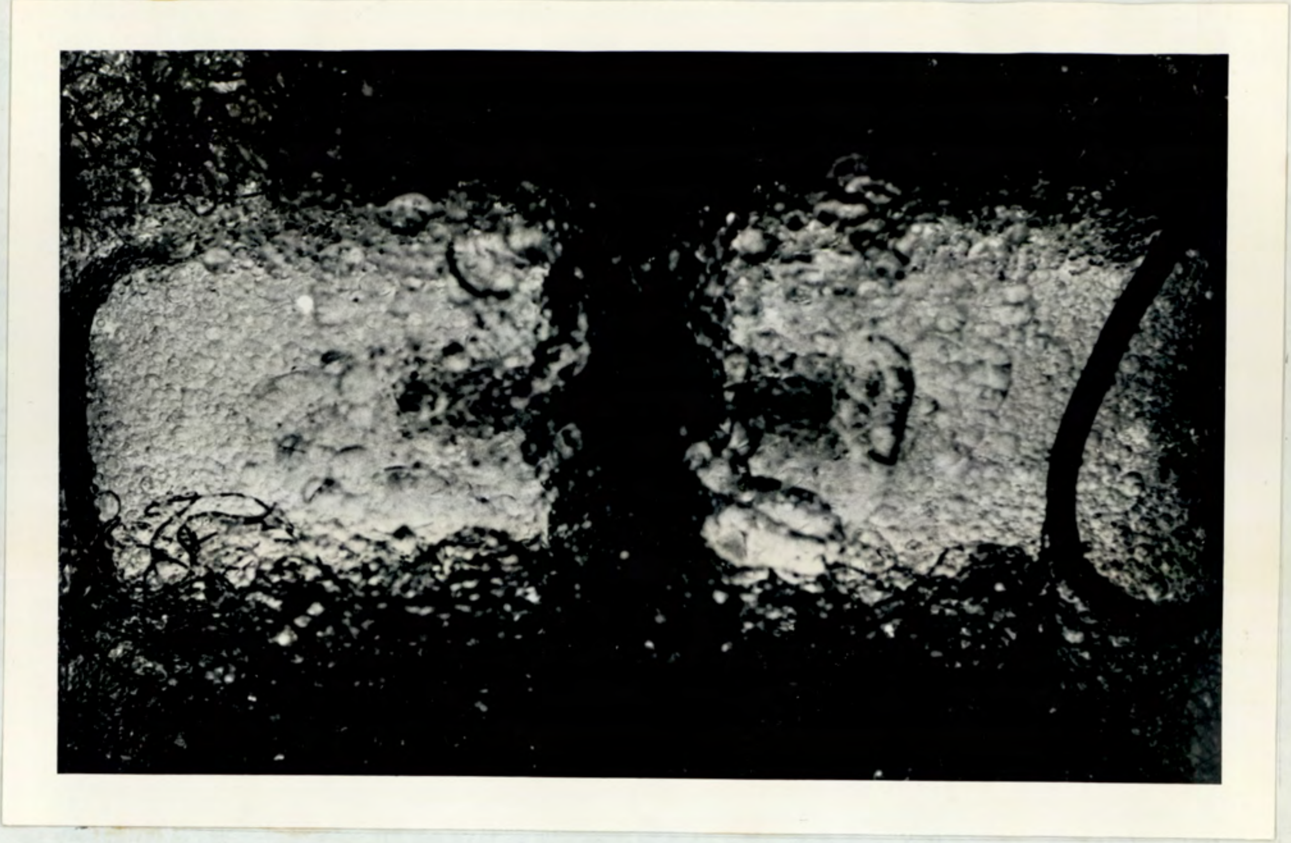
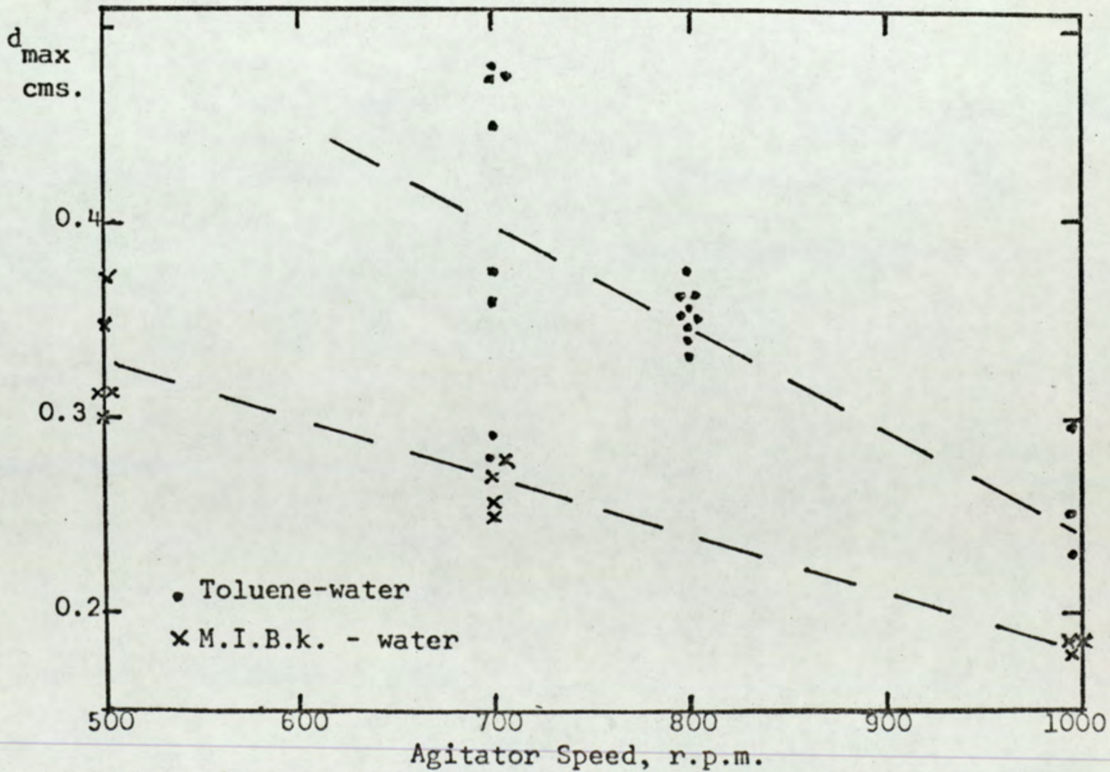


Figure 10. Formation of globules of dispersed phase by shearing off from packing (System; M.I.B.k. - water, N = 500 r.p.m.)



igure 11. Maximum drop-size vs. rotor speed; Scheibel Column.

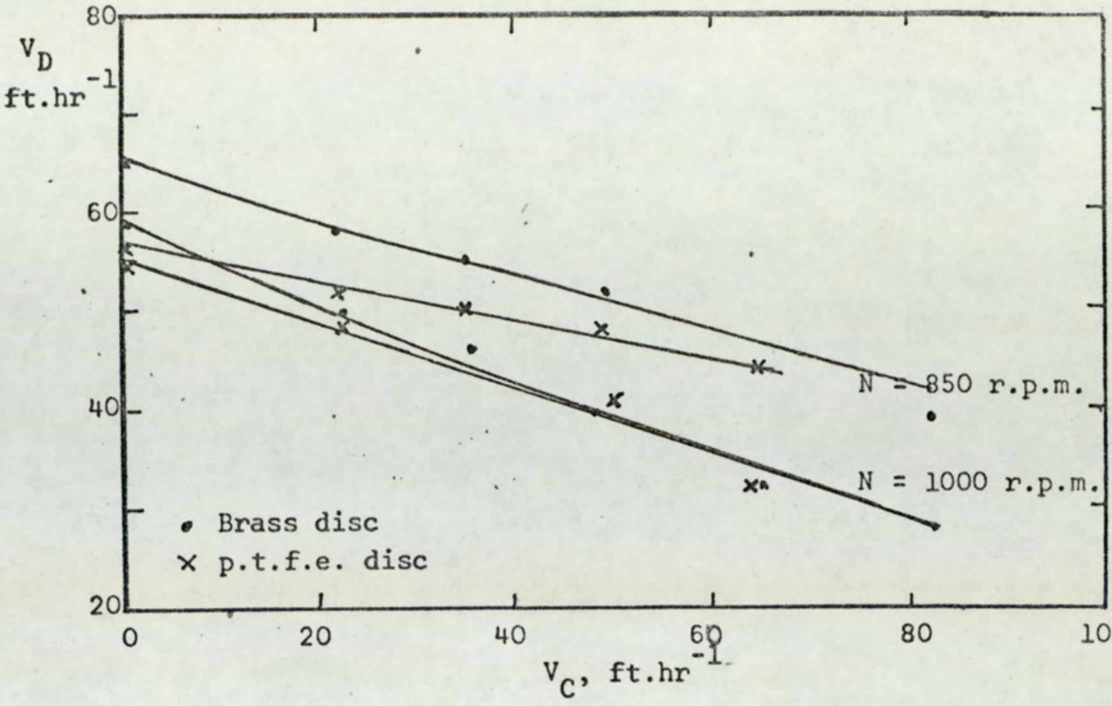


Figure 12. Flooding curves for R.D.C. section (System; toluene-water)

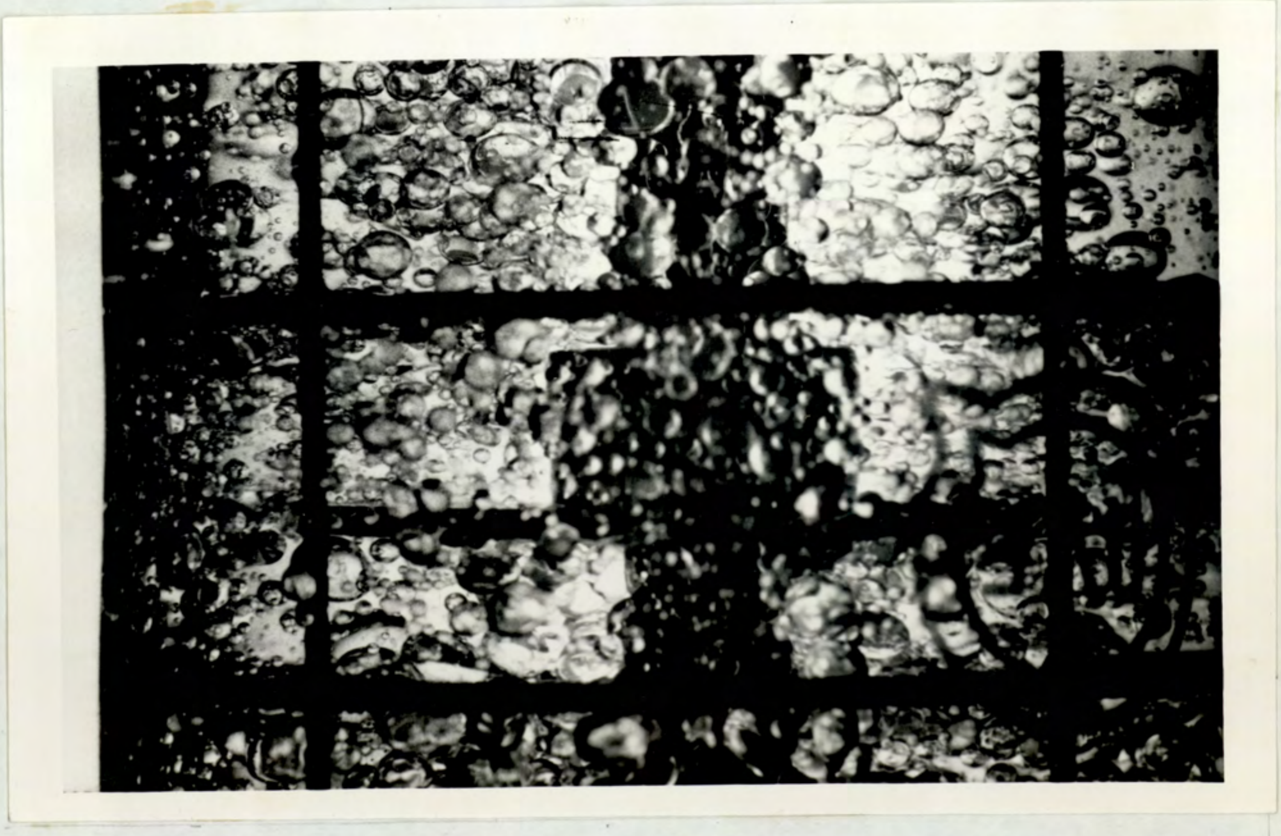


Figure 13. R.D.C. section with p.t.f.e. rotor disc showing attachment of dispersed phase. (System; toluene-water, N = 800 r.p.m.)

by drip-point formation. This mechanism is similar to that described by Haynes et. al. from their work on droplet formation at orifices (12). The size of the globules leaving the stators was apparently independent of rotor speed in the range 500 to 1000 r.p.m. Similar phenomena were reproduced qualitatively in separate runs with a film of oil covering the brass stators and in the Oldshue-Rushton section with p.t.f.e. stator rings as illustrated elsewhere (10). The combined wetting effects in an unclean R.D.C. section are shown in Figure 14.

With the Scheibel column observations were made using polypropylene packing type 9093 supplied by Knitmesh Ltd. At low hold-ups drop phenomena were similar to those reported for stainless steel packing but with drip point formation from an increased number of sites. This is undesirable since it produces random globules of indeterminate size which when ruptured from the packing elements produce small satellite drops. Such secondary drops persisted in the column due to their low settling velocities.

At the higher agitator speeds with increased throughputs prior to flooding a transition point occurred at which the packing surface forming the top and bottom of the agitated compartment became covered with a film of dispersed phase. An emulsion of fine droplets was formed in the agitated compartment. In effect under these conditions the column's mode of operation changed to that of a continuous mixer-settler. Conditions just prior to this transition are illustrated in Figure 15.

Coalescence

The extent of interdroplet coalescence was investigated. A regulated number of coloured drops of substantially the same size as those from the distributor were introduced via a single sharp-edged orifice from a 100 cc. hypodermic syringe. The organic soluble dye used was Calco Oil Red N-1700 which is non-surface active (25). A spread of dye amongst significant numbers of droplets leaving the contact zone was taken as evidence of a droplet break-up-recoalescence mechanism

In the R.D.C., runs were performed with all three systems at constant rotor speeds of 500 r.p.m., 700 r.p.m., 850 r.p.m. and 1000 r.p.m. and with varying flowrates. In no case was drop-drop coalescence found to be significant until flowrates approached the flood points determined earlier. The absence of coalescence was borne out by a study of all the high speed cine film taken, albeit necessarily, at low hold-up values.

Similar observations weremade using the Oldshue-Rushton section. At practical operating speeds, spread of dye was almost complete throughout the dispersed phase at all but the lowest hold-ups. Thus, a coalescence-redispersion mechanism was predominant A study of high speed cine film, necessarily taken at low hold-ups showed, however, that even at a Reynolds Number of 6 x 10^3 droplet break-up and coalescence was localised in the region of the turbines.

A few droplets always maintained their identity, a phenomena also observed by Bibaud and Treybal with the system toluene-water in a 12 stage column (23). At low operating speeds the onset of drop-drop coalescence was not easily determinable due to bulk coalescence effects in the dynamic flocculation zone created beneath the stator rings as described earlier.

A coalescence-redispersion mechanism was also found with all systems in the Scheibel section providing the agitator speed was above a minimum necessary to distribute droplets across the column cross-section.

Conclusions

In the R.D.C. the absence of drop-drop coalescence up to the flood point in systems having a range of interfacial tensions between 9.7 and 55.4 dynes/cm explains the persistence of small droplets when present among drops of all sizes. Since practical column operation is normally at less than 80% of flooding rates then, in the absence, of any special interfacial effects associated with mass transfer, droplet coalescence and redispersion are not significant factors affecting extraction efficiency. Therefore, the column operates as a discrete droplet contacting device and can be designed on this basis. The major problem is then the prediction of mean drop diameters. Based on limited data, the drop size produced in the laminar region, corresponding to Re $<$ 1.75 x 10^4 for a non-dispersed phase wetted disc, is independent of hold-up and is given approximately by Equation 5. Further work isneeded before drop sizes in the transition range can be predicted with confidence by Equation 4. Allowance must be made for the effect of hold-up. In the case ofdispersed phase wetted internals the drop size

Figure 14. Gross wetting effects in an uncleaned R.D.C. column section. (System toluene-water, N = 1000 r.p.m.)

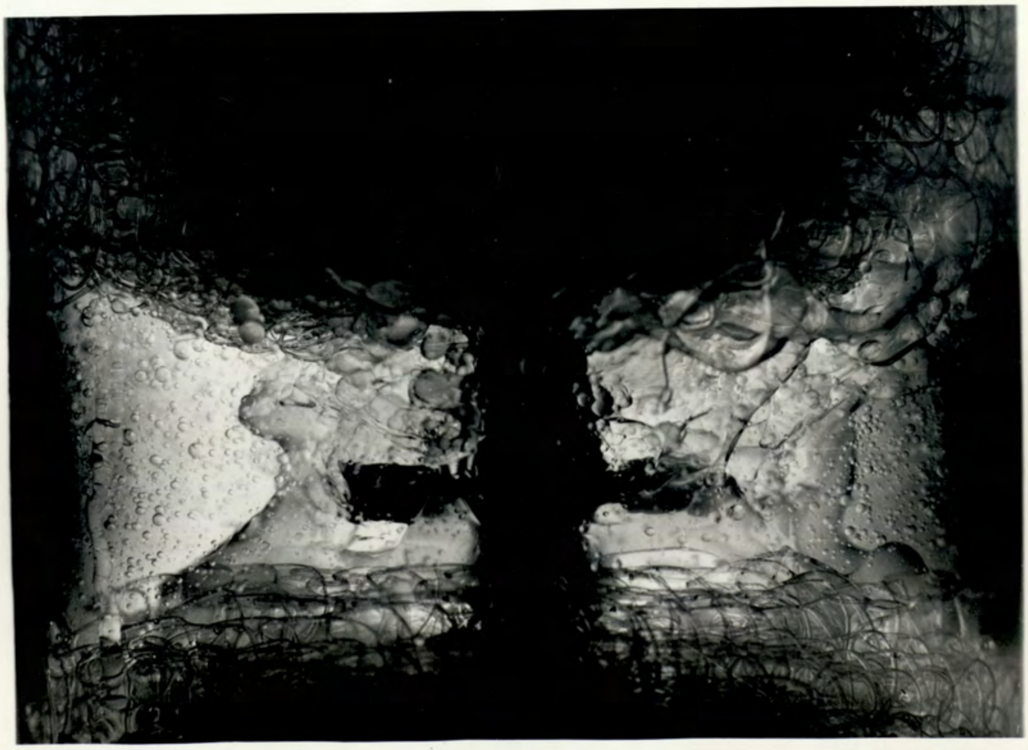


Figure 15. Scheibel column with polypropylene packing wetted by dispersed M.I.B.K. phase.

distribution and hold-up cannot be predicted satisfactorily.

In an Oldshue-Rushton column under normal operating conditions a coalescence-redispersion mechanism predominates. Therefore it is to be expected that, whereas at the lower end of the size distribution range, single droplet data would indicate a molecular transport process for solute transfer the rate in practice should be more in accordance with an eddy diffusional mechanism. This is apparently contradicted by the limited results given by Bibaud and Treybal (23) but these workers predicted drop sizes from the work of Thornton for an agitated vessel (24), under such conditions the mean droplet size is of a different order of magnitude. On the basis of the limited data reported it appears that for $Re < 4 \times 10^3$ the mean drop size is largely independent of hold-up and can be predicted by means of Equation 5. Within the range of normal operating speeds, the data are represented by forms of Equation 7. In practice, the circulating mixing in a compartment produces a high average residence time for dispersed phase droplets and therefore concentration changes between compartments should be approximately stagewise. This is confirmed by the observations of Oldshue (8). Therefore, columns are designed commercially using the H.E.T.S. concept (34).

The work on the Scheibel column indicates that for systems having an interfacial tension $>$ 9.7 dynes/cm, it is desirable to omit the packed sections. No efficiency measurements were made in this work, but Scheibel has also proposed replacement of the packings by baffled calming zones for high interfacial tension or high viscosity systems (16) and similar columns have been designed (1). Within the normal operating range a coalescence-redispersion mechanism has been found to predominate in the Scheibel column. Due to globule formation the mean interfacial area in the agitated zone is unpredictable as indeed is that in the packed sections. Therefore, resort must still be made to the stagewise design procedure proposed elsewhere (35). The problem of estimating volumetric capacity remains (32). This could be eased and some of the undesirable droplet phenomena eliminated by employing $\frac{1}{4}$ inch diameter Berl saddles as packing. This would have a similar order of voidage, i.e. 90%. Established flooding correlations would then be applicable (36). One design employing 15 mm Lessing rings is known. In general, from the phenomena observed with dispersed phase wetted internals it is concluded that when such conditions obtain in practice no reliable estimate can be made of efficiency and volumetric capacity for the column concerned. This is one factor dictating the provision of variable speed agitators on commercial agitated columns. The other reasons are to allow for extraneous phenomena due to surfactants (9) and to provide operating flexibility with regard to phase ratios and terminal concentrations (34). The R.D.C. is probably most flexible in these respects. Such wetting phenomena, together with coalescence problems which may arise at the phase separation interface, may also determine in practice the frequency at which column cleaning is necessary.

Nomenclature

C_R = factor accounting for column restriction, equal to S^2/D^2 or $(D^2-R^2)/D^2$.

D = column diameter

d = diameter of drop

d_o = diameter of uncombined drop

d_{12} = hydraulic mean drop diameter

d_{vs} = average surface-volume drop diameter

d_{32}

d_{max} = maximum drop diameter

E = energy of dissipation per unit mass

H = stator spacing

L = microscale of turbulence

M = velocity exponent

N = impeller speed

P_v = power dissipated per unit volume of vessel

R = impeller diameter

ΔR = distance between agitator and column wall, equal to $(D-R)/2$

S = diameter of stator opening

V = superficial velocity of phase in column

$\overline{V}_N$ = characteristic velocity, i.e. the mean vertical droplet velocity at substantially zero flow rates and at rotor speed N

X = fractional hold-up of dispersed phase

Z = coefficient of coalescence

σ = interfacial tension

ρ = density

μ = viscosity

γ = kinematic viscosity

Subscripts

c = continuous phase

d = dispersed phase.

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APPENDIX 2.

SUMMARY OF MISCELLANEOUS STUDIES.

Column Type	System	Authors
R.D.C.	lubricating oil fraction - oil-acetone/water.	King and Rhodes (91)
R.D.C.	water - phenol - butyl acetate.	Goerz et.al. (117)
R.D.C.	Numerous.	Shtrobel et.al. (118)
R.D.C.	phenol - water - benzene or Fenosolvan.	Misek and Roskos (119)
A.R.D.C.	ammonium sulphate soln. - caprolactam - toluene.	Marek et.al. (26)
R.D.C.	diethylene - glycol - benzene - hexane.	Pechstein et.al. (120)
R.D.C.	water - caprolactam - trichlorcethylene.	Kagan et.al. (105)
R.D.C.	lubricating oil fraction - oil - furfural.	Reman and van de Vusse (109)
R.D.C.	lubricating oil - asphaltic compounds - propane.	Marple et.al. (121)
R.D.C.	ditto	Ditman et.al.(122)
R.D.C.	ditto	Thegze et.al.(123)
R.D.C.	High partition coefficient systems.	Pajack (124)
R.D.C.	diethylene glycol - benzene - heptane.	Charlton (125)

APPENDIX 3.

DETAILS OF KNITMESH PACKINGS.

Wire used is 33 s.w.g. (0.010") in round sections.

Type No.	Density	Surface Area/Unit Volume	Free Volume
Stainless Steel Packing.			
	lb.ft.$^{-1}$	ft.ft.$^{-1}$	%
9030	9	90	98
9036	6	60	98.75
Polypropylene Packing.			
9036	$2\frac{3}{4}$	150	95

Note: The density of the sections employed would be approximately 10% higher than given above because (a) In avoiding an area of weakness in the centre it is impossible to avoid some increase. (b) The sections are made slightly oversize to give a good wall fit and this is a bigger proportion of the total volume (97).

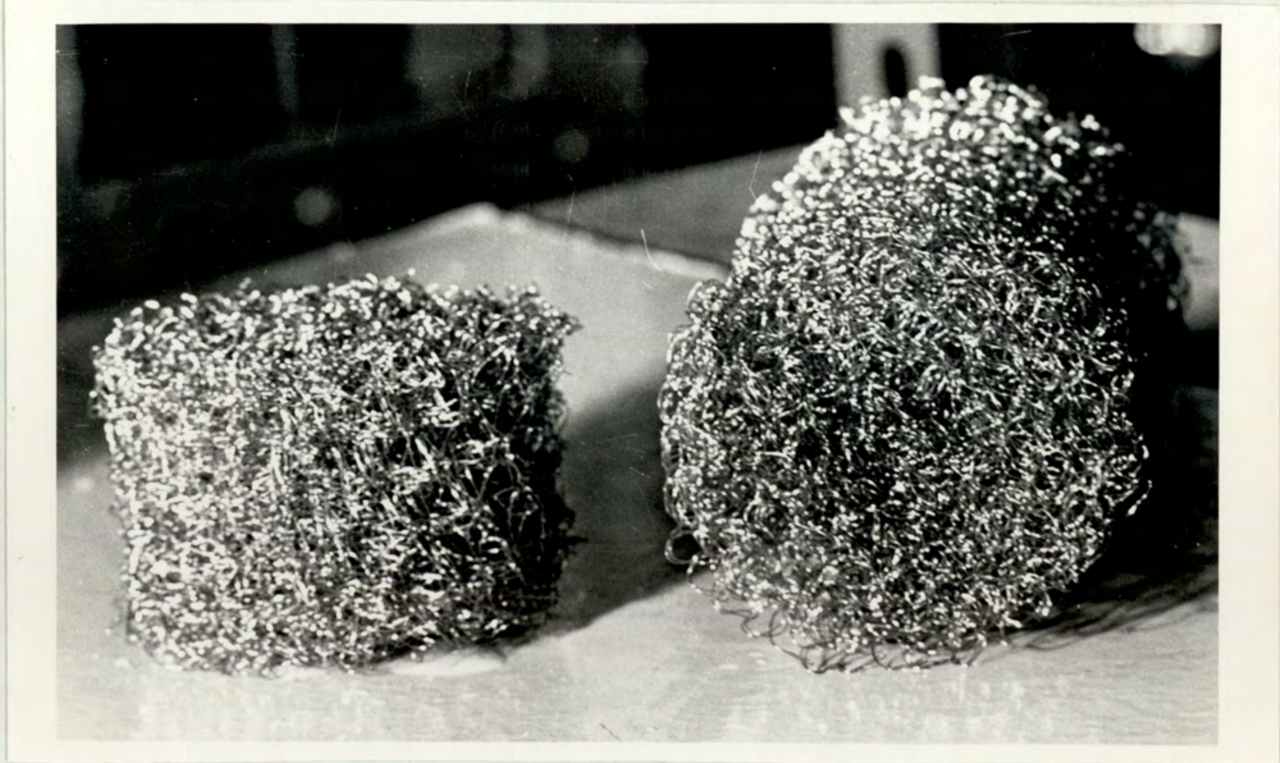

FIGURE 98. KNITMESH PACKING

Stainless Steel, Type 9036.

APPENDIX 4.

PHYSICAL PROPERTIES OF LIQUID-LIQUID SYSTEMS.

The following physical properties have been taken from International Critical Tables.

Interfacial Tension - Dynes/cm.

System		Temperature
Toluene - water	36.1	25°C
M.I.B.K. - water	9.7	20°C
Trimethylpentane - water	55.4	25°C

Density - gm./cc.

Liquid		
Toluene	0.865	20°C
M.I.B.K.	0.80	20°C
Trimethylpentane	0.692	20°C

Viscosity - centipoise

Toluene	0.58	20.6°C
M.I.B.K.	0.30	21.3°C
Trimethylpentane	0.50	25 °C

Materials were purchased to the grades and specifications given below:-

Toluene

Pure Solvent Grade

B. Pt. Range	109° - 112°C
Wt. per ml.	0.857 - 0.864 at 20°C.

Maximum limits of impurities:

Thiophenes and other S compounds	0.0005
Volatile matter	0.005

Methyl-isobutyl Ketone

Minimum Assay Ketone	98%
Boiling Range	95% distils between 114° - 117°C
Refractive index	1.3945 - 1.3963
Free Acid	< 1 ml. $\frac{N}{1}$ %

APPENDIX 4. (Cont'd)

Trimethylpentane

wt. per ml.	0.6
Non-volatile matter	0.005% max.

Conforms to I.P. and A.S.T.M. Specifications

Decon 75 Concentrate used for cleaning was obtained from Medical Pharmaceutical Developments Limited.

APPENDIX 5.

Table 2 - PUMPING EFFECT OF DISCS IN R.D.C. SECTION.

R.P.M.	Head m.m. water	Volts	Amps
	(Average of Duplicate Runs)		
0	0	-	-
200	0	130	0.1
400	0.5	140	0.2
600	1.0	145	0.25
800	1.0	150	0.3
1000	1.0	160	0.35

Table 3 - MAXIMUM DROP SIZES RECORDED IN SCHEIBEL COLUMN.

System: Toluene -water

V_d, ft.h^{-1}	9.5	10.8	15.2	21.6	28.2	35.5	49.0	61.2
N = 700 r.p.m. dmax. m.m.	4.8	3.6	2.8	4.8	4.8	2.9	4.5	3.75
N = 800 r.p.m. dmax. m.m.	3.75	3.6	3.6	3.4	3.3	3.5	3.5	3.6
N = 1000 r.p.m. dmax. m.m.	3.0	2.5	2.3	-	-	-	-	-

System: M.I.B.K. - water

V_d, ft.h^{-1}	10.9	13.0	20.6	27.0	34.8	-	-	-
N = 500 r.p.m. dmax.m.m.	3.75	3.5	3.0	3.15	3.15	-	-	-
N = 700 r.p.m. dmax.m.m.	2.8	2.7	-	2.57	-	-	-	-
N = 1000 r.p.m. dmax.m.m.	1.82	-	-	-	-	-	-	-

System: Trimethylpentane - water

V_d, ft.h^{-1}	13.8	20.3	27.2	41.0	50.8	60.5	68.7	77.5
N = 700 r.p.m. dmax.m.m.	6.2	6.7	4.7	5.1	5.9	5.4	4.8	4.8
N = 850 r.p.m. dmax.m.m.	4.0	4.0	4.2	4.3	3.7	3.7	-	-
N = 1000 r.p.m. dmax.m.m.	2.7	3.2	3.2	2.4	2.4	2.7	-	-

APPENDIX 5. (Cont'd)

Table 5, R.D.C. - DROP SIZE MEASUREMENTS BELOW THE CRITICAL MINIMUM SPEED. (m.m.)

System: Toluene - water

N = 500 r.p.m.

X	d_{max}	d_{min}	d_{12}
-	3.9	0.3	2.93
-	3.7	0.3	3.3
-	3.6	0.3	2.62

System: M.I.B.K. - water

N = 500 r.p.m.

0.015	2.3	0.25	1.08
0.02	2.3	0.25	1.29
0.09	2.3	0.25	1.19
0.18	2.15	0.25	1.32

N = 400 r.p.m.

-	2.61	0.46	1.49

N = 600 r.p.m.

-	1.86	0.23	0.98

System: Trimethylpentane - water

N = 500 r.p.m.

-	3.07	0.46	2.50
0.09	3.07	0.46	2.16
0.13	3.07	0.46	2.25
0.21	2.79	0.46	2.0
0.29	3.17	0.46	2.34

N = 350 r.p.m.

-	2.8	0.9	2.28	
-	3.2	0.9	2.31	With increasing hold-up.
-	3.07	0.9	2.23	
-	3.2	0.9	2.41	
-	2.8	0.9	2.22	

N = 200 r.p.m.

-	3.22	1.4	2.7	
-	3.22	1.4	2.54	With increasing hold-up.
-	3.15	1.4	2.32	
-	3.06	1.4	2.38	

APPENDIX 5. (Cont'd)

Table 6. R.D.C. - DROP SIZE MEASUREMENTS ABOVE THE CRITICAL MINIMUM SPEED. (m.m.)

System: M.I.B.K. - water				
N = 700 r.p.m.				
X	d_{max}	d_{min}	d_{12}	d_{32}
0.06	1.5	0.15	0.88	1.0
0.082	1.5	0.15	0.71	0.88
0.117	1.5	0.05	0.84	0.98
0.23	1.5	0.25	0.84	0.95
0.34	1.75	0.15	0.86	1.05
N = 850 r.p.m.				
0.09	1.25	0.15	0.60	0.83
0.11	1.5	0.15	0.67	0.76
0.31	1.75	0.15	0.75	0.96
0.15				
N = 1000 r.p.m.				
-	1.5	0.1	-	-
System: Toluene - water				
N = 700 r.p.m.				
0.027	3.45	-	-	-
0.029	3.5	-	-	-
0.032	3.5	0.3	1.68	2.5
0.071	3.65	0.3	2.03	2.59
0.12	3.85	0.3	1.97	2.7
0.28	3.9	0.5	1.74	2.62
0.43	4.05	0.3	1.84	2.56
N = 850 r.p.m.				
0.034	2.8	0.16	1.65	2.01
0.04	2.8	-	-	-
0.048	2.95	-	-	-
0.09	3.15	0.3	1.58	1.96
0.17	3.3	0.5	1.72	1.88
0.31	3.4	0.5	1.62	2.28
0.4	3.4	0.6	1.75	2.32

APPENDIX 5. (Cont'd)

Table 6 (Cont'd)

System: Toluene - water

N = 1000 r.p.m.

X	d_{max}	d_{min}	d_{12}	d_{32}
0.08	2.35	-	-	-
0.09	2.45	-	-	-
0.094	2.5	0.3	1.1	1.34
0.157	2.65	0.16	1.23	1.66
0.251	2.8	0.35	1.17	1.63
0.37	2.96	0.35	1.18	1.63
0.42	3.1.	0.5	1.29	1.85

System: Trimethylpentane - water

N = 700 r.p.m.

0.03	3.48	0.46	-	-
0.06	3.67	0.46	-	-
0.08	3.67	0.46	-	-
0.145	3.67	0.46	1.99	2.46
0.20	3.17	0.46	1.99	2.31
0.244	3.07	0.46	1.94	2.32
-	3.48	0.46	1.97	2.24

N = 850 r.p.m.

0.05	3.25	0.46	1.93	2.33
0.10	3.68	0.23	1.60	2.28
0.17	3.72	0.7	1.89	2.33
0.21	3.25	0.46	2.08	2.35
0.24	3.68	0.23	1.88	2.42
-	3.72	0.46	2.06	2.42

N = 1000 r.p.m.

0.06	3.07	0.46	1.49	1.86
0.08	2.78	0.46	1.71	1.98
0.10	3.07	0.46	-	-
0.19	3.48	0.46	1.69	2.12
0.19	3.72	0.46	1.70	2.15
0.255	3.26	0.46	-	-
-	3.72	0.46	1.78	2.14
-	3.72	0.46	1.67	2.12

APPENDIX 5 (Cont'd)

Table 7. Oldshue-Rushton - Drop Size Measurements, Laminar Region (mm).

System: Toluene - water

N = 350 r.p.m.

X	d_{max}	d_{min}	d_{12}	
0.03	3.2	0.2	1.46	
0.04	2.7	0.1	1.15	
0.05	3.2	0.2	1.23	
0.075	3.1	0.2	1.15	
0.137	3.2	0.2	1.07	
0.21	3.2	0.2	1.35	

N = 200 r.p.m. ('Very low' rotor speed)

X	d_{max}	d_{min}	d_{12}	
-	3.5	0.4	2.18	With increasing hold-up
-	3.5	0.2	2.0	

System: Trimethylpentane - water

N = 350 r.p.m.

X	d_{max}	d_{min}	d_{12}	
-	3.1	0.3	2.0	With increasing hold-up
-	2.9	0.3	1.96	
-	2.9	0.3	1.60	
-	3.1	0.3	1.90	

N = 200 r.p.m. ('Very low' rotor speed)

X	d_{max}	d_{min}	d_{12}	
-	3.2	0.5	2.29	With increasing hold-up
-	3.2	0.5	2.46	
-	3.1	0.5	2.47	
-	3.45	0.5	2.50	
-	3.45	1.1	2.34	

System: M.I.B.K. - water

N = 350 r.p.m.

X	d_{max}	d_{min}	d_{12}	
-	1.47	0.22	0.78	With increasing hold-up
-	1.33	0.22	0.69	
-	1.47	0.22	0.73	
-	2.05	0.22	0.70	

APPENDIX 5. (Cont'd)

Table 7. (Cont'd)

System: M.I.B.K. - water

N = 200 r.p.m. ('Very low' rotor speed)

-	2.4	0.22	1.12	
-	2.6	0.22	1.19	
-	2.4	0.22	1.07	With increasing hold-up
-	2.1	0.22	1.25	
-	2.6	0.22	1.31	
-	2.2	0.22	1.14	
-	2.0	0.22	1.19	

APPENDIX 5. (Cont'd)

Table 8. Oldshue-Rushton - Drop Size Measurements, Turbulent Region (mm.)

System: Toluene - water

N = 500 r.p.m.

X	d_{max}	d_{min}	d_{12}	d_{32}
0.043	2.3	0.1	0.85	1.19
0.048	3.2	0.1	0.96	1.17
0.063	3.2	0.1	0.99	1.38
0.095	3.2	0.2	0.84	1.45
0.095	2.3	0.2	-	1.41

N = 600 r.p.m.

X	d_{max}	d_{min}	d_{12}	d_{32}
0.077	1.5	0.13	0.64	0.79
0.080	1.5	0.13	-	0.84
0.090	1.7	0.2	0.70	0.94
0.10	2.2	0.2	0.74	1.04
0.137	2.0	0.2	-	1.26

N = 700 r.p.m.

X	d_{max}	d_{min}	d_{12}	d_{32}
0.125	1.7	0.2	0.67	0.75
0.128	1.7	0.2	0.64	0.79
0.135	1.3	0.2	0.65	0.79
0.15	1.3	0.2	0.74	0.91

System: M.I.B.K. - water

N = 500 r.p.m.

X	d_{max}	d_{min}	d_{12}	d_{32}
0.101	0.89	0.22	0.45	0.49
0.139	1.33	0.22	0.58	0.69
0.152	1.33	0.22	0.61	0.72
0.165	1.5	0.25	0.54	0.77

System: Trimethylpentane - water

N = 500 r.p.m.

X	d_{max}	d_{min}	d_{12}	d_{32}
0.05	2.1	0.26	1.21	1.47
0.095	2.1	0.26	1.33	1.55
0.21	2.6	0.26	1.38	1.60
0.29	2.4	0.26	1.25	1.63

APPENDIX 5. (Cont'd)

Table 8. (Cont'd)

System: Trimethylpentane - water

N = 700 r.p.m.

0.02	2.1	0.26	-	-
0.048	1.6	0.26	0.96	0.99
0.15	2.1	0.26	0.95	1.03
0.24	1.6	0.26	1.01	1.07
0.26	1.6	0.26	0.96	1.12

N = 850 r.p.m.

0.062	1.6	0.26	0.95	0.96
0.098	1.6	0.26	0.84	0.97
0.136	1.6	0.26	0.84	1.00

APPENDIX 5. (Cont'd)

Table 9 - MEAN DROP SIZES FROM DISTRIBUTOR.

Basis - Between 15 and 30 drops counted only (∴ of uncertain accuracy)

Toluene - water

$ft.s^{-1}$	0.214	0.322	0.525	0.69	0.865	1.1	-
d_{av}, m.m.	4.35	4.37	3.9	3.5	3.26	3.1	-

M.I.B.K. - water

$ft.s^{-1}$	0.184	0.29	0.38	0.61	0.79	0.94	-
d_{av}, m.m.	2.6	2.4	2.22	2.04	1.78	1.69	-

Trimethylpentane - water

$ft.s^{-1}$	0.172	0.193	0.29	0.39	0.58	0.85	1.1
d_{av}, m.m.	4.65	4.45	4.3	4.15	3.8	3.4	3.1

APPENDIX 6.

Distributor Design

The size of holes in the distributor plate was estimated initially from the equation (11),

$$V_p + 9.68\,(10^{-10})\,\frac{V_p^{\frac{2}{3}}\,\rho_D\,V^2}{\Delta\rho}$$

$$= \frac{4.92\,(10^{-9})\,\sigma\,d}{\Delta\rho} + \frac{4.95\,(10^{-4})\,d^{1.12}\,V^{0.547}\,\mu_c^{0.279}}{\Delta\rho^{1.5}}$$

A number of 1/16th inch thick removeable brass plates were fabricated and tested. The one found satisfactory throughout this work comprised 45, 1/16th inch dia. holes on 3/16th inch square pitch inside a $1\frac{1}{2}$ inch dia. circle.

The holes were drilled undersize (3/64 inch) and then counterpunched to the correct size. This provided a small upward projection at each hole and hence more uniform droplets.

APPENDIX 7.

DESCRIPTION OF CINE-FILMS

1. Droplet Characteristics in Agitated Extraction Columns

Approx. 7 mins.

Stationary aqueous continuous phase. Toluene dispersed.

No.	Description	Note
1.	R.D.C., 700 r.p.m. Normal operation (64 t.p.s.)	No 'trapped' drops. Toroidal typical of low r.p.m.
2.	R.D.C., 1000 r.p.m. Normal operation (64 t.p.s.)	Typical of high speed hydrodynamics. Large number of small drops. Increased tendency for axial mixing.
3.	R.D.C., 1000 r.p.m. Flooding (64 t.p.s.)	Intense axial mixing.
4.	R.D.C., 500 r.p.m. i.e. $Re < 1.75 \times 10^4$ (4000 t.p.s.)	Drops built-up beneath rotor, not stator. Discrete and changing population. Thrown off with in-sufficient turbulence for break-up. Do not reach wall. No coalescence.
5.	R.D.C., 700 r.p.m. Normal operation. Tip of disc (4000 t.p.s.)	Small drops accelerated in region of disc tip. Large drops deformed in shear field. No coalescence. Drop spin and break-up in about 0.02 secs.
6.	O.R., 200 r.p.m. Low speed (64 t.p.s.)	Insufficient turbulence to remove drops trapped below stator. Inefficient distribution of drops over X-section.
7.	O.R., 350 r.p.m. $Re < 3.8 \times 10^3$ (64 t.p.s.)	Agitator acting as 'paddle' not turbine. Break-up by hitting agitator in the main.
8.	O.R., 500 r.p.m. $Re > 3.8 \times 10^3$ (64 t.p.s.)	Normal operation. Wide distribution of drop sizes. Axial mixing significant.
9.	O.R., 500 r.p.m. (4000 t.p.s.) Tip of turbine	Turbulence then quiescence. Localised coalescence (correlations ∴ for specific column geometry).

APPENDIX 7. (Cont'd)

2. Miscellaneous Phenomena

No.	Description	Note
1.	R.D.C. 500 r.p.m. acrylic spheres in water (500 t.p.s.)	Flow pattern
2.	R.D.C., 850 r.p.m. p.t.f.e. & brass discs Carbon tetrachloride dispersed in water (500 t.p.s.)	Streaming from tip of p.t.f.e. disc. Some large drops.

APPENDIX 8.

Derivation of Equation 78 (108).

The basis used for the derivation of Equation 78, was as follows:- Assume that spherical drops fall through a stationary volume element of cross-section F and height h with a velocity V_s referred to the element. Let n_k droplets per unit time enter the element equally distributed over F. During the time to cover distance h, i.e. h/V_s a further $h.n_k/V_s$ drops will enter the element. Therefore,

$$\text{Total volume of drops in volume element} = \frac{h.n_k.k_1.\ d^3}{V_s} \qquad 64.$$

$$\text{Hold-up } X = \frac{h.n_k.k_1\ d^3}{V_s.F.} \qquad 65.$$

The velocity of rise or fall of the drops is the relative velocity V_x diminished by the linear velocity of the continuous phase.

$$V_s = V_x - \frac{Wc.}{F\ (1-X)} \qquad 66.$$

where Wc is the continuous phase volumetric flow rate. Eliminating V_s

$$V_x = V_s - \frac{Wc}{F(1-X)} = \frac{n_k.k_1\ d^3}{F.X} - \frac{Wc}{F(1-X)} \qquad 67.$$

$$V_x = \frac{Wd}{F.X} - \frac{Wc}{F(1-X)} = \frac{Vd}{X} - \frac{Vc}{1-X} \qquad 68.$$

The relative velocity will be affected by collisions between the droplets in two ways.

(a) The velocity is decreased by inter-droplet collisions. Misek applied an empirical equation for hindered settling to describe this effect:

$$Vox = Vo\ (1-X)\ e^{-4.1x} \qquad 69.$$

(b) Coalescence results in increased droplet sizes with increased velocities.

Considering first the case for equal size drops; the coalescence of two droplets of equal initial diameter d_o yields a droplet of diameter d_1, where,

$$d_1 = \sqrt[3]{2\ d_o} = d_o\ \sqrt[3]{2} \qquad 70.$$

The droplets will split in the region of the agitator but coalesce to a final diameter $\bar{d}$. The critical diameter affecting velocity is the hydraulic mean diameter,

$$\bar{d} = \frac{\sum d^2}{\sum d} \qquad 71.$$

If an initial number of droplets n_o require q binary collisions to reach the final size $\bar{d}$, the system will then contain q coalesced droplets and $n - 2q$ non-coalesced droplets. Hence the final droplet size,

$$\bar{d} = \frac{(n_o - 2q)\ d_o^2 + q\ (\sqrt[3]{2.}\ d_o)^2}{n_o - 2q\ d_o + q\ \sqrt[3]{2}\ d_o} \qquad 72.$$

$$= d_o .\ \frac{n_o - q\ (2 - 2^{\frac{2}{3}})}{n_o - q\ (2 - 2^{\frac{1}{3}})} \qquad 73.$$

Substituting $Q = q/n_o$ and evaluating

$$\bar{d} = d_o\ \frac{1 - Q\ 0.41}{1 - Q\ 0.74} \qquad 74.$$

For the more precise case when after each coalescence the mean droplet diameter is changed and coalescence is between droplets of the mean diameter corresponding to the previous collision, assuming a large number of drops are present, Misek obtained the simplified expression,

$$\frac{\bar{d}}{d_o} = \frac{1}{(1 - Q)^{0.328}} \qquad 75.$$

Comparison of calculated values of $\bar{d}/d_o$ from Eqns. 74 and 75 showed that the difference between the mechanisms was small.

Since droplet size does not change over wide limits,

$$\frac{V_x}{V_{ox}} = \frac{1}{(1 - Q)^{m.0.328}} \qquad 76.$$

where m is a function of $N_{Re} = \frac{V_o \, d_o \, \rho}{\mu}$

as for the terminal falling velocities of rigid spheres (65).

Misek replaced the function in Equation 76, by an approximation,

$$\ln \frac{V_x}{V_{ox}} = k_2.m.X = \frac{Z}{m} X \qquad 77.$$

assuming the number of collisions Q is proportional only to the dispersed phase hold-up (i.e. $Q = k_2 X$) and the magnitude of k_2 depends on the physical properties of the system, agitation and coalescence time.

Combining Equations 68, 69 and 77,

$$\frac{V_d}{X} + \frac{V_c}{1-X} = \bar{V}(1 - X).\, \exp\left[\frac{Z}{m} - 4.1\right] X \qquad 78.$$

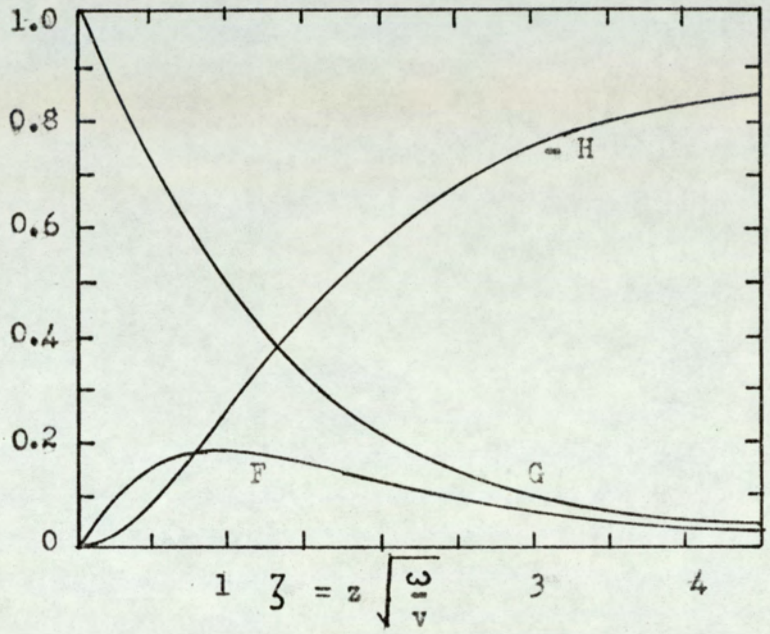

FIGURE 91. VELOCITY DISTRIBUTION NEAR A ROTATING DISC ;

Values of functions F, G and H for use in Equation 100 (142)

APPENDIX 8 (Cont'd)

Table 11. R.D.C. Laminar Region - Summary of Calculated Radial and Tangential Velocities.

Plane					r = R' cm.			
		0.5	0.75	1.0	1.25	1.5	1.75	2.0
N = 500 r.p.m.								
z = 0	u	0	0	0	0	0	0	0
	v	26.25	39.4	52.5	65.6	78.8	91.9	105.0
z = d	u	4.7	7.05	9.4	11.75	14.1	16.45	18.9
	v	12.3	18.5	24.6	30.7	36.9	43.0	49.2
z = 2d	u	3.1	4.65	6.2	7.75	9.3	10.85	12.4
	v	5.35	8.02	10.7	13.37	16.05	18.72	21.4
N = 600 r.p.m.								
z = 0	u	0	0	0	0	0	0	0
	v	31.5	47.3	63.0	78.7	94.5	110.0	126.0
z = d	u	5.65	8.45	11.3	14.1	16.9	19.8	22.7
	v	14.8	22.2	29.6	36.8	44.3	51.5	59.0
z = 2d	u	3.72	5.6	7.45	9.3	11.2	12.4	14.9
	v	6.43	9.6	12.8	16.05	19.2	22.4	25.6

Note: u, v in cm. s^{-1}.

APPENDIX 8 (Cont'd)

Table 12. R.D.C. Laminar Region

Resultant Differences in Velocities across droplets.

	r = R'						
	0.5	0.75	1.0	1.25	1.5	1.75	2.0
	V cm. s^{-1}						
N = 500 r.p.m.							
z = 0 to z = d	14.8	24.6	29.6	37. 0	44.3	51.5	58.9
z = d to z =2d	7.15	10.8	14.3	17.8	21.4	24.9	28.5
N = 600 r.p.m.							
z = 0 to z = d	17.7	26.4	33.8	44.2	53.0	61.5	71.8
z = d to z =2d	8.6	12.9	17.3	23.2	25.7	30.0	34.3

APPENDIX 8 (Cont'd)

Table 13. R.D.C. Laminar Region

Stable drop diameters beneath disc

	r = R'						
	0.5	0.75	1.0	1.25	1.5	1.75	2.0
	$d_{s.d.}$ cm.						
N = 500 r.p.m.							
System: toluene - water.							
z = 0 to z = d	2.9	1.57	1.26	0.97	0.78	0.62	0.53
z = d to z = 2d	6.9	4.75	3.02	2.34	1.87	1.56	1.33
System: M.I.B.K. - water.							
z = 0 to z = d	1.37	0.74	0.59	0.45	0.36	0.28	0.24
z = d to z = 2d	3.28	2.26	1.44	1.11	0.89	0.74	0.63
System: Trimethylpentane - water.							
z = 0 to z = d	4.25	2.31	1.84	1.42	1.14	0.94	0.80
z = d to z = 2d	10.20	7.00	4.45	3.44	2.74	2.28	1.94
N = 600 r.p.m.							
System: toluene - water.							
z = 0 to z = d	2.35	1.45	1.08	0.78	0.63	0.55	0.44
z = d to z = 2d	5.60	3.44	2.42	1.71	1.52	1.25	1.07
System: M.I.B.K. - water.							
z = 0 to z = d	1.11	0.68	0.51	0.36	0.29	0.26	0.21
z = d to z = 2d	2.53	1.62	1.14	0.80	0.71	0.59	0.50
System: Trimethylpentane - water.							
z = 0 to z = d	3.46	2.14	1.58	1.14	0.93	0.81	0.65
z = d to z = 2d	8.25	5.05	3.56	2.52	2.24	1.84	1.57

APPENDIX 9.

Typical Drop Counts.

R.D.C.; System; toluene - water.
N = 850 r.p.m., X ≙ 0.034

d	n	nd	nd^2	nd^3
2.8	21	59.0	166.0	462.0
2.7	3	8.1	21.9	57.0
2.5	4	10.0	25.0	62.0
2.2	18	39.6	87.5	189.0
1.9	2	3.8	7.8	13.7
1.7	35	59.5	101.0	172.0
1.4	11	15.4	21.6	30.1
1.3	5	6.5	8.5	11.0
1.1	52	57.2	63.0	69.0
0.83	19	15.8	13.1	10.8
0.55	72	39.6	21.6	11.9
0.3	39	11.7	3.5	1.1
0.16	15	2.4	-	-
0.1	6	-	-	-
		328.6	540.5	1089.6

$d_{12} = \frac{540}{328} = 1.65$ mm. $d_{32} = \frac{1089.6}{540.5} = 2.01$ mm.

Oldshue - Rushton; System, toluene-water.
N = 350 r.p.m., X ≙ 0.04

d	n	nd	nd^2	nd^3
2.7	2	5.4	14.6	39.6
2.3	3	6.9	15.9	36.6
1.8	4	7.2	13.0	23.2
1.4	9	12.6	17.7	21.5
1.1	1	1.1	1.21	1.3
0.9	11	9.9	8.91	8.1
0.7	2	1.4	0.98	0.6
0.5	46	23.0	11.5	5.7
0.2	29	5.8	1.16	0.2
0.1	4	0.4	0.04	-
		73.7	85.0	137.0

$d_{12} = \frac{85}{73.7} = 1.15$ mm. $d_{32} = \frac{137}{85} = 1.61$ mm.

APPENDIX 9 (Cont'd)

Table 14, R.D.C. - Drop Size Measurements with Flow of Continuous Phase (m.m.)

See also Table 6.

System: Trimethylpentane - water

N = 850 r.p.m. V_c = 20.8 ft.hr.$^{-1}$

V_d ft.hr.$^{-1}$	d_{max}	d_{min}	d_{12} approx.	d_{32} approx.
13.8	3.90	0.37	2.05	2.37
20.3	3.75	0.37	2.09	2.48
27.2	3.75	0.37	1.99	2.50
41.0	3.75	0.37	2.03	2.56
60.5	3.90	0.37	1.92	(2.35)
77.5	3.42	0.37	1.84	(2.18)

N = 850 r.p.m. V_c = 34.7 ft.hr.$^{-1}$

V_d ft.hr.$^{-1}$	d_{max}	d_{min}	d_{12} approx.	d_{32} approx.
13.8	3.82	0.37	-	-
20.3	4.14	0.37	1.95	2.24
27.2	3.80	0.37	2.02	2.32
41.0	4.14	0.37	2.00	2.46
60.5	3.75	0.37	2.20	2.56

NOMENCLATURE.

Symbols.

a = Interfacial area of contact (ft^2 per ft^3 column volume).

A = Total interfacial area in contactor (ft^2).

B = $h.x.$

C = Concentration

C_R = Factor accounting for column restriction equal to S^2/D^2 or $(D^2 - R^2) / D^2$.

D = Column diameter

D' = Diffusion coefficient

d = drop diameter

d_{vs}, d_{32} = mean volume - surface drop diameter

d_h, d_{12} = hydraulic mean drop diameter

d_c, d_{cr} = critical drop size

$d_{f.c.}$ = critical packing dimension

d_L = ligament diameter

E = energy dissipation per unit mass

E' = eddy diffusivity

F = $h.\theta$

f = total growth of wave = $\ln T/T_o$.

f_x = ratio of actual mean drop diameter to diameter at $X = 0.1$

g = acceleration due to gravity

H = stator opening or column height

h = sheet thickness

H.T.U.= height of a transfer unit

K = overall mass transfer coefficient

k = film mass transfer coefficient

k = constant

L = macroscale of turbulence

m = distribution coefficient

N = rotor or impeller speed, rev./time

N' = average number of droplet collisions per unit time.

N'' = frequency of collisions per unit volume

N_{Re} = Reynolds number for droplets, equal to $\frac{d.V_t.\rho c}{\mu c}$

n = number of compartments

n' = number of drops per unit volume

n'' = wave number

P, τ = force per unit area

P_V = power input per unit volume

P_D = power input per unit mass of dispersed phase, equal to $\frac{\rho N^3 R^5}{\rho_D X.H.D^2}$

Pe = Peclet number, equal to V.H./E'

R = rotor or impeller diameter

R' = effective distance of drop from column centreline

ΔR = distance between agitator and column wall, equal to (D-R) /2.

r = drop radius

Re = Reynolds number for impeller, equal to $\frac{N^2 R \rho c.}{\mu c}$

S = stator ring opening

T = wave amplitude

To = amplitude of initial disturbance

t = contact time

U = radial velocity

u(d) = relative velocity between two points in fluid separated by distance d.

u,v,w = fluctuating components of velocity

$\bar{u},\bar{v},\bar{w}$ = root mean square values of u,v,w

V = superficial velocity of phase in column

V = velocity

$\bar{V}_N$ = characteristic velocity, i.e. the mean vertical droplet velocity at substantially zero flow rates and at rotor speed N.

V_o = peripheral velocity of agitator

v_o = characteristic velocity of turbulence fluctuations

ν = shear stress; coalescence frequency

w^* = critical relative velocity along the line of centres between colliding drops

X = fractional hold-up of dispersed phase

x = distance, linear of diffusion

Z = coefficient of coalescence

δ = thickness of fluid layer carried by disc.

$\mathcal{E}$ = energy dissipation

ϵ = fractional voidage of column packing

ε = turbulent diffusion coefficient

γ = kinematic viscosity

η = microscale of turbulence

ϕ = empirical correction factor for motion of restricted drops

μ = viscosity

σ = interfacial tension

ρ = density

$\Delta\rho$ = density difference between phases

ζ = dimensionless distance from disc surface

Subscripts.

c = continuous phase

d = dispersed phase

t = terminal

s.d. = stable drop

f = at the flood point

f.c. = critical size (packing)

cr = critical size (droplets)

θ = time

ω = angular velocity

L = ligament

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