THE INFLUENCE OF PREFERENTIAL DIFFUSION ON THE BLOW-OFF OF LAMINAR AERATED BURNER FLAMES

by

DAVID OSMOND BIZLEY

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The Department of Chemical Engineering.
The University of Aston in Birmingham.
England.

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SUMMARY

The blow-off of a laminar aerated burner flame in which Hydrogen is present cannot be characterised by the flame stretch correlation proposed by Reed. A literature review has indicated that the preferential diffusion of Hydrogen towards the point of stability in the region of maximum primary flame front curvature at the base of the flame may cause this anomaly.

The influence of the introduction of Hydrogen into the Propane-Air system on blow-off has been studied and correlations of critical boundary velocity gradient and burning velocity against fuel concentration obtained for eight different Propane/Hydrogen ratios. It has been proposed that the degree to which preferential diffusion will occur may be characterised by a dimensionless group termed the preferential diffusion factor. This has been correlated against critical flame stretch factor for the Hydrogen-Propane-Air system and hence an equation has been developed which may be used to predict critical flame stretch factors for aerated systems from a knowledge of fuel concentration and preferential diffusion factor.

Physical and transport properties of the Hydrogen-Propane-Air system have been varied by the replacement of the Nitrogen present in the Air by Helium and Argon. These data, together with that for the Hydrogen-Methane-Air system from the literature, have been analysed using the preferential diffusion factor approach and have revealed considerable agreement between predicted and experimental critical flame stretch factor-fuel concentration relationships.

Hydrogen concentration isopleths have been studied in the stabilising region of three laminar Hydrogen-Propane-Air flames with 60% Hydrogen present in the fuel gas mixture. Isopleths plotted from this investigation are consistant with the preferential diffusion mechanism.

Similar studies of the Ethylene-Air system have been made at all stages of the investigation for comparison purposes.

NOMENCLATURE.

NOMENCLATURE

Symbol	Meaning
A	Area of flame front (m^2)
a	Parameter defined in section 1.1.4 (unless otherwise stated).
C	Parameter defined in section 4.2.1.
C_p	Specific Heat (J/kg.deg.C).
d	Diameter (m).
D	Molecular diffusivity (m^2/s).
F	Preferential diffusion factor (unless otherwise stated).
gb	Critical boundary velocity gradient (s^{-1}).
k	Thermal conductivity (w/m^2 deg.C.).
K	Flame stretch factor as defined in equation (1.8).
K^1	Flame stretch factor as defined in equation (4.2).
K*	Predicted critical flame stretch factor.
Le	Lewis Number.
L_E	Entry length for fully developed laminar velocity profile (m).
M	Molecular weight.
P	Pressure (N/m^2).
Q	Volumetric flow rate (m^3/s).
R	Burner Radius (m).
Re	Reynolds number.
S	Local burning velocity (m/s).
Su	Normal burning velocity (m/s).
t	Time (s).
T	Temperature (deg.C.).

Symbol	Meaning
U	Gas velocity (m/s).
x	Concentration expressed as a mole fraction.
X	Fuel concentration expressed as a fraction of stoichiometric.
α	Parameter defined in section 1.1.4.
β	Parameters defined in sections 1.1.4 and 1.2.3.
μ	Viscosity (Ns/m^2).
η_o	Preheat zone thickness (m).
σ	Standard deviation.
ρ	Density (kg/m^3).

Subscripts.

1	Low molecular weight species in combustion products.
2	Low molecular weight species in unburned gas mixture.
b	Burned gases.
i	Point of ignition (unless otherwise stated).
u	Unburned gases.
max	Maximum value.

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INTRODUCTION

Blow-off is said to occur when a flame lifts off from a burner and is extinguished. Until recently, Town gas was the principal fuel gas utilised both domestically and industrially in Britain. Town gas is a mixture of a number of gases but normally contains about 50% Hydrogen and has consequently a relatively high maximum value of burning velocity. Natural gas contains no Hydrogen, however, and so has a maximum value of burning velocity less than half that of Town gas. The introduction of Natural gas in Britain has forced a change of emphasis in burner design from light back to blow-off as the most important flame stability consideration. This change of fuel gas has also resulted in the almost exclusive use of aerated rather than diffusion flames in burner design.

Research work has recently been directed towards the development of a means of predicting when blow-off of an aerated flame will occur from any burner using any fuel gas. This has necessitated clarification of the fundamental mechanism governing flame stability in general and blow-off in particular. The Flame Stretch theory has gone some way to attaining these objectives. It was proposed that the critical flow rate of the fuel-oxidant mixture at which blow-off would occur could be predicted from a general correlation (Equation 1.10). It has since been shown that most laminar aerated systems adhere fairly closely to the behaviour predicted by the general correlation but that when Hydrogen is present in the fuel gas, the method breaks down. It has been proposed that the anomaly may be attributed to the preferential diffusion of Hydrogen. By this mechanism, Hydrogen would diffuse towards the point on the flame front governing blow-off, thus increasing the local unburned Hydrogen concentration to a value in excess of that present in the bulk of the unburned gas mixture.

During the last five years, investigations have been carried out into the use of existing Towns gas production plant for the manufacture of substitute Natural Gas both as a standby system and as a means of supplementing normal supplies at times of high demand. Such substitute gases would contain quantities of Hydrogen dependent on the mode of operation of the process in question. The problem of the prediction of stability characteristics of the fuel gas mixtures containing Hydrogen is therefore clearly not without practical relevance.

The main aims of this research project have been as follows:-

(i) To determine if preferential diffusion can account for the anomolous blow-off characteristics of aerated flames produced with fuel gas mixtures containing Hydrogen.

(ii) To modify the flame stretch correlation in such a way as to take account of the preferential diffusion process and thus render it applicable to all laminar aerated flames.

Section 1.

LITERATURE REVIEW.

1. LITERATURE REVIEW.

1.1 Theories of Blow-off

1.1.1 General principles of Blow-off

For a flame front to be stabilised on a burner there are two pre-requisites:-

(i) At some point on the flame front the local burning velocity must equal the velocity on the unburned gases.

(ii) At all other points on the flame front the unburned gas velocity must exceed the local gas velocity. Thus, that point described by the first pre-requisite will provide a source of ignition for the remainder of the flame front.

Should the local gas velocity exceed the local burning velocity over the entire flame front, blow-off will occur. However, should the first pre-requisite be met but the burning velocity exceed the local gas velocity at other points on the flame front, the flame will light back. Whether either of these types of instability will take place is dependant on the flow régime that exists, and the device used to produce a suitable régime is the gas burner. The gas industry has used a variety of ways to represent stability limits, but perhaps the most well known of these is Weaver's method, which has been discussed in detail by Gilbert and Prigg (1).

Thus it is clear that the most important parameters governing blow-off are likely to be:-

(i) Burning velocity.

(ii) A parameter which in some way characterises the flow régime.

1.1.2 Early theories of Blow-off

The first important attempts to develop theories of stability of burner flames were made some thirty years ago. Adam(2), Damkoehler (3), Heiligenstaedt (4), Delbourg (5), Garside et al (6) and Lewis and Von Elbe all published their respective ideas on the subject. Most of these are today largely of historic interest, but the work of Lewis and Von Elbe stands out as being by far the most significant contribution to our knowledge of the subject until the last decade.

1.1.3 The Boundary Velocity Gradient Theory of Lewis and Von Elbe

The theory (7, 8, 9) is based on the following assumptions:-

(i) The gas velocity varies linearly with distance from the stream boundary.

(ii) The burning velocity of the combustion mixture is zero at the burner wall and remains constant as the distance from the burner wall is increased until a point is reached where the effects of quenching of the flame reactions by the burner, which decreases with distance from the wall, and dilution of the combustion mixture by diffusion from the surrounding atmosphere, which increases with distance from the burner wall, are counterbalancing each other in such a way that a finite burning velocity can exist. This burning velocity then increases with distance from the burner wall until the maximum burning velocity is reached at a point where the quenching effect of the burner is zero.

A flame is stabilised where the local gas velocity and local burning velocity are equal. If the gas velocity is increased, the point at which the flame is stabilised is removed from the burner wall until the reduced quenching effect permits the burning velocity to increase to a sufficient degree to equal the new local gas velocity. Thus a new equilibrium position is reached by the flame front and an increase in the dead space at the base of the flame has occurred. An increase in dead space, however, may cause an increase in diffusion from the surrounding atmosphere to the outer regions of the flame and this causes a corresponding change in the burning velocity in this region. It will become clear, therefore, that a point will be reached where further increases in flow rate cannot be matched by a corresponding increase in burning velocity. Hence, blow-off will occur. Lewis and Von Elbe proposed that the critical blow-off point could be characterised by the velocity gradient of the unburned gases at the burner wall. It follows that for different burners, the critical boundary velocity gradient remains constant provided that the mixture composition and other physical properties are unaltered. This has been shown to be largely true with the possible exception of very small burner ports where interaction may occur between different regions of the flame front.

It follows that there are two requirements for the prediction of the blow-off characteristics of a flame of known composition from a burner by this theory:-

(i) An experimentally determined correlation must exist of critical boundary velocity gradient against fuel concentration for the fuel gas in question.

(ii) It must be possible to relate the boundary velocity gradient to a volumetric flow rate. In the majority of cases, this necessitates the characterising of developing flow in some kind of channel. The efforts of a number of workers e.g. Sparrow et al (10) have made this possible and earlier work (11) by the author has shown that such predictions of developing flow can be used to predict blow-off correlations which agree closely with those found experimentally.

The theory has been used by many workers to interpret their blow-off data and Reed has listed a number of these in one of his papers (12). The good correlations obtained from the theory tended to discourage enquiry into the validity of the mechanism proposed. There are, however, two reasons why further work has been carried out during the last decade on the subject of blow-off. The first of these is that the critical boundary velocity gradient theory is dependant on a series of experimentally determined correlations and each fuel gas mixture has its own unique correlation. This is a considerable obstacle to the designer who will frequently wish to design burners for fuel gas mixtures of varying composition. The second reason is a series of observations which appear fundamentally to differ from those which would be expected from the proposed mechanism:-

(i) Taylor (13) has observed no detectable change in the dead space at the base of the flame when the gas flow rate is increased in some cases.

(ii) Reed has observed increases in dead space when the burner diameter is decreased and no change in critical boundary velocity gradient is observed (14). He has also observed that by reducing ambient pressure and thus causing a large increase in dead space, he has failed to observe a correspondingly large decrease in critical boundary velocity gradient.

(iii) More recently, Datta et al (15) have studied the stabilising region of a two dimensional near stoichiometric natural gas flame by analysis of temperature, gas velocity and composition.

They have shown that in this case, the burning velocity which is considered as a measure of heat release rate, does not differ significantly in the stabilising region from that in other parts of the flame. This is directly in contrast with the theory of Lewis and Von Elbe which predicts a significant reduction of burning velocity in the stabilising region. The alternative mechanism of "aerodynamic quenching" often called "flame stretch" has been proposed.

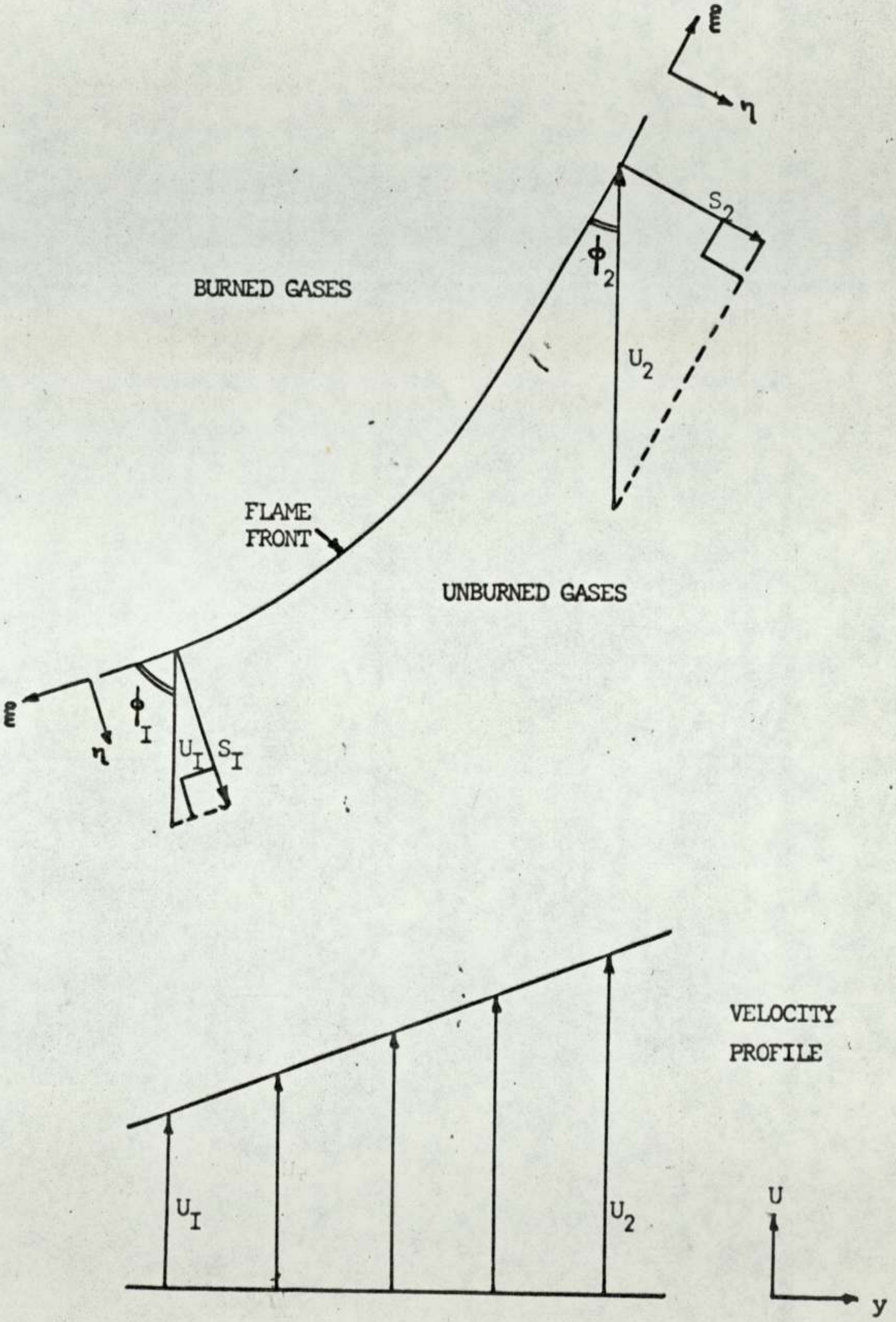
1.1.4 The Flame Stretch theory of Blow-off.

The concept of aerodynamic quenching or flame stretch was first proposed by Karloritz and his colleagues (16, 17). They studied the effects of two dimensional flow on flame propagation and developed equations from the continuity equation for two dimensions, and a heat balance assuming conduction in the η direction only (at right angles to the flame front), convection in the η and ξ directions and negligible radiation (Fig 1.1). A relationship was also developed relating mass flow rate to distance from the flame front in the η direction. The following assumptions were then made:-

(i) $$\sin\phi = \frac{S}{u}\,(\eta = o) \qquad (1.1)$$

(ii) $$\cos\phi = 1 \qquad (1.2)$$

The former assumption is clearly correct. The latter assumption implies a vertical flame front with $\phi = 0^{o}$. Although this is clearly never correct it can be shown that this is a good approximation when blow-off occurs ($\cos\phi > 0.9$ if $\phi < 25^{o}$) Karlovitz combined the three basic relationships and integrated twice



FLAME FRONT PROPAGATION ACROSS A VELOCITY GRADIENT.

FIG. I.I

arriving at the following equation:-

$$\beta = 1 + a\eta_o \tag{1.3}$$

where $a = \frac{1}{u(\eta = o)} \frac{du}{dy}$ (1.4)

and $\eta_o = \frac{k}{Cp \ \rho(\eta = o) Su}$ = preheat zone thickness (1.5)

The concept of the preheat zone thickness is derived from the simplified one-dimensional heat balance (Fig 1.2)

$$\rho(\eta = o) \ Cp \ Su \ (Ti - Tu) = k \left(\frac{dT}{d\eta}\right)_i \tag{1.6}$$

The preheat zone thickness η_o is defined

$$\left(\frac{dT}{d\eta}\right)_i = \frac{Ti - Tu}{\eta_o} \tag{1.7}$$

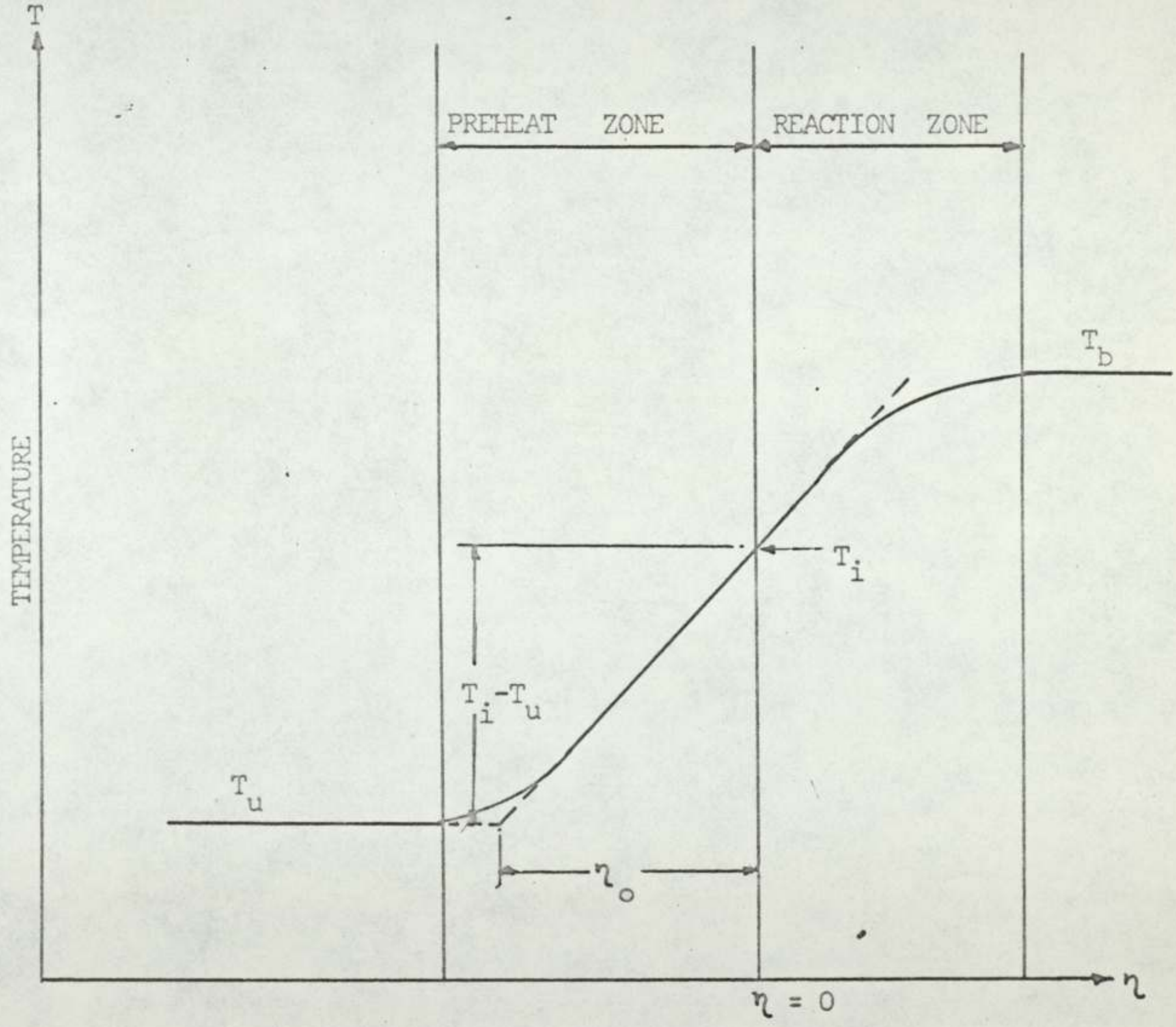
Equation (1.5) follows by combining equations (1.6) and (1.7).

A new parameter K was now defined such that

$$K = \beta - 1 = \frac{1}{u(\eta = o)} \frac{du}{dy} \frac{k}{Cp \ \rho(\eta = o) Su} \tag{1.8}$$

K was named the "Karloritz Number" or "Flame Stretch Factor". Because of the curvature of a flame front, the area of the unburned side of the flame front is greater than the burned gas side. This "stretching" effect gives the theory its name. A consequence of this is that the amount of heat flowing from the reaction zone to the unburned gas is distributed over a greater volume of unburned gas than for a flame front without curvature. This results in a corresponding reduction of chemical reaction rate, heat release rate, and hence burning velocity.

T_b - TEMPERATURE OF BURNED GASES
T_i - IGNITION TEMPERATURE
T_u - TEMPERATURE OF UNBURNED GASES



TEMPERATURE PROFILE OF REACTION AND PREHEAT ZONES OF LAMINAR FLAME

FIG. I.2

The flame stretch factor characterises the increase in area when the flame front propagates over the distance equal to the preheat zone thickness. A critical value will exist therefore which represents the greatest increase in area that the flame front can withstand before rupturing will occur.

Lewis and Von Elbe used the flame stretch concept as proposed by Karlovitz to interpret blow-off data for inverted flames stabilised on wires, and thus they may be considered to have piloted the application of the flame stretch concept to the blow-off of flames (7, 18, 19, 20). They have not, however, attempted to interpret the blow-off of flames from normal burner ports in this way preferring their boundary velocity gradient approach. It seems illogical to dismiss flame stretch as insignificant in this context, particularly when flame stretch in no way invalidates the concept of a critical boundary velocity gradient. It seems unlikely that this point escaped Lewis and Von Elbe and it is more likely that they did not feel the need to introduce flame stretch considerations in a situation in which the boundary velocity gradient provided adequate correlations.

Reed has likened the conditions shown by Karlovitz to lead to the extinction of a combustion wave to that present at the base of a bunsen type flame when blow-off occurs (12, 21). Over a distance equal to the width of the preheat zone, very large changes in gas velocity occur because of the large boundary velocity gradient always present at this point. The region adjacent to the burner rim at the base of the flame is known as the stabilising region, for in this region the largest velocity gradients occur, and therefore the greatest flame stretch will be present. It follows, therefore, that at this point, the flame front rupture causing blow-off will occur. Photographs of the lift-off of flames have shown that this is in fact the case (22). Reed proposed that provided the preheat zone term, ηo, could characterise the combustion wave, and u and $\frac{du}{dy}$ the flow field, a constant value of flame stretch factor might exist that could characterise blow-off independant of other factors such as temperature, pressure and composition. It was then necessary to make two further assumptions to evaluate flame stretch factors for practical cases, these being that at the point of stability where flame front rupture was most likely to occur:-

(i) $\frac{du}{dy} = gb$ - the boundary velocity gradient used by Lewis and Von Elbe in their work.

(ii) $u = Su$ - This is in contrast with Lewis and Von Elbe's theory in which it is stated

$u = S$ where $S =$ local burning velocity, but $Su > S$

Both assumptions are open to criticism. The former may be criticised because gases emerging from the burner tend to "spill out" and the flame front itself may cause a distortion of the velocity profile by the back pressure it exerts, even though this is usually very small. Such criticism is equally valid, however, when discussing the boundary velocity gradient theory and yet this has been shown to give good correlations and so for this reason, the assumption may be considered acceptable. The second assumption is more readily criticised. The flame stretching effect causes a reduction of burning velocity and a thickening of the preheat zone. It is equally true to say, however, that it would be very difficult indeed to apply and test the theory if this assumption is not made. A precise test of this assumption could be made using a particle track technique.

The work of Datta et al (15) has already been discussed and they have shown that the assumption is not unreasonable for the limited case that they have studied. They have also cited other relevant references and discussed this point in some detail.

Reed has thus redefined the flame stretch factor as follows:-

$$K = \frac{\eta_o \; gb}{Su} = \frac{gb \quad k}{Cp \; \rho \; Su^2} \qquad (1.9)$$

He has used critical boundary velocity gradient and burning velocity data obtained by a number of workers over a wide range of mixture compositions, fuel gases, temperatures and flow states (the majority of which were in the laminar region) to test his hypothesis. He plotted gb ηo against Su and found a fairly good correlation although three systems he tested differed noticably from the general pattern of behaviour, these being Hydrogen-Air, Methane-Oxygen, and Propane-Oxygen. He correlated flame stretch factor against fuel concentration and arrived at the following equation:-

$$K = 0.23 \left[1 + (X^{6.4} - 1)\alpha \right] \text{ for } X < 1.36 \qquad (1.10)$$

where $\alpha = 0$ for systems with no secondary reaction zone

$\alpha = 1$ for systems with both primary and secondary combustion zones.

He was unable to test the correlation for $X < 0.7$ but has suggested that there is no theoretical reason for the imposition of such a limit of applicability. He accounted the behaviour of the three anomolous systems to the phenomenon known as preferential diffusion although this suggestion has received considerable criticism (28, 70).

The correlation characterised by equation (1.10) is good as Reed's statistical analysis endorses, but its chief fault lies in that it is based on many results obtained at different times and in different ways by a number of different workers. There are likely to be, therefore, a number of sources of error present in the various sets of data and this may tend to mask the underlying trend of the correlation. A major factor in advocating the general validity of the correlation for practical use, however, lies in this very wide range of systems and conditions used to obtain the correlation.

Mak (23, 24) has commented on the mathematical derivation and the assumptions made and has suggested that a precise test of the theory requires studies of the blow-off of flames using optical techniques. Studies of the blow-off of flames using high speed photography might certainly provide interesting results, but it is doubtful whether any modification made to the definition of the flame stretch factor could avoid rendering the theory more complex and so more difficult to apply practically.

1.2 Analysis of blow-off data using the flame stretch theory

1.2.1 Variation of the burning velocity of Methane-Air mixtures by the use of an additive.

Edmondson and Heap (25, 26) have varied the burning velocity of Methane-Air mixtures by the addition of small known concentrations of Methyl Bromide to the unburned gas mixture. Blow-off data from cylindrical burners and burning velocity data have been determined and critical boundary velocity gradient correlated against fuel concentration for each additive concentration employed. All data was then analysed by the flame stretch method and a single correlation of critical flame stretch factor against fuel concentration was found to exist. This provided a particularly good and convincing test of the flame stretch theory as it avoids the possibility of variations in diffusional fluxes caused by changing the fuel gas so altering the concentrations of the various molecular species present and hence the concentration driving forces for diffusion.

1.2.2 Variation of the nature of the surrounding atmosphere

Edmondson and Heap (27, 28) have carried out determinations of critical boundary velocity gradients for the blow-off of laminar Methane-Air flames from cylindrical burners in atmospheres of Air, Nitrogen, Oxygen, Helium and Carbon Dioxide. They later repeated the determinations for the blow-off of Ethylene-Air flames in atmospheres of Air, Nitrogen and Oxygen. The work was confined to flames leaner in fuel than stoichiometric. Attempts were made to correlate the data by the flame stretch equation(1.10) and the failure to do so was attributed to the influence of the surrounding atmosphere. There can be little doubt that this behaviour is caused by diffusion of the secondary atmosphere through either the dead space at the base of the flame, or through the secondary combustion zone, or possibly even by both routes. Diffusion through the secondary reaction zone is an important mechanism in fuel rich flames, for it is only by this route that oxygen can reach this zone to enable secondary combustion to take place. Clearly, therefore, provided a suitable concentration driving force exists for such diffusion, it need not be confined to Oxygen nor indeed to fuel rich flames.

Reed (29, 30) has presented a logical and well ordered argument against the possibilities of diffusion through the dead space. With the aid of particle track photographs, he has shown that with flames leaner than stoichiometric, the bulk flow of combustible gas is away from the burner port through the dead space at the base of the flame. The concentration driving forces of both Oxygen and Nitrogen are small between the primary

Fuel-Air mixture and the secondary atmosphere. A situation therefore exists where diffusion is very unlikely to occur against such a sizable bulk gas flow. Reed presents the following alternative explanation. Because secondary combustion occurs in fuel rich flames to a far greater degree than in fuel lean flames, an energy flux exists towards the primary reaction zone in the former, and away from the primary reaction zone in the latter. Any change in ambient conditions will alter these fluxes which will in turn affect the heat release rate in the primary reaction zone, i.e. the burning velocity. The work of Hottel and his colleagues (31) is cited as substantiating this argument. Hottel stabilised gas lean inverted flames part way along a rod. The rod provides a heat sink which reduced the energy flux towards the primary reaction zone. The critical boundary velocity gradient was found to be only one tenth of that for a similar flame stabilised above the end of the rod. The presence of the rod penetrating the flame front in this way provides an effective energy sink which in turn gives rise to a substantial reduction in burning velocity and hence stability. This effect would be anticipated even by the advocates of diffusion through the dead space on fuel lean burner stabilised flames, however, for such an effective heat sink could not fail to cause a substantial reduction in stability. It is felt therefore that this work has little to offer in the clarification of diffusion mechanisms.

Of far more importance are the various concentration measurements made by different workers in the stabilising region of flames. Smith (32) has increased the Argon concentration in the Air surrounding a bunsen type burner on which a Methane-Air flame was stabilised. Analysis was carried out using a continuous sampling system and a mass spectrometer. He found no significant diffusion of Argon into the primary mixture with a fuel concentration (X) less than 1.1 stoichiometric. However, sampling 2mm. above the port of a rectangular burner, the Argon concentration in the primary mixture was found to rise significantly as X was increased from 1.2 to 1.9. Increasing the gas velocity was found to increase the Argon concentration slightly in richer mixtures. The work of Datta et al (15) on a near stoichiometric two dimensional flame has already been discussed in another context. Their concentration measurements in the stability region showed no indication of significant diffusion through the dead space. A similar study carried out by the same authors (33) on a two dimensional fuel rich Methane-Air flame showed that as expected there is an increase in diffusion through the dead space in this situation.

In contrast with the above observations, Edmondson and Heap (27, 28) have proposed that their work on Carbon Monoxide-Oxygen flames shows that diffusion through the dead space is significant in fuel lean flames. The Carbon Monoxide-Oxygen system was used because of the way in which the burning velocity of a specific mixture varies with water vapour concentration. Wires et al (71) have shown that little or no combustion will occur when absolutely dry Carbon Monoxide-Oxidant mixtures are used. Edmondson and Heap used mixtures dried using silica gel and they proposed that where diffusion from a moist secondary atmosphere occurred, an increase in burning velocity would be observed. Initial experiments were carried out using a dry Oxygen atmosphere. The critical flame stretch factors calculated from critical boundary velocity gradient and burning velocity data obtained, increased rapidly as the fuel concentration was increased towards stoichiometric. They then determined burning velocities from particle track photographs for a fuel lean dry primary mixture burning in a moist secondary atmosphere of Nitrogen and found burning velocity increased significantly as the burner rim was approached.

It seems extremely hazardous to draw any general conclusions from this work for the following reasons:-

(i) Water vapour molecules are far more mobile than those present in other situations and might therefore diffuse through the dead space of a flame under conditions in which a less mobile molecule, e.g. Oxygen, would not diffuse to a significant degree.

(ii) There is a far larger concentration driving force for diffusion of water vapour present than would exist in most of the other situations under consideration.

(iii) The situation is not dissimilar to that present in a diffusion flame where the magnitude of the burning velocity is dependant on the diffusion of a molecular species from the secondary atmosphere to the primary flame front.

It may be concluded that diffusion through the dead space is unlikely to take place in an aerated flame burning in Air. It is also unlikely to take place where the secondary atmosphere is made up of low mobility molecular species or where there is an insufficient driving force for diffusion. In the case of a fuel lean aerated flame burning in a Helium atmosphere, however, it is quite likely that diffusion might occur against the bulk gas flow through the dead space.

1.2.3 Vitiation of Primary and Secondary Air

The subject of vitiation is of considerable practical importance, particularly when the diluent is water vapour as might be the case with a bathroom water heater, or combustion products, as may be the case when a defective flue is in use. It is very surprising, therefore, that the subject has received little attention since Masterman et al (34) studied the subject more than thirty years ago.

Only recently have Reed et al (35, 36, 37) carried out a detailed study of the influence of vitiation of primary and secondary Air both separately and together on critical boundary velocity gradient, burning velocity, and critical flame stretch factors. In order to express their results in a convenient manner, they have defined a parameter β as follows:-

$$\beta = \frac{\text{Critical boundary velocity gradient under vitiated conditions}}{\text{Critical boundary velocity gradient under non-vitiated conditions}} \qquad (1.11)$$

It follows from equation (1.10) that

$$\beta = \frac{\left(\rho \frac{Cp\ Su^2}{k}\right)_{\text{Vitiated}}}{\left(\rho \frac{Cp\ Su^2}{k}\right)_{\text{Non-vitiated}}} \qquad (1.12)$$

Reed et al have used this relationship for the case in which only primary vitiation occurs to predict the effects of vitiation on blow-off. They have obtained fair agreement between predicted and experimental values of β within the limits of accuracy to which Reed has suggested flame stretch factor can be determined ($\pm$ 60%). This comparatively high figure is attributable largely to the wide variations in values of burning velocity cited in the literature which may be as great as $\pm$ 20% of the currently accepted values.

It has been shown in section 1.2.2 that the flame stretch equation (1.10) cannot predict the behaviour when the secondary atmosphere is not Air. It follows therefore that the same equation cannot be used to predict the influence of vitiation of the secondary atmosphere. The parameter β has been expressed as a function of fuel concentration for a variety of vitiants. It is interesting to note that a substantial reduction of Oxygen concentration in the secondary atmosphere is required before any influence is noted on fuel-lean flames. This reduction is greater than 6% when the vitiant is Nitrogen although slightly less than this when the vitiant is Carbon Dioxide. However, this trend was not reflected when flames richer in fuel than stoichiometric were studied, for Carbon Dioxide caused a far greater reduction in stability than Nitrogen. The effect is to be expected, for vitiation will reduce the heat release rate and hence the temperature in the secondary reaction zone. This in turn will reduce heat transfer to the primary reaction zone and hence stability will also be reduced.

Finally the effects of simultaneous primary and secondary vitiation have been studied and experimental results compared with predictions from the individual effects of primary and secondary vitiation already characterised. Considerable success has been achieved in the agreement of the predicted and experimental values of β .

1.2.4 Studies of Inverted Flames

To this point, the work discussed has been confirmed to the subject of authodox burner stabilised flames. In this type of flame, the unburned gases are on the inside of the cone and the burned gases on the outside. The latter can thus readily mix with the surrounding atmosphere. It is possible, however, to produce an inverted flame in which the situation is effectively reversed. Such flames are normally stabilised on the ends of wires mounted in tubes or on thin plates mounted between a pair of narrow parallel channels of sufficient length to eliminate "end effects". They should not be confused with devices used to produce a "bluff body" stabilised flame, the fundamental stabilising mechanism of which is entirely different. The latter depends on turbulence engendered by the body in its wake to stabilise the flame. The former type use the wire or plate to produce a laminar flow régime condusive to the stabilising of a flame.

The study of inverted flames is of significance for two reasons. Firstly, it produces a means of studying the behaviour of a flame while eliminating the influence of the secondary atmosphere on the stabilising region (Inverted flame studies are frequently made in a Nitrogen atmosphere as this facilitates easier stabilising of the flame). For this reason, inverted flames have been described as the purest practical instance of the flame stretch concept. A second reason for interest in inverted flames, particularly those stabilised on parallel slot burners, concerns flame front interaction. It has been suggested that the situation may be likened to two interacting flame fronts separated by a distance equal to the thickness of the plate (38). Little work has been carried out on this subject to date, although this, among other aspects of flame interaction has been studied by Ewins (39, 40).

The earliest work by Lewis and Von Elbe with wire in tube burners has already been discussed. They calculated flame stretch factors for lean natural gas flames and found that the critical flame stretch factor (which they called the Karlovitz Number) decreased as fuel concentration was increased from $X = 0.62$ to $X = 0.85$. The values of critical flame stretch factor were all greater than unity, however, which is substantially greater than the corresponding value for a "bunsen" type flame of a similar composition.

Edmondson and Heap (28) have studied the influence of plate thickness on the critical boundary velocity gradients of Methane-Air flames stabilised on parallel slot burners. Not only did they observe increased stability as the plate thickness was decreased, but they also claim to have noted that the maximum stability occurs with increasingly fuel-lean mixtures. On inspection, however, it seems that this latter observation is not justified from the experimental data and arises only from the lines they have chosen to draw through the data points on their graphs. The author feels that no increase in standard deviation would have been observed if lines through all the data points reaching a maximum critical boundary velocity gradient at about X = 1.04 had been constructed. Variation of slot width was found to have no influence on stability. This is to be expected and is in close agreement with the observations of Lewis and Von Elbe who varied both the wire and tube diameters of their burners.

Edmondson and Heap (41) extended their work to other fuels and have correlated their data using the flame stretch principle. It is difficult to draw any firm conclusions from their results as they have restricted their work to one burner only. Nevertheless, it is apparent that unlike Lewis and Von Elbe, they have observed blow-off to occur at a comparatively constant value of critical flame stretch factor. From their work already discussed it would be expected that increased plate thickness would increase the critical boundary velocity gradient for all fuel gases and the value of critical boundary velocity gradient for zero plate thickness might be found by extrapolation. Edmondson and Heap have explained this phenomenon by suggesting that the flow lines of the gas emerging from the burner port adjacent to the plate, divert to fill the gap above the plate and thus the correct boundary velocity gradient is lower than that calculated. As the plate thickness is reduced the error also reduces.

The correlation of critical flame stretch factor with fuel concentration is far less good for inverted flames than for cylindrical burners and although it seems likely that flame stretch is the controlling blow-off mechanism, a considerable quantity of work will be required before any firm conclusions can be drawn as to the nature of a general correlation. A possible explanation of the poor correlation may be in the presence of small eddies above the wire or plate. These have been demonstrated to exist by both Lewis and Von Elbe (7) and Dugger and Gerstein (42). They lie directly below the flame front and so cannot influence the laminar nature of the flame nor the significance of the boundary velocity gradient. They may, however, influence the heat transfer from the primary flame front to the unburned gas mixture in the region of maximum flame stretch and as wire or plate thickness is likely to influence the size and indeed the very existence of these eddies, they may in some way contribute to the variation in critical flame stretch factor with wire or plate thickness.

1.2.5 The correlation of blow-off data from cylindrical burners

Reed's original test of the flame stretch theory was carried out using a large quantity of data from a number of different workers which was analysed statistically to obtain the basic flame stretch correlation. It was left to Edmondson and Heap (27, 28) to investigate the precise form of the relationship between fuel concentration and critical flame stretch factor for the blow-off of laminar aerated flames from cylindrical burners using a variety of fuel gases. In order to do this it was necessary to determine consistent burning velocity data as values published in the literature sometimes vary by as much as ± 20% of the currently accepted values. Edmondson and Heap (43) did this for a number of different gases employing a "flat flame" technique. A recent study of most of the well known methods of burning velocity determination has thrown considerable light on the inherent inaccuracies of each method (44). This work suggests that the method employed by Edmondson and Heap may give values as much as 20% below the correct value which will in turn lead to the under-estimation of critical flame stretch factor by as much as 40%. Consequently, the absolute values of critical flame stretch factor published by these authors are substantially different from those predicted by Reed's correlation. This does not detract from their value as a guide to the true nature of the critical flame stretch factor-fuel concentration relationship.

They have found a series of closely related correlations for different fuel gases rather than the single correlation which is predicted by the theory. The highest values of critical flame stretch factor have been exhibited by Ethylene for any particular fuel concentration. Reed has proposed that if $X < 1.0$, the critical flame stretch factor is independant of fuel concentration and has a constant value of 0.23 (equation 1.10). In contrast, however, Edmondson and Heap have found that progressively increasing values of critical flame stretch factor are observed as X approaches unity.

Reed has already provided a possible explanation for both of these phenomena. Clearly, if preferential diffusion is to account for the anomolous behaviour of Hydrogen, it must be expected to play a minor role with all fuel gases and would be expected to influence the flame stretch correlation. Of the gases Edmondson and Heap tested, Ethylene would be expected to most readily preferentially diffuse to the stabilising region and it is this gas that was observed to exhibit the highest values of critical flame stretch factor. The rapid increase in critical flame stretch factor with fuel concentration for $X > 1.0$ is attributed to heat transfer from the secondary combustion zone to the primary flame front. In practice, a certain amount of secondary combustion will take place even in a fuel-lean flame and this will increase as X approaches unity. Clearly then, this will be observed in the form of a progressively increasing critical flame stretch factor although the rate of increase with fuel concentration will be small in comparison with that in the fuel rich region.

1.2.6 Conclusions

It may be concluded that although equation (1.10) does not give a precise means of predicting critical flame stretch factor, it does provide us with the best means available for characterising the blow-off behaviour of laminar aerated flames. The only situation that cannot be characterised by this equation exists when Hydrogen is present in the fuel gas. Whether this method can be extended to non-aerated systems is not clear, although there is no theoretic reason for precluding such a step. Reed (14) has attempted to correlate data for Methane-Oxygen and Propane-Oxygen flames and these have given rise to critical flame stretch factors differing considerably from those predicted by the theory. Such data for laminar flames must have been obtained on extremely small burners and it has already been pointed out in section 1.1.3 that such burners are known to give erroneous values of critical boundary velocity gradient. No general conclusion can be drawn on this point therefore from these isolated observations.

1.3 Preferential Diffusion

1.3.1 Observation of Phenomena associated with Preferential Diffusion

1.3.1.1 Flame Front propagation in tubes

Observations of various phenomena attributable to preferential diffusion have been made throughout the last seventy years and a few of the more relevant examples to be found in the literature are cited in the following paragraphs.

Coward and Brinsley (45) observed that when they burned Hydrogen-Air mixtures in propagation tubes, that a portion of the Hydrogen remained unburned. Clusius et al (46, 47) observed the limits of flamability for upward propagation of a flame front in mixtures of Hydrogen and Oxygen, and Deuterium and Oxygen. They found that the lowest Hydrogen concentration permitting a flame to travel throughout the length of the tube was 3.8% whereas the figure was 5.3% when Deuterium was the fuel gas. The ratio of these minimum values was almost exactly that of the diffusivities of Hydrogen and Deuterium in Oxygen. In a mixture of Deuterium, Hydrogen and Oxygen, the Hydrogen was found to burn preferentially.

Manton et al (48) have photographed spherical flames under conditions of both isotropic and non-isotropic propagation. Non-isotropic propagation is the term normally used to describe the situation in which some kind of cellular or polyhedral flame structure is formed. They concluded that the propagation was non-isotropic when the deficient component of a mixture also possessed the larger diffusivity. Markstein has carried out an investigation into the upward and downward propagation of cellular flames in tubes (49, 50) using a variety of fuel gases. In this work he has correlated cell size against pressure, fuel concentration, and fuel molecular weight.

1.3.1.2 Burner stabilised flames

Polyhedral flames have been observed where both Hydrogen and Hydrocarbon fuels are burned in aerated flames. Many workers have commented on these phenomena and typical of the observations made were those reported by Smith and Pickering (51) who observed and photographed rich Propane-Air flames, and Broida and Kane (52) who observed open topped weak Hydrogen-Air flames.

It may be concluded that observations were confined to Hydrogen flames leaner than stoichiometric and Hydrocarbon flames richer than stoichiometric and also that the number of sides that the flame possessed varied with fuel concentration, burner size, and the nature of the fuel gas.

1.3.1.3 Explanation for the formation of non-isotropic flames

Gaydon and Wolfhard (53) have suggested that "differential diffusion effects" can account for the majority of incidences of non-isotropic flame fronts but they point out the striking similarity between the visual appearance of cellular flames and the patterns caused by convective heat transfer in liquids observed by Prandtl (54). Recent mathematical models have demonstrated the way that both heat transfer and diffusion can play an important part in governing stability characteristics of flame fronts but that heat transfer by conduction plays a more important role than by convection. The term "differential diffusion effects" is a particularly good one as it is apparent that "preferential diffusion" has been used to describe a variety of phenomena with some confusion having arisen as a result.

Concentration measurements have been made in laminar flames where it was suspected that preferential diffusion was taking place and composition shifts have been detected (55-59). Perhaps the most thorough study has been made by Markstein (59) of a 1.44 stoichiometric Propane-Air flame. He has plotted concentration profiles through the flame front in a "valley" and a "ridge" of a polyhedral flame. The species he has plotted include Oxygen, Hydrogen, Methane, C_2 and C_3 saturated and unsaturated hydrocarbons, and carbon monoxide. He has shown that considerable differences exist in the profiles of the different regions.

1.3.2 The influence of non-isotropic flame structure on combustion characteristics.

Harris and Lovelace (60) have pointed out that the varying fuel concentration in the flame front of polyhedral flames will engender variations in burning velocity and hence it will not be possible to predict stability characteristics from correlations developed from observations of isotropic behaviour. It is clearly essential therefore to know if the flame being studied is isotropic or non-isotropic and if the flame is not readily visible to the naked eye, as is sometimes the case with fuel lean Hydrogen-Air mixtures, a means must be found of projecting an image of the flame front.

M yers has also pointed out (61) that it is not possible to readily characterise the heat transfer by radiation from non-isotropic flames.

1.3.3 Characterising Preferential Diffusion by Mathematical Models

Early mathematical models of one dimensional flame front propagation made a number of simplfying assumptions and failed to take account of the possibility of preferential diffusion. Many of these have been reviewed by Evans who has cited 116 references on such models (62).

Landau (63, 64), Emmons (65) and Eckhaus (66, 67) all made important steps forward by developing equations to characterise heat, mass and momentum transfer as well as chemical reaction. They applied a disturbance to the flame front and determined the effects of this by a perturbation analysis. Markstein (72) has discussed the work of Eckhaus which is itself based on the work of Landau and Emmons.

All these workers have been forced to study idealised situations because of the lack of available chemical reaction mechanism data. Recently Dixon-Lewis (68) has succeeded in characterising the Hydrogen-Air reactions sufficiently to enable him to reliably predict burning velocity data with a considerable degree of accuracy.

The most recent and sophisticated attempt at analysing the influence of a light molecular species on flame front propagation has been carried out by Parlange (69). He studied three situations in which the light mobile species was present. These were:-

(i) When this species was present in the unburned gases in defect.

(ii) When it was present in the unburned gases in excess.

(iii) When it was formed as a product of combustion.

He found it necessary to make many of the simplifying assumptions made by previous workers about the physical and transport properties and then developed the various transport equations applicable. He next carried out a perturbation analysis disturbing the flame front slightly and seeing if this disturbance decayed with time. From this, he developed stability criteria for each of the three cases, (ii) and (iii) being mathematically similar. Heat conduction and molecular diffusion mechanisms were shown to be fundamentally similar, both influencing burning velocity by means of the chemical reactions taking place. Once again this emphasises the need for reliable chemical kinetics data and only qualitative results can be obtained without such data.

1.3.4 Conclusions

In the past, preferential diffusion has been studied and discussed only in situations in which it is believed to give rise to the formation of non-isotropic flame fronts. Because of this, it appears that some authors believe the two phenomena to be inseparable (28). However, there is no theoretical or experimental evidence to suggest that the absence of a non-isotropic flame structure is an indication of the absence of preferential diffusion. The criteria Parlange has layed down are criteria for flame front instability and not preferential diffusion itself.

It is perhaps because of this basic misconception that the subject of preferential diffusion has been neglected quantitatively in all blow-off theories to date.

Section 2.

ANALYSIS OF THE PREFERENTIAL DIFFUSION PROCESS IN LAMINAR FLAMES.

2. ANALYSIS OF THE PREFERENTIAL DIFFUSION PROCESS IN LAMINAR FLAMES.

2.1 The preferential diffusion mechanism

An understanding of the mechanism is essential in order to characterise the preferential diffusion process. Markstein (72) has pointed out that diffusion of reactants in the preheat and reaction zones can be caused only by concentration gradients that are created by diffusion of combustion products into the unburned gas. This point appears not to have been recognised by most authors, although Manton et al (48) have clearly done so. The process is therefore one of counter-diffusion and a mobile molecular species must be present in the combustion products as well as the unburned gas mixture in order for its effects to be significant.

The products of combustion are likely to consist of a mixture of the inert constituents of the primary fuel-oxidant mixture, oxides of carbon, water vapour, and small concentrations of oxides of Nitrogen when air is the primary oxidant. No other molecular species are likely to be present in sufficient concentrations to provide a driving force for significant mass transfer. Of these molecular species, only water vapour has a molecular weight significantly lower than that of Nitrogen which will constitute the bulk of the gas when air is the primary oxidant. In his study of a Methane-Air flame, Datta et al (15) found water vapour in the preheat zone of the flame significantly further into the unburned gases than any of the other species formed by chemical reaction.

Consider a flame front which meets the bulk gas flow at an angle ϕ (Fig 2.1). Diffusion of mobile combustion products will occur in a direction at right angles to the flame front causing a concentration gradient in this direction of all molecular species present in the unburned gas mixture. There will, however, clearly be a greater

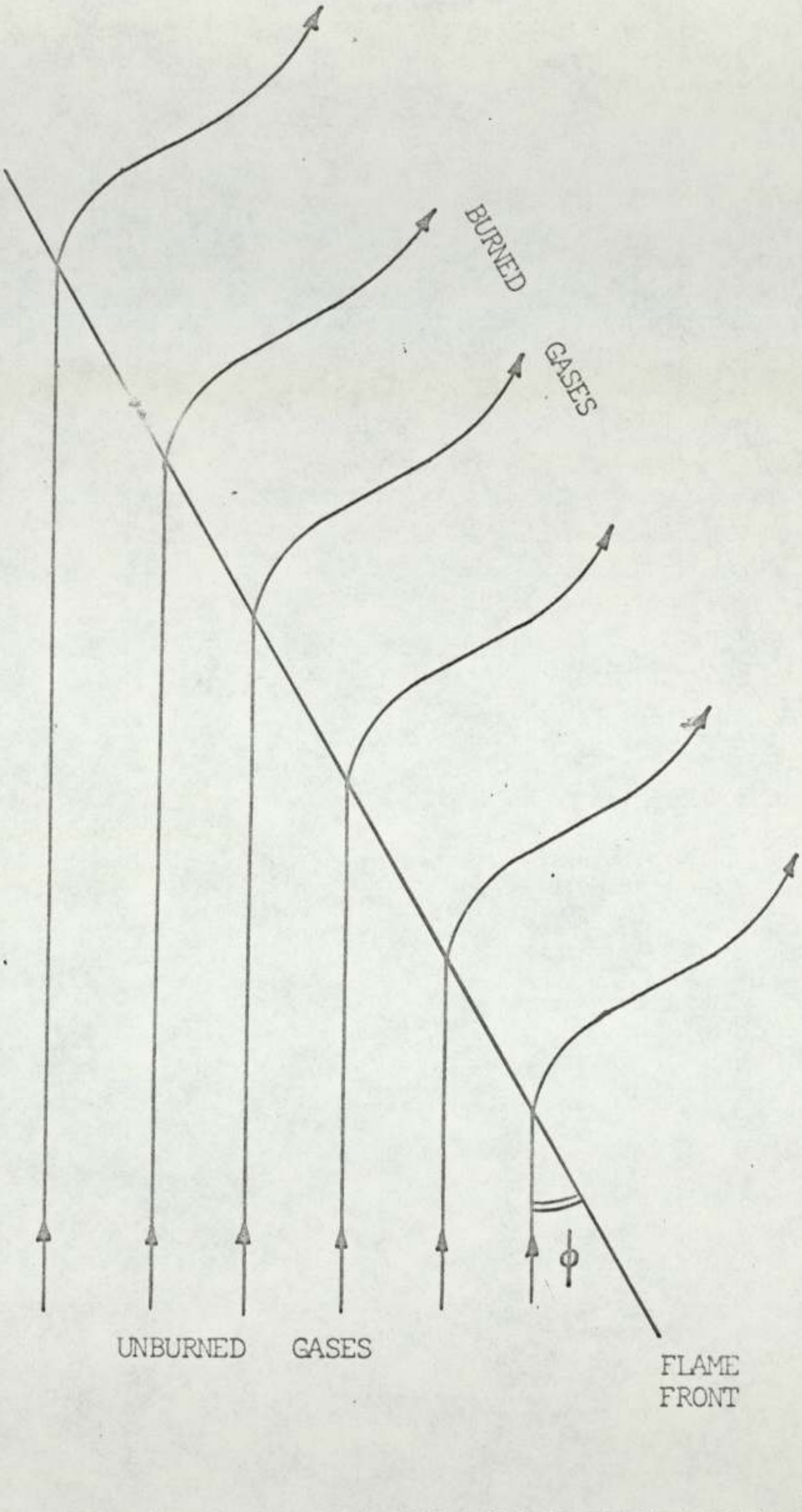


DIAGRAM OF FLOW LINES TRAVERSING LAMINAR FLAME FRONT

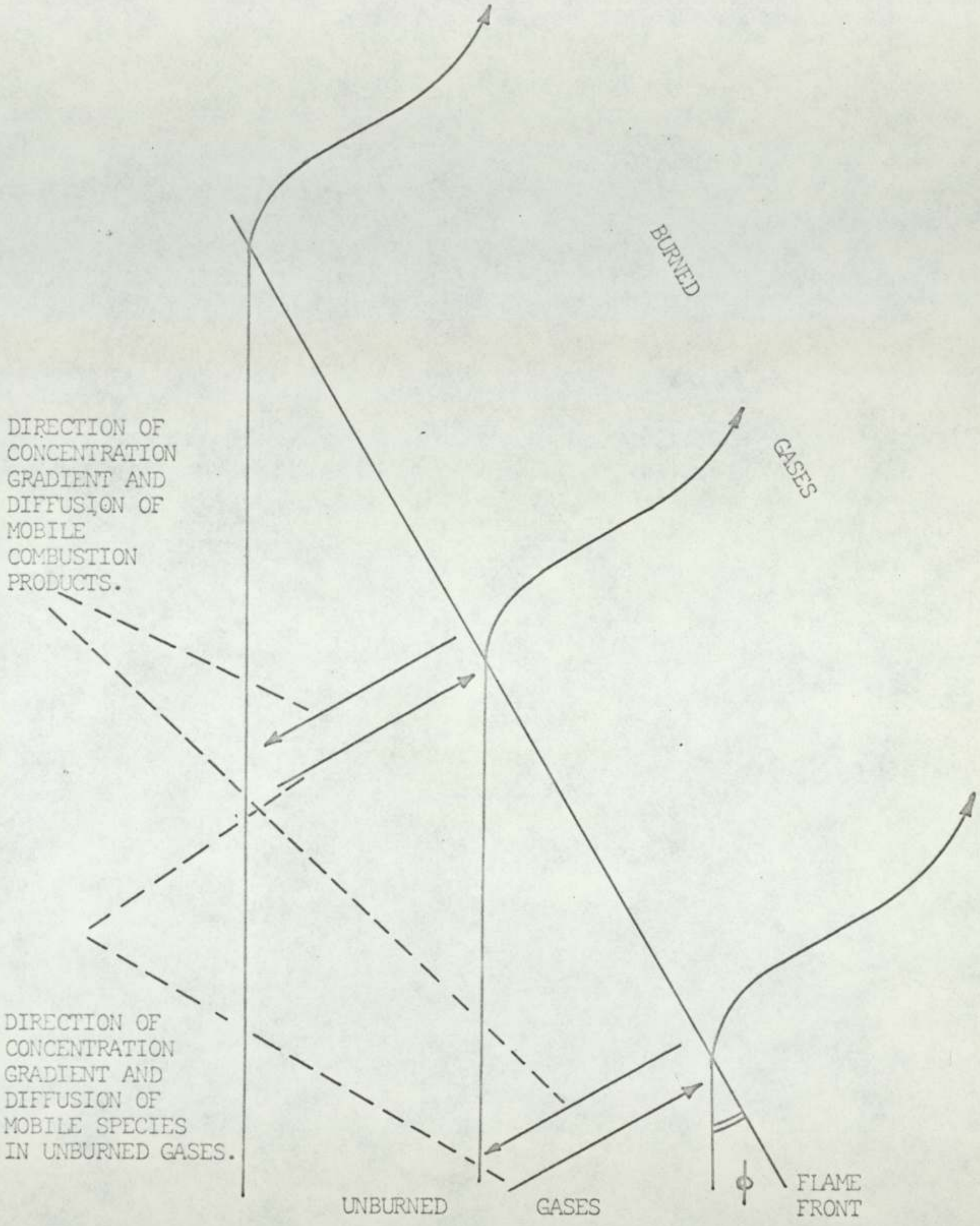
FIG. 2.I

concentration gradient for those species consumed in the combustion reactions. Thus a light molecular species is likely to diffuse towards the flame front in the direction of the driving force, i.e. the concentration gradient. This direction will only be that of the bulk gas flow when $\phi = 90^\circ$.

Where a section of flame front of constant angle ϕ exists, the loss of a light species from a flow line will be exactly balanced by the gain of that species from an adjacent flow line as the flame front is approached. Thus there will be no concentration variations in a direction parallel to the flame front (Fig 2.2). In practice the idealised situation illustrated in Figs 2.1 and 2.2 is unlikely to exist as preheating of the unburned gases will distort the flow lines as the flame front is approached gradually increasing ϕ. However, the value of ϕ is unlikely to vary along the flame front provided there is no flame front curvature present, and so the principle remains unaltered.

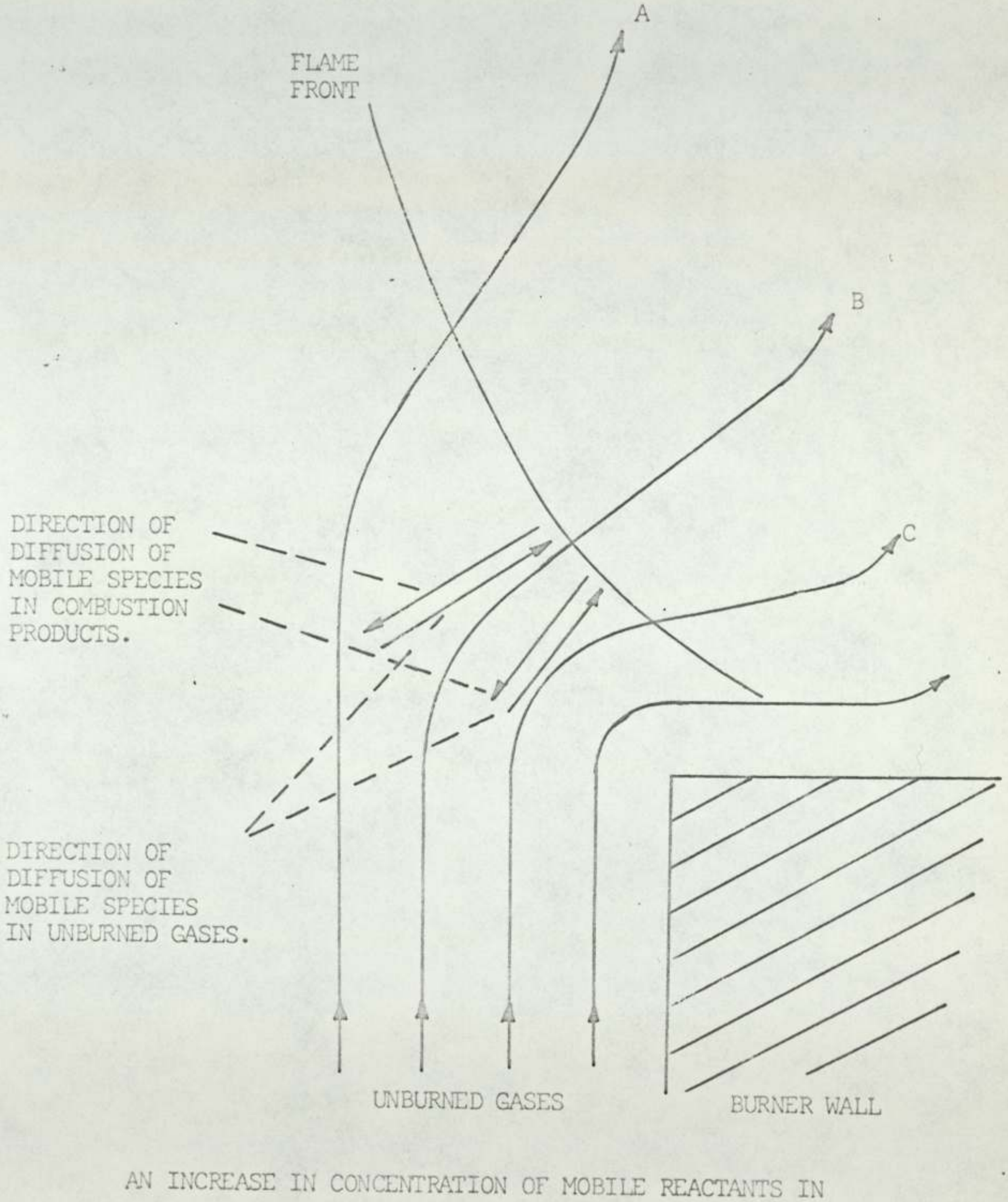
Consider now the region of flame front curvature at the base of a laminar aerated burner stabilised flame (Fig 2.3). Flow line B passes through the point of stability. This is the only point on the flame front at which a flow line passes through it at right angles to the tangent to the flame front and at which the gas velocity exactly equals the local burning velocity. Flow lines A and C on either side of B diverge from B as they pass through the preheat and reaction zones. A concentration driving force therefore exists for the migration of the light molecular species in the combustion products which will occur from flow line B to flow lines A and C by molecular diffusion. Similarly the light molecular species in the unburned fuel-oxidant mixture will diffuse towards flow line B from flow lines A and C.

The directions of these fluxes is indicated on Fig 2.3. It may be



CONCENTRATION GRADIENTS AND FLUXES ASSOCIATED WITH FLOW RÉGIME INDICATED IN FIG. 2.I

FIG.2.2



AN INCREASE IN CONCENTRATION OF MOBILE REACTANTS IN FLOWLINE B IS CAUSED BY MIGRATION OF MOBILE MOLECULES FROM FLOWLINES A AND C.

CONCENTRATION GRADIENTS AND FLUXES IN STABILISING REGION OF A LAMINAR AERATED FLAME.

FIG. 2.3

concluded that flame front curvature itself is not a pre-requisite for preferential diffusion to occur, but that the same type of flow régime is likely to give rise to the presence of both phenomena.

It has already been indicated that it is the behaviour at the point of stability which governs the blow-off of the flame, and so the migration of a light species towards flow line B will influence the chemical, transport, and physical properties governing blow-off, i.e. Burning Velocity, and the Density, Specific Heat and Thermal Conductivity of the unburned gas mixture.

2.2 The influence of Preferential Diffusion on the flame stretch correlation

In the calculation of physical and chemical properties for the evaluation of flame stretch factors, the compositions of the unburned gases at the point of stability are assumed to be those of the bulk of the fuel-oxidant mixture. It has been shown that when preferential diffusion occurs, this assumption is not justified and an indication of how the assumption will influence the flame stretch correlation may be readily assessed. The errors arising in the evaluation of density, specific heat, and thermal conductivity would be small in comparison with the accuracy to which a flame stretch factor may be evaluated. Burning velocity is extremely sensitive to small changes in concentration, however, e.g. in some systems, a 2% change in Hydrogen concentration may give rise to a change in burning velocity of almost 50%. Flame stretch factor is inversely proportional to the square of burning velocity and so this could give rise to substantial errors in critical flame stretch factor which would be quite unacceptable for burner design purposes.

If burning velocity were the only parameter influenced to a significant degree in blow-off considerations, however, values of critical flame stretch factor predicted by Reed's theory would be too high where an increase in fuel concentration gave rise to an increase in burning velocity, and too low when the converse was true. This is not consistant with the experimental data, however, for increases in flame stretch factor are observed in both fuel-weak and fuel-rich systems. The reason for this is clear. In fuel-rich systems, preferential diffusion will increase the fuel concentration giving rise to a decrease in burning velocity but also an increase in secondary combustion and hence an increase in heat transfer from the secondary to the primary reaction zones. This heat transfer process has already been shown to be responsible for the rapid increase in critical flame stretch factor with fuel concentration in fuel-rich systems by Reed and is clearly of equal importance in considerations of the influence of preferential diffusion.

A method is therefore required to characterise the preferential diffusion process and assess the degree to which it will influence Critical Boundary Velocity Gradient.

2.3 Quantitative assessment of the preferential diffusion process

From the mechanism discussed in Section 2.1 it may be deduced that the following variables are likely to influence the degree to which preferential diffusion occurs.

(i) D_1 - Diffusivity of the mobile molecular species in the combustion products $\left[\text{Dimensions} - \frac{L^2}{T}\right]$

(ii) x_1 - Concentration driving force of this species expressed as a mole fraction [Dimensionless]

(iii) D_2 - Diffusivity of the mobile species in the unburned gases $\left[\text{Dimensions} - \frac{L^2}{T}\right]$

(iv) x_2 - Concentration driving force of this species expressed as a mole fraction [Dimensionless]

(v) ρ - Density of unburned gas mixture $\left[\text{Dimensions} - \frac{M}{L^3}\right]$

(vi) Cp - Specific Heat of unburned gas mixture $\left[\text{Dimensions} - \frac{H}{M\theta}\right]$

(vii) k - Thermal Conductivity of unburned gas mixture $\left[\text{Dimensions} - \frac{H}{\theta LT}\right]$

It may be postulated that the degree to which preferential diffusion will occur may be characterised by a dimensionless group (F) termed the preferential diffusion factor.

By the "π" theorem of dimensional analysis, which has been discussed in detail by Massey (73) it may be stated:-

$$(\pi_1 \;\; \pi_2 \;\; \ldots . \;\; \pi g) = 0 \qquad (2.1)$$

where π = dimensionless group

and $g = n - m$

where n = number of independant variables

m = number of fundamental magnitudes

In this case $n = 8 \quad m = 5 \quad \therefore g = 3$

Let $\pi_1 = (D_1)^{n_1} \; (k)^{n_2} \; (\rho)^{n_3} \; (Cp)^{n_4}$

In terms of fundamental magnitudes:-

$$\left[L^o \; M^o \; T^o \; H^o \; \theta^o\right] = \left[\frac{L^2}{T}\right]^{n_1} \left[\frac{H}{LT\theta}\right]^{n_2} \left[\frac{M}{L^3}\right]^{n_3} \left[\frac{H}{M\theta}\right]^{n_4}$$

Then, by comparison of dimensional coefficients:-

$L : 0 = 2n_1 - n_2 - 3n_3$

$M : 0 = n_3 - n_4$

$T : 0 = -n_1 - n_2$

$H \; : \; 0 \; = \; n_2 - n_4$

$\theta \; : \; 0 \; = \; -n_2 - n_4$

Thus $n_1 = -n_2 = n_3 = n_4$

if $n_1 = 1$, $\pi_1 = \dfrac{D_1}{k/\rho C_p}$

Similarly $\pi_2 = \dfrac{D_2}{k/\rho C_p}$

By definition $\pi_3 = F$

We wish to incorporate all the relevant variables into the analysis and as x_1 will relate to π_1 and x_2 will relate to π_2 the concentration terms will be considered as part of the groups π_1 and π_2. Furthermore, Fick's first law of diffusion states:-

$$J \propto D \frac{dx}{dy} \tag{2.2}$$

where J = Flux

D = Diffusivity

$\frac{dx}{dy}$ = Conc. gradient driving force

and so x_1 and x_2 will be raised to the same power as D_1 and D_2. Thus:-

$$\Phi\left[\frac{D_1 x_1}{k/\rho C_p} \; , \; \frac{D_2 x_2}{k/\rho C_p} \; , \; F \right] = 0 \tag{2.3}$$

This may be rewritten:-

$$F = \Phi^1\left[\frac{D_1 x_1}{k/\rho C_p} \; , \; \frac{D_2 x_2}{k/\rho C_p} \right] \tag{2.4}$$

Preferential Diffusion factor is a measure of fluxes and it therefore follows from Equation (2.2) that

$$F \propto \left(\frac{D_1 x_1}{k/\rho C_p}\right) \left(\frac{D_2 x_2}{k/\rho C_p}\right) \tag{2.5}$$

For convenience F will be defined as follows:-

$$F = \left(\frac{D_1 x_1}{k/\rho C_p}\right)\left(\frac{D_2 x_2}{k/\rho C_p}\right) = \frac{D_1 x_1 D_2 x_2}{\eta_o^2 \quad Su^2} \tag{2.6}$$

The constant of proportionality which will arise from Equation (2.5) will be incorporated into the stability correlations in which F is present. It may be noted that F can be considered as the product of two Lewis Numbers and two concentration terms. Thus Equation (2.6) becomes

$$F = Le_1 x_1 \quad Le_2 x_2 \qquad (2.7)$$

The Lewis Number is normally used as a measure of the ratio of molecular to thermal diffusivities where "thermal diffusivity" in this instance refers to the analogous heat transfer process to mass transfer by molecular diffusion and not the mass transfer process induced by a temperature gradient driving force.

It would be possible to attempt to relate the preferential diffusion factor to one of the following:-

(i) A change in concentration of the mobile molecular species in the unburned gas mixture

(ii) A change in burning velocity

(iii) A change in flame stretch factor

(i) would have to be related to a change in stability which would present considerable problems in itself and so this idea was discarded. Of the remaining possibilities only flame stretch factor takes into account the changes in both burning velocity and heat transfer from the secondary combustion zone when this is significant. Furthermore, correlation with (iii) would provide the simplest and most easily applied solutions for both burner design and the assessment of interchangability of fuel gases on existing burners.

2.4 The Preferential Diffusion Factor as applied to aerated flames

2.4.1 Mobile Species in the Fuel Gas

From Equation (2.6) it will be seen that for aerated systems the following approximation may be made:-

$$F \propto D_2 x_1 x_2$$

The values of Thermal conductivity, Density and Specific Heat are unlikely to vary greatly from those of Air as this gas will form the bulk of any primary mixture to which the flame stretch concept is applicable. The Suffix 1 will almost certainly refer to water vapour and so D_1 may also be considered constant.

x_1 will be influenced by the nature of the fuel gas, e.g. how much Hydrogen is present in each molecule, the concentration of the fuel gas, and the amount of Oxygen present, although this is only important if there is insufficient for complete combustion.

D_2 will be influenced by the molecular weight of the light species in the unburned gases.

x_2 will be influenced by the stoichiometric ratio of the fuel gas or gases, and the degree of aeration.

Table 1 has been compiled as a comparison of the values of D_2, x_1, and x_2 for stoichiometric mixtures of the two lowest molecular weight fuel gas, Hydrogen and Methane, with Air. Diffusivities were evaluated at the unburned gas temperature (in this case 20°C) as this cool region of the flame provides the greatest resistance to mass transfer. It follows that:-

$$\frac{F\ (\text{Hydrogen})}{F\ (\text{Methane})} \simeq 15.2 \qquad (2.8)$$

In flames in which Hydrogen forms only part of the fuel gas mixture, x_2 will be reduced, but x_1 is likely to retain a value of a similar order to that of a Hydrogen-Air flame as water will be formed from other Hydrocarbon fuels present in the unburned gas mixture. It has been shown

Fuel Gas	Diffusivity in Air D_2 (M^2/S)	x_1	x_2
Hydrogen	0.532×10^{-4}	0.346	0.296
Methane	approx. 0.2×10^{-4}	0.19	0.095

Table 1.

Comparison of parameters D_2, x_1 and x_2 for stoichiometric flames of Hydrogen and Methane with Air.

that the behaviour of Methane-Air flames can be predicted with reasonable accuracy by the flame stretch correlation, Equation (1.10), and so it may reasonably be deduced that as only Hydrogen has a lower molecular weight than Methane, Hydrogen is the only fuel gas likely to more readily preferentially diffuse. From the comparison of preferential diffusion factors given in Equation (2.8) it may be seen why only Hydrogen appears not to behave as predicted by the flame stretch theory of blow-off.

2.4.2 Preferential Diffusion of Oxygen

Oxygen is unlikely to preferentially diffuse in aerated systems as its diffusivity is approximately one third that of Hydrogen and less than that of Nitrogen and many light hydrocarbon fuels. Furthermore, the concentration driving force x_2 can never exceed 0.21 and is usually only a fraction of this value. The only situation in which Oxygen is likely to diffuse more readily than a fuel gas is when the driving force for such a diffusion process is sufficiently large that it overrides the effect of the low diffusivity. In such situations, however, the water vapour concentration driving force may not be large enough to enable enough Oxygen to diffuse to influence stability.

An increase in fuel gas concentration in an aerated system will always give rise to increased flame stability. An increase in Oxygen concentration, however, will not automatically do so. A flame with an increased Oxygen concentration will have a higher burning velocity than a normal flame provided the two have the same fuel concentration expressed as a fraction of stoichiometric. The diffusion of Oxygen will effectively reduce the fuel concentration, however, and this may give rise to either an increase or a decrease in burning velocity depending on whether the flame is leaner or richer than the fuel concentration at which maximum burning velocity occurs. How these factors together with the changes in heat transfer from secondary to primary reaction zones combine to influence stability can only readily be determined by the study of specific cases. As Oxygen diffusion in aerated systems is unlikely to be of any practical significance, the problem has not been pursued either theoretically or experimentally.

Apart from that cited by Reed and discussed in Section 1.2.6 and shown to be of doubtful validity, there is no data available on the correlation of blow-off characteristic of fuel-oxygen systems by the flame stretch concept. It is possible, however, that the large concentration gradients available for the diffusion of molecular species of comparatively low mobility may make preferential diffusion significant in such situations.

2.5 Experimental Programme

In order to assess the influence of preferential diffusion from a practical point of view, three pieces of experimental work have been carried out.

(i) In order to correlate calculated values of preferential diffusion factor with experimental data, a study has been made of laminar aerated Hydrogen-Propane-Air flames stabilised on cylindrical burners. Critical boundary velocity gradient and burning velocity data were obtained and correlated by the flame stretch method, and from this a technique was developed to predict the influence of Hydrogen on the blow-off laminar aerated flames. The Hydrogen-Propane-Air System was selected as it afforded the opportunity to study mixtures with burning velocities which varied by as much as a factor of ten and some of the mixtures studied would have somewhat similar properties to those which may be used as Synthetic Natural Gas. It was also known that the Propane-Air System adhered closely to the behaviour predicted by the flame stretch theory.

(ii) The prediction method developed in the first section of experimental work was tested by two techniques. Firstly, experimental data of other workers was correlated and statistically compared with data predicted by the method. Secondly, in order to test the method using a wide range of physical properties, the inert constituent in the Air (Nitrogen) was replaced in turn by Argon and Helium, and critical boundary velocity gradient and burning velocity data redetermined for the Hydrogen-Propane-"Air" System. This has also been analysed and compared statistically with predictions made by the method developed.

(iii) Hydrogen concentration profiles in the stabilising region of Hydrogen-Propane-Air flames have been studied with 60% Hydrogen in the fuel gas mixture and degrees of aeration equivalent to 0.8, 1.0 and 1.2 stoichiometric. This particular fuel gas mixture was selected as it provides a reasonable compromise between the increase in Hydrogen flux with increasing Hydrogen concentration and the corresponding decrease in preheat zone thickness in which to detect concentration changes attributable to preferential diffusion. Sampling was carried out with a quartz microprobe and a gas chromatograph was used for sample analysis.

In both sections (i) and (iii) similar data for the Ethylene-Air System was obtained for the purpose of comparison. However, in section (iii) Ethylene, rather than Hydrogen concentrations were determined.

Section 3.

EXPERIMENTAL APPARATUS AND TECHNIQUES.

3. EXPERIMENTAL APPARATUS AND TECHNIQUES.

3.1 Flow Metering

3.1.1 Choice of Flow Meters

It was required to meter accurately and blend thoroughly a maximum of two different fuel gases, supplied from high pressure cylinders, with a primary oxidant supplied either from a high pressure cylinder or a main at 70 lb/in^2 gauge. A wide range of flow rates of each gas were to be metered and so to avoid the necessity for a large number of meters for each gas a type of meter was sought which could be used for all of the flow-rates likely to be encountered in the work. Three types were considered:-

(i) Variable area flow meters.
(ii) Porous plug flow meters.
(iii) Orifice plates (subcritical jewelled type).

Of these, the subcritical jewelled orifice, the design, construction, and use of which have been investigated by Sprange (74), are likely to provide the most accurate results, as corrections for variations in temperature and pressure can be made without loss of accuracy. They are, however, not readily available and are therefore expensive, require sophisticated pressure drop monitoring equipment, and each orifice is applicable to only a very limited range of flows. Porous plug flow meters can be criticised similarly for their lack of versatility. It was therefore decided to use variable area flow meters as they are well tried and known to be reliable when calibrated carefully, are fairly inexpensive, and can be used over a wider range of flows than any of the alternatives considered. A total of nine meters were used, five of which were manufactured by Rotameter and four by Fischer and Porter. Tables 2 - 5

Number	Manufacturer	Tube type	Float	Approximate flow range (m^3/s)
F_3	Rotameter	Metric 7P	Dural	0.4×10^{-4} - 5.8×10^{-4}
F_4	Fischer & Porter	Tri-flat 1/8-16-G-5/36	1/8" Diameter Sapphire	0.052×10^{-4} - 0.83×10^{-4}
F_5	Fischer & Porter	Tri-flat 1/16-10-G-5/36	1/16" Diameter Stainless Steel	0.012×10^{-4} - 0.067×10^{-4}

Table 2 - Hydrogen Flow Meters

Number	Manufacturer	Tube type	Float	Approximate flow range (m^3/s)
F_6	Rotameter	Metric 7P	Dural	0.15×10^{-4} - 1.5×10^{-4}
F_7	Fischer & Porter	Tri-flat 1/8-16-G-5/36	1/8" Diameter Tantalum	0.033×10^{-4} - 0.5×10^{-4}
F_8	Fischer & Porter	Tri-flat 1/16-10-G-5/36	1/16" Diameter Stainless Steel	0.0092×10^{-4} - 0.375×10^{-4}

Table 3 - Propane Flow Meters

Number	Manufacturer	Tube type	Float	Approximate flow range (m^3/s)
F_1	Rotameter	Metric 10P	Korannite	0.7×10^{-4} - 6.6×10^{-4}
F_2	Rotameter	Metric 7 x P	Dural	0.11×10^{-4} - 1.35×10^{-4}

Table 4 - Primary Air Flow Meters

Number	Manufacturer	Tube type	Float	Approximate flow range (m^3/s)
F_9	Rotameter	Metric 14P	Korannite	1.67×10^{-4} - 15×10^{-4}

Table 5 - Secondary Air Flow Meters

Number	Gas	Approximate flow range (m^3/s)
F_2	21% Oxygen 79% Helium	0.104×10^{-4} - 2.1×10^{-4}
F_2	21% Oxygen 79% Argon	0.092×10^{-4} - 1.18×10^{-4}
F_7	Ethylene	0.067×10^{-4} - 0.184×10^{-4}
F_8	Ethylene	0.0098×10^{-4} - 0.0417×10^{-4}

Table 6 - Flow Meters calibrated for gases other than those listed in Tables 2 - 5

show the meters employed for each gas and their approximate volumetric flow ranges measured at atmospheric pressure and 18°C. Table 6 lists flow meters which were also calibrated for other gases over the ranges indicated.

3.1.2 Pressure and Temperature monitoring and control

High pressure cylinders were fitted with pressure regulators recommended for use with the gas in question by the gas supplier. Primary and secondary Air was supplied from the University main at 70 lb/in^2. This pressure was reduced by means of a pressure control valve to about 20 lb/in^2 and a pressure relief valve was installed to relieve at 25 lb/in^2 downstream of this valve for safety purposes. Norgren pressure regulators were installed on the flow control panel to enable accurate pressure control of fuel gases and primary oxidant. Flow control was carried out using needle valves of which the majority were 1/4 in. sized. The pressure of each gas was monitored at the flow meter outlet by means of a water manometer which could be read to an accuracy of ± 0.05 in. water gauge.

The gas temperature was also monitored on the outlet of each flow meter with a mercury thermometer calibrated to an accuracy of ± 0.1°C. This was verified by comparison with a standard thermometer. A similar thermometer was installed on the inlet to the burner to provide an accurate indication of the final temperature of the unburned gas mixture.

No temperature control was provided but the laboratory was of a sufficiently constant temperature to ensure that the final temperature of the unburned mixture was always in the range 293°K ± 3°.

The atmospheric pressure was measured by a Mercury barometer and a small correction made for the difference in siting of the barometer and experimental rig.

A detailed flow diagram is shown in Fig 3.1 and a photograph of the flow control panel is included in Plate 1. The secondary atmosphere control was not situated on the main control panel and no temperature and pressure monitoring equipment was fitted on the outlet of flow meter F.9. This was because only a rough indication of secondary atmosphere flow rate was required.

3.1.3 Gas Drying, Mixing and safety equipment

3.1.3.1 Drying

The fuel gases and primary oxidant were passed through drying towers containing silica gel. These towers were sited upstream of the flow control panel pressure regulators and a sintered glass filter was connected to the outlet of each drying tower to remove particles entrained in the gas. In order to gauge when the silica gel was spent, and also to remove any moisture diffusing into the dry gases from the manometers, a small U-tube packed with self-indicating silica gel was fitted in the line after the point at which the primary fuel-oxidant mixture was blended. When this indicated that moisture was present in the mixture, the drying agent in all three towers was replaced.

The secondary oxidant was also passed through a silica gel packed drying tower which was situated on the inlet to the secondary oxidant distributor. No filter was provided as it was felt that the distributor system would itself fulfill this function.

3.1.3.2 Mixing

The primary mixture was blended in two stages. The fuel gases were

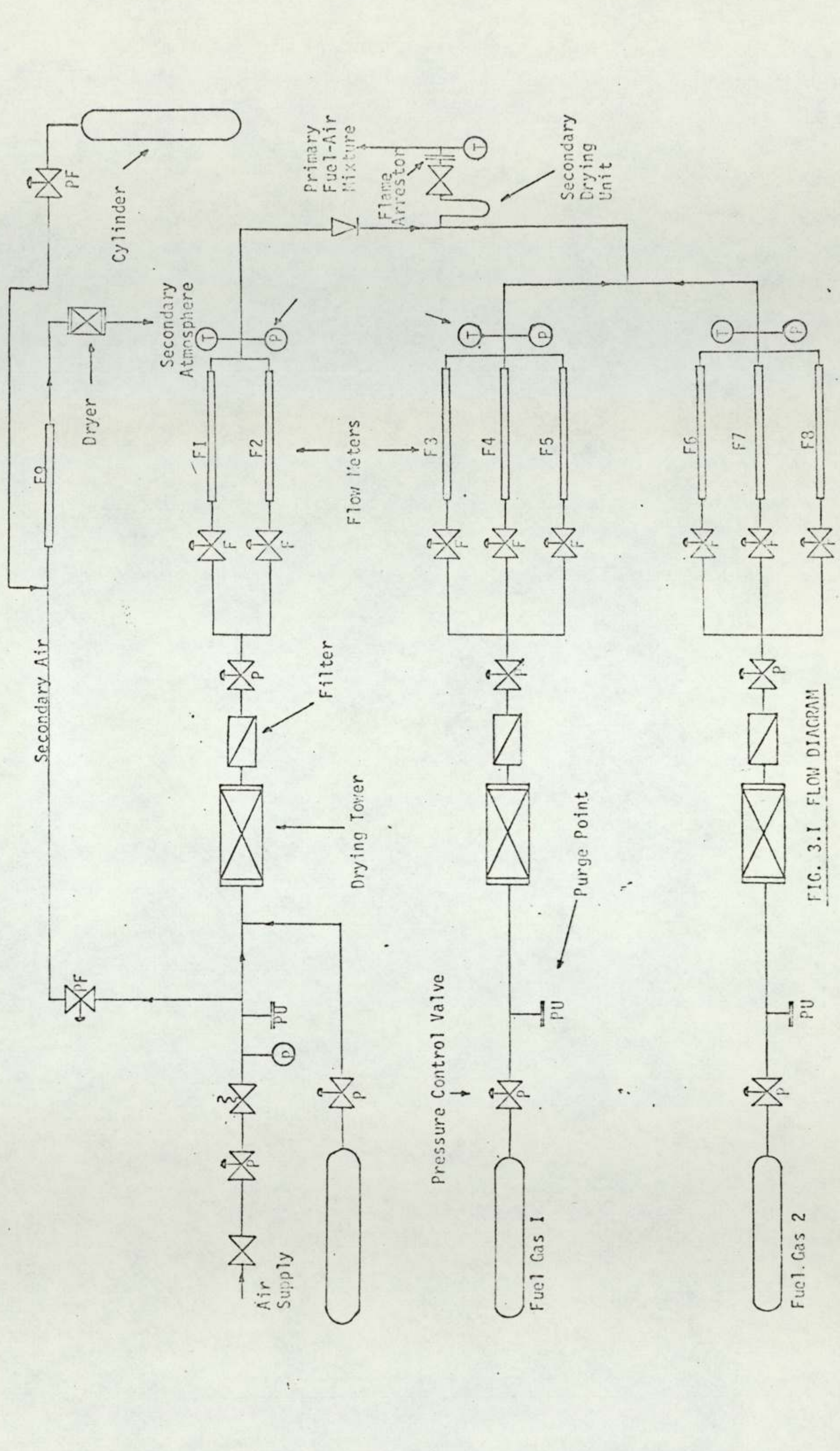


FIG. 3.1 FLOW DIAGRAM

KEY TO FLOW DIAGRAM (FIG. 3.I)

⋈	Isolation Valve.
⋈ F	Flow Control Valve.
⋈ P	Pressure Control Regulator.
⋈ PF	Pressure and Flow Control Valve or Regulator.
⋈	Pressure Relief Valve.
◁	Non-return Valve.
Ⓣ	Temperature Indicator (Thermometer).
Ⓟ	Pressure Indicator (Manometer or Bourdon Gauge).
⊠	Drying Tower.
⧄	Sintered Glass Filter.
⊣⊢	Flame Arrestor.
⊤ PU	Purge Point.

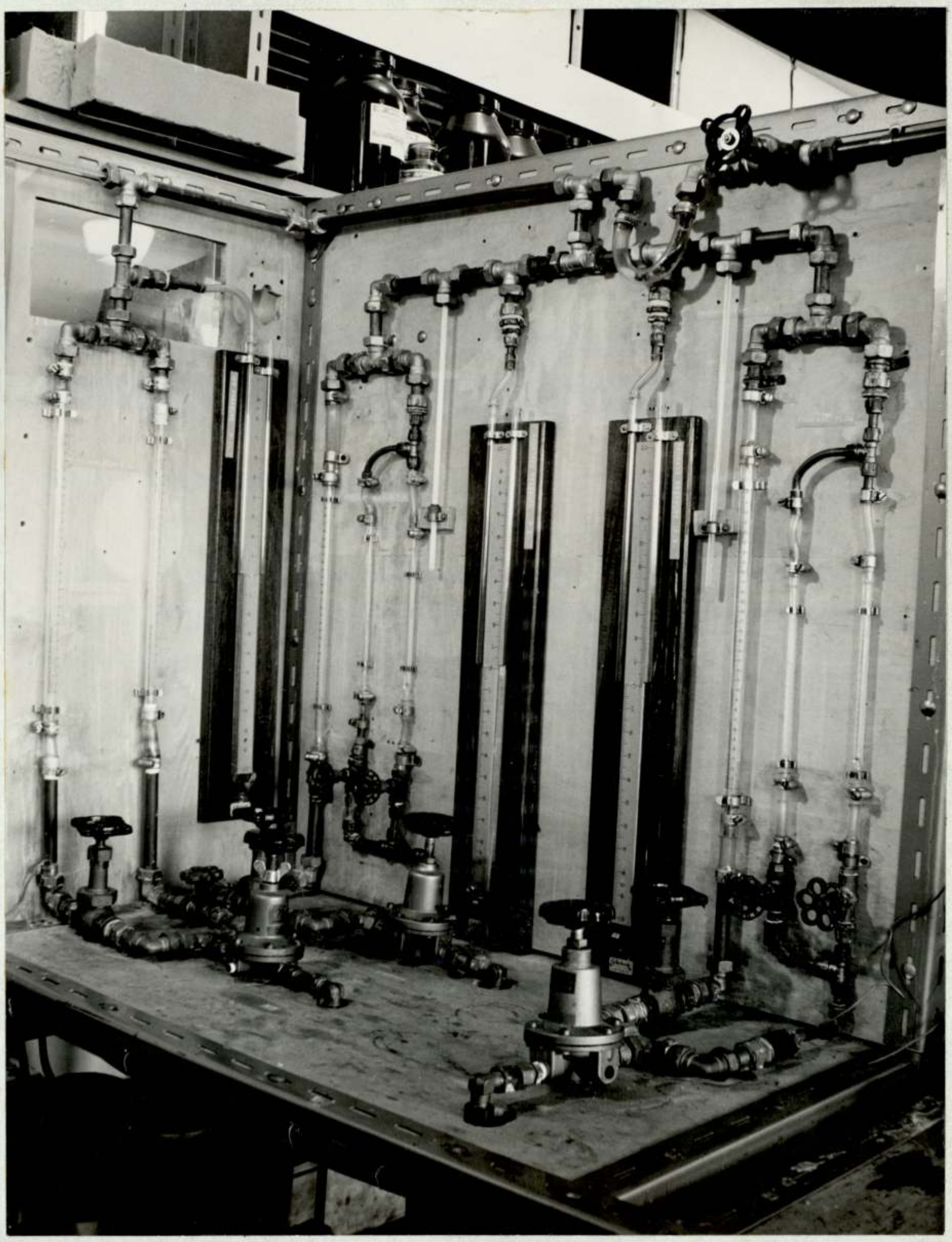


PLATE I FLOW CONTROL PANEL FOR PRIMARY FUEL-OXIDANT MIXTURES.

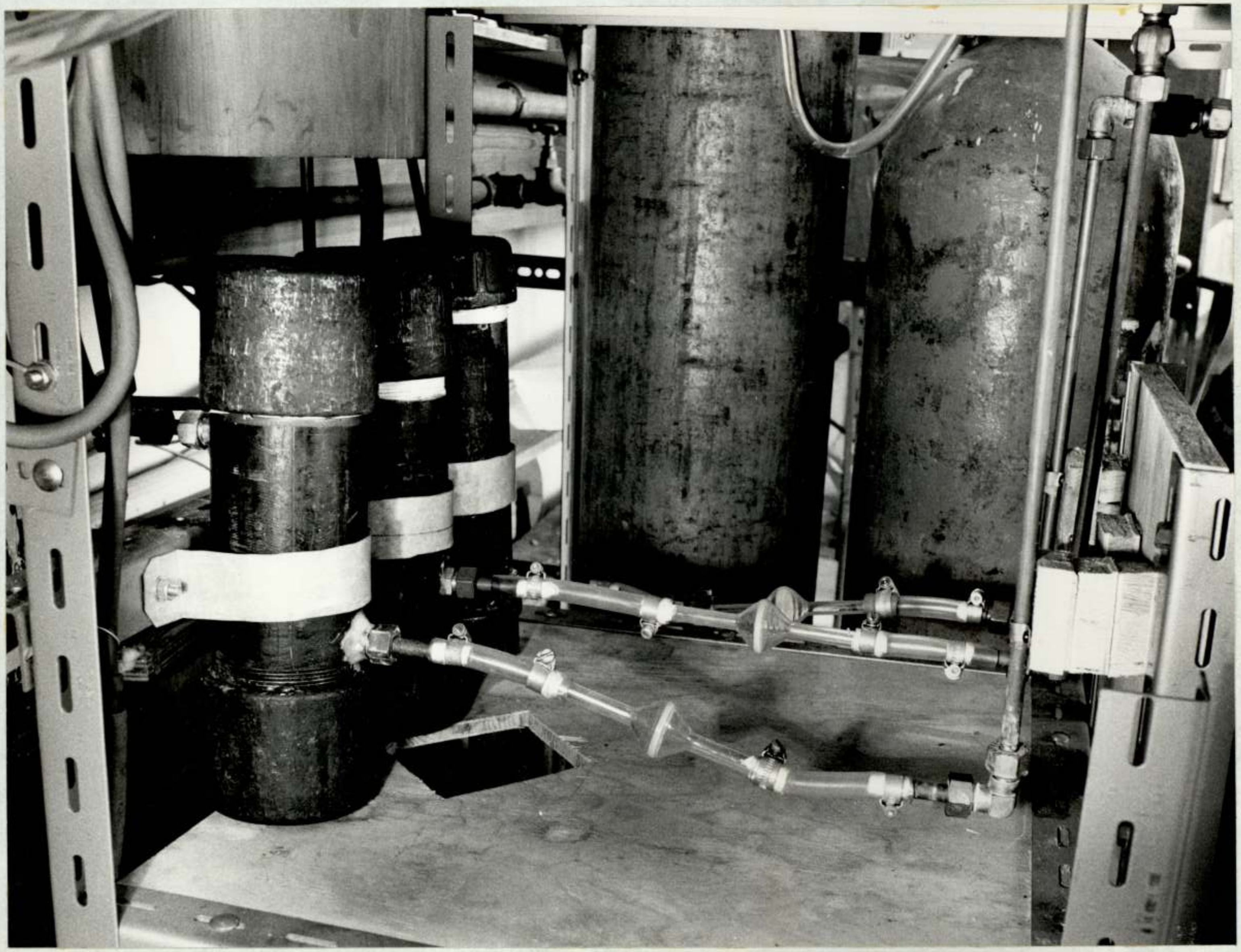


PLATE 2 DRYING TOWERS AND FILTERS. THE TWO LARGE CYLINDERS AT THE REAR OF THE FILTERS CONTAIN THE FUEL GASES.

first mixed by means of a normal "tee" compression fitting. The fuel gas mixture was then introduced into the primary Air by means of a pitot tube sited in the centre of the primary Air tube pointing upstream. The fuel-oxidant mixture then flowed through approximately 5 m. of 0.5 in. diameter copper tubing in which were fitted seven 90° elbows to ensure perfect mixing.

This long length of tube caused a delay between adjustment of flow meter settings and the corresponding change in primary mixture composition at the burner inlet. However, it was felt that this minor inconvenience was of little importance compared to the need to ensure satisfactory mixing.

3.1.3.3 Safety

Before entering the burner, the primary mixture passed through a flame arrestor manufactured by "Knitmesh" and designed for use with all fuel gases including Hydrogen. As an additional safety precaution a non-return valve was fitted in the primary Air line so as to ensure that fuel gas could not under any circumstances flow into the University compressed Air Main system.

Leak testing was carried out by pressurising the entire system with compressed Air to a pressure in excess of that used under normal operating conditions and each joint was tested with soap solution. This procedure was repeated regularly.

3.1.4 Flow Meter Calibration

Flow meter calibration curve prediction methods were specified by both manufacturers of flow meters. Because of the need for a very high degree of accuracy, however, it was decided to calibrate each flow meter experimentally with the actual gases to be metered. A standard wet gas meter was used for the purpose.

It was necessary to determine whether the moistening of the gas in the meter would be sufficient to influence the accuracy if the volumes recorded were assumed to be on a dry basis. To do this, several experiments were carried out by coupling a dry gas meter, first to the inlet, and then to the outlet of the wet gas meter. Different flow rates of a number of gases were passed through both flow meter configurations and the flow rates calculated from the volumes recorded by each meter were compared. It was concluded that when the dry gas meter was coupled to the wet meter outlet, the flow rates were similar, as expected. When the arrangement was reversed, a maximum discrepancy, of 0.2% of the dry meter reading was recorded. This figure was considered small in comparison with inaccuracies in flow metering introduced by other factors and so no corrections were made to the gas volumes recorded by the wet meter.

The flow meters were calibrated by measuring the time for the wet gas meter pointer to complete a minimum of one complete revolution. If one complete revolution of the pointer took less than two minutes, the least number of complete revolutions taking longer than two minutes was recorded. Timing was carried out using a stop watch accurate to ± 0.1 s. Each flow meter reading was reset three times and the average of the three flowrates used for plotting the calibration curves. Small flow rate increments were used so that normally about twenty-five different flows were used to plot each calibration curve. When expensive gas mixtures were being used, however, it was not possible to use as many points to plot the curve as for cheap gases because of the large volumes of gas required for such determinations. In the case of Propane, a cylinder of slightly less pure gas was obtained for flow meter calibration than was used in the experimental work itself.

Cremer and Davis (75) have indicated that when a variable area flow meter indicates the same reading for two gases, 1 and 2, of differing density, it follows that:-

$$\frac{Q_2}{Q_1} \propto \left[\frac{(\rho f - \rho_2)}{(\rho f - \rho_1)}\frac{\rho_1}{\rho_2}\right]^{1/2} \qquad (3.1)$$

where ρf = density of flow meter float.

When the fluids being metered are gases, it follows that

$\rho f \gg \rho_1$ and $\rho f \gg \rho_2$

in which case

$$\frac{Q_2}{Q_1} \propto \left[\frac{\rho_1}{\rho_2}\right]^{1/2} \qquad (3.2)$$

Equation (3.2) has been used both in the production of calibration curves and correction of indicated flow rates to values applicable to the particular conditions of temperature and pressure under which experiments were being carried out. An analysis of the maximum error arising in flow metering is given in Appendix 2 and has been estimated to be ± 1.5%.

3.2 Gas Purity

The gases detailed in table 7 were used in the course of the work. The purities quoted were those specified by the suppliers and were not verified experimentally.

Gas	Supplier	Grade	Purity
Air	University compressed Air main	-	-
Hydrogen	British Oxygen Company	High Purity	99.99%
Propane	British Oxygen Company (from Matheson Gas Corporation)	Instrument	99.5%
Ethylene	British Oxygen Company	Commercial	99.85%
21% Oxygen 79% Argon	British Oxygen Company	Special Gas Mixture	± 1% of specification
21% Oxygen 79% Helium	British Oxygen Company	Special Gas Mixture	± 1% of specification
Argon (for gas chromatograph)	British Oxygen Company	Industrial	99.995%

Table 7 - Specifications of gases used in experimental work

3.3 Determination of critical boundary velocity gradient data

3.3.1 Choice, design and construction of gas burners

Burners were required which would enable the critical boundary velocity gradient to be easily and accurately determined from a knowledge of the volumetric flowrate of the primary mixture at blow-off. For this reason it was decided to use four cylindrical tubes of different diameters sufficiently long to ensure a fully developed laminar velocity profile of gases emerging from the ports. The burners were made from drawn stainless steel tube and were attached to the primary mixture line by means of brass compression fittings. Bird, Stewart, and Lightfoot (76) have stated that to ensure a fully developed laminar velocity profile, the minimum tube length (L_E) is given by the following formula.

$$L_E = 0.035\ Re.d \qquad (3.3)$$

d = tube diameter

It may be assumed that the maximum value of Reynolds Number will be 2100 and therefore the minimum tube length will be given by

$$L_E = 73.5\ d \qquad (3.4)$$

It may be seen from the burner specifications in table 8 that some are substantially longer than required by equation (3.4). This was to enable easy interchange of the burners without the necessity for modification of the secondary atmosphere distribution system. External diameters were determined with a micrometer and internal diameters with a travelling microscope.

3.3.2 Secondary Atmosphere distribution

The burner was situated in a cylindrical tower 0.175 m diameter up which the secondary atmosphere was passed at an average velocity of

Burner Number	External Diameter (m)	Internal Diameter (m)	Length (Pipe Diameters)
1	0.00958	0.00782	115
2	0.00798	0.00615	145
3	0.00480	0.00313	290
4	0.00320	0.00154	585

Table 8 - Cylindrical Burner Data

0.02-0.04 m/s. This velocity was not critical, the only requirement being that it was sufficiently high to prevent recirculation of combustion products and sufficiently low to prevent disturbance of the flame. The tower extended to a height approximately 0.5 m above the burner port. The secondary atmosphere was distributed by a sparge ring at the base of the tower and above this was a fine wire gauge supporting a layer of 0.006 m diameter glass ballotini about 0.03 m thick. This secondary distributor was present to iron out any major flow irregularities which might be present. As there was no need for accurate flow metering of the secondary oxidant, the flow meter manufacturer's calibration for air was used and corrections made for the gas density changes by equation (3.2) when other oxidants were used.

Two windows were cut on diametrically opposed sides of the tower to enable a beam of light to be shone through the tower passing through the flame. The windows were covered with pyrex plate glass which, while not being optically perfect, were of sufficient quality for the burning velocity determination technique adopted. This will be discussed in more detail in section 3.4.3. The windows were also essential for observation of the flame when determining critical boundary velocity gradients.

3.3.3 Experimental technique

The following experimental technique was adopted. The fuel gas flow rates were set to give the required volumetric flow and ratio of the constituents. The Air was then set at a low enough value to enable a laminar flame to be stabilised on the burner. This flow rate was gradually increased making the flame progressively leaner until blow-off occurred. Sufficient time was allowed after each flow rate increase to take account of the lag between the alteration of the flow rate and the change in composition of the flame. This procedure was repeated several times and the average critical flow rate established. The following formula was then used to calculate the critical boundary velocity gradient and may be derived from the Hagen-Poiseuille equation.

$$gb = \frac{32\ Q}{\pi\ d^3} \qquad (3.5)$$

The upper limit for blow-off determinations was the onset of the transitional flow régime. This was visible by the slight pulsations of the flame which were observed when Reynold's Number exceeded 2000. As an additional check, Reynold's Number was calculated for all blow-off determinations, even when no visible sign of turbulence was observed.

To check the reproducibility of the experimental procedure, a few experiments were carried out using only one fuel component, by holding the Air flow constant and reducing the fuel gas flow rate. These were compared with the values obtained by holding the fuel flow constant and increasing the Air flow rate, but no variation in the critical flows were detectable between the two cases.

Investigations were carried out on each burner for each fuel gas mixture over a range limited by the onset of turbulence at one extreme (fuel rich) and the weakest mixture with which a flame could be readily stabilised at the other. The latter limit was caused either by flow metering problems or the difficulty of avoiding light back in the determination of the critical flow rates. The formation of polyhedral flames also proved to be a limiting factor in some instances.

Polyhedral flames were sometimes observed in rich flames when the fuel gas had a high Propane content or in lean flames when the fuel gas had a high Hydrogen content. Some Hydrogen-Air flames could not be readily observed with the naked eye and so a shadow image of the flame was projected onto a screen using a high pressure mercury arc source, a concave mirror, and a screen as shown in(Fig 3.2). A more detailed description of the items of optical equipment used is given in section 3.4.3.1. Where polyhedral flames were observed, no critical boundary velocity gradient determinations were carried out as it is known already that such flames give rise to erroneous blow-off data. Normally, a flame of similar fuel concentration was stabilised on a larger burner and this was sometimes found to eliminate the non-isotropic structure. Where this was not the case, however, the formation of the polyhedral structure provided the limit for blow-off determinations.

3.3.4 Details of experiments carried out

Table 9 lists the fuel oxidant systems which were studied over the ranges of fuel concentrations indicated. The values of critical boundary velocity gradient have been plotted against fuel concentration on the graphs indicated in table 9.

The investigations represented in Graphs 10-15 were initiated only after the detailed analysis and interpretation of the data represented in Graphs 1-9 and 16-23.

3.4 Determination of Burning Velocity Data

3.4.1 Choice of method

In order to analyse the critical boundary velocity gradient data by the flame stretch concept it was felt essential to determine burning

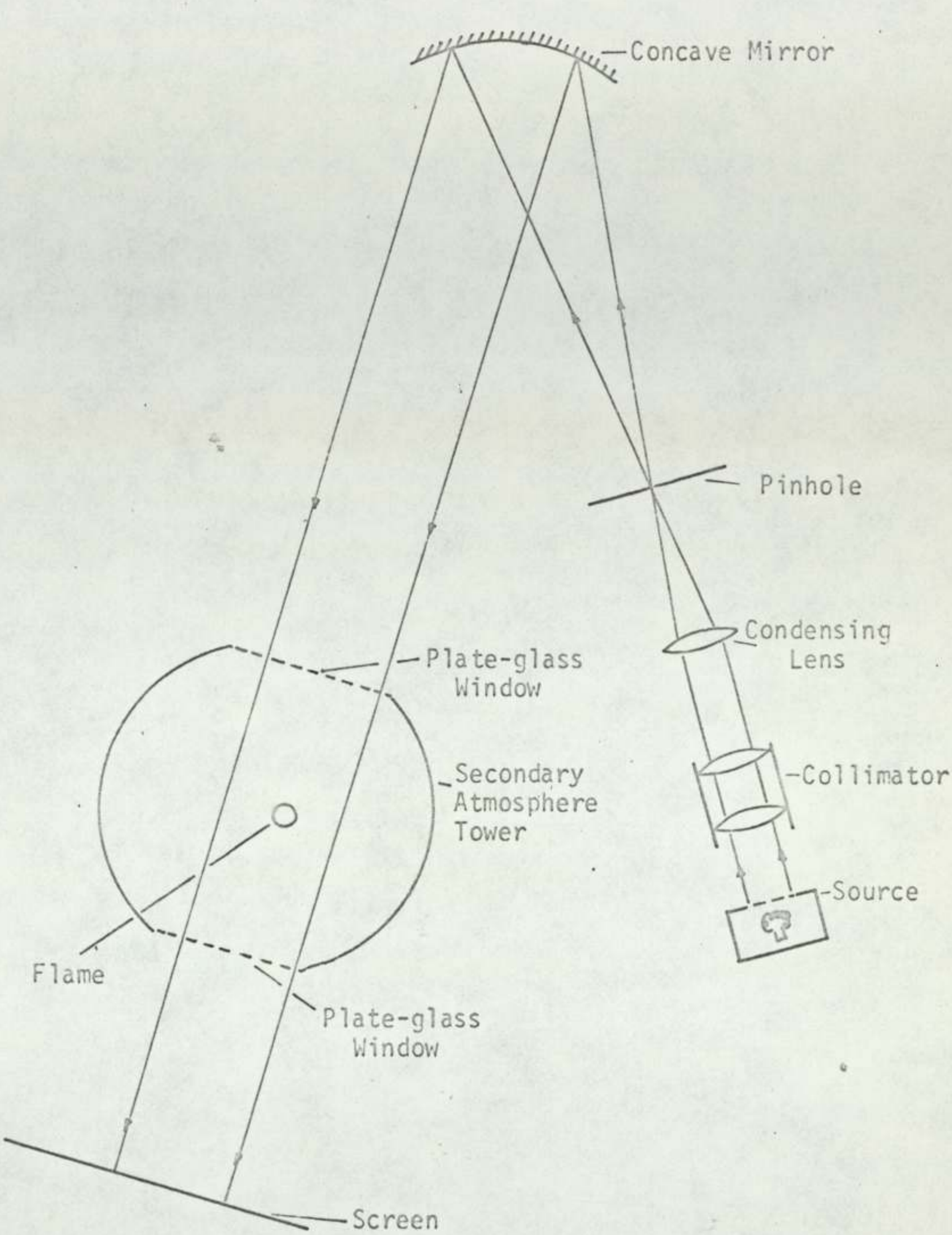


FIG 3.2 OPTICAL SYSTEM USED TO PROJECT IMAGE OF HYDROGEN-AIR FLAMES

Fuel Gas	Range of fuel concentrations studied expressed as a fraction of stoichiometric - X	Oxidant (Primary & Secondary)	Graph No.
Propane	0.82 - 2.09	Air	1
80% Propane 20% Hydrogen	0.85 - 2.33	Air	2
60% Propane 40% Hydrogen	0.72 - 2.37	Air	3
40% Propane 60% Hydrogen	0.65 - 2.40	Air	4
20% Propane 80% Hydrogen	0.57 - 1.46	Air	5
10% Propane 90% Hydrogen	0.53 - 1.15	Air	6
5% Propane 95% Hydrogen	0.45 - 0.51	Air	7
Hydrogen	0.3 - 0.51	Air	8
Ethylene	0.67 - 2.27	Air	9
Propane	0.54 - 2.06	21% Oxygen 79% Argon	10
60% Propane 40% Hydrogen	0.53 - 1.79	21% Oxygen 79% Argon	11
20% Propane 80% Hydrogen	0.43 - 0.78	21% Oxygen 79% Argon	12
Propane	0.52 - 1.12	21% Oxygen 79% Helium	13
60% Propane 40% Hydrogen	0.53 - 1.27	21% Oxygen 79% Helium	14
20% Propane 80% Hydrogen	0.44 - 0.94	21% Oxygen 79% Helium	15

Table 9 - Systems for which critical boundary velocity gradient fuel-concentration relationships were studied

velocity data for all of the systems detailed in section 3.3.4. It has already been shown that burning velocity inconsistencies give rise to the greatest errors in flame stretch factor determination and so in choosing a method for this study, consistency was a more important requirement than absolute accuracy, although clearly the latter was also not undesirable. The second major requirement was for rapidity of data determination as to study all the systems would need a minimum of in excess of one hundred and fifty separate determinations.

It was felt that a burner method best met these requirements. These methods may be subdivided into methods employing flat or button shaped flames, and methods employing conical flames. Doubts have recently been cast on the accuracy of values obtained by flat flame methods. The matter has been discussed in general by Andrews and Bradley (44) and a particular technique which was used for the interpretation of blow-off data by Edmondson and Heap has been discussed by Pritchard (77). It was therefore decided to employ a technique which used a conical flame. Two alternative approaches were available.

(i) The first approach was to establish a flame which approximated reasonably to a perfect cone. By determination of the local gas velocity (V) either by a particle track or pitot tube method, and the cone angle (ϕ) by direct measurement from a photograph, burning velocity may be calculated from equation (3.6).

$$Su = V \sin\phi \qquad (3.6)$$

To establish a perfect cone, most workers have used a "bell" shaped nozzle burner to produce a flat topped velocity profile of the gases emerging from it. Rutherford and Fells (78) have suggested that such a velocity profile may be produced on a burner somewhat more easily and cheaply constructed in which the "bell" shaped section was replaced by a flat plate in which the port is drilled. This claim has not been verified by other researchers, however.

(ii) The second approach relies on the assumption that a constant value of burning velocity exists over the entire flame front. This assumption is not unreasonable for flame fronts with little curvature as is the case over the greater part of a conical flame. Only at the base and tip of the flame would the assumption not be valid and so the average value of burning velocity over the flame front will be effectively the same as the required value. By determination of the flame front area (A) and the volumetric flow rate of the unburned gases (Q), burning velocity may then be determined from equation (3.7)

$$Su = \frac{Q}{A} \qquad (3.7)$$

Three methods are available to locate the flame front in order to determine its area, these being by photographing the visible cone, the shadow cone, or the schlieren cone. The merits of each have been discussed by a number of authors including Garner et al (79) and the schlieren cone shown to be most likely to provide accurate results. Even this method has been shown recently, however, (44) to give too high a value of the flame front area and thus too low a value of burning velocity.

Method (i) was thought to give the most accurate values of burning velocity but was also likely to be time consuming. Method (ii) was therefore selected for use and it was decided to employ a schlieren optical system for flame front location. There are many optical arrangements suitable for schlieren photography and the selection was governed by cost and versatility.

As an indication of the errors arising from the use of these and other methods of burning velocity determination, data published by different authors has been compared for the Propane-Air system by Gray et al (80) and for the Methane-Air system by Andrews and Bradley (81).

3.4.2 Choice of burner

It has already been pointed out that the accuracy of the technique adopted is dependent on the reduction of the influence of flame front curvature to a minimum. With this in mind, two alternative types of burner were considered for use. The first of these was the normal tubular burner described in section 3.3.1 which produces a parabolic laminar velocity profile. The second type considered for use was the "bell" shaped nozzle burner discussed briefly in section 3.4.1 which it was hoped could be made so as to produce a flat topped velocity profile. It was felt that if a burner of the latter type could be successfully constructed, a slight increase in accuracy could be obtained over results obtained using the former type of burner.

With this in mind, a nozzle burner very similar in dimensions to that described by Scholte and Vaags (82) was constructed and preliminary tests were carried out. Experiments were performed both with and without the use of washers to vary the distance between the top of the bell shaped section, which was water cooled, and the burner port. In neither case could a consistently conical flame be produced. It was felt that this might be caused either by surface imperfections in the "bell" shaped section, or the presence of an insufficient number of flow straighteners in the main section of the burner. Two modifications were therefore carried out. The bell shaped section was removed and the inside electroplated and then polished. Additional flow straighteners were also installed. This enabled an almost perfect conical flame to be produced but it was found that the bell shaped section required regular removal and repolishing in order to maintain a satisfactory flame.

It was therefore decided that the marginal increases in accuracy which might arise from the use of this burner did not warrant the additional time required to regularly remove and repolish the bell shaped section. It was thus abandoned in favour of the tubular burners. The majority of the work was carried out using burner No.1, although No.3 was used in some instances where flames of high burning velocity mixtures were being studied. In such instances, attempts to stabilise a laminar flame of the desired composition resulted in light back on burner No.1.

3.4.3 Description of experimental equipment and techniques

3.4.3.1 Schlieren optical system

The system utilised is illustrated in (Fig 3.3) and was mounted on two parallel optical benches each 2 m long and placed on either side of the secondary atmosphere distribution tower.

The light source was a 250 watt high pressure mercury arc the light from which was emitted through a 0.025 m diameter window. A collimator

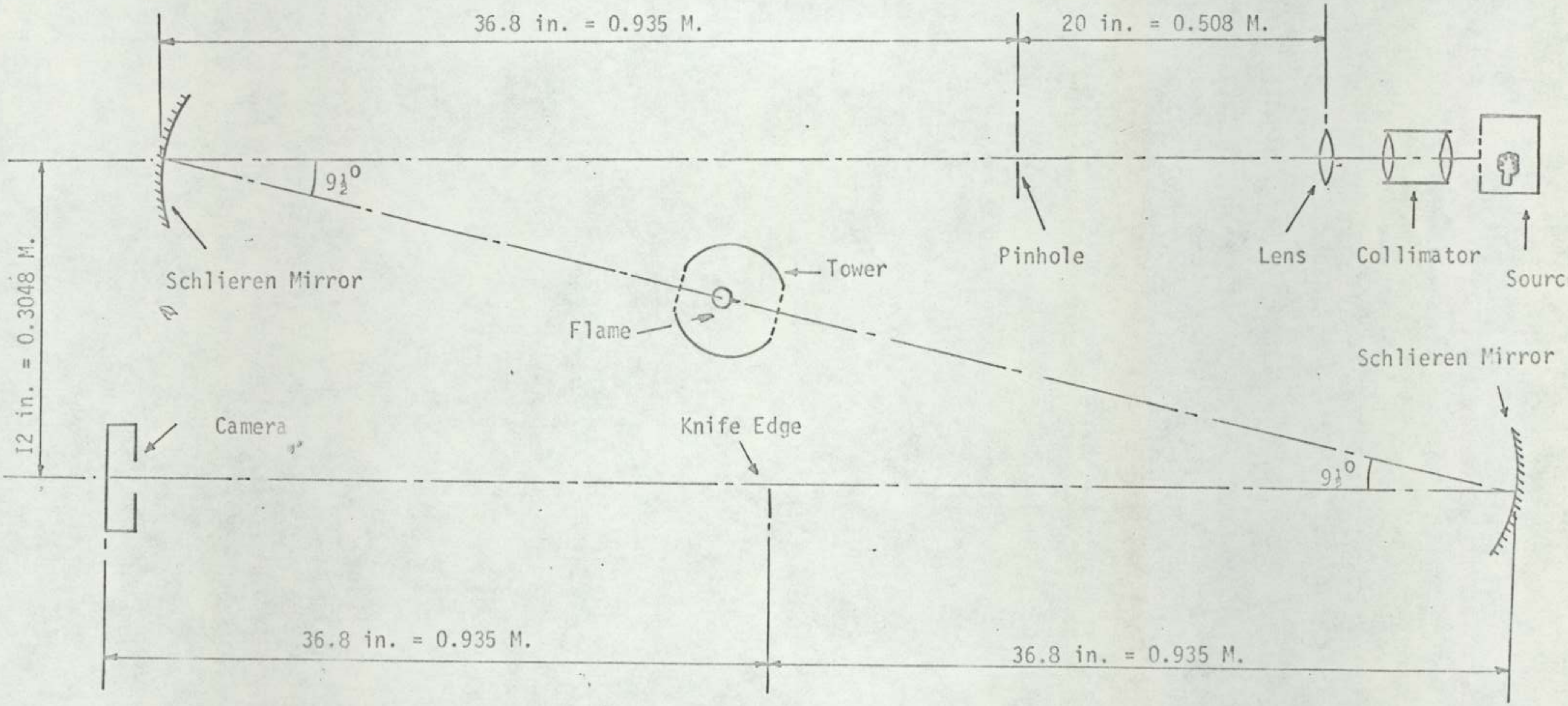


FIG. 3.3 SCHLIEREN OPTICAL SYSTEM

was used to produce an almost parallel beam of light which was focused onto a pinhole with a 0.508 m focal length biconvex lens. The size of the pinhole used governed both the clarity and brightness of the schlieren image produced. An increase in brightness automatically gave rise to a decrease in clarity and the choice of pinhole diameter was of necessity governed by a compromise between these two. These pieces of optical equipment thus combined to provide a high intensity point source of light.

A schlieren concave mirror 0.116 m in diameter and with a focal length of 0.935 m was used to produce a parallel beam of light which passed through the flame to an exactly similar concave mirror which refocused the beam to a point 0.935 m from the second mirror. At this point, a knife edge or graded filter was positioned when required. The schlieren image of the flame was produced 0.935 m beyond the knife edge position and it was at this point that the camera was situated. A single lens reflex camera from which the lens had been removed was used and the image of the flame was projected directly onto the shutter which was in the focal plane. Kodac Plus-X film was used (125 ASA) and exposure times were determined by trial and error but were normally in the range 1/125 - 1/1000 s.

For perfect schlieren pictures, optically perfect windows would have been needed for the secondary atmosphere tower. Because of the very high cost of these, however, high quality pyrex plate glass was used and this produced a slightly mottled pattern superimposed on the entire schlieren photograph. This did not, however, give rise to any loss of accuracy and was merely a minor inconvenience.

The complete optical system, together with the burner, was housed in a framework approximately 2 m x 2 m x 2 m. covered by fire resistant blackout curtains and fitted with a lightproof flue.

3.4.3.2 The use of knife edges

The photograph produced when no knife edges were used consisted of an image of the visible flame cone on which was superimposed the schlieren image. Knife edges have been used by many authors to eliminate the visible image and make interpretation and measurement of the schlieren image easier. Three knife edge configurations have been used and all have been discussed by Pickering and Linnett (83). Of these configurations, the horizontal knife edge is of little practical use. Both single and double vertical knife edges have been used with success and of these the single edge has been by far the most popular arrangement. The schlieren image produced appears to have one light side and one dark side and the image becomes progressively darker as the knife edge is moved to exclude more of the light. A good schlieren image by itself can be obtained if a point source of very high intensity is used. In this case, however, financial limitations had not made the procurement of such a source possible and so it was decided not to use a knife edge but to interpret the superimposed visible and schlieren cone image. Determinations carried out on the Propane-Air system using this approach gave values of burning velocity similar to those of other workers who used the schlieren technique and therefore the method was considered acceptable. Plates 3 and 4 show flames photographed with and without the use of a single vertical knife edge.

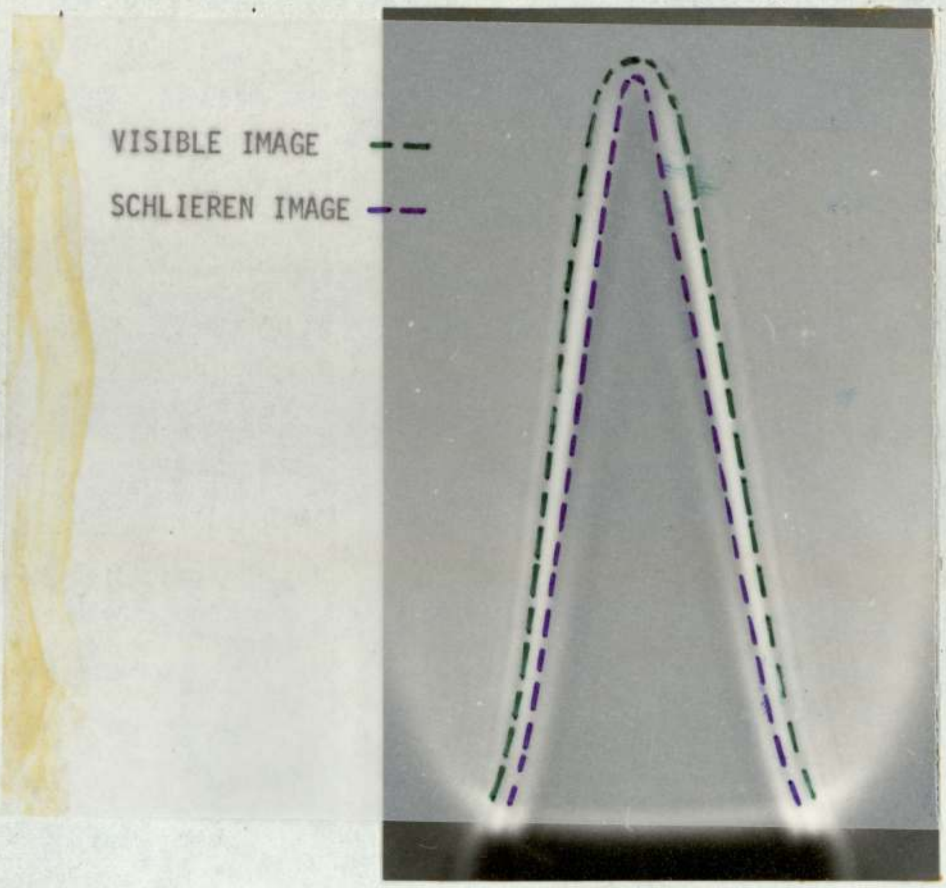


PLATE 3 SUPERIMPOSED VISIBLE AND SCHLIEREN IMAGES OF AN ETHYLENE-AIR FLAME.

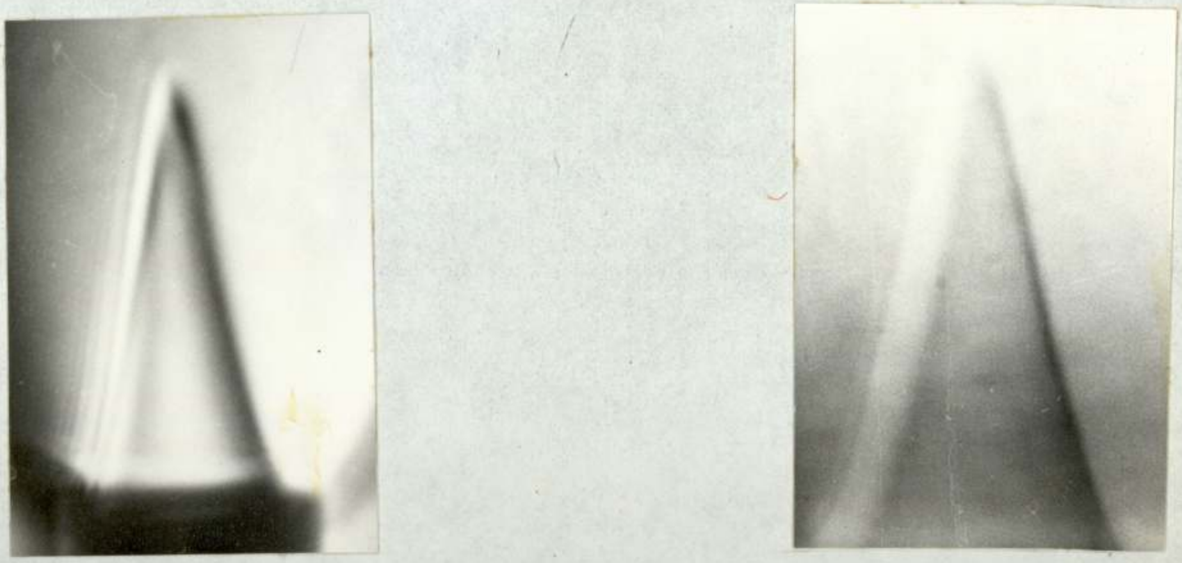


PLATE 4 PROPANE-AIR FLAMES. THE RIGHT HAND SIDE OF THE VISIBLE IMAGE HAS BEEN REMOVED BY THE IMPOSITION OF A KNIFE EDGE. THE POOR DEFINITION IS A CONSEQUENCE OF THE NECESSITY TO INCREASE THE PIN HOLE SIZE IN ORDER TO ENABLE THE SCHLIEREN IMAGE TO BE PHOTOGRAPHED IN THIS WAY. NOTE ALSO THE PRESENCE OF INTERFERENCE PATTERNS ON THE LEFT HAND PHOTOGRAPH.

3.4.3.3 Experimental technique

After the fuel gas flow rates had been set, the Air flow rate was adjusted to give a flame of the desired degree of aeration. Photographs of the image produced were then taken using several different exposure times. The degree of aeration was then altered to a new value and the procedure repeated. Normally three photographs of each of twelve different flames were taken on each film. After developing, enlargements were made of the best photograph of each flame and although the size to which the flame was enlarged was not critical, care was taken to ensure that the degree of magnification was kept constant for all photographs printed from the same film. The burner diameter was then measured from each print and the values averaged to enable the degree of magnification to be determined accurately. A magnification of approximately five times was normally used. The height of the flame was governed by the volumetric flow rate of the fuel oxidant mixture and was set at a value at which the image of the flame and burner occupied the whole frame of the camera (0.036 m high). Normally, at least 0.005 m of the burner was photographed for scaling purposes.

3.4.4 Method of analysis of photographs

The method used was that utilised previously by Senior (84). Firstly the cone was split into a large number of frustra by stepping down the flame front from the apex with dividers. The exact step length (a) was determined by measurement of several such steps and averaging. This procedure results in a small step (b) left at the base of the cone. Radii r_1 to r_n are then constructed as shown on(Fig 3.4.) If distance'a'is small enough it may then be assumed that the cone

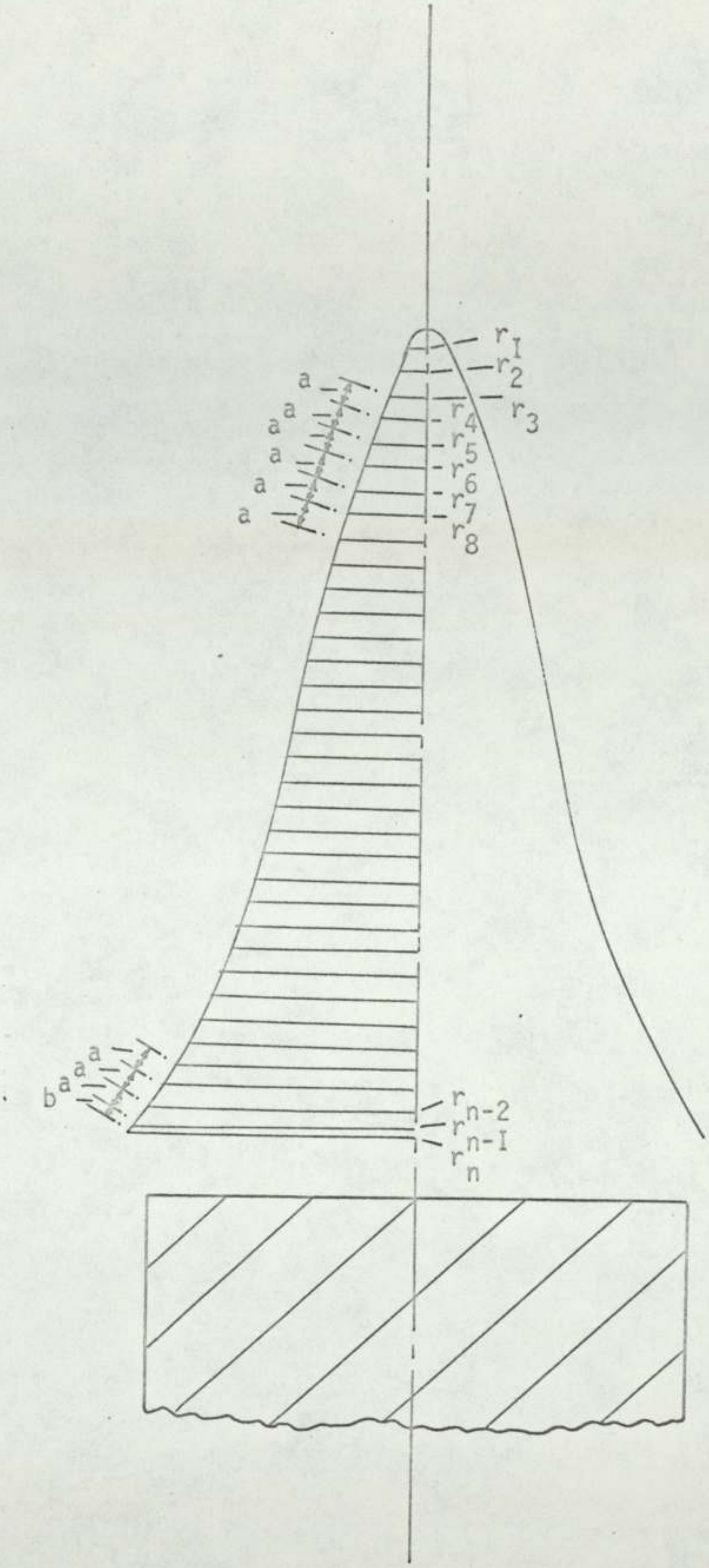


FIG. 3.4 ANALYSIS OF SURFACE AREA OF CONE FROM SCHLIEREN PHOTOGRAPH.

surface between adjacent radii is without curvature. Hence the surface area (A_1) of the frustrum bounded by radii r_1 and r_2 may be approximated as follows :-

$$A_1 = \pi a(r_1 + r_2)$$

similarly

$$A_2 = \pi a(r_2 + r_3) \quad \text{etc.}$$

$$A_{n-2} = \pi a(r_{n-2} + r_{n-1})$$

$$A_{n-1} = \pi b(r_{n-1} + r_n)$$

$$A_o = \pi a\, r_1$$

Now the surface area of the cone (A) is given by

$$A = \sum_{i=o}^{n-1} A_i$$

$$\therefore \quad A = \pi b(r_{n-1} + r_n) + 2\pi a \left[\sum_{i=o}^{n-2} r_i + \frac{r_{n-1}}{2} \right] \qquad (3.8)$$

Most methods require the measurement of the cone angle at a number of points on the flame front. Senior felt that the errors introduced by the assumptions made in his method would be small compared to those arising from such measurements provided 'n' was large. In these experiments, distance 'a' was not more than 0.003 m and normally $n > 15$. Distances 'a' and 'b' were measured with a travelling microscope and radii r_1 to r_n with a graticule scale subdivided to 0.0001 m.

3.4.5 Details of experiments carried out

Table 10 lists the fuel-oxidant systems which were studied over the ranges of fuel concentration indicated and graphs of fuel concentration against burning velocity plotted as detailed in the table.

There are considerable discrepancies in the burning velocity data

Fuel Gas	Approximate range of fuel concentrations studied expressed as a fraction of stoichiometric - X	Oxidant (Primary & Secondary)	Graph No.
Propane	0.84 - 1.29	Air	16
80% Propane 20% Hydrogen	0.91 - 1.29	Air	17
60% Propane 40% Hydrogen	0.75 - 1.35	Air	18
40% Propane 60% Hydrogen	0.65 - 1.35	Air	19
20% Propane 80% Hydrogen	0.65 - 1.45	Air	20
10% Propane 90% Hydrogen	0.64 - 1.55	Air	21
5% Propane 95% Hydrogen	0.70 - 1.40	Air	22
Ethylene	0.70 - 1.52	Air	23
Propane	0.55 - 1.69	21% Oxygen 79% Argon	24
60% Propane 40% Hydrogen	0.54 - 1.61	21% Oxygen 79% Argon	25
20% Propane 80% Hydrogen	0.43 - 2.07	21% Oxygen 79% Argon	26
Propane	0.57 - 1.62	21% Oxygen 79% Helium	27
60% Propane 40% Hydrogen	0.69 - 1.56	21% Oxygen 79% Helium	28
20% Propane 80% Hydrogen	0.60 - 1.60	21% Oxygen 79% Helium	29

Table 10 - Systems for which burning velocity - fuel concentration relationships were studied

cited in the literature by different authors for the Hydrogen-Air system. The determination of such data experimentally would have proved extremely inaccurate using the technique by which the other data had been obtained, however, because of the impossibility of stabilising a laminar flame on all but the smallest of cylindrical burners and then only by using the leanest of flames. Graph No. 22 shows a considerable scatter of points which occurred for this reason when the fuel with 95% Hydrogen was used and it was anticipated that the scatter would be even greater with the Hydrogen-Air system. Dixon-Lewis (68) has recently computed burning velocity data for this system from chemical reaction rate and transport phenomena considerations and close agreement has been obtained with experimental data determined by Edmondson and Heap and Guenther and Janisch. The combined experimental data of these authors was therefore used in this work and has been plotted on a comparable basis to the data determined experimentally in this project in Graph No. 30. The maximum burning velocity is considerably greater than most values cited in the literature but is recognised as the best value currently available.

3.5 The analysis of concentrations of stable species in flames

3.5.1 The choice of method and principles by which it operates

The major considerations in the choice of a suitable method of analysis were:-

(i) The flame front should not be disturbed in such a way as to alter the stability characteristics of the flame.

(ii) The method should be capable of measuring point concentrations of Hydrogen or Ethylene to an acceptable degree of accuracy.

(iii) A high degree of resolution was required because of the extremely small thickness of the preheat and reaction zones of the flame front.

Ideally, the first requirement would have best been met by a method which did not require the removal of a gas sample from the flame. Spectroscopy has been used to study radical concentrations in flames but was considered unsuitable for this type of work as it could be used neither to analyse the molecular species in question nor to provide the resolution required. It was therefore necessary to employ a method requiring the removal of a small sample of gases from the flame front and the analysis of this sample.

Such a system has been described by Fristrom and Westenburg (85) and subsequently used by several other workers and it was from these studies the system used in this work was developed. A quartz microprobe was positioned along a flow line to remove a sample from the flame. Provided the probe orifice was small and the probe was shaped in an aerodynamically favourable way, no disturbances of the flame front were detectable. In order to prevent further reaction of the gases after sampling, the gases were withdrawn at a sufficiently high rate to cause sonic velocity through the probe orifice. The adiabatic expansion and subsequent cooling of the gases was sufficient to quench the flame reactions. This sampling technique has been investigated in detail by members of the Gas Council Combustion Research Group at Watson House and their calculations have shown that in a fairly typical situation, such an adiabatic expansion might be expected to reduce the sample temperature of a flame at 1500^{o}K to as low as 350^{o}K. They have also investigated the influence of probe orifice diameter on resolution, and sample pressure on reproducibility.

Two instruments were considered suitable for sample analysis, these being a gas chromatograph or a mass spectrometer. No instrument of the latter type was readily available. However, a suitable gas chromatograph was available and so was selected for use.

3.5.2 Construction, testing and positioning of microprobes

The probes were constructed from 0.003 m outside diameter quartz tubing. Tubes of both 0.001 m and 0.002 m internal diameter were tested and the former found to be more satisfactory for reasons discussed in section 3.5.6. The first stage of construction was to cut a length of tube about 0.01 m long. One end of this was bent to form a hook and the tube was then clamped vertically with the hook at the bottom. It was then drawn to a fine capillary just above the hook with the use of a glass blower's Town gas-Oxygen burner to which was attached a micro-torch. Small weights, constructed from small nuts and washers hung on wire loops, were hung on the hook to produce an even taper of the tubing. Thus the tube internal diameter was reduced from 0.001 m to 0.0001 m evenly over a distance of 0.015 - 0.02 m. The size of the weight hung on the tube was reduced as the size of the neck decreased. The tube was then cut with a fine quartz fibre at the end of the tapered section and inspected through a microscope to ensure the cut was at right angles to the tube axis. The orifice diameter was then further reduced to a diameter in the range 0.00008 - 0.00003 m by careful use of the micro-torch. During this stage of the construction, the probe orifice was viewed through a travelling microscope pointed along the tube axis. The orifice appeared as a black dot in the centre of the hot quartz and its diameter could thus be determined after each application of the micro-torch by direct measurement. In this way a probe could be constructed such that its orifice diameter was within ± 0.00001 m of the desired value. The most difficult aspect of construction was found to be the cutting of the tube without splintering after it had been tapered. Even after practice, a success rate of less than 50% was achieved in this operation. Any excess quartz present at the tip of the probe was ground away with a fine moist Arkansas Stone to give a more aerodynamically favourable shape which would minimise the chances of flame front disturbance.

The probes were mounted and positioned in a Prior micro-manipulator capable of movement in three perpendicularly opposed planes. The manipulator was capable of measurement by means of a vernier scale to 0.0001 m and had an additional facility which enabled the probe to be held at any angle. Probes were tested by connection to the sampling system which is described in detail in section 3.5.4. The probe was then positioned so that the tip was approximately 0.0001 m on the burned gas side of the primary flame front and 0.0005 m above the burner rim. A poorly constructed probe caused the flame to lift at this point from the burner rim. If no lifting was observed under these conditions, it was not found possible to detect any other form of flame front disturbance. Whether a probe proved satisfactory appeared not to depend on the orifice diameter but rather on the shape of the tip. The orifice diameter did, however, affect both sampling rate and resolution, an increase in one of these automatically causing a decrease in the other. Some compromise was thus necessary in order that the time lag between a sample entering the probe and reaching the gas chromatograph sample loop was not too great.

3.5.3 Sample system construction and testing

A diagram of the system is shown in (Fig 3.5) and a photograph in Plate 5. In order to determine sample compositions, it was necessary to know the pressure and temperature of the sample. Rather than measure the sample temperature, it was decided to use a constant temperature bath through which the sample passed. The tubing from the bath outlet to the sample loop was lagged to minimise heat loss and the constant temperature bath controller was set to control at the temperature of the chromatograph oven. The sample pressure was measured with a transducer rather than a manometer so that the dead volume present in the limb of a manometer could be eliminated. An instantaneous pressure reading was required and so the transducer convertor was coupled directly to a digital voltmeter. The digital voltmeter readings were calibrated against a manometer connected to the tee adjacent to the transducer as shown on the flow diagram. The transducer selected for use was an inductive transducer with a differential pressure range of 0-20 lb/in^2. An Edwards vacuum pump was used to withdraw samples on a continuous basis and the sample pressure was controlled by the use of the valve attached to a tee on the pump inlet.

It has been suggested that in order to obtain sonic flow through the probe orifice, a pressure ratio of at least 2.2 is required across the orifice. Preliminary tests showed that the pressure drop in the pipework and fittings between the probe and transducer was less than 100 mm Hg and so it was estimated that the maximum pressure in the sample loop should not exceed 200 mm Hg. In practice, however, experiments were carried out with a sample loop pressure of approximately 150 mm Hg so that

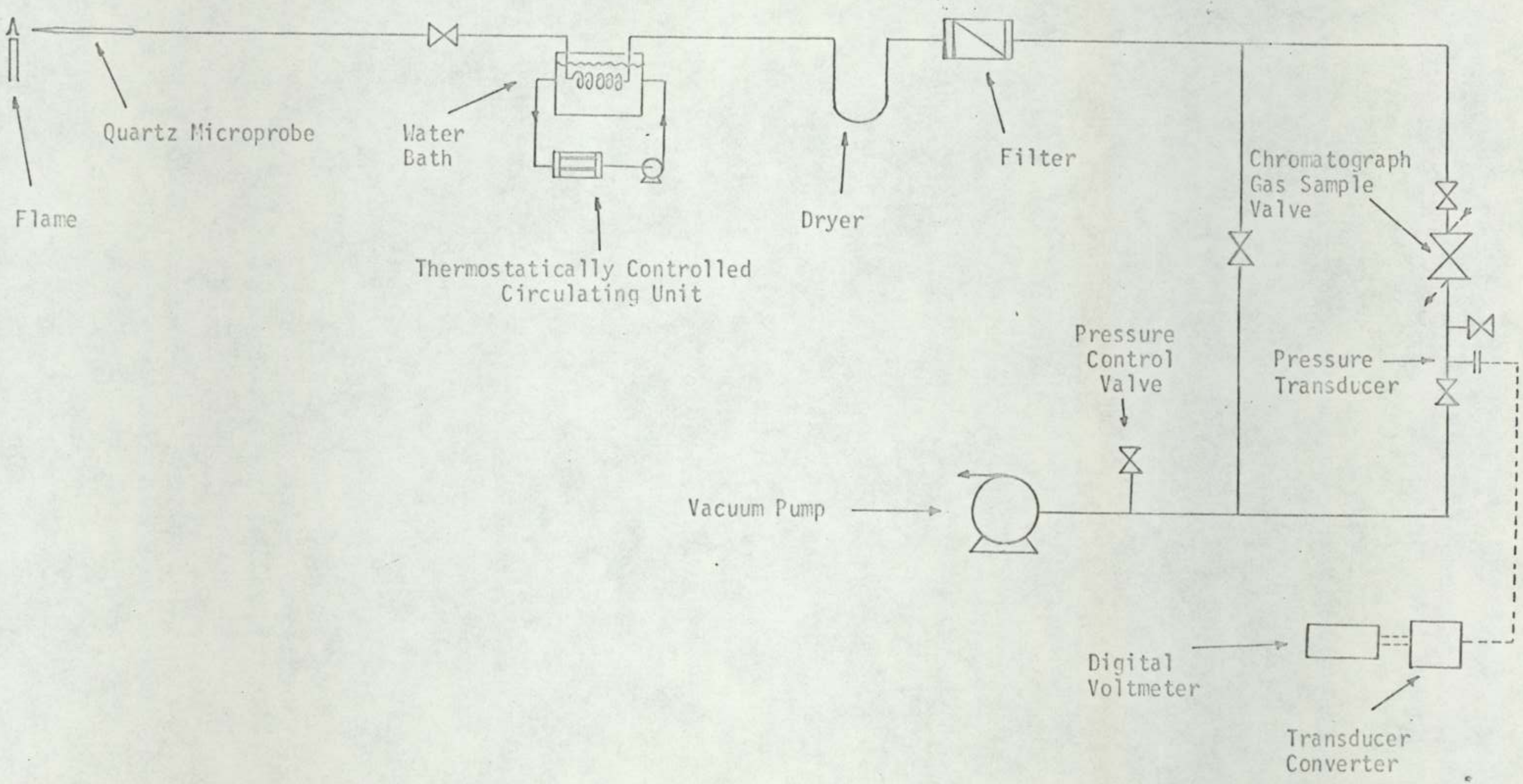


FIG. 3.5 CONTINUOUS SAMPLING SYSTEM.

a margin for safety existed . This figure implies a pressure ratio of 3.04 when the atmospheric pressure is 760 mm Hg.

The chromatograph sample valve had originally been fitted with two sample loops each 0.0005 m^3 capacity,one of which was always in line with the carrier gas supply and column and the other with the sample line. The loop positions could be reversed by simply switching the sample valve control to its alternative position. It was felt that when, on injection, the sample pressure was increased to that of the column, the effective sample volume would be too small for really accurate concentration measurements. The two loops were therefore replaced by one of 0.0015 m^3 capacity and a low capacity bypass. Insufficient space was available for the installation of two loops of the larger size.

It was recognised that moisture would be present in the sample and as this would adversely affect the chromatograph column packing, a drying unit was installed. This consisted of a stainless steel U-tube packed with self indicating Calcium Sulphate (Drierite) and a Nupro stainless steel filter constructed to remove particles greater than 0.000007 m diameter. The filter was present to protect the chromatograph sample valve against damage caused by entrained particles of drying agent. The drying agent was replaced regularly.

All tubing and fittings were constructed from stainless steel except for a short section of flexible P.T.F.E. tubing between the probe and the valve on the temperature controller inlet. All valves used were Hone stainless steel needle valves fitted with replaceable P.T.F.E. seats. Tubing was either 1/16 in or 1/4 in outside diameter depending on location. Swagelok fittings were used but the stainless steel olives

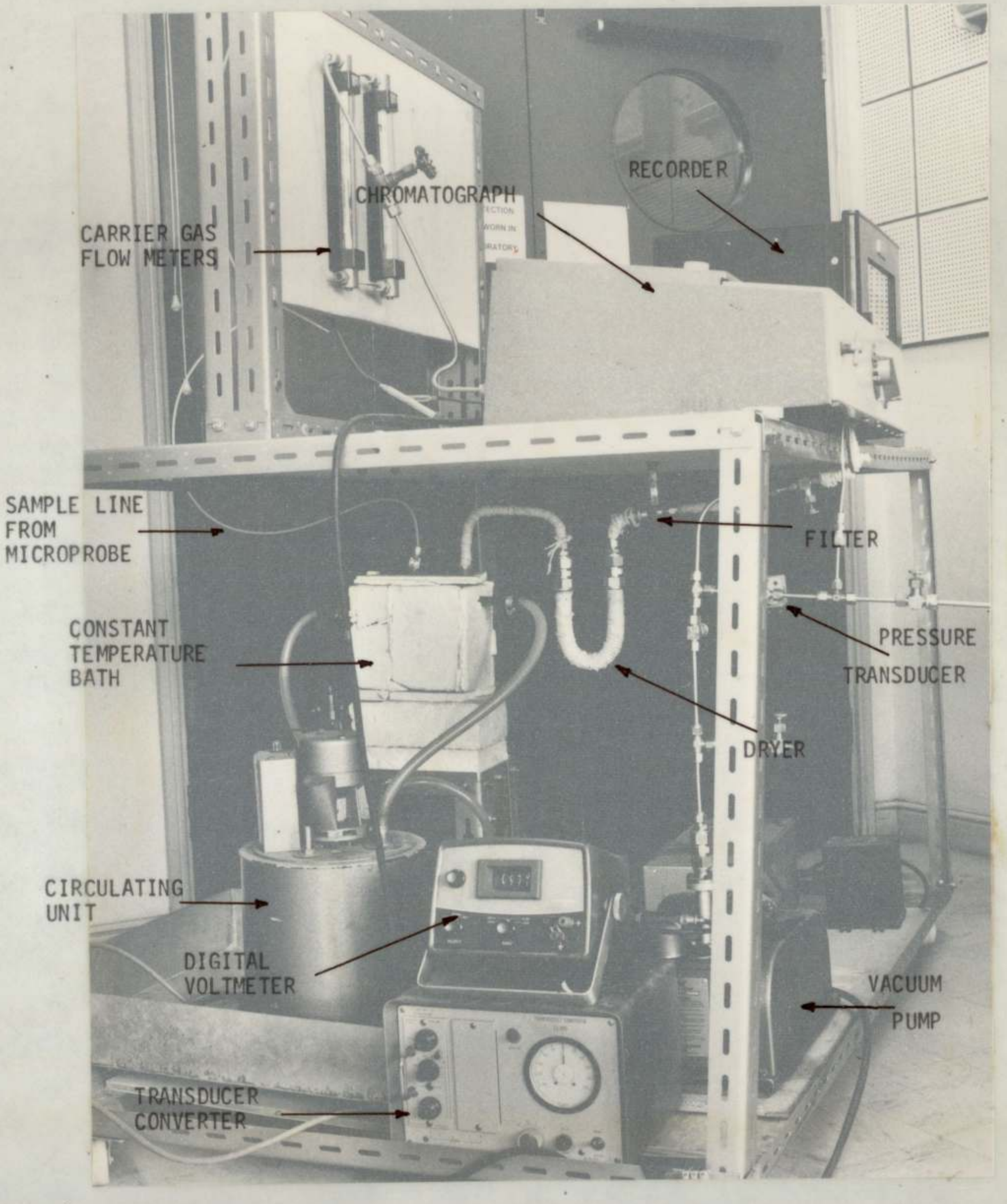


PLATE 5 CONTINUOUS SAMPLING SYSTEM AND ANALYTICAL EQUIPMENT FOR CONCENTRATION PROFILE INVESTIGATIONS.

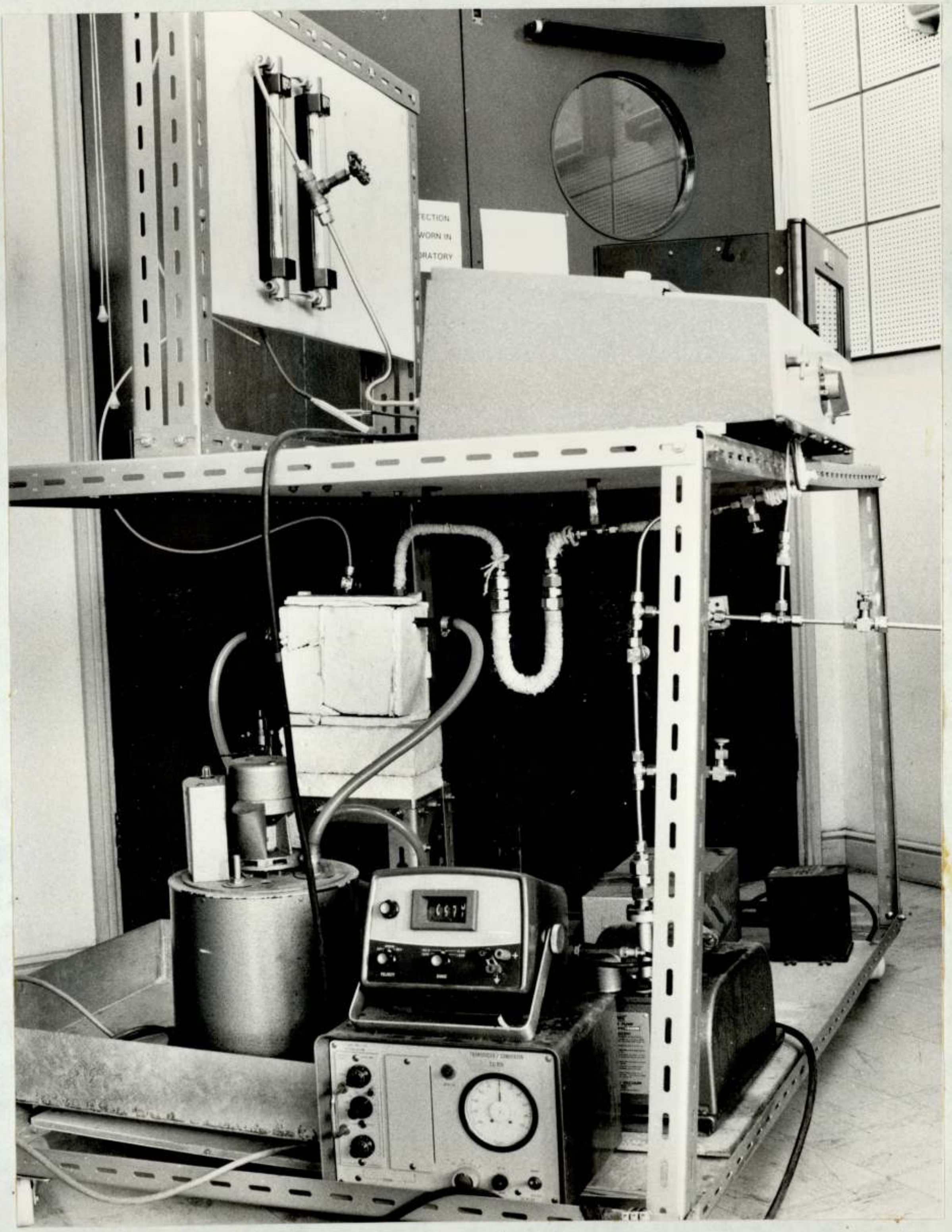


PLATE 5 CONTINUOUS SAMPLING SYSTEM AND ANALYTICAL EQUIPMENT FOR CONCENTRATION PROFILE INVESTIGATIONS.

were replaced with viton O-rings as it was found that the latter were less likely to give rise to leaks.

Leak testing was carried out regularly by closing the valve on the temperature controller inlet, evacuating the sample system, and closing the valve closest to the vacuum pump in the main sample line. The pressure was then monitored at regular time intervals. Two methods were used to locate leaks. The system could be pressurised with compressed air and leaks located with soap and water. Alternatively, the sample system was operated under vacuum in the normal way and inflated plastic bags stuck over each fitting. Deflation of the bag indicated the presence of a leak. The latter approach was also used to test for leaks on parts of the sampling system which could not be tested by the pressure drop method because of the positions of the isolation valves.

3.5.4 Analytical instrument

A Beckman G.C.2 gas chromatograph fitted with a katharometer detector was used. The carrier gas was Argon, the specifications of which appear in section 3.2., and the following instrument settings were employed.

Oven temperature - 40°C
Detector current - 200 ma
Carrier gas pressure - 30.5 lb/in^2 gauge

A Honeywell single speed chart recorder was used with an input range of 0-1.0 mv. and a chart speed of 30 in per hour. Table 11 details the attenuation settings and columns used in the analysis of each gas.

This instrument has been previously used by both Woodcock (86) and Atamar (87) who have given more detailed descriptions of its construction

Gas to be analysed	Column Packing	Attenuation setting	Column Length (ft)
Hydrogen	Porapak Q	50	21
Ethylene	Porapak Q	2	6

Table 11 - Chromatograph Column Data

and operating characteristics. They concluded that it was possible to obtain equal accuracy by the correlation of either peak height or peak area against concentration and as the former approach is the simpler, it was used in this project. In the case of both gases analysed, the complete analysis took approximately ten minutes. A typical Hydrogen-Air sample trace is shown in (Fig 3.6).

3.5.5 Calibration and reproducibility of chromatograph

Because of the variable sample pressure it was necessary to calibrate peak height against effective volume of Hydrogen (V_H) or Ethylene (V_E) where

$$V_H = x_H \frac{\text{sample pressure}}{\text{atmospheric pressure}} \qquad (3.9a)$$

x_H = Hydrogen concentration expressed as a mole fraction

$$V_E = x_E \frac{\text{sample pressure}}{\text{atmospheric pressure}} \qquad (3.9b)$$

x_E = Ethylene concentration expressed as a mole fraction

and

$$\begin{bmatrix}\text{sample}\\ \text{pressure}\end{bmatrix} = \begin{bmatrix}\text{atmospheric}\\ \text{pressure}\end{bmatrix} - \begin{bmatrix}\text{indicated differential}\\ \text{pressure}\end{bmatrix} \qquad (3.10)$$

Equation 3.9a and b imply an isothermal compression of the sample on injection which is reasonable as the column is situated in a thermostatically controlled oven. Graph No. 31 is the calibration graph obtained by sampling gas mixtures of known Hydrogen concentration.

The reproducibility was estimated to be ± 1% of the maximum concentration under a single set of ambient conditions. This value was reduced to ± 5% when large variations in ambient conditions occurred probably due to inefficient lagging of tubes giving rise to variations in

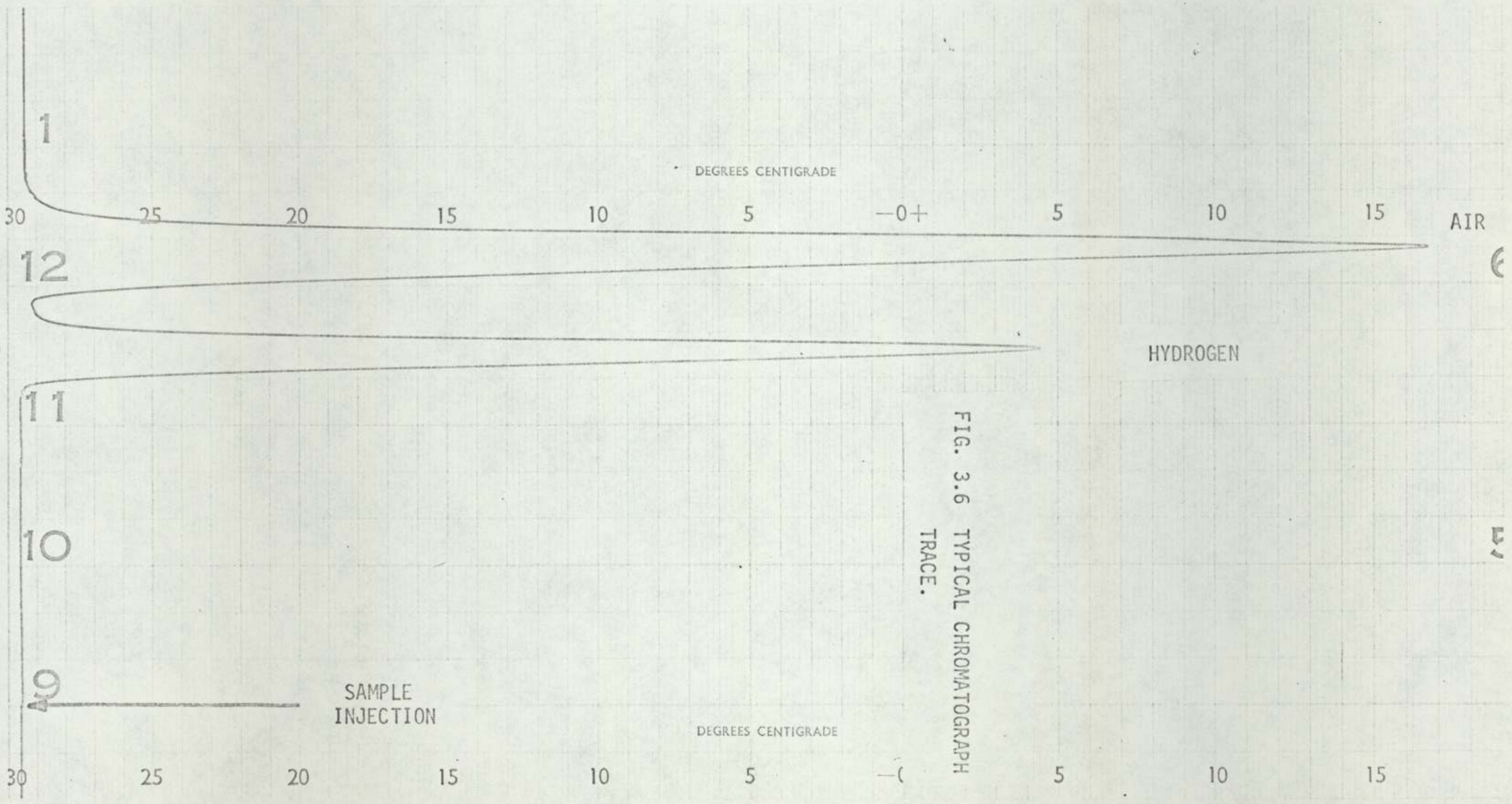


FIG. 3.6 TYPICAL CHROMATOGRAPH TRACE.

sample temperature. All concentrations were expressed as a fraction of the unburned gas concentration, however, and as the relationship between effective sample volume and peak height was linear, the concentrations expressed as a fraction of the unburned gas concentration were estimated to be ± 1% under all conditions. Such variations in ambient temperature of the laboratory were experienced in this part of the work only because a new ventilation and heating system was being installed in the building at the time.

3.5.6 Experimental procedure

A laminar flame of the desired composition was stabilised on burner No.1, a description of which appears in section 3.3.1. The probe was set at an angle considered to be similar to that of the flow lines in the region of the flame to be studied. Although no experiments were carried out to indicate the direction of the flow lines, their paths could be estimated with a considerable degree of confidence from particle track photographs of aerated flames published by both workers.

Distances were measured relative to a point on the outside of the burner port at which the tangent to the burner port intersected the probe axis at right angles. The first stage in an experiment was therefore to note the micro-manipulator readings in all three directions when the probe tip was touching this point. The probe was then moved vertically to the desired distance above the burner and horizontally to the point at which the secondary combustion zone appeared to merge with the surrounding atmosphere.. One of the plate glass windows in the secondary atmosphere tower had been replaced with polythene sheeting through which the probe passed.

The continuous sampling system was allowed to withdraw samples from a single probe position for at least ten minutes before injection into the chromatograph was carried out. This was more than twice the length of time that preliminary tests had shown to be necessary but as a single analysis took ten minutes, the probe could be moved to its new position immediately after injection of the previous sample and the new sample withdrawn by the time the instrument was available for the next injection. Because only one 0.0015 m^3 sample loop was being used, it was necessary to switch the sample valve back to its original position five minutes after the injection. This had the effect of injecting a very small sample into the column from the bypass loop but the recorder traces that this produced did not appear until well after the traces from the original sample injection.

From its original position on the edge of the secondary combustion zone, the probe was moved horizontally in small steps towards the axis of the burner tube. Each step was normally 0.0001 m in length although in regions of the flame where concentration varied only slightly with horizontal distance, the step lengths were sometimes increased to several times this value. The digital voltmeter reading was noted and a sample injected after each step and the probe position was noted when the tip of the probe made contact with the primary flame front. The concentration profile was considered to be complete when a constant concentration of the species being monitored equivalent to that present in the unburned gases was recorded. After each concentration profile had been completed, the probe was removed and inspected for damage under a microscope. If any damage was found, the probe was replaced and all experiments carried out since the previous inspection were disregarded and repeated.

It was found that in flames richer than stoichiometric in fuel a point was reached as the probe was moved vertically at which sufficient of the probe was passing through the secondary combustion zone for the sample to be reheated enough to re-ignite the gases in the probe. This was clearly visible to the naked eye and occurred only after the probe had penetrated the primary reaction zone. It was also clear from the concentration profiles recorded under such conditions. In such cases, the profile was entirely different in shape to those determined under normal conditions. This provided a second check as to whether re-ignition was taking place and marked the maximum height above the burner at which profiles could be determined reliably. This phenomena was found to take place far more readily when the probes were constructed from thin walled quartz tubing than when the thicker tube was used. It was for this reason that probes made from the latter tubing were preferred.

Little trouble was experienced with probe orifices closing because of the high temperatures in the flame reaction zone. When this did occur, however, it was clearly evident by a rapid decrease in pressure in the sample line and a corresponding reduction in recorder peak height.

3.5.7 Details of experiments carried out

Concentration profiles were studied as detailed in table 12. In all cases, concentrations were expressed as ratios where

$$R_H = \frac{\text{local Hydrogen concentration}}{\text{concentration of Hydrogen in unburned gases}}$$

$$R_E = \frac{\text{local Ethylene concentration}}{\text{concentration of Ethylene in unburned gases}}$$

Local and unburned gas concentrations were expressed as mole fractions.

Fuel Gas	Oxidant (Primary & Secondary)	Species Analysed	Fuel concentration Fraction of stoichiometric - X	Height above burner (m)	Graph No.
60% Hydrogen 40% Propane	Air	Hydrogen	0.8	0.0006	32
"	"	"	"	0.0009	33
"	"	"	"	0.0015	34
"	"	"	"	0.0021	35
"	"	"	1.0	0.0006	36
"	"	"	"	0.0009	37
"	"	"	"	0.0012	38
"	"	"	"	0.0015	39
"	"	"	1.2	0.0006	40
"	"	"	"	0.0009	41
"	"	"	"	0.0012	42
"	"	"	"	0.0015	43
Ethylene	Air	Ethylene	1.0	0.0006	44
"	"	"	"	0.0009	45
"	"	"	"	0.0012	46
"	"	"	"	0.0015	47

Table 12 - Concentration Profile Studies

3.6 Analysis of temperature profiles in the stabilising region of laminar aerated flames

It was decided to attempt to measure the corresponding temperature profiles to the concentration profiles already determined. The method used proved to be unsatisfactory and the work is recorded briefly for the benefit of future workers.

3.6.1 Experimental method and findings.

The technique adopted was to use a fine wire thermocouple constructed from 80% Pt/20% Rh and 60% Pt/40% Rh wires 0.00025 m in diameter. A detailed description of the construction of such thermocouples has been given by Fristrom and Westenburg (85) and their use has been commented on by Datta et al (15). The use of the method depends on two important criteria:-

(i) The thermocouple must be aligned with an isotherm so as to eliminate heat conduction along the wires.

(ii) The couple must not disturb the flame front.

It was felt that provided the wires lay along a tangent to the flame front, the method might prove successful in this study. This was not the case, however, for three reasons. Firstly, the flame front was observed to lift locally from the burner when the couple was in the stabilising region. Secondly, the Pt/Rh wire chosen were found to melt in the fuel rich flames studied. Thirdly, conduction effects were clearly not negligable. While the two latter problems would not have arisen in Datta's study it is not clear why he did not encounter the problem of flame front disturbance.

Section 4.

ANALYSIS AND DISCUSSION OF RESULTS.

4. ANALYSIS AND DISCUSSION OF RESULTS.

4.1. Comparison of experimental results for Hydrogen-Propane-Air system with those of other workers

To facilitate easy comparison of critical boundary velocity gradient and burning velocity data with that of other workers, Graphs 48 and 49 have been compiled from the data presented in Graphs 1-8 and 16-22.

4.1.1 Critical Boundary Velocity Gradient data

Reiter and Wright (88) have made a study of the Hydrogen-Propane-Air system and their data has been plotted on a similar basis to this investigation in Graph No. 50. By comparison of Graphs 48 and 50 it will be seen that there is considerable agreement between the critical boundary velocity gradients of mixtures common to both studies. It should be pointed out that Reiter and Wright used Propane of only 95% purity and although it is not clear what constituted the other 5% it is possible that this may account for any discrepancies which do exist. Their smallest burner was 0.00573 m internal diameter and so they were unable to study mixtures with as wide a variation in degree of aeration as in this investigation. Nevertheless this work provides a valuable corroboration of the accuracy of the data presented in Graph 48.

Grumer et al (89) have carried out a few critical boundary velocity gradient determinations for this system. Their investigation was not a comprehensive one and none of their mixtures were directly comparable to those studied in this work. Their results were used by Van Krevelen and Chermin (90) in the development of a method for prediction of critical boundary velocity gradients for multi-component gaseous fuels. The curves presented in Graph 51 have been computed by application of this method. The agreement with Graph 48 is generally poor. The discrepancy is greatest between experimental and predicted critical boundary velocity gradients with fuels of high Hydrogen content and so the method might be of use for prediction of the blow-off characteristics of low Hydrogen content fuels provided a high degree of accuracy was not essential. In general it would seem unwise to place a great deal of confidence in blow-off data predicted by this method.

4.1.2 Burning Velocity Data

Leason (91) has carried out measurements of burning velocity for Hydrogen-Propane-Air mixtures with 0, 5%, 10%, 20% and 30% Hydrogen in the fuel gas mixture. He determined his data by application of equation (3.7) using the visible image of the separated primary flame front in order to evaluate the flame front area. The value of maximum burning velocity reported by Leason for the Propane-Air system is approximately 20% below currently accepted literature values. Furthermore, the maximum burning velocity occurs with an exactly stoichiometric flame, whereas most workers have found that this occurs with flames slightly richer than stoichiometric (in this study at X = 1.065). It is not surprising, therefore, that Leason's curve for 20% Hydrogen present in the fuel differs considerably from that in Graph 17. The ratio of the maximum burning velocities of Propane with Air and a 20% Hydrogen, 80% Propane fuel gas mixture with Air in the two studies is similar, however, and this indicates that the discrepancies are probably due to the difference in experimental methods only.

It has been found possible to correlate maximum burning velocity (Su max) against mole fraction of fuel in the fuel oxidant mixture (x_F)

by the relationship

$$Su_{max.} = 0.3195\, e^{0.0619\, x_F} \qquad (4.1)$$

Maximum burning velocity is plotted against percentage Hydrogen in the mixture in Graph 52 and against total fuel concentration expressed as a mole fraction in Graph 53. Also on Graph 53 are plotted the values of maximum burning velocity determined for the Hydrogen-Methane-Air system by Scholte and Vaags (82) and it will be seen that a similar type of equation to (4.1) may be used to correlate their data also. It is possible that this might provide a method of predicting the maximum burning velocity for other Hydrogen-Hydrocarbon mixtures although clearly this would require experimental verification using a variety of Hydrocarbon fuels.

4.2 Flame Stretch analysis of Hydrogen-Propane-Air data

Physical properties of the mixtures studied were calculated by the methods indicated and discussed in Appendix 3. Flame stretch factors were then calculated using equation (1.9) and the experimental data from Graphs 48 and 49. The burning velocity-fuel concentration graphs for fuel gas mixtures 0, 20% and 40% Hydrogen were extrapolated slightly to enable larger concentration ranges to be analysed. A typical set of calculations is shown in table 13 and the results of all calculations are plotted on Graph 54. Insufficient data was available to produce any flame stretch analysis for the fuel gas mixture containing 95% Hydrogen because of flow metering limitations.

Reed's correlation (equation 1.10) indicates that critical flame stretch factor is independent of fuel concentration for flames leaner

Fuel Concentration X	Critical Boundary Velocity Gradient g_b (s^{-1})	Thermal Conductivity k (w/m^2deg.C.)	Density ρ (kg/m^3)	Burning Velocity Su (m/s)	Specific Heat Cp (J/kg deg.C.)	Critical Flame Stretch Factor K
0.75	1280	0.02773	1.195	0.320	1057	0.274
0.80	1820	0.02783	1.194	0.357	1061	0.313
0.85	2390	0.02794	1.194	0.395	1064	0.337
0.90	3090	0.02804	1.194	0.432	1068	0.364
0.95	3800	0.02814	1.194	0.464	1071	0.388
1.00	4620	0.02824	1.194	0.483	1074	0.436
1.05	5470	0.02834	1.194	0.491	1078	0.500
1.10	6400	0.02844	1.193	0.486	1081	0.598
1.15	7320	0.02854	1.193	0.476	1084	0.713
1.20	8330	0.02863	1.193	0.457	1087	0.881
1.25	9360	0.02872	1.193	0.423	1091	1.154
1.30	10,350	0.02881	1.193	0.382	1094	1.566
1.35	11,300	0.02890	1.193	0.331	1097	2.776

Table 13 - Flame stretch analysis of blow-off and burning velocity data for fuel gas mixture containing 40% Hydrogen and 60% Propane

than stoichiometric. Edmondson and Heap (28) showed that in practice this was never the case but that the dependance was least significant in a region X = 0.85 to X = 1.0. If the fuel concentration was further reduced, however, the dependance on fuel concentration appeared to increase again in a number of cases. This trend was present in their study of the Propane-Air system and has been reproduced in this investigation of the same system. The corresponding values of critical flame stretch factor differ slightly in the two investigations but the reason for this discrepancy has already been indicated in section 1.2.5. Neither correlation differs substantially from that predicted by Reed's theory, however.

In this context, the work of Van Heyningen is of some significance (97). He has attempted to determine the true nature of this relationship by the evaluation of parameters in equation (1.3) by solution of the Navier-Stokes equation without the assumptions made in the derivation of equation (1.9). The work is open to considerable criticism, however, as the author appears to have failed to comprehend the assumptions made in the derivation of equation (1.3) and hence the upper fuel concentration limit of applicability of this equation. His theoretical values of flame stretch factor for rich flames are of very limited value therefore.

The progressive introduction of Hydrogen into the system causes a rapid increase in critical flame stretch factor so that in a stoichiometric flame with a fuel gas mixture of 80% Hydrogen and 20% Propane, the critical flame stretch factor is nearly five times that of the corresponding Propane-Air flame and eleven times that value predicted by equation (1.10). It is postulated that such increases are attributable to preferential diffusion of Hydrogen into the stabilising region of the flame by the mechanism which is discussed in section 2. Substantially larger increases in Hydrogen concentration would be required in the high Hydrogen content flames to cause these stability anomalies than in the low Hydrogen content flames. It may be concluded from Graph 54 that the increase in Hydrogen in the stabilising region is dependent on the Hydrogen concentration in the unburned gas mixture. This is indicative that a molecular diffusion process may be responsible.

It is perhaps at this stage worth indicating how a thermal diffusion process might account for the changes in critical flame stretch factor caused by the introduction of Hydrogen. Thermal diffusion refers in this instance to the diffusion of a molecular species because of the presence of a thermal driving force, i.e. a temperature gradient.

Thermal diffusivity is substantially independant of the concentrations of the molecular species present in the mixture. The introduction of Hydrogen will give rise to a reduction in preheat zone thickness which is proportional to burning velocity. The maximum Hydrogen flux caused by thermal diffusion will therefore vary with burning velocity. Large temperature gradients exist only over a very short distance in the preheat zone of the flame however and it is doubtful whether thermal diffusion could account for the transport of Hydrogen over the distances necessary to influence stability to the degree necessary to give rise to the observed increases in critical flame stretch factor. This argument is supported by the work of Dixon-Lewis and his colleagues.

Dixon-Lewis et al (94, 95) have computed fluxes of molecular Hydrogen due to both molecular and thermal diffusion in Hydrogen-Oxygen-Nitrogen flames. The method employed was based on a technique described in an earlier paper (96). They have shown that fluxes due to thermal diffusion can at times reach a similar order of magnitude to those due to molecular diffusion and by substantially reducing the Oxygen to Nitrogen concentration ratio in the unburned mixture to below that of Air they have recorded instances where the thermal diffusion fluxes exceed the molecular diffusion fluxes. In such situations, however, the magnitude of both fluxes is small. They have shown that fluxes of atomic Hydrogen are of considerable significance in flame structure and flame reaction kinetic considerations and have always found that molecular diffusion fluxes of this species are always very much greater than the corresponding thermal diffusion fluxes. This has also been shown to be the case with Oxygen, Nitrogen and water vapour. With these three species the two fluxes oppose each other whereas for molecular Hydrogen the fluxes complement each other. Clearly, whether fluxes of atomic Hydrogen oppose or complement each other is dependent on the position in the flame front under consideration.

4.2.1 Correlation of flame stretch factor and preferential diffusion factor

It has been suggested that the preferential diffusion factor as defined in equation (2.6) may be considered as a measure of the degree to which preferential diffusion will occur in the stabilising region of a laminar aerated burner flame. It has been evaluated for Hydrogen-Propane-Air mixtures as detailed in table 14 and the following assumptions have been made:-

(i) D_1 = diffusivity of water vapour in Air.

% Hydrogen in Fuel Gas	Fuel Concentration X	Mole Fraction Hydrogen x_2	Mole Fraction Water x_1	Preheat Zone Thickness η° X 10^4 (m)	Burning Velocity Su (m/s)	Preferential Diffusion Factor F	Critical Flame Stretch Factor K
0	0.8	0	0.1259	0.6757	0.292	0	0.2314
	0.9	0	0.1405	0.5456	0.358	0	0.2896
	1.0	0	0.1550	0.4770	0.406	0	0.3360
	1.1	0	0.1545	0.4608	0.416	0	0.3954
	1.2	0	0.1538	0.5025	0.378	0	0.5849
	1.3	0	0.1533	0.6444	0.292	0	1.1520
20	0.8	0.00787	0.1329	0.7115	0.299	0.002545	0.287
	0.9	0.00881	0.1455	0.5710	0.372	0.003125	0.338
	1.0	0.00975	0.1603	0.4946	0.428	0.003841	0.3802
	1.1	0.0107	0.1595	0.4750	0.445	0.004206	0.4793
	1.2	0.0116	0.1587	0.5094	0.414	0.004558	0.7099
	1.3	0.0125	0.1581	0.6166	0.341	0.004923	1.2711
40	0.8	0.0199	0.1368	0.6148	0.357	0.006223	0.3134
	0.9	0.0223	0.1526	0.5090	0.432	0.007751	0.3641
	1.0	0.0246	0.1681	0.4559	0.483	0.009392	0.4361
	1.1	0.0269	0.1673	0.4538	0.486	0.01089	0.5975
	1.2	0.0292	0.1663	0.4831	0.457	0.01097	0.8806
	1.3	0.0314	0.1651	0.5779	0.382	0.01171	1.5657
60	0.7	0.0361	0.1313	0.5816	0.396	0.009840	0.2188
	0.8	0.0409	0.1487	0.5099	0.455	0.01244	0.3328
	0.9	0.0456	0.1659	0.4479	0.522	0.01524	0.4178
	1.0	0.0502	0.1826	0.4043	0.582	0.01823	0.5140
	1.1	0.0548	0.1810	0.4013	0.590	0.01948	0.7244
	1.2	0.0593	0.1796	0.4463	0.534	0.02065	1.1951
	1.3	0.0637	0.1780	0.5520	0.434	0.02176	2.4040

Table 14 - Cont'd. on next page.

% Hydrogen in Fuel Gas	Fuel Concentration X	Mole Fraction Hydrogen x_2	Mole Fraction Water x_1	Preheat Zone Thickness η_o X 10^4 (m)	Burning Velocity Su (m/s)	Preferential Diffusion Factor F	Critical Flame Stretch Factor K
80	0.6	0.0661	0.1343	0.6219	0.402	0.01564	0.2491
	0.7	0.0760	0.1550	0.5515	0.463	0.01990	0.4740
	0.8	0.0857	0.1751	0.4890	0.532	0.02442	0.7262
	0.9	0.0952	0.1950	0.4342	0.610	0.02914	1.0642
	1.0	0.1044	0.2164	0.3914	0.689	0.03421	1.3064
	1.1	0.1133	0.2114	0.3590	0.763	0.03515	1.7642
	1.2	0.1221	0.2087	0.3547	0.785	0.03620	2.5532
	1.3	0.1306	0.2060	0.3872	0.730	0.03708	4.2433
	1.4	0.1389	0.2034	0.4764	0.602	0.03783	8.7046
90	0.7	0.1206	0.1828	0.4692	0.608	0.02985	1.0959
	0.8	0.1353	0.2062	0.3734	0.788	0.03548	1.2701
	0.9	0.1494	0.2291	0.3184	0.951	0.04111	1.5065
	1.0	0.1630	0.2513	0.2858	1.088	0.04665	1.8964
	1.1	0.1761	0.2467	0.2732	1.168	0.04699	2.6430
	1.2	0.1887	0.2420	0.2765	1.182	0.04708	4.1636
100	0.35	0.1282	0.1370	0.4104	0.642	0.02786	0.256
	0.4	0.1439	0.1551	0.3062	0.880	0.03385	0.696
	0.5	0.1736	0.1901	0.2209	1.261	0.04684	1.752

Table 14 - Preferential Diffusion Factor analysis of Hydrogen-Propane-Air system

(ii) D_2 = diffusivity of Hydrogen in Air

(iii) x_1 = water vapour concentration in combustion products

(iv) x_2 = Hydrogen concentration in unburned gases.

(v) physical properties are those of the unburned gas mixture

(vi) D_1 and D_2 have been evaluated at the unburned gas temperature as it was anticipated that this region would provide the greatest resistance to mass transfer.

(vii) x_1 has been evaluated assuming complete combustion of that proportion of the fuel gas mixture for which there was sufficient Oxygen present in the primary mixture.

(viii) A constant Hydrogen to Propane concentration ratio has been assumed at all points in the primary combustion zone.

All concentrations have been expressed as mole fractions. This means of evaluation of x_1 was shown by Datta (14, 32) to approximate closely to the maximum concentration difference across the primary and secondary combustion zones in the stabilising region of both fuel lean and fuel rich Methane-Air flames. Thus the apparent inconsistency in the evaluation of x_1 and x_2 provides a means of arriving at a reasonable value of the concentration driving force which exists for each molecular species.

Three additional values of preferential diffusion factor were calculated, from data obtained by the extrapolation of Graph 54 as detailed in table 15.

Graphs 55-61 have been plotted and it has been found possible to characterise the relationships between critical flame stretch factor and

Fuel Concentration X	% Hydrogen in Fuel-Gas	Critical Flame Stretch Factor K	Preferential Diffusion Factor F
0.7	0	0.16	0
0.7	20	0.18	0.00192
0.7	40	0.20	0.00488

Table 15 - Additional data obtained by extrapolation of experimentally determined critical flame stretch factor-fuel concentration graphs

preferential diffusion factor by equations of the following type for constant fuel concentration (X).

$$K = K^1 e^{CF} , \quad 0.7 < X < 1.3 \qquad (4.2)$$

K = flame stretch factor for situation in which preferential diffusion occurs.

K^1 = flame stretch factor determined experimentally for situation in which no significant preferential diffusion occurs.

C = parameter dependant on fuel concentration only.

The various values of K^1 and C on Graphs 55-61 were computed by a "least-squares" method on the University Honeywell 316 digital computer. 95% confidence limits are also shown on the graph. The evaluation of confidence limits from the variance of log K is justified as the probable errors in flame stretch factor could be expressed as constant percentage of the value of flame stretch factor. Standard deviations and correlation coefficients are listed in table 16. The author, however, is of the opinion that the correlation coefficients are of little statistical meaning as the validity of the correlations is unlikely to be disputed.

The parameter C has been plotted against fuel concentration expressed as a fraction of stoichiometric in Graph 62 and the following relationship derived empirically:-

$$C = 38.6 + (1 - a)\left[\frac{2.64}{X - 0.615} - 6.85 \right] \qquad (4.3)$$

$$0.7 < X < 1.3$$

The nature of this relationship reflects the different mechanisms by which preferential diffusion influences flame stability in fuel weak and fuel rich flames.

Fuel Concentration X	Standard Deviation σ	Correlation Coefficient
0.7	0.1119	0.984
0.8	0.0989	0.986
0.9	0.0983	0.984
1.0	0.1024	0.988
1.1	0.0787	0.988
1.2	0.0544	0.997
1.3	0.0394	0.997

Table 16 - Statistical analysis of data used to obtain relationships of the type indicated in equation (4.2)

In fuel rich flames, preferential diffusion will increase secondary combustion which in turn will increase the heat transfer to the primary flame front. It would be anticipated therefore that this would override any influence that primary combustion would have in this instance. It is unlikely that the influence on critical flame stretch factor of a change in concentration, reflected by a particular value of preferential diffusion factor will depend on fuel concentration itself. This gives rise to a value of C for $X > 1.0$ independent of fuel concentration.

In fuel lean flames, the influence of preferential diffusion will act on the primary flame front reactions altering burning velocity and hence critical flame stretch factor. Secondary combustion effects will be negligible in comparison. The influence on the primary combustion reactions of a concentration change reflected by a particular value of preferential diffusion factor will be complex and will be a function of the fuel concentration itself. This is reflected by the variation of C with fuel concentration for $X < 1.0$.

By combining equations (4.2) and (4.3) it follows that

$$K = K^1 \exp \Big[\Big(38.6 + (1 - a)\Big(\frac{2.64}{X - 0.615} - 6.85\Big)\Big)F\Big]_{0.7 < X < 1.3} \qquad (4.4)$$

If equation (1.10) is used to characterise K^1 then it also follows that

$$K = 0.23 \Big[1 + (X^{6.4} - 1)\Big] \exp \Big[\Big(38.6 + (1 - a)\Big(\frac{2.64}{X - 0.615} - 6.85\Big)\Big)F\Big]_{0.7 < X < 1.3} \qquad (4.5)$$

Equation (4.5) may be expressed in a more convenient form for evaluation of critical boundary velocity gradients as follows:-

$$gb = \frac{0.23 \rho C_p Su^2}{k} \Big[1 + (X^{6.4} - 1)\Big] \exp \Big[\Big(38.6 + (1 - a)\Big(\frac{2.64}{X - 0.615} - 6.85\Big)\Big)F\Big]_{0.7 < X < 1.3} \qquad (4.6)$$

It is proposed that this equation should be valid for the prediction of the influence of Hydrogen on any laminar aerated burner flame provided that the fuel concentration lies within the stated limits. It has not been tested for mixtures in which $F > 0.047$ and so this provides a further limitation on its applicability although there is no theoretical reason for the imposition of such a limit. This limit would be exceeded only when very high Hydrogen content fuels were being used and in such cases the burners used would be so small that the situation is very unlikely to exist in practical situations.

Equation (4.4) was used to calculate the values of critical flame stretch factor corresponding to those determined experimentally and listed in table 14. An average discrepancy of ± 7% of the experimental value was found to exist between calculated and experimental values.

4.3 Prediction of the blow-off characteristics of the Hydrogen-Methane-Air system

As a test of the applicability of equation (4.6) to systems other than the Hydrogen-Propane-Air system from which the equation was developed, an analysis has been carried out on data for the Hydrogen-Methane-Air system. This system was selected as both critical boundary velocity gradient and burning velocity data were available in the literature. Scholte and Vaags (82) determined the burning velocity data by a "cone angle" method and their work has already been discussed in earlier sections. Their method has provided accurate data for other systems and there is no reason to doubt the validity of this particular set of data. Grumer et al (89) have investigated critical boundary velocity gradient-fuel concentration relationships for this system. It has already been pointed out that their data for the Hydrogen-Propane-Air system was used in the derivation of the Van Krevelen and Chermin method for the prediction of such relationships for multicomponent fuels. This method has proved inaccurate for the latter system and this may reflect on the accuracy of the data from which the method was derived. If the accuracy of the Hydrogen-Propane-Air data is poor, then it is quite possible that the Hydrogen-Methane-Air data is equally inaccurate.

From table 17 it may be observed that there are two particularly curious results. The first concerns the experimental critical flame stretch factor for a 0.7 stoichiometric flame with a fuel gas mixture containing 43.9% Hydrogen. The value is far smaller than is to be expected. In the system with 79.9% Hydrogen in the fuel gas, the critical flame stretch factor-fuel concentration relationship is unlike any studied in this or any other investigation. The critical flame stretch factor for a 0.7 stoichiometric flame is very much greater than for a 0.8 stoichiometric flame. There is no reasonable explanation for this and it must be concluded that both of the anomalous results discussed are accountable to experimental error.

If these two results are included, an average discrepancy of ± 37.8% of the experimental value exists between predicted and experimental critical flame stretch factors with a maximum discrepancy of 192%. If the anomalous results are ignored, however, these two figures are reduced to ± 25.6% and 54.6% respectively. The largest discrepancies occur with the fuel mixture containing 79.9% Hydrogen and it was with high Hydrogen content fuels that the data of Grumer et al was found to be least

Percentage Hydrogen in fuel	Fuel Concentration X	Critical flame stretch factor		Discrepancy $\frac{\lvert K - K^* \rvert}{K} \times 100\%$
		Predicted K^*	Experimental K	
43.9	0.7	0.38	0.13	192.0
	0.8	0.58	0.39	48.8
	0.9	0.73	0.67	7.46
	1.0	0.96	0.84	14.3
	1.2	2.12	1.74	21.8
63.9	0.7	0.77	0.72	6.95
	0.8	1.01	0.80	26.2
	0.9	1.04	1.11	6.30
	1.0	1.67	1.76	5.11
	1.2	3.68	4.55	19.1
79.9	0.7	1.43	2.51	43.0
	0.8	1.68	1.61	43.5
	0.9	1.91	2.95	34.5
	1.0	2.64	4.62	43.8
	1.2	5.30	11.68	54.6

Table 17 - Analysis of data for Hydrogen-Methane-Air system obtained from the literature

accurate for the Hydrogen-Propane-Air mixtures. It is possible therefore that a Hydrogen flow metering error exists in a number of the experimental results of these workers.

In the circumstances, the agreement between experimental and predicted critical flame stretch factors is good. Reed has suggested that flame stretch factors can be evaluated only to an accuracy of ± 60% and in all but one case the agreement is well within these limits. Graph 64 presents these data graphically.

4.4 The replacement of Nitrogen in the primary and secondary Air by Argon and Helium

4.4.1 Critical boundary velocity gradient and burning velocity data

The replacement of the inert constituent of the Air, i.e. Nitrogen, by Argon or Helium causes change in density, specific heat, and thermal conductivity of the unburned gas mixture and the diffusivities of the various molecular species present. Chemical reactions in the primary flame front will be influence only to the extent that those concerned with the production and reaction of oxides of Nitrogen will be eliminated. The changes in physical, chemical and transport properties will ensure, however, that burning velocity, critical flame stretch factor and preferential diffusion factor may all differ from the corresponding values associated with normal aerated systems.

As far as the author is aware, Helium and Argon "Airs" have not been used in blow-off studies prior to this investigation. They have, however, been used by Mellish and Linnett (92) and Clingman et al (93) in attempts to clarify the mechanisms governing burning velocity and by a number of authors in studies of the upper and lower fuel concentration limits for upward and downward propagation of flame fronts with a variety of fuels. The latter studies have been presented in a table by Mellish and Linnett (92) together with data obtained using a number of other "Airs".

Clingman et al (93) have calculated adiabatic flame temperatures for a number of Methane-"Air" flames with Nitrogen, Helium, and Argon as the inert constituent. The values using the "Helium" and "Argon" Airs are identical but in some cases 350°K greater than that for a Nitrogen-Air flame of the same fuel concentration. They concluded from their experimental results that no simple burning velocity theory; e.g. that of Tanford and Pease, can account for the observed ratios of the burning velocities of mixtures of a similar fuel concentration but different inert gases.

The values of burning velocity using the various inerts would be anticipated to be in increasing order of magnitude :- Nitrogen, Argon, Helium. The experimentally determined ratios of these values for Methane-Air flames would not be expected to be the same as those for the Hydrogen-Propane-Air flames. This has been found to be the case in practice.

4.4.2 Flame stretch analysis

The data presented in Graphs 10-15 and 24-29 have been analysed on a flame stretch basis and the results are presented in Graphs 65 and 66. The general shape of the curves of critical flame stretch factor plotted against fuel concentration are similar to those of normal aerated systems.

When no Hydrogen is present, the values of critical flame stretch factor are significantly lower when either Helium or Argon is the inert than when Nitrogen is used. The flame stretch theory predicts that if no preferential diffusion is likely to occur, the values of critical flame stretch factor should correspond to those calculated using equation (1.10) irrespective of the nature of the oxidant. Equation (1.10) was derived, however, from an analysis of aerated systems in which the inert constituent of the Air was always Nitrogen. It must be concluded, therefore, that equation (1.10) is applicable to normal aerated systems only and that Reed's anomalous results for the Propane-Oxygen and Methane-Oxygen systems may not necessarily be caused by preferential diffusion as he has suggested.

In searching for a reason why Reed's correlation is not applicable when Argon or Helium is the inert, it is worth noting that the adiabatic flame temperature is substantially higher when either of these inerts is present than when the inert is Nitrogen. This may provide an explanation for the variations in critical flame stretch factor.

The boundary velocity gradient theory assumed that heat conduction to the burner was of prime importance in blow-off considerations. The flame stretch theory has shown that aerodynamic quenching is the more important mechanism but makes the further assumption that the local burning velocity at the point of stability is equivalent to the normal burning velocity. The amount of heat conduction to the burner and the curvature of the flame front will influence how good this assumption is. Datta showed that it was not unreasonable for the two dimensional Methane-

Air flames that he studied but that nevertheless the local burning velocity in this region of the flame was lower than the normal value. The flame temperature, and thus the temperature driving force for heat conduction will to some degree influence just how much lower than the normal value the local burning velocity is. As a result of this, the assumption will give rise to lower values of critical flame stretch factor for flames of similar degrees of aeration than would be the case if the local burning velocity were used. Thus the hottest flame will give rise to the lowest critical flame stretch factor if the normal burning velocity is used in calculations.

As the adiabatic flame temperatures are identical when either Helium or Argon is the inert, similar critical flame stretch factor-fuel concentration relationships would be expected if no preferential diffusion occurs, although differences in dead space height might influence the heat conduction to the burner. Graphs 65 and 66 show that there is considerable similarity over part of the concentration range studied and that the two correlations differ only for fuel concentrations at the lean end of the range. It has already been pointed out that when Nitrogen is the inert, the adiabatic flame temperature is lower and as anticipated, the critical flame stretch factor-fuel concentration relationship is unlike those which have been found to exist when Helium or Argon is the inert.

On the introduction of Hydrogen, substantial increases of critical flame stretch factor were noted when Argon was the inert (Graph 65) but little or no change was noted when Helium was used (Graphs 67-68). In order to test whether preferential diffusion could account for these observations, preferential diffusion factors have been calculated and by application of equation (4.4), critical flame stretch factors have been predicted as detailed in table 18. In order to evaluate K^1 in this equation, the experimentally determined values of critical flame stretch factor for the Propane-"Air" system with the appropriate inert have been used. The discrepancy between predicted and calculated critical flame stretch factors have been calculated as detailed in table 18 and all lie within Reed's ± 60% limit which is discussed in section 4.3 and all but four values within the limits discussed in Appendix 2. It is suggested therefore that this work with Helium and Argon Airs provides additional support for both the preferential diffusion concept and also the method of analysis proposed in section 2 and applied to aerated flames containing Hydrogen in section 4.2. The use of these "Airs" has enabled physical and transport properties to be varied over a far wider range of values than would normally be encountered in aerated flames.

It is clear from the preferential diffusion factors calculated for systems using Helium "Air" that preferential diffusion of Hydrogen is relatively unimportant in comparison with systems using Argon or Nitrogen "Air". It would be anticipated therefore that other phenomena associated with preferential diffusion would show comparable behaviour. The data presented by Mellish and Linnett (92) of concentration limits for Hydrogen-"Air" and Deuterium-"Air" flames propagating in tubes using Helium, Argon and Nitrogen as inerts support this suggestion. The differences between limits for upward and downward flame front propagation for the two fuel gases were substantially smaller when Helium "Air" was

Inert Gas	Percentage Hydrogen in fuel gas	Fuel Concentration X	Preferential Diffusion Factor F	Critical flame stretch factor		Discrepancy $\frac{\lvert K^* - K \rvert}{K} \times 100\%$
				Experimental K	Predicted K*	
Argon	40	0.7	0.005183	0.206	0.231	12.1
		0.8	0.006602	0.241	0.250	3.73
		0.9	0.008215	0.269	0.267	0.745
		1.0	0.009970	0.304	0.288	5.26
		1.1	0.01082	0.331	0.349	5.14
		1.2	0.01165	0.414	0.453	9.42
		1.3	0.01242	0.618	0.597	3.40
	80	0.7	0.01872	0.643	0.546	15.1
		0.8	0.02270	0.712	0.531	25.4
		0.9	0.02290	0.751	0.494	34.2
		1.0	0.02752	0.778	0.566	27.2
		1.1	0.03022	0.838	0.774	7.65
		1.2	0.03331	1.021	1.112	8.90
		1.3	0.03544	1.373	1.374	0.073
Helium	40	0.7	0.0002499	0.140	0.135	3.57
		0.8	0.0003311	0.147	0.128	12.9
		0.9	0.0004307	0.138	0.122	11.6
		1.0	0.0005427	0.131	0.132	0.76
		1.1	0.0006121	0.140	0.158	12.85
		1.2	0.0006848	0.164	0.203	23.8
		1.3	0.0007570	0.236	0.319	35.1
	80	0.7	0.001245	0.162	0.144	11.1
		0.8	0.001623	0.154	0.136	11.7
		0.9	0.002057	0.137	0.131	4.38
		1.0	0.002556	0.127	0.143	12.6
		1.1	0.002771	0.139	0.172	23.7
		1.2	0.003006	0.157	0.224	42.6
		1.3	0.003236	0.252	0.349	38.5

Table 18 - Analysis of blow-off data determined using Helium and Argon "Airs"

Inert Gas	Percentage Hydrogen in fuel gas	Average Discrepancy (± %)	Maximum Discrepancy (± %)
Argon	40	5.68	12.1
	80	16.93	34.2
Helium	40	14.37	35.1
	80	20.65	42.6

Table 19 - Statistical Analysis of data presented in table 18

used than when either of the other "Airs" were employed.

4.5 Flame stretch analysis of Ethylene-Air data

The data presented in Graphs 9 and 23 has been analysed on a flame stretch basis and replotted in Graph 69. Also on this graph are plotted four other correlations for the same system.

These are:-

(i) The experimental data of Edmondson and Heap.

(ii) Reed's general correlation - equation (1.10).

(iii) The experimental correlation for the situation in which no significant preferential diffusion will occur (Propane-Air data obtained in this investigation).

(iv) The relationship obtained by calculating preferential diffusion factors of Ethylene and applying these to equation (4.5).

Curve (iv) provides the closest approximation to the experimental curve for Ethylene-Air. In its derivation, however, it has been assumed that equation (4.5) may be used to characterise the influence of preferential diffusion of fuels other than Hydrogen. This is an assumption clearly open to criticism and would require verification by further investigation. The influence that preferential diffusion of Ethylene will have is comparatively insignificant, however, and may for most practical applications be ignored. For this reason, the applicability of equation (4.5) to other hydrocarbon fuels in aerated systems is largely of academic rather than pratical interest and for this reason has not been investigated further.

4.6 Composition studies in stabilising region of flames

4.6.1 Analysis of concentration measurements and likely sources of error

In order to present the concentration profiles of Hydrogen and Ethylene in a more readily assessable form, the following procedure was adopted. Photographs were taken of the flames used for each study. Enlargements were produced and from these the location of the primary flame front in the stabilising region could be plotted in relation to the burner as shown on Graphs 70-73. This enabled lines of constant concentration to be plotted from Graphs 32-47 and the position of these could be related to both the burner and the primary flame front.

Three major possible sources of error were thought to exist in these measurements.

(i) In any method which requires a finite sample to be extracted from the flame, there is an inherent lack of resolution. In this work, a sample composition has been assumed to be representative of the point at which the probe tip was located. In practice, however, it is likely that it will be representative of a point displaced a small distance from the tip along the flow line on the unburned gas side of the probe.

(ii) In regions of high concentration of the species being monitored, the rate of change of concentration with distance is very low and thus the location of the lines $R_H = 1.0$ and $R_E = 1.0$ are likely to be far less accurate than the other lines of constant concentration.

(iii) The micro-manipulator vernier scale was sub-divided into divisions for measurement to 0.0001 m only and thus each concentration measurement has a possible location error of ± 0.00005 m. These errors will to some extent be eliminated, however, by the drawing of smooth curves through the point concentration measurements in the establishing of the overall concentration profile. This type of error may also be minimised by measuring as many point concentrations for each profile as is practically feasible.

4.6.2 Interpretation of Hydrogen concentration isopleth diagrams

Graph 73 shows that in a stoichiometric Ethylene-Air flame, the lines of constant Ethylene concentration lie effectively parallel both to each other and the primary flame front. This is typical of the type of concentration profile that would be expected using any one component fuel-Air mixture if the fuel concentration were monitored. As the fuel concentration is increased above that of the stoichiometric flame, fuel will be detected progressively further from the primary flame front on the burned gas side.

In the Hydrogen-Propane-Air flame, the situation is complicated by the production of Hydrogen from the thermal cracking of Propane. This gives rise to an initially less rapid rate of decrease of Hydrogen concentration with distance as the primary flame front is approached from the unburned gas side than might be expected by comparison with Graph 73. Once the production of Hydrogen by cracking ceases to be significant, however, the Hydrogen concentration decreases very rapidly indeed in a 0.0001 m thick region on the unburned gas side of the primary flame front to leave unburned only Hydrogen for which there is insufficient

Oxygen present for combustion to take place. The rate of Hydrogen consumption is apparently greater than the corresponding Ethylene consumption and this is a reflection on the difference in mechanism of the combustion chemical reactions for the two gases and the associated temperatures involved.

Preferential diffusion of Hydrogen might be expected to show itself in two possible ways as follows:-

(i) In the stabilising region of fuel rich and stoichiometric flames, the concentration of Hydrogen in the secondary reaction zone would be greater than at any other point further up the flame but the same distance from the primary flame front.

(ii) In fuel rich, stoichiometric and fuel lean flames, the Hydrogen concentration in the stabilising region primary reaction zone would appear to be greater than at a similar distance from the primary flame front in another part of the flame. This effect would appear as a kink in the lines of constant concentration so that the line was closer to the primary flame front in this region than at any other point along the flame front. Unfortunately, the "flame stretch" effect and the quenching effect of the burner may both influence the concentration profiles in this part of the flame and the interpretation of the lines of constant concentration within the primary reaction zone is extremely difficult.

In Graphs 71 and 72, increases in fuel concentration in the secondary combustion zone of the type described in paragraph (i) are evident.

In attempting to identify profiles of the type described in paragraph (ii) it is advisable to disregard the lines for $R_H = 1.0$ for the reasons discussed in section 4.6.1. If this is done there is some evidence of an apparent decrease in reaction zone thickness in the stabilising region of all three Hydrogen-Propane-Air flames. This effect may be seen more clearly in the 1.0 and 1.2 stoichiometric than the 0.8 stoichiometric flame. By itself this decrease in primary reaction zone thickness would not be conclusive, but when considered with the observed increases in fuel concentration in the secondary reaction zone, provides considerable support for the preferential diffusion hypothesis.

Section 5.

CONCLUSIONS AND SUGGESTIONS FOR FUTURE WORK.

5. CONCLUSIONS AND SUGGESTIONS FOR FUTURE WORK.

5.1 Conclusions

1. The flame stretch correlation as proposed by Reed is capable of predicting the critical boundary velocity gradient at blow-off of laminar premixed burner flames if all of the following pre-requisites are met:-

   (i) The fuel gas contains no Hydrogen

   (ii) The primary oxidant is Air

   (iii) The secondary atmosphere is either Air or Nitrogen.

2. The preferential diffusion of Hydrogen towards the point of stability at the base of the flame can account for the anomalous critical flame stretch factor-fuel concentration relationships which are found if Hydrogen is present in the fuel gas.

3. Hydrogen concentration profiles determined in the stabilising region of laminar Hydrogen-Propane-Air flames are consistent with the preferential diffusion hypothesis.

4. The preferential diffusion factor as derived in section 2 may be considered as a measure of the degree to which preferential diffusion will occur in the stabilising region of a laminar burner flame.

5. Preferential diffusion factor has been correlated against critical flame stretch factor for the Hydrogen-Propane-Air system and from this, equation (4.4) has been derived.

6. It is proposed that equation (4.4) may be used to predict the influence of preferential diffusion of Hydrogen on the blow-off of any laminar flame. This equation relates the critical flame stretch factor to that which might be expected to exist if no significant preferential

diffusion occurred. There is therefore no reason to apply the three limitations detailed in conclusion 1, to the applicability of equation (4.4). This latter point has been confirmed using Helium/Oxygen and Argon/Oxygen mixtures as the primary oxidant and secondary atmosphere but only when the same gas mixture was employed in both of these functions.

7. Preferential diffusion of the most mobile species in the primary fuel-oxidant mixture will always occur to some degree but this is unlikely to influence stability to a significant degree unless the fuel gas is Hydrogen.

5.2. Suggestions for future work

The suggestions for future work may be sub-divided into two categories:-

(i) Work aimed at clarifying matters which have arisen during this investigation.

(ii) Work which will enable the mechanisms governing the blow-off of burner flames to be more fully understood and more accurately characterised.

5.2.1 Future work related to this investigation

Under the above heading, investigations into the following would facilitate a better understanding of the limitations of the correlations and proposals presented in this dissertation:-

(i) The applicability of the preferential diffusion factor concept to the blow-off of aerated flames in inert atmospheres.

(ii) The applicability of the preferential diffusion factor concept to the blow-off of inverted flames.

(iii) Clarification of the reasons why the flame stretch correlation proposed by Reed is not applicable when the inert constituent of the Air is replaced by one with different physical and transport properties.

5.2.2 Long term research objectives

From the discussion of experimental results and the subsequent conclusions it is apparent that Reed's flame stretch theory of blow-off contains assumptions which, when applied to laminar burner stabilised, premixed flames have proved in appropriate in some situations. It is the opinion of the author that further attempts to modify the theory would be of little value. In its present form, the original flame stretch correlation (equation 1.10) combined with the preferential diffusion factor modification provides a useful method for the prediction of blow-off characteristics in most situations encountered practically. Only rarely will a situation arise in practice for which this approach is inapplicable.

Long term efforts must therefore be directed towards a more sophisticated approach to the problem which would eliminate the necessity for the simplifying assumptions of the present theory. The first stage in this approach has already been undertaken by Dixon-Lewis and his colleagues. The propagation of Hydrogen-Oxygen-Nitrogen flames has already been characterised and this work has been discussed in sections 1 and 4. Such an approach must be extended to more aerodynamically complex situations and more kinetically complex systems.

It is worth while noting, however, that the application of sophisticated computation techniques, which are required for investigations of this type, is expensive. It is likely, therefore, that even when such methods can predict flame stability characteristics with considerable accuracy, there may still be a requirement for less rigorous methods which are simple and therefore inexpensive to apply.

The investigations discussed in this dissertation have been confined to premixed flames leaner in fuel thnn $X = 1.36$, the limit placed by Reed on the applicability of equation (1.10). In practice, many burners are operated with fuel concentrations greater than this value. It is to be hoped therefore that future theories will be applicable to a wider range of fuel concentrations and possibly even to diffusion flames.

Section 6.

REFERENCES.

6. REFERENCES

1. Gilbert, M.G., Prigg, J.A., U.K. Gas Council Research Communication G.C. 35 (1956).
2. Adam, A., Electrotechnick, 12th Sept. 1946.
3. Damkoehler, G., Z. Electrochem. und Angew.Chem. 46 (11), 80 (1940).
4. Heiligenstaedt, W., Gas (Essen) 14, 73 (1942).
5. Delbourg, P., Ass.Tech. de l'Industrie du Gas en France Congress (1946).
6. Garside, R.M., Forsythe, J.F., Townsend, D.T.A., J.Inst.F., 18, 175 (1945).
7. Lewis, B., Von Elbe, G., "Combustion, Flames and Explosions in Gases" 2nd Edition, Academic Press (1961).
8. Lewis, B., Von Elbe, G., J.Chem.Phys. 11, 75 (1943).
9. Von Elbe, G., Mentser, M.J., J.Chem.Phys., 13 89 (1945).
10. Sparrow, E.M., Lin, S.H., Lundgren, T.S., Physics of Fluids, 7, 338 (1964).
11. Bizley, D., South Eastern Gas Board Central Laboratories Report CL/255/70 (1970).
12. Reed, S.B., J.I.Gas.E. 8, 157 (1968).
13. Taylor, B.G., "A study of ignition temperatures", Thesis, University of Leeds (1958).
14. Reed, S.B., J.Inst.F. 36, 105 (1963).
15. Datta, P., Haywood, B.M., Reed, S.B., Comb. & Flame, 17, 399 (1971).
16. Karlovitz, B., Denniston, D.W., Knapschaefer, D.H., Wells, F.E., 4th. Int. Symp. on Combustion, 613 The Combustion Institute (1953).
17. Karlovitz, B., Chem.Engng.Prog. 61 (8), 56 (1965).
18. Lewis, B., Termotecnica (Milan) 3 , 201 (1963).
19. Lewis, B., Von Elbe, G., Z.Phys.Chem. (Frankfurt) 37, 287 (1963).
20. Lewis, B., Address to 7th Int.Symp. on Combustion, The Combustion Institute (1958).
21. Reed, S.B., Comb. & Flame 11, 177 (1967).

22. Weinberg, F.J., Wilson, J.R., Comb. & Flame 10, 89 (1966).

23. Mak, F., U.K.Gas Council Report No. I381 (Midlands Research Station).

24. Mak, F., J.I.Gas E. 11, 305 (1971) Discussion Contribution.

25. Edmondson, H., Heap, M.P., Gas World, 166, 288 (1968).

26. Edmondson, H., Heap, M.P., 12th Int. Symp. on Combustion, 1007, The Combustion Institute (1969).

27. Edmondson, H., Heap, M.P., Comb. & Flame 15, 179 (1970).

28. Edmondson, H., Heap, M.P., J.I.Gas.E. 11, 305 (1971).

29. Reed, S.B., J.I.Gas.E. 11, 305 (1971) Discussion Contribution.

30. Reed, S.B., Comb. & Flame 17, 105 (1971).

31. Hottel, H.C., Toong, T.Y., Martin, J.J., Jet Propulsion 1, 28, (1957).

32. Smith, R.A., J.I.Gas.E., 11, 305 (1971) Discussion Contribution.

33. Datta, P., Reed, S.B., Comb. & Flame 19, 89 (1972).

34. Masterman, C.A., Dunning, E.W.B., Densham, A.B., Trans. Inst. Gas Eng. 85, 95 (1935).

35. Reed, S.B., Wakefield, R.P., J.I.Gas.E. 10 (2), 77 (1970).

36. Reed, S.B., Datta, P., Mineur, J., J.Inst.F., 44 (361), 111 (1971).

37. Reed, S.B., Mineur, J., McNaughton, J., J.Inst.F. 44 (362), 149 (1971).

38. Hoggarth, M.L., J.I.Gas.E., 11, 305 (1971) Discussion Contribution.

39. Ewins, R.A., Private Communication.

40. Ewins, R.A., Comb. & Flame 18 (3), 476 (1972).

41. Edmondson, H., Heap, M.P., Comb. & Flame 14, 191 (1970).

42. Dugger, G.L., Gerstein, M., N.A.C.A. Report No. 1300 (1959).

43. Edmondson, H., Heap, M.P., Pritchard, R., Comb. & Flame 14, 195 (1970).

44. Andrews, G.E., Bradley, D., Comb. & Flame 18, 133 (1972).

45. Coward, H.F., Brinsley, F., J.Chem.Soc. 105, 1859 (1914).

46. Clusius, K., Kölsch, W., Waldmann, L., Z. Physik. Chem. (Leipsig), A 189, 131 (1941).

47. Clusius, K., Faber, G., Z. Naturforsch. 2 A, 97 (1947).

48. Manton, J., Von Elbe, G., Lewis, B., J.Chem.Phys. 20, 153 (1952).

49. Markstein, G.H., J.Chem.Phys. 17, 428 (1949).

50. Markstein, G.H., J. Aeronaut. Sci. 18, 199 (1951).

51. Smith, F.A. Pickering, S.F., J. Research Natl. Bur. Standards 3, 65 (1929).

52. Broida, H.P., Kane, W., J.Chem. Phys., 20, 1042 (1952).

53. Gaydon, A.G., Wolfhard, H.G., "Flames, their structure, radiation and temperature", Chapman and Hall (1960).

54. Prandtl, L., "Ströhmungslehre", Viewegbrunswick, 370 (1942).

55. Bubnoff, N., Z.Phys. Chem., 88, 641 (1914).

56. Haber, F., Z.Phys. Chem. 88, 670 (1914).

57. Jost, W., Krischer, B., Z.Phys. Chem. (N.F.) 5, 398 (1955).

58. Jost, W., Krug, J., Sieg. L., 4th Int. Symp. on Combustion, 535, The Combustion Instutute (1953).

59. Markstein, G.H., 7th Int. Symp. on Combustion, 289, the Combustion Institute (1958).

60. Harris, J., Lovelace, D.E., J.I.Gas.E., 8, 169 (1968).

61. Myers, A., Ph.D.Thesis, University of Aston (1972).

62. Evans, M.W., Chem. Revs. 51, 363 (1952).

63. Landau, L.D., Acta. Physicochim. (U.S.S.R.) 19, 77 (1944).

64. Landau, L.D., Liftschift, E.M., Mechanics of Continuous Media, Moscow (1953).

65. Emmons, H.W., Fundamentals of Gas Dynamics, Princetown University Press, (1953).

66. Eckhaus, W., M.I.T. Fluid Dynamics Research Group Report No.59-9 (1959).

67. Eckhaus, W., J.Fluid Mech. 10, 80 (1961).

68. Dixon-Lewis, G., Garside, J.E., Kilham, J.K., Roberts, A.L., Williams, A., I.Gas.E. Research Communication 861 (1971).

69. Parlange, J-Y., J.Chem.Phys. 48 (4) 1843 (1968).

70. Blandon, E.E., J.I.Gas.E. 8, 157 (1968) Discussion Contribution.

71. Wires, W., Watermeier, L.A., Strehlow, L.A., J.Phys. Chem. 63, 989, (1959).

72. Markstein, G.H., "Non-Steady Flame Propagation", Pergamon Press (1964).

73. Massey, B.S., "Units, Dimensions, Dimensional Analysis, and Physical Similarity", Van Nostran Reinhold (1971).

74. Sprange, M., Reed, S.B., J.Ind. & Eng. Chem. (Fundamentals) 7, 651 (1968).

75. Cremer, H.W., Davies, T., "Chemical Engineering Practice" 4, Fluid States, Butterworth (1957).

76. Bird, R.B., Stewart, W.E., Lightfoot, E.N., "Transport Phenomena", Wiley (1960).

77. Pritchard, R., J.I.Gas.E. 11, 305 (1971) Discussion Contribution.

78. Fells, I., Rutherford, A.G., Comb. & Flame 13. 131 (1969).

79. Garner, F.H., Long, R., Thorley, B., 4th Int. Symp.on Combustion, 306, the Combustion Institute (1952).

80. Gray, K.L., Linnett, J.W., Mellish, C.E., Trans. Faraday Soc. 48, 1155 (1952).

81. Andrews, G.E., Bradley, D., Comb. & Flame 19 (2) 275 (1972).

82. Scholte, T.G., Vaags, P.B., Comb. & Flame 3 (4) 511 (1959).

83. Pickering, H.S., Linnett, J.W., Trans. Faraday Soc. 47, 981 (1951).

84. Senior, D.A., Comb. & Flame 5, 7 (1961).

85. Fristrom, R.M., Westenburg, A.A., "Flame Structure", McGraw-Hill (1965).

86. Woodcock, C., Ph.D. Thesis, University of Aston (1971).

87. Atamar, S., Ph.D. Thesis, University of Aston (1972).

88. Reiter, S.H., Wright, C.C., J.Ind. & Eng. Chem. 42, 691 (1950).

89. Grumer, J., Harris, M.E., Rowe, V.R., Rep.Investigations of U.S. Bureau of Mines No.5225 (1956).

90. Van Krevelen, D.W., Chermin, H.A.G., 7th Int.Symp. on Combustion, 358, The Combustion Institute (1958).

91. Leason, D.B., 4th Int. Symp. on Combustion, 369, The Combustion Institute (1952).

92. Mellish, C.E., Linnett, J.W., 4th Int. Symp. on Combustion, 407, The Combustion Institute (1952).

93. Clingman, W.H., Brokaw, R.S., Pease, R.N., 4th Int. Symp. on Combustion, 310, The Combustion Institute (1952).

94. Dixon-Lewis, G., Proc. Roy. Soc. A 317, 235 (1970).

95. Dixon-Lewis, G., Proc. Roy. Soc. A 330, 199 (1972).

96. Dixon-Lewis, G., Proc. Roy. Soc. A 307, 111 (1968).

97. Van Heyningen, G.J.J., Gas 89, 284 (1969).

Appendix 1.

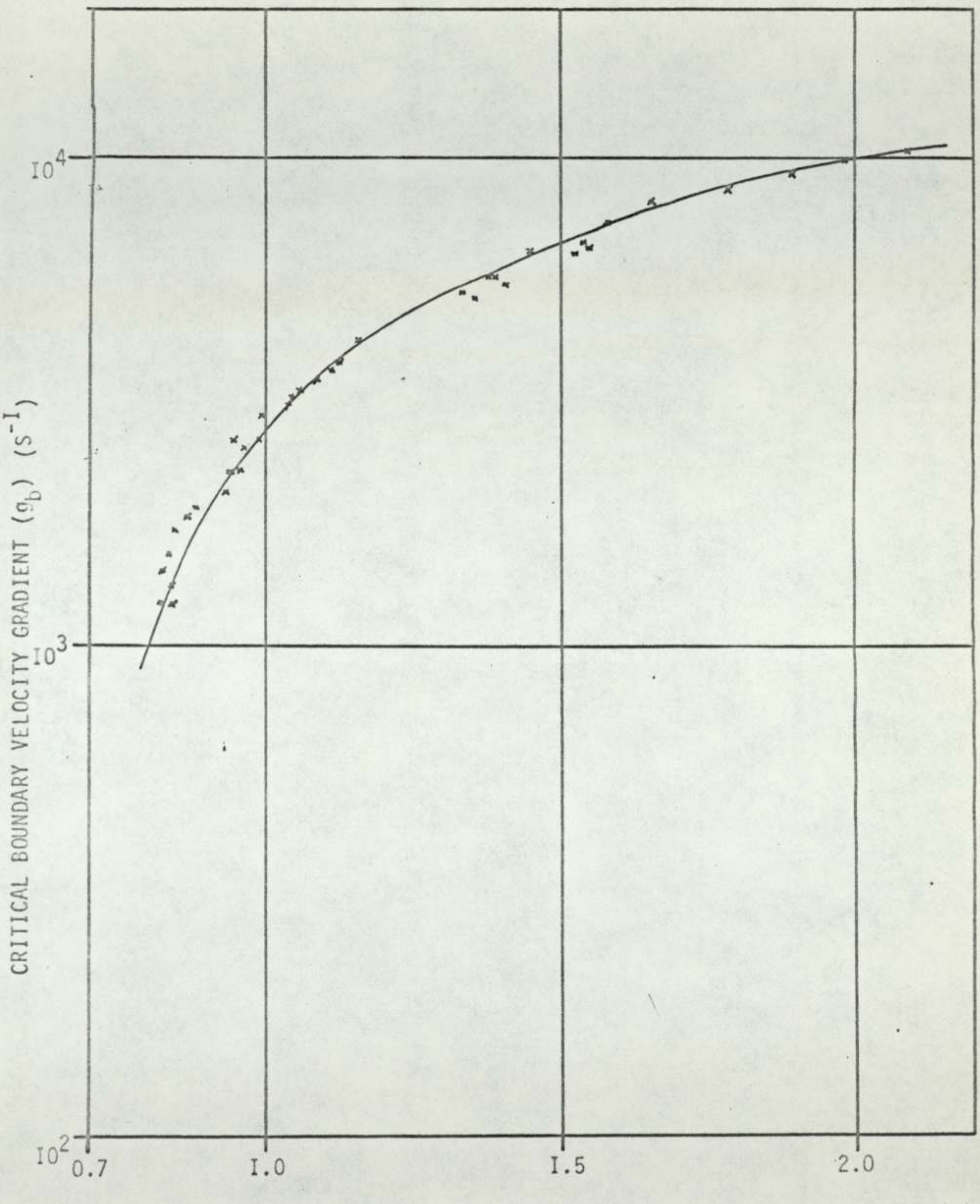
GRAPHS.

GRAPHS NOS. I-8

CRITICAL BOUNDARY VELOCITY GRADIENT PLOTTED AGAINST FUEL CONCENTRATION FOR BLOW OFF OF LAMINAR BURNER FLAMES USING THE HYDROGEN-PROPANE-AIR SYSTEM.

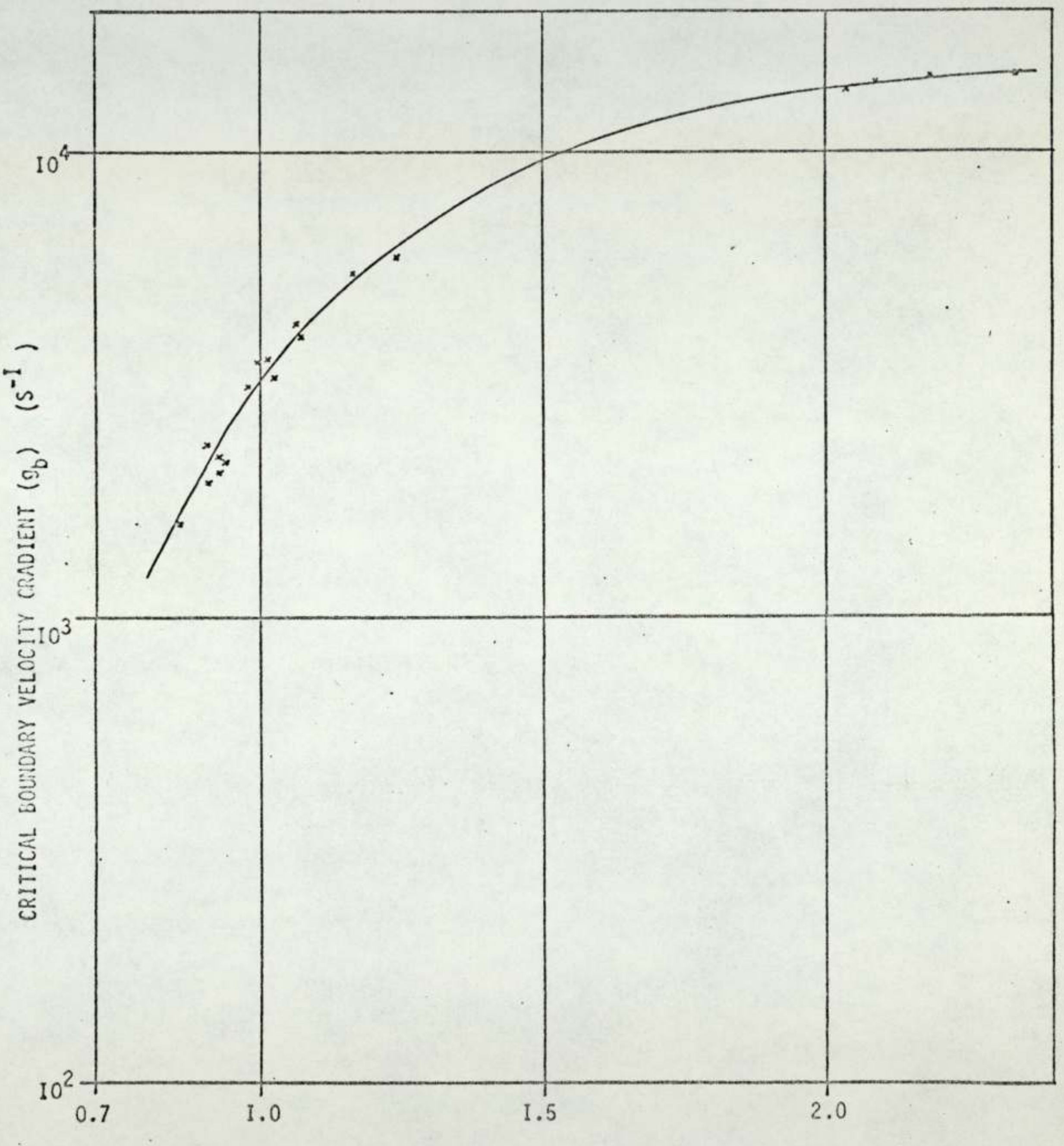
PRIMARY OXIDANT: AIR

SECONDARY ATMOSPHERE: AIR



FUEL GAS: 100% PROPANE

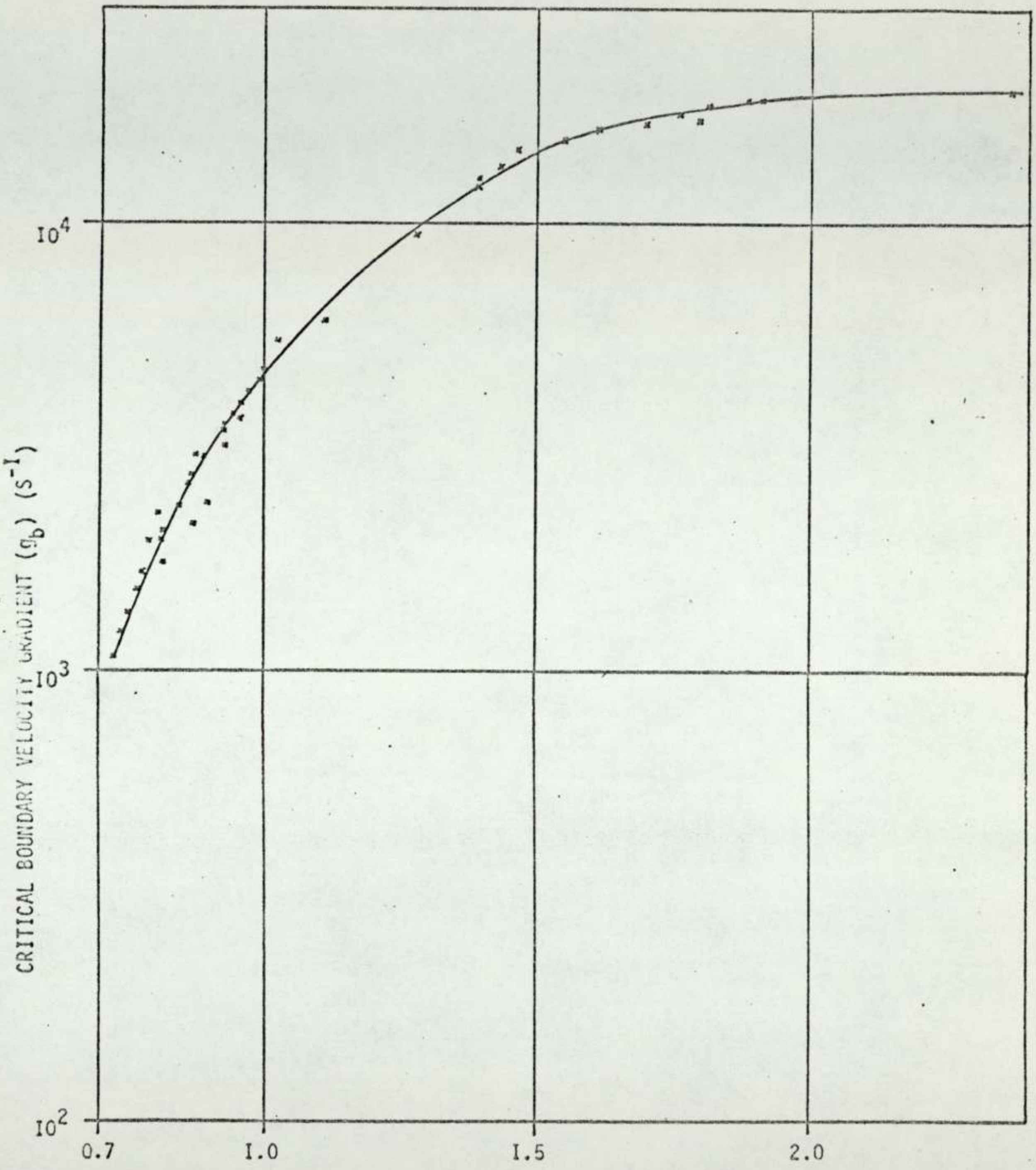
GRAPH NO. I



FUEL CONCENTRATION EXPRESSED AS A FRACTION OF STOICHIOMETRIC (X)

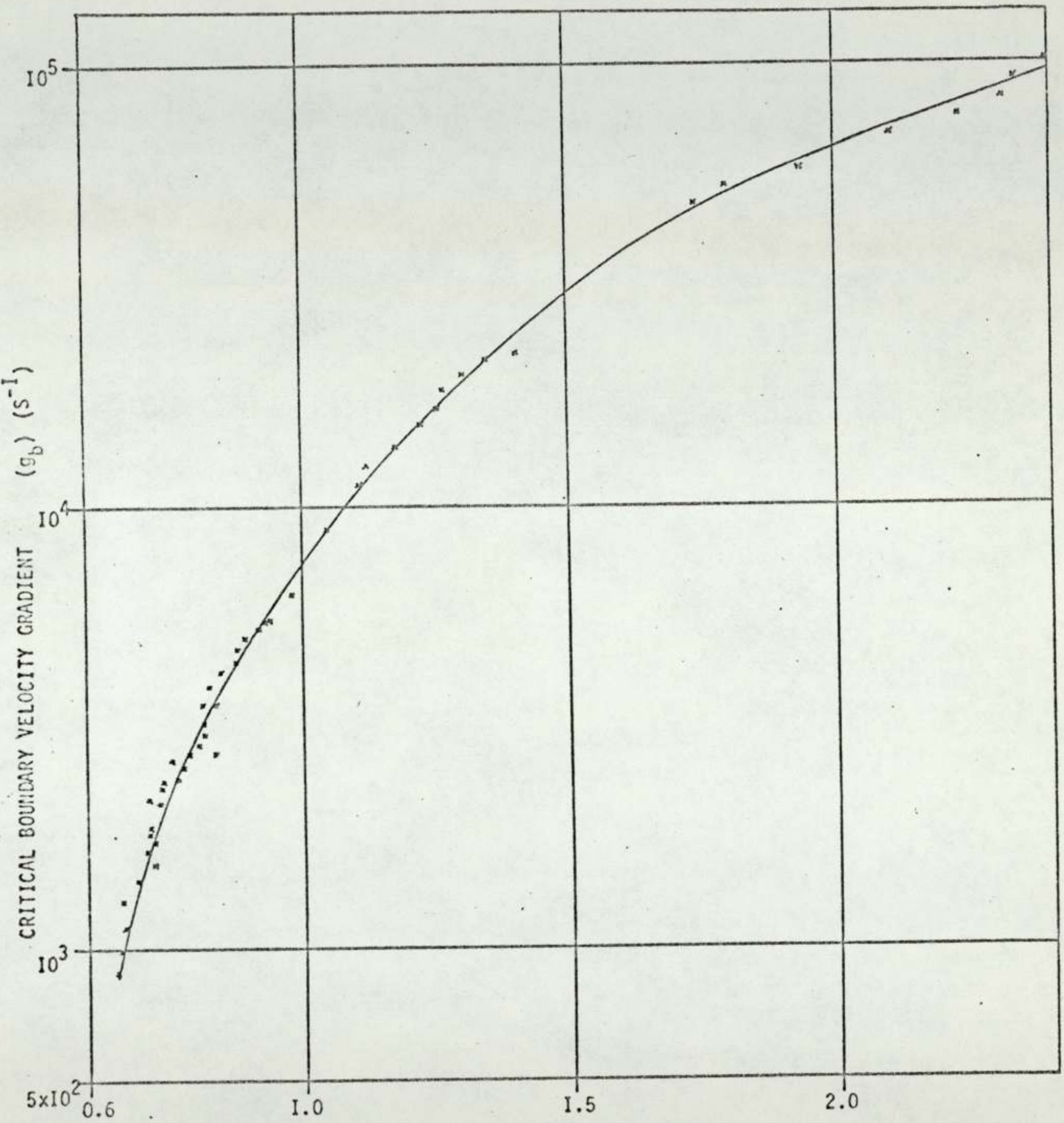
FUEL GAS: 80% PROPANE 20% HYDROGEN

<u>GRAPH NO. 2</u>



FUEL GAS: 60% PROPANE 40% HYDROGEN

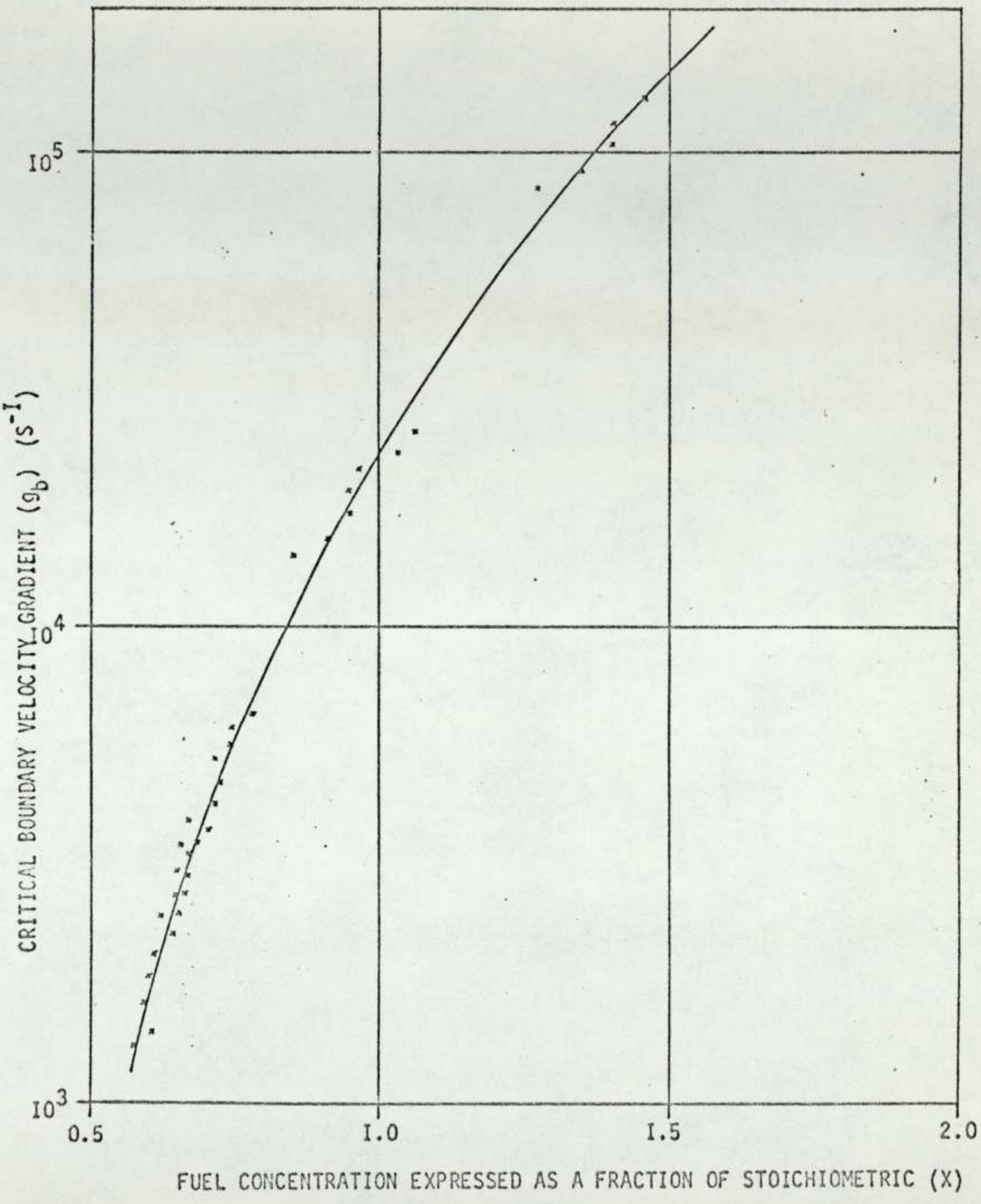
GRAPH NO. 3



FUEL CONCENTRATION EXPRESSED AS A FRACTION OF STOICHIOMETRIC (X)

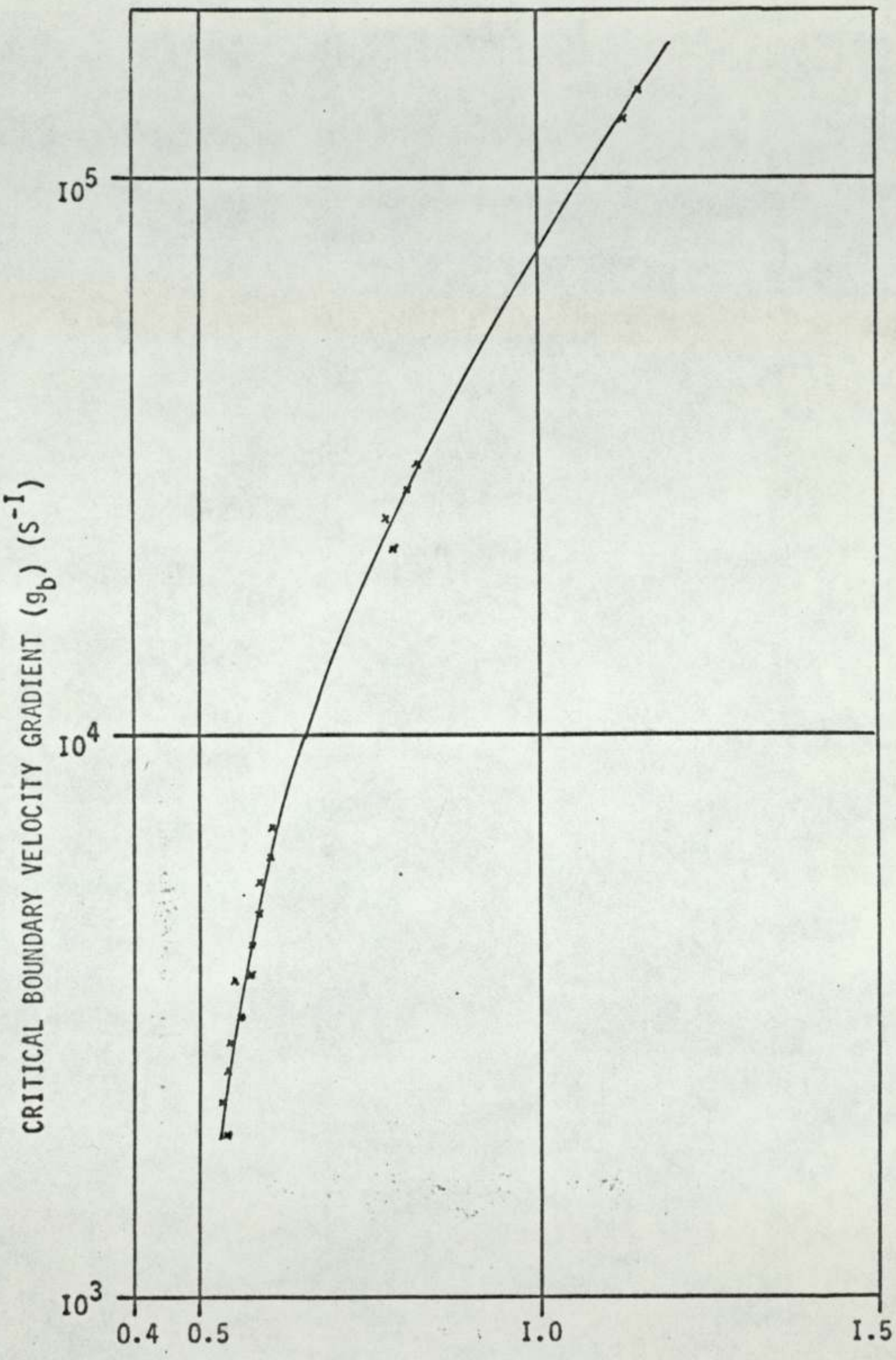
FUEL GAS: 40% PROPANE 60% HYDROGEN

GRAPH NO. 4



FUEL GAS: 20% PROPANE 80% HYDROGEN

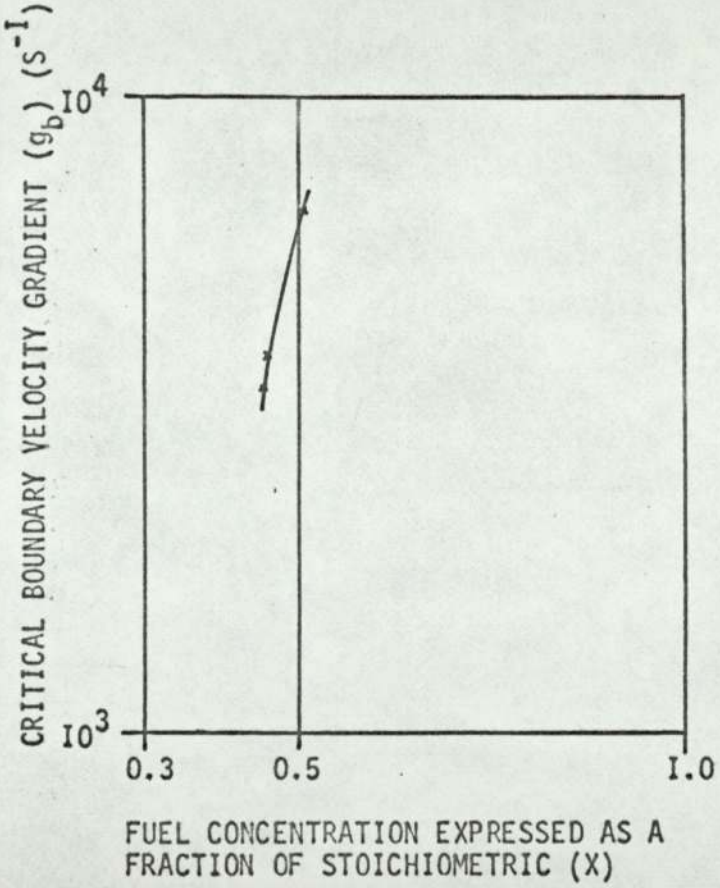
GRAPH NO. 5



FUEL CONCENTRATION EXPRESSED AS A FRACTION OF STOICHIOMETRIC (X)

FUEL GAS: I0% PROPANE 90% HYDROGEN

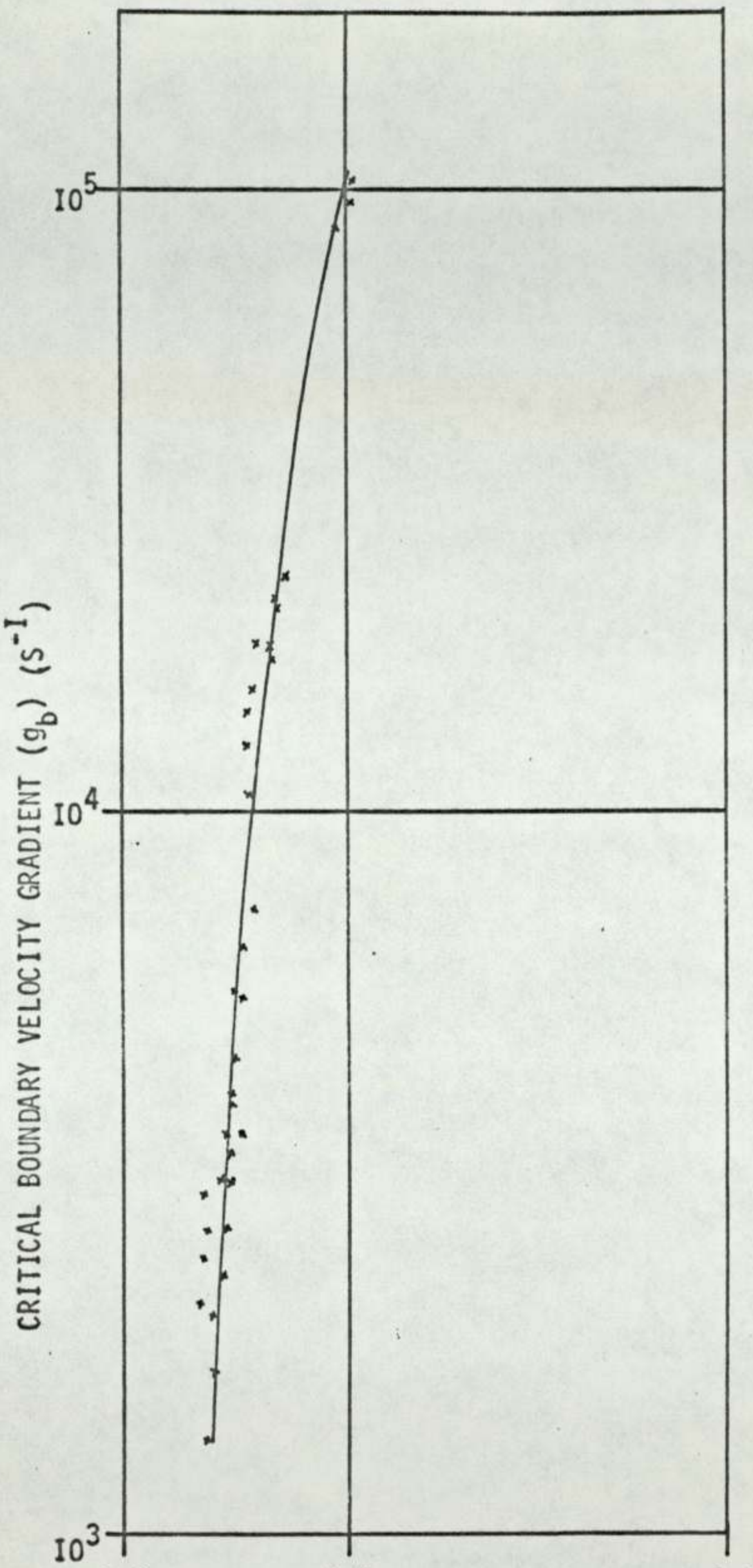
GRAPH NO. 6



FUEL CONCENTRATION EXPRESSED AS A FRACTION OF STOICHIOMETRIC (X)

FUEL GAS: 5% PROPANE 95% HYDROGEN

GRAPH NO. 7



FUEL GAS: I00% HYDROGEN

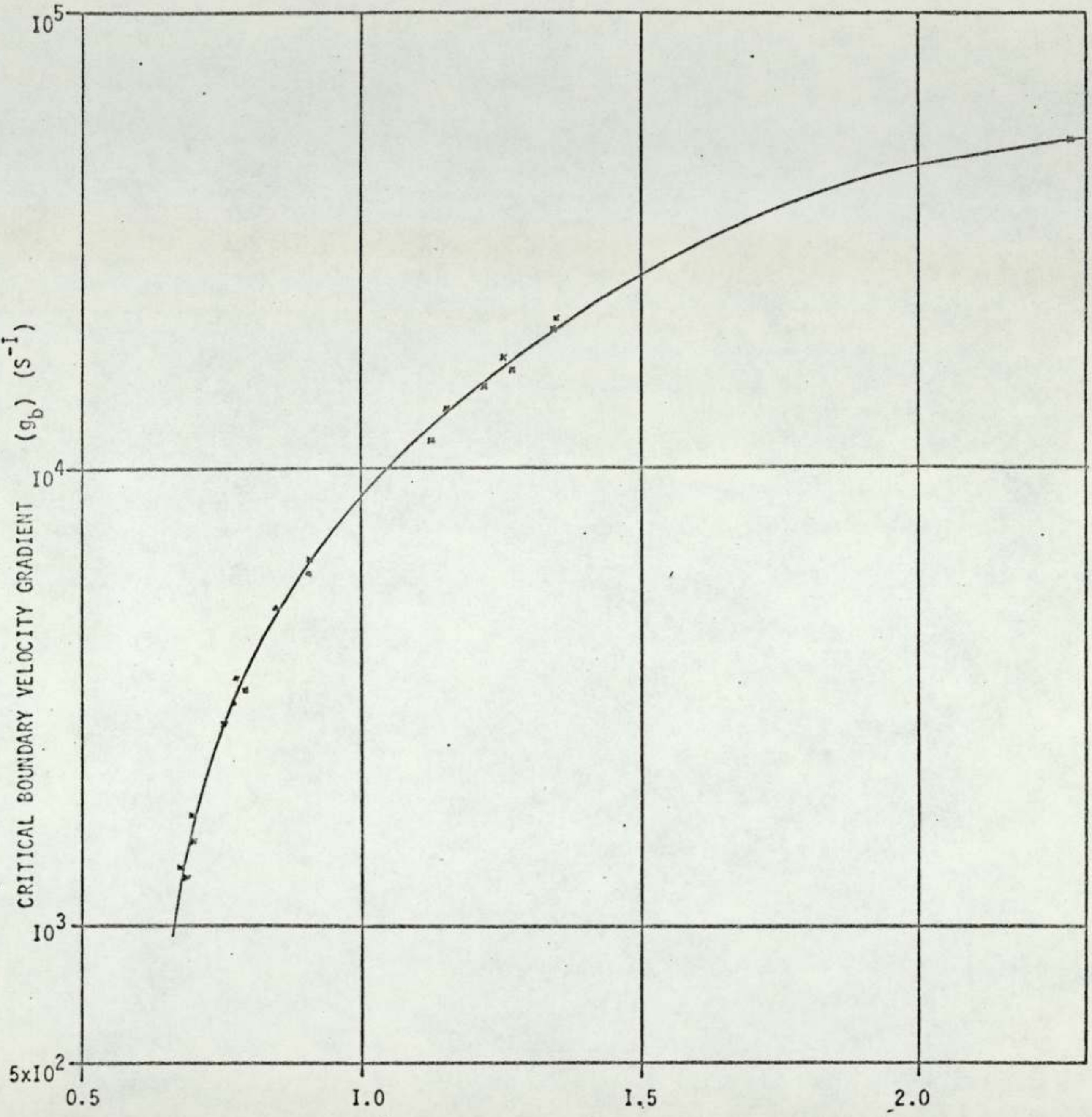
GRAPH NO. 8

GRAPH NO. 9

CRITICAL BOUNDARY VELOCITY GRADIENT PLOTTED AGAINST FUEL CONCENTRATION FOR BLOW OFF OF LAMINAR BURNER FLAMES USING THE ETHYLENE-AIR SYSTEM.

PRIMARY OXIDANT: AIR

SECONDARY ATMOSPHERE: AIR



FUEL GAS: ETHYLENE

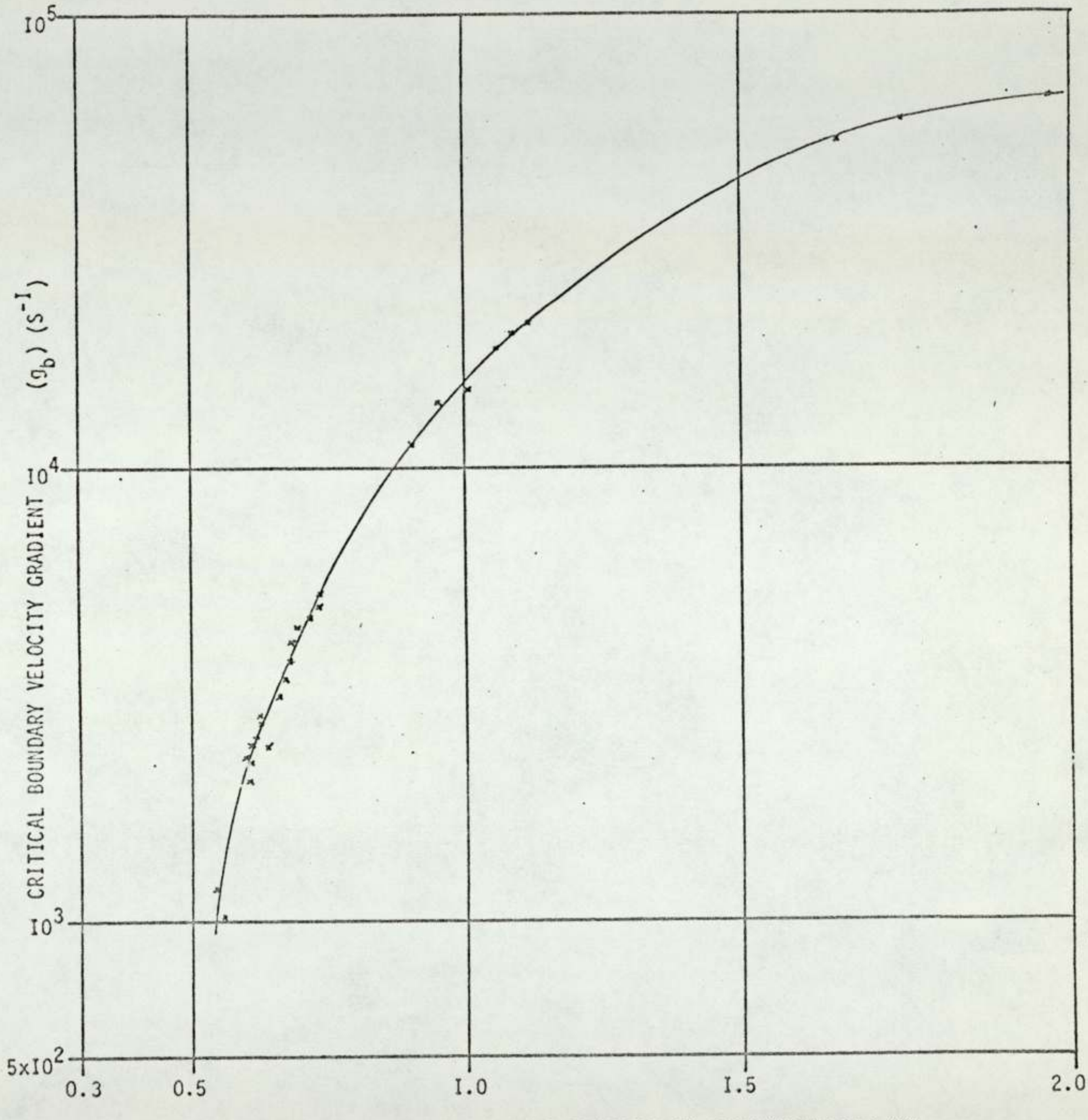
GRAPH NO. 9

GRAPHS NOS. IO-I2

CRITICAL BOUNDARY VELOCITY GRADIENT PLOTTED AGAINST FUEL CONCENTRATION FOR BLOW OFF OF LAMINAR BURNER FLAMES USING PROPANE AND HYDROGEN AS FUEL GASES.

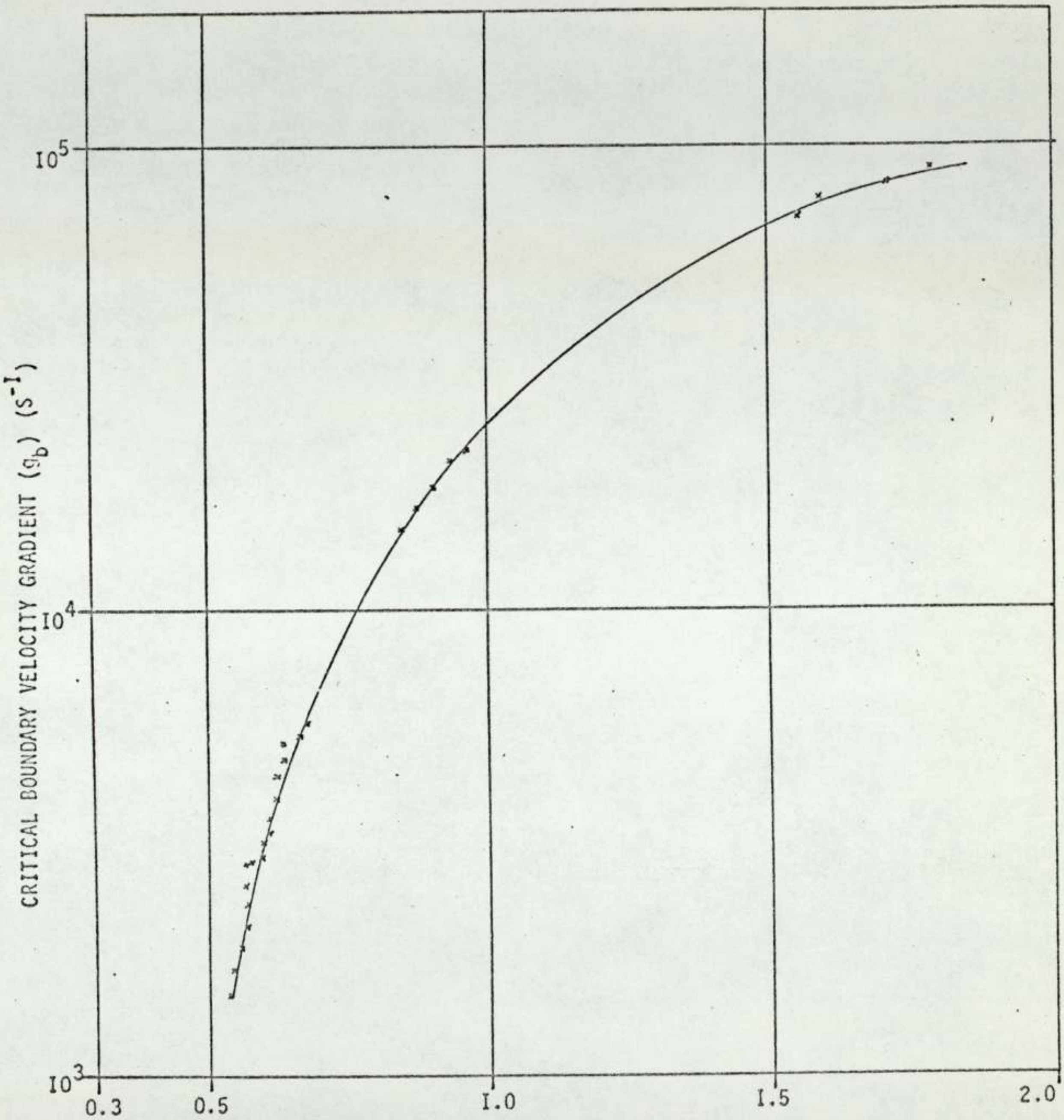
PRIMARY OXIDANT: 2I% OXYGEN 79% ARGON

SECONDARY ATMOSPHERE: 2I% OXYGEN 79% ARGON



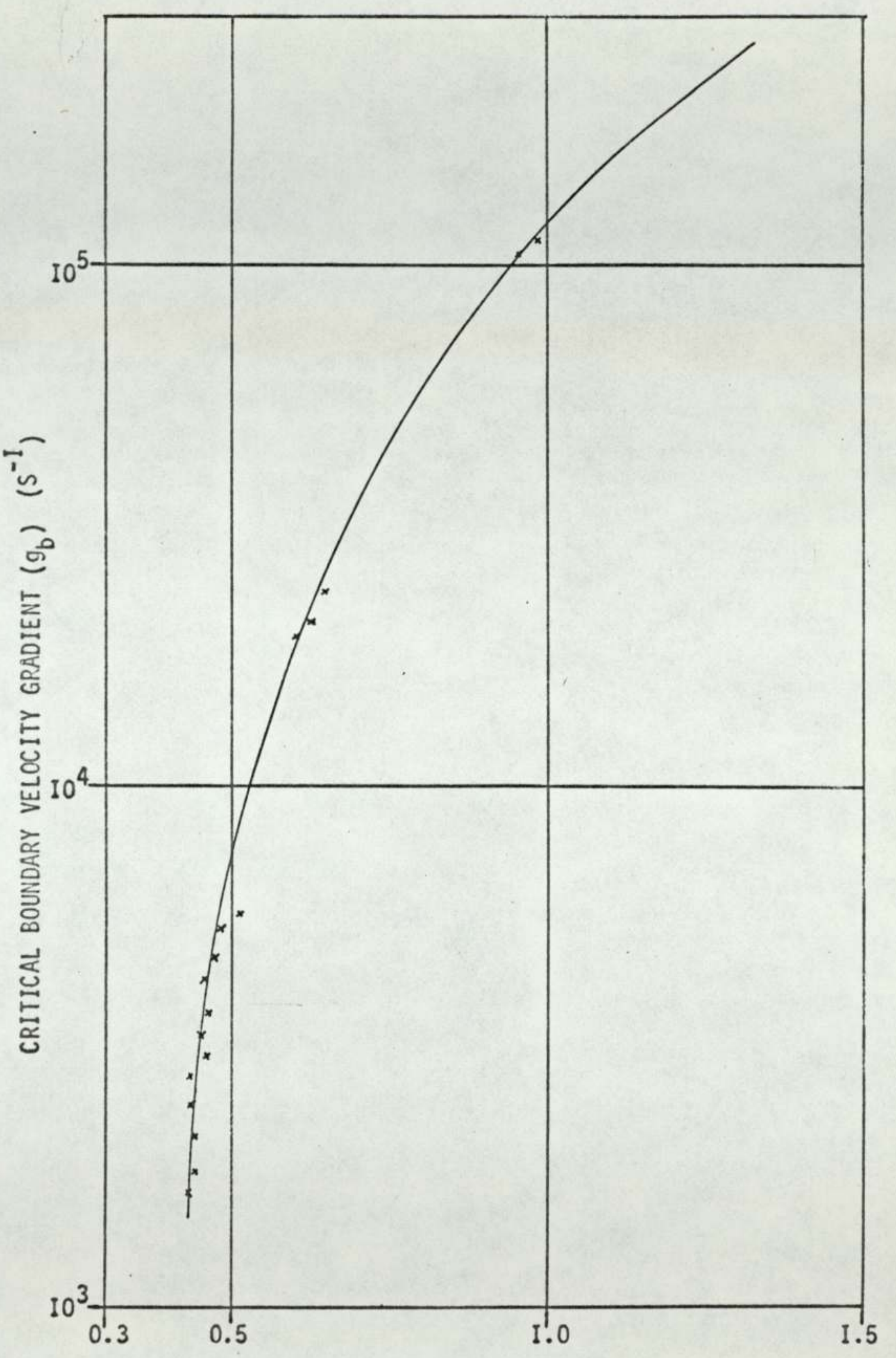
FUEL GAS: 100% PROPANE

GRAPH NO. 10



FUEL GAS: 60% PROPANE 40% HYDROGEN

GRAPH NO. II



FUEL CONCENTRATION EXPRESSED AS A FRACTION OF STOICHIOMETRIC
(X)

FUEL GAS: 20% PROPANE 80% HYDROGEN

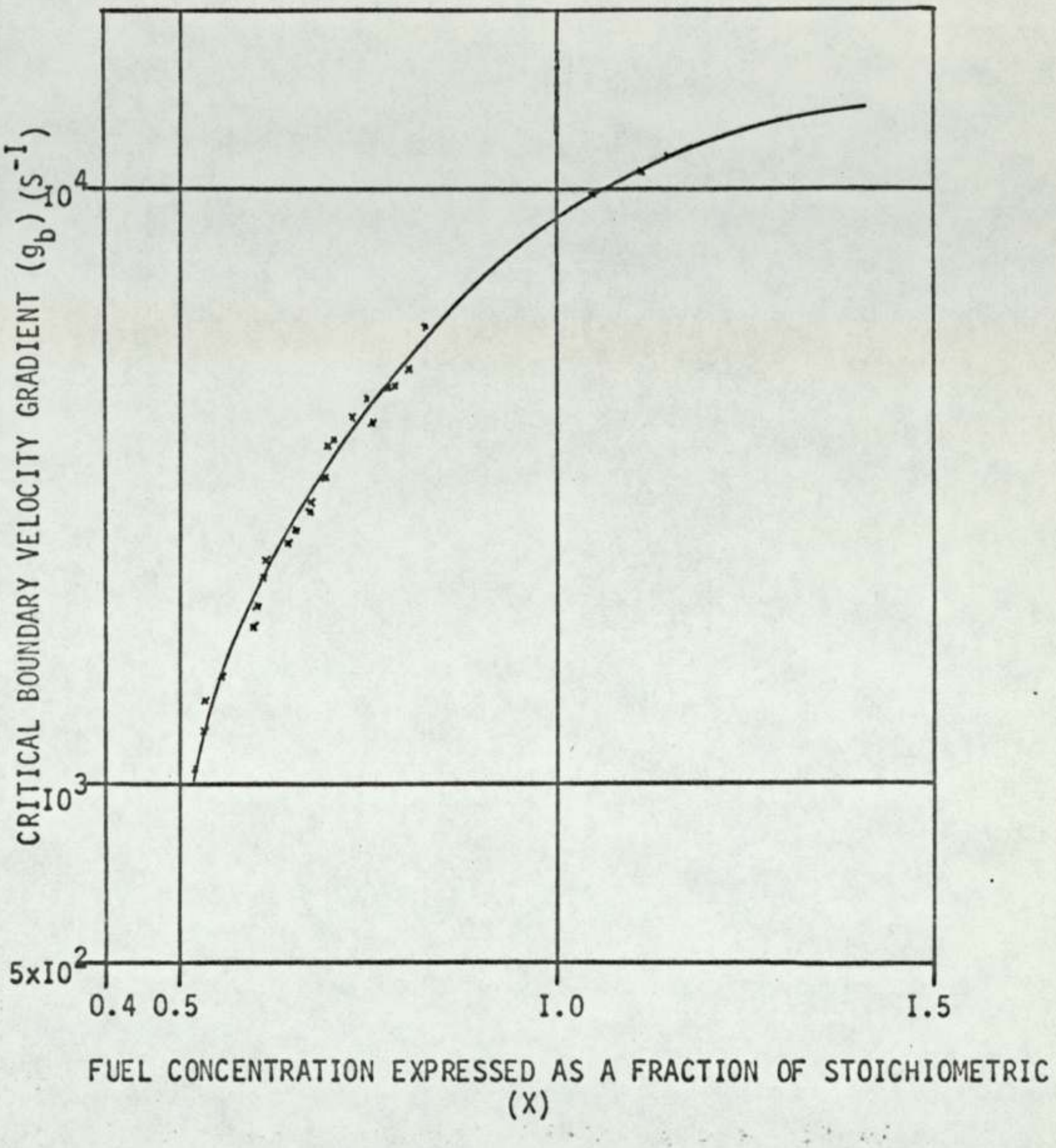
GRAPH NO. 12

GRAPHS NOS. 13-15

CRITICAL BOUNDARY VELOCITY GRADIENT PLOTTED AGAINST FUEL CONCENTRATION FOR BLOW OFF OF LAMINAR BURNER FLAMES USING PROPANE AND HYDROGEN AS FUEL GASES.

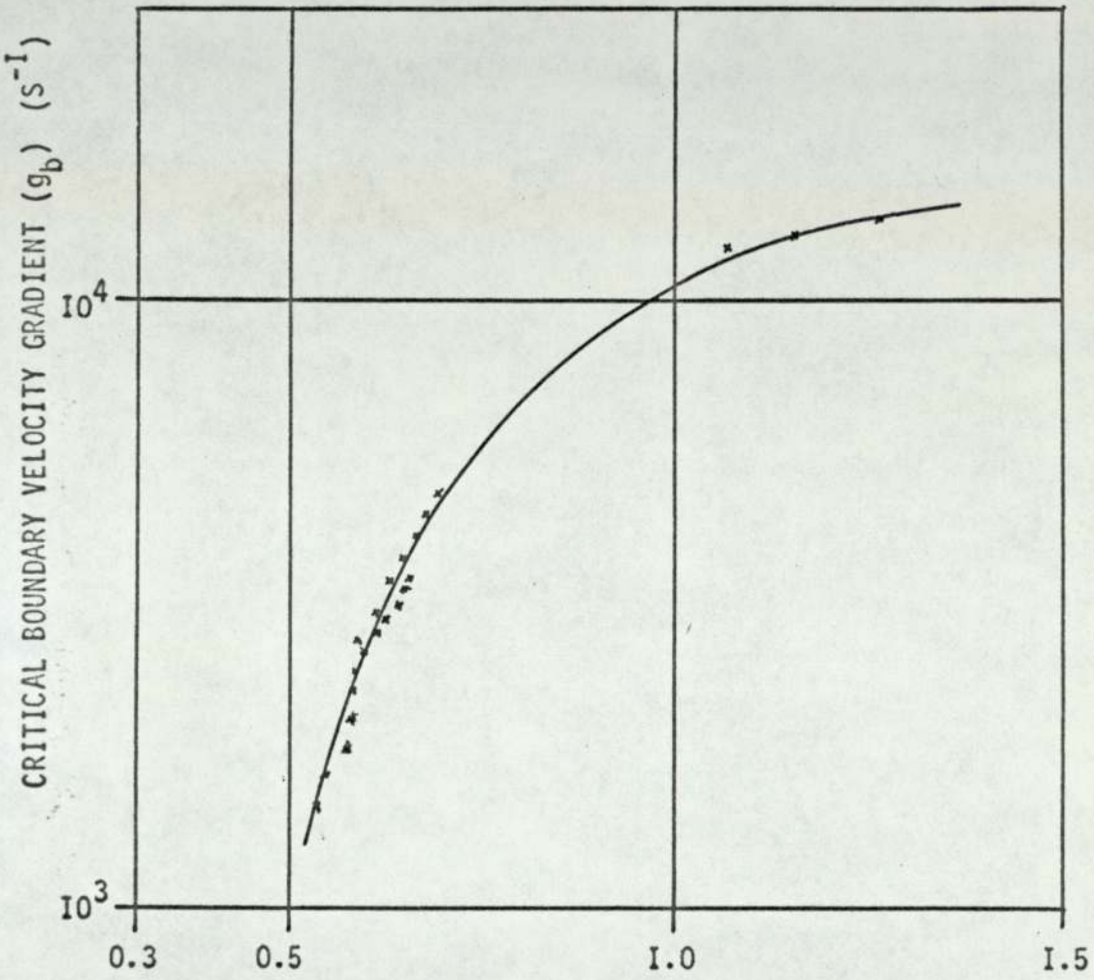
PRIMARY OXIDANT: 21% OXYGEN 79% HELIUM

SECONDARY ATMOSPHERE: 21% OXYGEN 79% HELIUM



FUEL GAS: 100% PROPANE

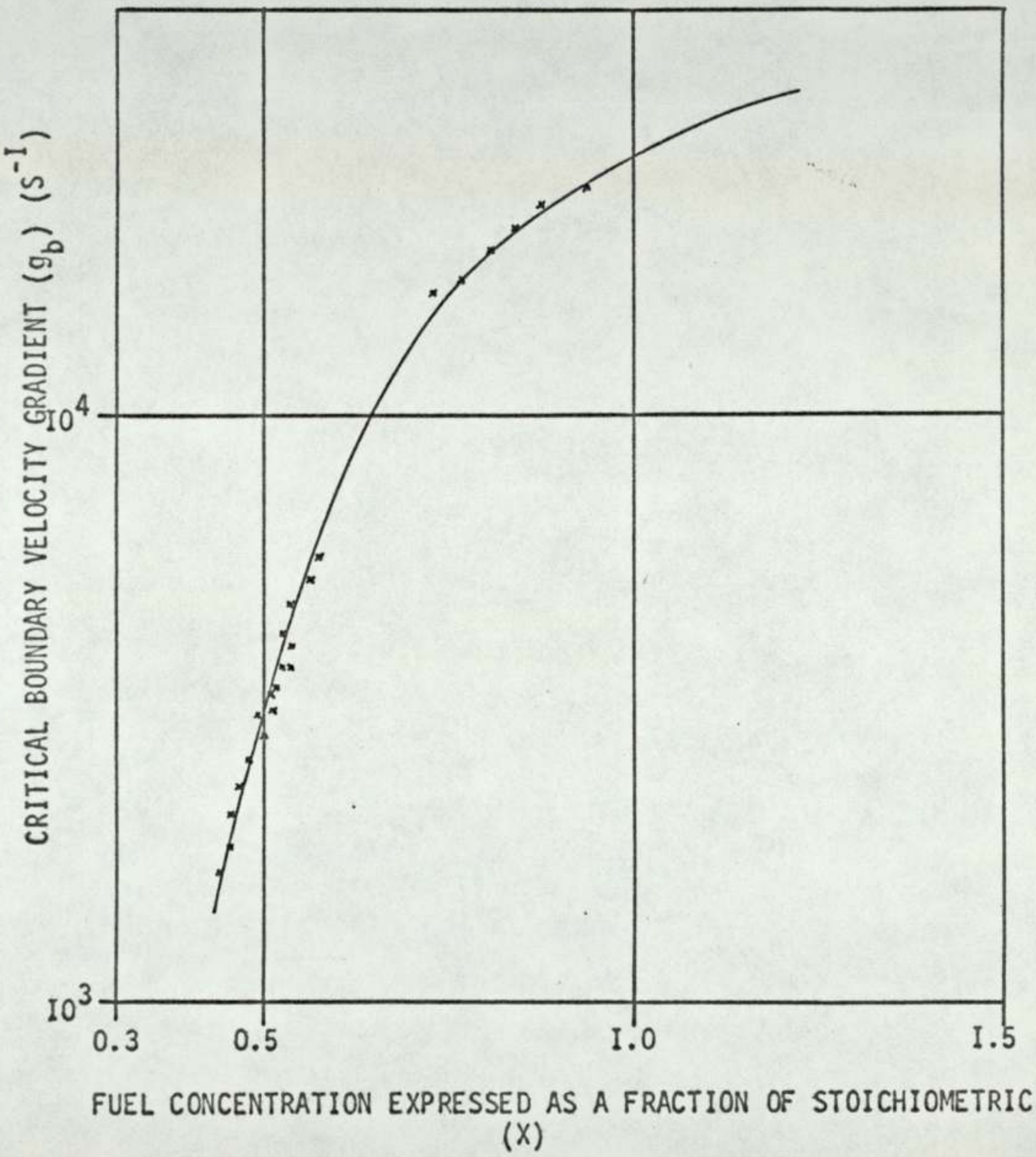
GRAPH NO. 13



FUEL CONCENTRATION EXPRESSED AS A FRACTION OF STOICHIOMETRIC (X)

FUEL GAS: 60% PROPANE 40% HYDROGEN

GRAPH NO. 14



FUEL GAS: 20% PROPANE 80% HYDROGEN

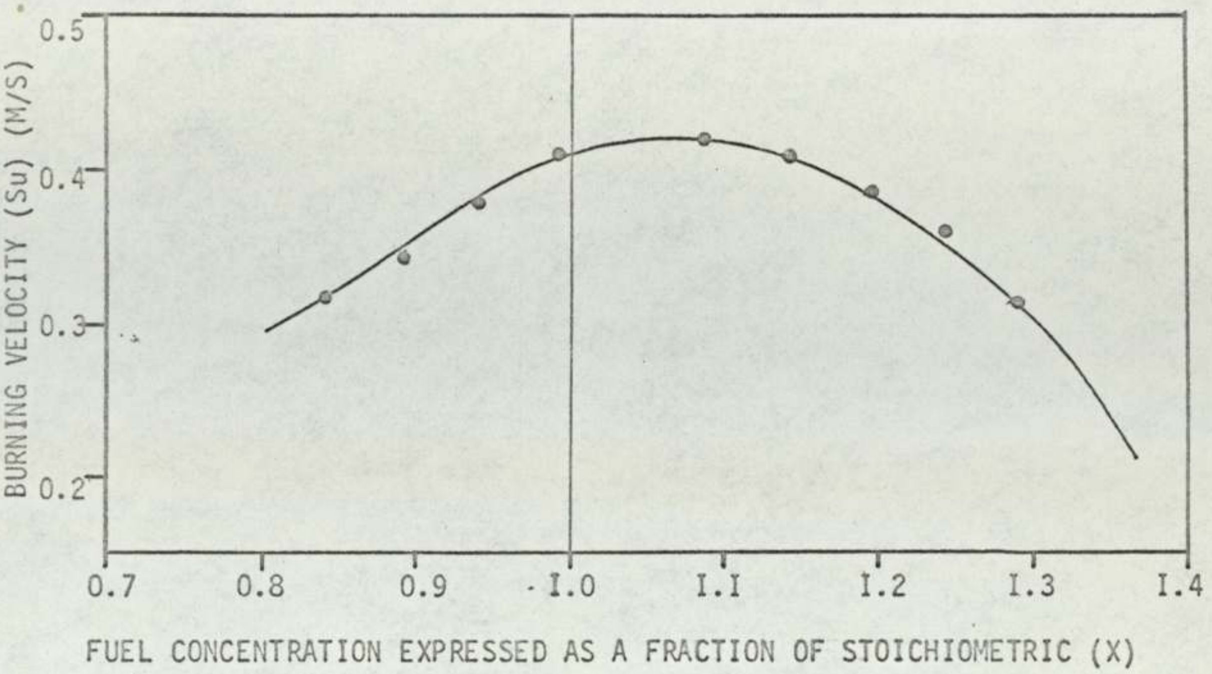
GRAPH NO. I5

GRAPHS NOS. I6-22

BURNING VELOCITY PLOTTED AGAINST FUEL CONCENTRATION FOR THE HYDROGEN-PROPANE-AIR SYSTEM.

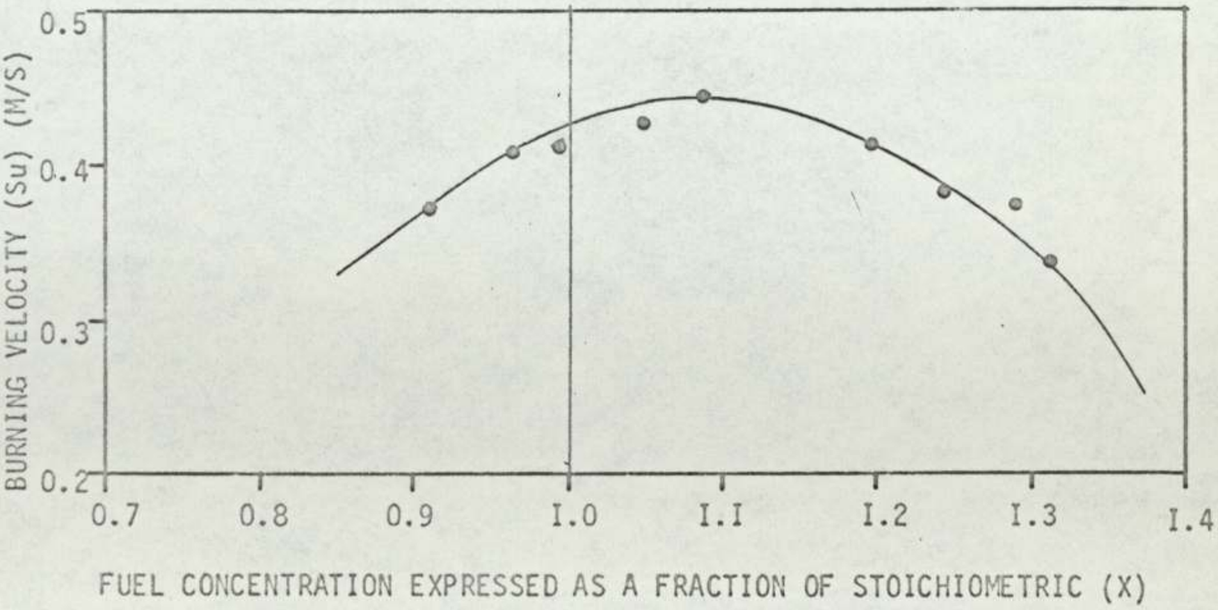
PRIMARY OXIDANT: AIR

SECONDARY ATMOSPHERE: AIR



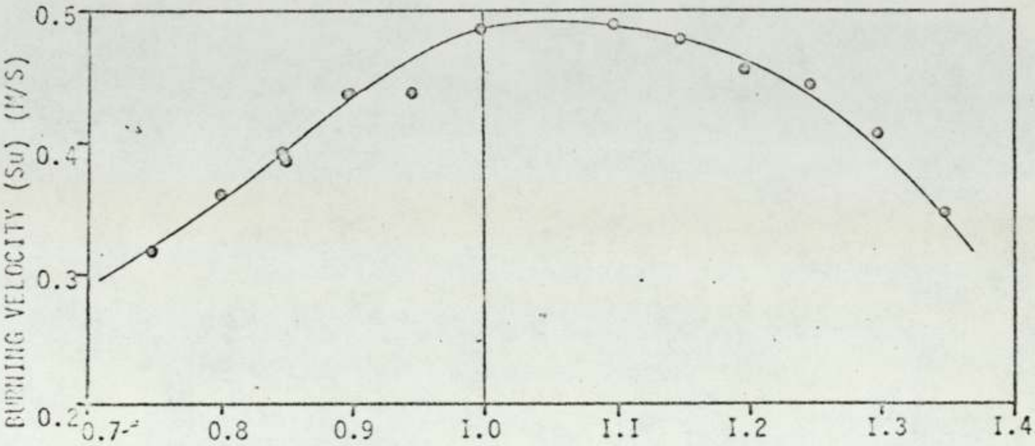
FUEL GAS: I00% PROPANE

GRAPH NO. I6



FUEL GAS: 80% PROPANE 20% HYDROGEN

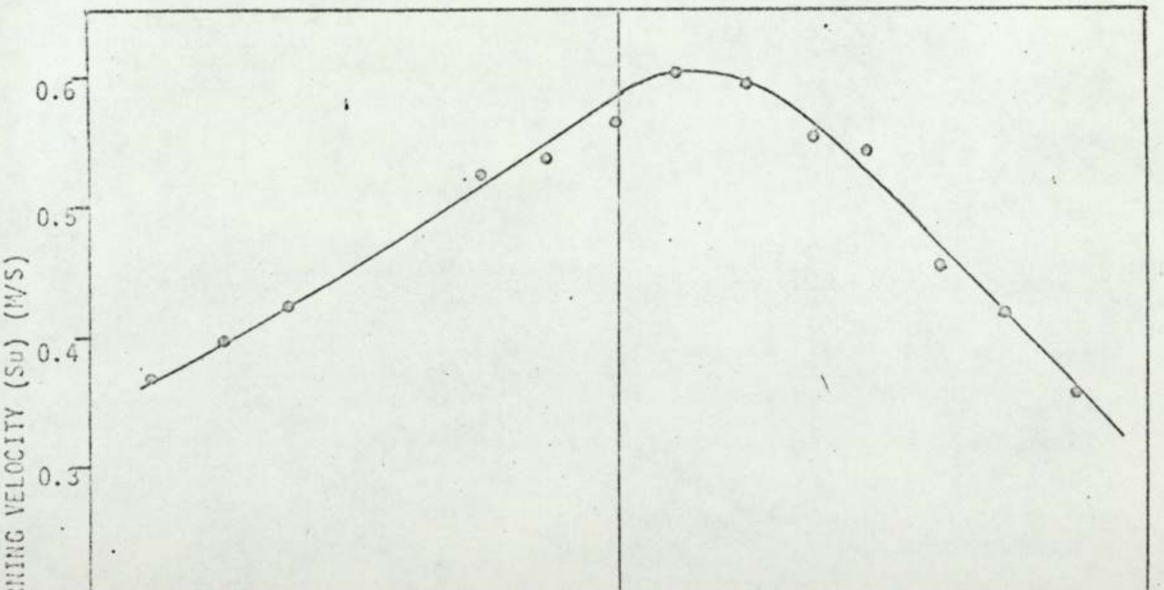
GRAPH NO. I7



FUEL CONCENTRATION EXPRESSED AS A FRACTION OF STOICHIOMETRIC (X)

FUEL GAS: 60% PROPANE 40% HYDROGEN

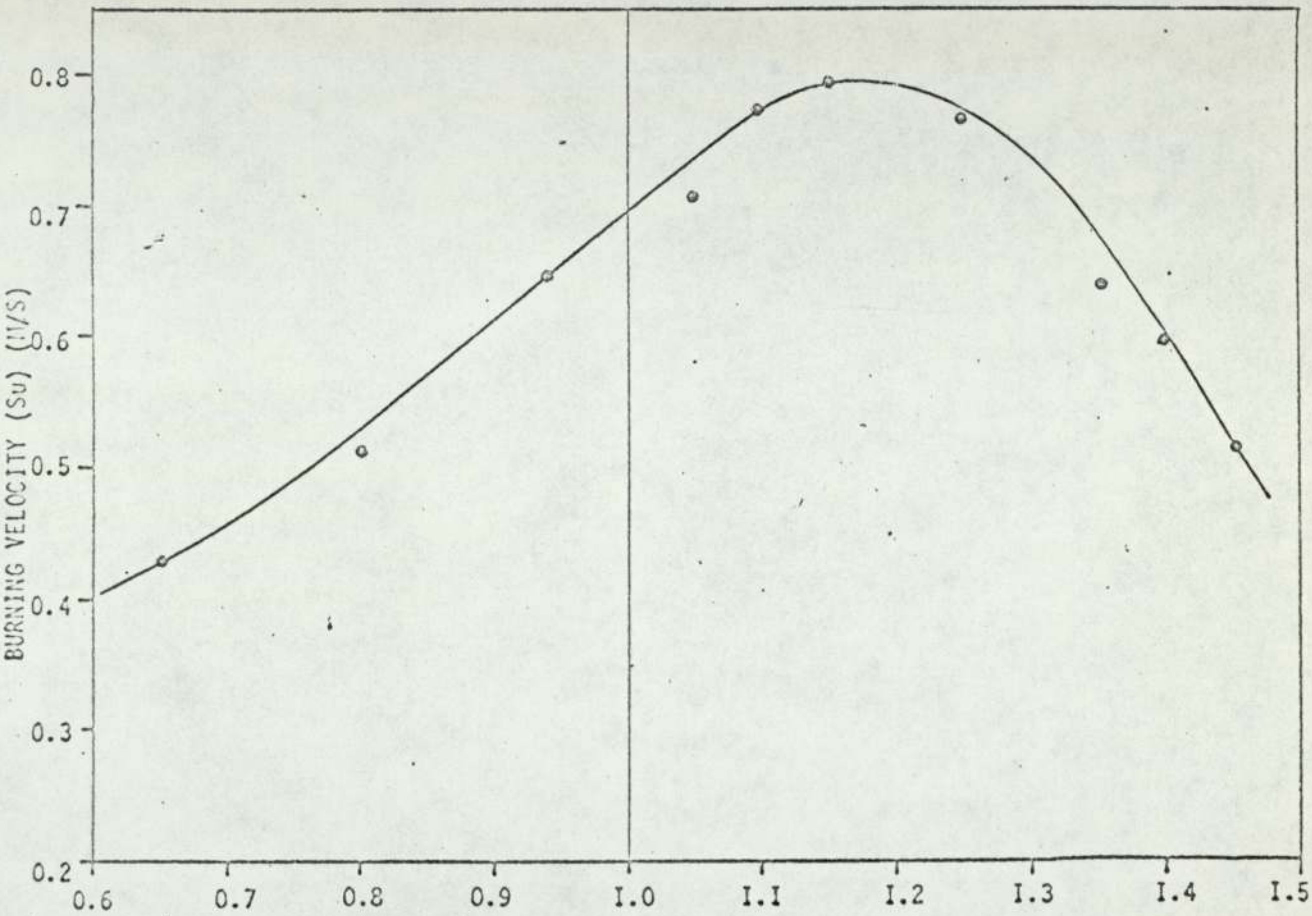
GRAPH NO. I8



FUEL CONCENTRATION EXPRESSED AS A FRACTION OF STOICHIOMETRIC (X)

FUEL GAS: 40% PROPANE 60% HYDROGEN

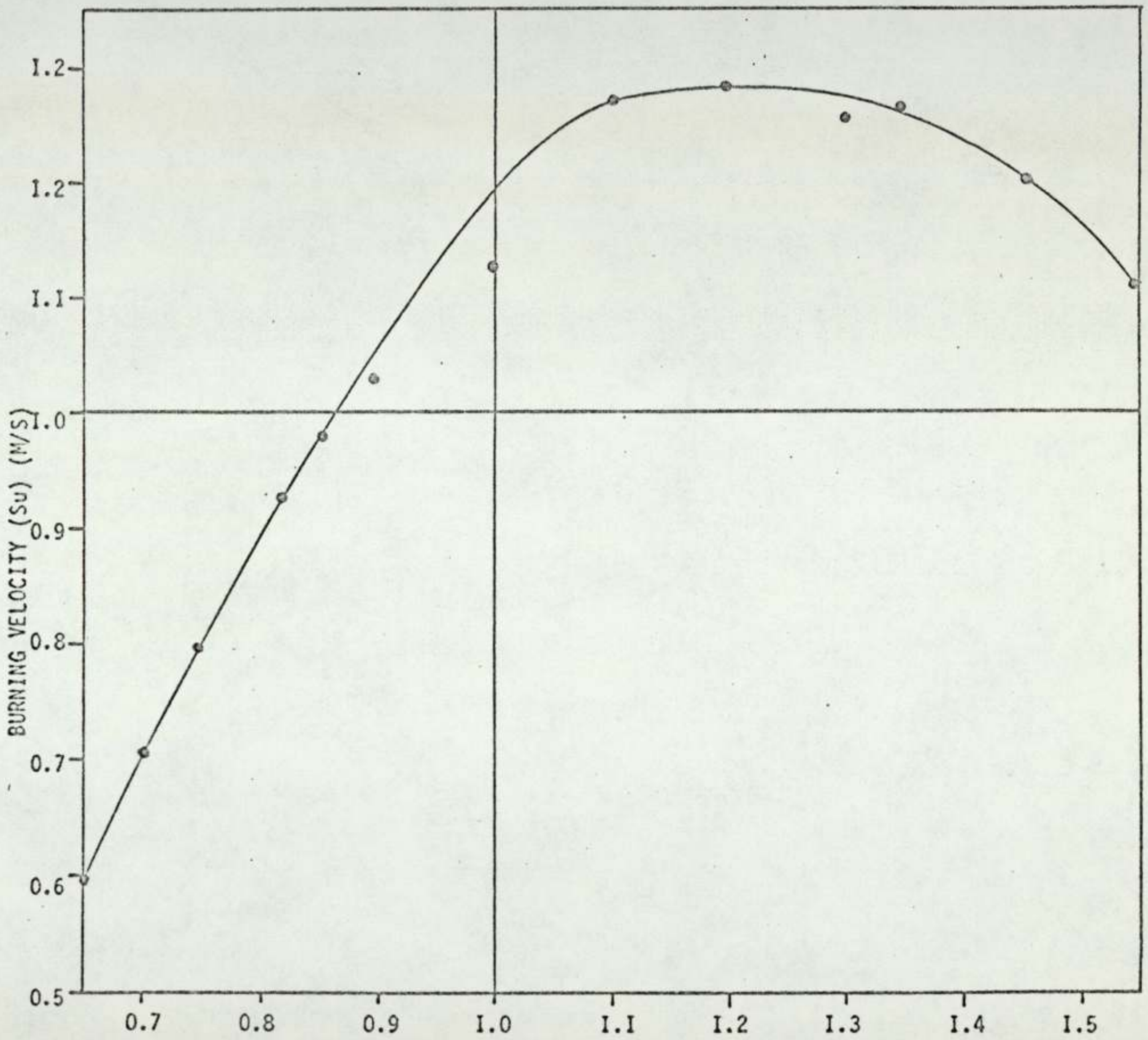
GRAPH NO. I9



FUEL CONCENTRATION EXPRESSED AS A FRACTION OF STOICHIOMETRIC (X)

FUEL GAS: 20% PROPANE 80% HYDROGEN

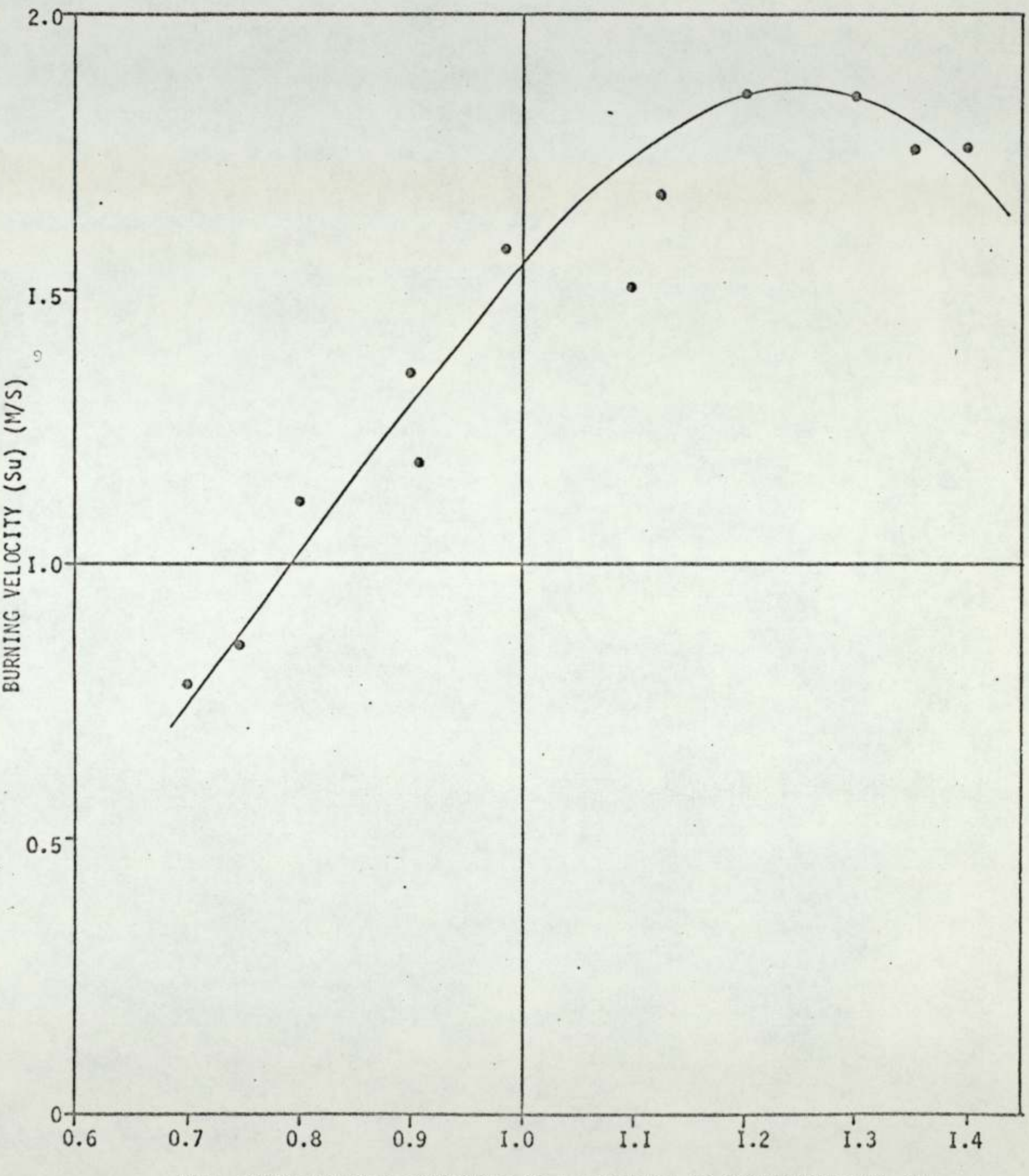
GRAPH NO. 20



FUEL CONCENTRATION EXPRESSED AS A FRACTION OF STOICHIOMETRIC (X)

FUEL GAS: I0% PROPANE 90% HYDROGEN

GRAPH NO. 2I



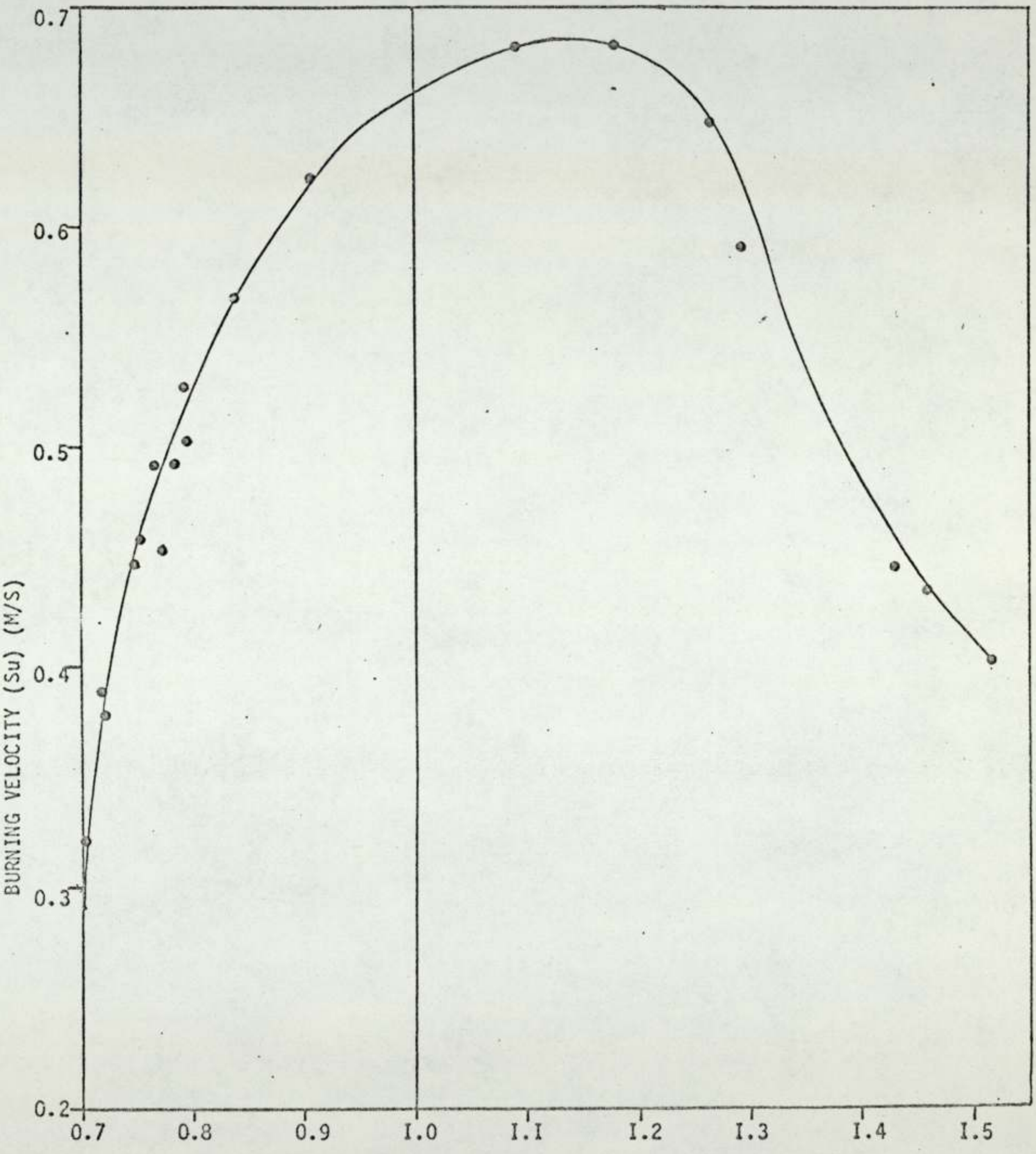
FUEL CONCENTRATION EXPRESSED AS A FRACTION OF STOICHIOMETRIC (X)

FUEL GAS: 5% PROPANE 95% HYDROGEN

GRAPH NO. 22

BURNING VELOCITY PLOTTED AGAINST FUEL CONCENTRATION FOR THE ETHYENE-AIR SYSTEM

PRIMARY OXIDANT: AIR SECONDARY ATMOSPHERE: AIR



FUEL CONCENTRATION EXPRESSED AS A FRACTION OF STOICHIOMETRIC (X)

FUEL GAS: I00% ETHYLENE

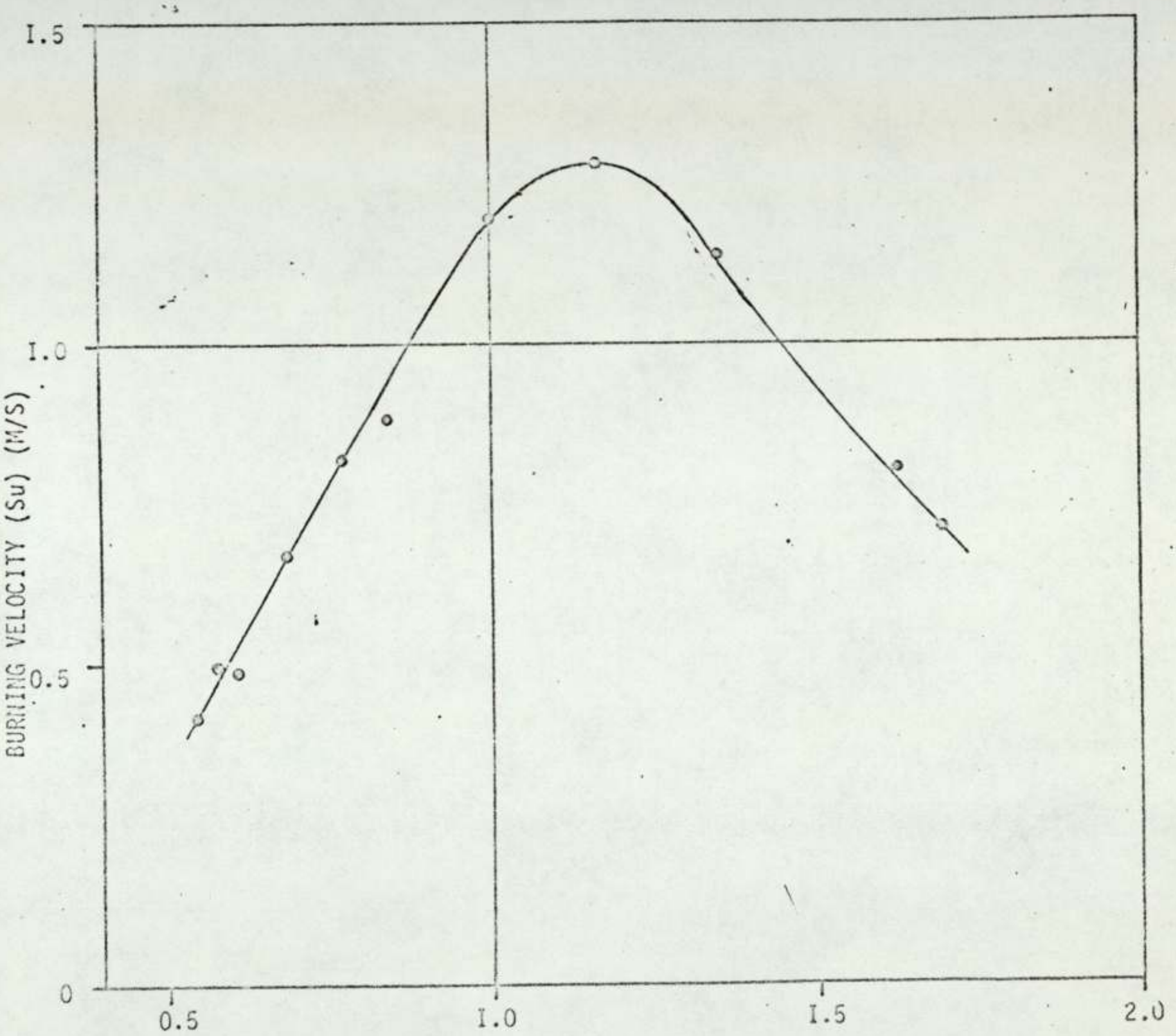
GRAPH NO. 23

GRAPHS NOS. 24-26

BURNING VELOCITY PLOTTED AGAINST FUEL CONCENTRATION USING PROPANE AND HYDROGEN AS THE FUEL GASES.

PRIMARY OXIDANT: 21% OXYGEN 79% ARGON

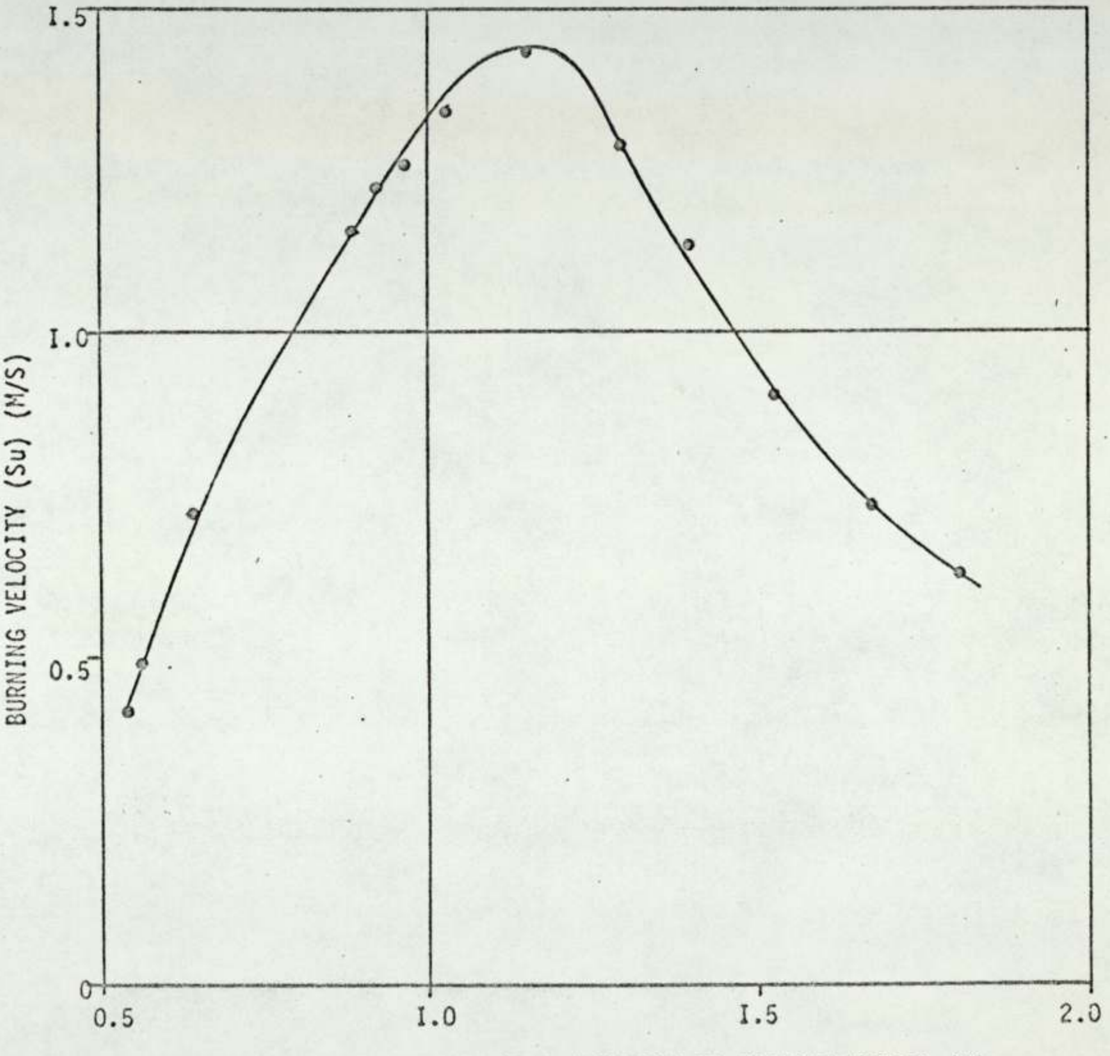
SECONDARY ATMOSPHERE: 21% OXYGEN 79% ARGON



FUEL CONCENTRATION EXPRESSED AS A FRACTION OF STOICHIOMETRIC (X)

FUEL GAS: I00% PROPANE

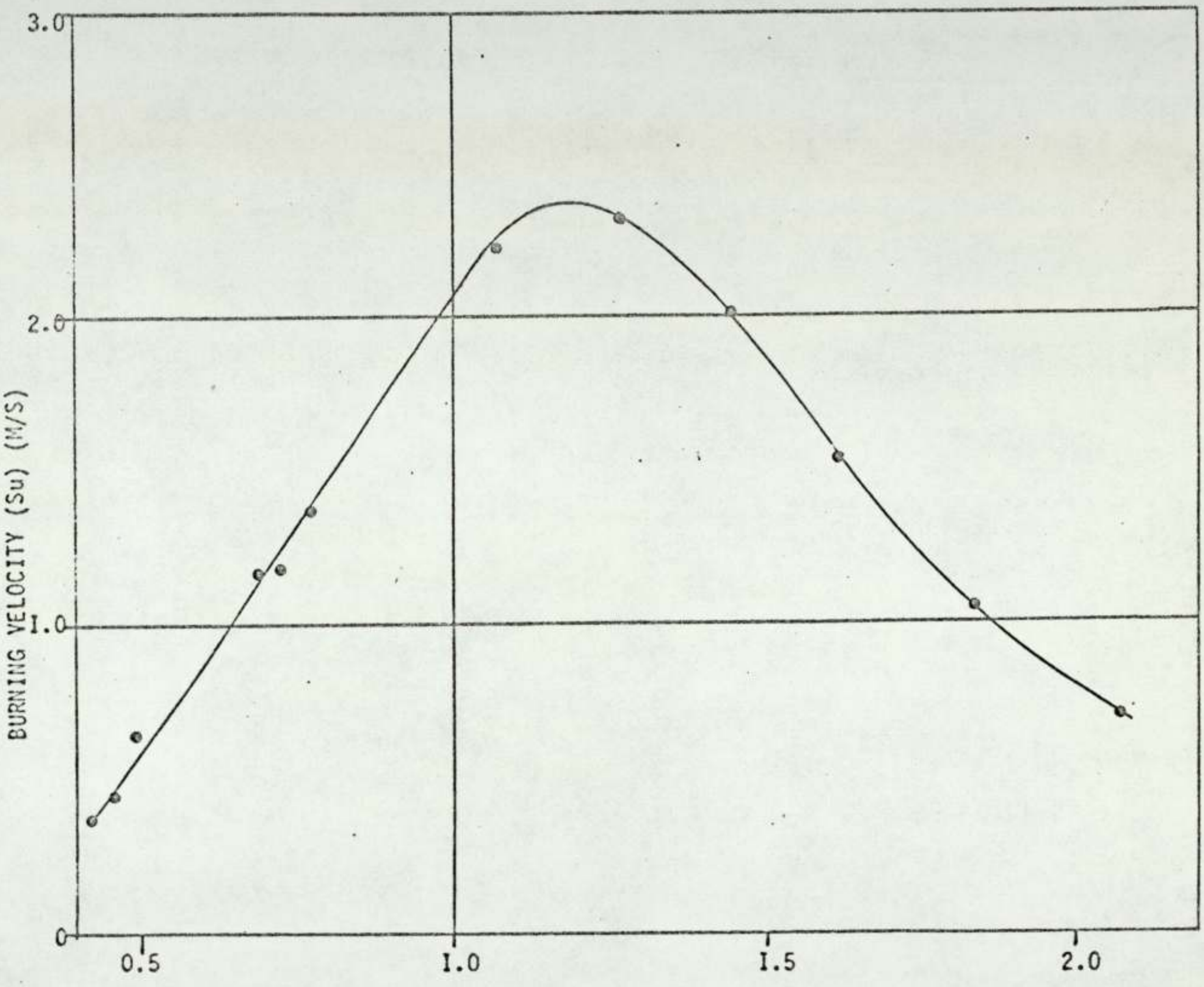
<u>GRAPH NO. 24</u>



FUEL CONCENTRATION EXPRESSED AS A FRACTION OF STOICHIOMETRIC (X)

FUEL GAS: 60% PROPANE 40% HYDROGEN

GRAPH NO. 25



FUEL CONCENTRATION EXPRESSED AS A FRACTION OF STOICHIOMETRIC (X)

FUEL GAS: 20% PROPANE 80% HYDROGEN

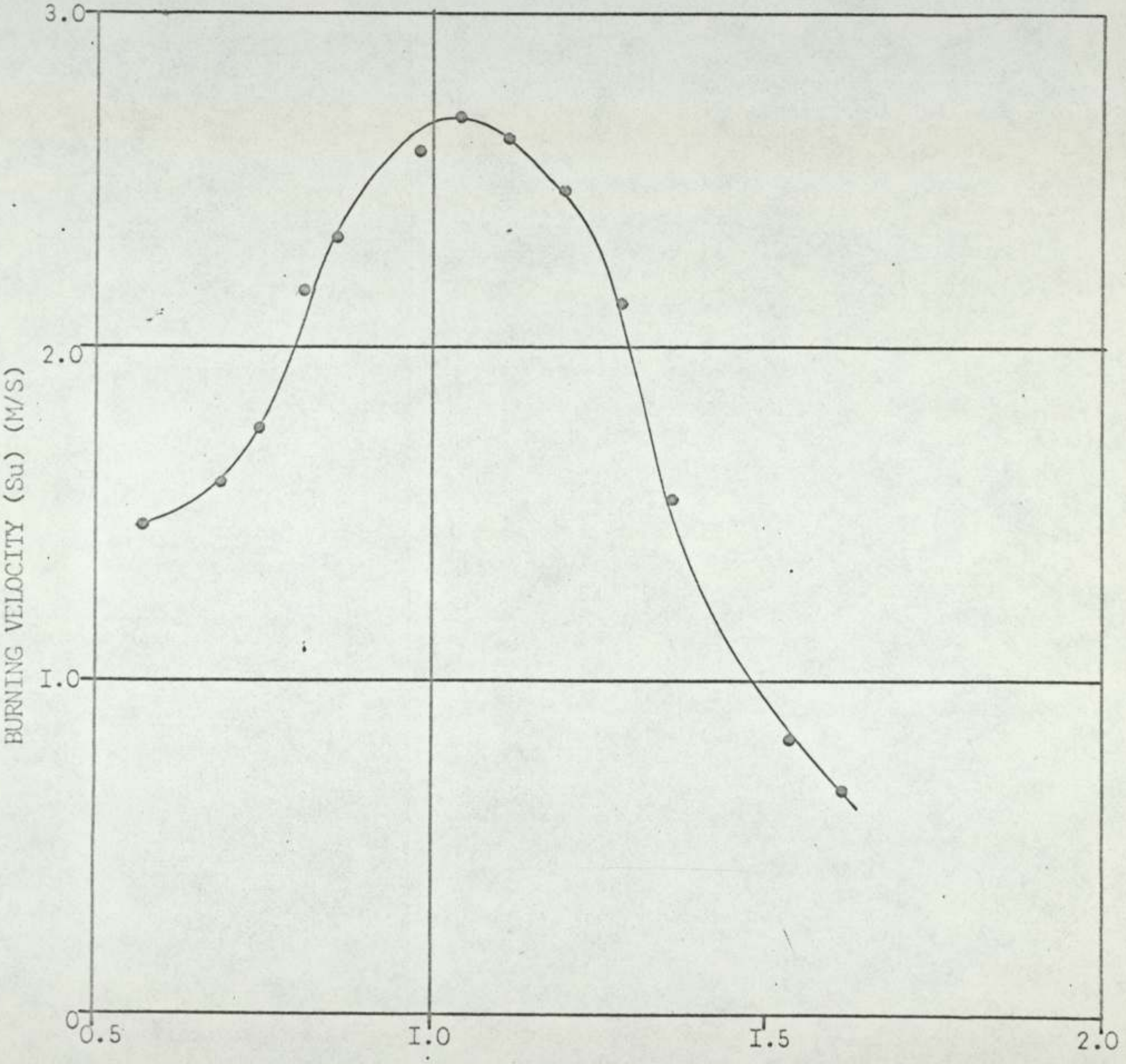
GRAPH NO. 26

GRAPHS NOS. 27-29

BURNING VELOCITY PLOTTED AGAINST FUEL CONCENTRATION USING PROPANE AND HYDROGEN AS THE FUEL GASES.

PRIMARY OXIDANT: 21% OXYGEN 79% HELIUM

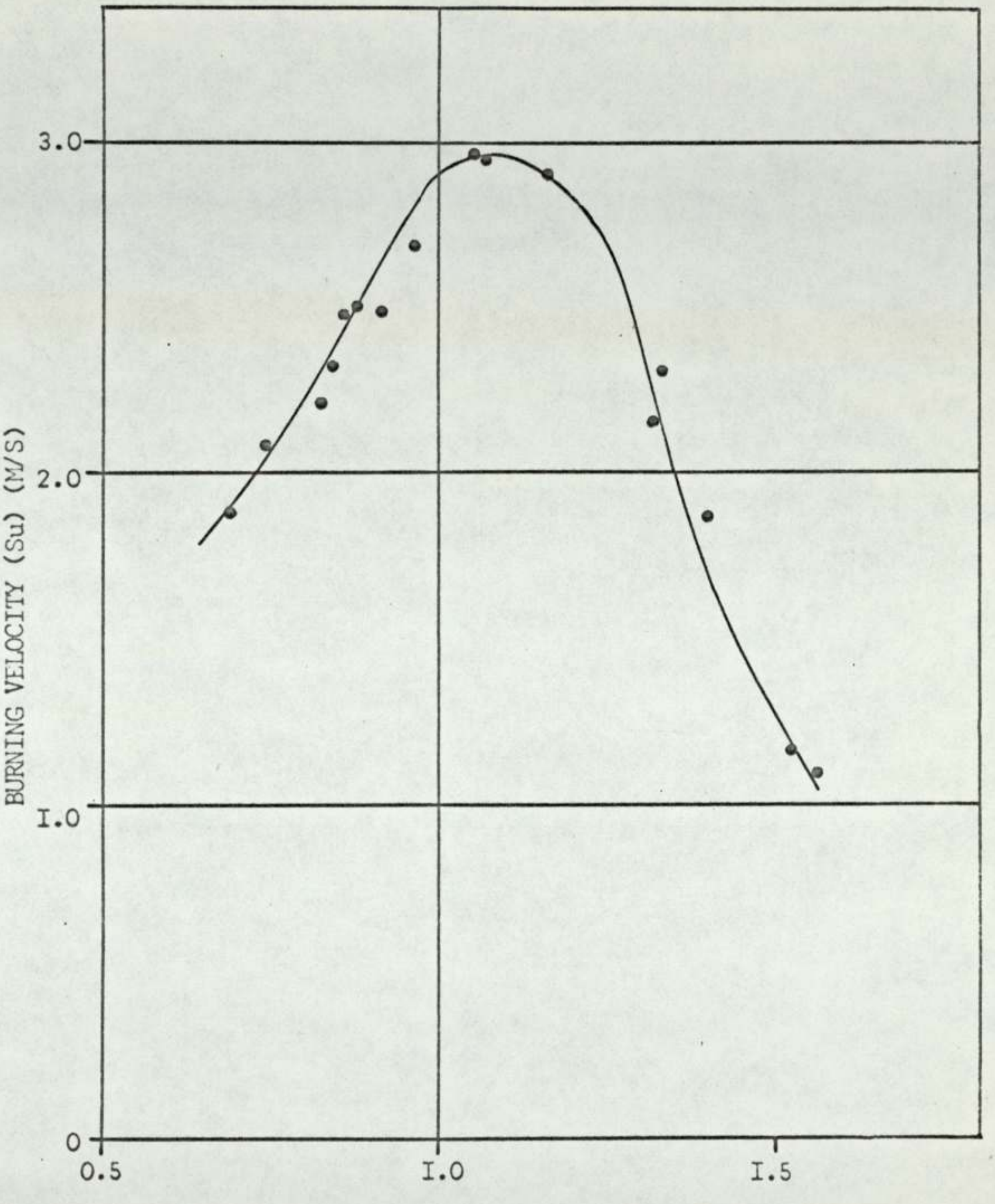
SECONDARY ATMOSPHERE: 21% OXYGEN 79% HYDROGEN



FUEL CONCENTRATION EXPRESSED AS A FRACTION OF STOICHIOMETRIC (X)

FUEL GAS: 100% PROPANE

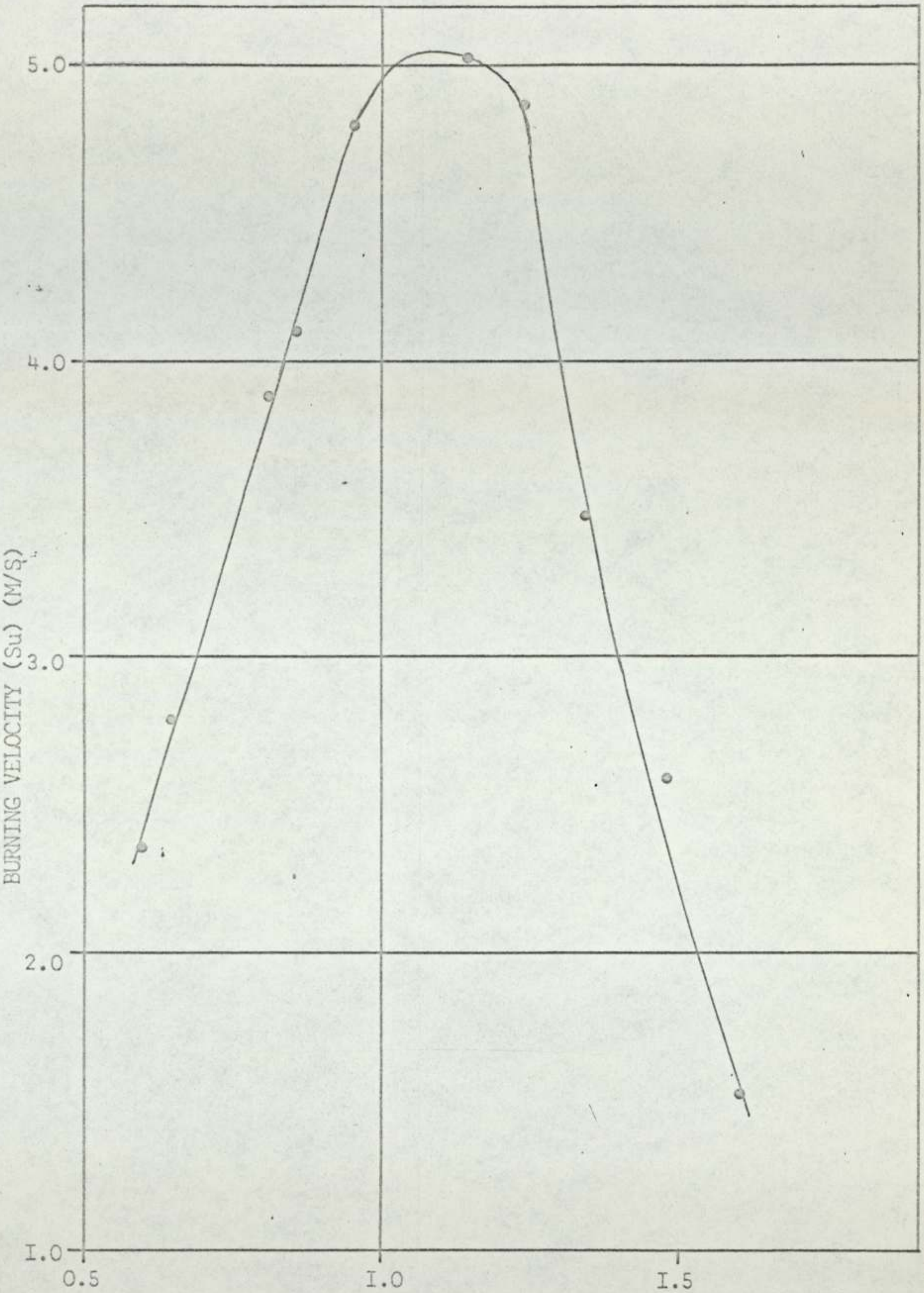
GRAPH NO. 27



FUEL CONCENTRATION EXPRESSED AS A FRACTION OF STOICHIOMETRIC (X)

FUEL GAS: 60% PROPANE 40% HYDROGEN

<u>GRAPH NO. 28</u>

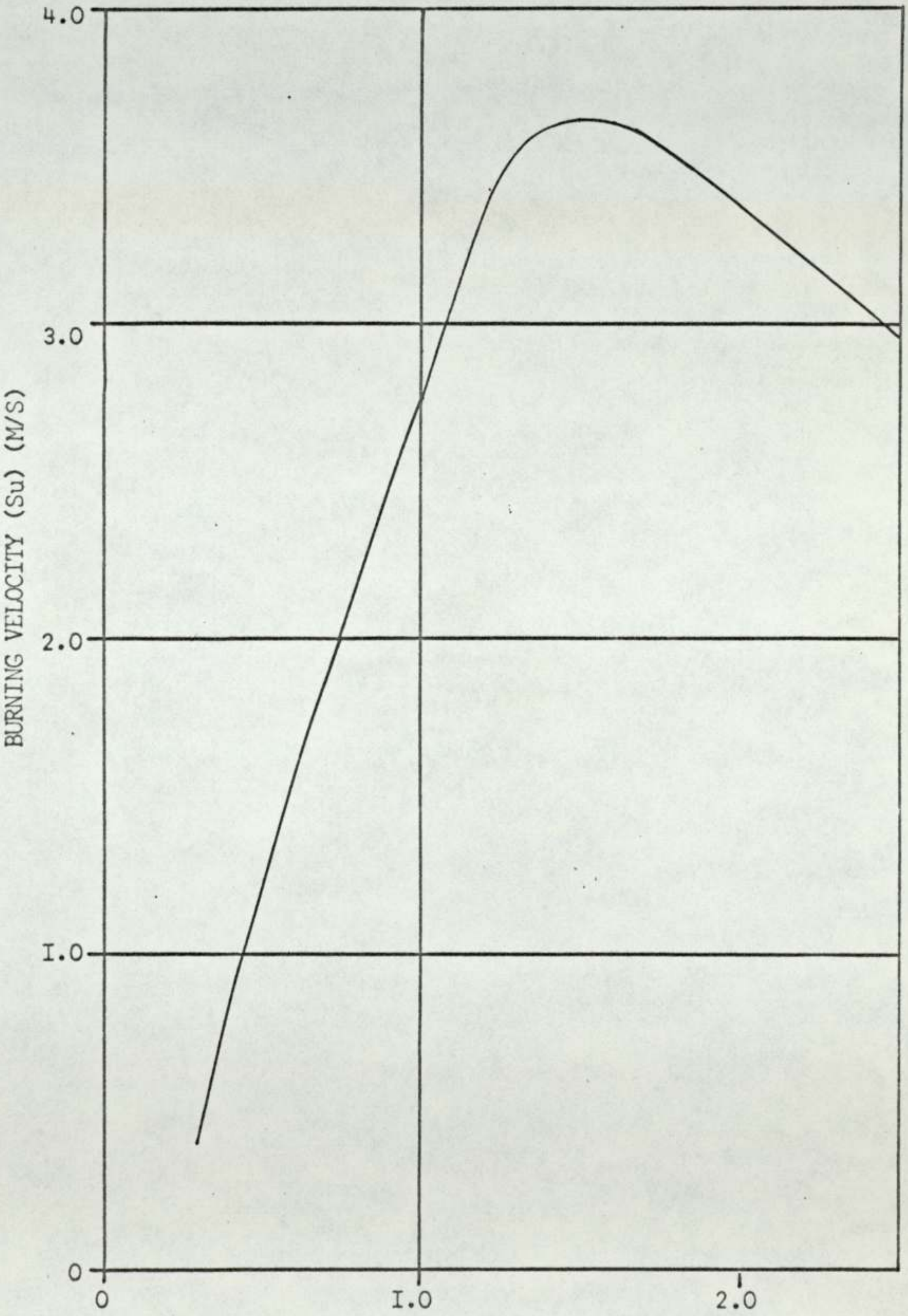


FUEL CONCENTRATION EXPRESSED AS A FRACTION OF STOICHIOMETRIC (X)

FUEL GAS: 20% PROPANE 80% HYDROGEN

GRAPH NO. 29

BURNING VELOCITY PLOTTED AGAINST FUEL CONCENTRATION FOR THE HYDROGEN-AIR SYSTEM. DATA OF GUENTHER & JANISCH, AND EDMONDSON & HEAP PRESENTED BY DIXON-LEWIS ET AL IN I.G.E. RESEARCH COMMUNICATION 86I.

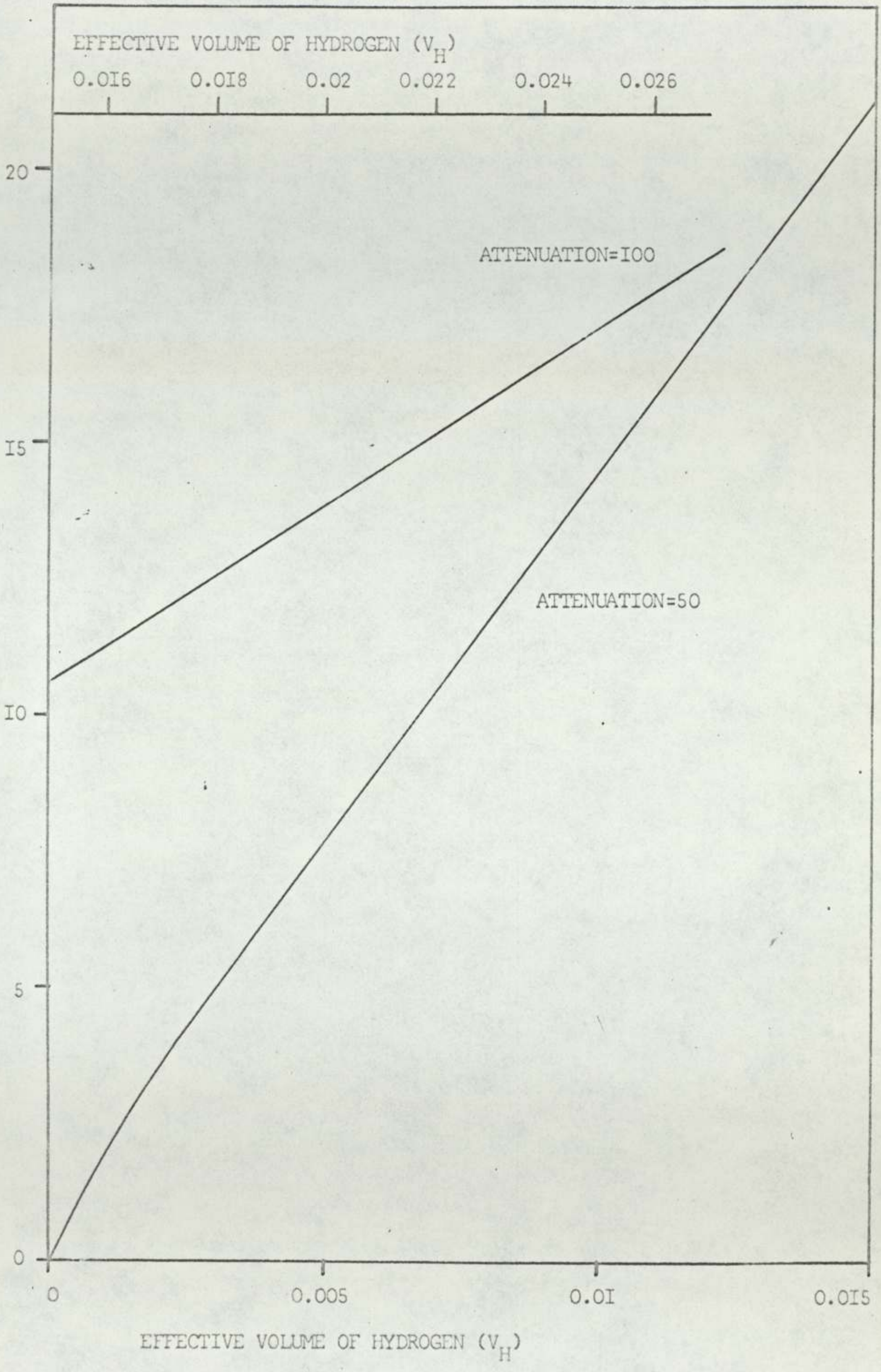


FUEL CONCENTRATION EXPRESSED AS A FACTION OF STOICHIOMETRIC (X)

FUEL GAS: I00% HYDROGEN

GRAPH NO. 30

CHROMATOGRAPH CALIBRATION GRAPH



$$V_H = \frac{(\text{SAMPLE PRESSURE}) \times (\text{MOLE FRACTION OF HYDROGEN IN SAMPLE})}{(\text{ATMOSPHERIC PRESSURE})}$$

GRAPH NO. 31

GRAPHS NOS. 32-43

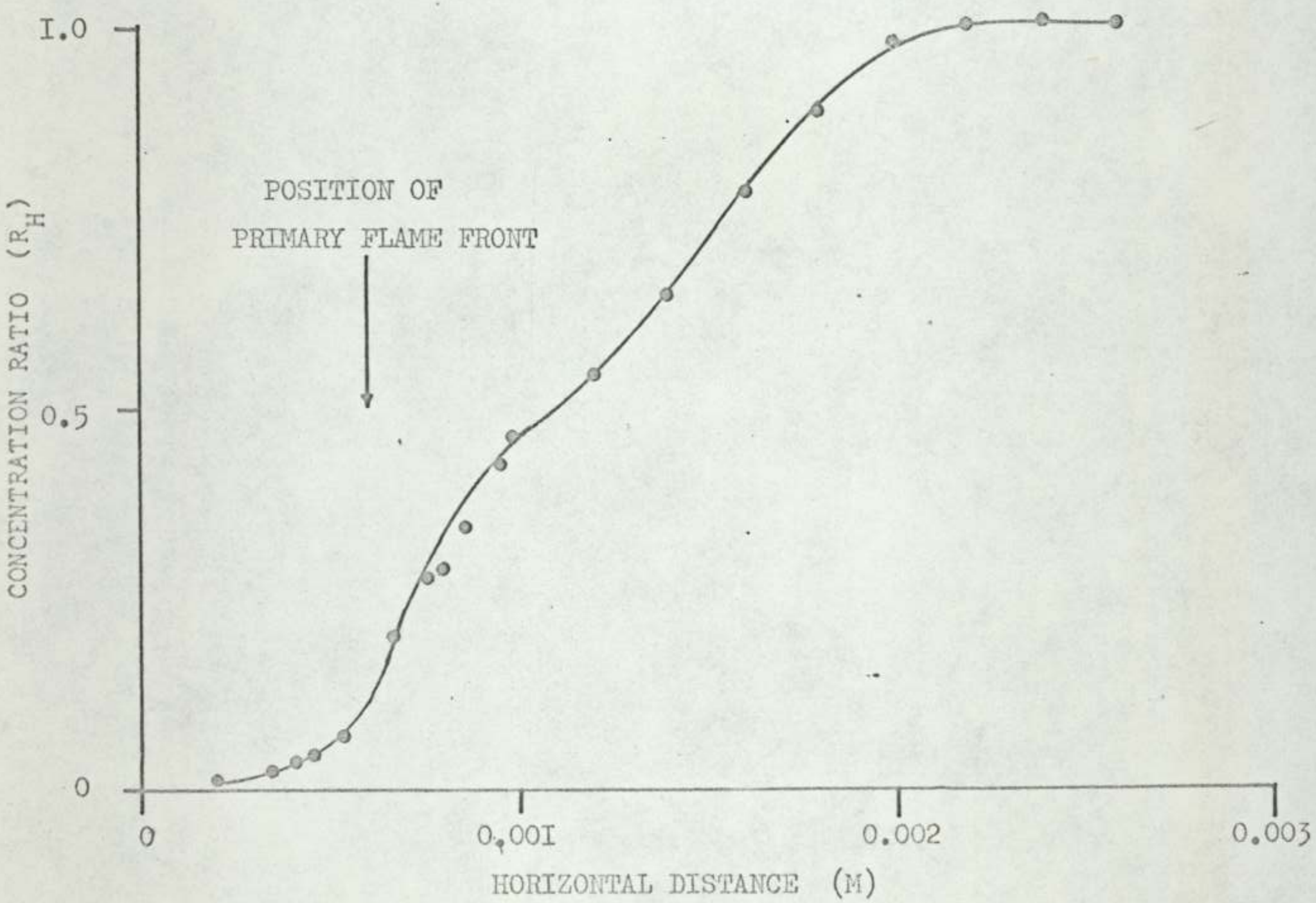
HYDROGEN CONCENTRATION PROFILES IN STABILISING REGION OF LAMINAR BURNER FLAMES.

FUEL GAS: 40% PROPANE 60% HYDROGEN

PRIMARY OXIDANT: AIR

SECONDARY ATMOSPHERE: AIR

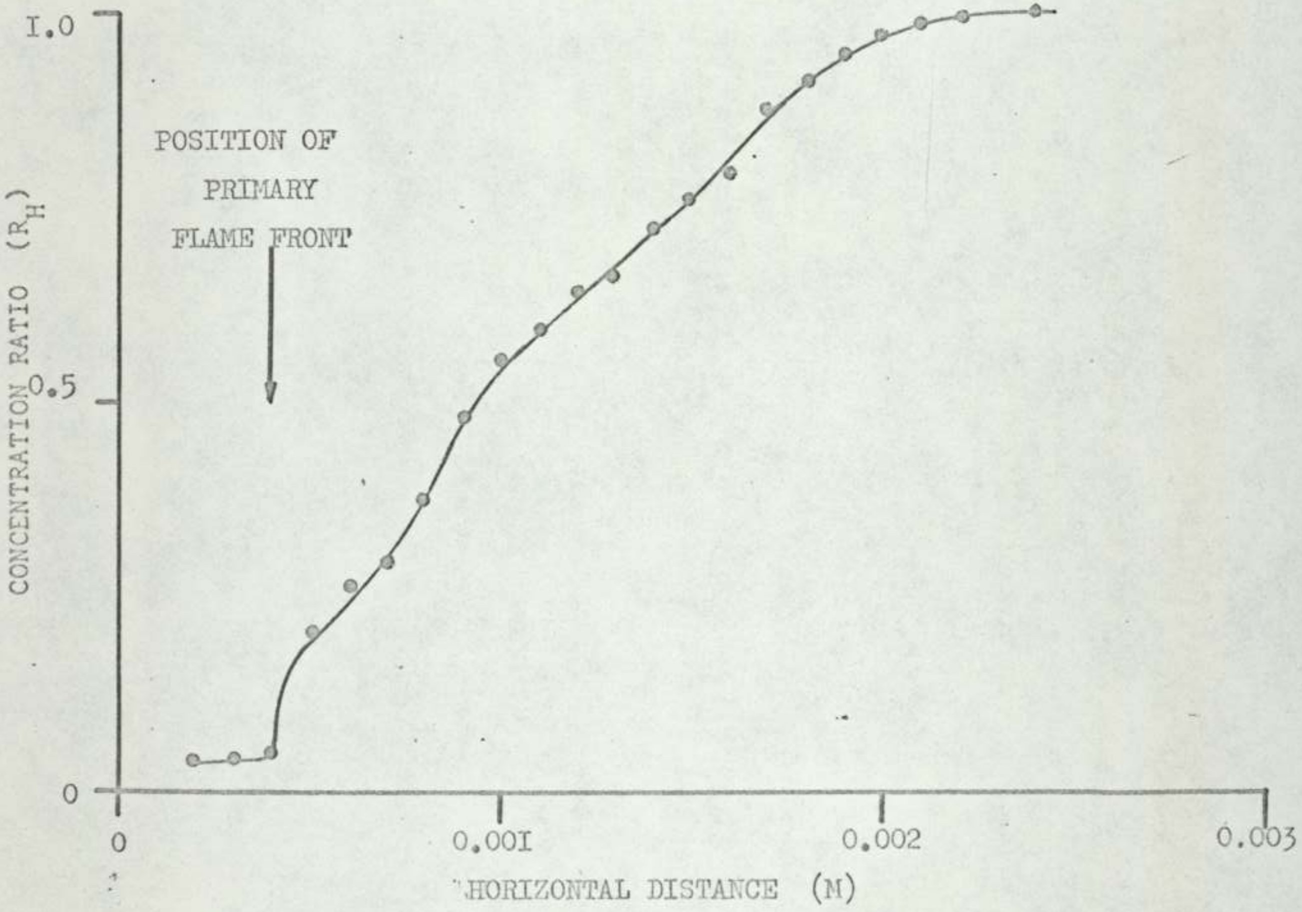
FUEL CONCENTRATION: GRAPHS NOS. 32-35 : 0.8 STOICHIOMETRIC
GRAPHS NOS. 36-39 : I.0 STOICHIOMETRIC
GRAPHS NOS. 40-43 : I.2 STOICHIOMETRIC



CONCENTRATION PROFILE OF HYDROGEN IN 0.8 STOICHIOMETRIC FLAME.

HEIGHT ABOVE BURNER:- 0.0006M.
FUEL GAS MIXTURE:- 60% H_2 , 40% C_3H_8

GRAPH NO.32

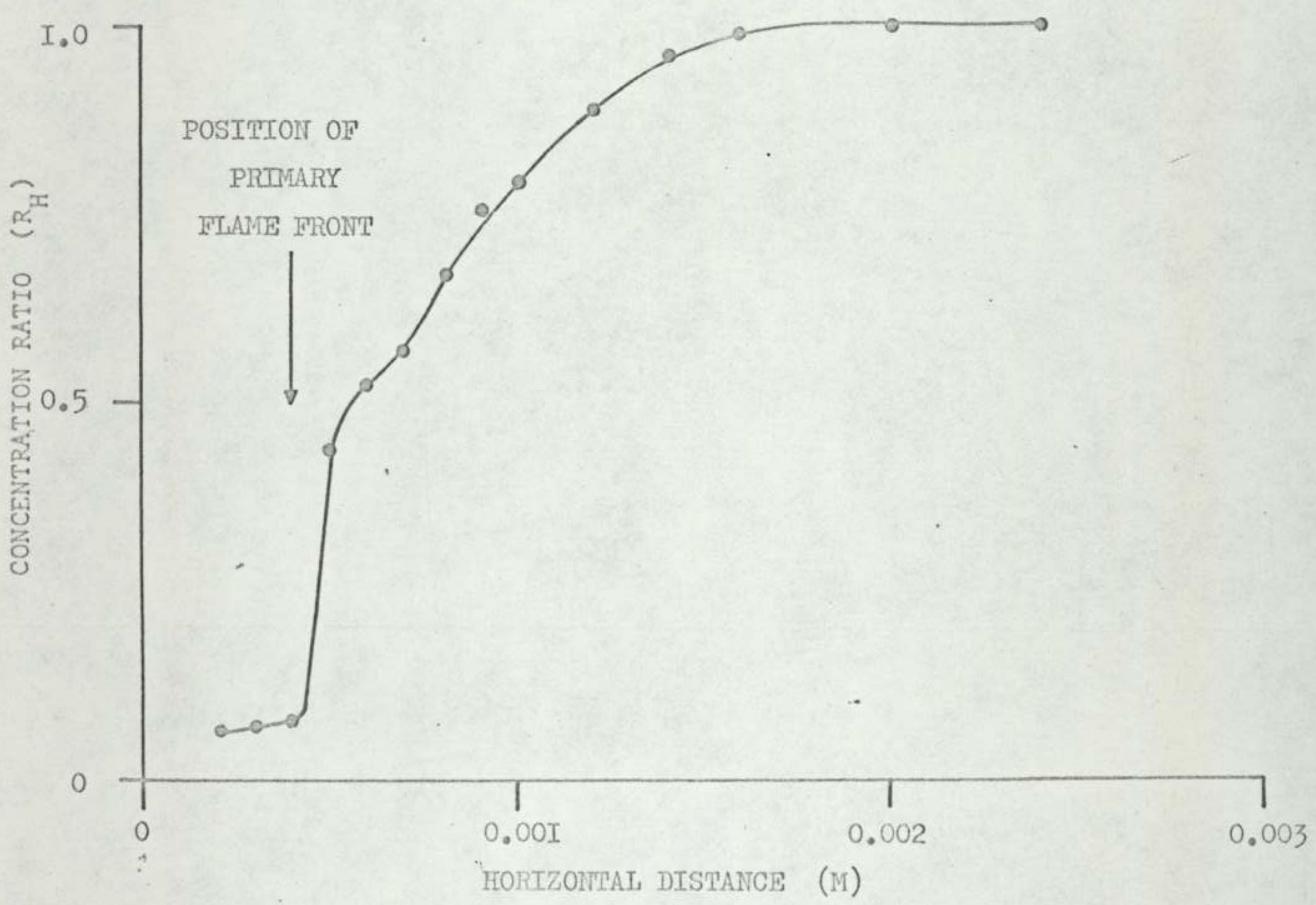


CONCENTRATION PROFILE OF HYDROGEN IN 0.8 STOICHIOMETRIC FLAME.

HEIGHT ABOVE BURNER:- 0.0009M.

FUEL GAS MIXTURE:- 60% H_2 , 40% C_3H_8

GRAPH NO. 33

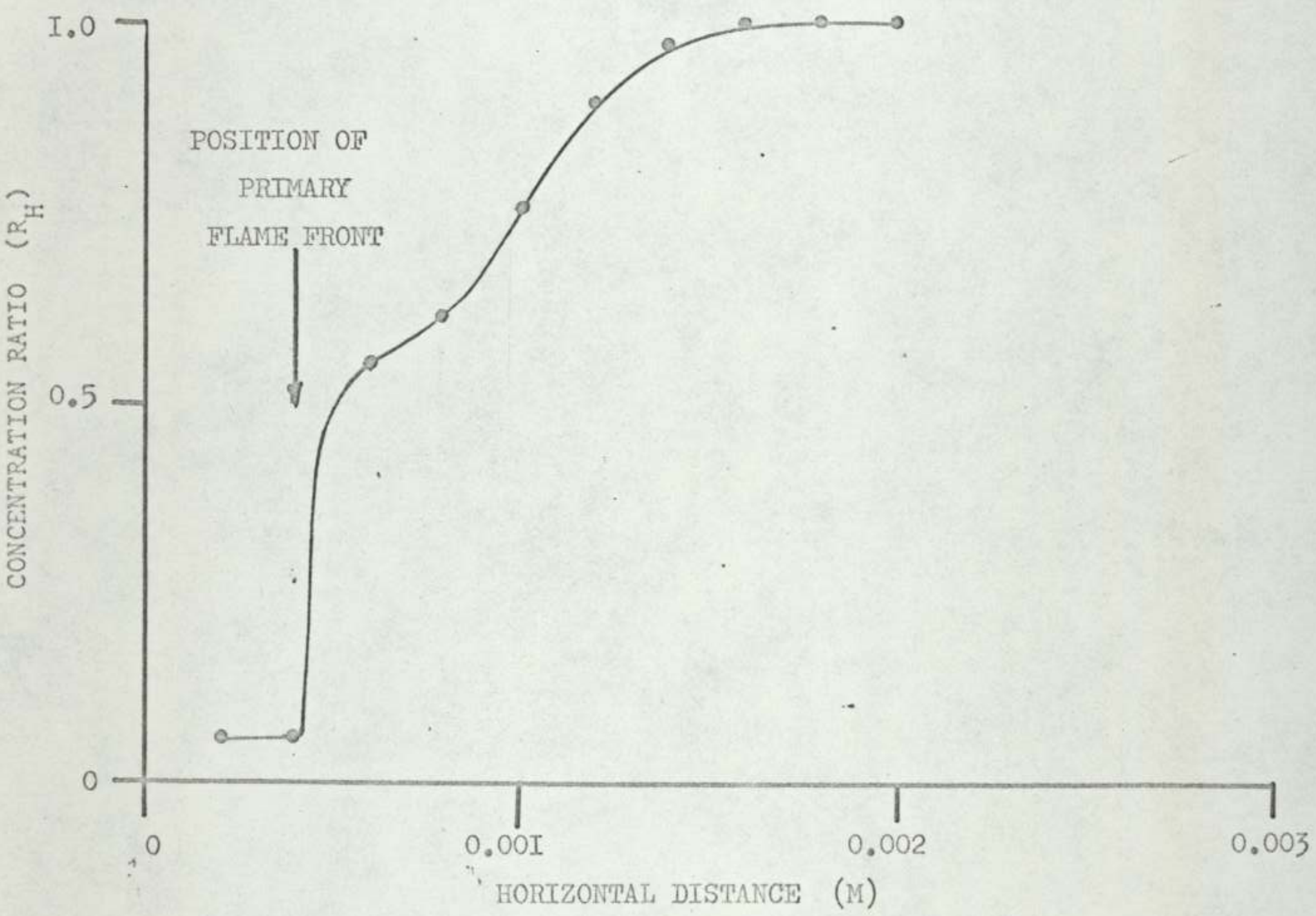


CONCENTRATION PROFILE OF HYDROGEN IN 0.8 STOICHIOMETRIC FLAME.

HEIGHT ABOVE BURNER:- 0.0015M.

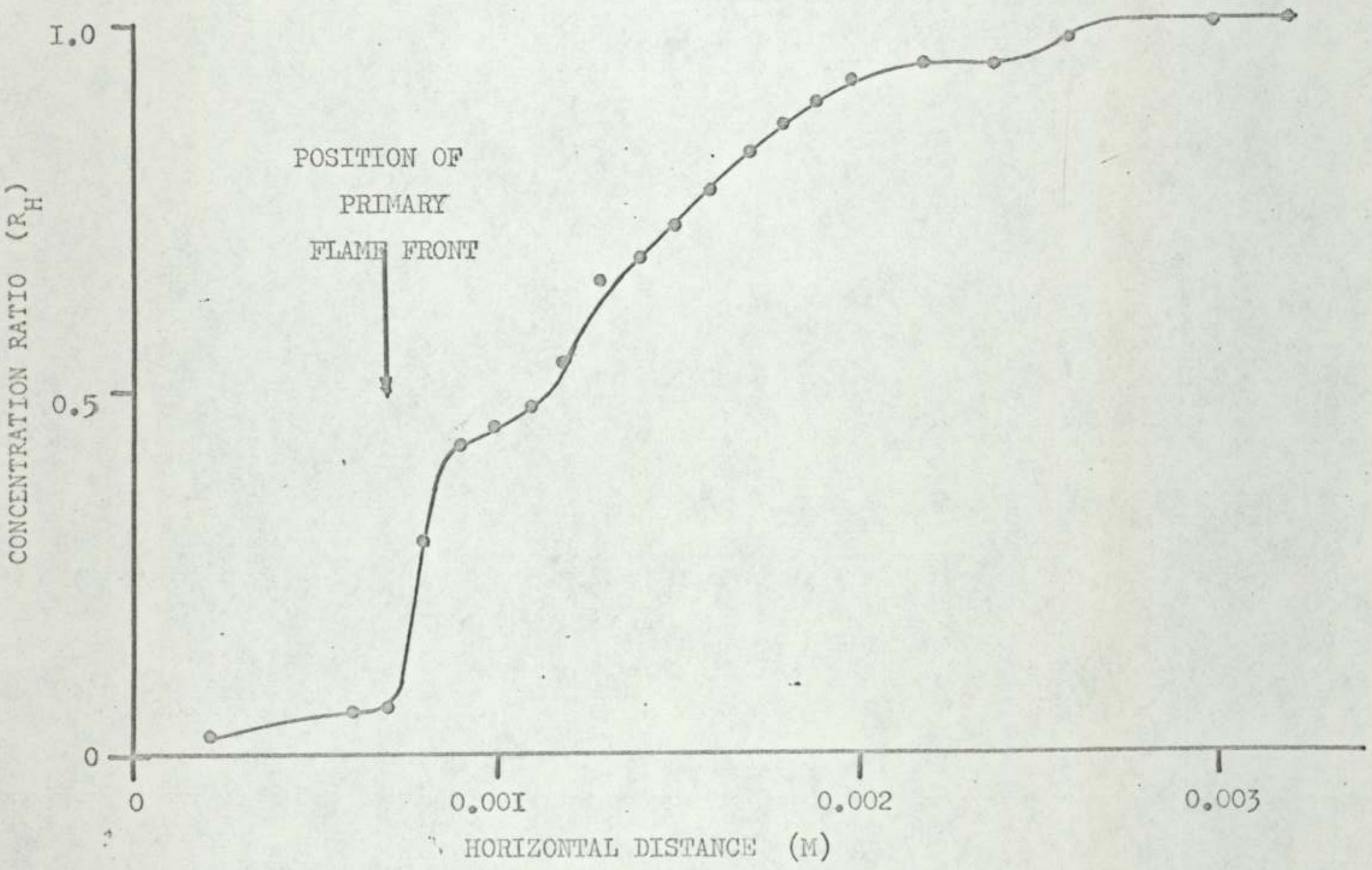
FUEL GAS MIXTURE:- 60% H_2 , 40% C_3H_8

GRAPH NO. 34



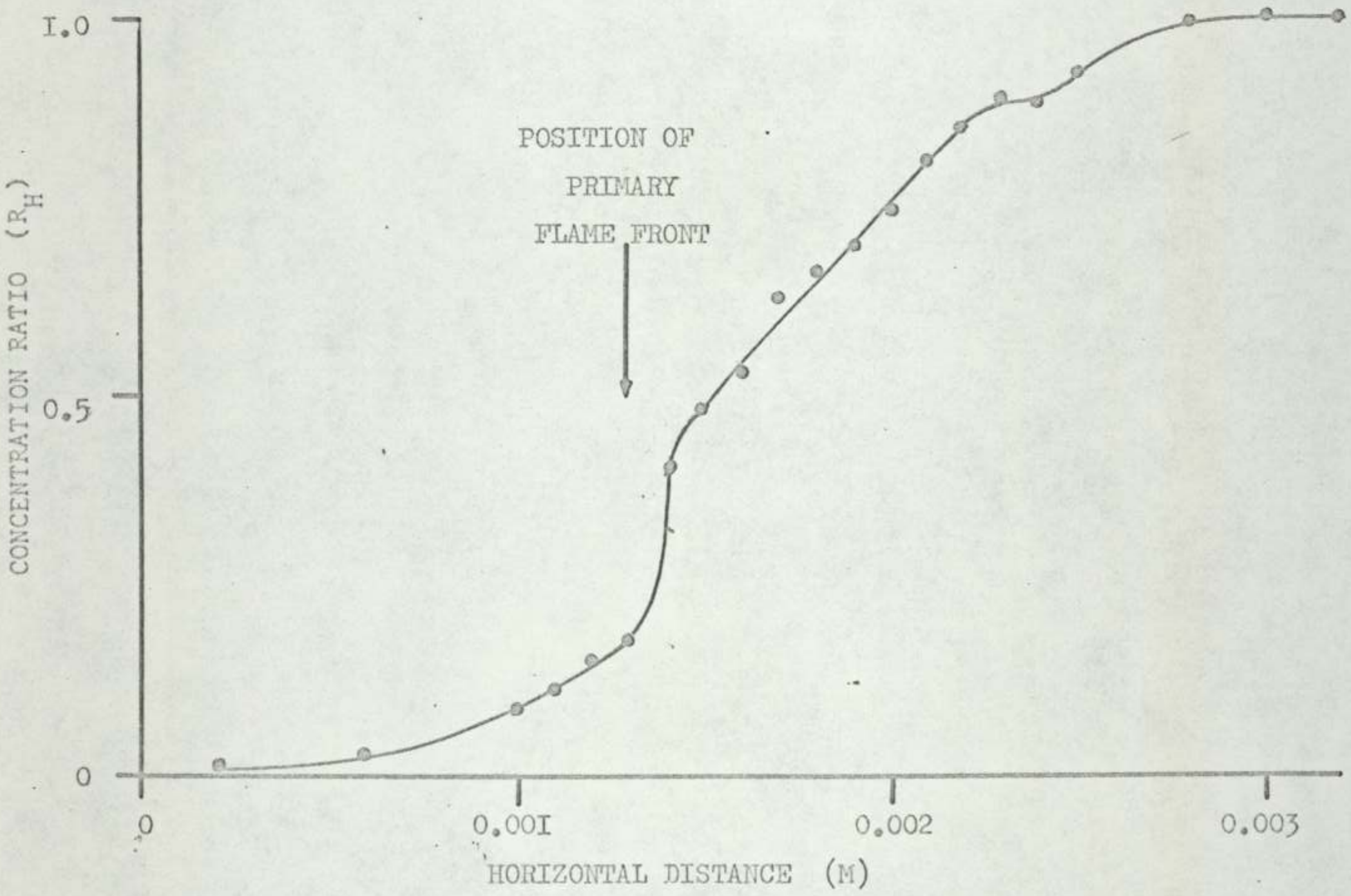
CONCENTRATION PROFILE OF HYDROGEN IN 0.8 STOICHIOMETRIC FLAME.
HEIGHT ABOVE BURNER:- 0.002IM.
FUEL GAS MIXTURE:- 60% H_2 , 40% C_3H_8

GRAPH NO. 35



CONCENTRATION PROFILE OF HYDROGEN IN I.0 STOICHIOMETRIC FLAME.
HEIGHT ABOVE BURNER:- 0.0006M.
FUEL GAS MIXTURE:- 60% H_2 , 40% C_3H_8

GRAPH NO. 36

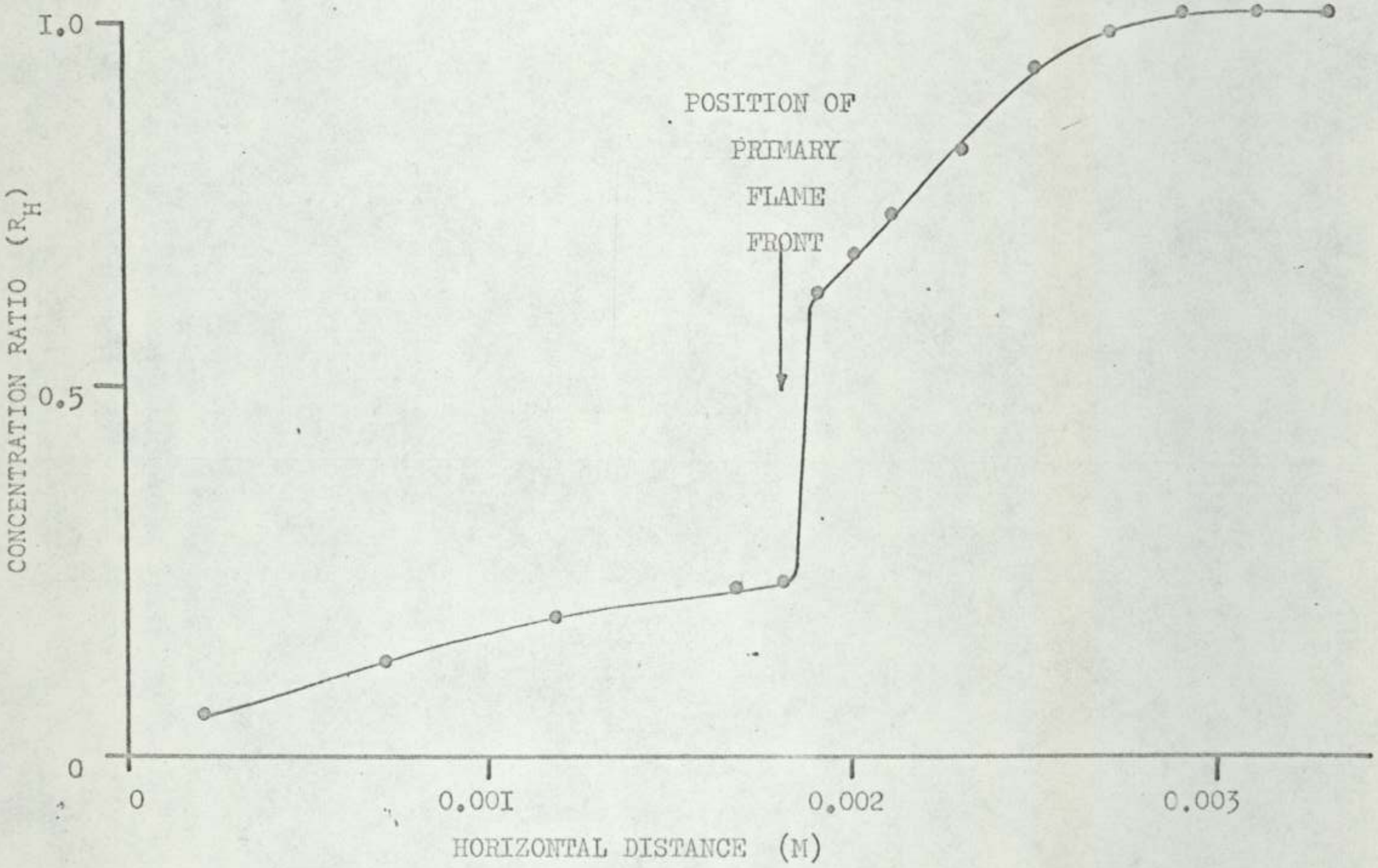


CONCENTRATION PROFILE OF HYDROGEN IN I.0 STOICHIOMETRIC FLAME.

HEIGHT ABOVE BURNER:- 0.0009M.

FUEL GAS MIXTURE:- 60% H_2 , 40% C_3H_8

GRAPH NO. 37

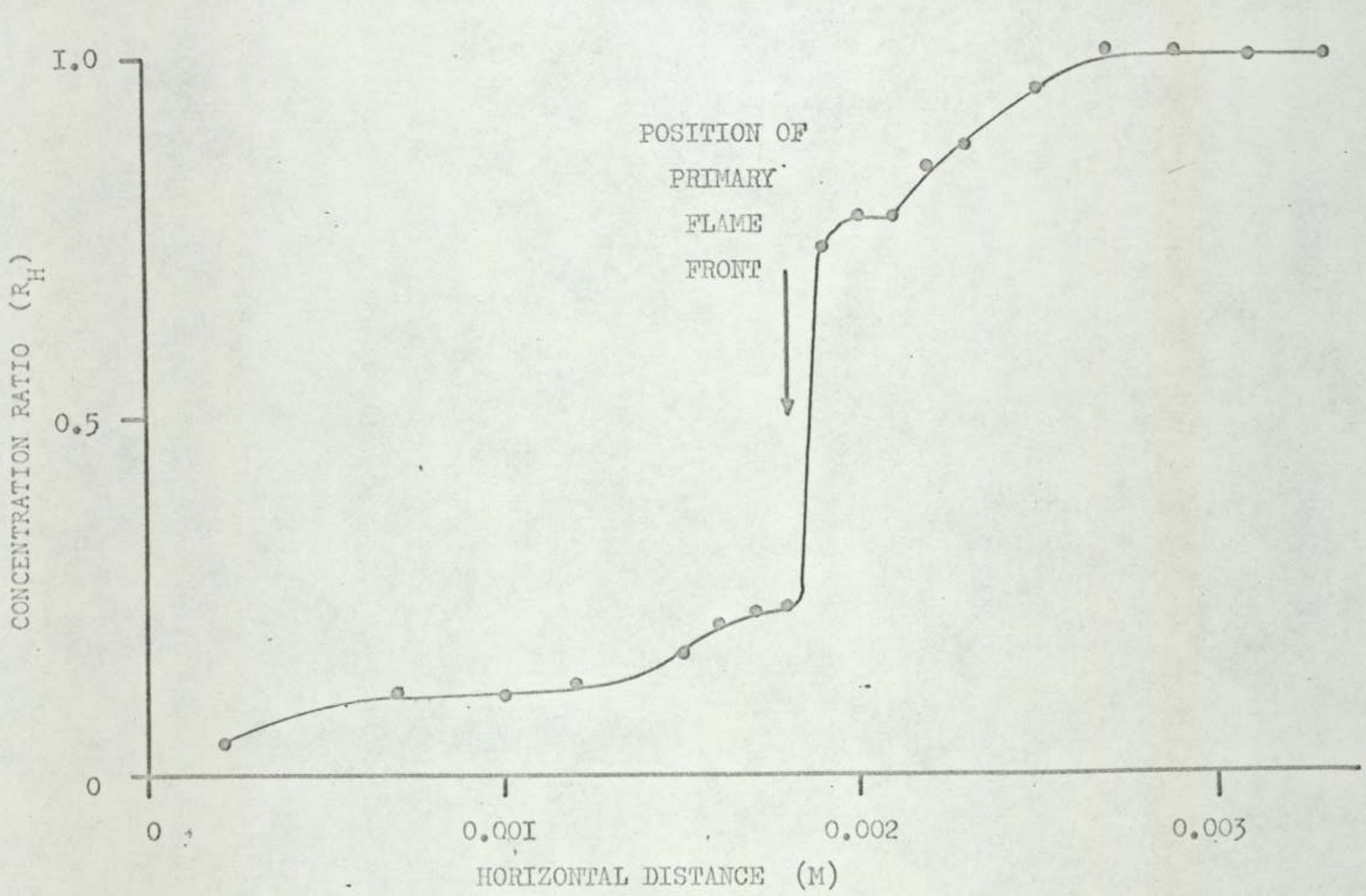


CONCENTRATION PROFILE OF HYDROGEN IN I.0 STOICHIOMETRIC FLAME.

HEIGHT ABOVE BURNER:- 0.00I2M.

FUEL GAS MIXTURE:- 60% H_2 , 40% C_3H_8

GRAPH NO.38

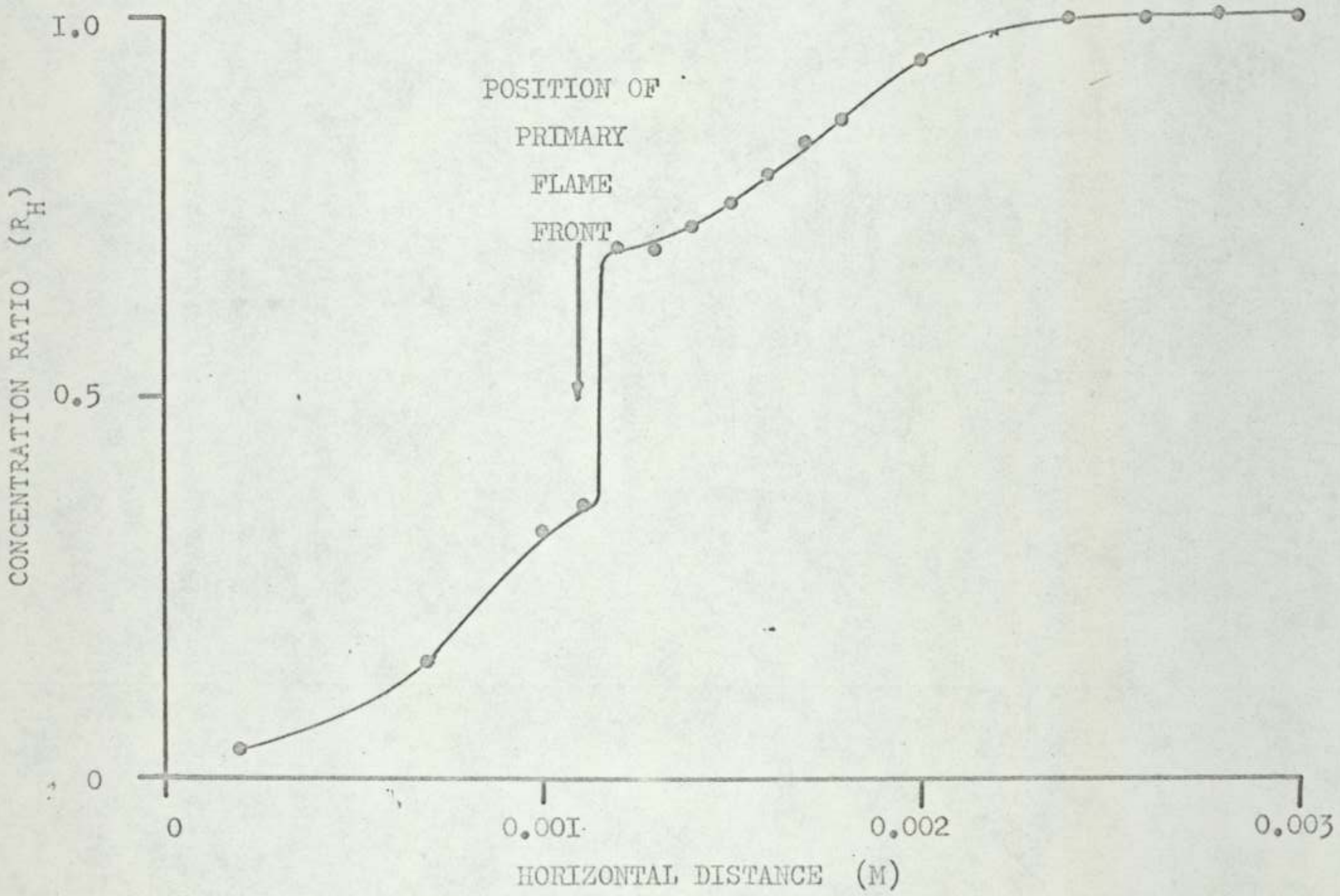


CONCENTRATION PROFILE OF HYDROGEN IN I.0 STOICHIOMETRIC FLAME.

HEIGHT ABOVE BURNER:- 0.00I5M.

FUEL GAS MIXTURE:- 60% H_2 , 40% C_3H_8

GRAPH NO. 39

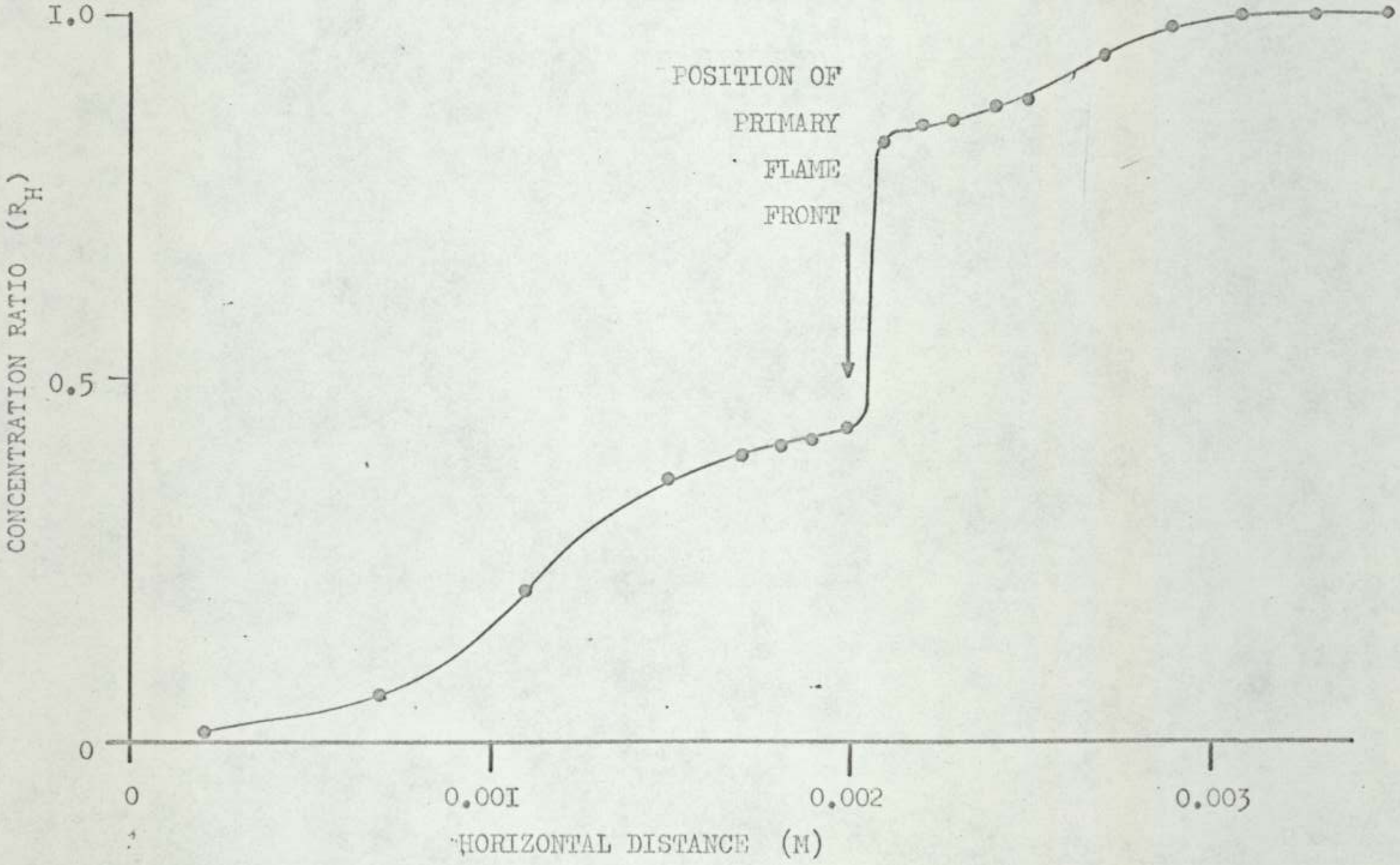


CONCENTRATION PROFILE OF HYDROGEN IN I.2 STOICHIOMETRIC FLAME.

HEIGHT ABOVE BURNER:- 0.0006M.

FUEL GAS MIXTURE:- 60% H_2 , 40% C_3H_8

GRAPH NO. 40

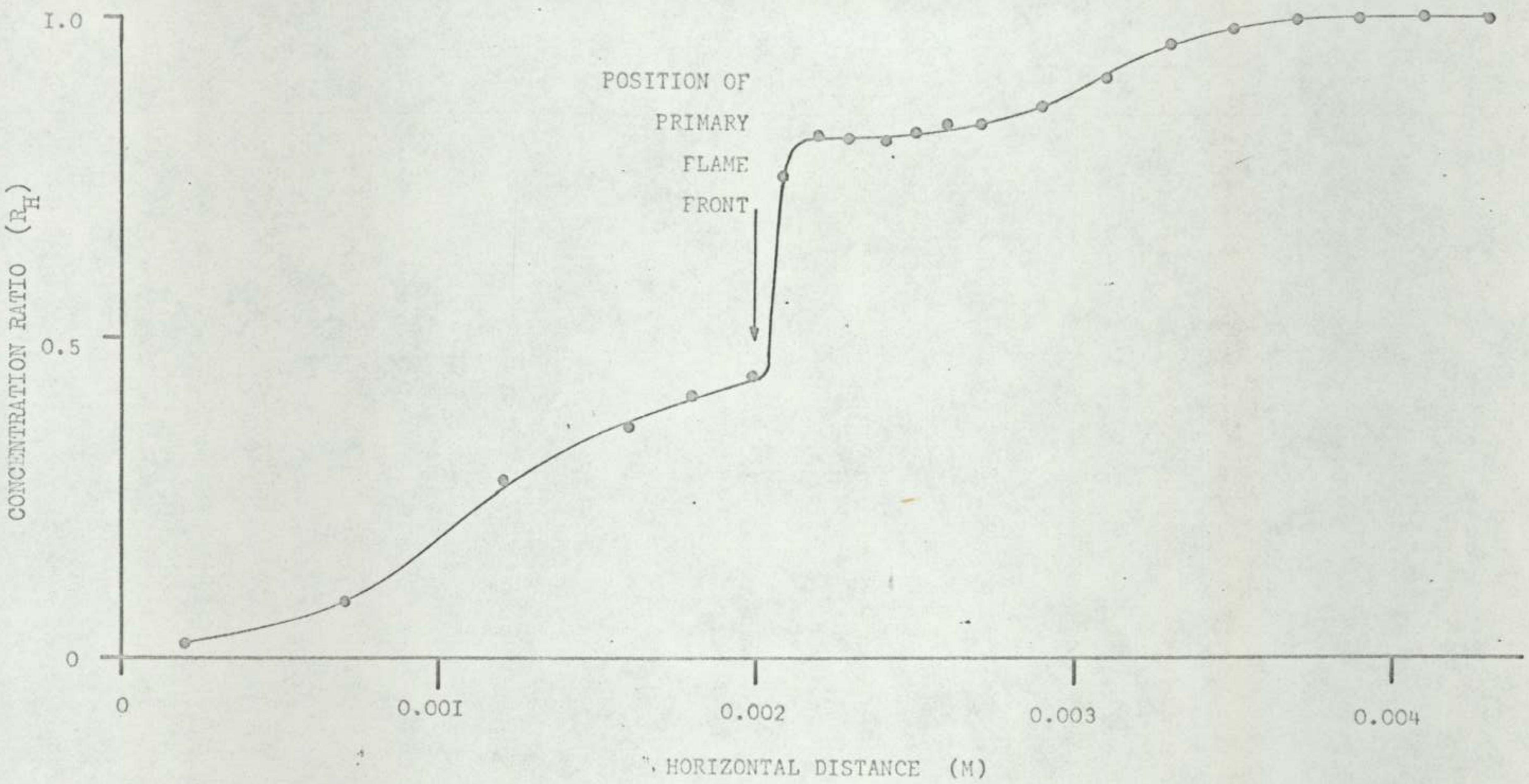


CONCENTRATION PROFILE OF HYDROGEN IN I.2 STOICHIOMETRIC FLAME.

HEIGHT ABOVE BURNER :- 0.0009M.

FUEL GAS MIXTURE:- 60% H_2 , 40% C_3H_8

GRAPH NO. 4I

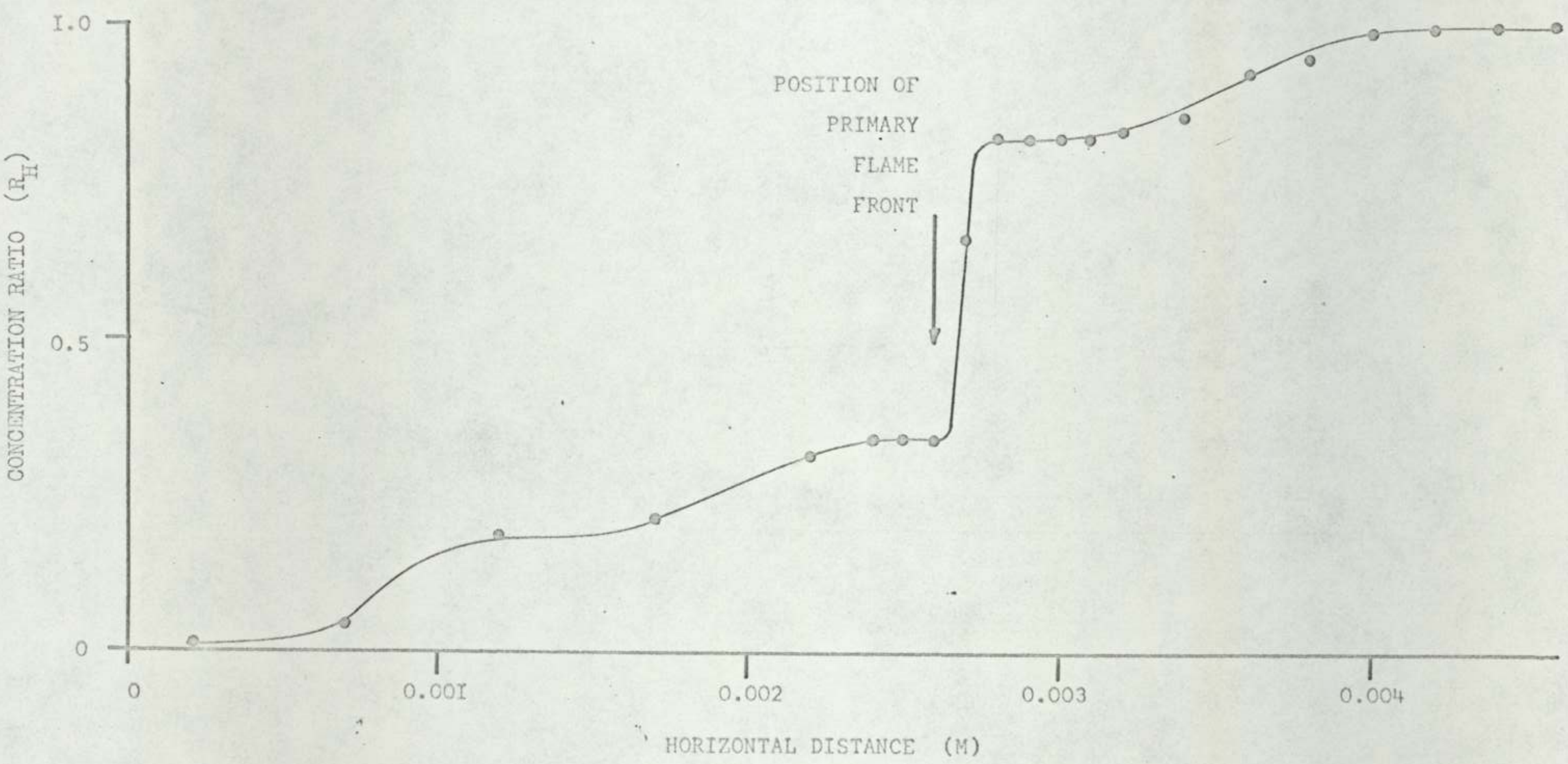


CONCENTRATION PROFILE OF HYDROGEN IN I.2 STOICHIOMETRIC FLAME.

HEIGHT ABOVE BURNER:- 0.00I2M.

FUEL GAS MIXTURE:- 60% H_2 , 40% C_3H_8

GRAPH NO. 42



CONCENTRATION PROFILE OF HYDROGEN IN I.2 STOICHIOMETRIC FLAME

HEIGHT ABOVE BURNER.- 0.00I5M.

FUEL GAS MIXTURE:- 60% H_2 , 40% C_3H_8

GRAPH NO. 43

GRAPHS NOS. 44-47

ETHYLENE CONCENTRATION PROFILES IN STABILISING REGION OF LAMINAR BURNER FLAME.

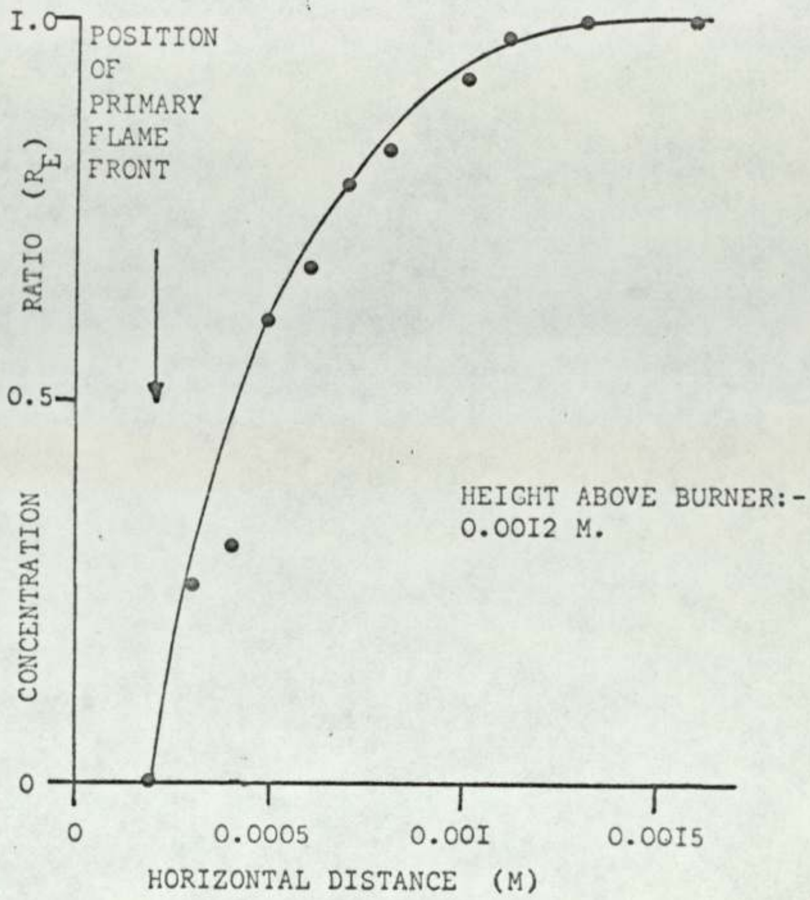
FUEL GAS: ETHYLENE

PRIMARY OXIDANT: AIR

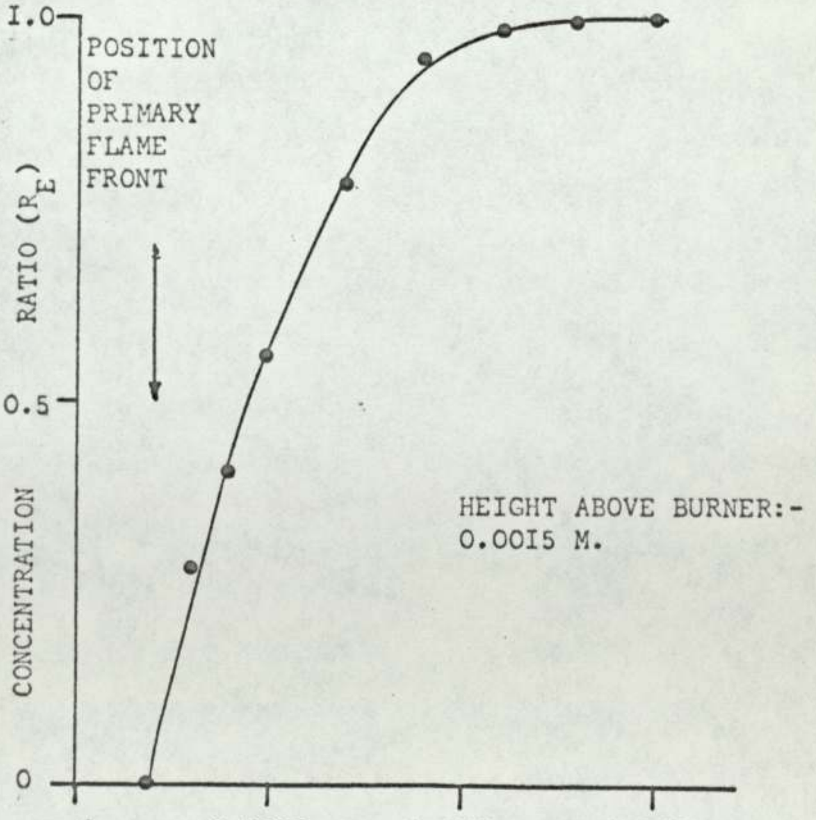
SECONDARY ATMOSPHERE: AIR

FUEL CONCENTRATION: I.O STOICHIOMETRIC

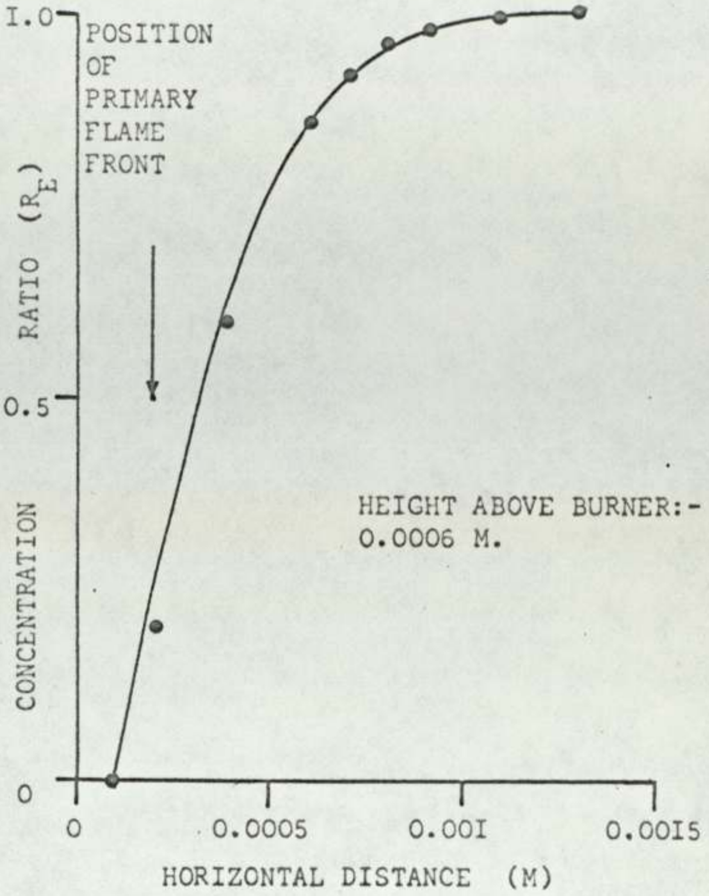
GRAPH NO. 44



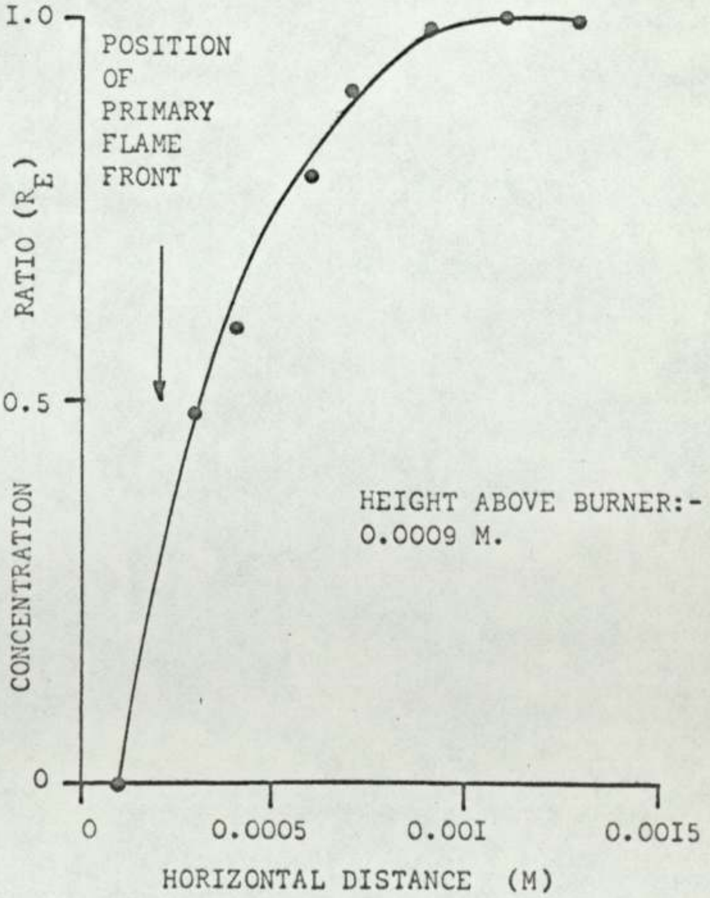
GRAPH NO. 45



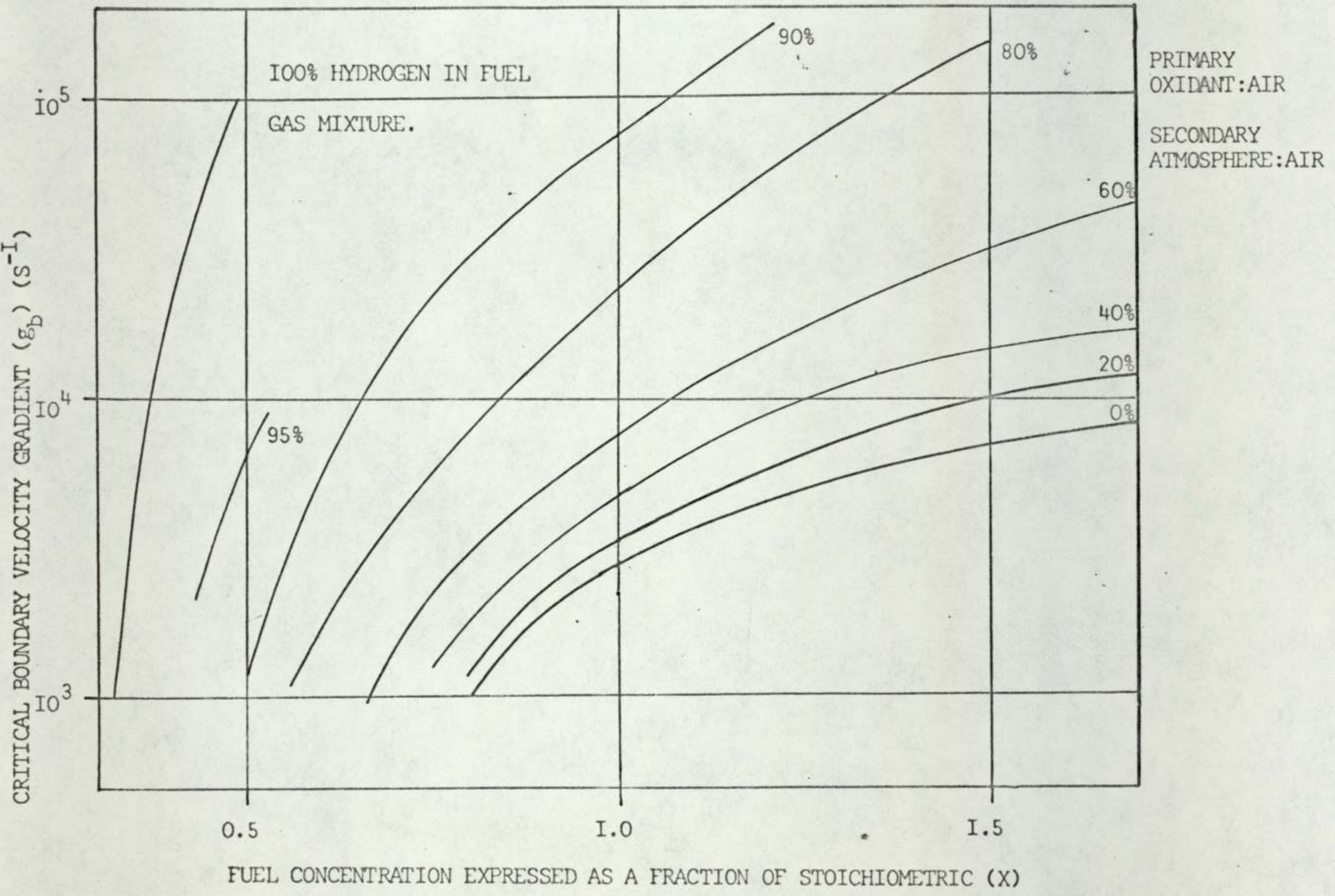
GRAPH NO. 46



GRAPH NO. 47

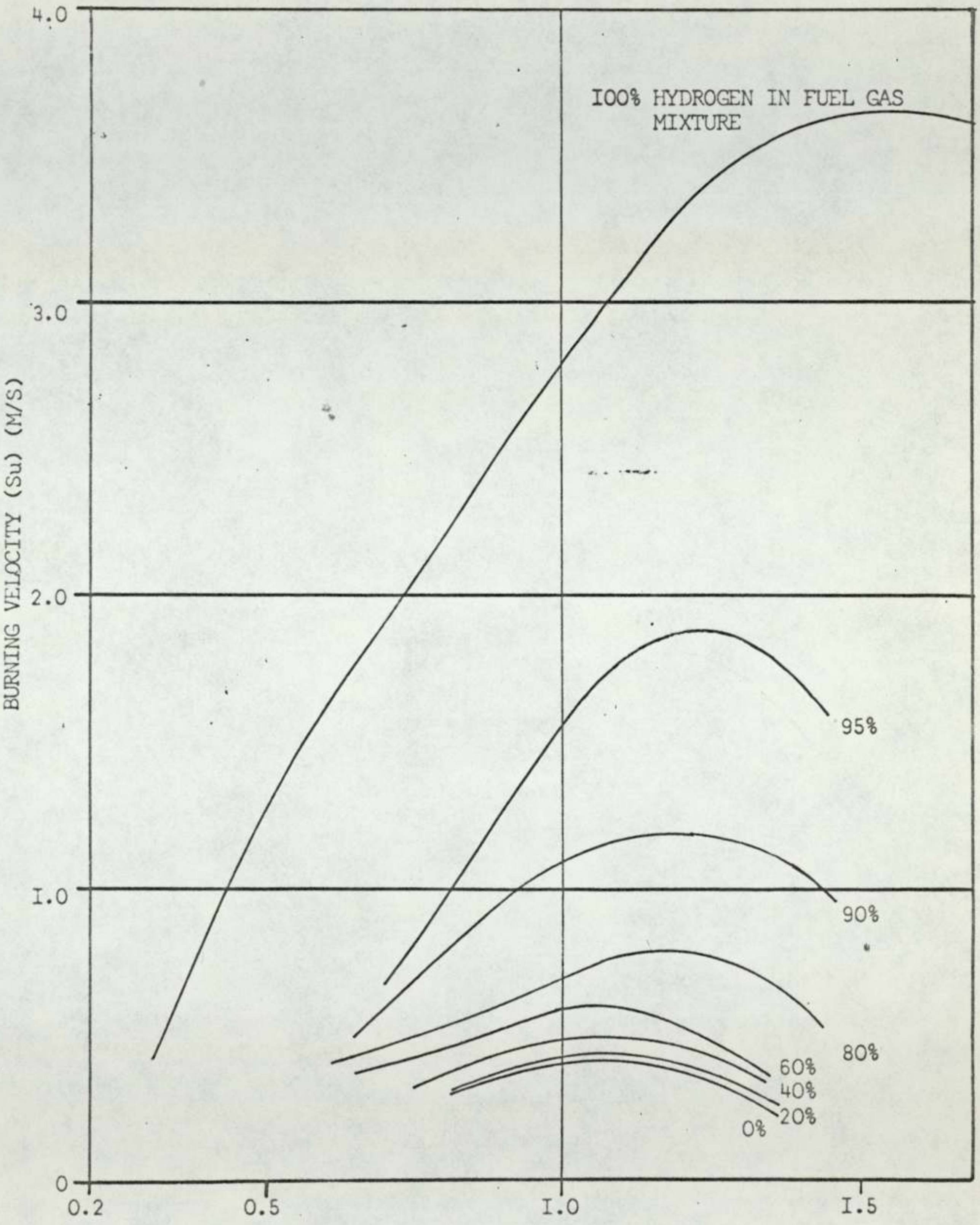


CRITICAL BOUNDARY VELOCITY GRADIENT PLOTTED AGAINST FUEL CONCENTRATION FOR HYDROGEN-PROPANE-AIR MIXTURES



GRAPH NO. 48

BURNING VELOCITY DATA PLOTTED AGAINST FUEL CONCENTRATION FOR HYDROGEN-PROPANE-AIR MIXTURES. DATA FOR 100% HYDROGEN SYSTEM IS THAT PRESENTED BY DIXON-LEWIS ET AL.

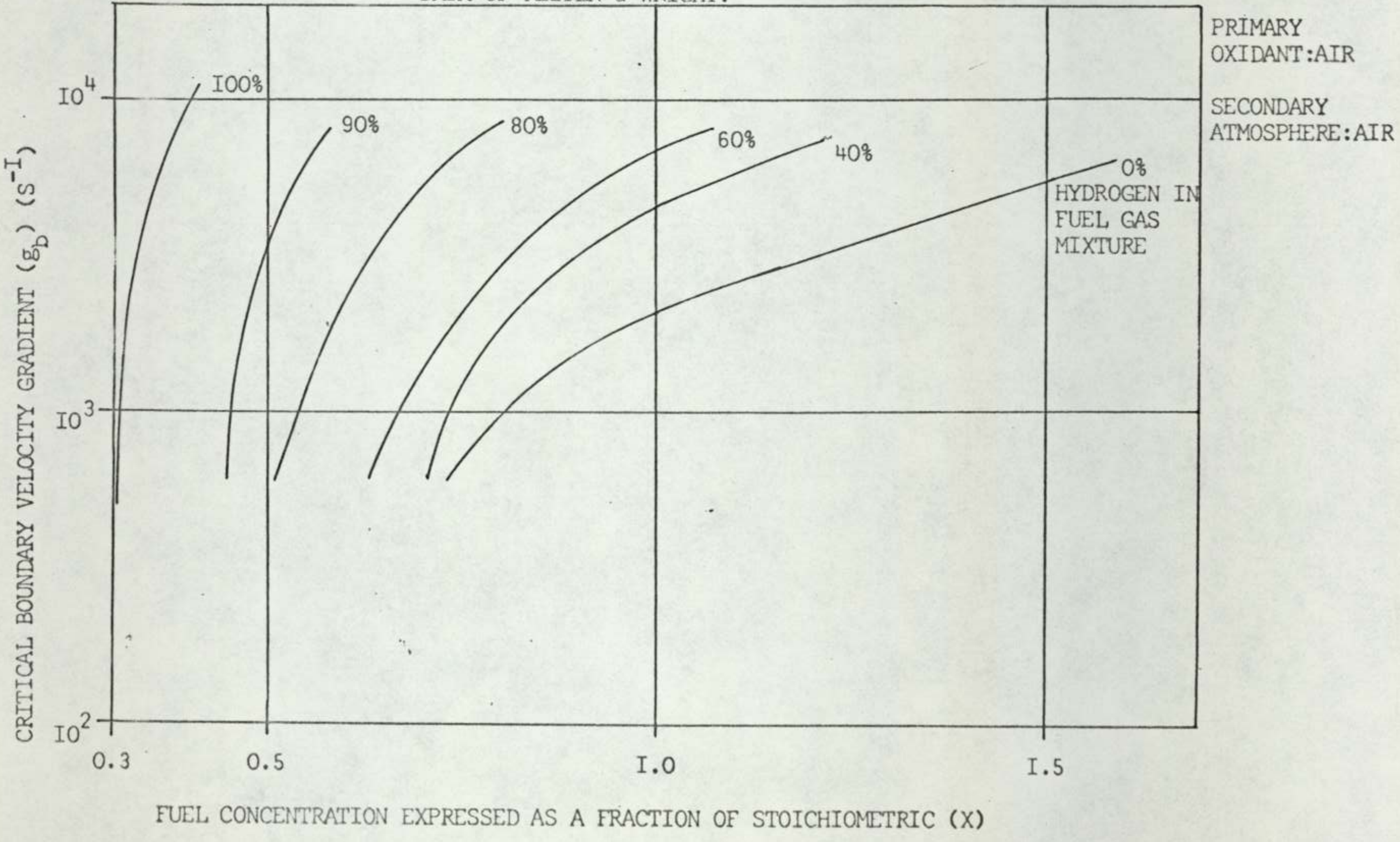


FUEL CONCENTRATION EXPRESSED AS A FRACTION OF STOICHIOMETRIC (X)

GRAPH NO. 49

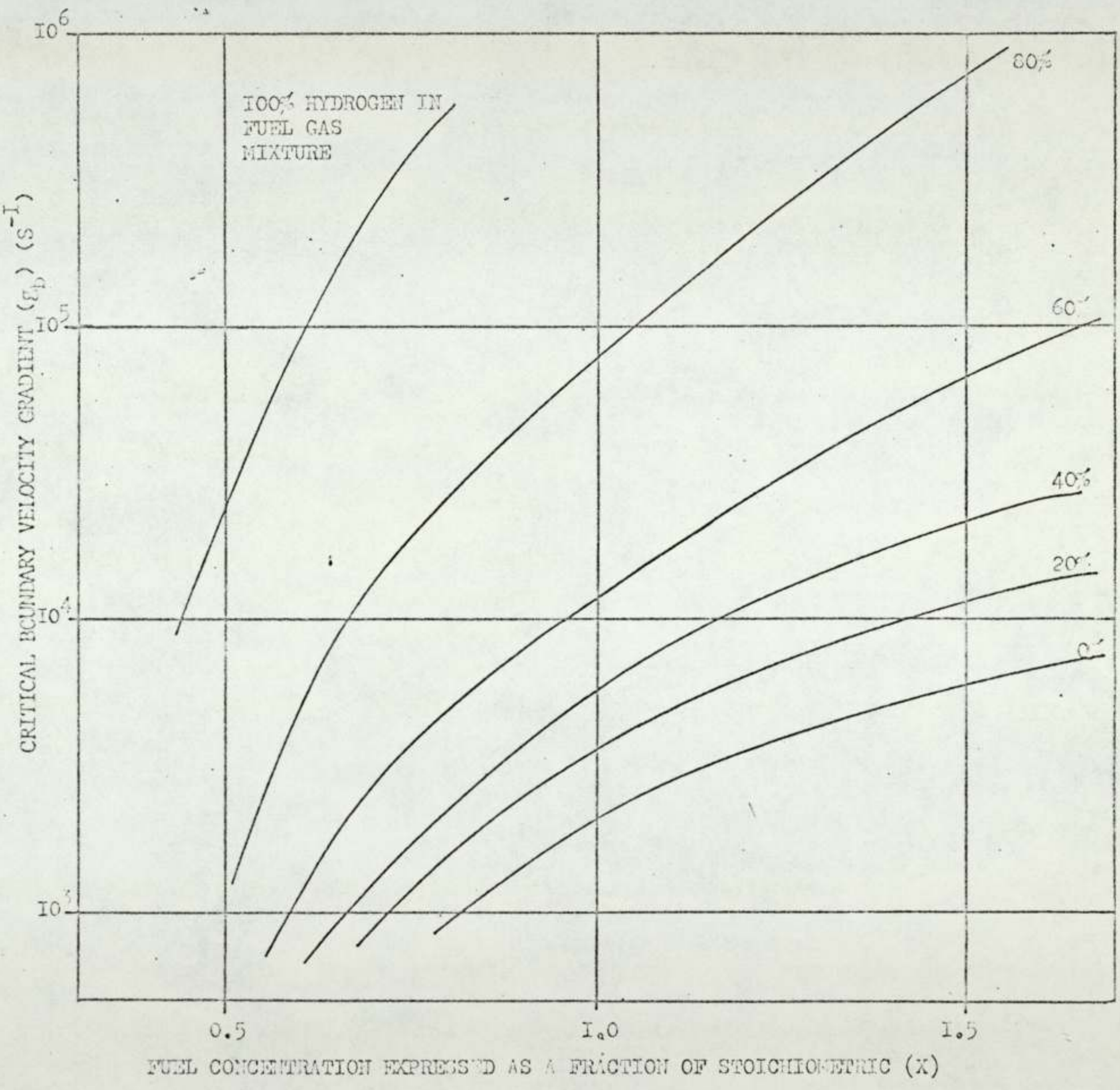
CRITICAL BOUNDARY VELOCITY GRADIENT PLOTTED AGAINST FUEL CONCENTRATION FOR HYDROGEN-PROPANE-AIR SYSTEM.

DATA OF REITER & WRIGHT.



GRAPH NO. 50

CRITICAL BOUNDARY VELOCITY GRADIENT PLOTTED AGAINST FUEL CONCENTRATION FOR BLOW OFF OF LAMINAR BURNER FLAMES USING THE HYDROGEN-PROPANE-AIR SYSTEM AS PREDICTED BY THE METHOD OF VAN KREVELEN AND CHERMIN.

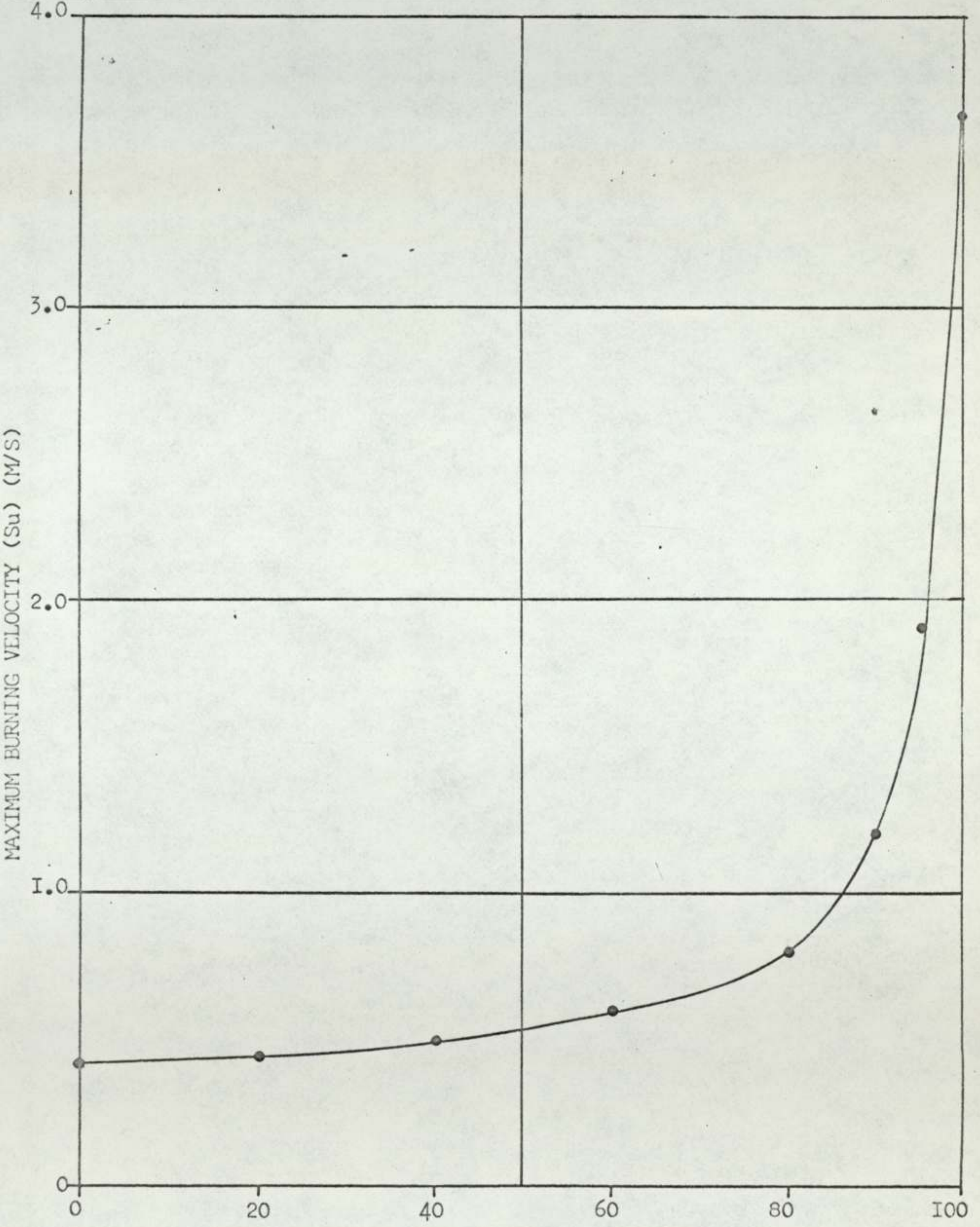


GRAPH NO: 51

GRAPH NO. 52

MAXIMUM BURNING VELOCITY PLOTTED AGAINST PERCENTAGE HYDROGEN PRESENT IN THE FUEL GAS MIXTURE. THE DATA WAS OBTAINED FROM GRAPH NO. 49 AND HAS BEEN PRESENTED IN TABULAR FORM ABOVE THE GRAPH.

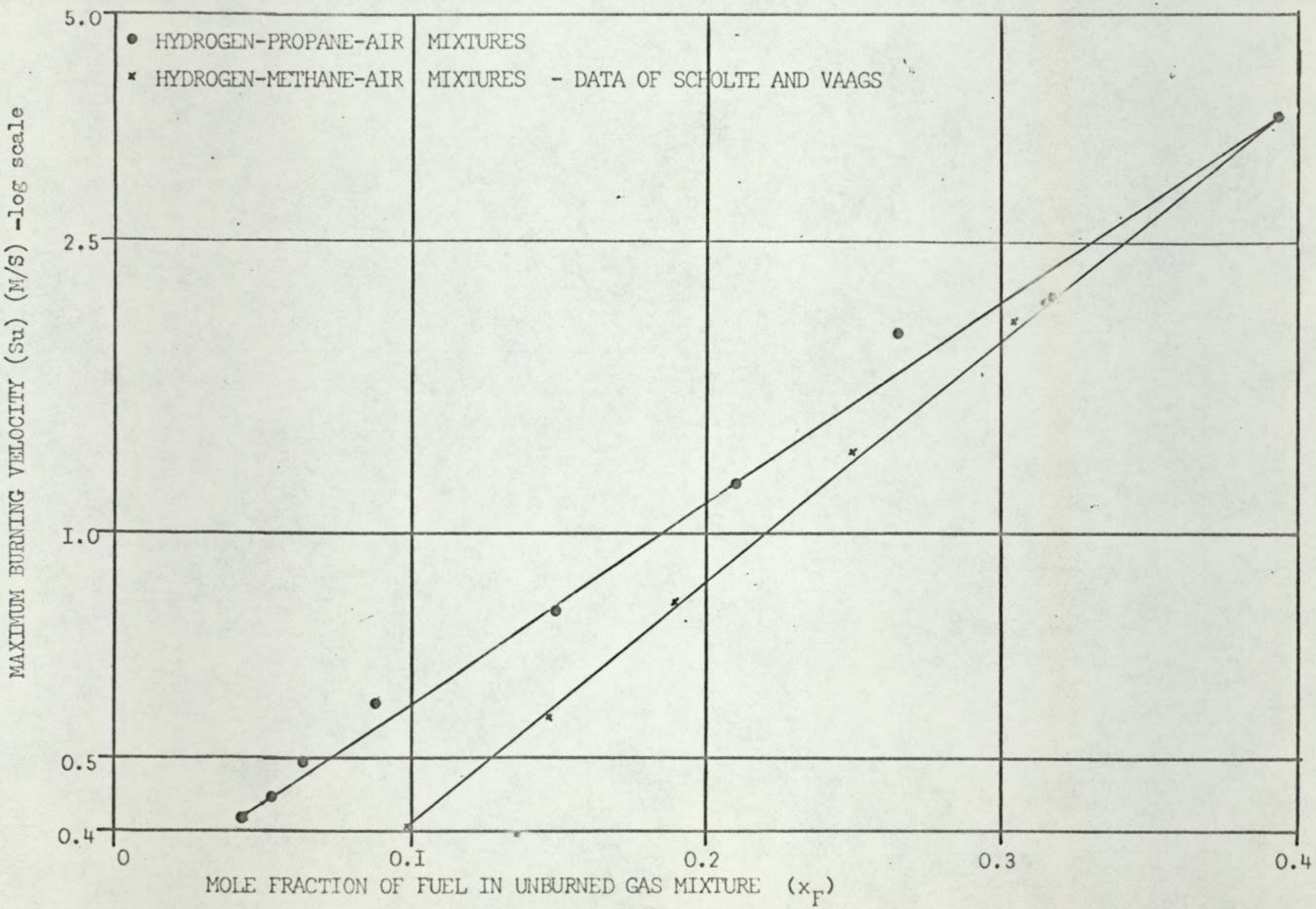
PERCENTAGE HYDROGEN IN FUEL GAS MIXTURE	FUEL CONCENTRATION AT MAXIMUM BURNING VELOCITY	PERCENTAGE FUEL IN MIXTURE	MAXIMUM BURNING VELOCITY (M/S)
0	I.07	4.30	0.418
20	I.09	5.29	0.445
40	I.05	6.44	0.49I
60	I.06	8.83	0.595
80	I.I7	I4.95	0.788
90	I.20	2I.00	I.I88
95	I.25	26.60	I.870
I00	I.54	39.30	3.640



GRAPH NO. 52

GRAPH NO. 53

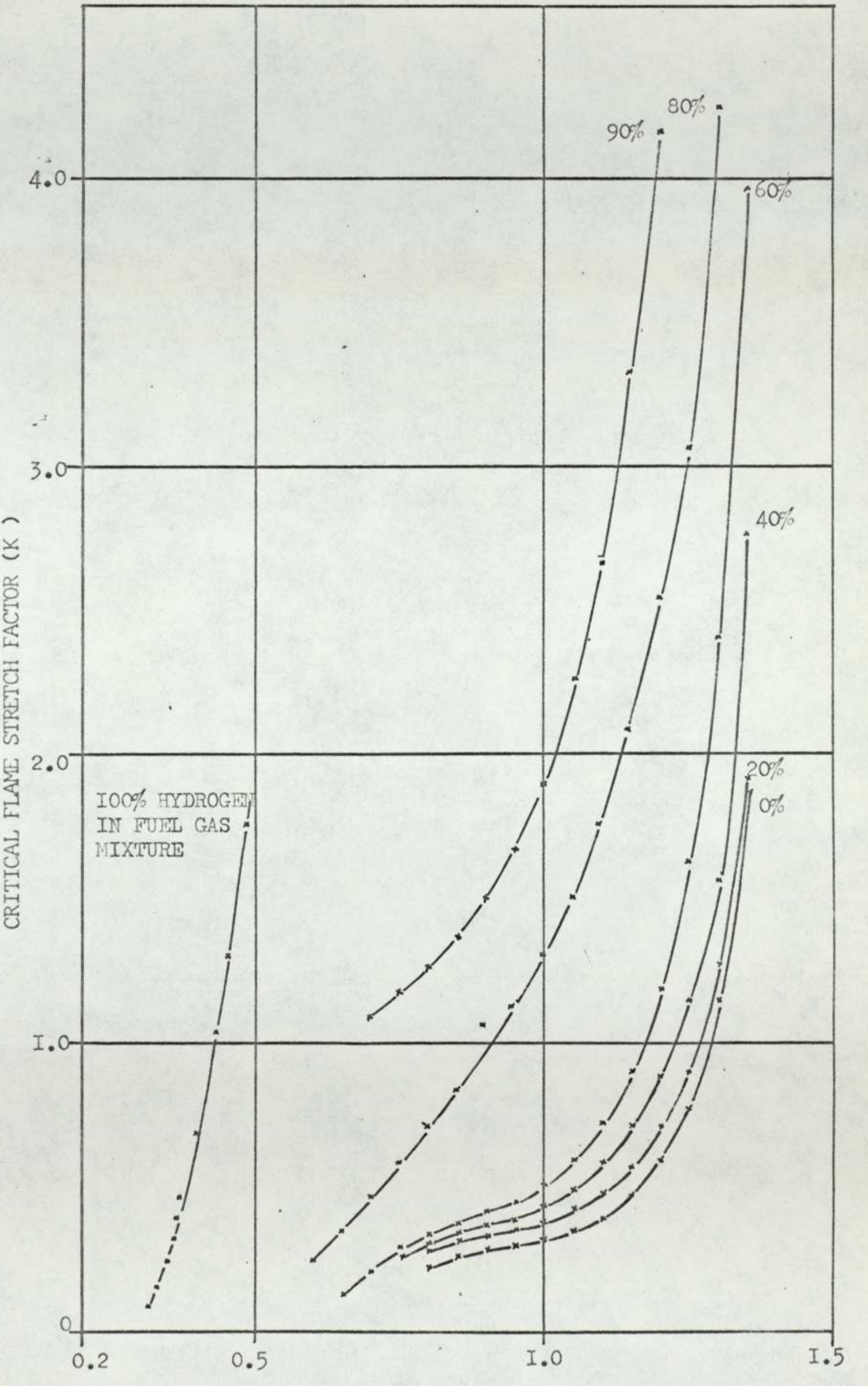
MAXIMUM BURNING VELOCITY PLOTTED AGAINST MOLE FRACTION OF FUEL IN THE UNBURNED GAS MIXTURE FOR THE HYDROGEN-PROPANE-AIR AND HYDROGEN-METHANE-AIR SYSTEMS.


MAXIMUM BURNING VELOCITY (Su) (M/S) -log scale
5.0
2.5
1.0
0.5
0.4
0
0.1
0.2
0.3
0.4
MOLE FRACTION OF FUEL IN UNBURNED GAS MIXTURE (x_F)
● HYDROGEN-PROPANE-AIR MIXTURES
× HYDROGEN-METHANE-AIR MIXTURES - DATA OF SCHOLTE AND VAAGS

GRAPH NO. 53

CRITICAL FLAME STRETCH FACTOR PLOTTED AGAINST FUEL CONCENTRATION FOR BLOW OFF OF LAMINAR BURNER FLAMES USING PROPANE AND HYDROGEN AS THE FUEL GASES.

PRIMARY OXIDANT: AIR SECONDARY ATMOSPHERE: AIR

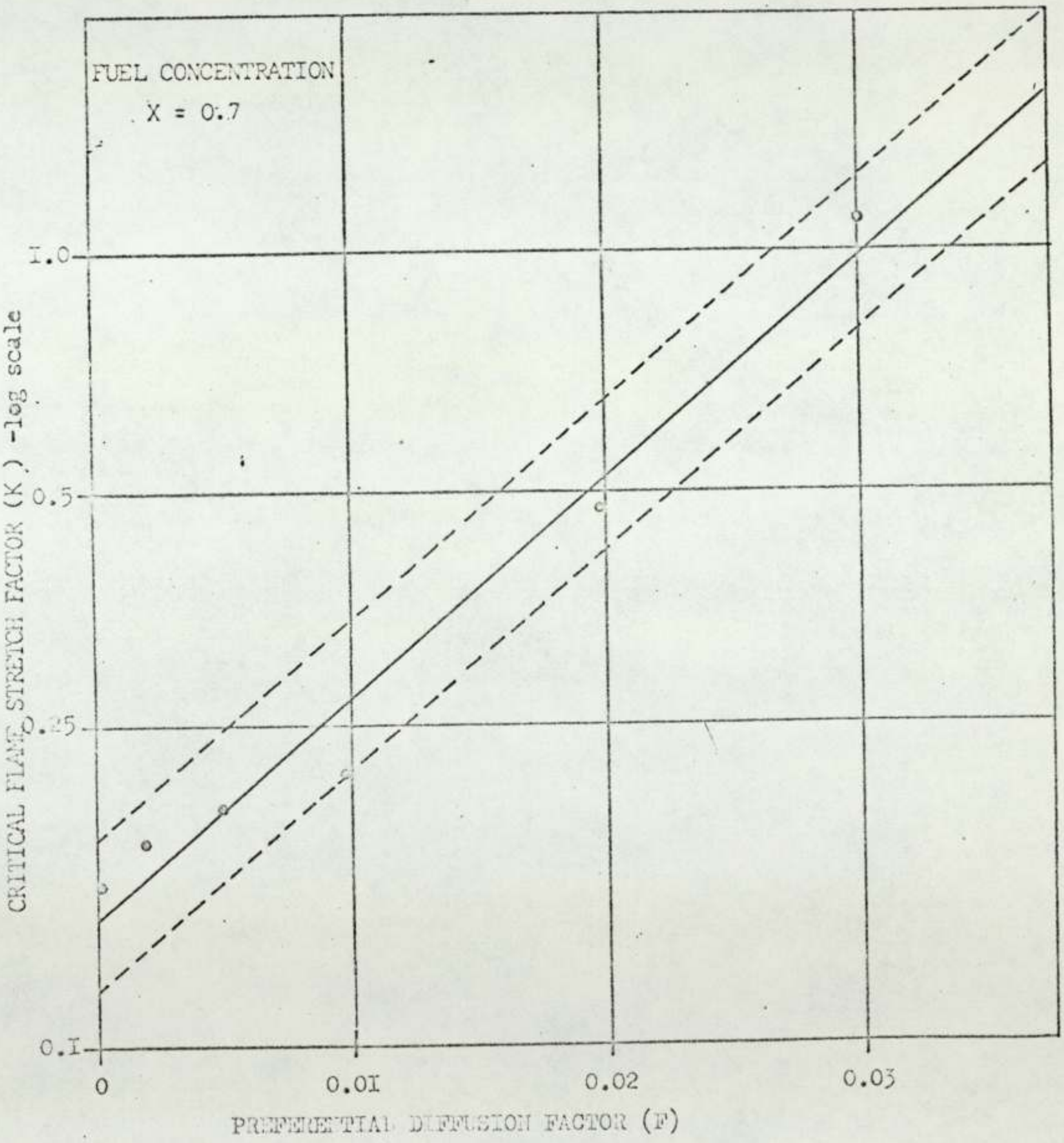


GRAPHS NOS. 55-61

CRITICAL FLAME STRETCH FACTOR PLOTTED AGAINST PREFERENTIAL DIFFUSION FACTOR FOR FLAMES OF CONSTANT AERATION USING THE HYDROGEN-PROPANE-AIR SYSTEM. EQUATIONS OF THE LINES HAVE BEEN COMPUTED BY A 'LEAST SQUARES' PROGRAM AND ARE INCLUDED ABOVE EACH GRAPH TOGETHER WITH STANDARD DEVIATION DATA. THE DOTTED LINES ARE 95% CONFIDENCE LIMITS.

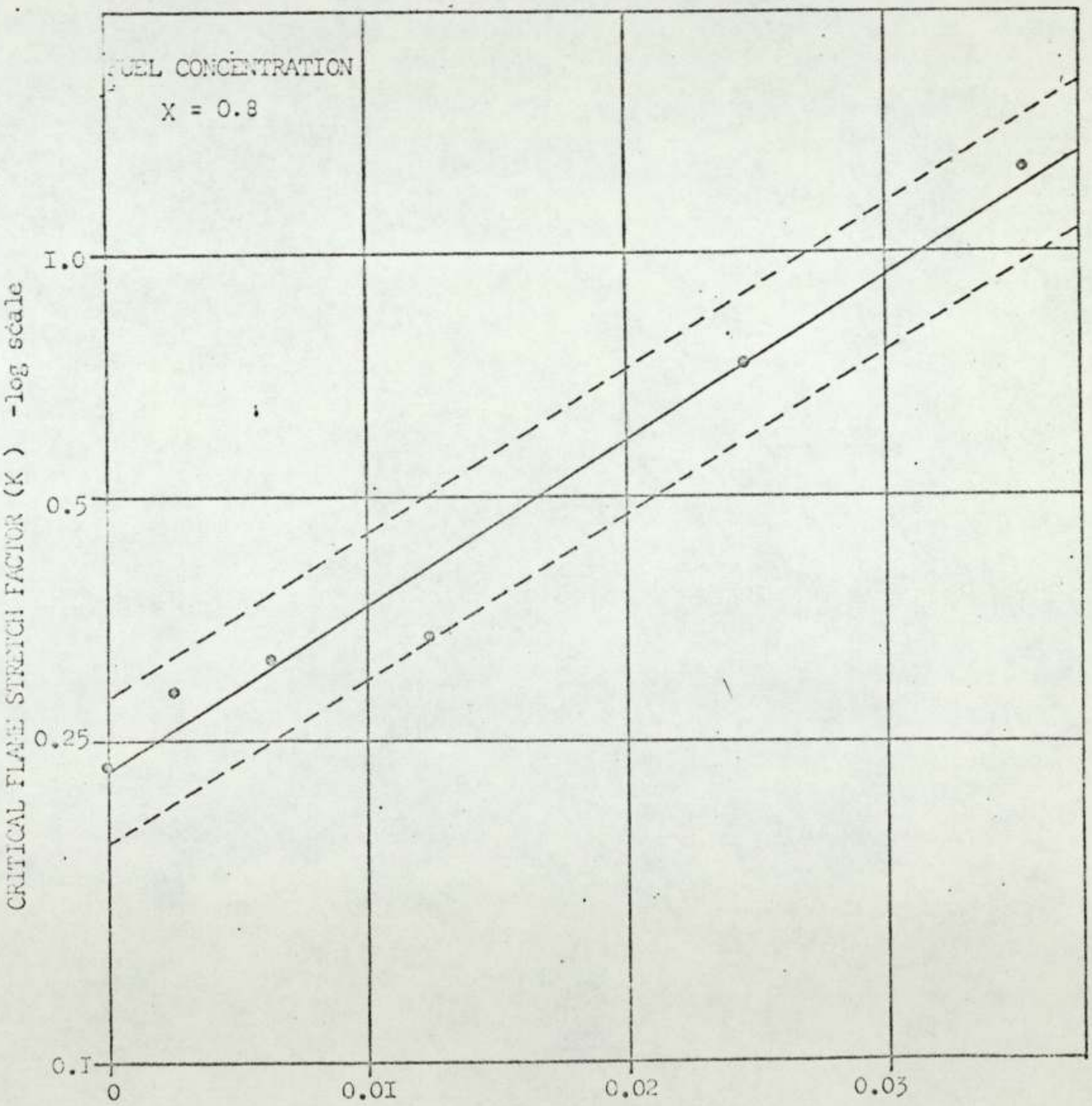
$K = 0.146\, e^{63.5\, F}$

$(\sigma_{\ln K} = 0.1119)$



GRAPH NO. 55

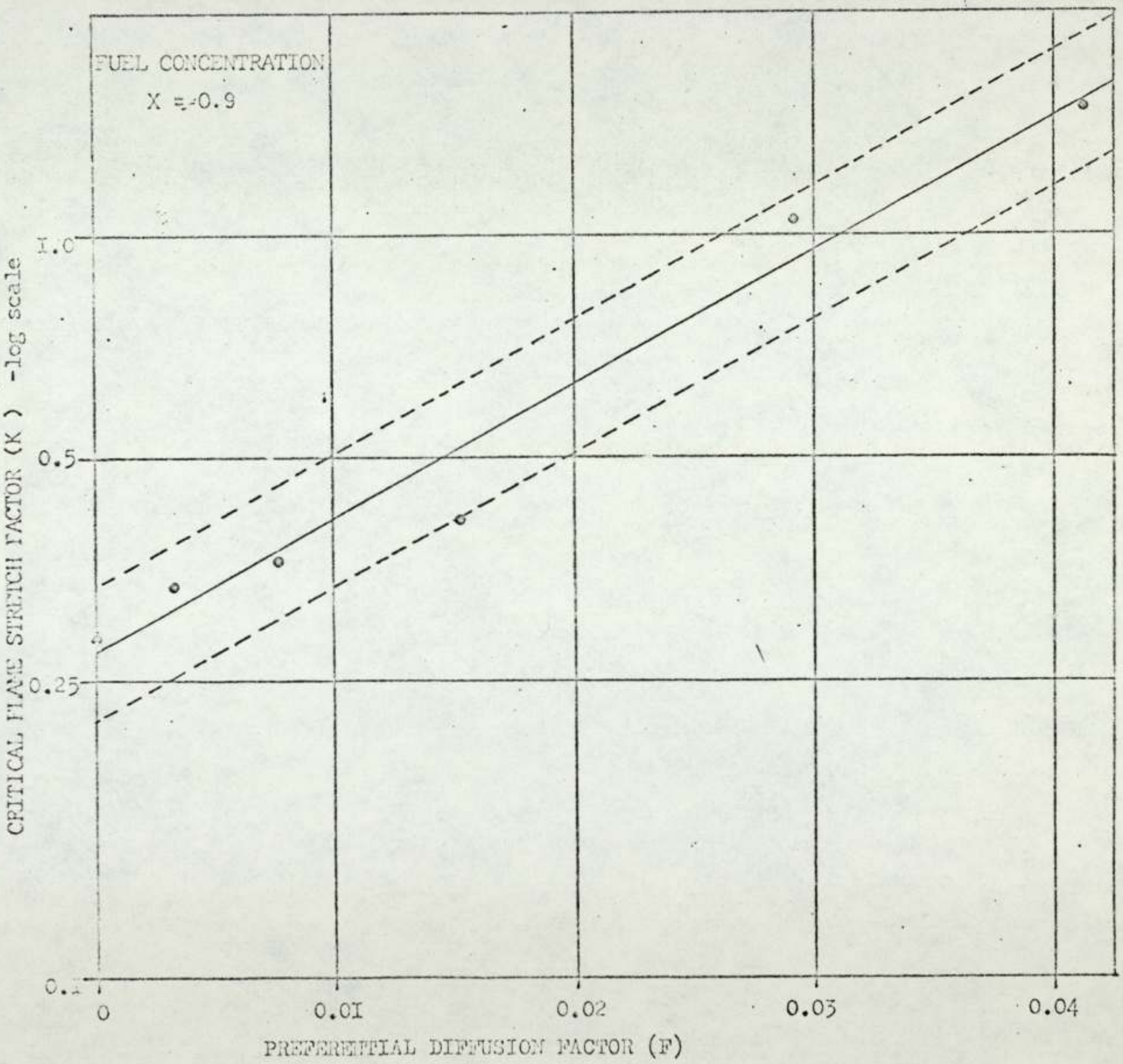
$$K = 0.229\, e^{46.8 F}$$
$$(\sigma_{\ln K} = 0.0989)$$



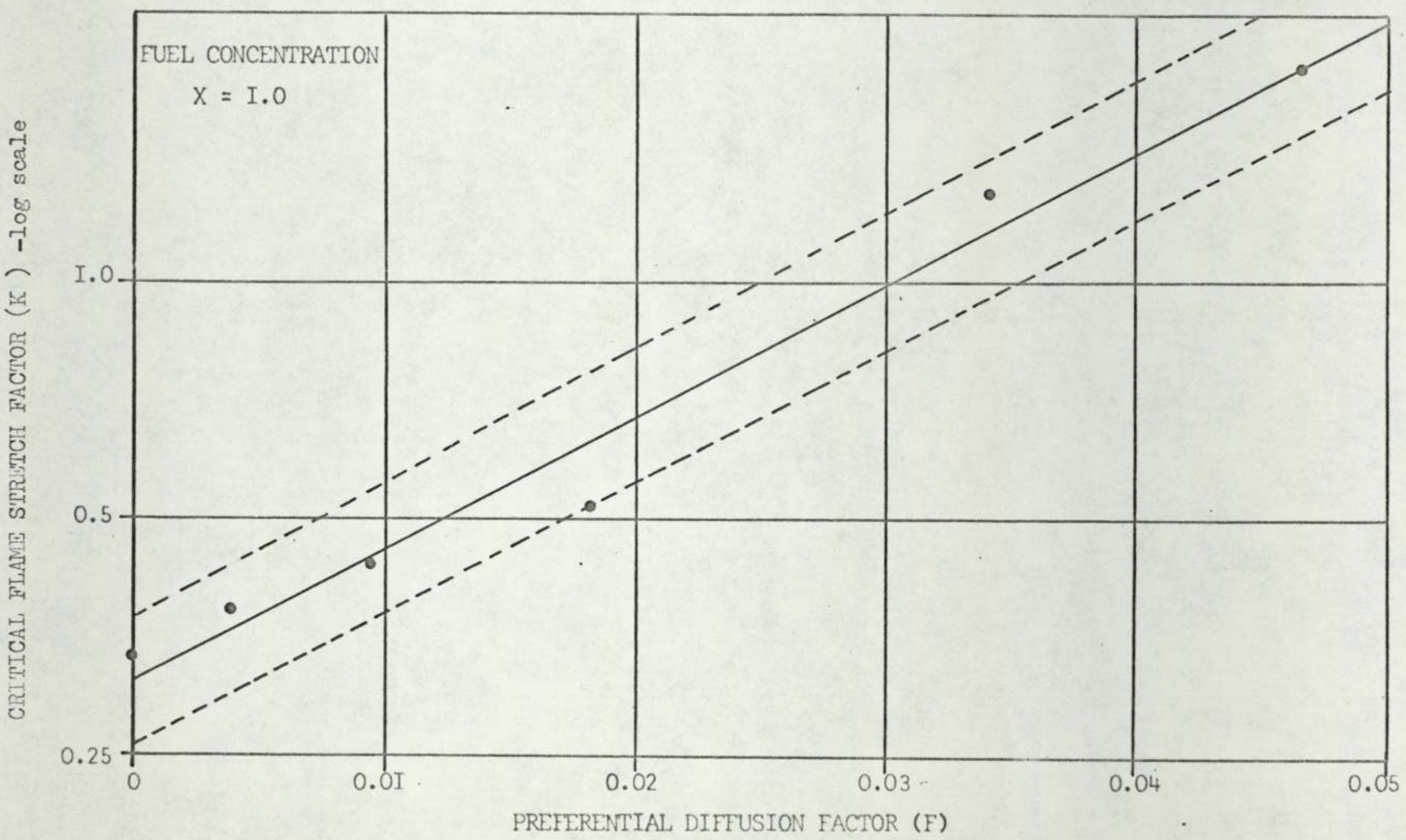
GRAPH NO. 56

$$K = 0.274\, e^{41.8 F}$$

$$(\sigma_{\ln K} = 0.0983)$$



GRAPH NO. 57



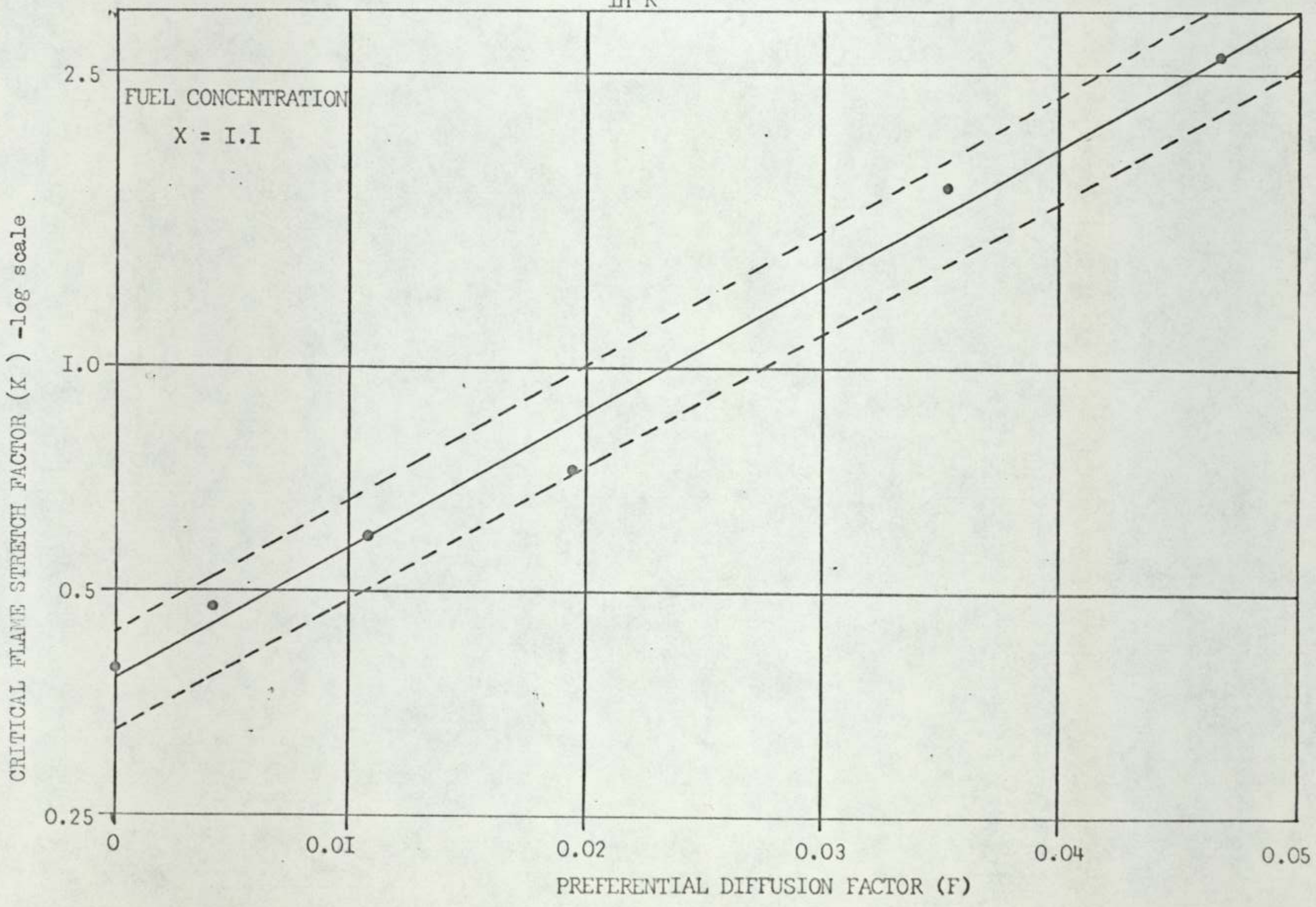
$K = 0.3I2\ e^{38.6\ F}$

$(\sigma_{\ln K} = 0.I024\ )$

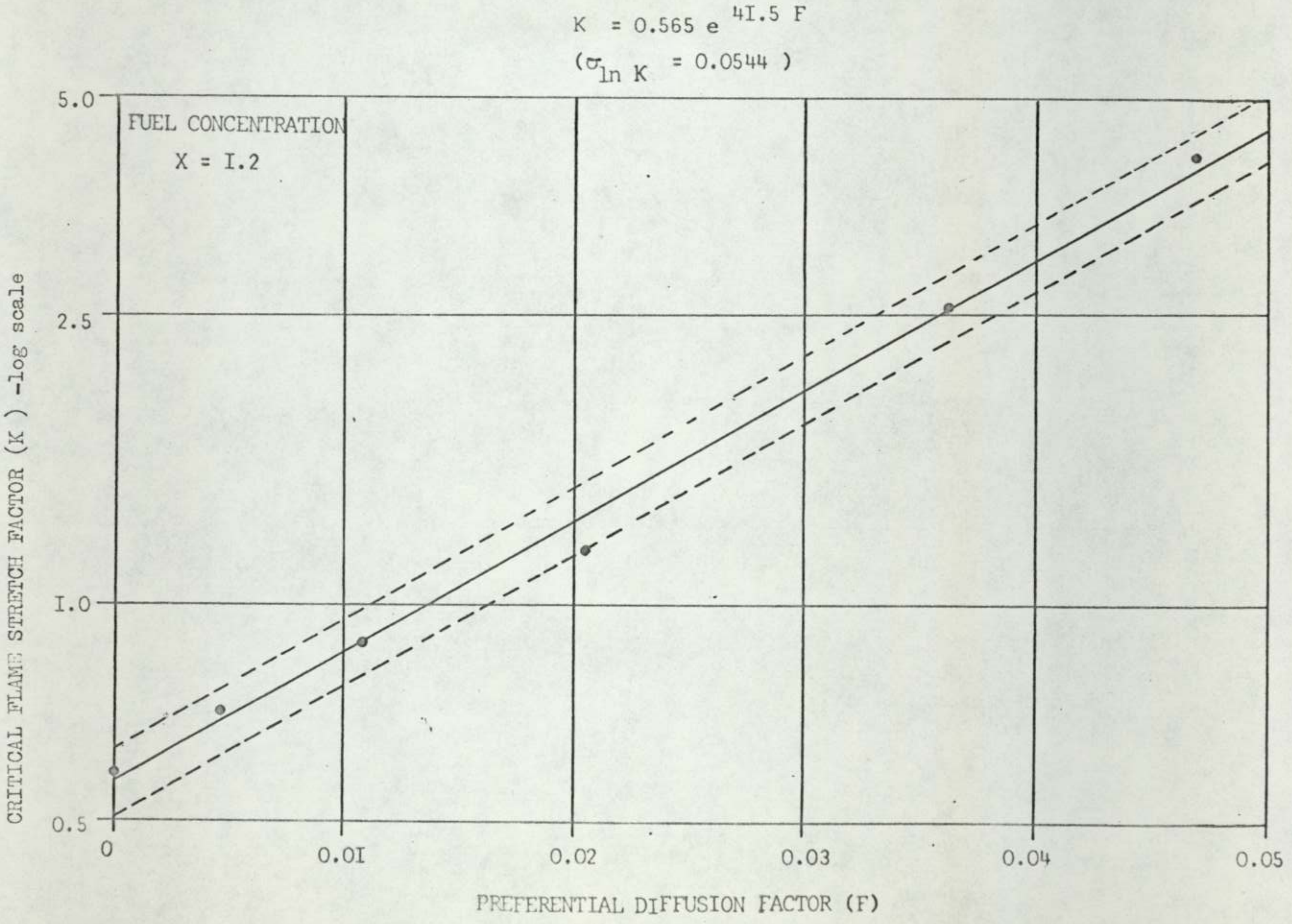
GRAPH NO. 58

$K = 0.383 \, e^{41.1 F}$

$(\sigma_{\ln K} = 0.0787)$



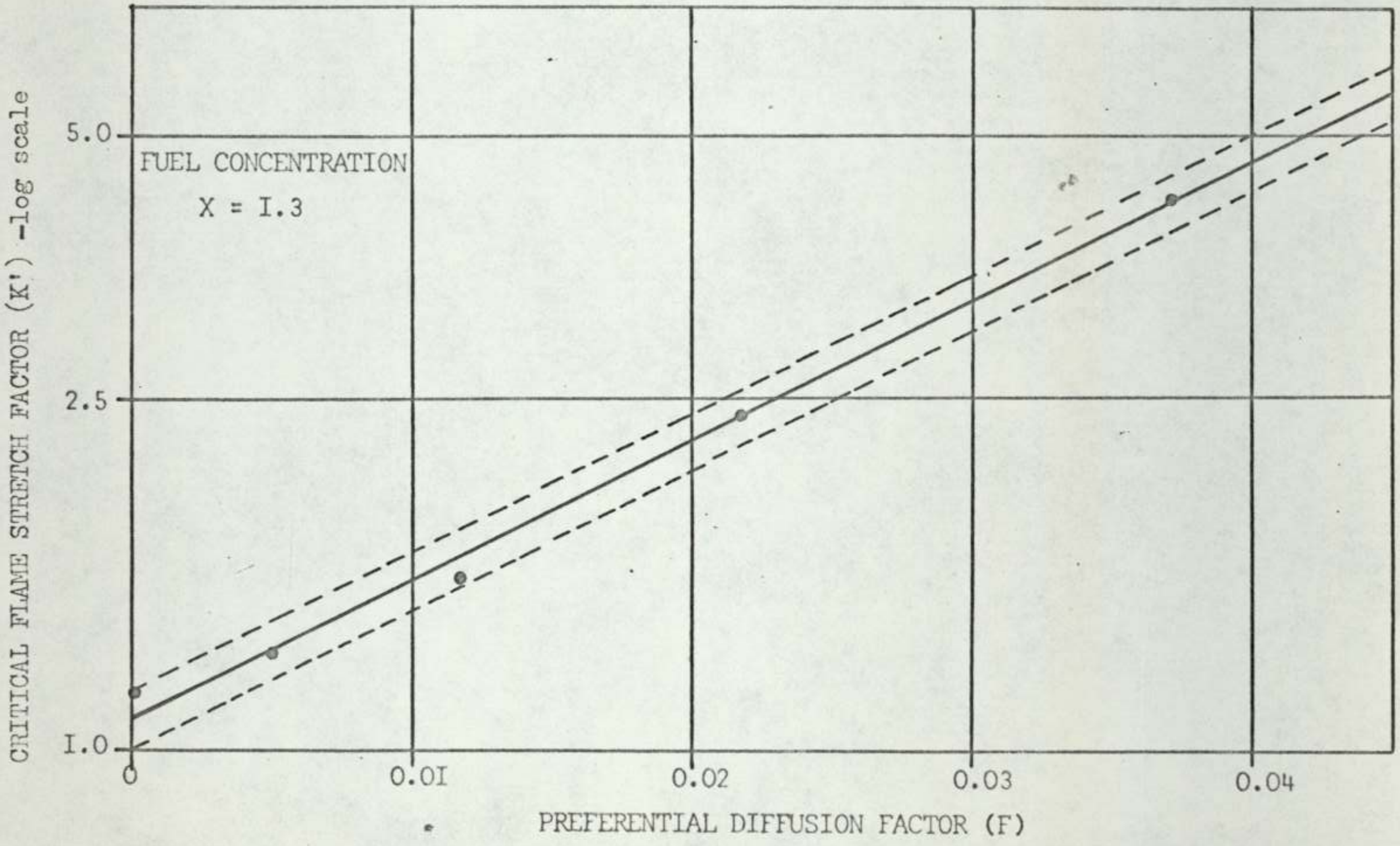
GRAPH NO. 59

$K = 0.565\,e^{41.5\,F}$
$(\sigma_{\ln K} = 0.0544)$
FUEL CONCENTRATION
X = I.2
CRITICAL FLAME STRETCH FACTOR (K) -log scale
5.0
2.5
I.0
0.5
0
0.0I
0.02
0.03
0.04
0.05
PREFERENTIAL DIFFUSION FACTOR (F)

GRAPH NO. 60

$K = 1.087\, e^{36.2 F}$

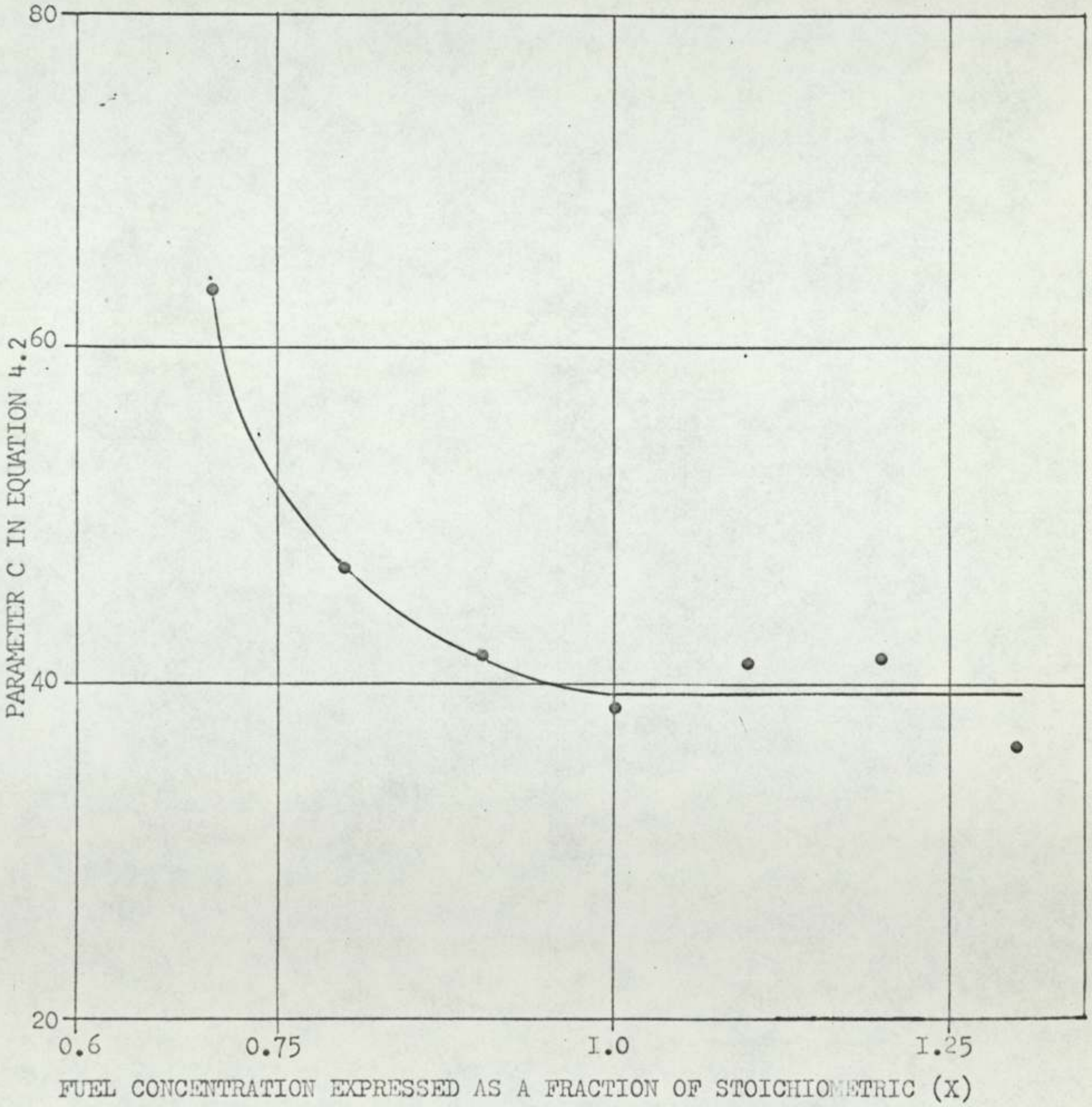
$(\sigma_{\ln K} = 0.0394)$



GRAPH NO. 6I

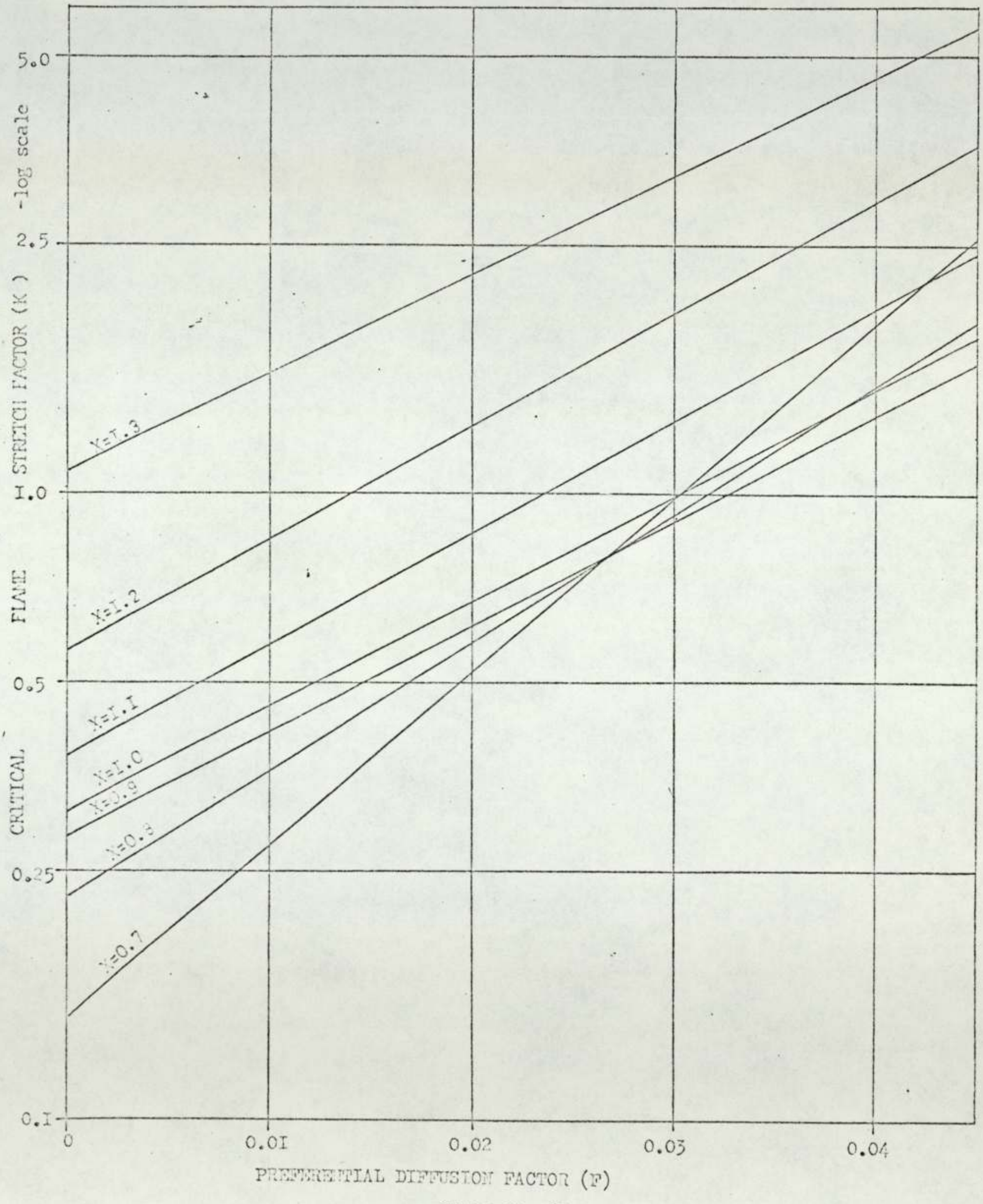
PARAMETER C IN EQUATION 4.2 PLOTTED AGAINST FUEL CONCENTRATION.

VALUES OF C OBTAINED FROM GRAPHS NOS. 55-61.



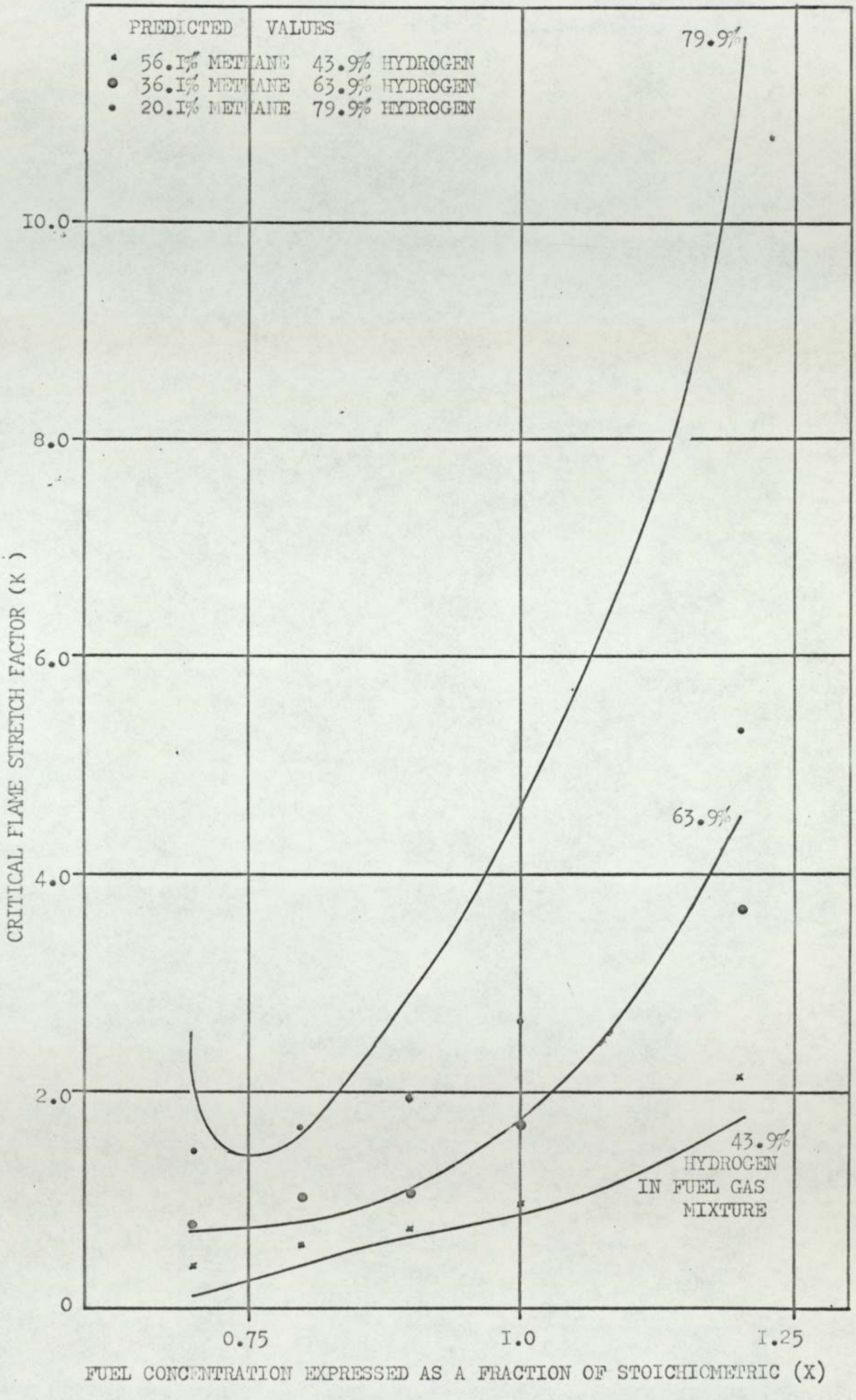
GRAPH NO. 62

CRITICAL FLAME STRETCH FACTOR - PREFERENTIAL DIFFUSION FACTOR CORELATIONS OBTAINED FROM GRAPHS NOS.55- 6I FOR THE FUEL CONCENTRATIONS STATED.



GRAPH NO. 63

CRITICAL FLAME STRETCH FACTOR PLOTTED AGAINST FUEL CONCENTRATION FOR BLOW OFF OF LAMINAR BURNER FLAMES USING HYDROGEN AND METHANE AS THE FUEL GASES.

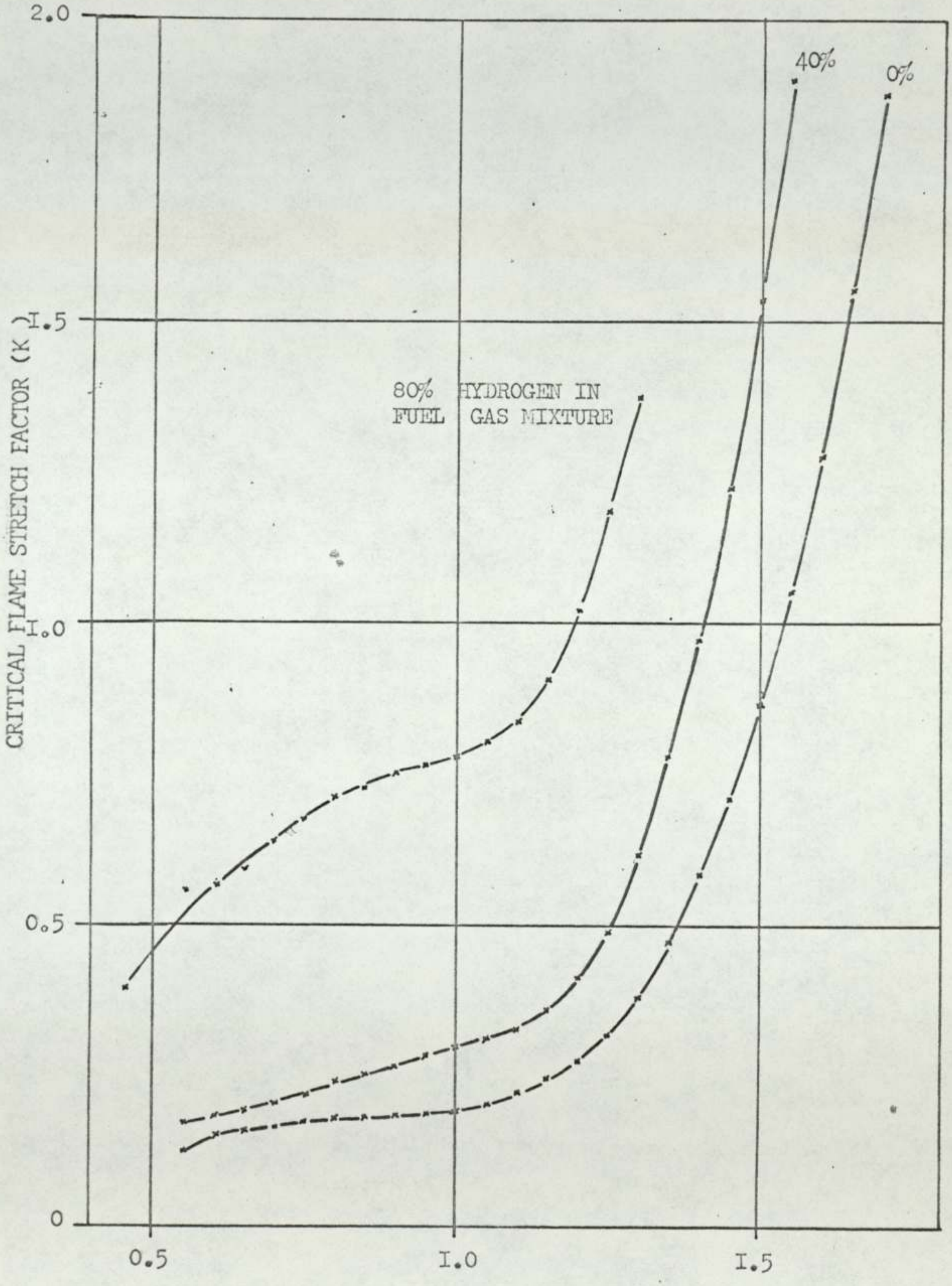


GRAPH NO. 64

CRITICAL FLAME STRETCH FACTOR PLOTTED AGAINST FUEL CONCENTRATION FOR BLOW OFF OF LAMINAR BURNER FLAMES USING PROPANE AND HYDROGEN AS FUEL GASES.

PRIMARY OXIDANT: 21% OXYGEN 79% ARGON

SECONDARY ATMOSPHERE: 21% OXYGEN 79% ARGON



<ins>GRAPH NO. 65</ins>

GRAPHS NOS 66-68

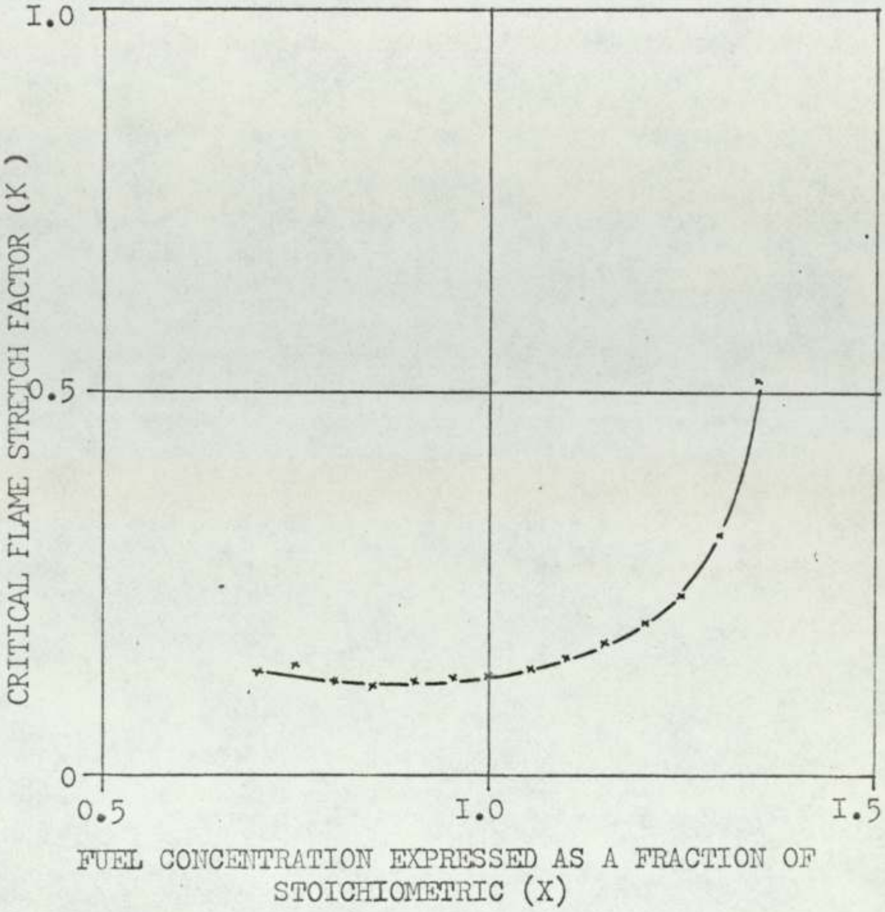
CRITICAL FLAME STRETCH FACTOR PLOTTED AGAINST FUEL CONCENTRATION FOR BLOW OFF OF LAMINAR BURNER FLAMES USING PROPANE AND HYDROGEN AS FUEL GASES.

PRIMARY OXIDANT: 21% OXYGEN 79% HELIUM

SECONDARY ATMOSPHERE: 21% OXYGEN 79% HELIUM

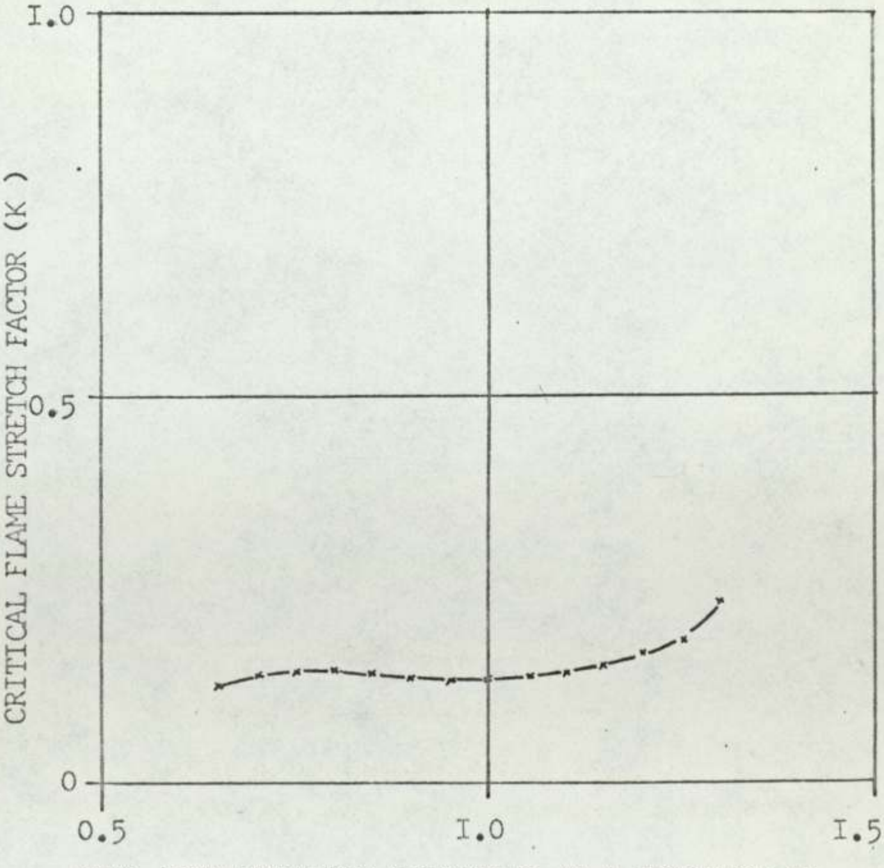
GRAPH NO. 66

FUEL GAS:
100% PROPANE



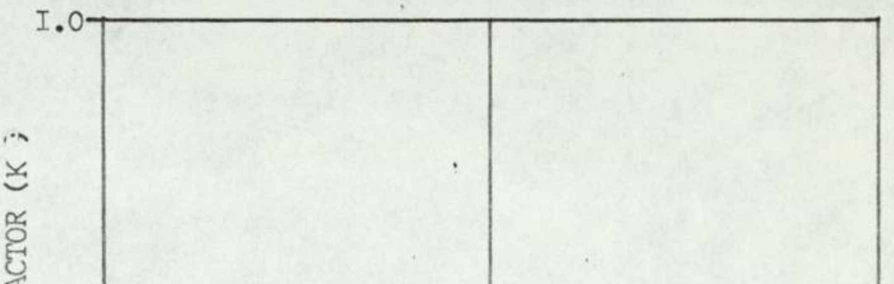
GRAPH NO. 67

FUEL GAS:
60% PROPANE
40% HYDROGEN



GRAPH NO. 68

FUEL GAS:
20% PROPANE
80% HYDROGEN



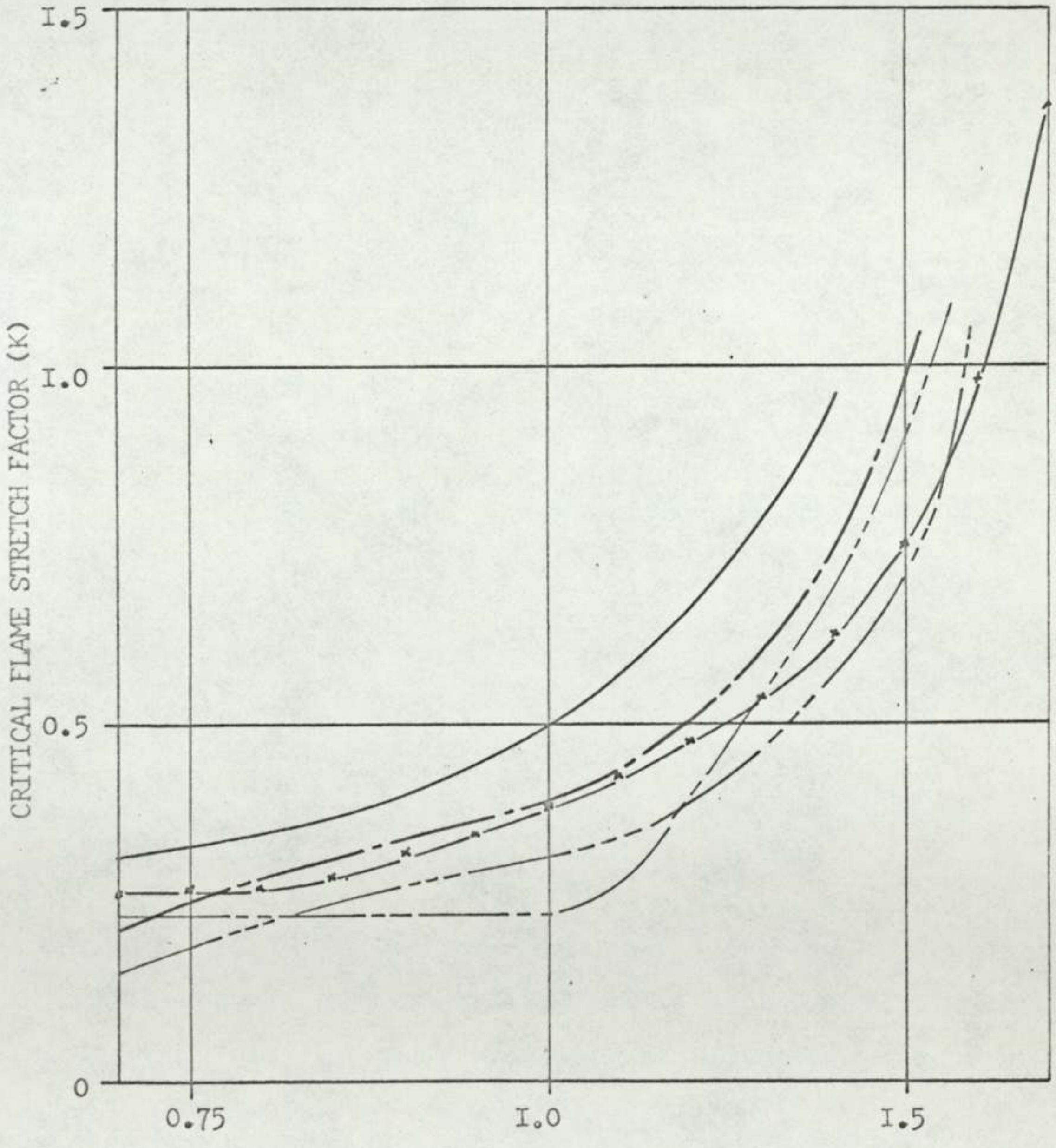
CRITICAL FLAME STRETCH FACTOR PLOTTED AGAINST FUEL CONCENTRATION

FOR THE ETHYLENE-AIR SYSTEM.

PRIMARY OXIDANT: AIR

SECONDARY ATMOSPHERE: AIR

—×— EXPERIMENTAL CORELATION
——— CORELATION OF EDMONDSON AND HEAP
— - — LINE PREDICTED BY EQUATION 4.4
— - - LINE PREDICTED BY REED'S CORELATION-EQUATION I.IO
- - - - EXPERIMENTAL CORELATION FOR PROPANE-AIR SYSTEM



FUEL CONCENTRATION EXPRESSED AS A FRACTION OF STOICHIOMETRIC (X)

GRAPH NO. 69

GRAPHS NOS. 70-73

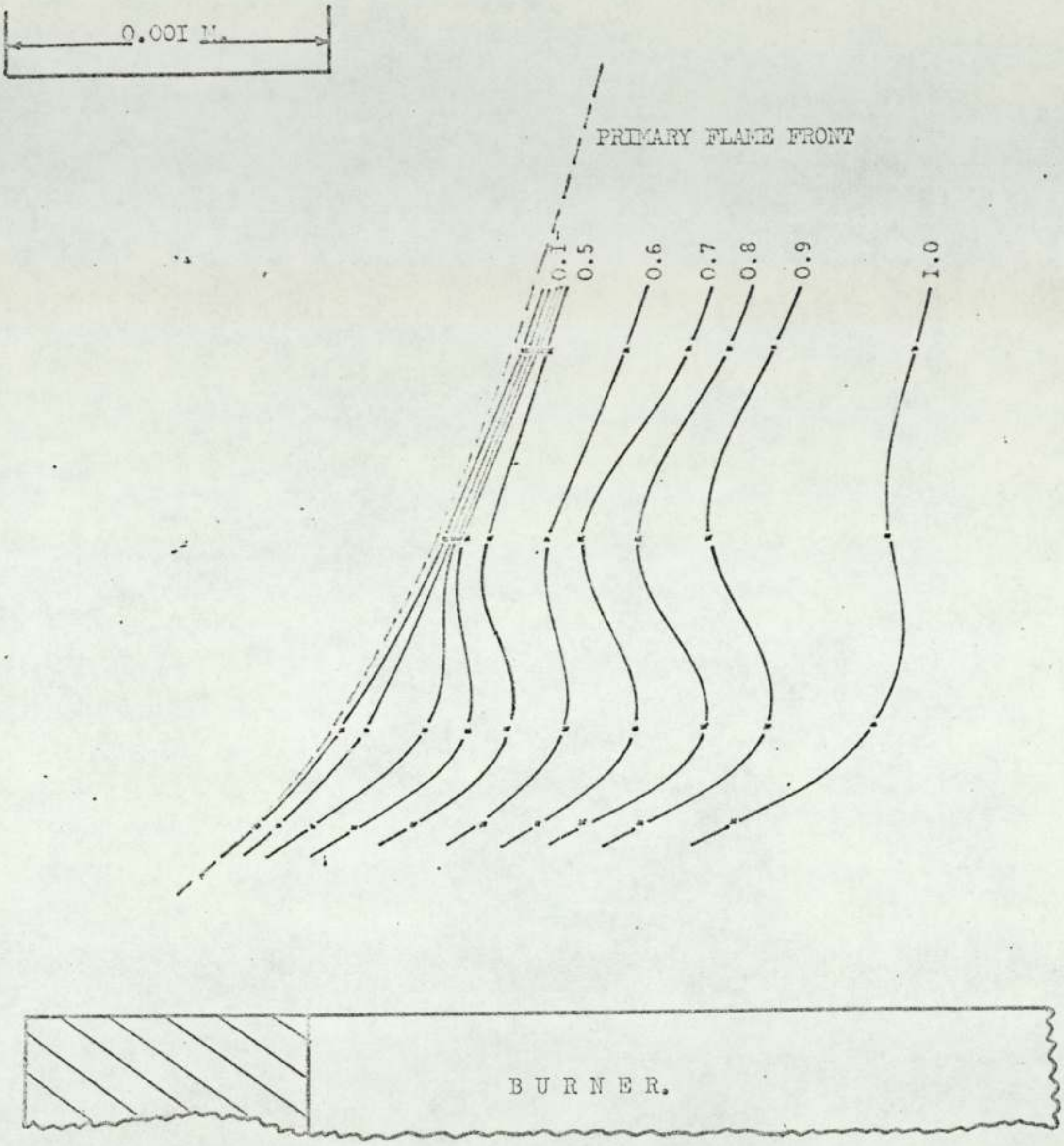
HYDROGEN CONCENTRATION PROFILES IN THE STABILISING REGION OF LAMINAR AERATED BURNER FLAMES.

FUEL GAS: GRAPHS 70-72: 60% HYDROGEN 40% PROPANE

GRAPH 73 : 100% ETHYLENE

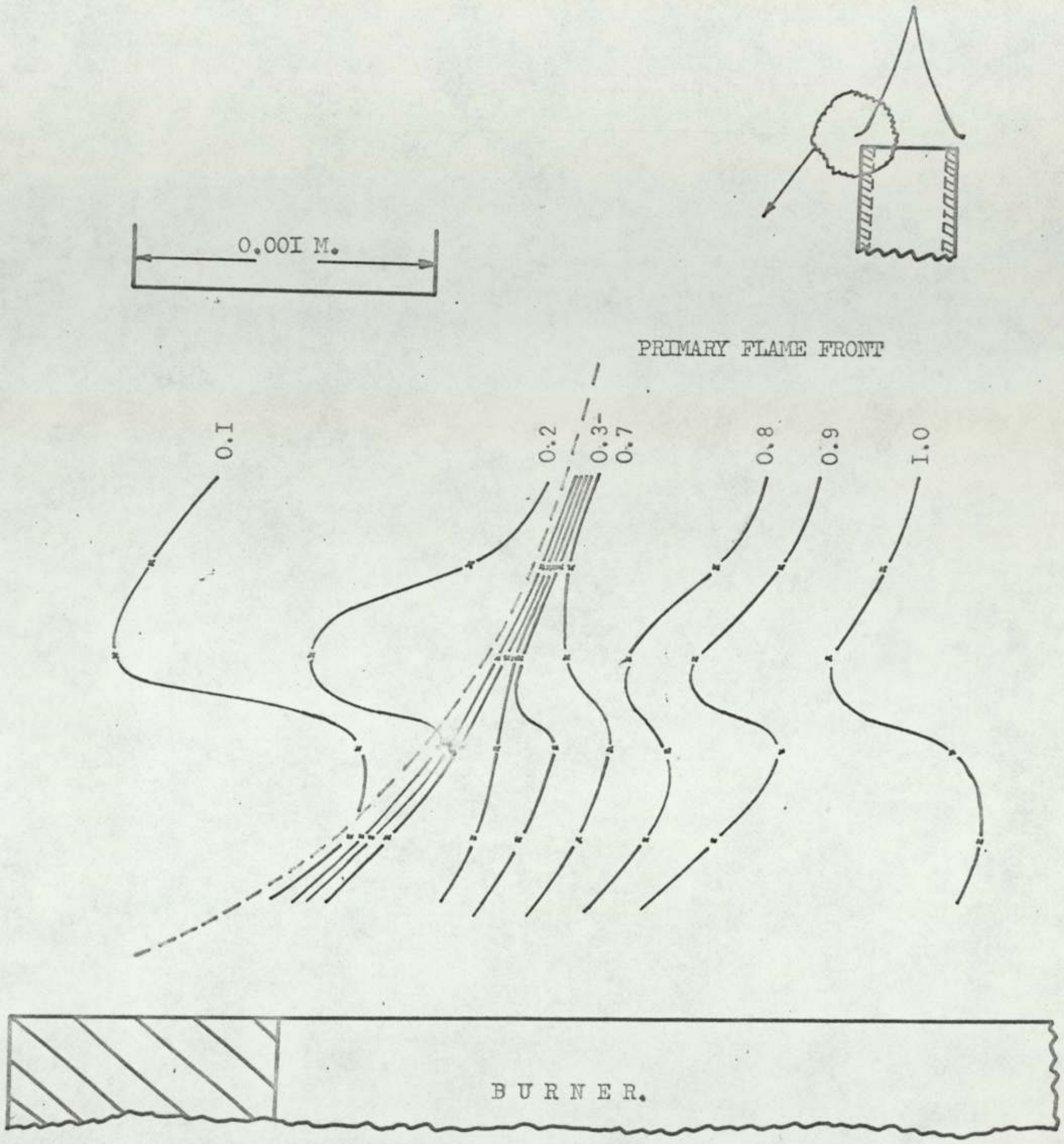
PRIMARY OXIDANT: AIR

SECONDARY ATMOSPHERE: AIR



CONCENTRATION PROFILES OF HYDROGEN IN STABILISING REGION OF 0.8 STOICHIOMETRIC FLAME. CONCENTRATIONS EXPRESSED AS A FRACTION OF THE UNBURNED GAS CONCENTRATION. FUEL GAS MIXTURE:- 60% H_2 , 40% C_3H_8

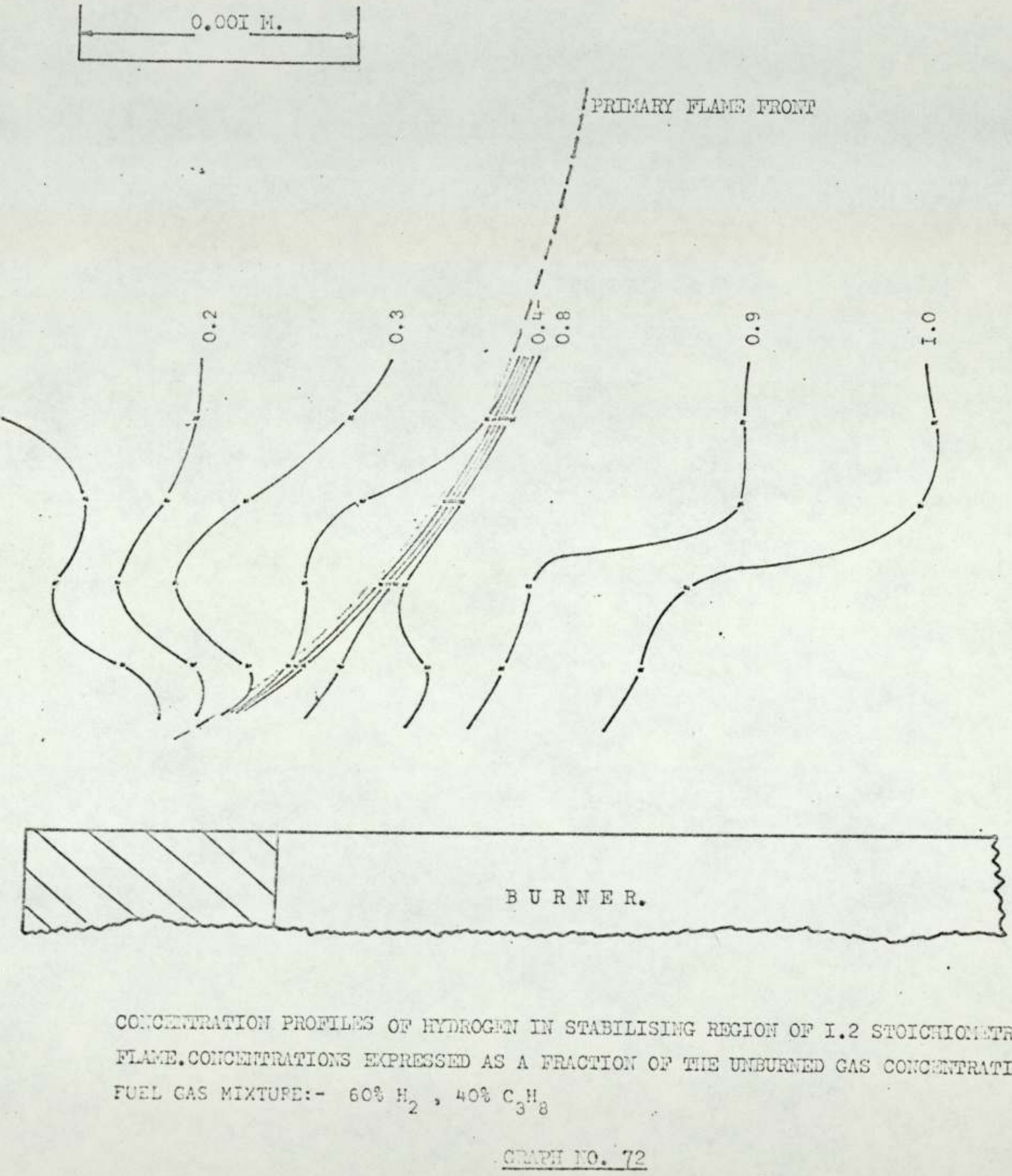
GRAPH NO. 70



CONCENTRATION PROFILES OF HYDROGEN IN STABILISING REGION OF 1.0 STOICHIOMETRIC FLAME. CONCENTRATIONS EXPRESSED AS A FRACTION OF THE UNBURNED GAS CONCENTRATION.

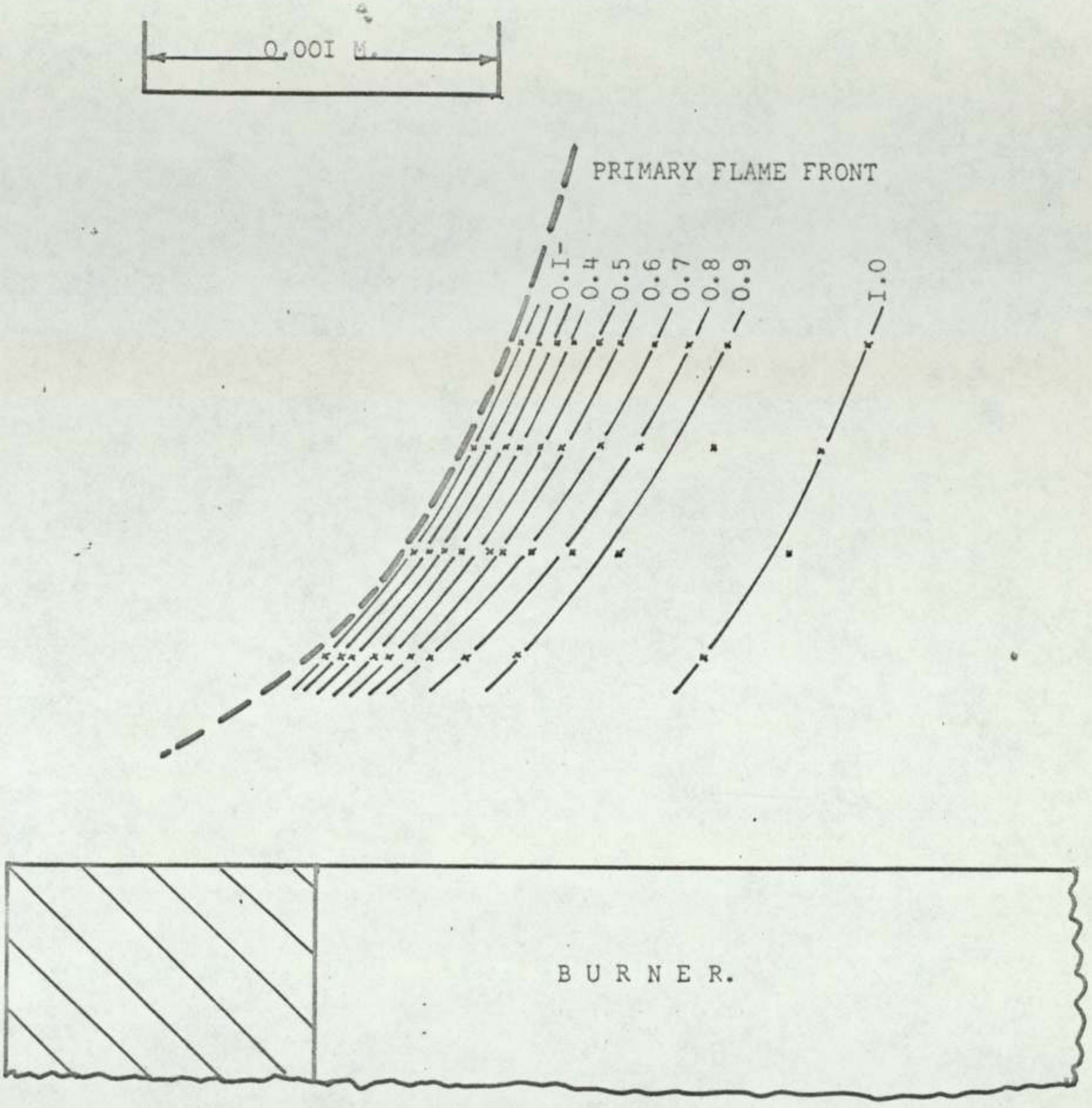
FUEL GAS MIXTURE:- 60% H_2 , 40% C_3H_8

GRAPH NO. 71



CONCENTRATION PROFILES OF HYDROGEN IN STABILISING REGION OF I.2 STOICHIOMETRIC FLAME. CONCENTRATIONS EXPRESSED AS A FRACTION OF THE UNBURNED GAS CONCENTRATION.
FUEL GAS MIXTURE:- 60% H_2 , 40% C_3H_8

GRAPH NO. 72



CONCENTRATION PROFILES OF ETHYLENE IN STABILISING REGION OF I.0 STOICHIOMETRIC ETHYLENE-AIR FLAME. CONCENTRATIONS EXPRESSED AS A FRACTION OF THE UNBURNED GAS CONCENTRATION.

GRAPH NO: 73

Appendix 2.

A QUANTITATIVE ASSESSMENT OF THE MAXIMUM ERRORS WHICH MAY ARISE FROM THE EXPERIMENTAL TECHNIQUES USED.

Appendix 2.

A quantitative assessment of the maximum errors which may arise from the experimental techniques used.

The method used has been described by Jensen and Jeffreys (A2.1). In section A2.1.1., the procedure has been reported in detail. There after, only the equations derived in a similar manner have been included.

A2.1 Flow metering.

A2.1.1 Correction of indicated flow rate to temperature and pressure at the outlet of a variable area flow meter

Equation (3.2) states as follows:-

$$\frac{Q_2}{Q_1} \propto \left|\frac{\rho_1}{\rho_2}\right|^{1/2} \qquad (3.2)$$

This equation may be rewritten:-

$$Q_1 \ \rho_1^{\ 1/2} = W^{11} Q_2 \ \rho_2^{\ 1/2} \qquad (A2.1)$$

If Q_2 and ρ_2 refer to the flow meter calibration and W^{11} is a constant, it follows that for any specific indicated flow rate

$$Q = \frac{W^1}{\rho^{1/2}} \qquad (A2.2)$$

where W^1 is a constant incorporating Q_2 and W_2.

Now $\rho \propto P$

and $\rho \propto \frac{1}{T}$

Therefore $$Q \propto W^1 \left|\frac{T}{P}\right|^{1/2} = W \left|\frac{T}{P}\right|^{1/2} \qquad (A2.3)$$

where W is a constant

Partially differentiating equation (A2.3):-

$$\frac{\partial Q}{\partial P} = -\frac{1}{2} \ WT^{1/2} \ P^{-3/2} \qquad (A2.4)$$

$$\frac{\partial Q}{\partial T} = \frac{1}{2} WT^{-1/2} P^{-1/2} \qquad (A2.5)$$

It may be shown that $Q = Q \quad (P,T)$

$$\delta Q = \frac{\partial Q}{\partial P} . dP + \frac{\partial Q}{\partial T} . dT \qquad (A2.6)$$

If P and T are small it follows that

$$\delta Q = \frac{\partial Q}{\partial P} .\delta P + \frac{\partial Q}{\partial T} \delta T \qquad (A2.7)$$

where δQ = error in flow measurement arising from errors in pressure measurement (δP) and temperature measurement (δT).

Substituting into equation (A2.7) from equations (A2.4) and (A2.5) and dividing throughout by Q, the following equation is obtained:-

$$\frac{\delta Q}{Q} = \frac{1}{2} \left(\frac{\delta T}{T} - \frac{\delta P}{P} \right) \qquad (A2.8)$$

(i) Evaluation of $\frac{\delta P}{P}$

Maximum error in water manometer reading = ± 0.1 mm.Hg.

Maximum error in barometer reading = ± 0.05 mm.Hg.

∴ Total maximum error in pressure reading = ± 0.15 mm.Hg.

$$\therefore \quad \frac{\delta P}{P} = \pm \frac{0.15}{760} \times 100\% \quad \pm 0.02\%$$

(ii) Evaluation of $\frac{\delta T}{T}$

Maximum error in thermometer reading = ± 0.05°C

$$\frac{\delta T}{T} = \pm \frac{0.05}{293} . \times 100\% \quad \pm 0.017\%$$

Substituting from (i) and (ii) into equation (A2.8)

$$\frac{\delta Q}{Q} = \frac{1}{2} (\pm 0.017 \pm 0.02)\%$$

This has a maximum value of

$$\frac{\delta Q}{Q} = \pm 0.0185\%$$

A2.1.2 Correction for change of temperature and pressure of gas during passage between flow meter and burner

It is already known that $Q = Q(P,T)$

by the gas laws it follows that $Q \propto \left(\frac{T}{P}\right)$

It may then be shown that

$$\frac{\delta Q}{Q} = \frac{\delta T}{T} - \frac{\delta P}{P} \qquad (A2.9)$$

The values of $\frac{\delta T}{T}$ and $\frac{\delta P}{P}$ evaluated in section A2.1.1 will be applicable to this correction also

$$\therefore \quad \frac{\delta Q}{Q} = (\pm 0.017 \pm 0.02)\%$$

This has a maximum value of

$$\frac{\delta Q}{Q} = \pm 0.037\%$$

A2.1.3 Addition errors arising in flow metering

(i) Maximum error arising in reading flow meter scale = 1/4 division

$$\frac{\delta Q}{Q} = \pm \frac{0.25}{50} \times 100\% = \pm 0.5\%$$

(ii) Maximum error arising in reading of calibration chart

= 1/5 division

$$\frac{\delta Q}{Q} = \pm \frac{0.2}{100}\% = \pm 0.2\%$$

A2.1.4 Total maximum error

The maximum error from the different sources detailed in sections A2.1.1., A2.1.2, and A2.1.3 will be the sum of the individual maximum values

$$\therefore \quad \Sigma \frac{\delta Q}{Q} = \pm (0.2 + 0.5 \pm 0.037 \pm 0.0185)\%$$

$$= \pm 0.756\%$$

This makes the basic assumption that the calibration chart is correct. An identical set of errors will have been introduced, however, during the production of the calibration graphs and an additional 0.08% error will have been introduced by the use of the stop watch. The correct maximum flow metering error will be given by the following expression:-

$$\left(\frac{\delta Q}{Q}\right)_{\text{Maximum}} = \pm (2 \times 0.756 \pm 0.08)\% - \pm 1.5\%$$

A2.2 Evaluation of fuel concentration as a fraction of stoichiometric

$$X = W\left(\frac{Q_H + Q_P}{Q_A}\right) \qquad \text{(A2.10)}$$

where Q_H, Q_P = Volumetric flow rates of component fuel gases

Q_A = Volumetric flow rate of Air

W = Constant (stoichiometric ratio)

It may be shown that

$$\frac{\delta X}{X} = \frac{\delta Q_H + \delta Q_P}{Q_H + Q_P} - \frac{\delta Q_A}{Q_A} \qquad \text{(A2.11)}$$

From section A2.1 it follows that

$$\frac{\delta Q_H}{Q_H} = \frac{\delta Q_P}{Q_P} = \frac{\delta Q_A}{Q_A} = \pm 1.5\%$$

$$\therefore \quad \frac{\delta Q_H + \delta Q_P}{Q_H + Q_P} = \pm 1.5\%$$

$$\therefore \quad \left(\frac{\delta X}{X}\right)_{\text{Maximum}} \pm (1.5 \mp 1.5)\% = \pm 3.0\%$$

A2.3 Evaluation of critical boundary velocity gradient

Equation (3.5) may be rewritten as follows:-

$$gb = \frac{W(Q_H + Q_P + Q_A)}{R^3} \qquad \text{(A2.12)}$$

where W = Constant

R = burner radius

It may be shown that

$$\frac{\delta gb}{gb} = \frac{\delta Q_H + \delta Q_P + \delta Q_A}{Q_H + Q_P + Q_A} - 3 \frac{\delta R}{R} \qquad (A2.13)$$

By a similar argument to that presented in section A2.2 it follows that

$$\frac{\delta Q_H + \delta Q_P + \delta Q_A}{Q_H + Q_P + Q_A} \simeq \pm 1.5\%$$

R was evaluated by measurement of the burner internal diameter

$$\therefore \quad \frac{\delta R}{R} = \frac{\delta d}{d} \qquad (A2.14)$$

The maximum error arising from the internal diameter measurement = 10^{-5}m.

$$\therefore \quad \frac{\delta R}{R} = \pm \frac{10^{-5} \times 100\%}{1.54 \times 10^{-3}} = \pm 0.65\%$$

Substituting in equation (A2.13)

$$\left(\frac{\delta gb}{gb}\right)_{\text{Maximum}} \simeq \pm (1.5 \pm 3 \times 0.65\%)\% \cdot \pm 3.5\%$$

A2.4 Burning velocity determination

Equation (3.7) states as follows:-

$$Su = \frac{Q}{A} \qquad (3.7)$$

It may be shown that

$$\frac{\delta Su}{Su} = \frac{\delta Q}{Q} - \frac{\delta A}{A} \qquad (A2.15)$$

now

$$A = \frac{2\pi a\left(\sum_{i=1}^{n-2} r_i + \frac{r_{n-1}}{2}\right) + \pi b\,(r_{n-1} + r_n)}{F'^2}$$

$$= \frac{\Sigma}{F'^2} \qquad (A2.16)$$

The maximum error in the evaluation of Σ may be approximated by equation (A2.17)

$$\frac{\delta \Sigma}{\Sigma} = \frac{\delta a}{a} + \frac{\delta r}{r} \qquad \text{(A2.17)}$$

Maximum error in evaluation of a $= 10^{-5}\text{m}$

$$\therefore \quad \frac{\delta a}{a} = \pm \frac{10^{-5}}{10^{-3}} \times 100\% = \pm 1\%$$

Maximum error in evaluation of r (average value) $= 10^{-4}\text{m}$

$$\therefore \quad \frac{\delta r}{r} = \pm \frac{10^{-4}}{10^{-2}} \times 100\% = \pm 1\%$$

From equation (A2.17) Maximum error in $\Sigma = \frac{\delta \Sigma}{\Sigma} = \pm 2\%$

$$\text{Maximum in} \quad F' = \frac{\delta F'}{F'} = \frac{\delta D_P}{D_P} - \frac{\delta d}{d} \qquad \text{(A2.18)}$$

where D_P = Burner diameter measurement on photograph

$$\therefore \quad \text{Maximum} \quad \frac{\delta D_P}{D_P} = \pm \frac{10^{-4}}{2 \times 10^{-2}} \times 100\% = 0.5\%$$

From section A2.3 $\frac{\delta d}{d} = \pm 0.65\%$

From equation (A2.18) the Maximum value of $\frac{\delta F'}{F'} = \pm 1.15\%$

It may be shown from equation (A2.16) that

$$\frac{\delta A}{A} = \frac{\delta \Sigma}{\Sigma} - 2\frac{\delta F'}{F'} \qquad \text{(A2.19)}$$

the Maximum value of $\frac{\delta A}{A} = \pm (2 \pm 2 \times 1.15)\% = \pm 4.3\%$

$\therefore$ from equation (A2.15) $\left(\frac{\delta Su}{Su}\right)_{\text{Maximum}} = \pm (1.5 \pm 4.3)\% = \pm 6\%$

A2.5 Evaluation of critical flame stretch factor

$$K = \frac{gb\ k}{\rho C_p\ Su^2} \qquad (1.9)$$

It may be shown that

$$\frac{\delta K}{K} = \frac{\delta gb}{gb} + \frac{\delta k}{k} - \left(\frac{\delta \rho}{\rho} + \frac{\delta C_p}{C_p} + 2\frac{\delta Su}{Su} \right) \qquad (A2.20)$$

$\frac{\delta gb}{gb}$ and $\frac{\delta Su}{Su}$ have been determined in sections A2.3 and A2.4 .

$\frac{\delta k}{k}$ may be as much as $\pm$ 12% and this is discussed in more detail in Appendix 3.

For the purpose of this calculation it will be assumed that

$$\frac{\delta C_p}{C_p} = \frac{\delta \rho}{\rho} = \pm 2\%.$$

This is not unreasonable as the deviation from ideality in this respect is small.

It follows therefore that

$$\left(\frac{\delta K}{K}\right)_{Maximum} = \pm 3.5 \pm 12.0 - (\pm 2.0 \pm 2.0 \pm (2 \times 6.0))\%$$
$$= \pm 31.5\%$$

A2.6 Summary of Appendix 2

Parameter	Maximum Error ± %
Volumetric flow rate (Q)	1.5
Fuel concentration (X)	3.0
Critical boundary velocity gradient (gb)	3.5
Burning velocity (Su)	6.0
Critical flame stretch factor (K)	31.5

The figure quoted for the third, fourth and fifth parameters listed above do not take account of errors arising from no correction having been made for fluctuations in the temperature of the unburned gases. The influence on critical boundary velocity gradient and burning velocity of the unburned gas temperature has been studied by a number of workers (A2.2., A2.3., A2.4) although not for the gas mixtures studied in this investigation. It is reasonable to conclude that the inaccuracies from this source are unlikely to give to an inaccuracy in critical flame stretch factor of more than an additional ± 3%.

References

A2.1 Jensen, V.G., Jeffreys, G.V., Mathematical Methods in Chemical Engineering, Academic Press (1963).

A2.2 Dugger, G.L., Nat. Advisory Comm. Aeronaut. Tech. Note 2170 (1950).

A2.3 Grumer, J., Harris, M.E., J.Ind. & Eng. Chem. 46 , 2424 (1954).

A2.4 Dugger, G.L., Weast, G.R., Heimel, S., J. Ind. & Eng. Chem. 47 , 114 (1955).

Appendix 3.

CALCULATION OF PHYSICAL AND TRANSPORT PROPERTIES OF MULTICOMPONENT GAS MIXTURES.

Appendix 3.

Calculation of physical and transport properties of multicomponent gas mixtures.

A3.1 Density and specific heat

Densities and specific heats of multicomponent gas mixture were determined assuming ideal behaviour.

$$\rho_{\text{mixture}} = \sum_{i=1}^{n} x_i \rho_i \qquad \text{(A3.1)}$$

$$Cp_{\text{mixture}} = \sum_{i=1}^{n} x_i Cp_i \qquad \text{(A3.2)}$$

i refers to ith component gas

Cp = molal specific heat.

It was felt that the errors arising from the assumption of ideal behaviour would be small, an opinion expressed also by a number of other authors.

A3.2 Thermal conductivity

For the purpose of the calculation of thermal conductivity, the Wassiljewa equation was used in conjunction with the Lindsay and Bromley modification

$$k_{\text{mixture}} = \sum_{i=1}^{n} \frac{k_i}{1 + \sum\limits_{\substack{j=1 \\ j \neq i}}^{n} A_{ij} \left(\frac{x_j}{x_i}\right)} \qquad \text{(A3.3)}$$

where $$A_{ij} = \frac{1}{4}\left[1 + \left\{\frac{\mu_i}{\mu_j}\left(\frac{M_j}{M_i}\right)^{3/4} \frac{1 + \frac{S_i}{T}}{1 + \frac{S_j}{T}}\right\}^{1/2}\right]^2 \left(\frac{1 + \frac{S_{ij}}{T}}{1 + \frac{S_i}{T}}\right)$$

(A3.4)

and $S_{ij} = (S_i.S_j)^{1/2}$ for non polar gases (A3.5)

$S_{ij} = 0.733\ (S_i.S_j)^{1/2}$ for polar gases (A3.6)

μ = viscosity of pure component gas

S_i = Sutherland constant for pure component

S_{ij} = Sutherland interaction constant

$S_i = 1.5\ TB_i$ (A3.7)

where TB_i = boiling point of pure component i ($^\circ K$). S_i, however, is designated as equalling $79^\circ K$ where equation (A3.7) predicts a value lower than this.

In order to test the accuracy of this method of thermal conductivity determination, a study has been made of the Hydrogen-Propane-Air system by Sarkar (A3.1). He has reviewed the prediction methods available and has carried out experimental determinations using the hot-wire method first devised by Andrews (A3.2). A comparison of predicted and experimental values shows that in most cases the experimental value was greater than the predicted values sometimes by as much as 11%. Sarkar noted discrepancies as high as 20% but only in mixtures which could not be used to stabilise a laminar flame. Thus a maximum error of ± 12% has been used in Appendix 2 for assessment of the maximum error in critical flame stretch factor determination.

A3.3 Viscosity

The method of Wilke has been used to predict viscosities of multi-component gas mixtures.

$$\mu_{mixture} = \sum_{i=1}^{n} \frac{\mu_i}{1 + \sum_{\substack{j=1 \\ j \neq i}}^{n} \frac{x_j}{x_i} \phi_{ij}} \qquad (A3.8)$$

where

$$\phi_{ij} = \frac{\left[1 + \left(\frac{\mu_i}{\mu_j}\right)^{1/2} \left(\frac{M_j}{M_i}\right)^{1/4}\right]^2}{2\sqrt{2}\left(1 + \frac{M_i}{M_j}\right)^{1/2}} \qquad (A3.9)$$

Wilke has claimed an accuracy of ± 2.0% for this method.

A3.4 Diffusivity

Because a simple method of preferential diffusion factor calculation was required, the diffusivities used in these calculations were binary coefficients of the molecular species in question in the appropriate "Air". In all cases, the latter constituted more than 80% of the unburned gas mixture and so in the circumstances the assumption was not unreasonable.

Binary diffusion coefficients were calculated by the method of Gilliland and Maxwell

$$D_{ij} = \frac{0.0043\ T^{1.5}\left(\frac{1}{M_i} + \frac{1}{M_j}\right)^{1/2}}{P\left(V_i^{1/3} + V_j^{1/3}\right)^2} \qquad (A3.10)$$

P is measured in atmospheres

T is measured in deg.K.

V_i = molecular volume of component i which may be approximated by the application of Kopp's law of additive volumes

D_{ij} = binary diffusivity measured in cm^2/s.

The alternative method of Wilke and Lee based on the work of Hirschfelder, Bird and Spotz was considered for possible use. Perry (A3.3) has compared the accuracy of the two methods as follows:-

Method	Deviation (±%)	
	Average	Maximum
Gilliland et al	20	47
Wilke et al	4	20

Perhaps a more useful comparison has been made by Jacobs and Peeters (A3.4) and some of their results are included in the following table:-

Gas Pair	Diffusivity X 10^4 (M^2/s) at 20°C		
	Gilliland et al	Wilke et al	Experimental
Oxygen/Air	0.151	0.2158	0.196
Propane/Air	0.0966	0.1167	
Propane/Oxygen	0.09813	0.1148	
Hydrogen/Air	0.532	0.7204	0.674
Methane/Oxygen	0.1805	0.2662	
Ethylene/Air	0.1335	0.1635	
Water/Air	0.207	0.2587	0.243
Water/Oxygen	0.2021	0.2616	

From this table it is apparent that the values predicted by the method of Gilliland et al are generally smaller than those predicted by that of Wilke et al. The discrepancies between experimental and predicted values correspond closely to the average values quoted by Perry. Clearly the method of Wilke et al provides more accurate results but is more complex to apply. The use of Gilliland's method would provide values of preferential diffusion factor of comparable accuracy to that to which critical flame stretch factor can be determined and is therefore satisfactory for use in this investigation.

References

A3.1 Sarkar, S., MSc. Thesis (Process Analysis and Development) University of Aston (1972).

A3.2 Andrews, T., Proc. Royal Irish Acad. 1, 465 (1840).

A3.3 Perry, J.H., Chemical Engineer's Handbook, McGraw-Hill, 4th Edition (1963).

A3.4 Jacobs, T., Peeters, L., Vermant, J., Bull. des Soc. Chimiques Belges 79, 337 (1970)

Appendix 4.

ANALYSIS OF A TYPICAL SET OF BLOW-OFF DATA FOR A HYDROGEN-PROPANE-AIR MIXTURE.

Appendix 4.

Analysis of a typical set of blow-off data for a Hydrogen-Propane-Air mixture.

A4.1 Experimental readings

The following experimental readings were obtained in the manner described in section 3.

Run No. :- BV/6 Data Set No. :- 17.

Date :- 12-7-71

Indicated Atmospheric Pressure :- 762.80 mm.Hg.

Corrected Atmospheric Pressure :- 760.10 mm.Hg.

Burner No. :- 1

Air flow :- Flow meter No. :- F2
Reading :- 10.20
Outlet Pressure :- 0.15 in.w.g.
Outlet Temperature :- 24.0°C

Propane flow :- Flow meter No. :- F5
Reading :- 6.32
Outlet Pressure :- 0.12 in.w.g.
Outlet Temperature :- 23.9°C

Hydrogen flow :- Flow meter No. :- F8
Reading :- 5.00
Outlet Pressure :- 0.12 in.w.g.
Outlet Temperature :- 24.1°C

Final gas temperature :- 22.5°C

From the flow meter calibration charts the following flow rates were indicated:-

Air :- 3.085 l./min.

Hydrogen :- 0.1263 l./min.

Propane :- 0.0842 l./min.

A4.2 Analysis of Data

For the analysis of these data, two computer programs were used. Both were written for the University I.C.L. 1905 digital computer and in both cases the programming language was Algol. The first of these (P1) applied the relevant equations to obtain the corrected gas flow rates for the conditions at the burner inlet. The second (P2) calculated critical boundary velocity gradient, Reynolds Number, physical properties, fuel concentrations etc. from the corrected gas flow rates and the burner diameter. The printout from the second of these programs for the data presented in section A4.1 is given in Fig. A4.1.

```
MOLE FRACTION AIR       0.9366

MOLE FRACTION HYDROGEN        0.0381

MOLE FRACTION PROPANE        0.0253


FRACTION OF STOIC          0.740


THERMAL CONDUCTIVITY           0.039052   W/M SQUARED DEG C


DENSITY          1.161786   KG/M CUBED


BOUNDARY VELOCITY GRADIENT        2388.89    /S


SPECIFIC HEAT          1074.3643   J/KG DEG C


MOLE PERCENTAGE HYDROGEN IN FUEL          0.6004


HYDROGEN FLOW    2.0766666667& -6    M CUBED/S

PROPANE FLOW    1.3820000000& -6    M CUBED/S

AIR FLOW    5.1116666668& -5    M CUBED/S


VISCOSITY      0.000018557   N S/M SQUARED


REYNOLDS NUMBER         707.077
```

PRINTOUT FROM COMPUTER PROGRAM P2 FOR DATA PRESENTED IN SECTION A4.I.

FIG. A4.I

PROGRAM PI

```
'BEGIN''COMMENT'BIZLEY?CEPSFOI6,ZFLOWCALC;
'INTEGER'N,M,R,S;
'REAL'ROTEMP,PRESS,ATMOS,P,FINTEMP,CORR,F,NEWFLOW;
R:=0;
S:=READ;  (Number of sets of data)
FIRST:N:=READ;  (Number of flows per set of data)
M:=0;
ATMOS:=READ;  (Atmospheric pressure)
SECOND:PRESS:=READ;  (Flowmeter outlet pressure)
P:=PRESS*25.4/I3.6+ATMOS;
ROTEMP:=READ;  (Flowmeter outlet temperature)
ROTEMP:=ROTEMP+273.0;
F:=READ;  (Indicated flow)
CORR:=F*((760.0*ROTEMP/(29I.0*P))'**'0.5);
FINTEMP:=READ;  (Burner inlet temperature)
FINTEMP:=FINTEMP+273.0;
NEWFLOW:=CORR*ATMOS*ROTEMP/(P*FINTEMP);
OUTPUT(NEWFLOW);
NEWLINE(2);
M:=M+I;
'IF'M'GE'N'THEN''GOTO'THIRD;
'GOTO'SECOND;
THIRD:R:=R+I;
NEWLINE(5);
'IF'R'GE'S'THEN''GOTO'FINISH;
'GOTO'FIRST;
FINISH:'END';
```

PROGRAM P2

```
'BEGIN''COMMENT'BIZLEY,CEPSFOI6,ZANALYSIS;
'INTEGER'N,M,R,S;
'REAL'VPC,VAC,VHC,VASTOIC,X,KM,KONE,KTWO,KTHREE,AONETWO,AONETHREE,ATWOON
E,ATWOTHREE,ATHREEONE,ATHREETWO,T,MUONE,MUTWO,MUTHREE,SONE,STWO,STHREE,S
ONTWO,SONTHREE,STWTHREE,YONE,YTWO,YTHREE,VOL,WONE,WTWO,WTHREE,PRESS,DENS
,DIA,MASSAIR,MASSHY,MASSPRO,BOUN,SPHT,SPHY,SPAIR,SPPRO,PERC,FIONETWO,FIT
WOONE,FIONETHREE,FITHREEONE,FITWOTHREE,FITHREETWO,MUM,RE;
R:=0;
S:=READ;  (Number of sets of data)
```

```
FIRST:M:=READ;  (Number of blow-off determinations per set)
PRESS:=READ;  (Atmospheric pressure)
DIA:=READ;  (Burner diameter)
N:=0;
EXECUTE:T:=READ;T:=T+273.0;  (Temperature of unburned gases)
VAC:=READ;  (Corrected air flow)
VHC:=READ;  (Corrected hydrogen flow)
VPC:=READ;  (Corrected propane flow)
VASTOIC:=23.8*VPC+2.38*VHC;
X:=VASTOIC/VAC;
VOL:=VAC+VHC+VPC;
PROPANE:=STHREE:=347.7;
YTHREE:=VPC/VOL;
KTHREE:=0.00000038*(T'**'I.79);
MUTHREE:=0.00I47+(0.0000223*T);
WTHREE:=44.0;
RESTART:YONE:=VAC/VOL;
YTWO:=VHC/VOL;
SONE:=79.0;STWO:=79.0;
KONE:=0.000II35*(T'**'0.859);
KTWO:=0.000902*(T'**'0.839);
MUONE:=0.0054+(0.000042*T);
MUTWO:=0.003I3+(0.0000I935*T);
WONE:=28.8;WTWO:=2.0;
SONTWO:=(SONE*STWO)'**'0.5;
SONTHREE:=(SONE*STHREE)'**'0.5;
STWTHREE:=(STWO*STHREE)'**'0.5;
CALCULATE:AONETWO:=0.25*(I.0+((MUONE/MUTWO)*((WTWO/WONE)'**'0.75)*(I+(SO
NE/T))/(I+(STWO/T)))'**'0.5)'**'2.0*((I+(SONTWO/T))/(I+(SONE/T)));
AONETHREE:=0.25*(I.0+((MUONE/MUTHREE)*((WTHREE/WONE)'**'0.75)*(I+(SONE/T
))/(I+(STHREE/T)))'**'0.5)'**'2.0*((I+(SONTHREE/T))/(I+(SONE/T)));
ATWOONE:=0.25*(I.0+((MUTWO/MUONE)*((WONE/WTWO)'**'0.75)*(I+(STWO/T))/(I+
(SONE/T)))'**'0.5)'**'2.0*((I+(SONTWO/T))/(I+(STWO/T)));
ATWOTHREE:=0.25*(I.0+((MUTWO/MUTHREE)*((WTHREE/WTWO)'**'0.75)*(I+(STWO/T
))/(I+(STHREE/T)))'**'0.5)'**'2.0*((I+(STWTHREE/T))/(I+(STWO/T)));
ATHREEONE:=0.25*(I.0+((MUTHREE/MUONE)*((WONE/WTHREE)'**'0.75)*(I+(STHREE
/T))/(I+(SONE/T)))'**'0.5)'**'2.0*((I+(SONTHREE/T))/(I+(STHREE/T)));
ATHREETWO:=0.25*(I.0+((MUTHREE/MUTWO)*((WTWO/WTHREE)'**'0.75)*(I+(STHREE
/T))/(I+(STWO/T)))'**'0.5)'**'2.0*((I+(STWTHREE/T))/(I+(STHREE/T)));
EVALUATE:KM:=(KONE/(I+((AONETWO*(YONE/YTWO))+(AONETHREE*(YONE/YTHREE))))
```

```
)+(KTWO/(I+((ATWOONE*(YTWO/YONE))+(ATWOTHREE*(YTWO/YTHREE)))))+(KTHREE/(
I+((ATHREEONE*(YTHREE/YONE))+(ATHREETWO*(YTHREE/YTWO)))));
KM:=KM*I.73I;
WRITETEXT('('MOLE%FRACTION%AIR')');
SPACE(5);
PRINT(YONE,I,4);
NEWLINE(2);
WRITETEXT('('MOLE%FRACTION%HYDROGEN')');
SPACE(5);
PRINT(YTWO,I,4);
NEWLINE(2);
WRITETEXT('('MOLE%FRACTION%PROPANE')');
SPACE(5);
PRINT(YTHREE,I,4);
NEWLINE(3);
WRITETEXT('('FRACTION%OF%STOIC')');
SPACE(5);
PRINT(X,3,3);
NEWLINE(3);
WRITETEXT('('THERMAL%CONDUCTIVITY')');
SPACE(5);
PRINT(KM,3,6);SPACE(I);WRITETEXT('('W/M%SQUARED%DEG%C')');
NEWLINE(3);
MASSAIR:=YONE*28.8/(YONE*28.8+YTWO*2.0+YTHREE*44.0);
MASSHY:=YTWO*2.0/(YONE*28.8+YTWO*2.0+YTHREE*44.0);
MASSPRO:=YTHREE*44.0/(YONE*28.8+YTWO*2.0+YTHREE*44.0);
DENS:=(YONE*28.8/22400.0+YTWO*2.0/22400.0+YTHREE*44.0/22400.0)*(273.0/T)
*(PRESS/760.0);
DENS:=DENS*I000.0;
WRITETEXT('('DENSITY')');
SPACE(5);
PRINT(DENS,4,6);SPACE(I);WRITETEXT('('KG/M%CUBED')');
NEWLINE(3);
VOL:=VOL*I000.0/60.0;
BOUN:=4.0*VOL/((22.0/7.0)*((DIA/2.0)'**'3.0));
WRITETEXT('('BOUNDARY%VELOCITY%GRADIENT')');
SPACE(3);
PRINT(BOUN,6,2);SPACE(I);WRITETEXT('('/S')');
NEWLINE(3);
SPHY:=0.5*(6.047-0.0002*T+(4.808'I0'-7.0*(T'**'2.0)));
```

```
SPPRO:=(0.4I0+0.0647I*T-(2.258'I0'-7.0*(T'**'2.0)))/44.0;
SPAIR:=(0.2I*(6.I48+0.003I02*T-(9.23'I0'-7.0*T'**'2.0)))+0.79*(6.449+0.
00I4I3*T-(0.0807'I0'-7.0*(T'**'2.0))))/28.8;
SPHT:=MASSHY*SPHY+MASSAIR*SPAIR+MASSPRO*SPPRO;
SPHT:=SPHT*I000.0*4.I87;
WRITETEXT('('SPECIFIC%HEAT')');
SPACE(5);
PRINT(SPHT,6,4);SPACE(I);WRITETEXT('('J/KG%DEG%C')');
NEWLINE(3);
PERC:=VHC/(VHC+VPC);
WRITETEXT('('MOLE%PERCENTAGE%HYDROGEN%IN%FUEL')');
SPACE(5);
PRINT(PERC,3,4);
NEWLINE(3);
VPC:=VPC/60000.0;
VAC:=VAC/60000.0;
VHC:=VHC/60000.0;
WRITETEXT('('HYDROGEN%FLOW')');
SPACE(3);
PRINT(VHC,0,I0);SPACE(I);WRITETEXT('('M%CUBED/S')');
NEWLINE(2);
WRITETEXT('('PROPANE%FLOW')');
SPACE(3);
PRINT(VPC,0,I0);SPACE(I);WRITETEXT('('M%CUBED/S')');
NEWLINE(2);
WRITETEXT('('AIR%FLOW')');
SPACE(3);
PRINT(VAC,0,I0);SPACE(I);WRITETEXT('('M%CUBED/S')');
NEWLINE(3);
FIONETWO:=(I+((MUONE/MUTWO)'**'0.5)*((WTWO/WONE)'**'0.25)'**'2.0)/((8.0'
**'0.5)*(I+(WONE/WTWO))'**'0.5);
FITWOONE:=FIONETWO*(MUTWO/MUONE)*(WONE/WTWO);
FIONETHREE:=(I+((MUONE/MUTHREE)'**'0.5)*((WTHREE/WONE)'**'0.25)'**'2.0)/
((8.0'**'0.5)*(I+(WONE/WTHREE))'**'0.5);
FITHREEONE:=FIONETHREE*(MUTHREE/MUONE)*(WONE/WTHREE);
FITWOTHREE:=(I+((MUTWO/MUTHREE)'**'0.5)*((WTHREE/WTWO)'**'0.25)'**'2.0)/
((8.0'**'0.5)*(I+(WTWO/WTHREE))'**'0.5);
FITHREETWO:=FITWOTHREE*(MUTHREE/MUTWO)*(WTWO/WTHREE);
MUM:=(MUONE/(I+FIONETWO*(YTWO/YONE)+FIONETHREE*(YTHREE/YONE)))+(MUTWO/(I
+FITWOONE*(YONE/YTWO)+FITWOTHREE*(YTHREE/YTWO)))+(MUTHREE/(I+FITHREEONE*
```

```
(YONE/YTHREE)+FITHREETWO*(YTWO/YTHREE)));
MUM:=MUM/1000.0;
WRITETEXT('('VISCOSITY')');
SPACE(5);
PRINT(MUM,1,9);SPACE(1); WRITETEXT('('N%S/M%SQUARED')');
NEWLINE(3);
RE:=VOL*DENS*28.0/(DIA*MUM*220000.0);
WRITETEXT('('REYNOLDS%NUMBER')');
SPACE(5);
PRINT(RE,5,3);NEWLINE(3);
TERMINATE:N:=N+1;
'IF'N'GE'M'THEN''GOTO'TWO;
'GOTO'EXECUTE;
TWO:R:=R+1;NEWLINE(8);
'IF'R'GE'S'THEN''GOTO'FINISH;
'GOTO'FIRST;
FINISH:'END';
```

Appendix 5.

"THE INFLUENCE OF PREFERENTIAL DIFFUSION
ON THE BLOW-OFF OF LAMINAR AERATED BURNER
FLAMES" - A PAPER PRESENTED AT THE EUROPEAN
SYMPOSIUM ON COMBUSTION IN SHEFFIELD, ENGLAND
ON SEPTEMBER 20TH 1973.

THE INFLUENCE OF PREFERENTIAL DIFFUSION ON THE BLOW OFF OF LAMINAR AERATED BURNER FLAMES

R.G. Temple and D.O. Bizley
Department of Chemical Engineering
University of Aston in Birmingham

Introduction

The concept of flame stretch was first applied to the analysis of blow off data by Lewis and Von Elbe (1). Reed (2) has applied the same concept to characterise the blow off of stabilised laminar, aerated burner flames by means of equation (i)

$$K = \frac{gb\, k}{\rho\, c_p\, Su^2} = \frac{gb\, \eta_o}{Su} = 0.23(1+(x^{6.4}-1)\alpha)$$

$$\text{for } x<1.36 \qquad \text{(i)}$$

where K = flame stretch factor
gb = critical boundary velocity gradient
η_o = preheat zone thickness
ρ = density
c_p = specific heat
k = thermal conductivity
Su = burning velocity
x = fuel concentration expressed as a fraction of stoichiometric
α = 0 for flames with a primary reaction zone only
= 1 for flames with both a primary and a secondary reaction zone

He found, however, that it is not possible to characterise Hydrogen-Air flames by this general correlation and attributed this behaviour to preferential diffusion. By this mechanism, the concentration of a light molecular species may be elevated in a region of a flame in which the flow lines diverge from one another while passing through the flame front. Such a situation exists in the stabilising region of a laminar aerated flame. This explanation has by no means gained universal acceptance. It is the purpose of this work therefore to clarify and characterise this mechanism.

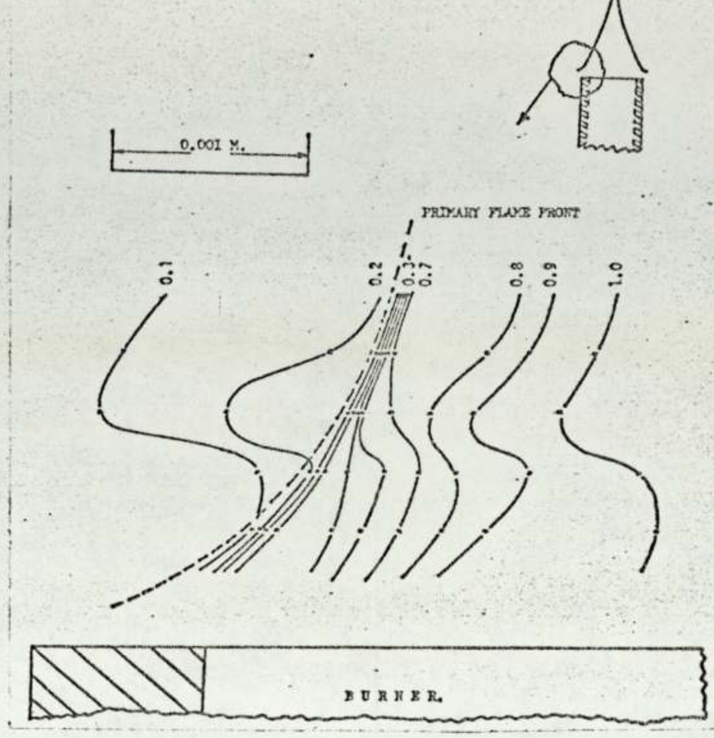


Fig. 1 Concentration profiles in stabilising region of 1.0 stoichiometric flame. Concentrations expressed as a fraction of unburned gas concentration

Evidence for Existence of Preferential Diffusion

Hydrogen concentration profiles have been plotted in the stabilising region of a Hydrogen-Propane-Air flame with 60% H_2 in the fuel mixture and degrees of aeration equivalent to 0.8, 1.0 and 1.2 Stoichiometric. The flames were stabilised on a 0.00782m diameter cylindrical burner. Sampling was carried out by means of a quartz microprobe system similar to that described by Fristrom and Westenburg (3) and samples were analysed with a gas chromatograph. Concentrations were expressed as a ratio of the local concentration to that in the unburned gases and Fig. 1. shows a typical set of profiles for the 1.0 stoichiometric flame. There is strong evidence of an increase of secondary combustion in the stabilising region which is indicative of an increase in fuel concentration attributable to preferential diffusion. Similar indications were found in the 0.8 and 1.2 Stoichiometric flames.

Characterising of Preferential Diffusion Process

In an earlier paper (4), the use of a dimensionless group termed the preferential diffusion factor (F) to characterise the degree to which preferential diffusion is likely to occur, has been discussed. The group was derived using a dimensional approach and may be considered as the product of two Lewis Numbers and two concentration terms

$$F = \frac{D_1X_1}{\eta_o^2}\,\frac{D_2X_2}{Su^2} = \left(\frac{D_1}{\eta_o Su}\right)\left(\frac{D_2}{\eta_o Su}\right) X_1\ X_2 \qquad \text{(ii)}$$

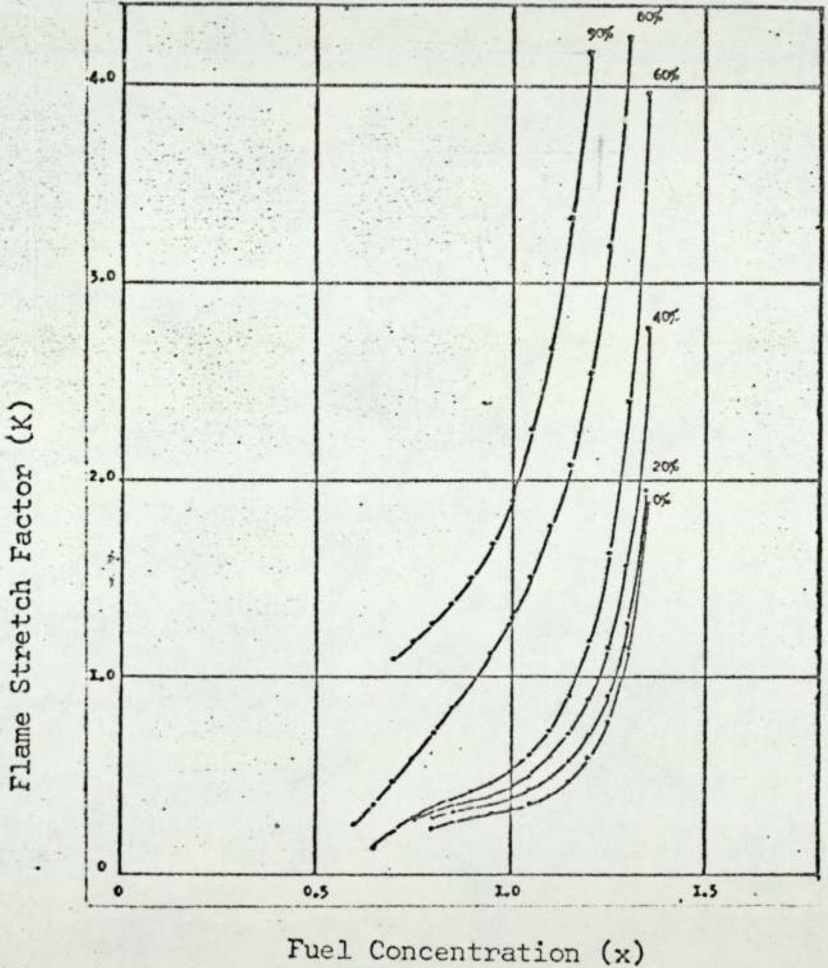


Fig. 2. Flame Stretch Factor - Fuel concentration correlations for fuel gas mixtures containing 20, 40, 60, 80 and 90% Hydrogen

D = Diffusivity
X = mole fraction

Subscripts 1 and 2 refer to light molecular species in the unburned gases and combustion products respectively.

The various physical and transport properties are evaluated at the temperature of the unburned gases as this region will provide a greater resistance to mass transfer than the hotter regions of the flame front.

The blow off characteristics of a typical system, Hydrogen-Propane-Air have been studied with fuel gas mixtures of 0, 20, 40, 60, 80 and 90% Hydrogen present, using cylindrical burners of sufficient length to ensure a fully developed laminar velocity profile. Burning velocity data has also been obtained using a "total area" method employing a Schlieren optical system.

These data have been analysed on a flame stretch basis (Fig. 2.) and it has been found possible to characterise the correlations obtained by the relationship in equation (iii)

$$K = K' e^{CF} \qquad 0.7 < x < 1.3 \tag{iii}$$

where K' is the flame stretch factor determined experimentally for the situation in which no preferential diffusion is expected and C may be expressed empirically as follows

$$C = 38.6 + (1-\alpha)\left(\frac{2.64}{x - 0.615} - 6.85\right) \tag{iv}$$

In this system, the subscript 1 represents Hydrogen and 2 represents water, the concentration of which has been evaluated assuming complete combustion of as much fuel as there is sufficient oxygen for. The average discrepancy between critical flame stretch factors predicted by these correlations and those determined experimentally was less than ±7% of the experimental value.

Evaluation of preferential diffusion factors for a variety of gases has indicated that Hydrogen is the only fuel gas of sufficiently low molecular weight to influence blow off characteristics to any significant degree. It seems likely, however, that it may account for the minor variations observed by Edmondson and Heap in their study of the blow off of a number of aerated hydrocarbon flames (5).

In order to vary the transport properties of the Hydrogen-Propane-Air System, the Nitrogen present in the Air has been replaced by both Helium and Argon. In both cases, fuel gas mixtures containing 0, 40 and 80% Hydrogen were studied and flame stretch correlations obtained in a similar manner to that already described. In the case of Argon the average discrepancy between experimental and predicted critical flame stretch factors was ±10.4% and the maximum discrepancy 34.2%. In the case of Helium these figures were ± 18.1% and 42.7% respectively.

It is perhaps worth noting that when using both Helium and Argon, the flame stretch factors obtained when no Hydrogen was present in the fuel were substantially lower than those that were predicted by Reed's theory, and this provides a further limitation to the applicability of the flame stretch theory.

In order to assess whether the correlations are applicable to all gas mixtures containing Hydrogen, the experimentally-determined blow off data of Grumer and Harris (6) and the burning velocity data of Scholte and Vaags (7) for the Hydrogen-Methane-Air system have been analysed on a flame stretch basis. An average discrepancy of ± 35% of the experimental values has been found between experimental and predicted critical flame stretch factors. It should be pointed out that Reed has already indicated that flame stretch factors may be determined only to an accuracy of ± 60%.

Conclusions

(1) These results indicate that preferential diffusion occurs in the stabilising region of laminar, aerated flames.
(2) For practical purposes, equations (iii) and (iv) may be used to predict blow off characteristics even in situations where preferential diffusion is likely to occur. For normal aerated systems K' may be approximated by equation (i) in which case

$$gb \doteq 0.23 \frac{\rho\, c_p\, Su^2}{k} (1 + (x^{6.4}-1)\,\alpha)\, \exp(\{38.6 + (1-\alpha)\left(\frac{2.64}{x-0.615} - 6.85\right)\}F) \text{ for } 0.7 < x < 1.3 \qquad (v)$$

Acknowledgements

The authors would like to thank the Gas Council for the provision of a scholarship for one of them (D.O.B.) which has enabled this work to be carried out, and to Dr. S.B. Reed and Dr. S. Radcliffe and their colleagues for the interest they have shown in this work.

References

(1) Lewis, B, Von Elbe G., Combustion Flames and Explosions in gases, Academic Press (2nd Edition 1961)
(2) Reed, S.B., J.I. Gas E. 8, 157 (1968)
(3) Fristrom R.M., Westenburg A.A., Flame Structure, McGraw Hill (1965)
(4) Temple, R.G., Bizley, D.O., - paper accepted for publication by J.Inst. Fuel.
(5) Edmondson, H., Heap, M.P., J.I. Gas E. 11, 305 (1971)
(6) Grumer, J., Harris M.E., J.Ind. & Eng.Chem. 44, 1547 (1952)
(7) Scholte, T.G., Vaags, P.B., Comb. & Flame 3, (4), 511 (1959)

Appendix 6.

"THE INFLUENCE OF HYDROGEN ON THE BLOW-OFF OF LAMINAR AERATED BURNER FLAMES" - A PAPER ACCEPTED FOR PUBLICATION BY THE JOURNAL OF THE INSTITUTE OF FUEL.

The Influence of Hydrogen on the Blow-off of Laminar Aerated Burner Flames

R.G. Temple, B.Sc.,Ph.D.,C.Eng.,F.Inst.F.,M.I.Chem.E., &
D.O. Bizley, B.Sc. (Department of Chemical Engineering,
The University of Aston in Birmingham).

1. Nomenclature

C - Parameter defined in equation (viii)
Cp - Specific heat of unburned gas mixture (J/kg.Deg.C.)
d - Burner diameter (m)
D - Molecular diffusivity (m^2/s)
F - Preferential diffusion factor as defined in equation (iv)
gb - Critical boundary velocity gradient (s^{-1})
k - Thermal conductivity of unburned gas mixture ($W/m^2Deg.C.$)
K' - Critical flame stretch factor for system in which no significant preferential diffusion occurs
K - Critical flame stretch factor for any system
Le - Entry length (m)
Q - Volumetric flowrate (m^3/s)
Re - Reynolds Number
Su - Burning velocity (m/s)
v - Local gas velocity (m/s)
x - Concentration expressed as mole fraction
X - Fuel concentration expressed as a fraction of stoichiometric
α - Parameter defined in equation (i)
η_o - Preheat zone thickness as defined in equation (ii)
ρ - Density of unburned gas mixture (kg/m^3)

2. Introduction

Some six years ago Reed proposed the Flame Stretch Theory of blow-off (1,2). The theory proposed that blow-off occurred as a result of excessive aerodynamic quenching, or "flame stretch", in the stabilising region of a flame. It further suggested that it was possible to predict when blow-off of any aerated burner flame would occur from the following equation:

$$K = \frac{gb\ \eta_o}{Su} = 0.23\left[1 + (X^{6.4} - 1)\,\alpha\right] \qquad \text{(i)}$$

$$0.7 < X < 1.36$$

where

$\alpha = 0$ for flames with a primary reaction zone only

$\alpha = 1$ for flames with both a primary and secondary reaction zone

and

$$\eta_o = \frac{k}{\rho C p S u} \qquad \text{(ii)}$$

There is no theoretical reason for the imposition of the lower limit of applicability in equation (i) and the limit merely defines the leanest flames for which experimental data was available for testing the validity of the equation.

The theory in no way invalidates experimentally determined correlations of critical boundary velocity gradient against fuel concentration, but did succeed in explaining the anomalies of the Boundary Velocity Gradient Theory of Lewis and Von Elbe (3). Edmondson and Heap (4) later showed that each fuel gas had its own unique critical flame stretch factor-fuel concentration relationship, but that the deviation from equation (i) was so small as to be comparatively insignificant from the burner designer's point of view.

Reed (1) has also shown, however, that the Hydrogen-Air System does not relate closely to the general flame stretch correlation. He has suggested that preferential diffusion, the phenomenon which is responsible for the formation of cellular and polyhedral flames, can also account for the anomalous blow-off behaviour of this system. Both Edmondson and Heap (5) and Blandon (6) have criticised this suggestion and the former workers have proposed the alternative explanation of 'intermixing with the surrounding atmosphere'. Work by Reed (7), Datta et al (8) and Smith (9) has shown that this alternative explanation is unlikely to be correct.

With the introduction of Natural Gas in Britain, interest has been shown in Synthetic Natural Gases which may contain significant concentrations of Hydrogen. It is important that the burner designer should be able to predict the blow-off characteristics of such gases without needing to carry out the time consuming work required to obtain an experimentally determined critical boundary velocity gradient correlation. This paper presents an attempt to characterise the influence of Hydrogen on the blow-off of laminar, aerated flames by redefining the flame stretch factor to incorporate a term characterising the tendency for preferential diffusion to occur.

3. The Effect of Preferential Diffusion on Laminar Flames.

An understanding of the mechanism by which preferential diffusion occurs is essential if one is to characterise this process. Let us consider a flame front which meets the bulk gas flow at an angle β (Fig.1). Then for preferential diffusion to occur it is necessary for one or more of the species present in the combustion products to diffuse against the bulk gas flow into the preheat zone of the unburned gases. The tendency for this to occur will be greater for a mobile species, like water vapour, than for instance Carbon Dioxide. Concentration profiles plotted by Datta et al (8) have shown that such fluxes do occur in practice. The presence of any such species in the preheat zone will cause a concentration gradient across the unburned gas at right angles to the flame front of all species present in the unburned gas mixture. The most mobile of these will diffuse towards the flame front (Fig.2). It is, of course, true that a concentration gradient will exist along the flow lines, but any diffusion in this direction will cause no overall concentration changes along the flame front. It is equally true that along a section of flame front of constant angle β, the loss of mobile species from a flow line will be exactly balanced by a gain from the adjacent flow lines. Thus there will be no concentration variations along the flame front. In practice this situation is unlikely to exist as preheating of the unburned gas will lead to distortion of the flow lines increasing β as the flame front is approached. In a region of flame front curvature, however, as is present in the stabilising region of a flame, there will be an increase in the concentration of the mobile species at the point of stability (Fig.3) caused by diffusion from adjacent flow lines. It may be noted that flame front curvature need not in theory be present for preferential diffusion to occur provided a suitable flow régime exists. Similarly, if all flow lines meet a region of flame front curvature at right angles, again no preferential diffusion will occur. The point of stability is the only point at which the local unburned gas velocity (v) is exactly equal and opposite to the burning velocity (Su). At all other points on the flame front the former exceeds the latter such that Su = v Sin β. It follows that blow-off of a flame is directly governed by the gas composition at the point of stability.

An increase in concentration of certain species at the point of stability to a value greater than that present in the bulk of the unburned gases may cause a change in burning velocity and possibly heat transfer from the secondary reaction zone to the primary flame front. Neither of these are taken into account in the normal evaluation of the flame stretch factor. It has already been pointed out that preferential diffusion is often associated with the formation of polyhedral or cellular flame fronts. From the above description, however, it may be concluded that the absence of such flame fromts does not automatically proclude the existence of preferential diffusion.

Several workers have tried to take preferential diffusion into account in their mathematical models of flame fronts. These include Markstein (10), Eckhaus (11) and Parlange (12). They all make such drastic simplifying assumptions, however, that their results, while being of great qualitative interest, do not provide us with a means of quantitatively assessing the effect of preferential diffusion in the stabilising region of a laminar flame. Parlange's model is perhaps the most sophisticated of those available. He has not only studied the case of a light species present in excess or deficiency in the unburnt gases, but also that of a light species formed as a product of combustion. In the latter case he has shown that conduction plays a very similar role to that played by diffusion in the former cases. It may further be pointed out that until we are able to fully characterise the kinetics of the chemical reactions taking place within the flame front, it will be unlikely that we can predict the concentrations of the various molecular species present with sufficient accuracy to adequately characterise the diffusion processes. It may also be concluded that any relationships which could be derived from such a mathematical model would be likely to be of such complexity as to render them useless to the designer.

It therefore seemed a more profitable approach to the problem, in the light of the available knowledge of the processes involved, to develop a dimensionless group which would characterise the tendency for preferential diffusion to occur and to attempt to correlate this with experimentally determined blow-off data.

The degree to which preferential diffusion will affect the flame stability will be related to the following parameters:-

(i) Diffusivity of mobile combustion products in the unburned gas mixture (D_1).

(ii) Concentration gradient of the above species. This will be related to the concentration (x_1) of these species in the combustion products. For this to be evaluated, perfect combustion is assumed.

(iii) Diffusivity of mobile species in the unburned gases (D_2)

(iv) Concentrations of these species in the unburned gases (x_2)

(v), (vi), (vii) Density, specific heat, thermal conductivity of unburned gas mixture (ρ, Cp, k)

Let the degree to which preferential diffusion will alter the concentration of a mobile species in the stabilising zone be characterised by a dimensionless group (F) termed the preferential diffusion factor. By the "π" method of dimensional analysis it may be shown that

$$F \quad \alpha \left(\frac{D_1}{k/\rho Cp}\right)^{n_1} {x_1}^{n_2} \left(\frac{D_2}{k/\rho Cp}\right)^{n_3} {x_2}^{n_4} \qquad \text{(iii)}$$

where n_1, n_2, n_3 and n_4 are the powers to which the various dimensionless groups are raised. It is clear that all four of these powers will be positive. However, to take the analysis further it has been necessary to make the following additional assumptions:-

(i) As the second and fourth dimensionless quantities in equation (iii) are closely associated with those immediately preceding them it has been assumed that $n_2 = n_1$ and $n_4 = n_3$.

(ii) The relationship between n_1 and n_3 could be found experimentally if x_1 and x_2 could be varied independently. As this is not possible, however, it has been assumed that $n_1 = n_3 = 1$. F has thus been defined as follows:-

$$F = \left(\frac{D_1 x_1}{k/\rho Cp}\right)\left(\frac{D_2 x_2}{k/\rho Cp}\right) \qquad \text{(iv)}$$

It may be seen that F can be considered as the product of two Lewis Numbers and two concentration terms. The Lewis number is the ratio of molecular to thermal diffusivities. Thermal diffusivity in this case refers not to diffusion of molecules caused by a thermal driving force but rather the analogous heat transfer process to mass transfer by molecular diffusion.

Our object was to relate the value of F to a change in stability. It was, therefore, decided to attempt to relate F directly to Critical Flame Stretch Factor as the latter was the only variable which could take account of the changes of both burning velocity and heat transfer to the primary flame front.

It is necessary to identify the molectular species most likely to affect the process. For the majority of cases,the species most likely to diffuse back into the unburned gases is water vapour. The only species likely to diffuse in the unburned gases will be a low molecular weight gas (e.g. Hydrogen, Ethylene, Acetylene, Methane). In practice only Hydrogen is found to diffuse to a sufficient degree to cause a marked change in stability in an aerated flame. It does follow, however, that all flames are subject to preferential diffusion phenomena to a greater or lesser degree and this is reflected in the series of closely related lines plotted by Edmondson and Heap (4,5) of critical flame stretch factor against fuel concentration on cylindrical burners for different fuel gases.

4. Experimental Work.

To relate flame stretch factor to preferential diffusion factor it was necessary to study the progressive influence of the addition of Hydrogen to a system which was known to obey fairly closely the general flame stretch correlation. For this purpose the Propane -Air system was chosen. Although limited data is available for both critical boundary velocity gradient at blow-off and burning velocity for the Hydrogen-Propane-Air system from the literature it was felt that it was essential that experimentally consistent data should be determined. This was because the absolute accuracy of flame stretch factor is normally only ± 60% and to base conclusions on such inaccuracies would be hazardous. If consistent data is used, however, it is possible to reduce the variation within the data to ± 20% and to draw reasonable conclusions even though the absolute accuracy is not increased beyond the original figure of ± 60%.

Fuel gases were supplied from high pressure cylinders and air was supplied from a central high pressure main. The gases were dried using silica gel and metered through variable area flow meters which were accurately calibrated. Temperature control was not employed but temperature was closely monitored both at the flow meter outlets and the burner inlet and the latter temperature was found to remain within the range 293^0 K ± 3^0. Thus it was estimated that flow metering accuracy was to within ± 1.5%. Four cylindrical burners of diameters varying between 0.008 and 0.0015 m were used for blow-off determinations. These were constructed of sufficient length to ensure a fully developed laminar velocity profile and thus enable accurate determination of the critical boundary velocity gradient. The entry length Le in a cylindrical tube for fully developed laminar flow is given by the equation

$$Le = 0.035\ d.Re \qquad \text{(v)}$$

Re = Reynolds Number, the maximum value of which will be ≈ 2000
The maximum value of Le is given by

$$Le = 70.d \qquad \text{(vi)}$$

The burners were situated in a 0.175 m diameter tower up which the secondary atmosphere was passed. A layer of glass ballotini supported on wire gauze was placed above the secondary atmosphere distributor at the base of the tower to ensure uniform flow distribution. Two diametrically opposed plate glass windows were fitted to facilitate the viewing of the flame. The critical flow rate was determined by presetting the required fuel flow rate and Propane/Hydrogen ratio and increasing the primary air flow until blow-off occurred. The procedure was repeated several times to ensure reproducibility. The boundary velocity gradient was predicted from the formula

$$gb = \frac{32Q}{\pi d^3} \qquad \text{(vii)}$$

The onset of turbulence could be observed visually but as an additional safeguard, Reynolds Number was determined for each measurement and cases rejected where Re > 1800. Fuel mixtures with 0, 20, 40, 60, 80, 90, 95 and 100% Hydrogen in the fuel were tested and the results obtained are shown in Fig.4. Over the ranges applicable these agree fairly closely with the results obtained by Reiter and Wright (12). There is little agreement, however, with the curves that can be predicted by the method of Van Krevelen and Chermin (14). Throughout the determination of critical boundary velocity gradients, care was taken to avoid data obtained from non-isotropic, polyhedral flames. These structures were encountered in fuel-rich flames with high Propane concentrations and fuel-lean flames where high Hydrogen concentractions were present, but it was sometimes found possible to obtain data for an isotropic flame of comparable composition by using a burner of a different diameter. Although such non-isotropic flames are of considerable interest, it was recognised that they might serve to mask the overall behaviour of the more normally encountered isotropic flames.

In the choice of a method of burning velocity determination the two chief considerations were accuracy and speed of determination. The latter was essential as the number of determinations was considerable to enable flame stretch factors to be computed for all the blow-off data obtained. It was decided to use a total area burner method, the schlieren image of the cone being used to determine the area of the flame front. The optical system consisted of a high pressure mercury arc, a collimator, a biconvex lens and a pinhole to produce a point source, two schlieren concave mirrors, and a schlieren knife-edge.

This equipment was mounted on two 2m. optical benches placed either side of the tower in which the burner was situated. The image of the flame was projected directly onto the film of a single lens reflex camera from which the lens had been removed. Enlargements were made from the photographs obtained and from these the total area of the schlieren cone was measured by the method described by Senior (15). This consists of treating the cone as comprising of a large number of frustra. Although some work was carried out using a nozzle burner, more consistent burning velocity data was obtained using the largest of the cylindrical burners and thus the latter was employed. It is known that this method of burning velocity determination gives results which are slightly too low, but this was considered acceptable for the use to which the data was to be put. Data of Guenther and Jarisch (16) and Edmondson and Heap (17)were used for the Hydrogen-Air system as the cylindrical burner employed was not suitable for the high burning velocities present in this system. The results are shown in Fig.5. The graph of maximum burning velocity against percentage Hydrogen present demonstrates the effect of progressive introduction of Hydrogen (Fig.6).

5. Flame Stretch Analysis

Critical flame stretch factors as defined in (i) and (ii) were computed for each of the systems studied. Densities and specific heats were determined assuming ideal gas mixtures. Thermal conductivities were determined using the Wassiljewa equation applied using the method of Lindsay and Bromley. The results obtained are shown in Fig.7.

Preferential diffusion factors were determined for each of the systems studied. The following assumptions have been made:-

(i) D_1 = diffusivity of water vapour in Air
(ii) D_2 = diffusivity of Hydrogen in Air

Concentrations of other gases are low relative to Air in all cases. D_1 and D_2 are evaluated at the unburned gas temperature as at this low temperature their values will be at their lowest and so this low temperature region will provide the controlling resistance to mass transfer.

(iii) x_1 = concentration of water vapour in the combustion products assuming perfect primary combustion
(iv) x_2 = concentration of Hydrogen in unburned gases
(v) k, ρ, C_p are unaffected and retain the values used in flame stretch factor determination.

Diffusivities have been computed by the methods of Gilliland and Maxwell, and Wilke and Lee based on the method of Hirschfelder, Bird and Spotz. A comparison of these methods together with experimentally determined values of diffusivities has been given by Jacobs et al (18).

It has been found possible to relate Preferential Diffusion Factor to experimentally determined values of Critical Flame Stretch Factor by relationships of the type (Fig.8):-

$$K = K' e^{CF} \qquad \text{(viii)}$$
$$\text{where } C = f(X)$$

The average discrepancy bdetween experimental values and those predicted by the above correlations has been calculated as ±6.93% of the experimental value.

It has already been stated that for design purposes K' may be approximated by equation (i). C may be expressed empirically by the relationship

$$C = 38.6 + (1 - \alpha) \frac{2.64}{(X - 0.615)} - 6.85 \qquad \text{(ix)}$$

Substituting equation (viii), the following equation is obtained

$$K = 0.23 \left[1 + (X^{6.4} - 1)\alpha\right] \exp \left[\left\{38.6 + (1 - \alpha)\left(\frac{2.64}{(X - 0.615)} - 6.85\right)\right\} F\right] \qquad \text{(x)}$$
$$0.7 < X < 1.3$$

The designer normally wishes to determine the critical flow rate and so requires to know the critical boundary velocity gradient which is expressed by the equation

$$gb = 0.23\,\frac{\rho CpSu^2}{k}\left[1+(X^{6.4}-1)\alpha\right]\,\exp\left[\left\{38.6+(1-\alpha)\left(\frac{2.64}{(X-0.615)}-6.85\right)\right\}F\right] \qquad (xi)$$

$$0.7 < X < 1.3$$

The value of C decreases as fuel concentration increases until stoichiometry is reached above which point it remains at a constant value. It is suggested that this behaviour reflects the fundamentally differing controlling mechanisms in these two regions. In fuel-lean flames the increased Hydrogen concentration alters the reaction rates in the primary flame front thus enhancing burning velocity. The influence is complex and it would not be anticipated that it could be characterised by a simple relationship. In fuel-rich flames, the controlling mechanism is one of heat conduction and it is not unreasonable that a particular value of preferential diffusion factor gives rise to an increase in critical flame stretch factor independent of fuel concentration. Fig.9 shows C as a function of X.

6. A Further Test of the Modified Flame Stretch Factor

In order to test the general validity of the modified flame stretch factor as a means of characterising the influence on blow-off of Hydrogen, the blow-off data of Grumer and Harris (19) and the burning velocity of Scholte and Vaags (20) for the Hydrogen-Methane-Air system have been analysed. The blow-off data was thought to be a little suspect as this, together with data for Hydrogen-Propane-Air obtained by the same authors, has been used in the derivation of the prediction method of Van Krevelen and Chermin (21). This has been shown to predict low values of critical boundary velocity gradient for the Hydrogen-Propane-Air system which suggests possible discrepancies in the data from which this empirical correlation was derived. The burning velocity data was believed to be fairly reliable as data obtained for different systems by the same authors on the same apparatus agree fairly closely with currently accepted literature values.

Predicted and practically determined values of flame stretch factor were computed and compared. The average discrepancy between the two values was found to be ± 35% of the predicted value. Reed (1) suggests that flame stretch factors may be calculated to an accuracy of only ± 60%, and the agreement between the predicted and experimental values is well within these limits. In addition it may be pointed out that where large discrepancies between these values occur, they do so in regions where burning velocity varies rapidly with fuel concentration and thus where small flow metering errors have the greatest effect on flame stretch factor. It is felt, therefore, that this work lends support to the validity of the modified flame stretch factor concept.

7. Conclusions

1. A study has been made of the effect of increasing Hydrogen concentration on the flame stretch factor-fuel concentration relationship for Hydrogen-Propane-Air mixtures.

2. A dimensionless group which characterises the degree to which preferential diffusion is likely to occur has been developed. This is termed the Preferential Diffusion Factor and incorporates the parameters most likely to affect the preferential diffusion process.

3. This dimensionless group has been used to characterise the deviation of the Flame Stretch factor from its predicted value by the flame stretch theory for the Hydrogen-Propane-Air System.

4. A modified flame stretch factor has been developed which should be capable of predicting the blow-off behaviour for all aerated laminar flames in which Hydrogen is present, although the validity of this conclusion has yet to be tested experimentally.

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References

1. Reed, S.B., J.I.Gas E., 8, 157 (1968),
"A unifying theory for the blow-off of aerated burner flames".

2. Reed, S.B., Comb & Flame 11, 177 (1967),
"Flame stretch - a connecting principle of blow-off data".

3. Lewis, B., Von Elbe, G.,
"Combustion Flames and Explosions of Gases", 2nd Edition, Academic Press (1961).

4. Edmondson, H., Heap, M.P., Comb & Flame 15, 179 (1970),
"The correlation of burning velocity and blow-off data by the Flame Stretch concept".

5. Edmondson, H., Heap, M.P., J.I.Gas E. 11, 305 (1971),
"The blow-off of burner flames".

6. Blandon, E.E., Discussion contribution to Ref. (1).

7. Reed, S.B., Discussion contribution to Ref. (5).

8. Datta, P., Hayward, B.M., Reed, S.B., Comb. & Flame 17, 399 (1971),
"A flame structure study of the stabilising region of a near stoichiometric laminar burner flame".

9. Smith, R.A., Discussion in contribution to Ref. (5).

10. Markstein, G.H., "Non steady flame propagation", Pergamon, Oxford (1964).

11. Eskhaus, W., J. Fluid Mech. 10, 80 (1961),
"Theory of flame front stability".

12. Parlange, J.Y., J. Chem. Phys., 48, (4) 1843 (1968),
"The influence of preferential diffusion on the stability of a laminar flame".

13. Reiter, S.H., Wright, C.C., Ind. & Eng. Chem., 42, 691 (1950),
"Stability of burner flames with propane hydrogen mixtures".

14. Van Krevelen, D.W., Chermin H.A.G., 7th Int. Symposium on Combustion, Oxford (1958).

15. Senior, D.A., Comb. & Flame, 5, 7 (1961),
"Burning velocities of Hydrogen-Air and Hydrogen-Oxygen mixtures".

16. Guenther, R., Jarisch, G., Reported by Dixon Lewis G. et al,
Inst. Gas Eng. Research Communication 861.

17. Edmondson, H., Heap, M.P., Comb. & Flame, 16, 161 (1971).

18. Jacobs, J., Peeters, L., Vermont, J., Bulletin des Societes Chimiques Belges, 79 (1970).

19. Grumer, J., Harris, M.E., J. Ind. & Eng. Chem., 44, 1547, (1952), "Flame stability limits of Methane, Hydrogen and Carbon Monoxide mixtures".

20. Scholte, T.G., Vaags, P.B., Comb. & Flame, 3 (4) 511 (1959), "Burning velocities of mixtures of Hydrogen, Carbon Monoxide and Methane with Air".

SUMMARY

In this paper, the flame stretch theory is discussed as a means of predicting the blow off of laminar aerated burner flames and it is concluded that the influence of hydrogen cannot be accounted for by the theory.
As preferential diffusion seems the most likely explanation of this, a dimensionless group has been developed which is a measure of the incidence of such diffusion. It has been found that this group can be related to experimentally determined data for the influence of hydrogen on a typical system. A modified flame stretch factor has been proposed to render the flame stretch theory applicable to all fuel gases and mixtures as a means of blow off prediction for design purposes.

FIGURES

Figures I-9 in the text of this paper refer to the following graphs and figures already included in this thesis:-

Figure Number	Thesis Figure or Graph Number
I	Fig. 2.I
2	Fig. 2.2
3	Fig. 2.3
4	Graph No. 48
5	Graph No. 49
6	Graph No. 52
7	Graph No. 54
8	Graph No. 63
9	Graph No. 62

The angle β refered to in the paper corresponds to the angle Φ in Fig. 2.I and 2.2.