

AN OPTICAL STUDY OF IGNITION IN GASES.

by

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## ERRATA

### Page

- 2 'devlopment' should read 'development'
- 56, 58  $L_1$  should read  $L_2$  ;  $L_2$  should read  $L_3$
- 73 equation 6.6 should read:  
$$\rho \lambda = 2 \Delta \delta \ell \cot \theta$$
- 90 bentomite should read bentonite.
- 165 'decelrating' should read 'decelerating'
- 14 Fig. 2.2: (b) should read 2.1 (b).
- 62 'quasi-stated states' should read  
'quasi-steady states'.

ABSTRACT.

The ignition of flowing gases by a hot surface is considered briefly in relation to the potentially hazardous environments associated with supersonic aircraft. A theoretical model is proposed as a basis for investigating pre-ignition conditions in the neighbourhood of a heated surface exposed to a combustible gas stream. By assuming a series of quasi-steady states prior to ignition onset, critical heat release rates are determined, which, it is suggested, may be used to establish safe conditions for any system in which the distributions of temperature and velocity are specified. The development of suitable experimental systems is described, along with the application of various optical methods to the study of conditions preceding ignition in the systems concerned. Interferometry is found to be ideally suited for this purpose, under the experimental conditions considered, and a detailed account is given of the relevant optics and analysis involved. A discussion of the results obtained from a thermal analysis for an ethylene-air mixture includes the deduction of potentially useful kinetic data. It is concluded that the method described in this research could be profitably extended and suggestions are given for developing the present work further.

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## CHAPTER 1.

GENERAL INTRODUCTION.

1. Data on spontaneous ignition are of interest for various reasons <sup>1</sup>. Ignition temperatures at long delay times are important from the standpoint of safety; such values indicate in a relative fashion which fuels constitute the greatest fire hazard, and suggest temperature limits which should not be exceeded. In addition, ignition temperature - ignition delay data may be significant with regard to various aspects of combustion as occur for example in numerous propulsion systems. Finally, thermal ignition data as a manifestation of basic chemical kinetics may yield clues as to the nature of the combustion process itself.

2. Background to the work.

An incidental consequence of high-speed flight is the aerodynamic heating of aircraft structures. This phenomenon and the ignition hazards associated with it arise as follows. In general, when relative motion exists between air and a solid body, a layer of air adjacent to the body is retarded causing a rise in air temperature as the surface of the body is approached. Heat is then transferred to the surface through the boundary layer. Stagnation points

are regions of particular high temperature generation because, here, very fast moving air relative to the body is adiabatically brought to rest. Figure 1.1 shows a typical graph of adiabatic stagnation temperature as a function of air stream velocity. The relationship between the adiabatic temperature  $T_A$  and relative speed of air with respect to a solid body is given by the equation <sup>2</sup> :

$$\frac{T_A}{T_s} = 1 + \frac{\gamma - 1}{2} m^2 \quad (1.1)$$

where  $\gamma$  is the ratio of specific heats,

and  $m$  is the Mach number of the air flow.

The actual temperature attained by the aerodynamic skin adjacent to the solid surface is always lower than the stagnation temperature, and depends on conditions in the boundary layer. For Reynolds numbers greater than a critical value, instability within the boundary layer causes a gradual transition from a laminar to a fully turbulent regime. The curves in figure 1.1 for the two extremes, demonstrates that the effect of aerodynamic heating is important over a wide range of boundary layer conditions.

The ducting of aircraft is invariably subject to heat transfer as a result of aerodynamic heating, either by direct conduction or as a consequence of convection heating by air circulating round the ducting walls. If combustible

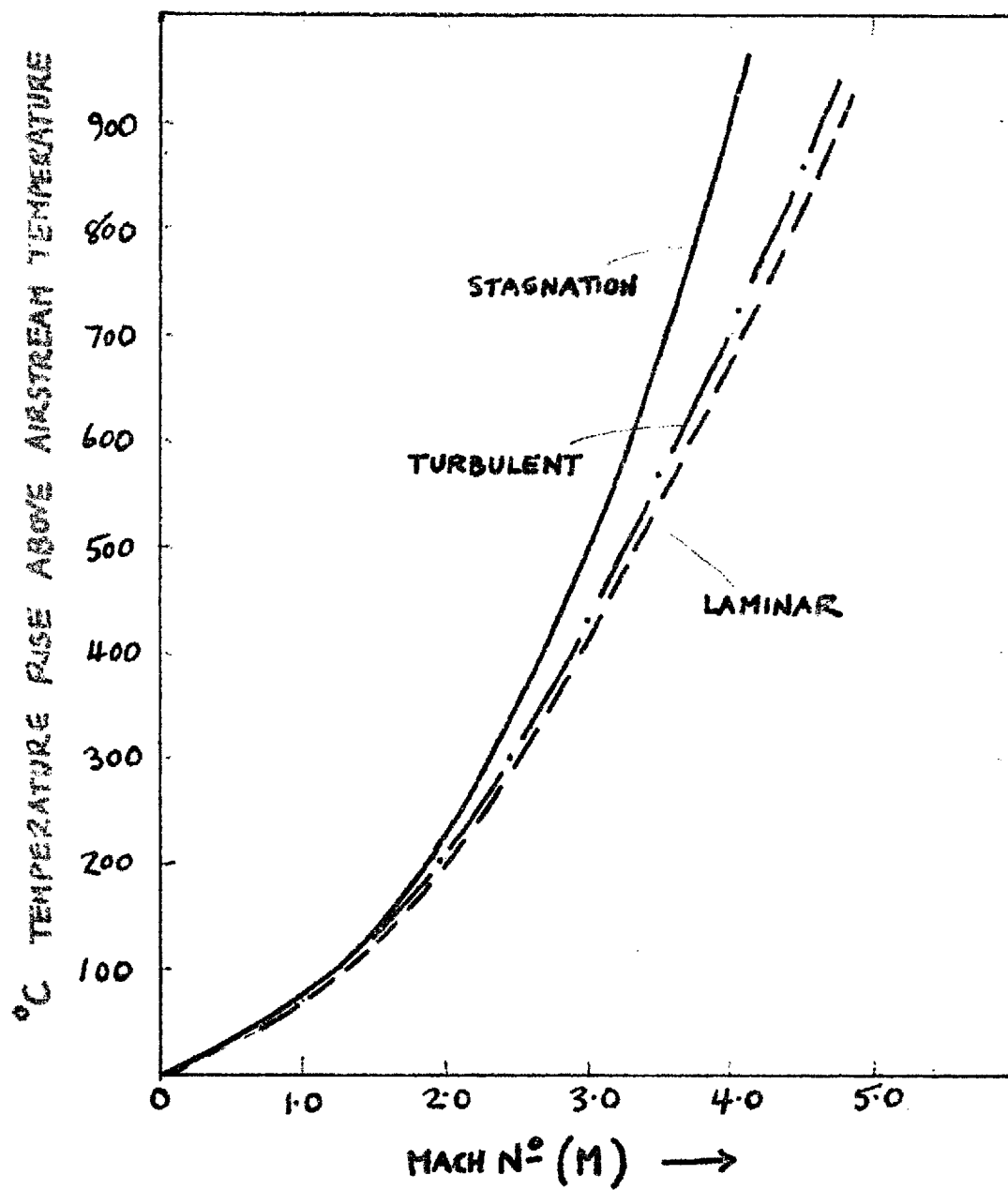


FIG.1.1

mixtures of fuel vapour and air come in contact with the hot ducting surface, spontaneous ignition may occur, with disastrous results. It is one purpose of the present work to investigate possible methods whereby criteria for safety in a flowing gas system could be predicted.

### 3. Theoretical model.

A simple thermal model was proposed as a basis for the present study, in which an exothermic reacting gas stream in laminar flow encounters a hot zone. Under steady-state conditions, the rate of heat release ( $q$ ) due to chemical reaction can be expressed as a function of temperature, by applying the generalised heat conservation equation

$$\nabla \cdot \underline{F} - q = 0 \quad (1.2)$$

where  $\underline{F} = -K \nabla T + \rho \bar{c} T \underline{v}$

and  $K, \rho, \bar{c}, T, \underline{v}$  are the conductivity, density, specific heat, temperature and velocity of the gas.

On the basis of a purely thermal model, an unstable state leading to spontaneous ignition of the gas arises when the rate of heat release within the gas just exceeds the rate of heat removal from the system. Knowledge of critical heat release data may then be used to establish what conditions are hazardous for any specified system no matter what geometry is involved, by calculating the net rate

of heat removal and deducing under what temperature conditions it exceeds or falls below the critical rate of heat release.

#### 4. Outline of the investigation.

It was decided to approach the problem by developing an experimental system to provide the data required for the theoretical model. Steady-state conditions just prior to ignition were to be studied by determining the temperature and velocity distributions within the gas and applying equation (1.2) to determine  $(q)$  as a function of temperature.

The obvious simplicity of the thermal model implies certain assumptions which need to be carefully considered when extending the model in general. For instance, attainment of steady-state conditions under critical pre-ignition conditions may be difficult to achieve in practice. This applies particularly to very reactive systems, where any time dependence is too high for the experimental techniques available to resolve. A further possible difficulty, which would demand modification of a purely thermal model, is catalysis at hot surfaces exposed to the reacting gas stream. In the present work steps were taken to minimize this effect. Special consideration must be given to ignition phenomena involving thermo-neutral reactions. All practically dangerous explosions are governed by overall

exothermic reactions in which the heat released chemically is the predominant cause of the rapid gas expansion. Thus, even if the initial stages of the reaction are thermo-neutral, heat release eventually sets in well before ignition, and would be measurable. Of course, a perfectly thermo-neutral ignition phenomenon could not be investigated in the way described, since reaction rates would not be detectable in terms of temperature increases. From the standpoint of safety, however, such reactions would be relatively harmless 'ignition hazards', as composition changes alone would be responsible for the explosion. The real risk, therefore, of an exothermic reaction which starts with thermo-neutral steps, is that a particular temperature will not define the rate of heat release, because that will depend on the accumulation of species, radicals, etc. The consequence of this will be to make the rate of heat release dependent on the previous thermal history and not just the instantaneous temperature of the gas. One way of establishing whether the rate of heat release is a unique function of temperature for a specific reacting system, would be to vary the flow velocity of the gas and observe whether the distribution of  $(q)$  as a function of temperature remained constant. Before extensive investigations can be carried out along the lines suggested, however, it is essential to first establish a method whereby the necessary data can be measured. Most of the present work is concerned with this latter problem.

## CHAPTER 2.

A CRITICAL SURVEY OF PREVIOUS WORK.1. General.

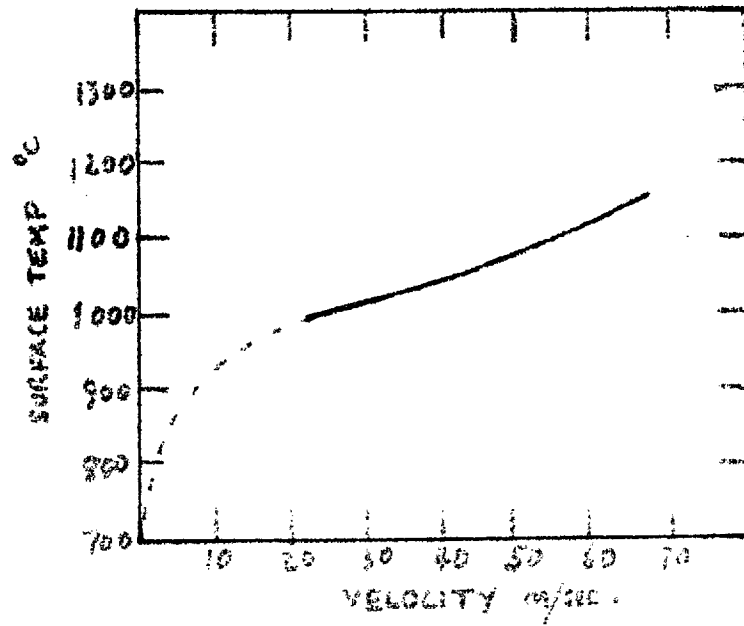
The ignition of combustible gas mixtures has been the object of a great number of theoretical and experimental investigations<sup>3</sup>. A broad division of the subject is necessary, based on the modes of ignition involved. These include spark discharges<sup>4,5,6</sup>, flames<sup>7,8</sup>, hot surfaces<sup>9,10,11</sup> and adiabatic compression ignition<sup>12,13</sup>, as well as more doubtful ignition agents such as radiation and ionization<sup>14</sup>. Theoretical studies<sup>15,16,17,18</sup> of the phenomenon have been attempted, but all involve basic assumptions about the physical state or chemical kinetics within the ignition system. As a result, there is frequently poor correlation between theory and the experimental facts. It is interesting to note that two main so-called 'ignition theories' have been postulated; namely a thermal and a chain theory. An ionization mechanism of ignition has also been proposed<sup>19</sup>. Digression into fuller discussion of the various theories will not be made here, but of these propositions the thermal theory seems to offer the most satisfactory basis for understanding ignition in general. This theory may also be considered to govern conditions

pre-requisite for a chain theory of ignition to operate <sup>20</sup>.

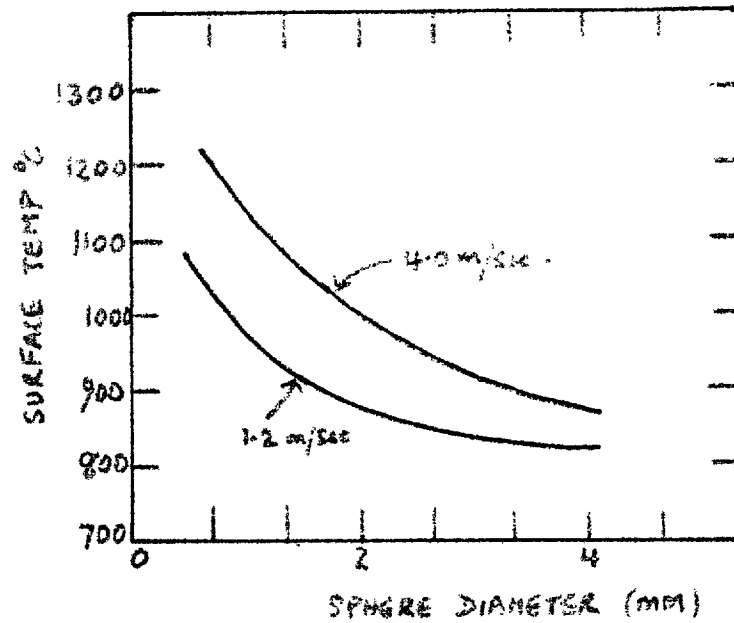
2. Before discussing previous research, it is useful to consider those aspects of the present work which are particularly relevant. The phenomenon under investigation is ignition of a combustible gas, while in laminar flow and under forced convection, near a hot surface. The study is essentially an optical one, directed primarily at the quantitative measurement of pre-ignition heat release rates, the purpose of which has been briefly discussed in Chapter 1. Thus, the following survey is mainly confined to systems in which ignition near a surface occurs under conditions of relative motion between the explosive gas and ignition source.

3. Silver <sup>21</sup> and Paterson <sup>22,23</sup> made the first serious attempts to correlate ignition criteria with experimental variables for such a system. In an earlier work <sup>24</sup> experimental observations were recorded for ignition of a flowing, combustible gas by an electrically heated wire. These workers concluded that the surface temperature of the wire, necessary to cause ignition, increased with gas velocity and decreasing wire diameter. In Silver's experiments, quiescent, combustible gas mixtures were ignited by hot, moving spheres projected into the gas. Various materials were used for the spheres. It was established

that the ignition temperature of the spheres varied inversely with sphere diameter, as shown in figure 2.2(a), but showed little dependence on the nature of the sphere material. (Paterson <sup>22</sup> showed, however, that polished Pt required much higher surface temperatures than other materials to effect ignition of the gas mixtures). The results could be crudely explained on the basis of a thermal ignition criterion. For a given gas mixture and sphere velocity, heat transfer to the gas may initiate thermal reaction near to the particle surface. If the rate of heat evolution in the initial reaction just exceeds the heat loss from the gas near to the hot surface, ignition subsequently results. By increasing the sphere diameter, the residence time of a reactive gas particle increases, thus favouring the attainment of a critical rate of heat release to meet the thermal criterion for ignition. Paterson <sup>22,23</sup> considered the same problem in more detail, particularly the variation of surface ignition temperature with sphere velocity (figure 2.2(b)). The results indicate an approximately linear dependence of ignition temperature on particle velocity for regimes in which the flow downstream of the sphere is turbulent. The lower dotted part of the curve represents a laminar flow regime. Paterson speculated that ignition probably started in the boundary layer region close to the sphere, or in the actual wake through upstream diffusion of



(a)

FIG.2.1

(b)

active species. With the exception of hydrogen, it was found that minimum ignition temperatures were obtained for approximately stoichiometric fuel/air mixtures. The results of Silver and Paterson indicate that the ability of a sphere to ignite a gas mixture depends on its surface temperature, size and velocity.

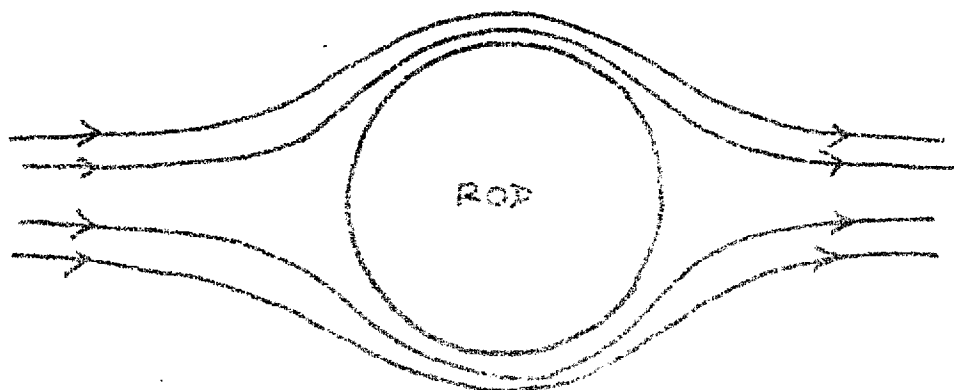
Mullen et al.<sup>25</sup> studied the conditions for fast stream ignition by electrically heated rods. Systematic measurements were made of rod surface temperature and stream velocity at the ignition threshold for different experimental conditions. Although the flow velocities employed were much higher than in the present work, it is interesting to note that in both studies optical pyrometry and interferometry were used to obtain information about rod surface and gas temperatures respectively. The interferometry in Mullen's work served only as a qualitative assessment of the temperature distribution in the gas. No attempts were made to study the velocity distribution in the flow regime and a single pitot tube measurement, just upstream of the igniter, constituted the total velocity data for each run. Results obtained are in broad agreement with those of Silver and Paterson. A pronounced concave up curve, with velocity taken along the abscissa, between rod surface temperature and stream velocity is characteristic of all their

results. The significance of this is not clear, but the authors suggested it is primarily a chemical effect and disregarded the influence of the flow patterns involved. They put forward the argument that an element of gas has only a finite time of contact with the heated rod, depending on the velocity of the gas stream. If a definite rate of reaction has to be achieved within the gas element for ignition to occur, the faster this element travels, the greater the initial rate of reaction needs to be, since contact time with the rod decreases. Assuming an Arrhenius type dependence of reaction rate on temperature (Rate  $\propto \exp(-E/RT_i)$ ), equal increments in  $T_i$  will not correspond to equal rate increases. So for equal reaction rate increments to accompany equal stream velocity increments, a greater than linear increase in  $T_i$  must apply. Mullen et al. proposed an ignition criterion based on correlation of rod temperature and stream velocity data. The contact time for a gas particle was taken to be proportional to  $D/u$ , where  $D$  and  $u$  are the rod diameter and gas stream velocity respectively. Assuming that the critical reaction rate was proportional to the contact time they obtained the following relationship :

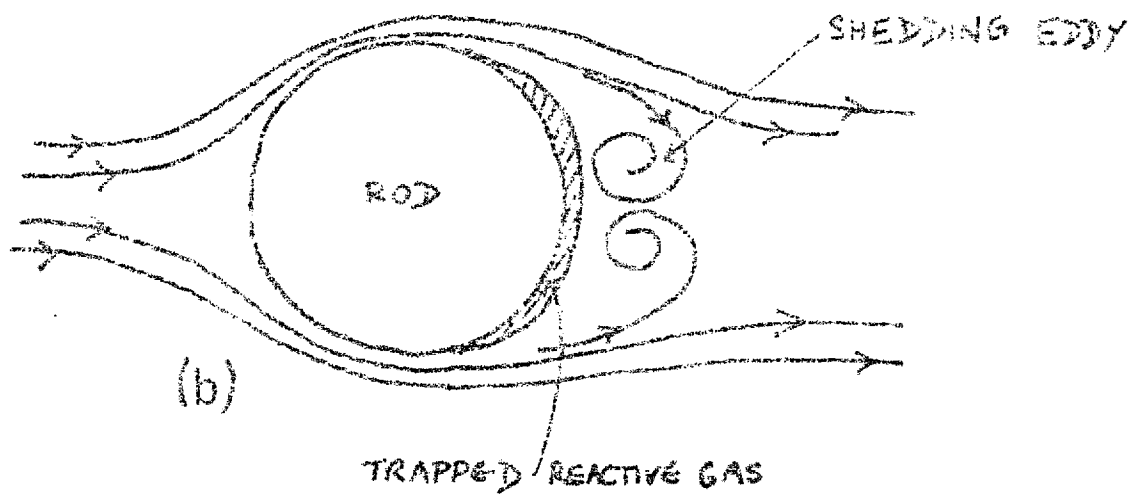
$$T_i \propto \frac{C \times D (T_r - T_s)}{u} \quad (2.1)$$

where  $T_i$ ,  $T_r$ ,  $T_s$  are the critical ignition, rod and gas stream temperatures.  $C$  is a constant.

Lewis and von Elbe <sup>26</sup> have argued that this correlation leads to results of no physical significance. In the opinion of the present author, the effect of the flow pattern, just downstream of the igniter rod, could play a significant role in the ignition process. The hydrodynamics of vortex formation at high Reynolds numbers behind a cylinder have been studied by Prandtl <sup>27</sup>. These conditions apply to Mullen's work, and figure 2.2 shows the downstream flow patterns observed for low and high Reynolds numbers. In figure 2.2b fluid from the boundary layer builds up on the downstream face of the rod. Mullen found, by interferometry and schlieren photography, that ignition is initiated in the downstream wake close to the rod surface. Under these conditions it is conceivable that increased heat transfer from the reacting gas trapped in the downstream face, to the relatively cold external flow, accompanies increased stream velocity. This would lower the temperature of the trapped gas and decrease the reaction rate. Entrainment of the reactive, trapped gas by the detached external flow may also reduce the effective 'steady state volume' in this region to below a minimum necessary to effect ignition at that temperature. An increase in rod surface temperature would be necessary in either case. It is suggested, therefore, that the effect of the flow pattern may be a serious factor in the relationship between rod surface temperature and stream



(a)



(b)

FIG 22

velocity, reported in (25).

Sage and Weinberg<sup>28</sup> attempted to measure 'absolute' spontaneous ignition temperatures, using a novel technique based on the following concept. When the temperature of a volume of ignitable gas at constant pressure is raised rapidly to some critical value, and maintained at that temperature, homogeneous ignition will occur throughout the gas. If the gas temperature is raised to, and maintained at, just below the critical value however, explosion will not occur without increasing the temperature. The thermal energy necessary to effect ignition was supplied in two stages. The first stage raised the temperature of the reactants homogeneously to a value, measured by means of a suction pyrometer, at which pre-ignition reaction commenced. Additional energy was then supplied by means of a second stage, causing ignition. By varying the energy input of the primary stage, adjustment of the second gave a method of extrapolating to zero energy for the second stage and hence to the homogeneous ignition temperature of the gas.

It is interesting to note that the ignition temperature of about 700°C found for ethylene-air mixtures in their work was considerably higher than the previously accepted values, and appeared to be unaffected by changes in fuel concentration. In the present work even higher

ignition temperatures were observed for ethylene-air mixtures, which displayed a much stronger dependence on mixture composition.

Kumagai <sup>29</sup> concluded from ignition experiments with heated nichrome wires in flowing gases that the ignition temperature is very nearly a linear function of contact time, especially for larger wire diameters. Other workers <sup>30,31,32</sup> have also studied hot wire ignition, but not for flowing gas systems.

Toong <sup>33</sup> gives a theoretical account of combustion development in a gas flowing parallel to an isothermal flat plate. Various approximations are made about the velocity and temperature distributions associated with the boundary layer regions, in which the ignition process takes place.

Goldenberg <sup>34</sup> considers a similar model on the basis of Khitrin's general treatment of ignition problems <sup>35</sup>. Again a number of simplifications are introduced. Up to the instant of ignition, both velocity and temperature boundary layers are assumed to develop in the absence of chemical reaction. Having solved the problem of heat transfer from the plate to the gas stream, the effect of reaction is introduced using Khitrin's general result. A familiar thermal ignition criterion is proposed; namely that heat evolved by the reaction adjacent to the plate must just exceed the heat transferred away from the vicinity of the

plate. Equation (2.2) shows the form of Goldenberg's results

$$\frac{X}{u_0} = K_1 \left( \frac{T_s - T_0}{T_0} \right)^2 \exp E/RT_s \quad (2.2)$$

where  $u_0$  is the free stream velocity

$T_0$ ,  $T_s$  the stream and surface temperatures

$E$  is a global activation energy

$X$  is the distance from the leading edge of the plate to the point of ignition.

This suggests that the distance a particle travels over an isothermal plane, before ignition occurs, depends only on  $E$  and  $u_0$ . There has been no experimental verification of such a general result as far as the author is aware.

Chambre<sup>36</sup> treats the problem of surface ignition using the relatively simple hydrodynamic field of a gas stream impinging on a flat plate, (figure 2.3). The analysis is a theoretical one, but of relevance to the present study, where a similar flow pattern was employed for much of the work. Some of the assumptions made by Chambre in his theoretical model apply equally to the experimental model used in the present work.

Sontaneous ignition of aviation fuels in particular has received much attention<sup>37,38,39,40</sup>. Mullins<sup>41</sup> has

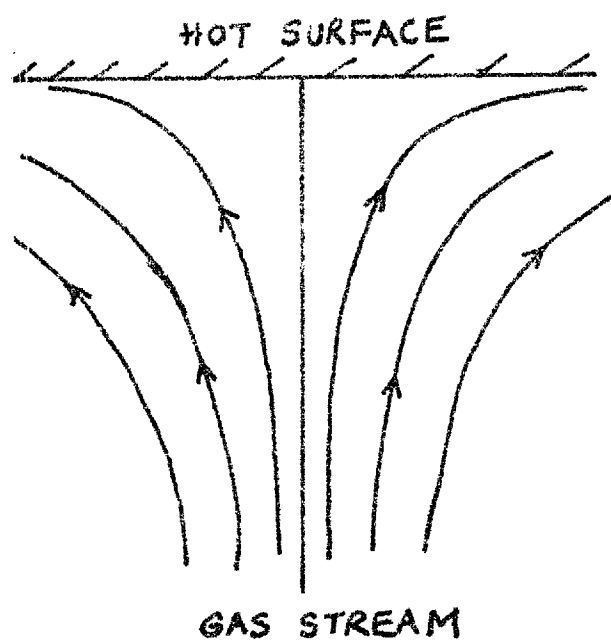


FIG.2.3

contributed greatly to this area of ignition. The methods used in this kind of work appear to be very empirical and frequently relate to static systems. Ignition delay times are mostly deduced. Randall <sup>38</sup> attempted, without success, to develop theoretical ignition criteria based on the correlation of limited experimental data available, with the object of extrapolating to untried experimental conditions. The method used was to postulate criteria for ignition in terms of a purely thermal model and then to introduce modifications which would account for chain branching mechanisms.

4. In much of the previous experimental work on ignition of flowing gases near a surface the primary measurement has been the limiting surface temperature, above which ignition of the flowing gas occurs. In practice this approach is of very limited use, because the critical surface temperatures so measured are generally found to be specific to the experimental systems used for the particular investigation. The reason for this state of affairs lies in the complex physico-chemical nature of the ignition process itself. Unless the influence on the chemical kinetics of factors such as the size, temperature and nature of the hot surface exposed to the gas, and the type, concentration and flow velocity of the gas concerned are understood, critical

ignition temperatures alone contribute little to elucidating the problems of ignition. In this respect the work of Salooja <sup>42</sup> is valuable.

Much more potentially useful information about the critical conditions preceding ignition in the gas itself can be obtained, if relatively simple experimental models are considered in which key parameters such as temperature and velocity can be specified. It will have been apparent from the foregoing discussion that the approach of the present work, outlined in Chapter 1, has little in common with previous ignition studies. In fact the method of analysis outlined earlier follows more closely along the lines of classical flame structure studies such as, <sup>43,44,45</sup>, in which heat release rate profiles were determined as a function of temperature. Pratt <sup>46</sup> has also investigated the pre-flame zone structure of pre-mixed methane-air flames using a composite probe technique. Although the experimental objectives of the present study are similar to those sought in many flame structure studies, a marked distinction between the two arises, in that the heat source in flame studies is an important source of highly active species such as radicals which must contribute by diffusion to the pre-ignition reactions, whereas in the present investigation the 'flame' is replaced by a heated zone, which is chemically inert, and thus permits a strictly thermal analysis to be carried out.

## CHAPTER 3.

CHOICE OF METHODS FOR DETERMINING EXPERIMENTAL VARIABLES.

1. From equation (1.2) it follows that in order to calculate a distribution of  $(q)$ , the parameters of temperature and velocity must be specified at every point. Values of local conductivity and thermal capacity for the gas are also required. The temperature dependence of these two physical quantities is well known for a number of gases <sup>47-50</sup>, but further complications arise in the presence of appreciable composition changes. In the present work, where lean ethylene/air mixtures were used, such effects are negligible, because of the small extent of reaction under pre-ignition conditions. This does not imply that the analysis to be described cannot be extended to systems where appreciable composition changes do occur. In such cases a deduction, or determination of species concentration would be an additional requirement. In fact, for flame reaction studies the secondary effects due to appreciable composition changes have to be taken into account in estimating local conductivity values. It is possible <sup>51</sup> to deduce the composition changes from the extent of reaction that has taken place, which can in turn be calculated from the amount of heat released and the temperature of the system.

## 2. Gas Temperature Measurement.

The methods available for gas temperature measurement are numerous and will be considered in three separate groups.

### a. Solid probe techniques.

These include thermocouples, hot wires, and suction pyrometers. The chief disadvantage of using probes for measuring gas temperatures are as follows. First, physical disturbance of both temperature and velocity fields within the gas is inevitable. Second, there is a risk of catalysis occurring at the probe surface exposed to a reactive gas stream, unless precautions such as described in (52) are rigorously and frequently observed. Finally, a limit is set on the spatial resolution of such methods, depending on the dimensions of the probe concerned.

### b. Methods based on the measurement of (i) the velocity of sound and (ii) the range of $\alpha$ -particles.

(i) The velocity of sound ( $c$ ) through a gas at temperature ( $T$ ) is given by the relationship

$$c = \sqrt{\frac{\bar{\gamma} RT}{\bar{M}}} \quad (3.1)$$

where  $\bar{\gamma}$  and  $\bar{M}$  are the average ratio of specific heats and mean molecular weight respectively. The main disadvantages

of using sound velocity measurements for determining gas temperatures are two-fold. The resolving power is limited by the wavelength of the radiation used, so unless high-frequency ultrasonic waves are employed, normal sound waves will give completely inadequate spatial resolution when compared with optical methods, for example. In addition, time varying temperature fields pose practical difficulties with regard to scanning mechanisms for sweeping across the test region.

(ii) The range of  $\alpha$ -particle method <sup>56</sup> depends on the 'stopping power' of the gas whose temperature is being measured and this is a simple function of gas density. It is difficult to make the technique accurate, and when compared with optical methods, which also measure gas density, is shown to be very inferior. Resolution is moderately better than in the case of sound velocity measurements, but the method is also unsuitable for investigating time varying phenomena.

### c. Optical methods.

An obvious attraction of all optical methods is their complete lack of physical interference with the test region. Various techniques have been successfully developed (51,53), including schlieren photography, deflection mapping and interferometry. Each of these methods depends on the fact that the refractive index of a gas is itself a function

of density related according to Gladstone and Dale as :-

$$\frac{n - 1}{\rho} = \text{const.} \quad (3.2)$$

where  $n, \rho$  are the refractive index and density of the gas respectively.

The density of a gas at constant pressure varies with the absolute temperature as

$$\rho_T = \rho_o T_o \quad (3.3)$$

Defining  $(n-1) = \xi$

(3.2) and (3.3) combined give :-

$$\xi_T = \frac{\xi_o T_o}{T} \quad (3.4)$$

where the subscripts (o) refer to some arbitrary reference state. This relationship (3.4) applies in the absence of appreciable composition changes. A weakness of optical and all other methods which measure temperature as a function of gas density, is the fall off in sensitivity with temperature.

Differentiating (3.4) gives

$$d\xi = - \frac{\xi_o T_o}{T^2} dT \quad (3.5)$$

indicating that at high temperatures, small errors in  $\xi$  correspond to large errors in temperature, though the fractional errors of course are the same, i.e.

$$\frac{d\xi}{\xi} = \frac{\xi}{\xi_o} = - \frac{T_o}{T}.$$

Two optical techniques in particular have been developed for quantitative gas temperature measurements. The first is deflection mapping which, as the name suggests, involves the measurement of light deflections by refractive index fields through which the light is passed. The technique has been successfully applied to flame structure studies<sup>54,55</sup>. The second is interferometry which has found wide application in the field of aerodynamics. These two methods are equivalent, measuring different functions of the same variable,<sup>6</sup>. A distinction between the two techniques is important and may be considered as follows.

### 3. Deflection Mapping.

The principle of deflection mapping can best be treated in terms of geometric optics. Figure 3.1 shows a simple optical arrangement suitable for the so-called 'inclined slit' deflection mapping method. A narrow slit or half-wave step, placed as shown, marks the wave front of a collimated beam of light which is arranged to traverse the refractive index field under investigation. In the absence of a refractive index field, a straight line image of the slit or half-wave step is observed on the screen (S). In the presence of a test region, such as a flame, the slit image becomes distorted due to deflection of the collimated beam of light. The deviation ( $\theta$ ) of a single ray passing

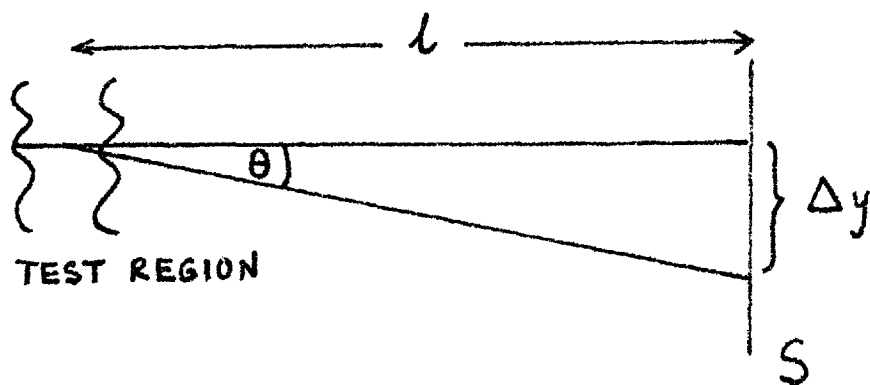
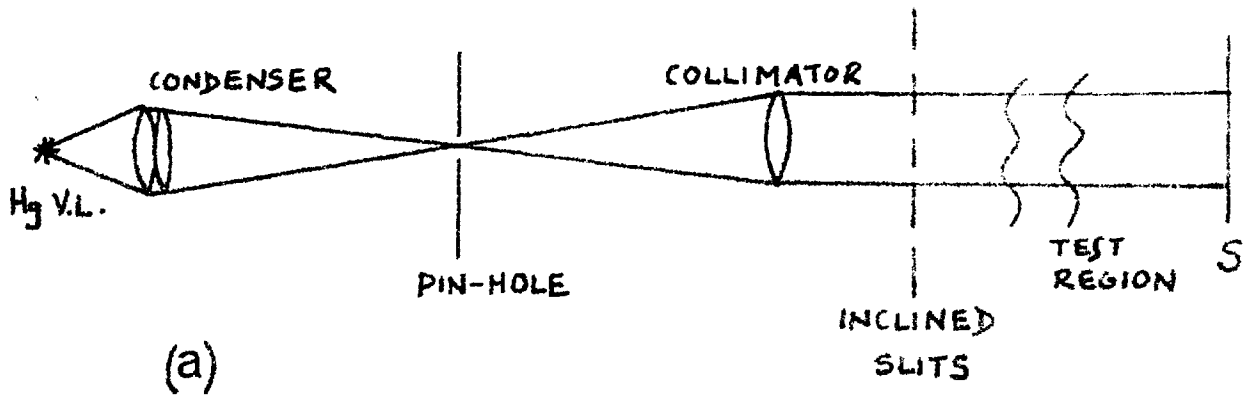


FIG.3.1

through a distance  $X$  of the refractive index field is given to a good approximation by :

$$\theta = \int_0^X \frac{dn}{dy} dx \quad (\text{see Appendix 1}) \quad (3.6)$$

where  $dn/dy$  is the refractive index gradient perpendicular to the direction coordinate  $x$  of the undeflected ray. Using the deflection  $\Delta y$ , observed on the screen, the following relationship is obtained for the local refractive index gradient

$$\frac{dn}{dy} = \frac{\Delta y}{lX} \quad (3.7)$$

It can be shown <sup>51</sup> that the sensitivity of the deflection mapping method increases with temperature gradient, but this is an over-simplification and it will depend also on the extent and nature of the refractive index field in question.

#### 4. Interferometry.

The principles are considered in terms of physical optics. Essentially, a plane light wave from a single source is separated into two, either by division of amplitude or wave front; one forming the test and the other a reference beam. The test beam only passes through a refractive index field and suffers phase changes along its path, while the reference beam remains unchanged. When the two beams are subsequently superposed, the phase changes in the test beam

are thrown into relief by the interference fringe pattern produced. Interference between the two wave fronts is governed by the condition ;

$$\int_{\text{Test}} n dx - \int_{\text{Ref}} n dx = p \lambda \quad (3.8)$$

where  $p$  is an integer - the 'fringe order'

$\lambda$  is the wavelength of the light

$n$  is the refractive index of the medium.

Where the combining wave-fronts are exactly parallel in the absence of a test field, the fringes, in its presence, represent contours of equal optical path differences between coincident points in the test and reference beams - as defined by (3.8). When there is a finite angle between the two wavefronts a linear fringe pattern is observed, even in the absence of a test refractive index field. In this case, in the presence of a field, the relative shift in position of the fringes as the result of phase changes is used in equation (3.8), instead of an absolute value of fringe order ( $p$ ).

The sensitivity of the method can be deduced by differentiating (3.8).

$$\text{Thus, } \lambda dp = d \left( \int_{\text{Test}} n dx - \int_{\text{Ref}} n dx \right) \quad (3.9)$$

which for a field where  $dn/dx = 0$  is equal to

$$X d (n_{\text{test}} - n_{\text{ref}})$$

where  $X$  is the geometric path length.

So that 
$$d\delta = \left( \frac{\lambda}{X} \right) dp$$

where 
$$\delta = (n-1).$$

The accuracy of  $\delta$  depends directly on the accuracy of reading changes in fringe order and on the ratio  $X/\lambda$ .  $X$  is generally accurately known, but at the edge of the test field  $dn/dx$  is never zero due to the presence of factors such as heat transfer.

Whereas deflection mapping appears to be most sensitive for fields of high refractive index gradients, the converse is true of interferometry. In the latter case, two difficulties arise in the presence of steep temperature gradients. The fringes become so closely spaced that resolving them is difficult and in addition the optical path lengths considered in equation (3.8) must be corrected for the appreciable deflections which are suffered by the test beam. The relative importance of these deflections is dealt with in Appendix 1.

For the purpose of the present work, these factors eventually favoured interferometry. It was anticipated that the refractive index gradients encountered would not be excessive. An additional advantage of using interferometry is that the entire refractive index field can be recorded

simultaneously, whereas this is not possible with deflection mapping. However, in case the actual test fields proved too steep for interferometry, it was decided to develop an optical system with sufficient flexibility to accommodate either technique. The development of such an arrangement will be described below.

##### 5. Accuracy of the method.

It was shown above, that, because of the fall off in sensitivity of optical methods with temperature, small errors in refractive index measurements produce large errors in temperature. Furthermore in the case of interferometry

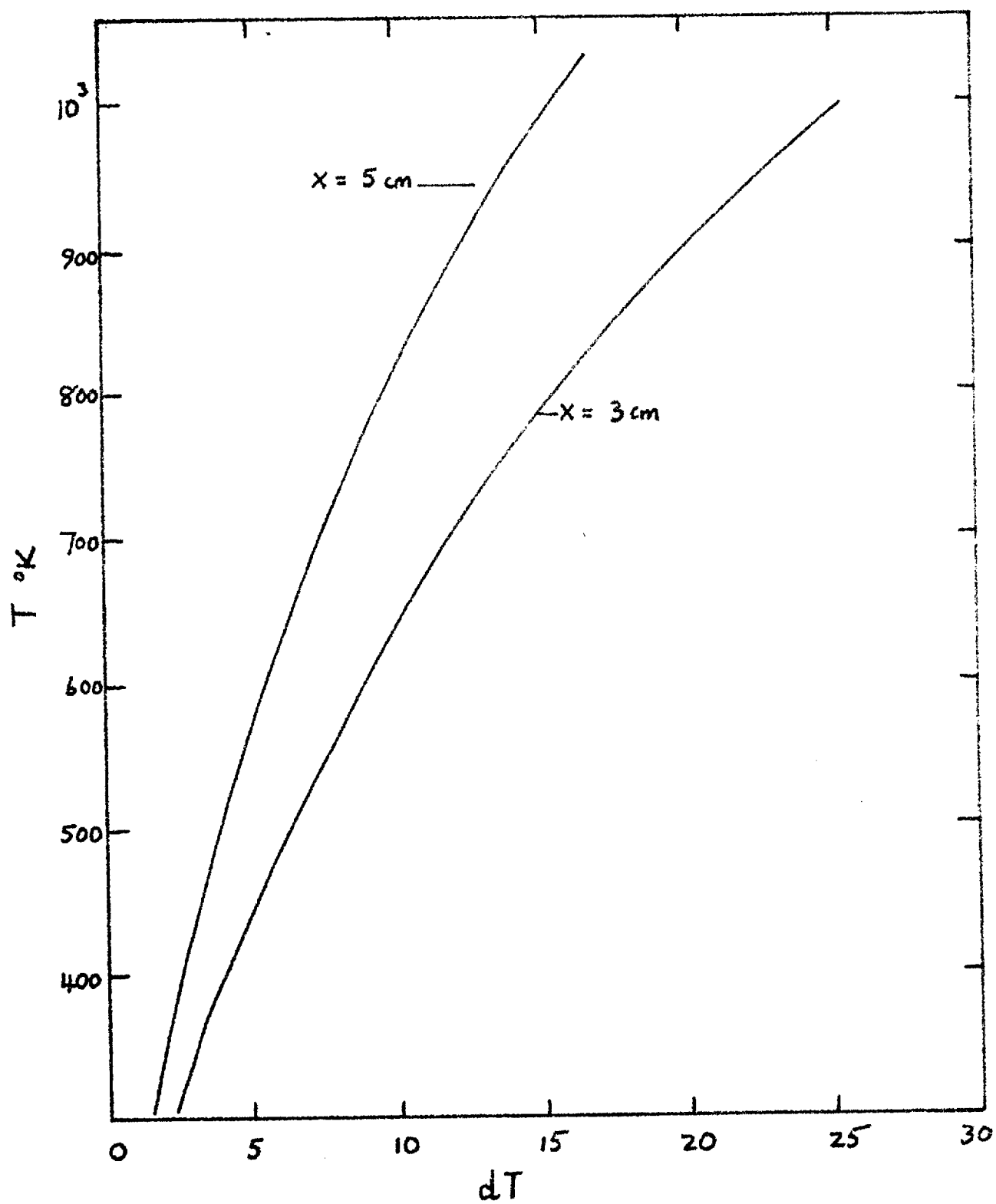
$$d\delta \propto dp$$

In the present work  $dp$  corresponded at best to about  $1/10$  of an order, at a wavelength of  $5.5 \times 10^{-5}$  cm. For a mean geometric test path length of 5 cm, and employing the relationship (3.5) gives :

$$\begin{aligned} dT &= - \frac{T^2}{\delta_0 T_0} d\delta \\ &= - \frac{T^2}{\delta_0 T_0} \frac{\lambda}{X} dp \end{aligned} \quad (3.10)$$

Substituting into (3.10) gives  $dT = 1.5 \times 10^{-5} T^2$  for air.

Figure 3.2 shows the fall off in sensitivity for a geometric path length of 5.0 cm and 3.0 cm. The calculations for figure 3.2 were based on the assumption that

FIG.3.2

$dn/dx = 0$  over the entire test field. In practice this is very far from the truth, and it will be shown that the calculation of optical paths,  $\int_{\text{test}} n dx$ , for a two-dimensional refractive index field involves errors which, taken in conjunction with the fall off in sensitivity of  $\epsilon$  with temperature, can lead to unacceptable overall errors.

The problem is most readily overcome by using (3.5) in reverse and pegging the refractive index field to the highest temperature boundaries, which are either known or can be deduced for the system. Details of the method are discussed in Chapter 8. In the present work, the highest temperature boundary coincided with the hot surface exposed to the gas stream. A suitable method for measuring the surface temperature distribution was therefore sought. For much of the work, a non-catalytic quartz surface was employed, which restricted the choice of methods.

## 6. Solid Surface Temperature Measurements.

### a. Solid probe techniques.

These were not practical, because the quartz surface was extremely delicate and polished to optical flatness. In any case, objections to using probes could be made on the grounds discussed in the section dealing with gas temperature measurement.

b. Radiation Methods.

None of the objections associated with probes apply to these methods <sup>57</sup>. All the methods in this group involve the use of a pyrometer and the measurements are based on the laws governing total, spectral or maximum wavelength radiation from a black body. The advantage of non disturbance of the test field associated with radiation techniques is obvious. A number of specific methods were considered. Spectral radiation is only applicable for colour temperatures  $> 1200^{\circ}\text{K}$ . Below this temperature, the error involved in matching a calibrated filament of a pyrometer against the background colour temperature of a hot surface is very large. Likewise, the maximum wavelength radiation method, based on Wien's displacement law, is only accurate for temperatures  $> 3000^{\circ}\text{K}$ . Spectral radiance-ratio measurements involve the determination of spectral radiance at two or more wavelengths, and additional knowledge about surface emissivities as a function of wavelength is required. An instrument of this sort is costly, but has the advantage over other radiation methods of high spatial resolution.

A total radiation pyrometer seemed most suitable for the present work, where, in the absence of actual flame formations, the surface temperatures were well below those encountered in flames. To operate the instrument, the

pyrometer is sighted onto the hot surface and receives a constant fraction of the radiant heat emitted. The energy received is focused onto a sensitive detector, which is usually a thermopile. For the instrument used in the present work, an accuracy of less than 2% is claimed over a temperature range of 800°K - 1300°K.

## 7. Gas Velocity Measurements.

A number of techniques employing classical probes, such as pitot tubes and hot wire or quartz fibre anemometers, were considered. For reasons similar to those already referred to in the discussion on the suitability of probes for gas temperature measurements, these methods were rejected in favour of the so-called particle track method. This is a special optical/probe technique which has been used for flame structure studies<sup>58-60</sup> and for general flow visualization purposes. Microscopic, inert particles are introduced into the gas flow and a strongly convergent Tyndall beam is focused into the region of interest in the gas flow. Light scattered by the particles which are taken up by the flowing gas is then photographed at right angles to the Tyndall beam. By suitable interruption of the illumination, a time base can be introduced to deduce the particle velocities from the photographs. Figure 3.3 shows

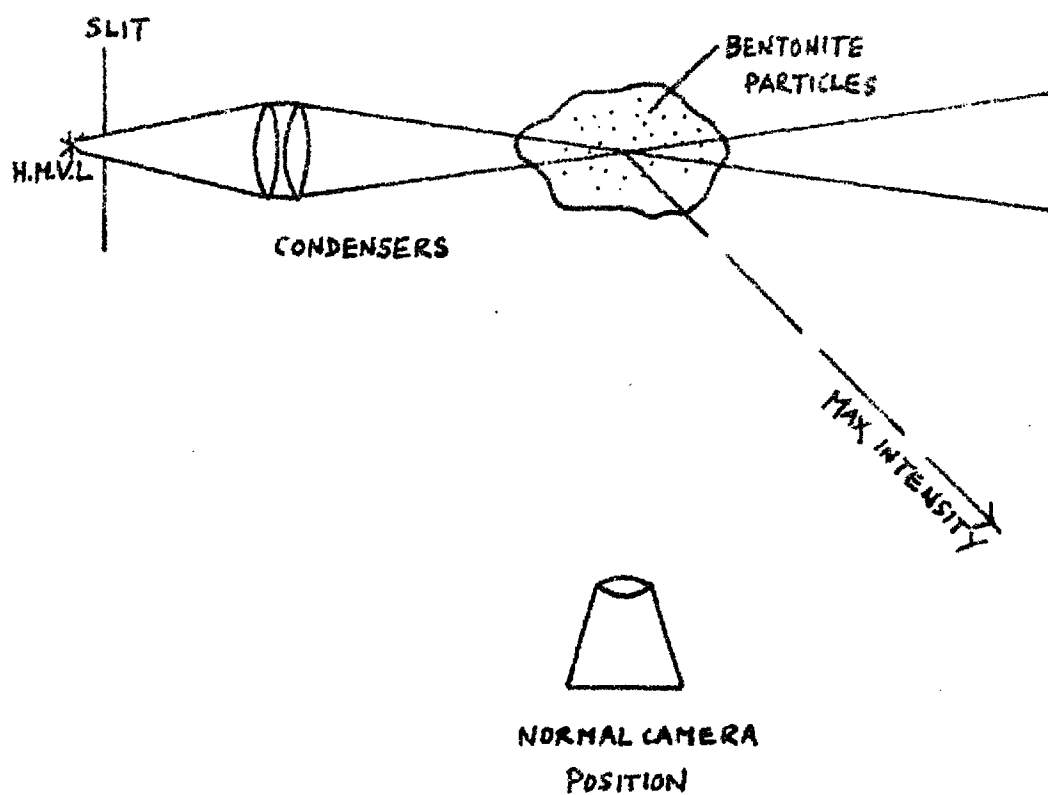


FIG.3.3

the simple experimental arrangement used in the present work. A slit about two millimetres wide was placed next to a high-pressure mercury arc source. An image of the slit was then focused into the test plane of the gas flow. It was observed that the most intense scattering of the Tyndall beam was a maximum with the camera positioned along a line approximately  $120^{\circ}$  to the Tyndall beam. In practice, some compromise between maximum scattered light intensity and size of field of view had to be made, so that the convergence of the beam was adjusted to fill the test region. For some of the work it was found necessary to employ two separate light sources to achieve this purpose. Particles were photographed at right angles for sharpest focusing over a wide viewing angle, because the camera had no facilities for altering the angle between the axis of the lens and camera back. Aerodynamic gas temperature measurements are possible under suitable conditions, using the particle track method <sup>61</sup>. This method of gas temperature measurement will be discussed at a later stage in relation to the present work.

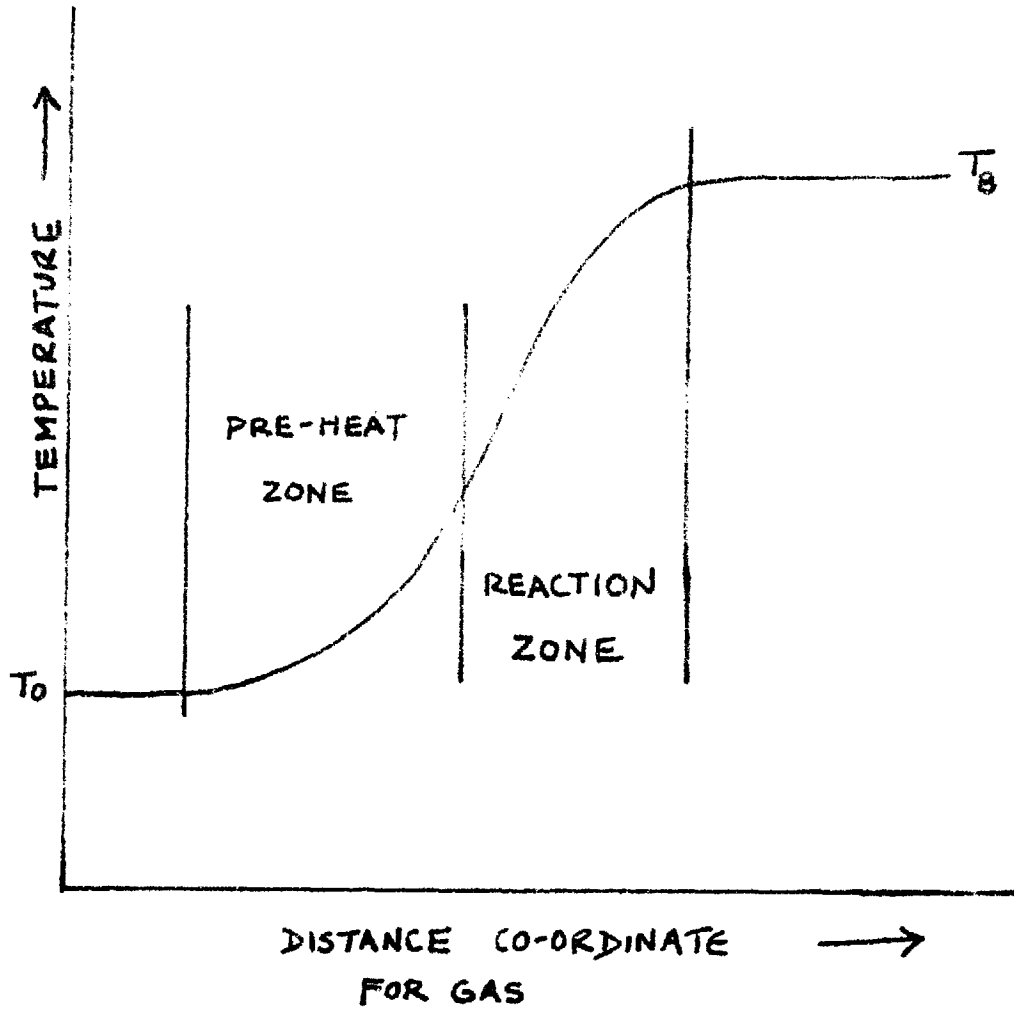
## CHAPTER 4.

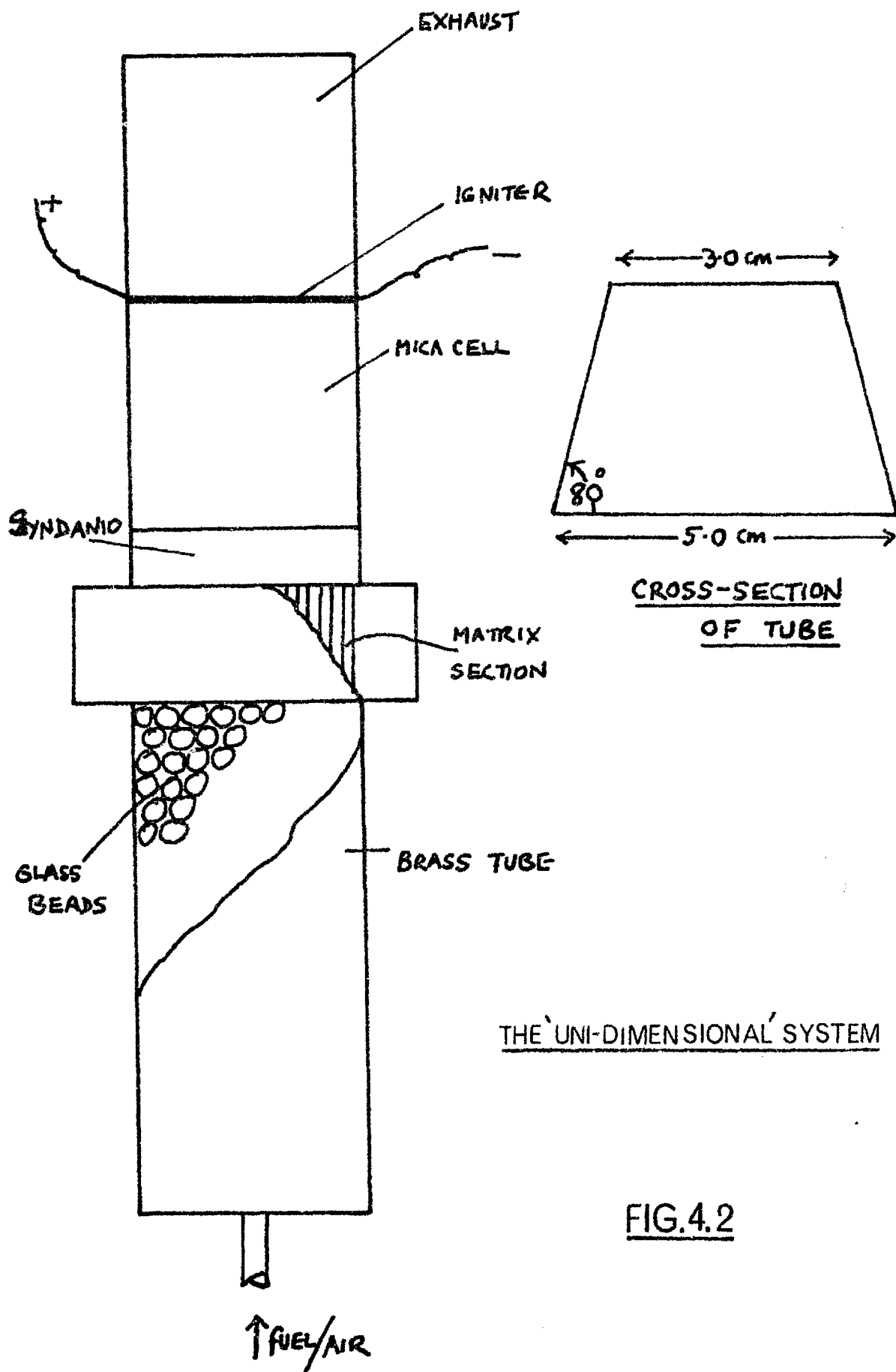
CHOICE OF EXPERIMENTAL SYSTEM.

Initially, it was proposed to work with a simple one-dimensional system. The results obtained indicated a need for certain modifications, which led to the development of a two-dimensional model.

1. The "Uni-dimensional Model".

Broadly speaking, the model proposed simulated the purely thermal part of a one-dimensional laminar flame, figure 4.1, in that the reaction zone of the flame was to be replaced by a hot, porous plug or grid, while avoiding the large diffusion flux of active species associated with flames. A steady flow of reactants approached the hot zone, which lay perpendicular to the gas flow, enabling a steady temperature distribution ahead of the porous plug to be established. Figure 4.2 shows the experimental model of the ignition tube and hot grid. The cross-section of the system is trapezium-shaped according to the dimensions given in the diagram, figure 4.2. The reason for this choice of geometry is related to the interferometric method used for determining the temperature distributions and will be explained subsequently. Operation of the apparatus was briefly as follows. Metered and dried ethylene and air were mixed

FIG.4.1

FIG.4.2

before entering the base of the ignition tube. The metering system is shown in figure 4.3. Glass beads occupying the lower section of the tube (figure 4.2) randomised the flow and after streamlining by a section of matrix tape, the gas mixture entered an observation cell. This consisted of a Syndanyo framework and thin mica windows, through which experimental observations were made. At the top of the cell the gas passed through into an exhaust section.

## 2. Problems of grid design.

In order to keep to a one-dimensional model, the hot grid has to heat the approaching gas uniformly. In practice this means that the 'pore' dimensions of the grid must be small in comparison with the thickness of the pre-heat zone. Construction of a suitable grid proved difficult, as had been anticipated. Various types of heating grid were tried, and two are shown in figure 4.4 Type (a) consisted of closely spaced nichrome wires fixed across a pair of insulating formers, which were grooved to keep the wire spacing constant at less than 1 mm. A series of light springs - not shown in figure 4.4(a) - kept the wires under slight tension, to prevent sagging due to temperature increases and consequent wire expansion. In addition, possible complications due to catalysis have to be considered, and steps were taken to coat the bare wire

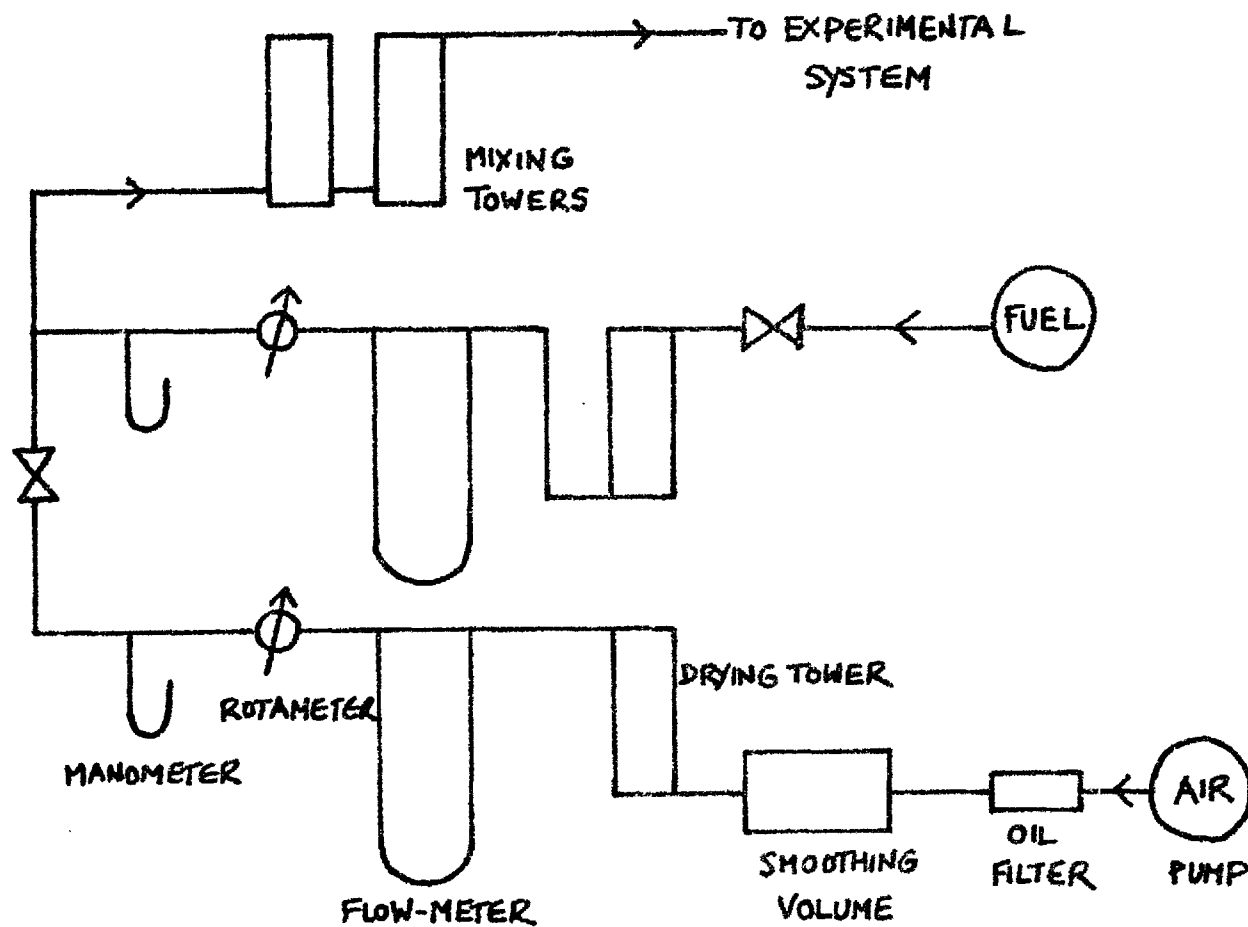


FIG.4.3

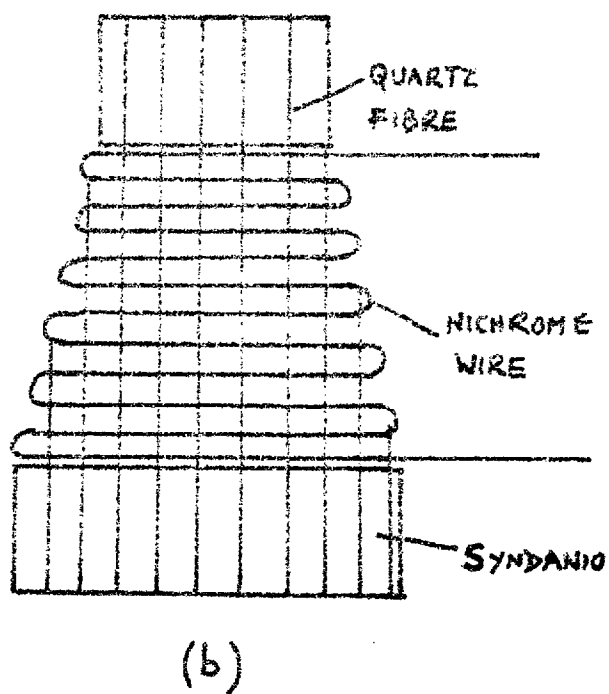
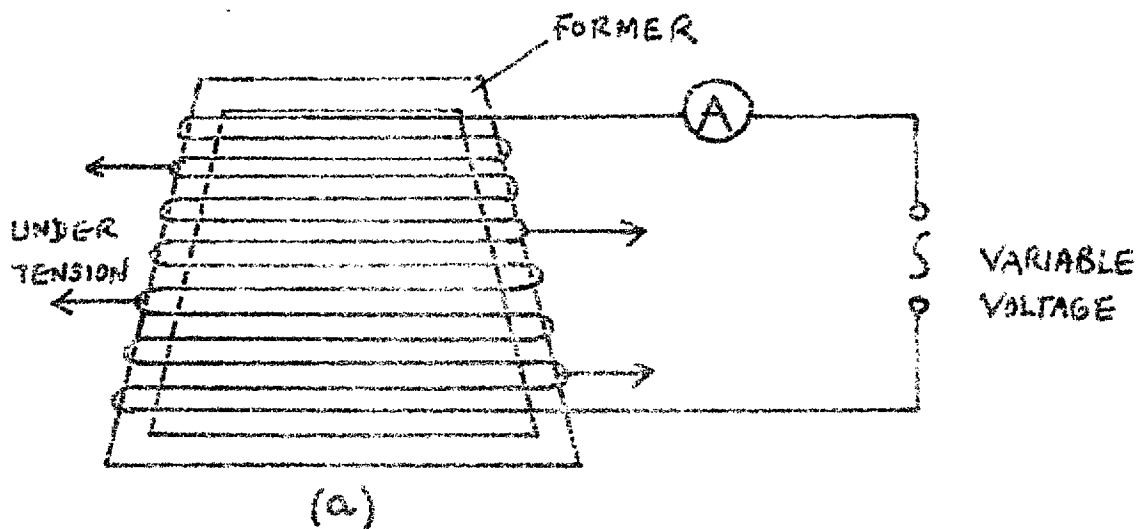


FIG.4.4

surface using the method due to Kaskan <sup>52</sup>, which has been used previously for protecting bare thermocouples exposed to reacting flame gases. The method used was to immerse the bare grid wire in a siloxane bearing premixed flame. The particles of silica present in the flame were deposited on the surface of the wire giving it a dull grey appearance. With the aid of a microscope, the thickness and homogeneity of the protective coating could be observed. It was found that, due to frequent thermal expansion and contraction of the wire, the silica surface cracked after a while and the process of recoating had to be repeated at intervals. Later, this form of grid was rejected because of practical difficulties associated with keeping closely spaced wires in one plane over a wide temperature range, without insulation breaking down. It was therefore decided to seek an alternative approach to this problem. Figure 4.4(b) shows the second type of grid developed. To construct such a heater, it was necessary to weave nichrome wire into an electrically insulated, porous medium. The technique adopted involved constructing a small 'loom'. Quartz fibre served as the warp, while the cross-threading or weft was introduced by means of a continuous feed of wire. A wire spacing of less than 0.5 mm. could be achieved in this way. On heating a grid of this kind no visible distortion arose,

because the wire network could expand freely in two dimensions within the plane of the quartz fibre without insulation breaking down. The heating current to the grid could be accurately controlled by means of a variac. Plate 1 shows the experimental assembly incorporating the second form of grid, figure 4.4(b). Certain difficulties arose with the 'uni-dimensional' model due, among other causes, to the presence of optical windows. The reason for ultimately rejecting the system in favour of a more complicated two-dimensional model will become clear, following the discussion in Chapter 6 on the preliminary work with the 'uni-dimensional' model. It was not fully appreciated a priori that there is a very fundamental point about obtaining a one-dimensional system in practice; namely that it is not possible to have at the same time, small holes, zero pressure drop and a stretched non-rigid system.

### 3. The 2-dimensional model.

In the 2-dimensional system, shown schematically in figure 4.5, optical windows were dispensed with. The arrangement is similar to the so-called Hoelscher burner<sup>62</sup>, where a flat, premixed flame burns against a solid plate facing the gas flow. Alternatively, two such flames are placed back to back and the solid plate is then dispensed with. The essential difference, in fact, between the one



PLATE 1.

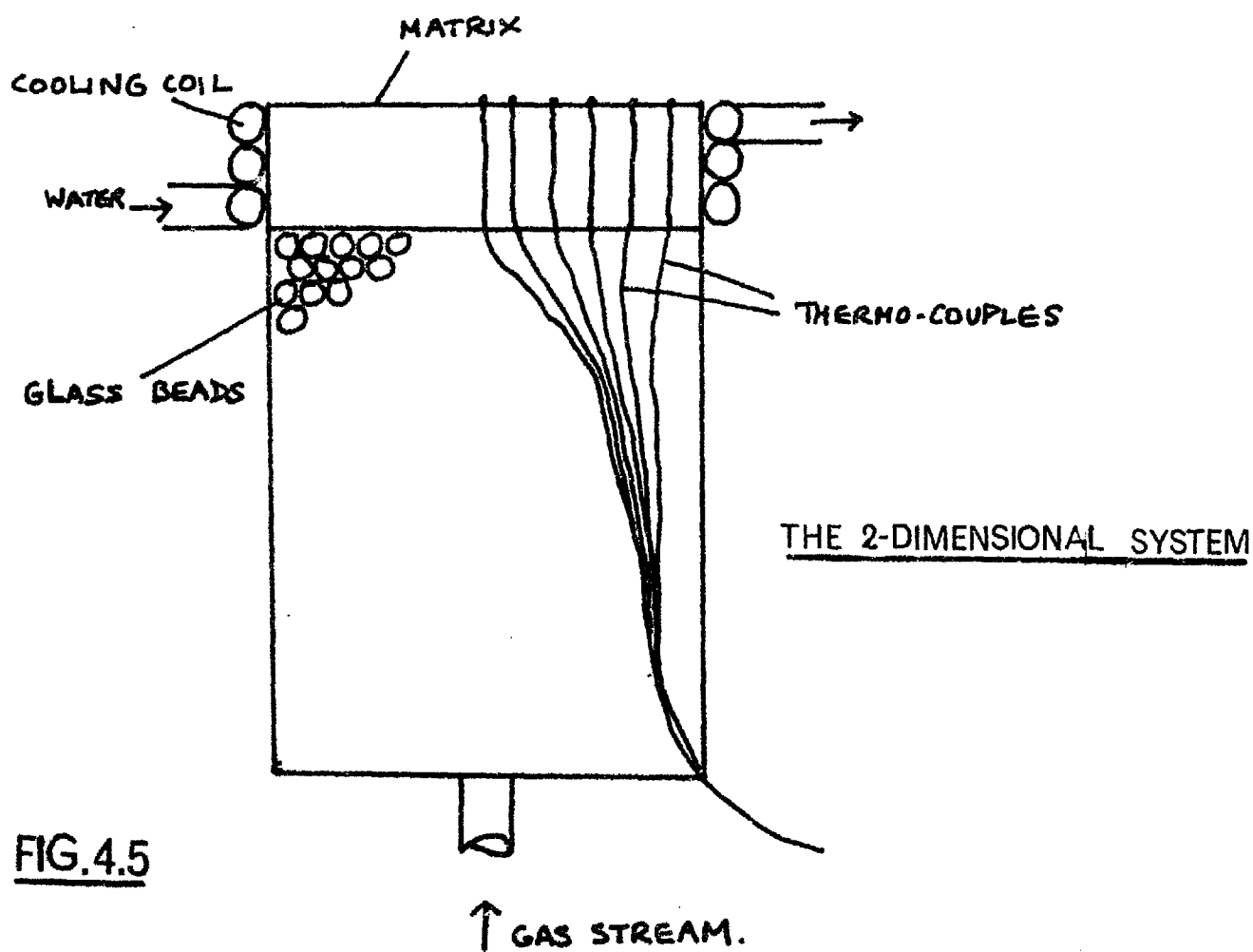
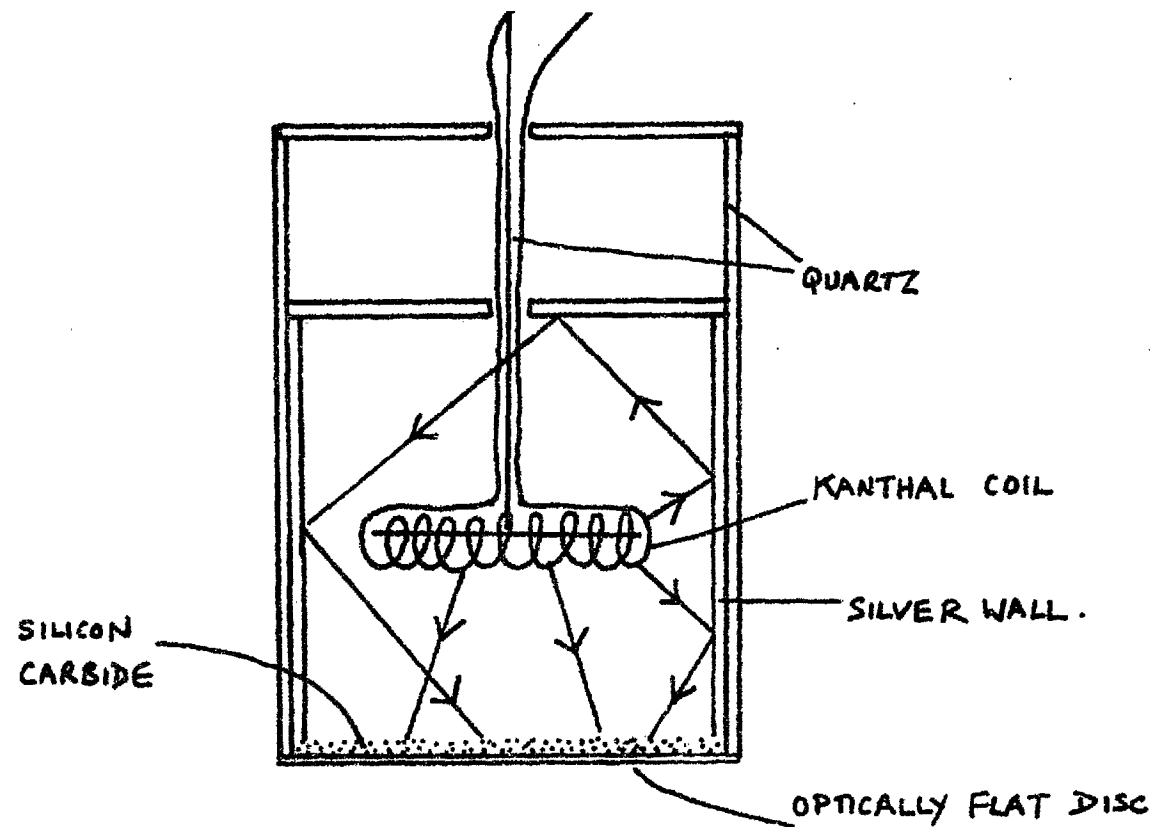


FIG.4.5

and two-dimensional systems used in the present work, insofar as they represent real flames, is that in one case the plate is porous and in the other the flow is made to diverge.

Referring again to figure 4.5, a stream of reactive gas, after passing through the lower ignition tube impinges on an optically flat base of a quartz cylinder. The base of the cylinder was designed to be less than 1 mm. thick, and approximately 6 cm. in diameter. Uniform heating of the thin disc offered a method for setting up an ideal non-catalytic hot surface, which would be capable of igniting the gas stream. Various methods of heating the disc were tried, including the use of a molten lead bath. Radiation heating however proved to be the most satisfactory approach to the problem. The upper portion of the quartz cylinder, shown in figure 4.5, comprised a radiant cavity. Thermal radiation from an electrically heated Kanthal wire coil, placed centrally as shown, was reflected in all directions by highly polished silver walls. An estimated 98% of the radiation emitted by the heater eventually fell on the base of the cylinder. In order to heat the quartz, which is transparent to infra red radiation, a thin layer of material with a high absorption coefficient needed to be placed in thermal contact with the upper surface of the disc. Silicon carbide proved ideal for this purpose, being an almost perfect black body with a very high conductivity. In

addition the material could be obtained as a fine powder, suitable for depositing evenly on the disc surface. Using the radiation method described, the surface temperature of the disc could be accurately controlled by varying the heater current. The entire assembly was supported vertically in a rigid stand, with the upper portion attached to an adjustable screw gauge, so that the distance between the lower matrix and hot surface could be altered. For most of the work, this distance was about 2 cm. The lower section of the apparatus was water cooled by means of a series of copper coils. Temperature distributions across the matrix could be determined using six thermocouples situated at the matrix surface as shown in figure 4.5. Plate 2 shows the experimental system in the test space of the interferometer used for the present investigation.

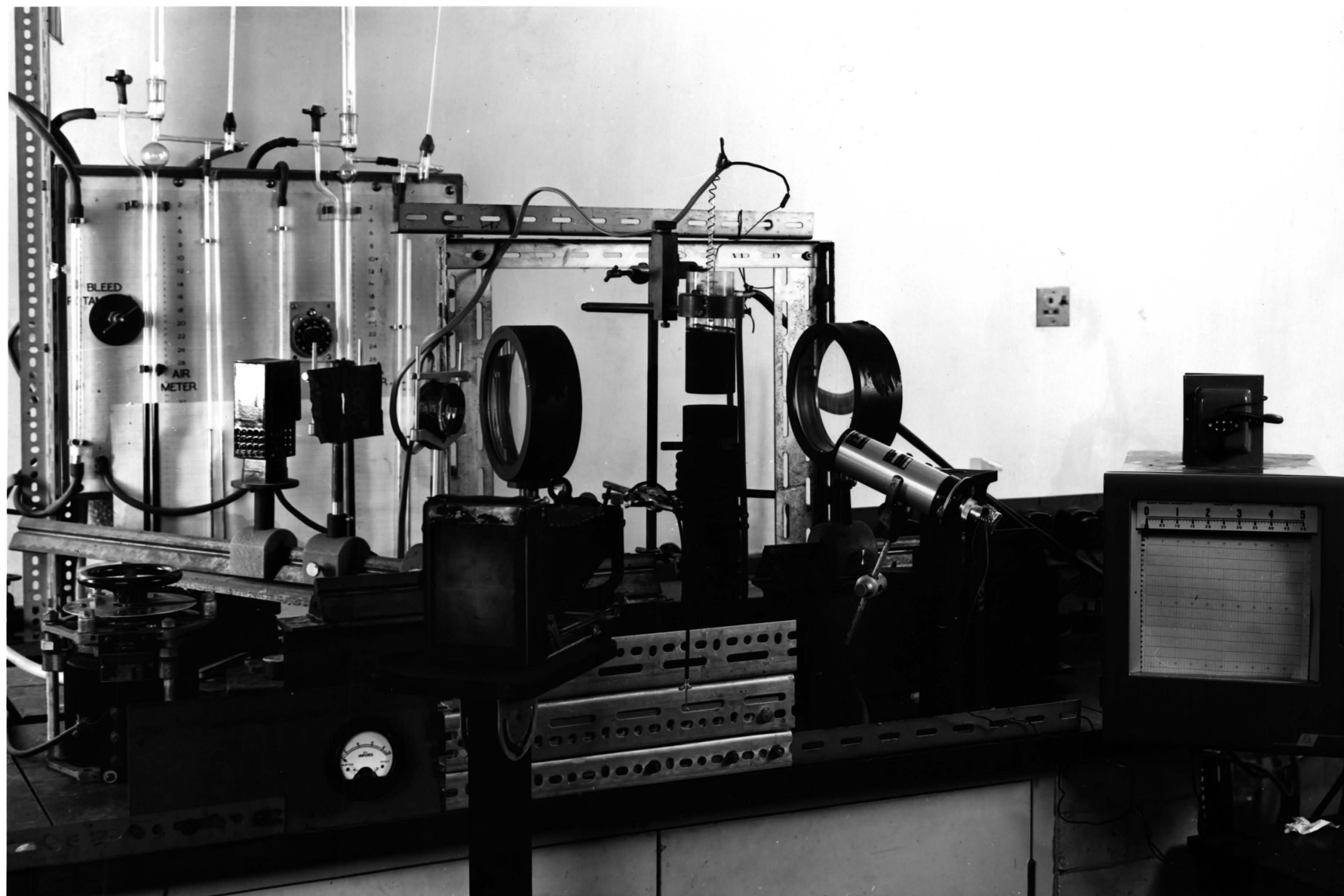


PLATE 2.

## CHAPTER 5.

DEVELOPMENT OF THE OPTICAL SYSTEM.

1. For the reasons outlined in Chapter 3, an optical system was developed to function primarily as an interferometer, but which could, if necessary, be modified for deflection mapping studies. Very few interferometers can be successfully applied to combustion studies for the following reasons. There must be appreciable separation between the test and reference beams in order to keep the optical path of the latter constant and independent of the test space. The test beam should traverse the test space once only, in order to avoid excessive deflection by the refractive index field through which it passes. The consequences of such deflection as does occur are minimised if it is possible to focus the test space onto a receptor.

The Mach-Zehnder interferometer<sup>63,64</sup> has found frequent application in aerodynamic studies. Two disadvantages of this optical instrument are the very high cost of large optical flats involved and the high sensitivity to misalignment. On the other hand, the optical arrangement facilitates large separation of the test and reference beams and maximum use of the available light flux. An instrument developed especially for combustion studies is the four grating interferometer<sup>65</sup>. The optical layout

is essentially similar to the Mach-Zehnder, employing four diffraction gratings in place of the optical flats.

Sensitivity and relative cost of the instrument are low, but, for time variant phenomena, the light losses involved are excessive on account of the discarded diffraction orders at the several gratings. The most suitable optical system for the present work appeared to be that based on the two diffraction grating interferometer due to Kraushaar<sup>66</sup>.

Major advantages of this system include relatively low cost, large field of view, low sensitivity to misalignment and ready conversion<sup>67</sup> to a deflection mapping system. In all past work<sup>67,68</sup> the Kraushaar system has been used with schlieren mirrors as the main optical components, but in the present investigation large aperture, long focal length lenses were used instead. One advantage of using lenses is immediately obvious by reference to figure 5.1 which shows schematically the difference in the two layouts. The test-space distance between the lenses  $L_2$ ,  $L_3$  can be made as small as desired (or as safe as the test object permits). With mirrors the corresponding distance is, of geometric necessity, greater than  $2f$ , and, due to the inevitable presence of small temperature gradients and consequent convection currents in the laboratory, path differences arise between portions of the initially coherent test and reference beams to give so-called 'floating' fringes<sup>69</sup>. This undesirable effect

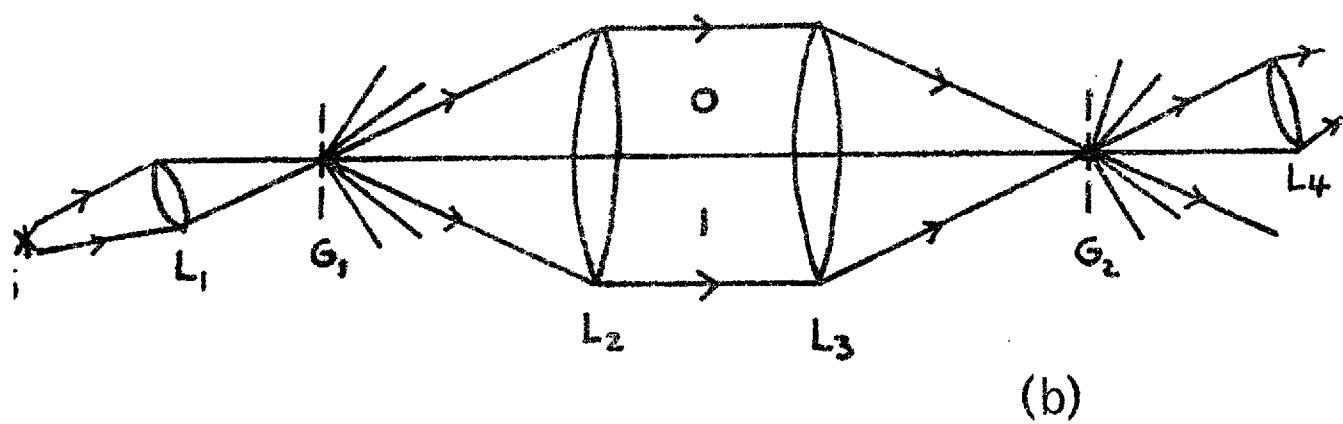
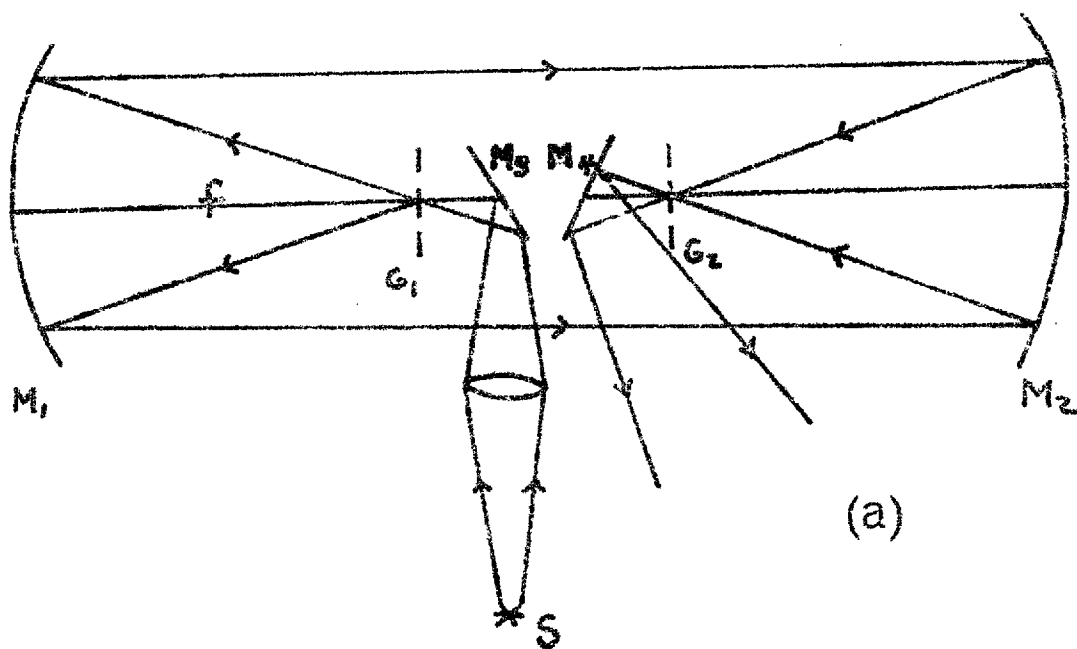


FIG. 5.1

is severely reduced, if not eliminated, by using lens components since the overall differential path lengths then involved are very small. The available test space area is also a maximum in the lens systems, unlike the mirror interferometer, where, for an axial system as shown in figure 5.1a,  $M_3$  and  $M_4$  partly obstruct the test beam. This loss of test-space area is avoidable by employing an off-axis system, but this gives rise to spurious fringes due to aberration effects.

## 2. Principle and Theory of the Interferometer.

The principle of the optical arrangement may be demonstrated by reference to figure 5.1(b). Light from an effective point source  $S$  is focused onto a diffraction grating  $G_1$ . Every ray within the bundle of light incident at  $G_1$  gives rise to its several diffraction orders. All but two orders, shown here as the zero and first, are lost to the system by arranging for these two orders to just fill the aperture of  $L_1$  symmetrically, with  $G_1$  at the focal point of  $L_1$ . The two orders then emerge from  $L_1$  as parallel light and serve as the test and reference beams of the interferometer. Beyond  $L_2$ , re-convergence of the two beams occurs at  $G_2$ , giving rise to two sets of diffraction orders. The orders of one set then overlap with the second set with a constant difference in order number of unity.

Straight lined 'field' fringes, like those shown in Plate 3, are observed on a screen when  $G_2$ , in figure 5.2, is displaced from the focal point of  $L_3$  by an amount  $\Delta x$  to  $G'_2$ . This effectively increases the angle between the initially parallel wavefronts to some finite value. In figure 5.2 the -1 order of the zero order beam overlaps with the zero order of the first order beam. Virtual displacement of the focal point at  $G_2$  perpendicular to the optical axis by a distance  $\Delta y$ , corresponds to an angle between the wave fronts of  $\alpha = \frac{\Delta y}{f}$ . The fringe spacing  $q$  is then given by :

$$\Delta y \, q = f \lambda \quad (5.1)$$

But  $\Delta y = \Delta x \tan \beta$  - see figure 5.2

$$= \frac{\Delta x \sin \beta}{\sqrt{1 - \sin^2 \beta}}$$

$$\text{and } \sin \beta = \frac{\lambda}{d}$$

$$\text{hence } \Delta y = \frac{\Delta x \lambda / d}{\sqrt{1 - \lambda^2 / d^2}}$$

$$\text{if } \left( \frac{\lambda}{d} \right)^2 \ll 1.$$

Substituting for  $y$  in (5.1)

$$q \approx \frac{fd}{\Delta x} \quad (5.2)$$

This suggests that the fringe spacing is, to a first approximation, independent of  $\lambda$  so that, in principle, white

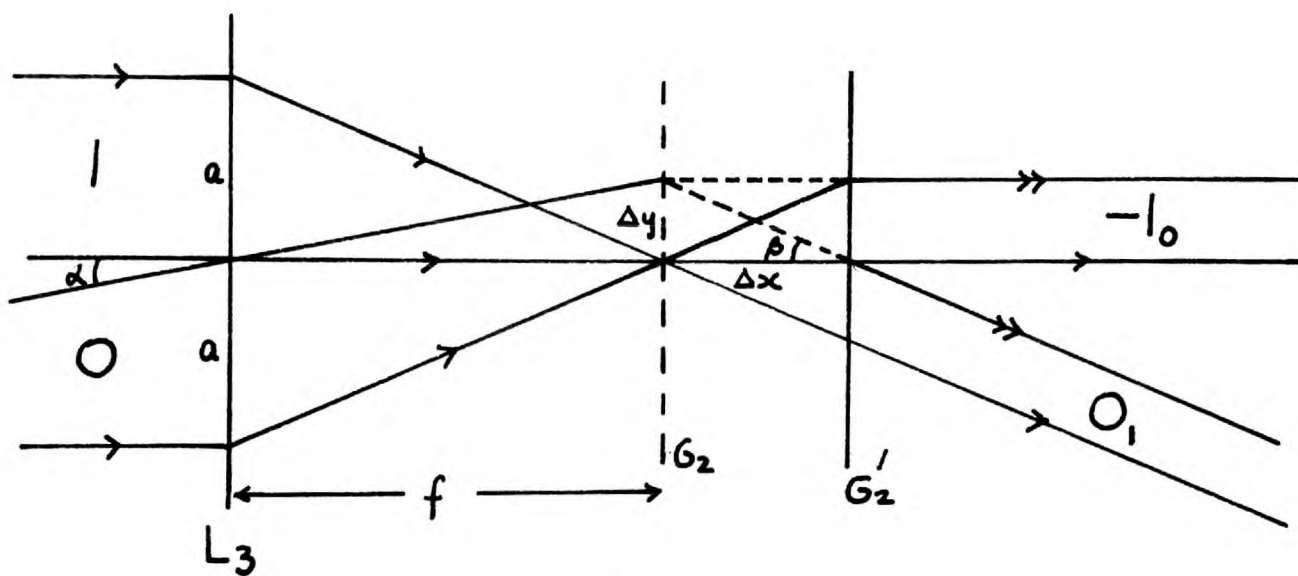
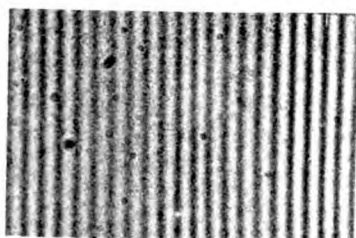


FIG.5.2



0,1 ORDERS PARALLEL

0,1 ORDERS NON-PARALLEL



fringes should be observed <sup>70</sup>. However, in practice it was found necessary to restrict the light from a high pressure mercury vapour lamp to a small wave-band, because the lens components showed appreciable chromatic aberration, making  $\Delta x$  a noticeable function of  $\lambda$ . Sets of overlapping, coloured fringes were recorded using white light.

To use the instrument for recording contours of equal optical path differences between the test and reference beams,  $G_2$  must be at the focal point of  $L_2$ , so that the interfering wavefronts are parallel. Ideally, a uniform distribution of light intensity should be observed across the screen in the region of overlapping orders. The system is then at the 'infinite fringe condition'. In practice a residual interference pattern was observed, due to optical imperfections in the manufacture of the schlieren lenses, but, in the absence of a changing refractive index field, these remained quite stationary. Plate 4 shows the field of view at the closest approach to the infinite fringe condition. When interpreting subsequent interferograms, any residual fringes due to optical defects in the system were taken into account. Conversion to a deflection mapping system can be very simply achieved by removing  $L_2$  and  $G_2$  and placing an inclined slit or half-wave step on either side of the test object. The versatility of the optical system is further demonstrated by reference to Plate 5, which is

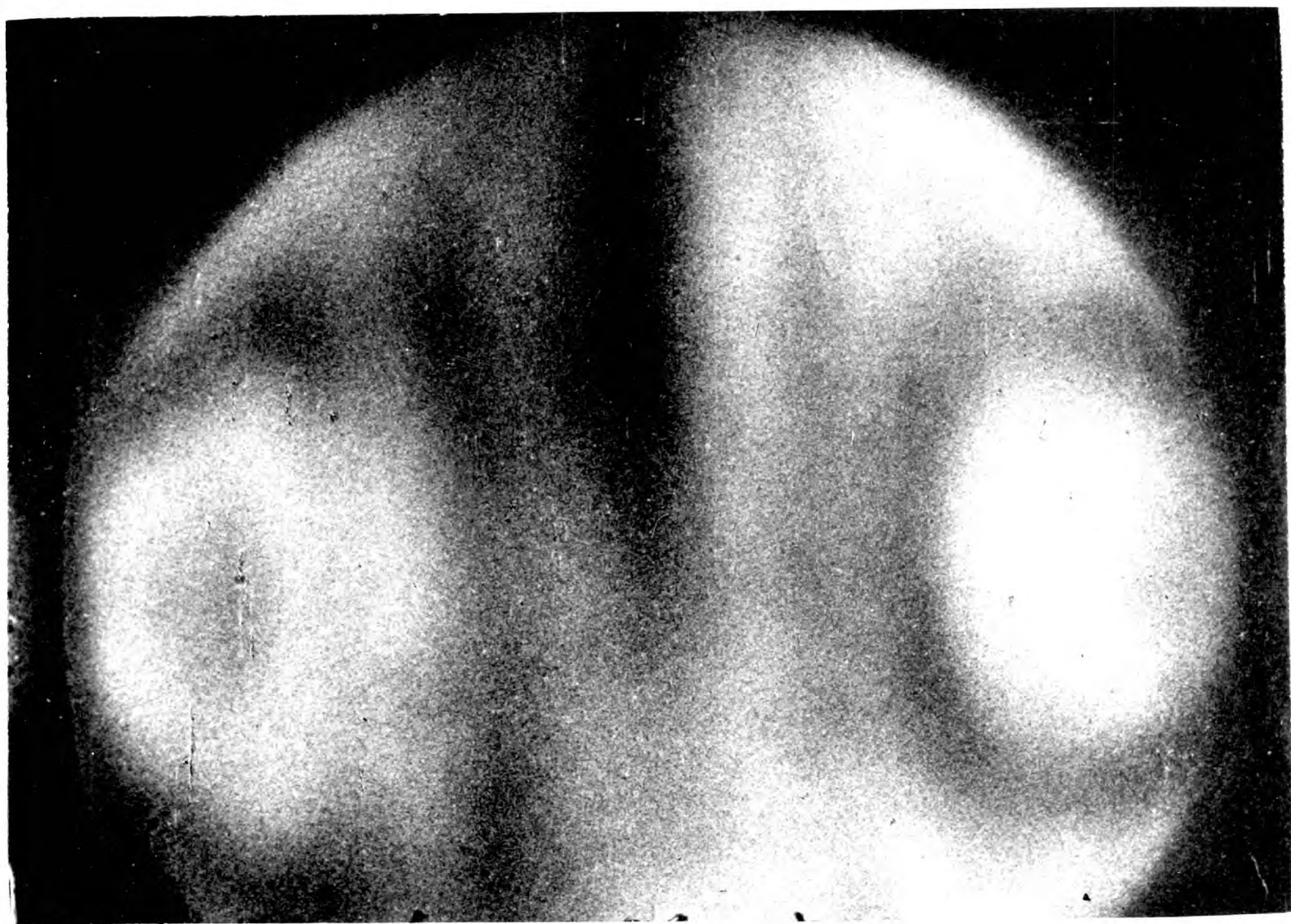


PLATE 4.

a schlieren photograph of a premixed flame obtained by placing a knife edge at  $F_2$  and removing the grating  $G_2$ .

An important feature of the two diffraction grating interferometer is the small sensitivity to misalignment at the infinite fringe setting. If  $\Delta x$  is the error in positioning  $G_2$  at  $F_2$ ,  $\Delta x q = \frac{f^2 \Delta}{a}$  - obtained by substituting  $d = \frac{\lambda}{\sin a/f}$  into equation (5.1) and making the approximation  $a/f \rightarrow 0$ . Calculation for the optical system used in this work gives  $\Delta x q \approx 0.2$  cm. This means that if  $G_2$  is out of position by as much as 0.2 cm, a fringe separation of only one order per cm would be observed. Positioning  $G_2$  to  $\pm .001$  cm presents no problem if a micrometer screw gauge is employed.

### 3. Preliminary setting up of the interferometer.

During initial setting up of the interferometer, it was noted that the effective test-space area was restricted, due to incomplete superposition of the two sets of interfering orders at  $G_2$ . This arose because the gratings and lens components were not matched correctly. Initially, two achromats, of 12.7 cm. and 15.2 cm. aperture and 183 cm. and 193 cm. focal length respectively, were used with 2,000 l.p.i. replica transmission gratings. For the two adjacent orders to just fill the aperture of the first schlieren lens, as described earlier, (see figure 5.1(b)),

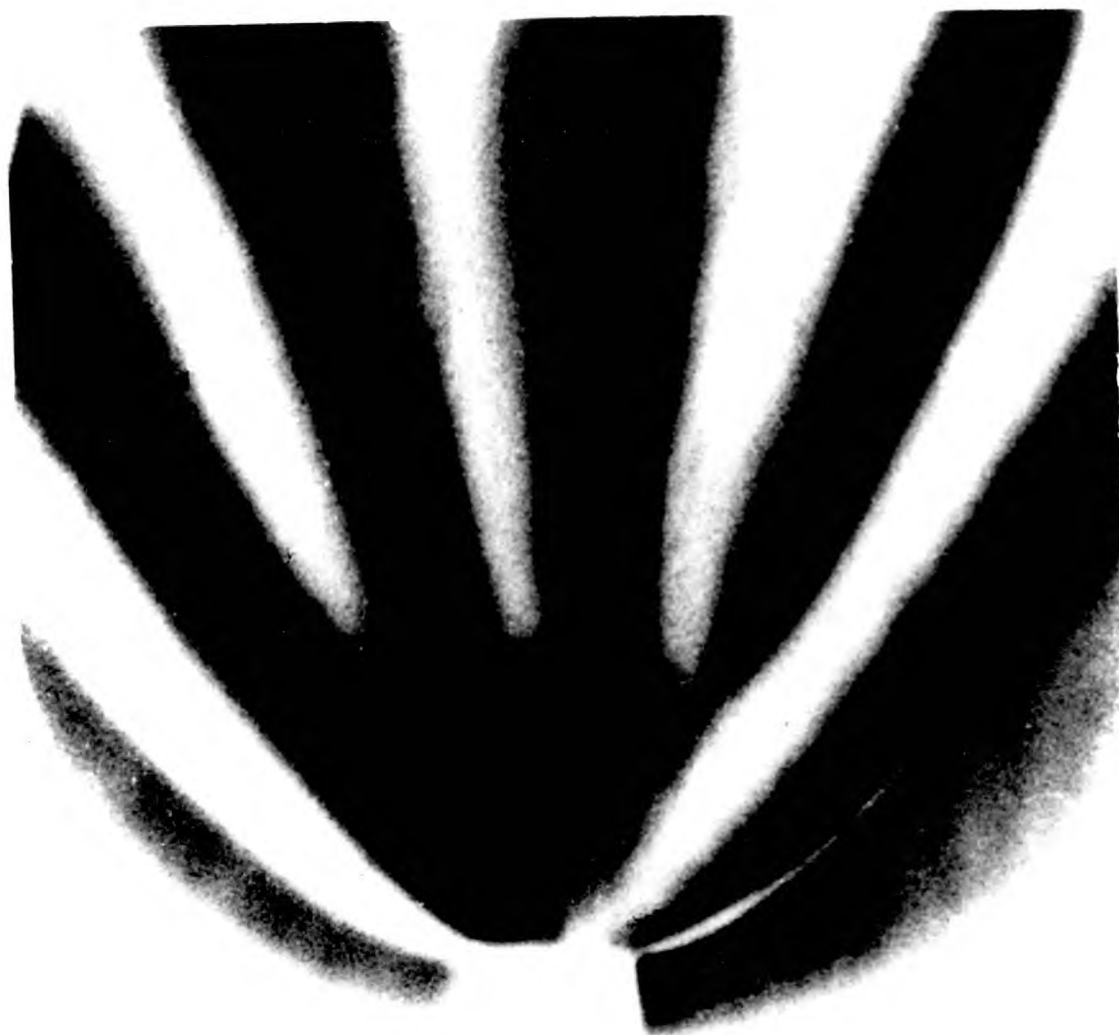


PLATE 5.

the following relationship has to hold :

$$\sin a/f = \frac{\lambda}{d}$$

where  $d$  is the ruling spacing of the grating,

$2a$ ,  $f$  are the aperture and focal length of the lens. The resulting restriction on test-space area was too severe to be useful, and a pair of almost identical achromats of 12.0 cm. aperture were therefore obtained, which exactly matched the gratings. The reason for choosing to match lenses to gratings was because to have done the converse, i.e. match the gratings to the two different lenses, would have been far too expensive, since the correct gratings would have to be made specially. In addition, the 2,000 l.p.i. replica gratings used were both cheap and readily replaceable. A number of 2,000 l.p.i. gratings were examined with a photo-cell device to compare the intensities of their various diffraction orders. Using this method, a suitable grating, with adjacent orders (3rd and 4th) of comparable transmission intensities, was selected and employed in subsequent work. The choice of orders had a great effect on the fringe legibility. Maximum contrast was observed when the interfering orders were of equal intensity. By using portions of the same grating for  $G_1$  and  $G_2$ , and reversing the ruling pattern on  $G_2$  with respect to  $G_1$ , slight differences in intensity between the chosen orders could be eliminated.

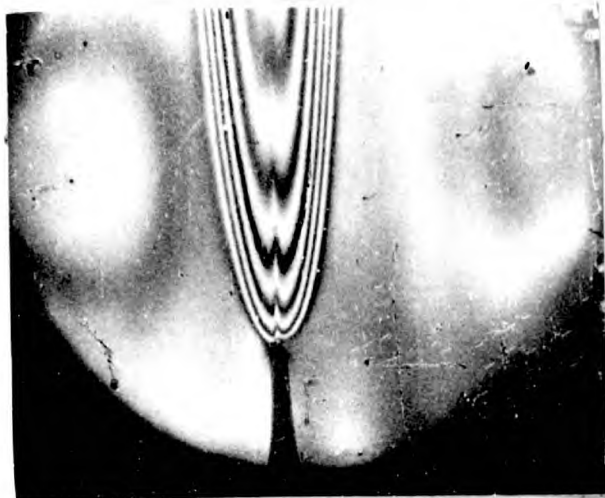
#### 4. Experimental Procedure for setting up the Interferometer.

To set up the interferometer, the following procedure was adopted. Light from a high-pressure mercury arc lamp was passed through a narrow band interference filter which transmitted at about 5500 Å. The filtered light was then focused onto a 2 mm. diameter aperture adjacent to the ruled surface of the first grating. Resulting diffracted images from the first grating were arranged to just touch each other by adjusting the effective aperture of the projection lens  $L_1$ . This section of the optical system was next rotated and positioned until  $G_1$  was at the focal point of  $L_2$  with the 3rd and 4th orders just filling the aperture of  $L_2$ . A plane mirror, placed on the far side of  $L_2$ , was used for this. If  $L_2$  and  $L_3$  were correctly lined up along a common optical axis, a series of multiple images of the hole at  $G_1$  were observed on looking in the direction of the source from the side of  $L_3$  nearest to  $G_2$ . All optical components up to and including  $L_3$  were found to be correctly aligned when these multiple images appeared along a single straight line. Once such a state had been achieved, the production of fringes required little more than placing  $G_2$  near the focal point of  $L_3$  and rotating the grating slowly. Fringes of maximum contrast were observed when the ruling on the two gratings were exactly

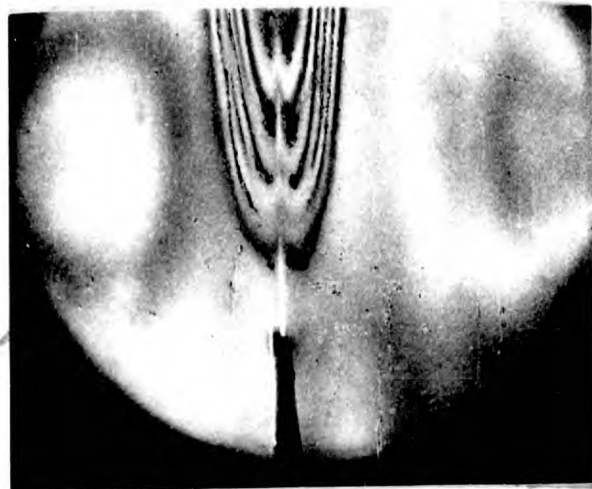
parallel. With the new set of lenses a residual fringe pattern was observed at the infinite fringe condition.

#### 5. Exposure Trials.

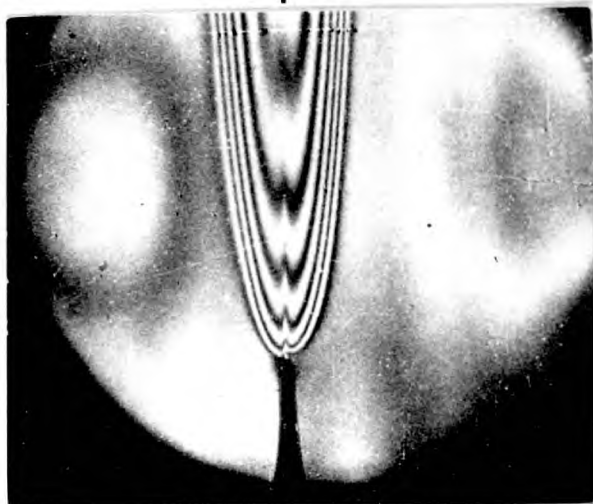
Preliminary ignition experiments with the 'uni-dimensional' model demonstrated certain difficulty in achieving an absolutely steady-state, just prior to ignition. In practice, the matrix temperature gradually increased due to radiation, and although the drift was gradual enough to assume a series of quasi-stated states, the system degenerated spontaneously to flame formation. In order to 'freeze' the critical conditions preceding ignition, therefore, it was important to record the process at an adequate framing rate. Trial exposures, using an H/16 Bolex 16 mm cine camera with H.P.3 and H.P.S. film, were carried out on time-variant refractive index fields. A combination of H.P.3 film at a framing rate of 24 f.p.s. was found to give the best results in terms of fringe legibility. One of the trial exposure experiments, using a small premixed flame burner, provided some interesting (though incidental to the subject of the present research) interferometric records of the blow-off conditions at a burner port <sup>71</sup>. This phenomenon occurs when the critical velocity gradient is exceeded in the approach stream of the premixed reactants. Plate 5 shows this sequence of events.



1



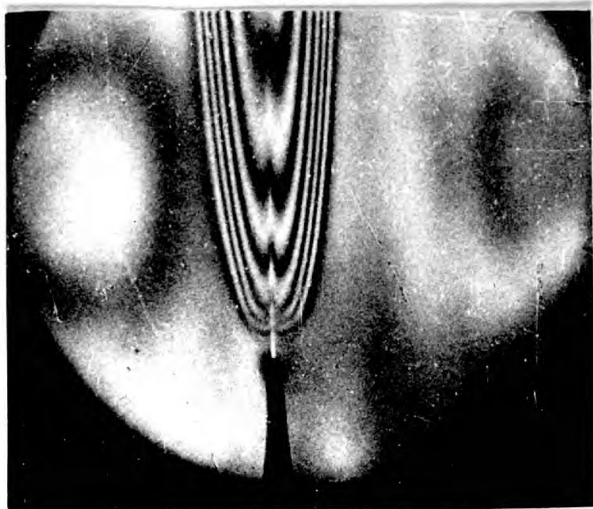
4



2



5



3

PLATE 6.

For the studies with the uni-dimensional model, a framing rate of only 16 f.p.s. was used, because the mica windows in the observation cell absorbed and reflected an appreciable fraction of the incident light. Even at 16 f.p.s. the fringe patterns appeared to be frozen, and therefore helped to justify the assumption of a series of quasi steady states prior to ignition onset.

## CHAPTER 6.

PRELIMINARY STUDIES WITH THE UNIDIMENSIONAL SYSTEM.

1. Before discussing the experimental aspects of the first system in greater detail, it would be convenient to consider briefly the essential steps involved in determining the volumetric heat release rate ( $q$ ) as a function of temperature as outlined previously. For this, the distributions of temperature and flow velocity upstream of the grid have to be determined. If these are approximately unidimensional, the equation for heat conservation, in the form of equation (1.2), can be applied to the system. Taking ( $z$ ) as the direction coordinate of gas flow, for a reacting fuel/air mixture we have :

$$-\frac{d}{dz} \left( -K \frac{dT}{dz} + v \rho \bar{c} T \right) - q = 0 \quad (6.1)$$

where  $K$ ,  $v$ ,  $\rho$ , and  $\bar{c}$  are the thermal conductivity, velocity, density and mean specific heat at constant pressure respectively. Using experimentally determined temperature and velocity profiles, graphical differentiation gives local values for  $\frac{dT}{dz}$ ,  $\frac{d^2T}{dz^2}$  and  $\frac{dv}{dz}$ .

But in order to solve for  $q$ ,  $K$ ,  $\rho$ ,  $\bar{c}$  must be known as functions of temperature as well. For any gas mixture it is theoretically possible to calculate the variation of

these quantities with temperature from known data <sup>72</sup>, but in principle it is possible to employ a less tedious method using the unidimensional system.

If the heat conservation equation is applied to a non-reactive system, such as air,  $q = 0$  and

$$-\frac{d}{dz} \left( -K \frac{dT}{dz} + v \rho \bar{c} T \right) = 0$$

Integrating gives :

$$-K \frac{dT}{dz} + v \rho \bar{c} T = v_0 \rho_0 \bar{c}_0 T_0$$

and dividing through by  $\rho \bar{c}$ ,

$$-\mathcal{D} \frac{dT}{dz} + vT = \frac{v_0 \rho_0 \bar{c}_0 T_0}{\bar{c}} \quad (6.2)$$

where  $\mathcal{D} = \frac{K}{\rho \bar{c}}$  is the thermal diffusivity.

But  $\rho_0 T_0 = \rho T$  for an ideal gas.

Substituting into (6.2) for  $\rho$

$$\mathcal{D} \frac{dT}{dz} + vT = \frac{\bar{c}_0}{\bar{c}_T} \cdot v_0 T$$

$$\mathcal{D} = T \frac{\left( v - \frac{\bar{c}_0}{\bar{c}_T} v_0 \right)}{dT/dz} \quad (6.3)$$

Distributions of  $\mathcal{D}$  as a function of temperature can therefore be obtained for a non-reacting gas - e.g. air, or fuel only, from knowledge of the temperature and velocity distributions in the gas. In the case of a reacting gas mixture the cold

thermal diffusivity can be deduced by plotting the function in (6.3) and extrapolating to the cold temperature boundary. In the present work, where very dilute ethylene/air mixtures were employed, the effect of reaction on thermal diffusivity could be assumed small and the variation with temperature may be obtained by using the deduced or calculated cold thermal diffusivity with the same temperature variation found for air. Substitution into the heat conservation equation for the conductivity data yields  $q$  as a function of temperature.

## 2. Experimental arrangement.

Investigations into the possibility of using the unidimensional system were carried out with a prototype wire grid, of the type shown in figure 4.4a. Most of the work at this stage was done with air only, in order to establish the method of measurement and form of temperature and velocity distribution in the experimental model, before extending the study to a reactive gas. The position of the model in relation to the interferometer and particle-track system is shown in figure 6.1. The extension of the cell optical windows into the reference section of the interferometer was necessary in order to keep the test and reference beam intensities comparable. If this precaution was not observed, very poor fringe contrast resulted.

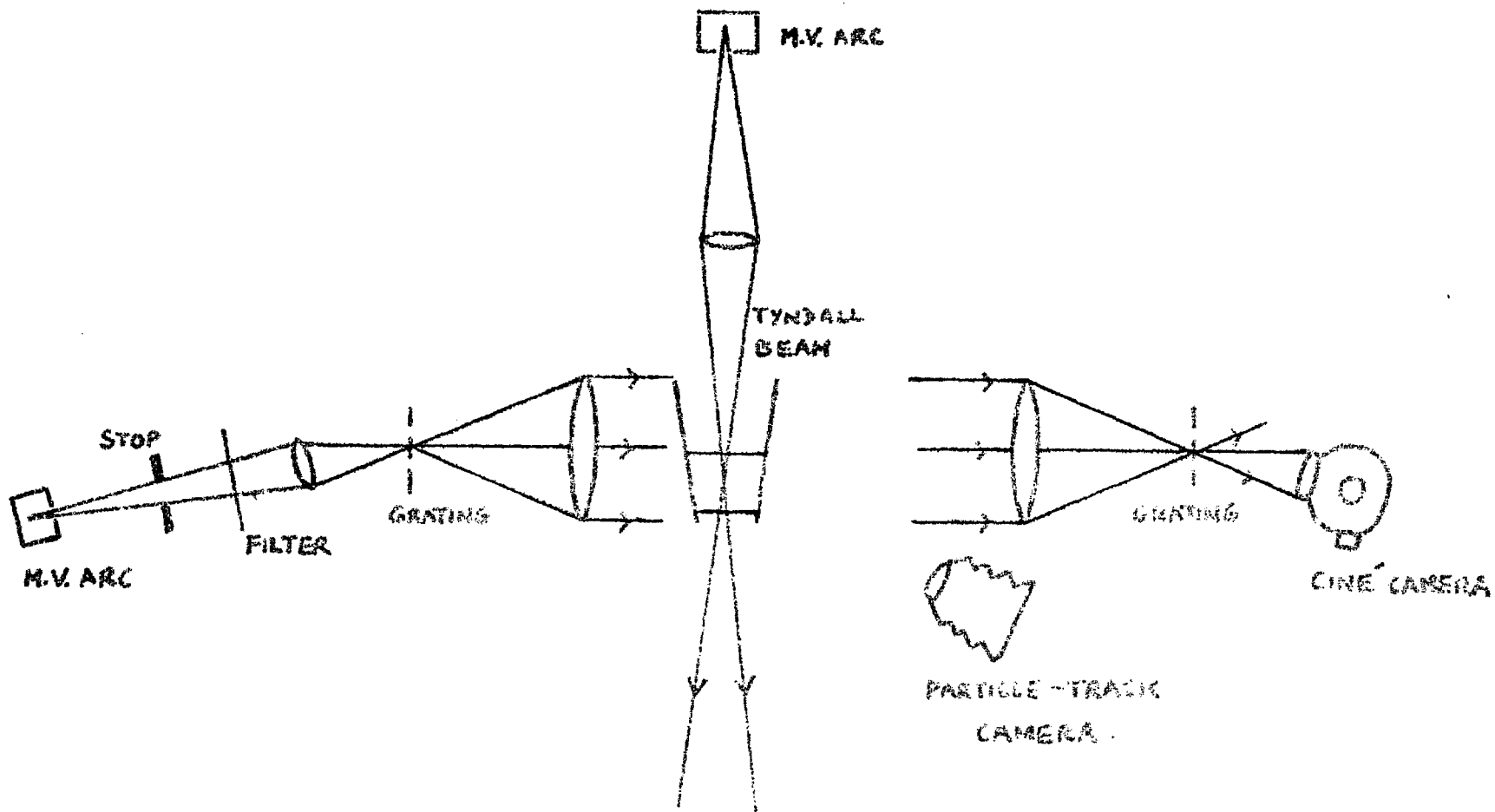


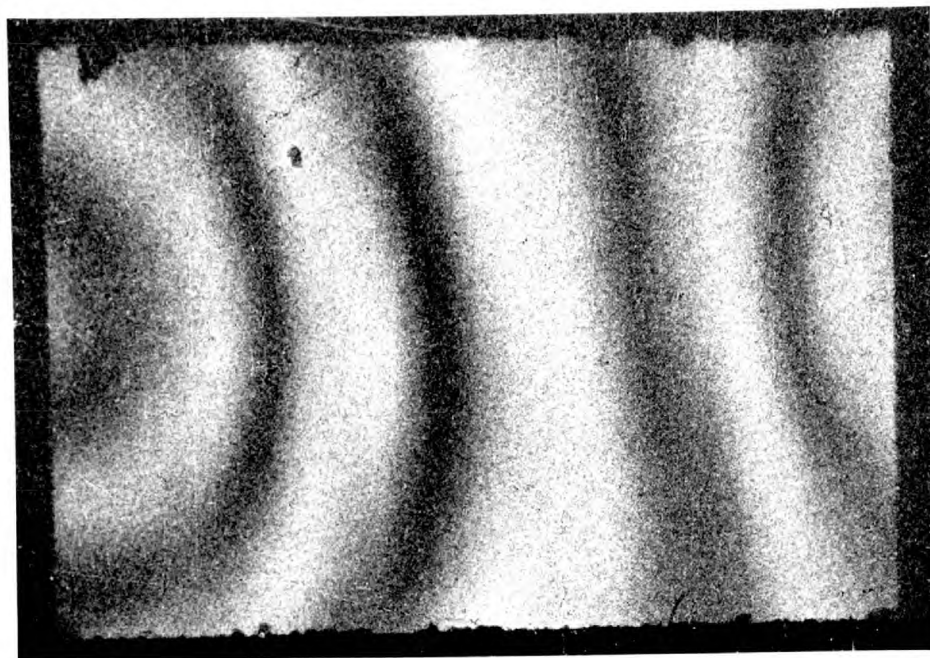
FIG. 6.1

### 3. Velocity profiles.

The velocity profiles were derived from measurements of particle-track photographs, using the A.C. mains frequency operating the light source as a time base. Measurements made over a wide flow range indicated that the profiles were reasonably flat. Figure 6.2 shows velocity profiles for various flow rates just upstream of the hot grid (broken line) and near to the entrance of the observation cell (full line). The velocities were plotted against a co-ordinate,  $\lambda$ , as defined in figure 6.2. These experiments were conducted by altering the position of the Tyndall beam. The slight asymmetry of the profile reflects the increase in viscous forces at the narrower end of the tube.

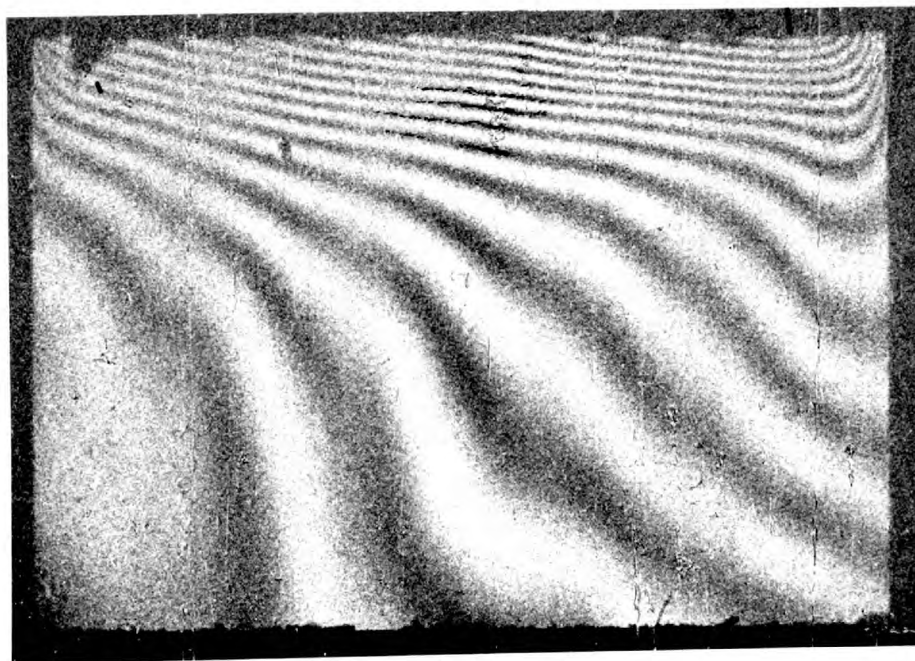
### 4. Temperature profiles.

Interferograms of the test region with the grid cold and hot are shown in Plate 7, with air flowing slowly through the system. The residual fringes in Plate 7(a) were caused by optical path differences across the mica optical windows and other optical defects. In the absence of a varying refractive index field the position and number of these fringes remained reasonably constant. For the purpose of interpreting subsequent test interferograms, it was important not only to record the residual fringes, but



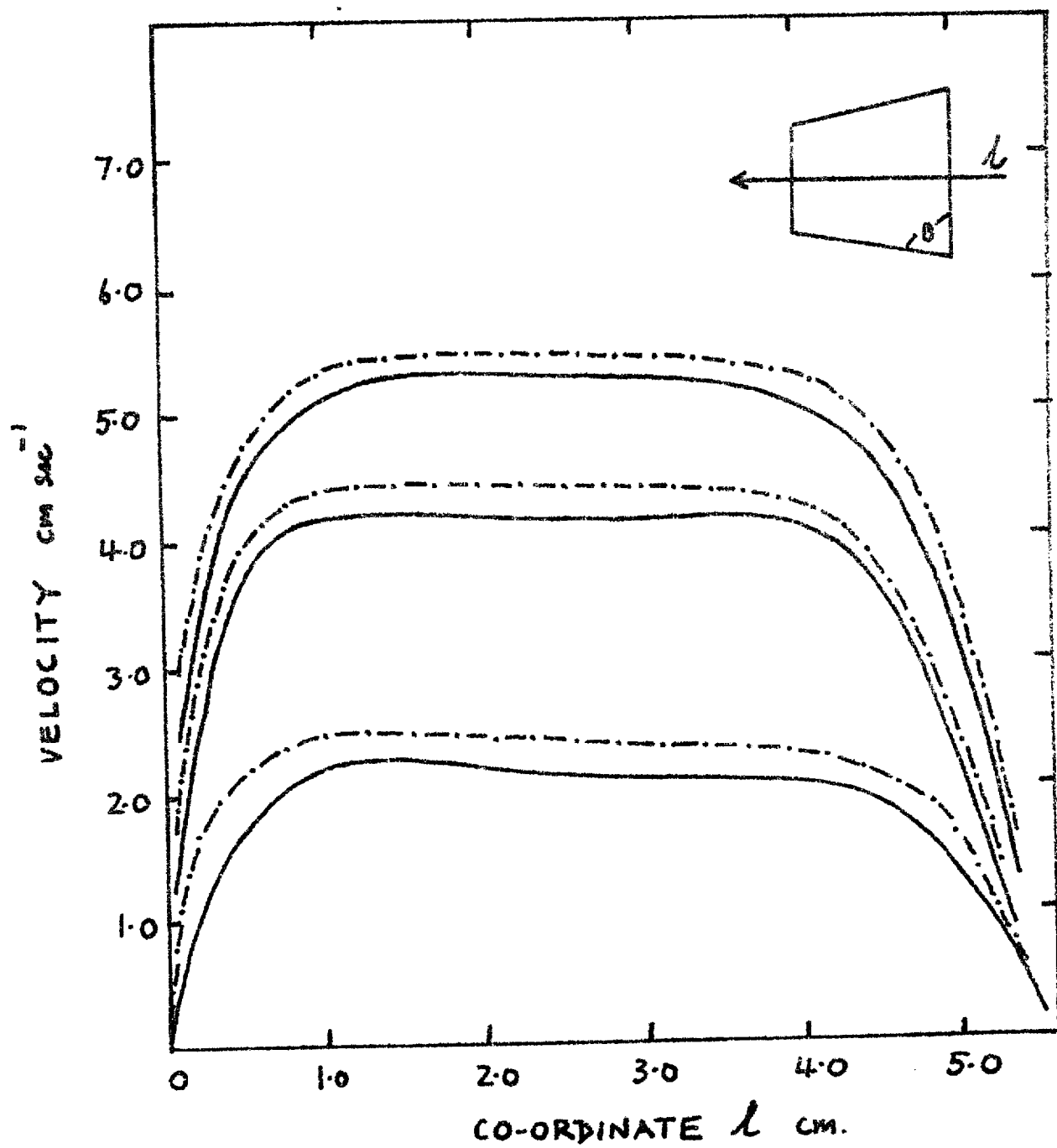
cold

(a)



hot

(b)

FIG.6.2

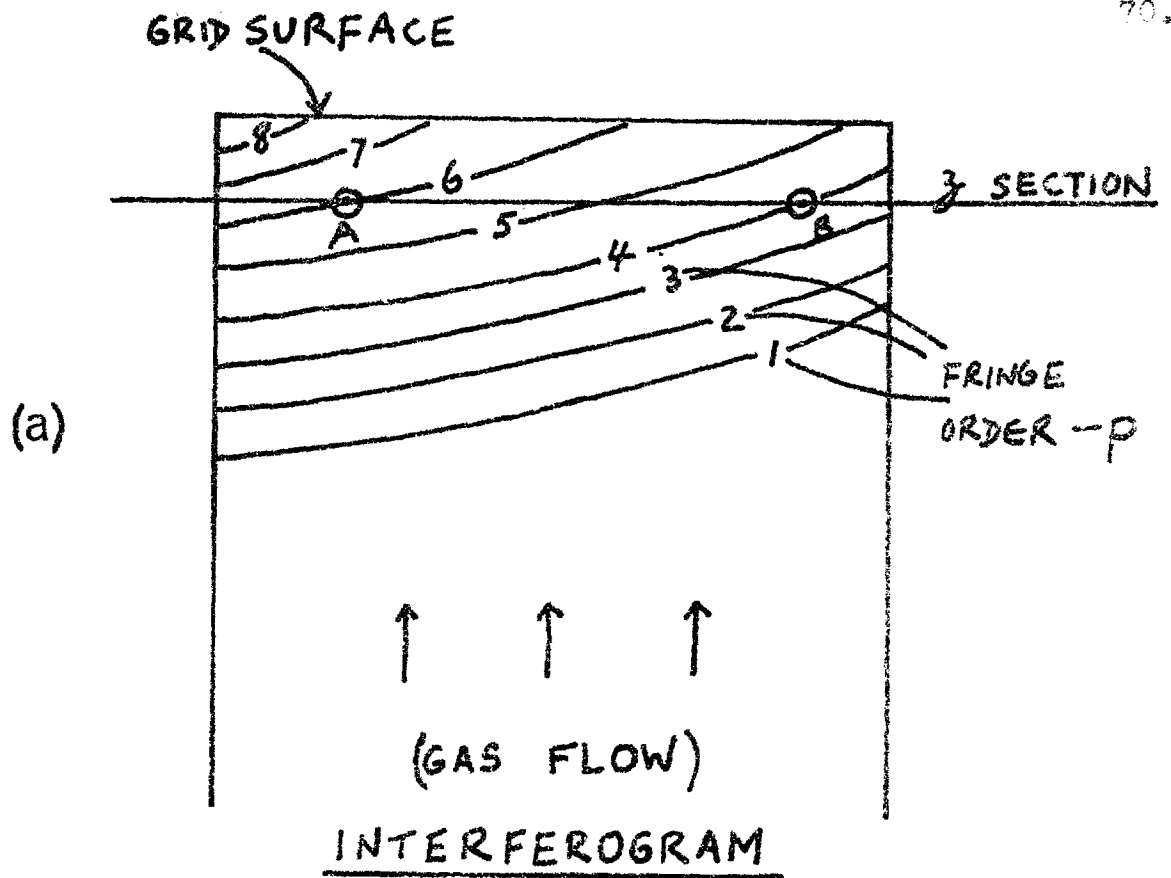
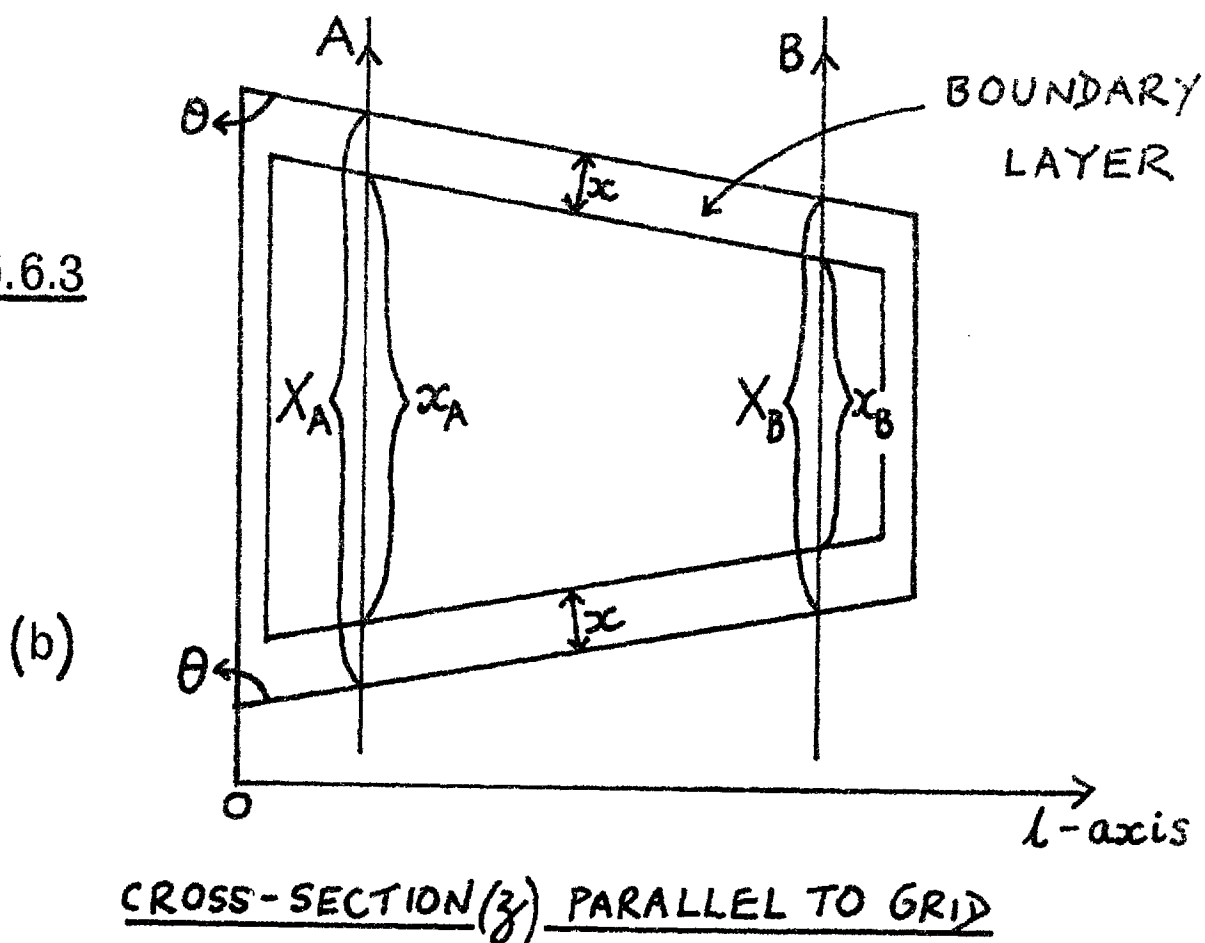


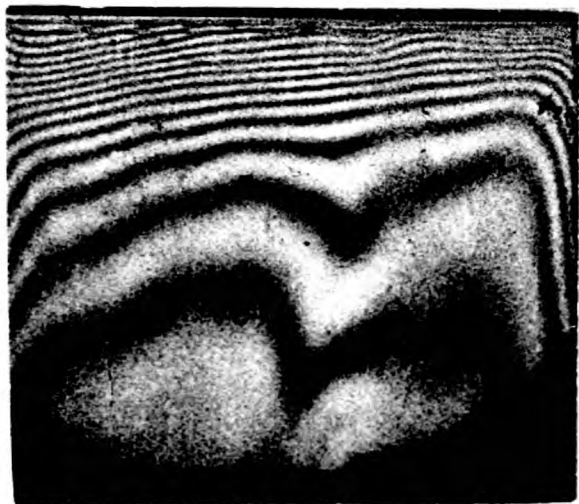
FIG. 6.3



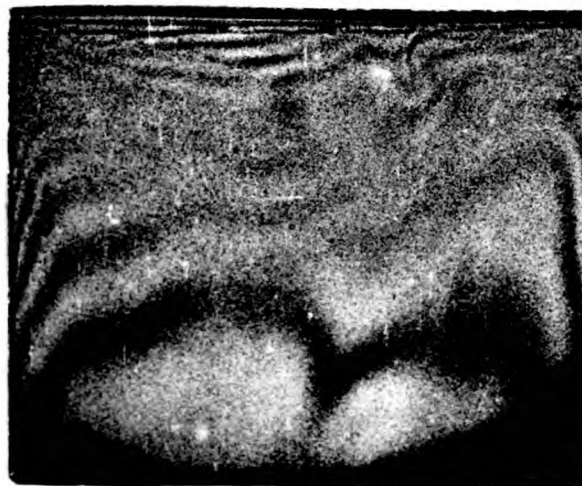
also to determine whether they should be added to or subtracted from the fringe order due to the test space, i.e. whether  $\int_{\text{Test}} n \, dx - \int_{\text{Ref}} n \, dx$  is negative or positive. Here,  $n$  refers to the refractive index of the mica. The legibility of the fringes near to the hot surface of the grid was reasonable, as can be judged by the sequence of consecutive interferograms in Plate 8, recording the ignition onset of an ethylene/air mixture at very low flow rates. The degeneration of the steady state to flame formation is reflected in the obvious change of fringe pattern. Vertical fringes in the latter frames suggest that, following ignition, all the gases remain at the final temperature for a time.

##### 5. Interpretation of interferograms for the unidimensional system.

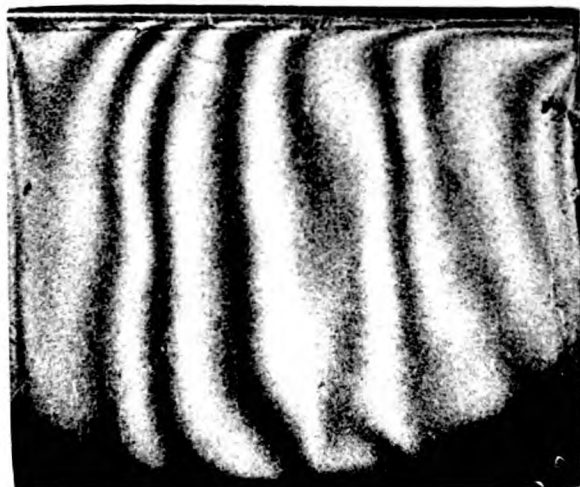
Consider a horizontal section through the cell parallel to the grid, as shown in figure 6.3. Under conditions of one-dimensional flow and heating, a large central section of any plane  $z$ , will be at a constant temperature  $T_z$ . However, a boundary layer adjacent to the containing walls will be present, in which the temperature is not constant. The difference in optical path length between a test ray A and corresponding reference ray in the reference beam can be expressed as :-



1



2



3

PLATE 8.

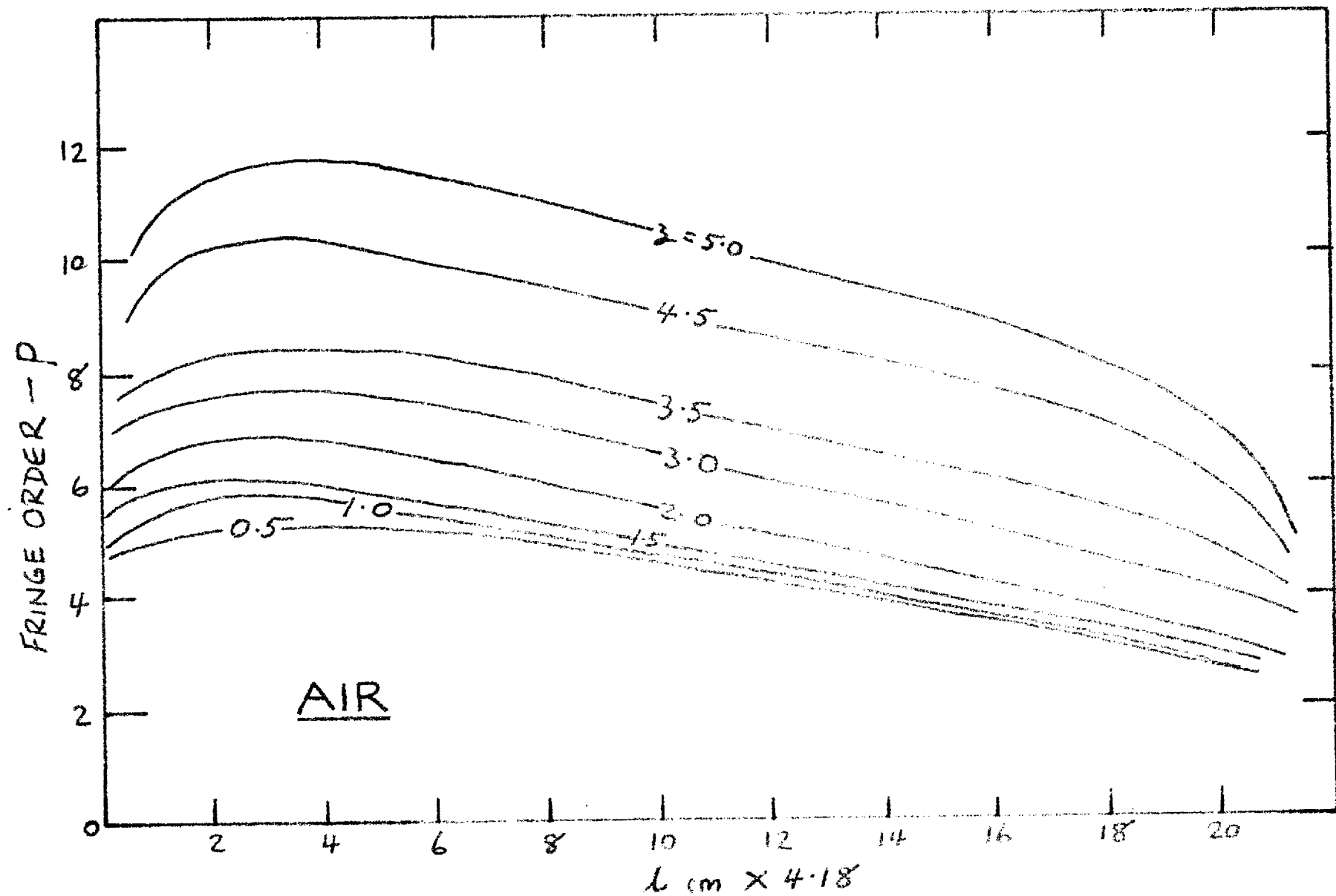


FIG.64

$$\int_{\text{Test}} \delta \, dx - \int_{\text{Ref}} \delta \, dx = p \lambda$$

where  $\delta = (n-1)$ ,  $n$  is the refractive index of the gas,  $p$  is the fringe order and  $\lambda$  the wavelength of the light.

Referring again to figure 6.3

$$\text{for ray A,} \quad \int_{\text{Test}} \delta \, dx = x_A \delta_T + 2 \int \delta \, dx$$

$$\text{so,} \quad x_A \delta_T + 2 \int \delta \, dx - x_A \delta_{T_0} = p \lambda \quad (6.4)$$

where  $x_A$  is the geometric path length of ray A.

Neither  $x_A$  or  $x$  are known with any certainty, so the equation (6.4) is not useful as it stands. But the difficulty, due to the presence of a boundary layer, can be resolved if the layer structure is constant within any chosen plane ( $z$ ).

Assuming this to be the case, the difference in optical path length between two parallel rays A and B, figure 6.3, is given

$$\begin{aligned} \Delta p &= [x_B \delta_T + 2 \int \delta \, dx] - [x_A \delta_T + 2 \int \delta \, dx] \\ &= (x_B - x_A) \delta_T \end{aligned} \quad (6.5)$$

where  $x_B$ ,  $x_A$  are accurately measurable distances across the cell, and  $\Delta p$  is the difference in fringe order number at A and B.

In terms of co-ordinate  $l$  and  $\theta$ , the base angle of the trapezium cross-section,

$$p \lambda = z \Delta \delta l \cot \theta \quad (6.6)$$

where  $\Delta \delta = (\delta_0 - \delta_T)$

Differentiating (6.6)

$$\lambda \frac{dp}{dl} = 2\Delta s \cot \theta$$

$$\Delta s = \frac{1}{2} \lambda \frac{dp}{dl} \tan \theta$$

Using  $s_o T_o = s T$

$$\frac{\Delta s}{s_o} = 1 - \frac{T_o}{T}$$

gives

$$T_z = \frac{T_o s_o}{s_o - \frac{1}{2} \frac{dp}{dl} \tan \theta} = \frac{T_o s_o}{s_o - K \frac{dp}{dl}} \quad (6.7)$$

where  $K = \frac{\lambda \tan \theta}{2}$  is constant for the system. (6.7)

gives a general expression for the constant temperature at any horizontal section (z).

The procedure for analysis adopted was therefore as follows. Residual fringes in the 'cold' interferogram were subtracted from or added to those in the 'hot' interferogram. The correct sign for the residual fringes was deduced by observing in which direction these fringes moved when a gas of very low density, such as hydrogen, was passed through the cold system.

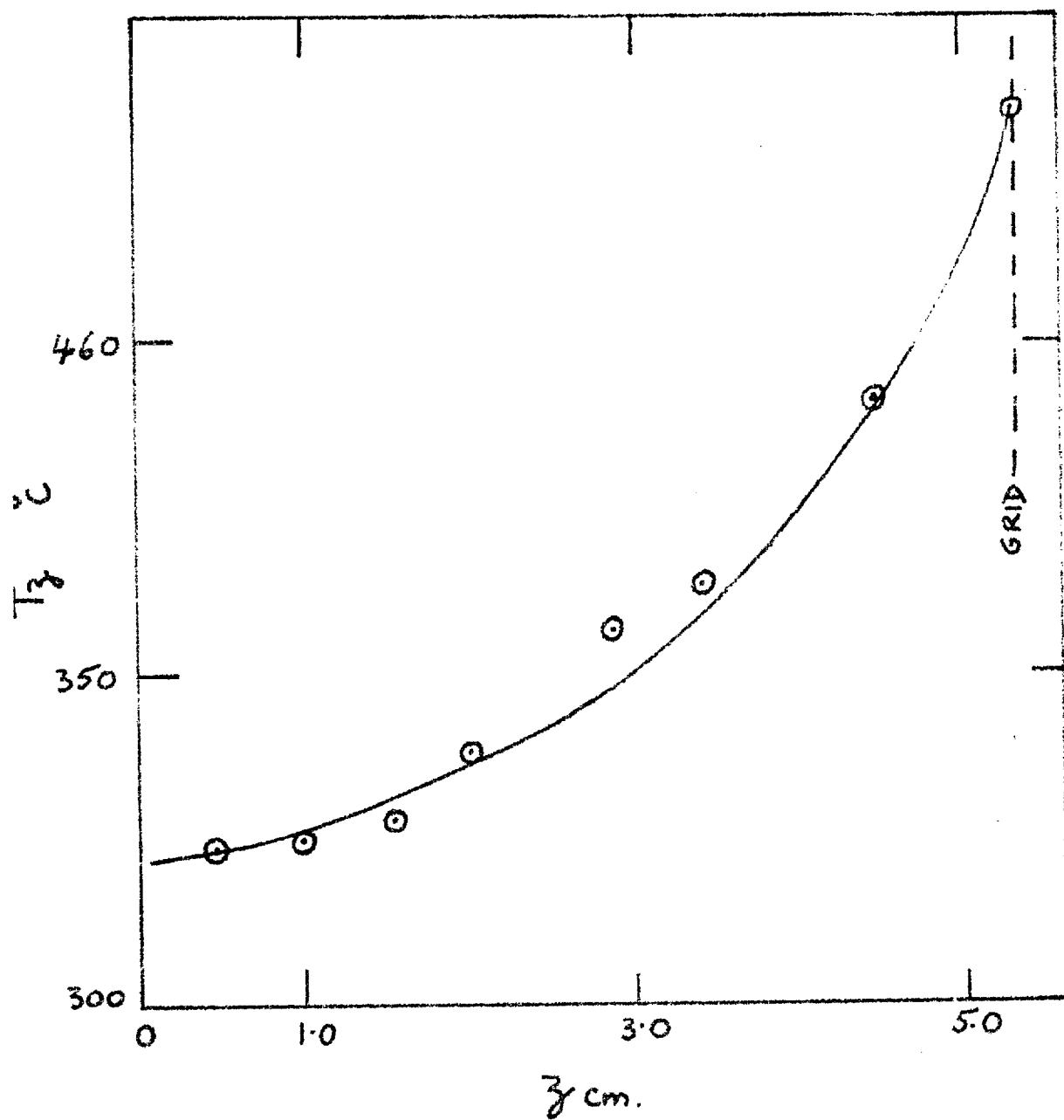
In actual experiments the residual fringes were observed to crowd closer together, moving towards the narrow end of the cell. It was concluded, therefore, that the optical path through the cell was less than that through the

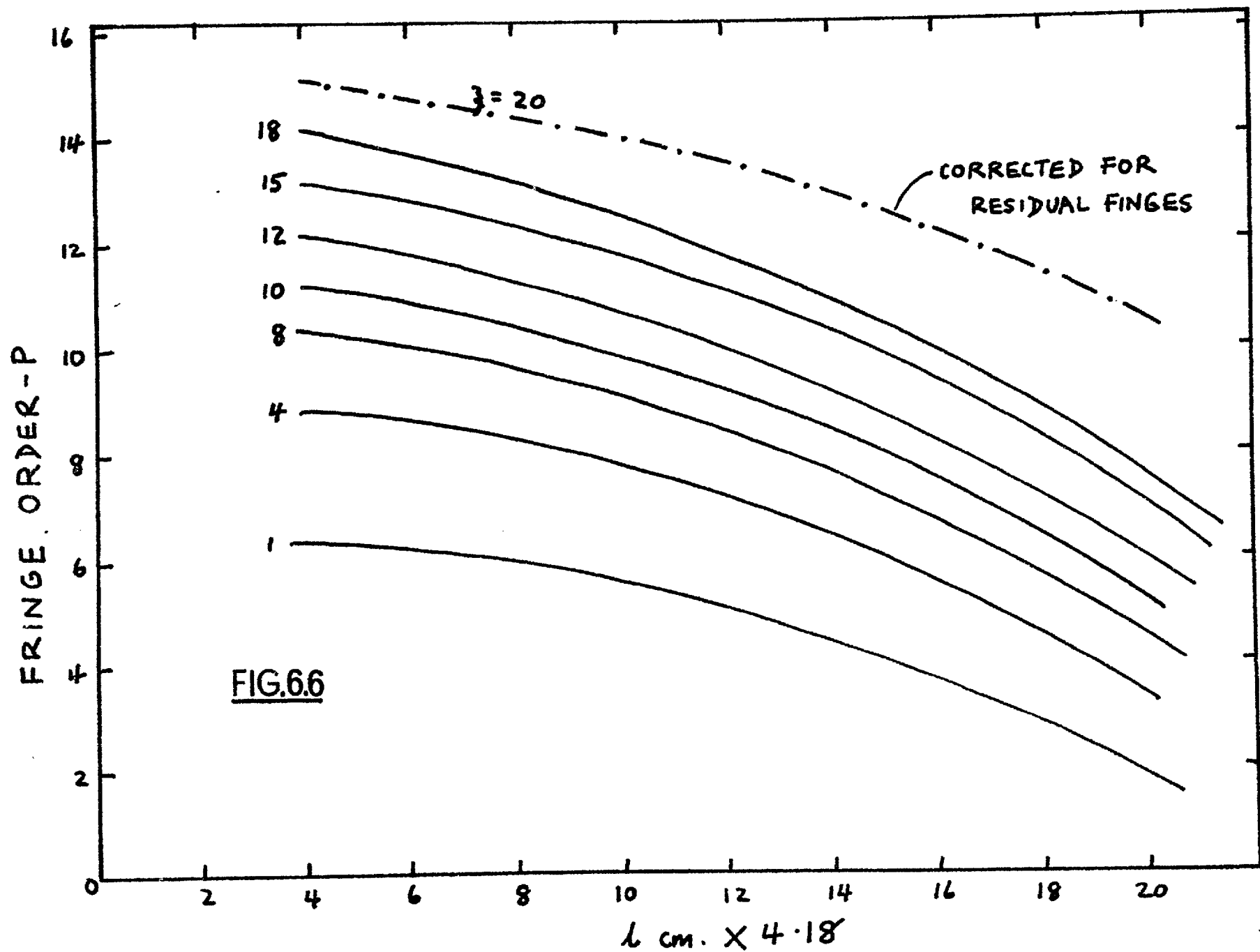
reference section. Since the "hot test fringes" arose due to decrease in the cell air density, as with hydrogen, the residual fringes were positive and should therefore be subtracted from the test interferograms. Once corrected, each interferogram was divided into a series of horizontal (z) sections. After numbering the fringes according to their orders, see figure 6.3a, graphs were drawn of fringe order (p) against co-ordinate (l) for each  $z = \text{constant}$ . Fig. 6.4 shows a typical result for one such analysis. From these plots, values of  $(\frac{dp}{dl})_z$  were obtained graphically and substituted into equation (6.7) for  $T_z$ . A temperature profile so derived is shown in figure 6.5.

Analyses of this sort were only found to be possible for interferograms taken under almost natural convection conditions, because when the flow rate was increased the fringe pattern became distorted, and appeared to follow the curved temperature field close to the surface of the grid wires. It was concluded that, in the case of the prototype wire grid systems tried, the temperature boundary layer round each wire changed rapidly with increased flow velocity. This difficulty led to modifications of the grid design referred to in Chapter 4.

## 6. Results for the modified unidimensional system.

The power output per  $\text{cm}^2$  from the second type of

FIG. 6.5



grid (fig.4.4(b)) was approximately 20 watts - almost twice that for the grid wire in the work described above. With the grid spacing reduced to 0.5 mm. and increased heat transfer across the grid, no appreciable change in fringe pattern occurred under forced convection conditions, other than a general compression of the fringes towards the heater surface. As with the first grid, the modified system was placed on top of the observation cell. Particle-track visualization of the flow under forced convection indicated that the velocity profiles were similar to those found previously. Several interferograms were recorded with the grid hot and gas flowing under forced convection. Plate 9 shows the form of closely spaced fringe pattern which was observed upstream of the grid surface. Analysis of interferograms, similar to that shown in Plate 9, however, yielded unreasonable order distribution profiles with no clear physical significance, figure 6.6. Non uniform flow in the vicinity of the heater surface was suspected, which led to non-unidimensional temperature distributions.

Closer inspection of the flow regime in this region using the particle-track method revealed that, as the temperature of the grid was increased, the particles no longer passed through the grid, but were deviated sharply through small gaps between the top of the cell and the heater surface. A probable explanation for this behaviour



PLATE 9.

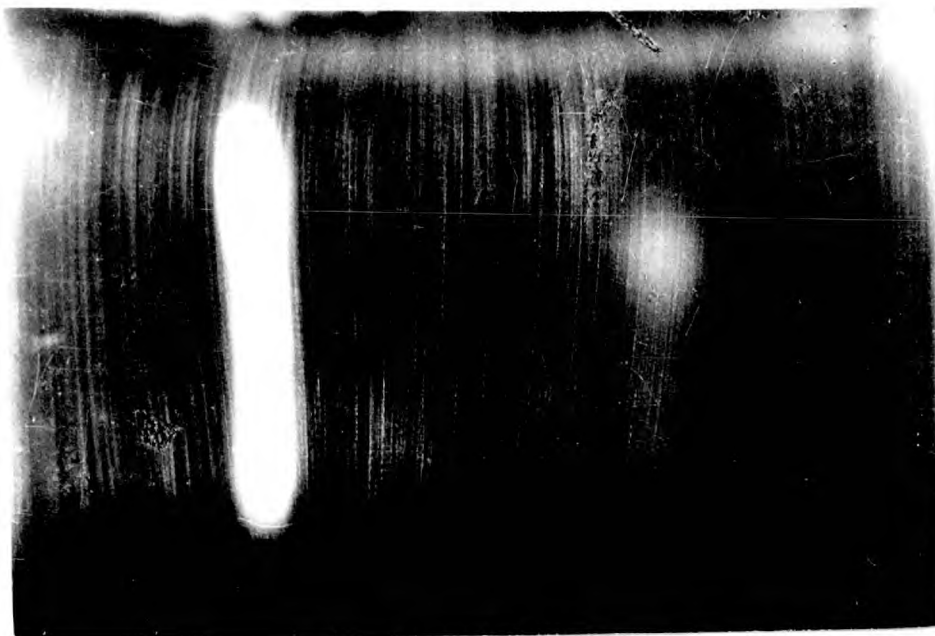
is that the pressure drop across the grid increases as the grid heats up, because the viscosity of a gas is approximately proportional to  $T^{\frac{1}{2}}$ .

Two methods were therefore tried to induce the gas through the pores in the grid. A suction tube system was attached to the grid and the downstream line pressure adjusted until the particles taken up by the flowing gas were observed to enter the grid surface. However, this idea had to be rejected because of severe aerodynamic disturbances which resulted due to air outside the observation cell being inducted and causing some eddy shedding. The presence of this phenomenon was reflected in rapid fluctuation of the fringe pattern. A second approach was to seal the grid permanently onto the mica cell, so that the whole system became completely air-tight. Gas now passed through uniformly, but only under conditions of high excess pressure upstream of the grid.

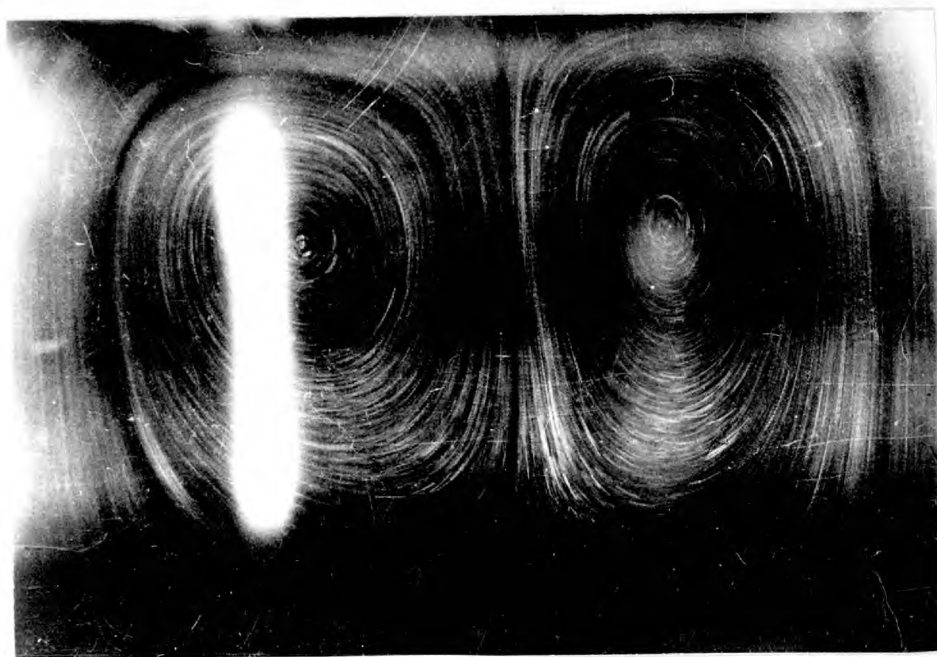
Interpretation of the interferograms taken with the modified system was complicated on two counts. The essential subtraction of residual fringes became uncertain, because their number and position changed with temperature. This was due to small stresses and distortions which arose in the mica windows as the now rigid system heated up. There was, in addition, suspected bulging upward of the grid

at appreciable flow rates which resulted in fringes being partly obscured. Restriction to working with very nearly natural convective flow could have removed the latter complication, but as Plate 10 shows, under these slow flow conditions, two large circulation systems in the form of vortices appeared, due to natural convection effects, superimposed on the supplied flow.

In view of the difficulties encountered, due to the failure of the 'unidimensional' model to behave in fact as a truly one-dimensional system, all subsequent work was based on a two-dimensional system which has already been described in Chapter 4.



COLD



HOT

PLATE 10.

## CHAPTER 7.

FURTHER STUDIES WITH THE 2-DIMENSIONAL MODEL.1. Theory.

Since the 2-dimensional system is axially symmetrical, the heat conservation equation (1.2) can be conveniently expressed in cylindrical polar co-ordinates.

$$\frac{1}{r} \frac{\partial}{\partial r} (r F_r) + \frac{\partial F_z}{\partial z} - q = 0 \quad (7.1)$$

where the heat flux  $F$  along any co-ordinate  $p$  is given by

$$F_p = -K \frac{\partial T}{\partial p} + \rho v_p H \quad (7.2)$$

and  $K$ ,  $\rho$ ,  $H$  and  $v$  are the thermal conductivity, density, enthalpy and velocity of the gas respectively.

Combining (7.1) and (7.2)

$$\frac{1}{r} \frac{\partial}{\partial r} \left( -rK \frac{\partial T}{\partial r} + r\rho v_r H \right) + \frac{\partial}{\partial z} \left( -K \frac{\partial T}{\partial z} + \rho v_z H \right) - q = 0$$

gives an expression for  $q$  in terms of the measurable variables,  $v$  and  $T$ .

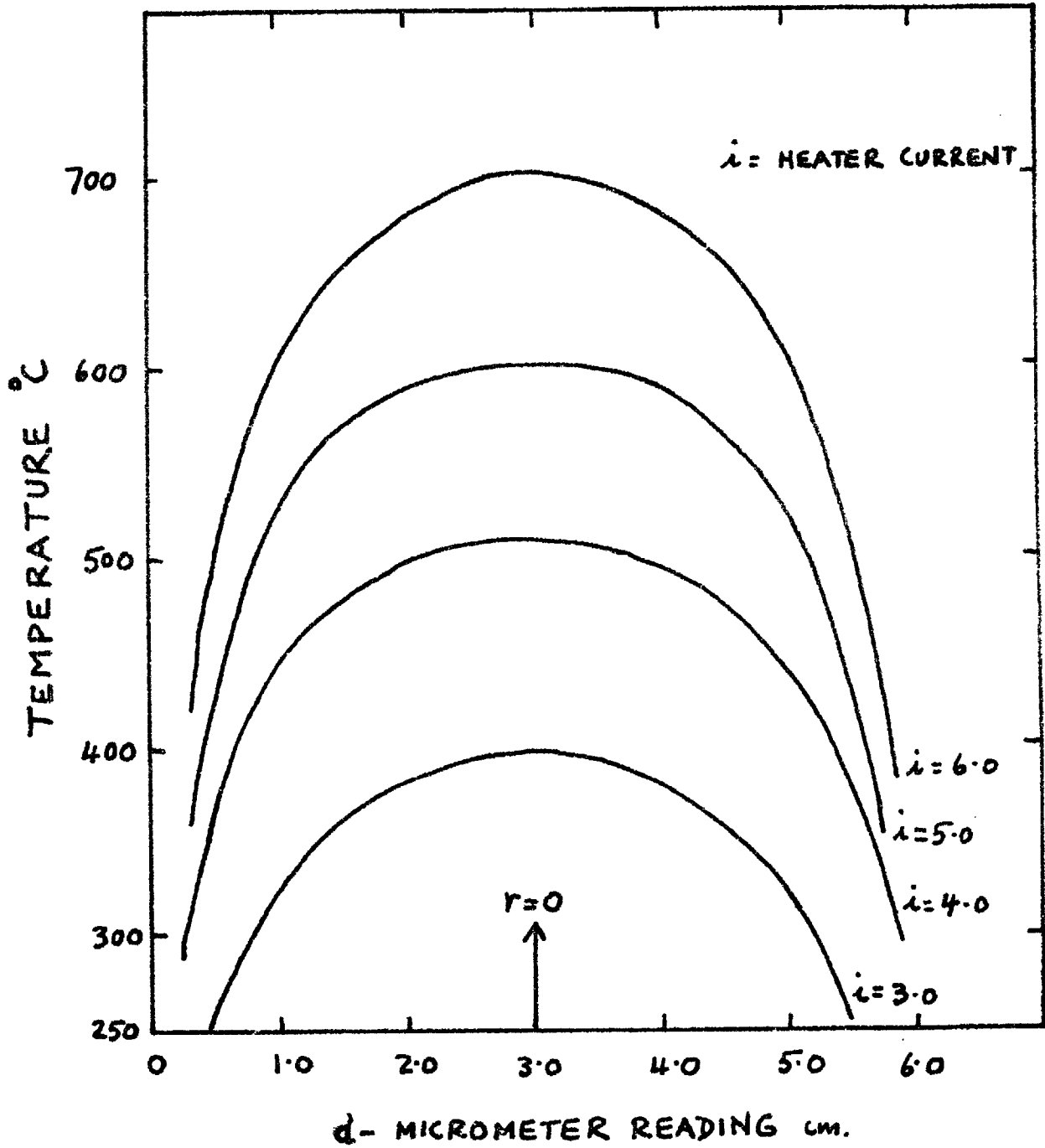
The other data required is the variation of local  $K$ ,  $\rho$ ,  $H$  values with temperature, which were readily available, (73).

The experimental model has been described in Chapter 4. Since it is based on the assumption of axial symmetry it was important to confirm this experimentally before proceeding further.

## 2. Surface temperature distribution.

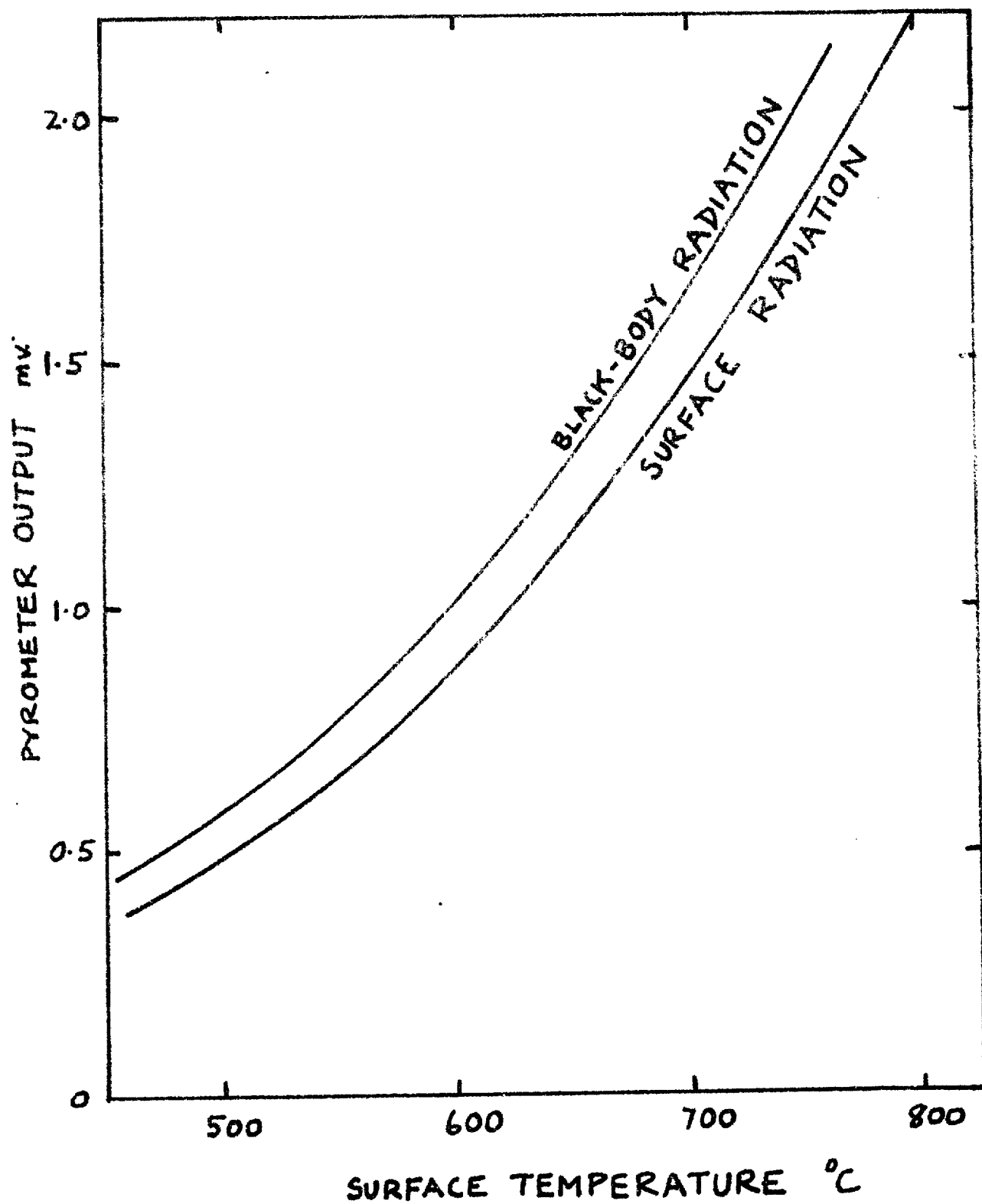
To establish the form of the temperature distribution across the lower surface of the quartz disc, traversals of the surface were made with a fine Pt/Rh thermocouple for a number of horizontal planes. No corrections were made for heat losses by the thermocouple due to radiation and conduction, but the results for a number of z-sections, shown in figure 7.1, indicated a radially symmetric temperature distribution. Because no attempt was made to eliminate heat losses, the radial temperature gradients in the profiles appear rather severe.

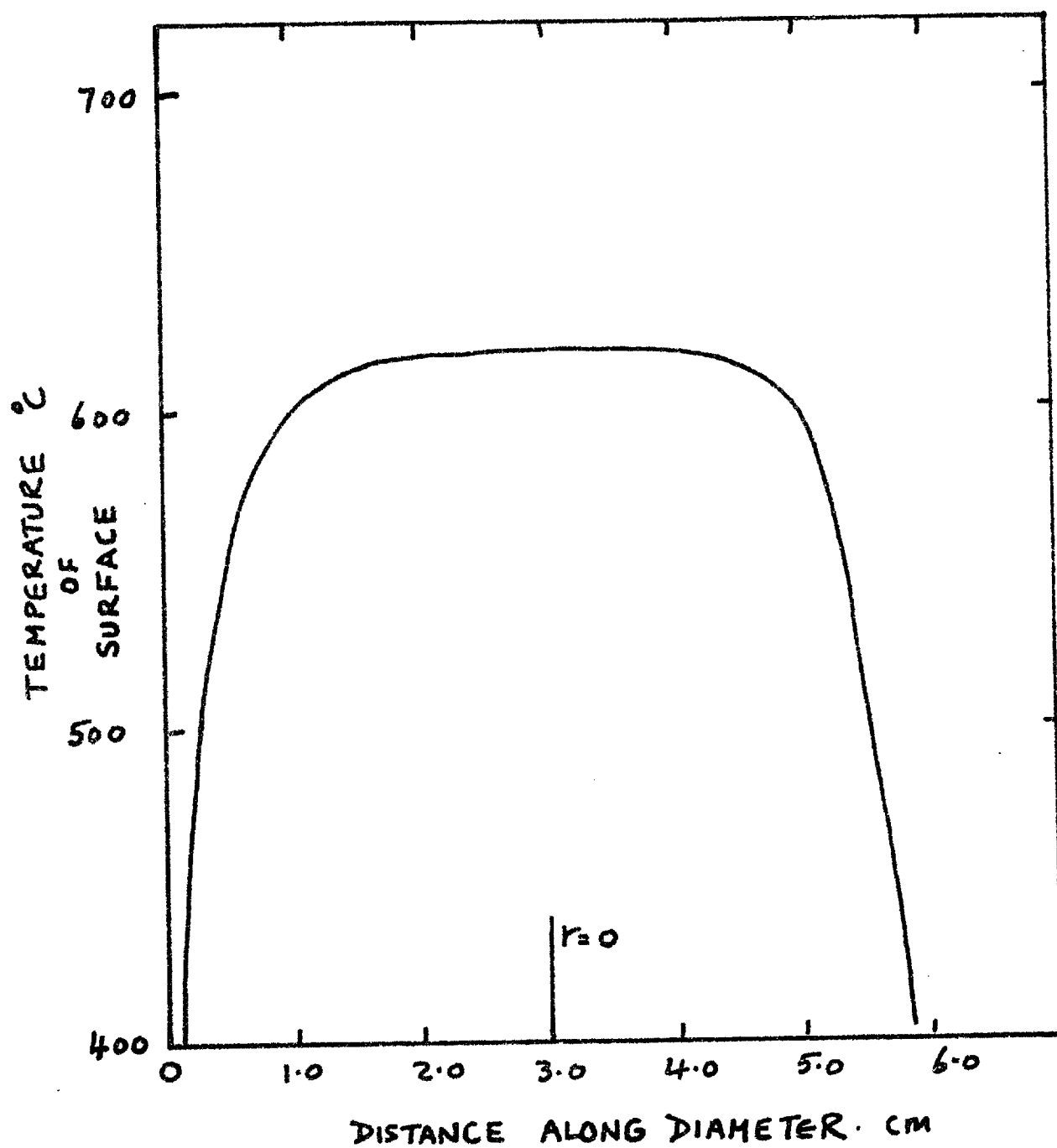
Preliminary experiments on the surface temperature distribution were continued on a quantitative basis using a total radiation pyrometer. The instrument employed was able to resolve 0.5 cm when focused on a hot surface at a distance of 15 cm, and had been calibrated for black-body radiation. It would therefore normally be essential to know the emittance of the disc surface used in the experiments before meaningful temperature measurements could be made with the pyrometer. A suitable method for obtaining emissivity values <sup>74</sup> was considered, which involves surrounding the sample by a black-body enclosure. The radiation received by the pyrometer is then recorded after the sample has been 'soaked' in the black body radiation.

FIG.7.1

This is the sum of the radiation emitted by the sample and the radiation reflected at the surface, and is equal to the black body radiation at the temperature of the enclosure. The sample is then placed in cool surroundings and the radiation received from it measured at frequent intervals. A plot of the cooling curve of radiation against time enables the radiation at zero time to be extrapolated. This is the radiation emitted by the sample when at the temperature of the black-body enclosure. The ratio of this to the original reading gives the emittance of the sample.

Fortunately, such auxiliary experiments became unnecessary because the manufacturers of the pyrometer agreed to recalibrate the pyrometer output directly for the disc surface used in the present experiments. The black-body and corrected calibration curves are shown in figure 7.2. Measurements of the radial temperature distribution over the disc surface were made by scanning the pyrometer across the disc. The temperature profiles indicated uniform heating over an extensive central region and confirmed the radial symmetry found using a thermocouple. Figure 7.3 gives a typical distribution of surface temperature against distance along a diameter of the disc. These measurements were made under forced convection conditions and are smoothed values, because of the finite target area of 0.5 cm over which the pyrometer would average.

FIG.7.2

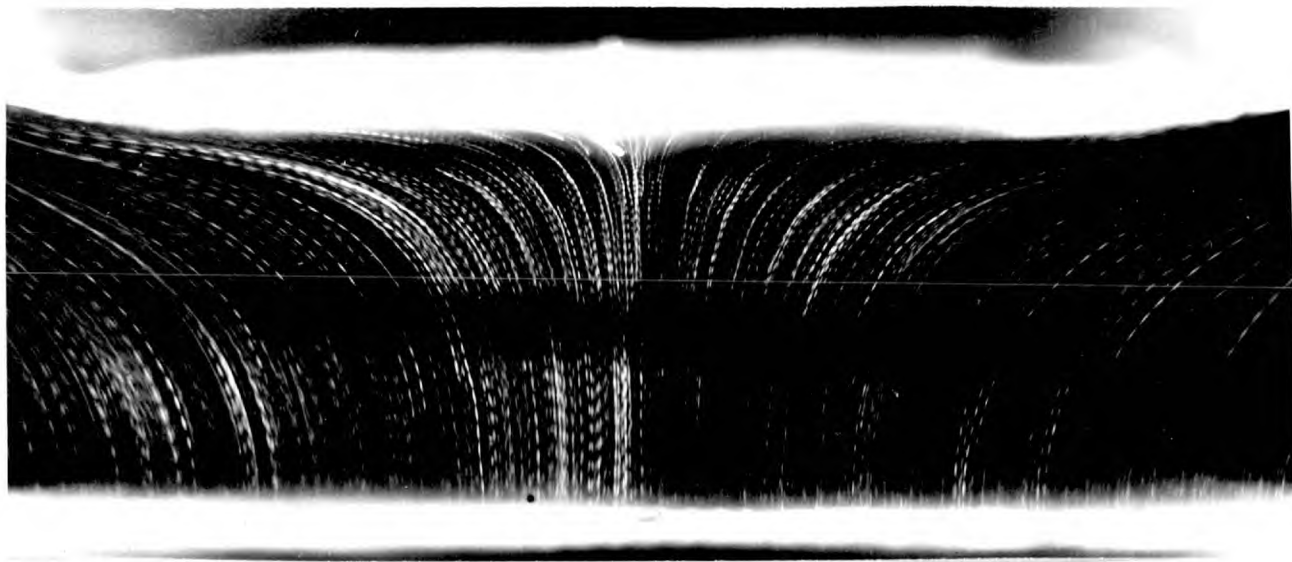
FIG.7.3

### 3. The flow pattern.

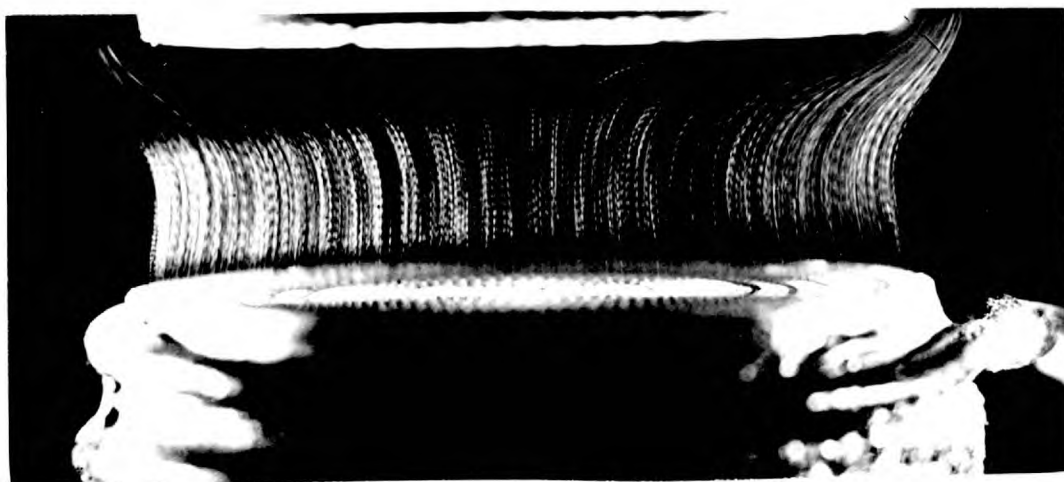
Several particle-track photographs were taken in order to study the symmetry of the flow pattern in the 2-dimensional system. It was very important that both sections of the model - i.e. the tube and matrix as well as the upper heated cylinder - were accurately aligned along a common, vertical axis (see figure 4.5). Only when this was so were symmetrical flow patterns observed. Plate 11 shows particle-track photographs, typical of the many taken. Judging by the photographs, the boundary layer and upstream stagnation zone do not appear to extend far from the disc surface.

### 4. Measurement of gas velocities.

To determine local gas velocities from the particle-track photographs, enlarged prints were made (approximately 10 x) of the region in which the gas velocities were to be determined. The exact magnification involved was found subsequently by reference to an identical enlargement of a scale photographed at the same time as the original particle tracks. Preliminary analysis of the velocity distribution was confined to a small central region along the axis. Rectangular axes,  $r$  and  $z$  were taken with the matrix centre as origin, figure 7.4. In determining the velocity components along co-ordinates ( $z$ ) and ( $r$ ) at every point,



(a)



(b)

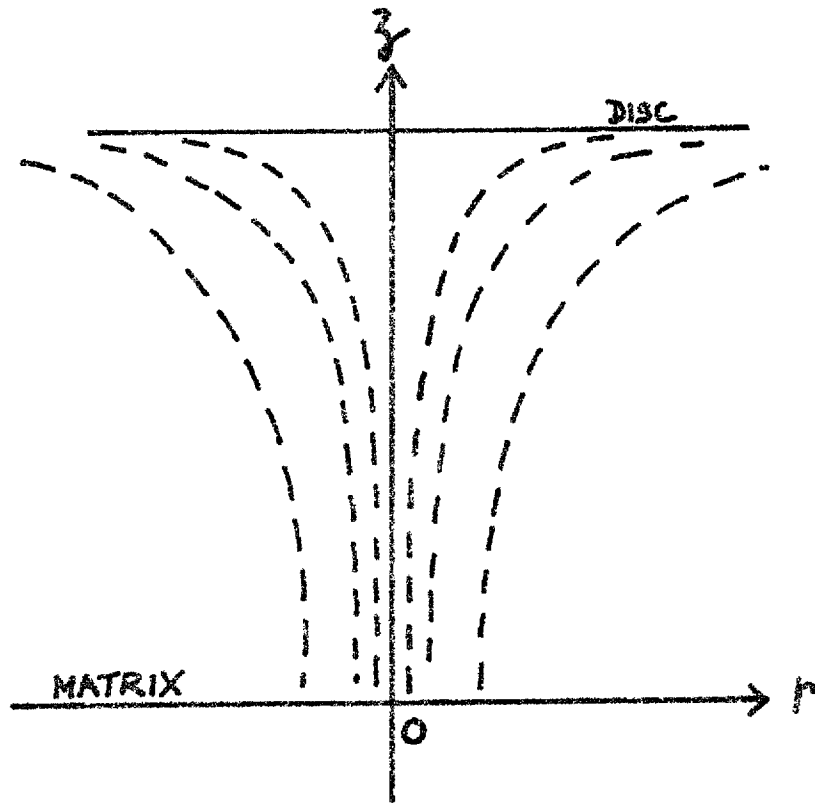


FIG.7.4

local curvature of the flow lines, over the finite distance for which the measurements were made, was neglected. In the fast flow regions, well upstream of the heated surface, the flow lines were almost linear and so the effective averaging of the direction in obtaining the velocity measurements was a negligible source of error. The approximation was also found to be reasonable in regions, just upstream of the heated disc, where flow stream curvature becomes appreciable. Here the maximum particle velocities were less than 3 cm/sec., and this corresponded to an interruption between adjacent tracks along the flow line of less than  $3 \times 10^{-2}$  cm. The accuracy to which the component velocities could be measured depended on the error involved in locating the centre of each track, and on errors in the magnification of the tracks themselves. The estimated error due to these factors ranged from about 9% in the low velocity region near the forward stagnation point, to under 3% in the relatively high velocity region near the matrix. Data from several particle-track photographs were required to determine a complete velocity distribution. The experimental scatter was appreciable, due in part to a wide range of particle size. Available data on the sample of bentonite particles used indicated a mean particle size of  $5 \mu$ . Agglomeration of the particles occurred, however, so that the resultant range of particle size included satellites with considerably bigger

diameters than  $5 \mu$ . Large particles well outside the  $5 \mu$  category showed a loss of inertia, appearing sluggish in regions of acceleration and showed a tendency to overshoot in regions of deceleration, especially near to the disc surface. In determining the velocity distribution from the particle -track photographs, those of conspicuously heavy particles were rejected.

#### 5. Sources of error.

The thermo-mechanical and velocity lag effects<sup>61,69</sup> are important sources of error in some flame studies where both the temperature and velocity gradients are very steep. These phenomena were considered in detail and shown to be unimportant in the present work where the temperature and velocity gradients were relatively mild, and the particle size much greater than the mean free path in the gas (see Appendix 2). Another possible source of error arises due to free fall experienced by the particles under gravity. An order of magnitude correction for this effect was estimated by equating the gravitational force on the particle to the Stokes-law viscous drag

$$\text{i.e.} \quad \frac{4}{3} \pi a^3 \rho (\sigma - \rho) g = 6 \pi \eta a \Delta v$$

where  $a$ ,  $\sigma$ , are the radius and density of the particle,  $\rho$  and  $\eta$  are the density and viscosity of the gas and

$\Delta v$  is the free-fall velocity of the particle

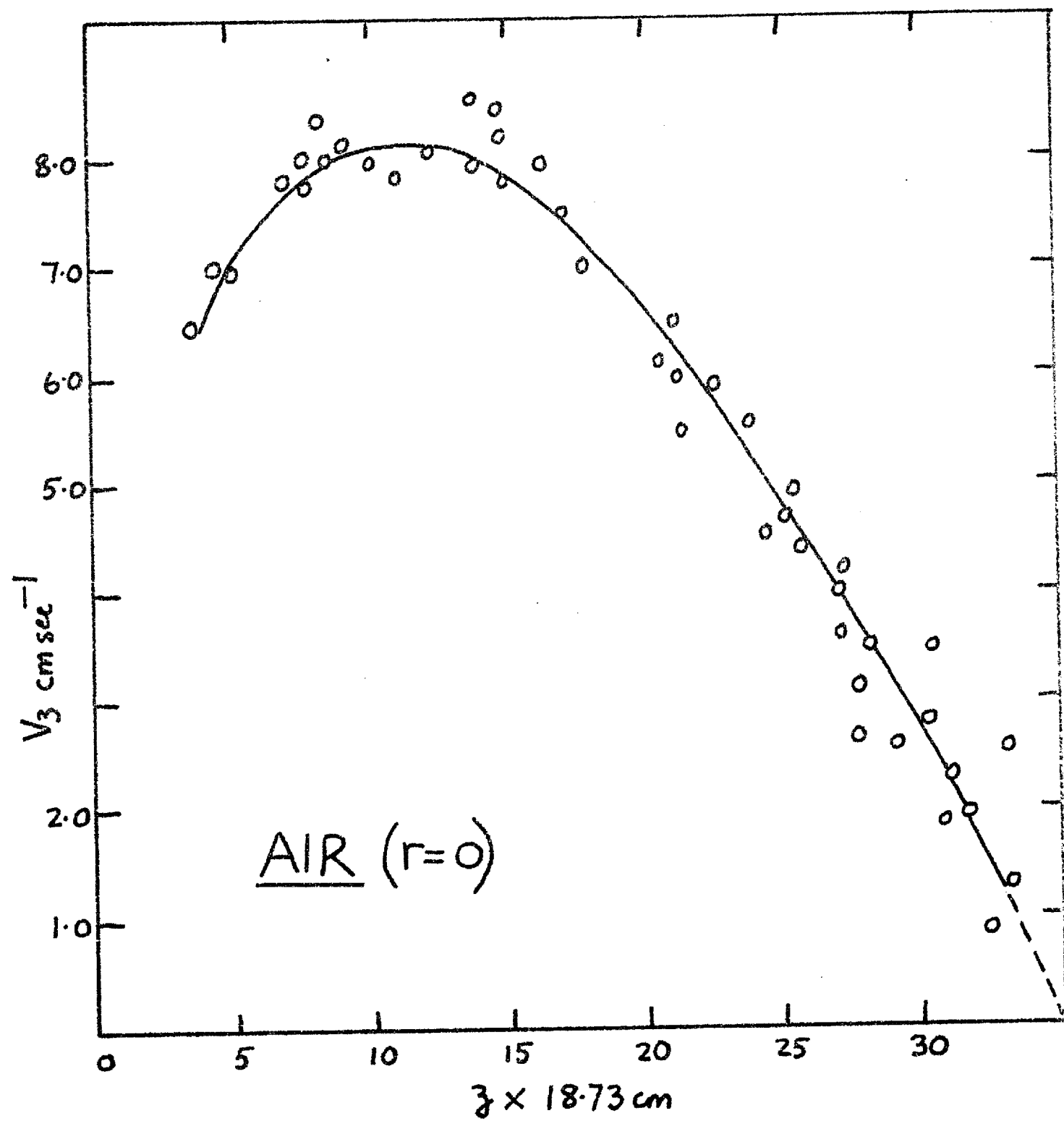
$$\Delta v = \frac{2a^2 (\sigma - \rho) g}{\dots}$$

For the present work  $\Delta v \approx 0.15 \text{ cm/sec.}$ , taking the density of the bentonite particles to be  $3.5 \text{ gm/cc.}$  This error was shown to be much smaller than the experimental scatter found in the particle-track velocity measurements.

## 6. Velocity distributions.

Figure 7.5 shows the velocity distribution found for air only at a cold flow rate of 12.5 litres/minute. The gas appears to accelerate initially as it leaves the matrix and then, after reaching a maximum velocity, is retarded strongly on approaching the disc surface. Initial acceleration is largely due to the gas expanding after being heated as it passes through the surface of the matrix, which strongly absorbs the radiation from the hot surface above it. Figure 7.6 shows the temperature distribution across the matrix measured by six thermocouples, which were constructed by welding nichrome wires to the matrix; the matrix acting as the second metal. (see figure 4.5).

Radial velocity distributions were also investigated as a function of the z-co-ordinate. Those in fig.7.7 are for air at the same flow rate as in the axial velocity distribution above. Both sets of velocity

FIG. 7.5

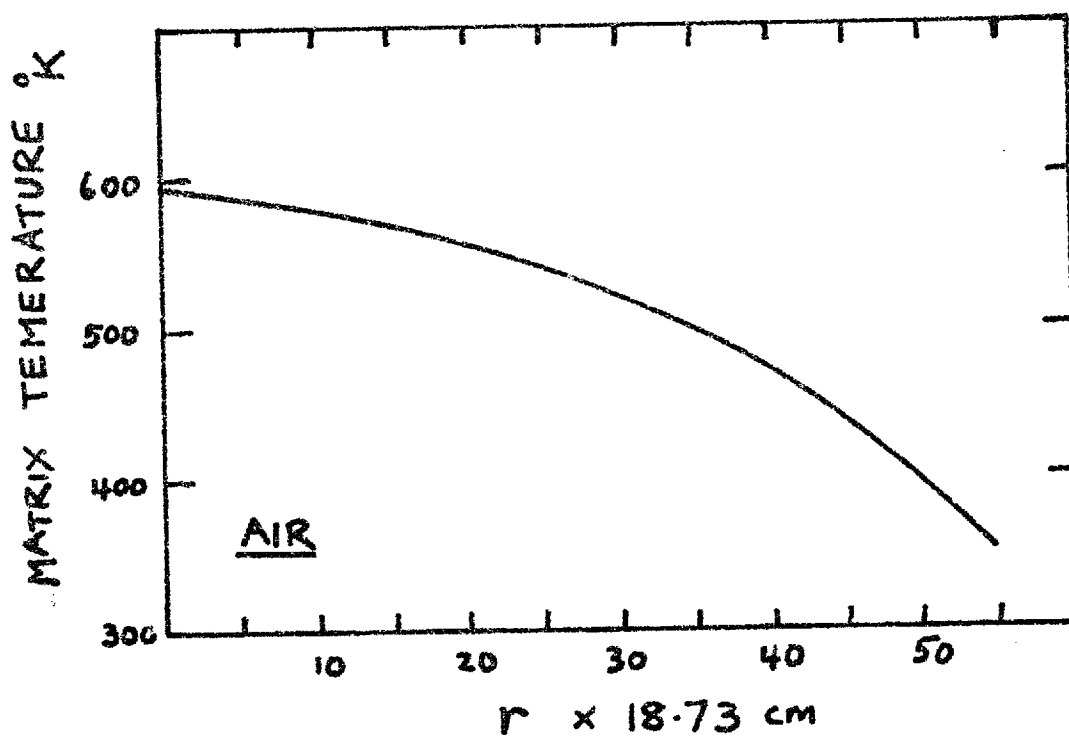


FIG. 7.6

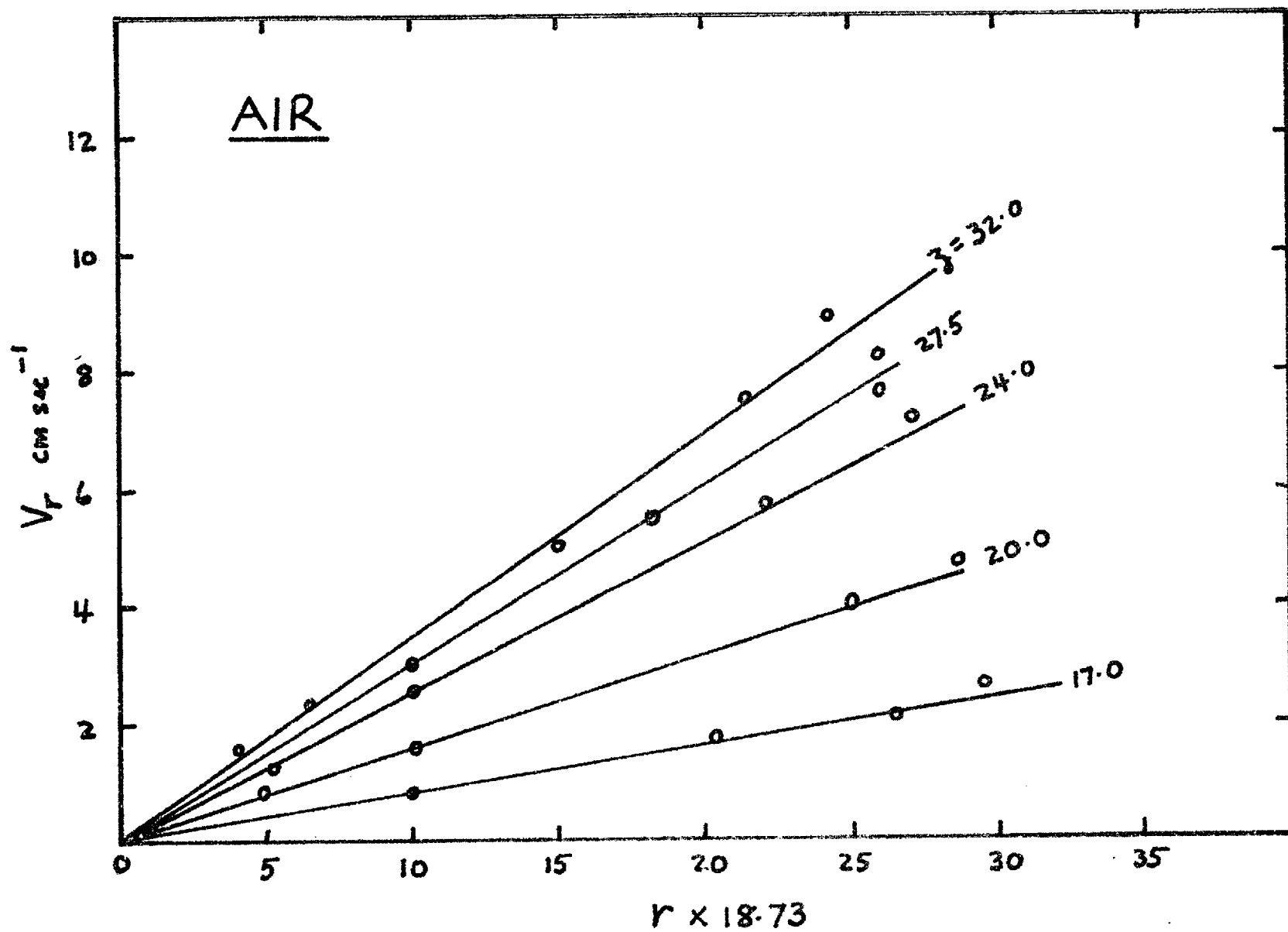


FIG.7.7

distributions were later used in a more detailed analysis of the system.

7. Sequence of operations for experimental runs.

A complete experimental run for both reactive and non-reactive gas mixtures entailed the following procedure :

- (1) The gas flow rate was adjusted to some specified value.
- (2) The heater current was switched on and slowly increased until the temperature of the disc reached a constant, pre-determined value. For ignition sequences with reactive ethylene/air mixtures, preliminary trial experiments were conducted to find the minimum surface temperature at which the mixture ignited, under known conditions of flow rate and mixture strength. The heating current and surface temperature were noted for subsequent experiments.
- (3) The matrix thermocouple readings were observed at short intervals until no change in their output was noted.
- (4) The reference space air temperature was recorded.
- (5) The pyrometer output was recorded.
- (6) When a steady state had been established, which took sometimes up to 2 hours, several particle track photographs were taken.
- (7) Without altering conditions of the experiments, the cine camera was started and the interferograms photographed.

For air only, the procedure followed exactly the steps described. In the case of an ethylene/air mixture, however, it was necessary to alter the air flow rate a little after the steady temperature conditions had been reached and quickly re-adjust to the initial overall flow rate by admitting the specified flow of ethylene. Immediately afterwards, the cine camera was switched on to record events which usually culminated in spontaneous ignition. While the interferograms were being photographed, the surface temperature of the disc was recorded automatically on the single pen recorder. The relevant particle-track photographs were also taken under these conditions.

## CHAPTER 8.

ANALYSIS OF INTERFEROGRAMS OBTAINED WITH THE  
2-DIMENSIONAL SYSTEM.

It may be noted here that even if the so-called 'unidimensional' system had worked experimentally, one would not really have assumed that it was unidimensional, anymore than flat flames are assumed to be one-dimensional. Thus a form of 2-dimensional analysis has always got to be carried out. The only difference is that if the system is approximately unidimensional, the errors produced at the higher temperature due to errors at the lower values are very much less.

1. Choice of method.      (a) Numerical integration.

Analysis of a 2-dimensional system is complicated by the need to integrate through changing refractive index fields. This offers prospects of a lengthy and tedious analysis. A numerical method for carrying out the analysis was therefore considered, with a view to using a computer for most of the work. The following discussion describes, very briefly, the initial formulation of the problem.

Considering figure 8.1, with the z-axis of the co-ordinate system chosen as the axis of cylindrical symmetry, the local value of refractive index ( $n$ ) in the

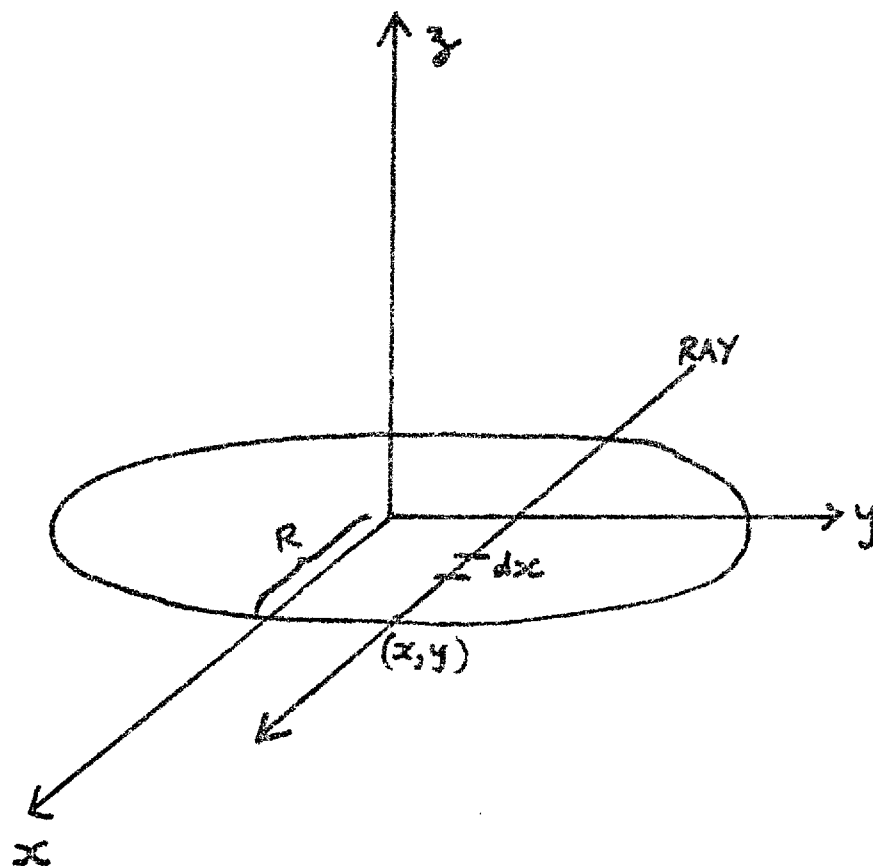


FIG.8.1

field is a function of  $(z)$  and  $(r)$  where  $r = \sqrt{y^2 + x^2}$ .  
 For a line element  $(dx)$  along the path of a ray parallel  
 to the  $(x)$  axis

$$dx = \frac{r dr}{\sqrt{r^2 - y^2}}$$

Neglecting deflection effects, the optical path difference  
 for a ray through the field is simply

$$2 \int_0^x n_r dx - 2 \int_0^x n_o dx = p(y) \lambda$$

where  $n_o$  refers to the reference state refractive index

$$\begin{aligned} \text{i.e. } p(y) \lambda &= 2 \int_y^{\sqrt{x^2+y^2}} (n_r - n_o) \cdot \frac{r dr}{r^2 y^2} \\ &= \int_y^R \frac{(n_r - n_o) \cdot d(r^2)}{\sqrt{r^2 - y^2}} \end{aligned} \quad (8.1)$$

(8.1) is an integral of the Abel type<sup>75</sup>. It can be shown<sup>76</sup>  
 that, by further manipulation, equation (8.1) may be expressed  
 in the form of a Stieltje's integral

$$n_r - n_o = - \frac{\lambda}{\pi} \int_r^R \frac{dP_y}{\sqrt{y^2 - r^2}} \quad (8.2)$$

Numerical determination of  $n(r)$  at specified radii  
 $r_i$  from equation (8.2), presents a sizeable computing task.  
 Details on how to treat equation (8.2) for such a numerical

analysis can be found in (77). The main problem is that the distribution function  $P_y$ , which is simply the order number, has to be programmed separately for each  $z = \text{const.}$  section of the test-space. In view of the size of the computing task involved, it was proposed to shelve setting up programmes for the 2-dimensional system, at least until it could be confirmed worthwhile.

(b) Graphical method.

The essential steps in the analysis discussed below apply to an interferogram which has been corrected for residual fringe errors. It is only necessary to consider one half of the test space, as shown in figure 8.2, since the system is axially symmetrical. Plane sections,  $z = \text{const.}$ , are drawn at right angles to the central axis. The next stage concerns each one of the  $z$ -sections as viewed in figure 8.2b. Concentric zones, 1,2,... n, are drawn as shown, with the outermost zone, 1, corresponding to the edge of the refractive index field. The latter is defined in terms of the first change in optical path detectable on the interferogram, which may be determined quite accurately from a plot of fringe order ( $p$ ) against co-ordinate ( $r$ ) as in figure 8.3. The first zone then corresponds to a chosen value of ( $r$ ), which is slightly less than  $r_{(p=0)}$ . A ray,

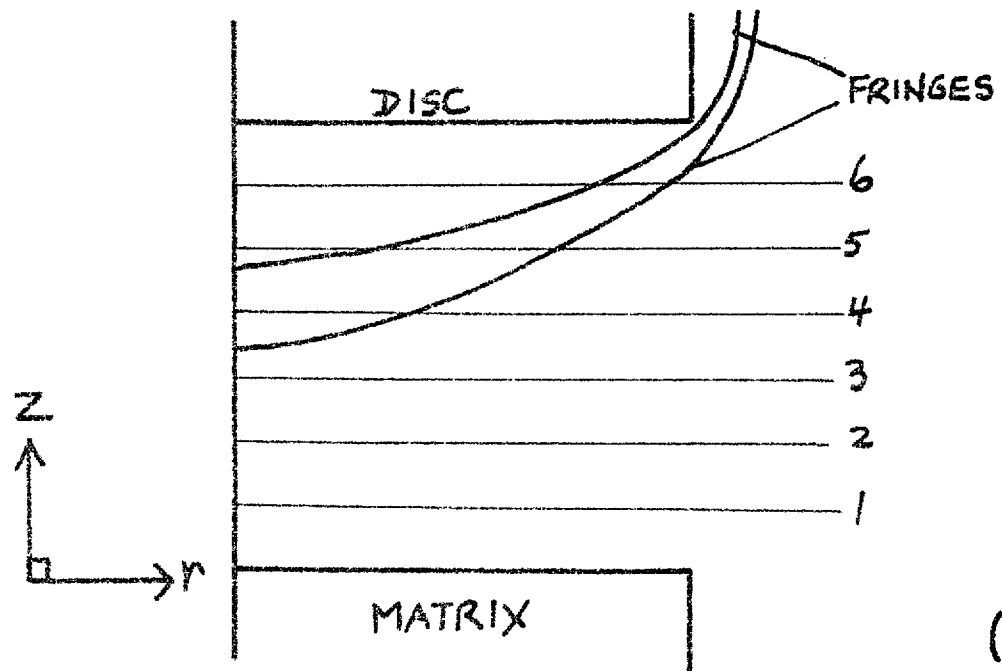
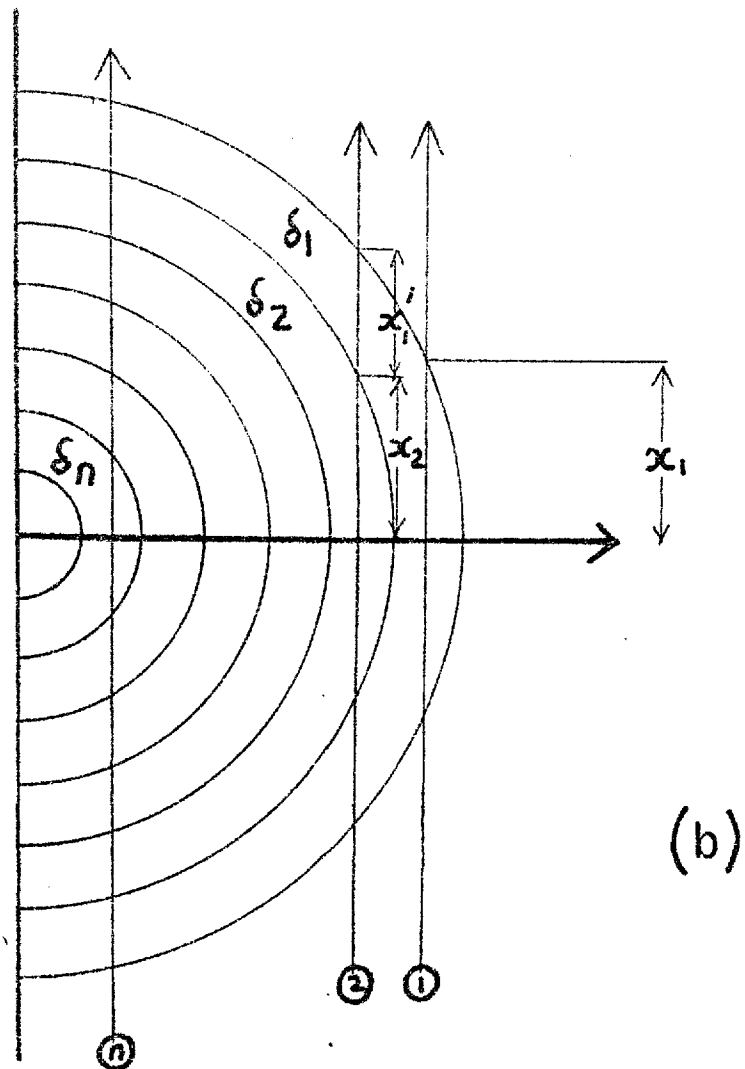
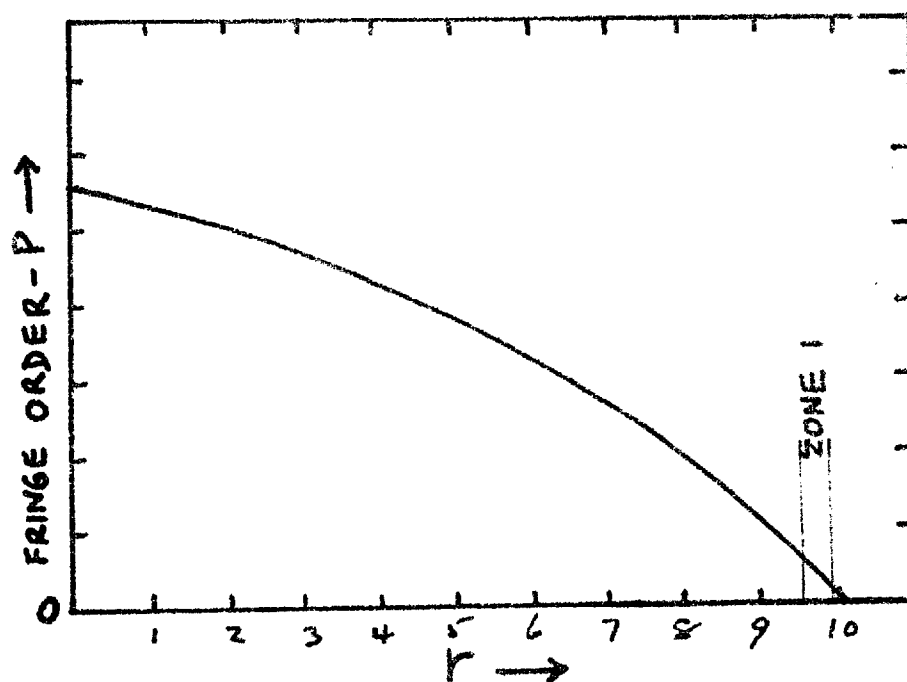


**FIG. 8.2**

A CROSS-SECTION  
(Z)



FIG. 8.3

(1) in figure 8.2b, is drawn perpendicular to the (r) axis and through the middle of zone (1). The refractive index  $\xi_1$  within the first zone is taken to be constant. Ray (1) corresponds to a known fringe order, obtained from the smoothed distribution curve in figure 8.3. The optical path difference between ray (1) and a counterpart reference ray, is given by

$$\begin{aligned} \frac{1}{2} P_{(1)} \lambda &= \int_0^{x_1} (\xi_0 - \xi_1) dx \\ &= (\xi_0 - \xi_1) x_1 \end{aligned}$$

Similarly for the second zone

$$\frac{1}{2} P_{(2)} \lambda = \xi_0 (x_1' + x_2) - (x_2 \xi_2 + x_1' \xi_1)$$

Hence knowledge of the geometric path lengths through each of the zones enables the same procedure to be adopted for the  $n^{\text{th}}$  zone until the axis is reached.

i.e. in the general case

$$\xi_n = \xi_0 \sum_{j=1}^n x_j - \sum_{j=1}^{n-1} x_j \xi_j - \frac{1}{2} P_{(n)} \lambda$$

$\xi_0$  must be accurately measured since a small error in this reference quantity will affect subsequent  $\xi$  values for each zone.

The number and width of the zones is arbitrary, depending on the fringe distribution concerned. In the

present work it was usually found necessary to construct 44 zones for each (z) section. Generally, the narrower the zones are the less severe is the approximation of constant refractive index over each. A limit on zone width is set however by the accuracy to which the fringe orders can be read; in the present study this was estimated to be about  $1/10$  of a fringe. On this basis, the zone widths were varied depending on the separation of the fringes. (i.e. in regions of crowded fringes, very narrow zones were employed, while wider zones applied in regions of more widely separated fringes). Another approximation was made by assuming the deviations of the light rays from a straight path to be infinitesimal. In reality, some refraction must occur at the zone boundaries and although this becomes a serious consideration near the edge of the disc where the refractive index gradients are steep, the relevant geometric path lengths involved here are extremely small, so the effect on the optical path lengths can be neglected.

In practice the analysis described was carried out on enlarged interferograms which were traced onto feint-lined graph paper. The magnification was determined from accurate measurement of the distance between the disc surface and matrix. In the present work a magnification factor of 18.73 was used.

## 2. Correction of interferograms.

An interferogram could hence be analysed to yield a temperature distribution in the way described, provided the reference space refractive index ( $\xi_0$ ) was known. Additional data on the temperature distribution across the disc surface and matrix was also available, since these distributions had been measured independently with the pyrometer and matrix thermocouples under experimental conditions relevant to the interferogram being analysed. This excess information was used to make the analysis of the interferograms more accurate, as discussed below.

Each interferogram was pegged at the disc surface and matrix planes. Pegging at the highest temperature zone was particularly desirable because, as has been discussed earlier, small errors in refractive index measurements result in large temperature discrepancies

$$\text{i.e.} \quad d\xi = \frac{-\xi_0 T_0}{T^2} dT$$

The general shape of a single, broad residual fringe, which occupied part of the test space when the system was cold, was also noted. The interferogram was then pegged at the matrix and disc surfaces by using the method of analysis described above in reverse. From a knowledge of the two temperature, and hence refractive index distributions, a

theoretical fringe order distribution was calculated in each case. The temperature distribution across the plane, but away from the circumference of the disc was deduced on the assumption that the fringe order distribution in this region was substantially correct. Since no part of the residual fringe was conserved here, this seems a reasonable assumption. Figure 8.4 shows the experimental and theoretical distributions across the disc and matrix planes. The residual fringe corresponded to the interpolated correction of about one fringe order at the disc surface. A similarly good fit was observed at the matrix. By following the general direction of the residual fringe it was now possible to estimate the error in fringe order for any chosen section ( $z$ ). For this the order distribution across the residual fringe was assumed to remain constant. Figure 8.5 shows the interpolated fringe order correction for a number of  $z$ -sections, which were subtracted from the experimental order distributions.

The importance of carrying out these corrections is reflected in the temperature distributions shown in figure 8.6 which were obtained when this important correction was not made. Not only is the temperature of the gas highest near the periphery of the disc, but it also exceeds the maximum temperature recorded at the time the interferogram was taken

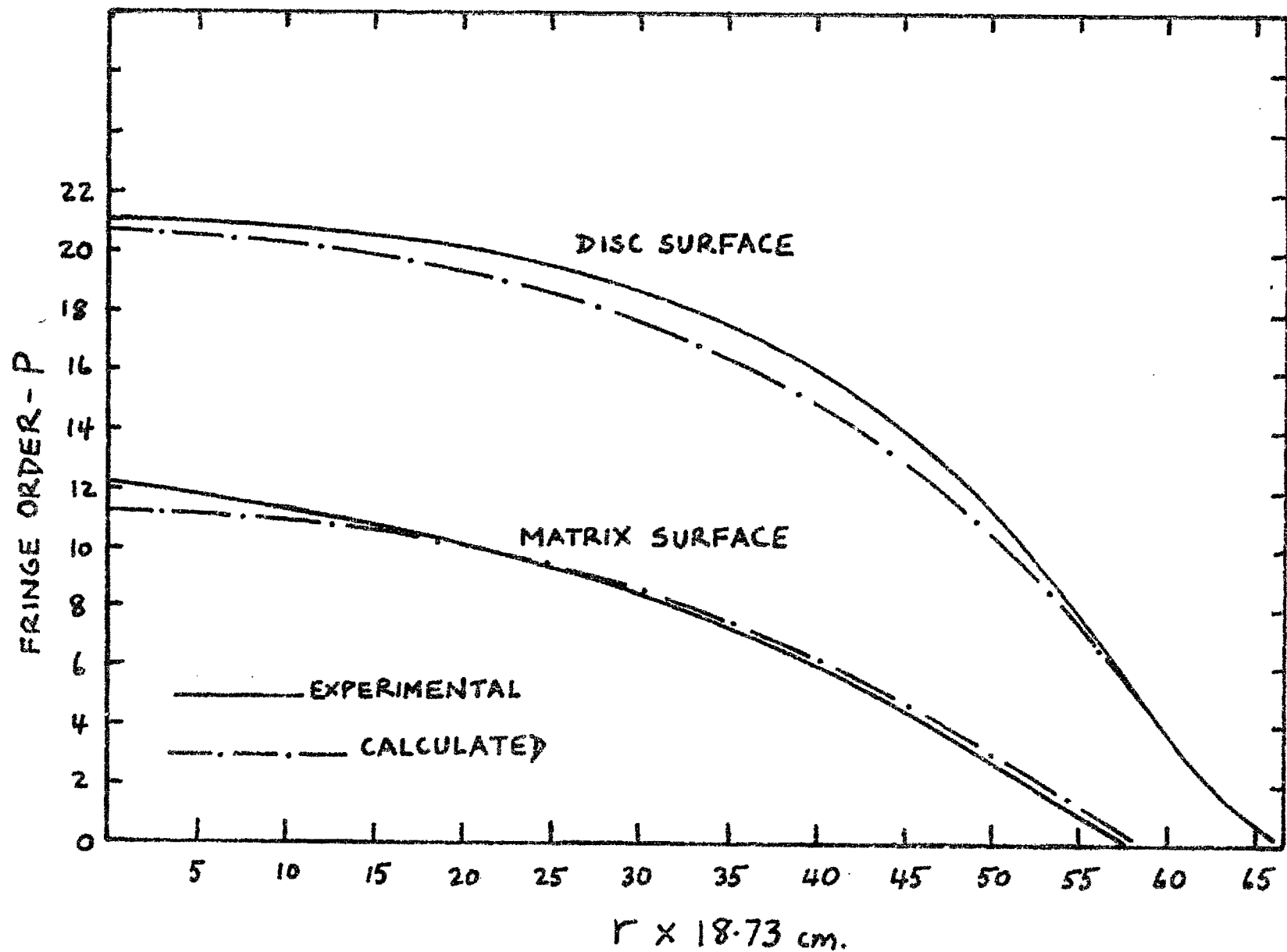


FIG.8.4

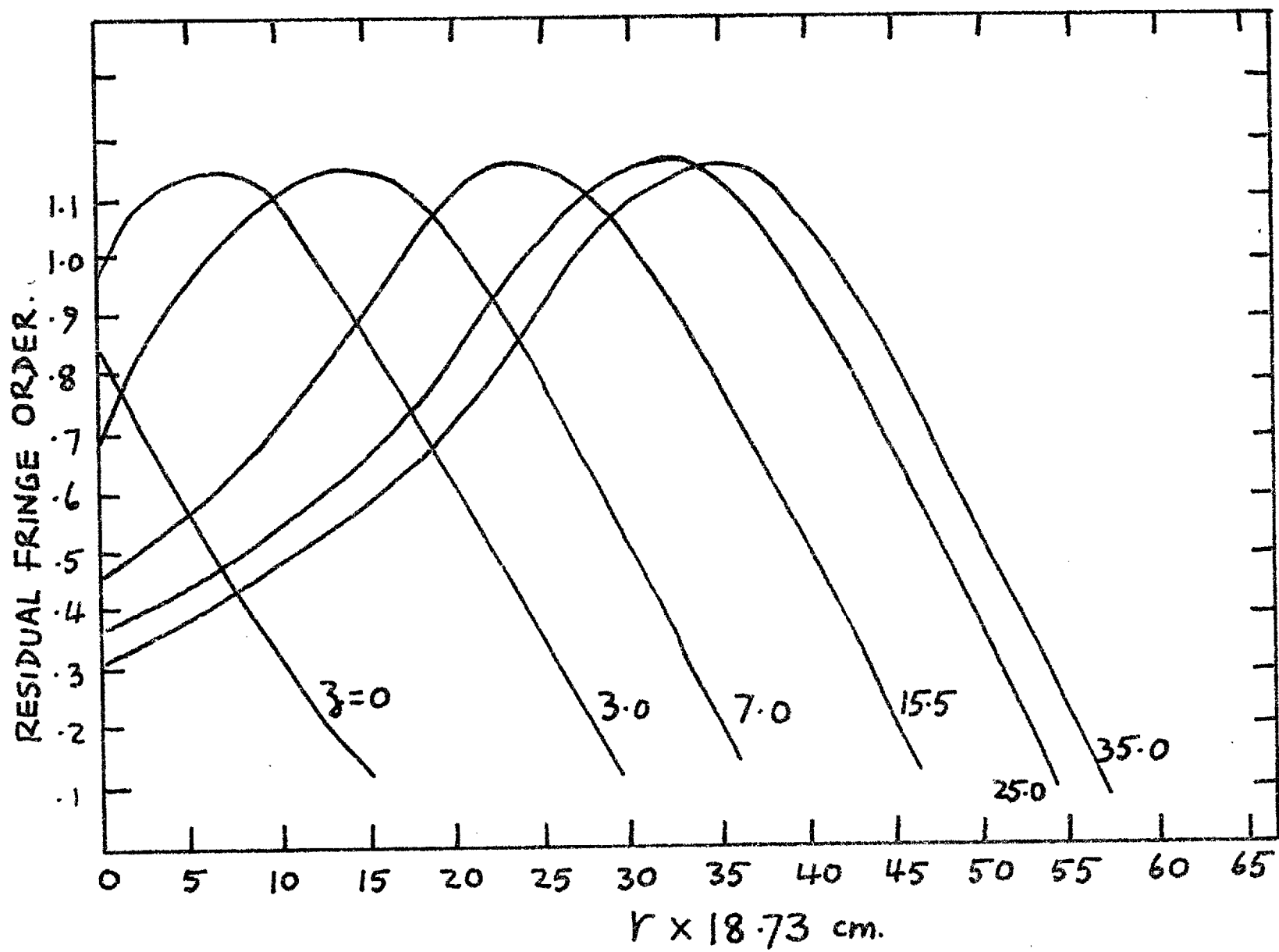


FIG.8.5

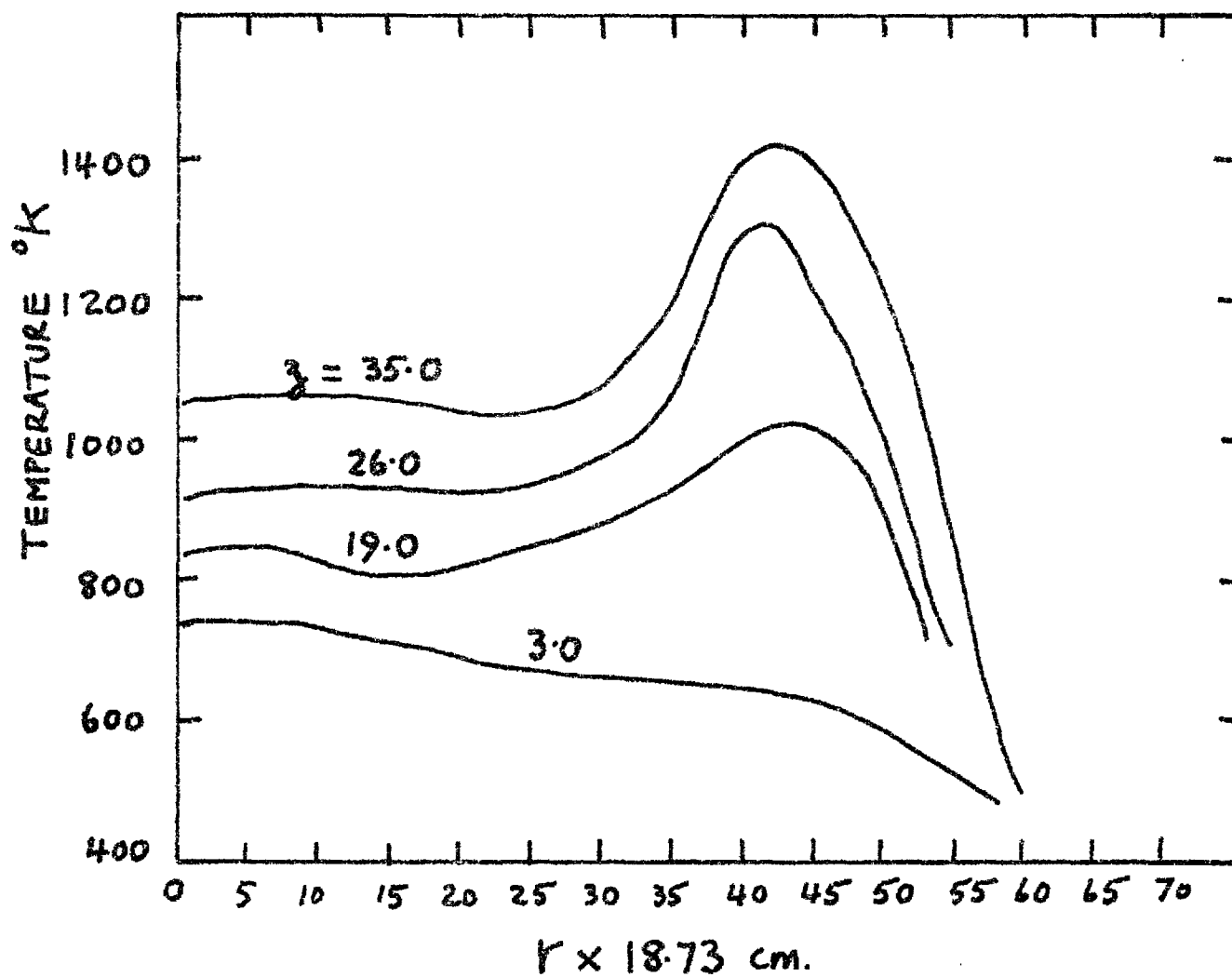


FIG.8.6

by almost  $300^{\circ}\text{K}$ ; which is hardly possible for pure air. This in fact illustrates what had been anticipated earlier theoretically; that because of the rapid attenuation of  $\delta$ , a small error in fringe order will give rise to a very large error in temperature, because in the analysis, a small quantity (the optical path across a single zone) had to be deduced by subtracting two large ones.

## CHAPTER 9.

RESULTS FOR AIR.

1. A preliminary investigation was conducted with air only flowing through the system. The reason for this was to establish whether the temperature and velocity data measured experimentally did in fact correspond to the theoretical model.

Since  $q = 0$  for air, the heat conservation equation for the system becomes :

$$\frac{1}{r} \frac{\partial}{\partial r} \left( -r K \frac{\partial T}{\partial r} + r v_r H \right) + \frac{\partial}{\partial y} \left( -K \frac{\partial T}{\partial z} + v_z H \right) = 0 \quad (9.1)$$

There are several ways in which the criterion of zero-heat release rate could be checked. For instance, by using the experimental velocity data and known values of  $K$ ,  $\rho$ ,  $H$ , a temperature distribution could be calculated and compared with the experimentally derived profile. Similarly, conductivity values for air as a function of temperature may be determined and then checked against established conductivity data. For the present purpose, a more obvious approach to the problem was made. Since conductivity, density and specific heat values are known with sufficient accuracy for air, it was possible to formulate equation (9.1)

to obtain the distribution of heat release, so that one could judge directly how closely this approached zero. The way in which this was done will now be discussed.

## 2. Temperature profiles.

The method of analysis, outlined in Chapter 8 , was applied to the interferogram shown in Plate 12 . The corrected fringe order distributions are shown in figure 9.1, for various z-cross-sections. Each z-section was now treated separately and a set of temperature profiles derived. Fig.9.2 gives the radial temperature distributions found. Tabulated results of the analysis are shown at the back of the thesis.

It will be noticed that the temperature distributions are all parabolic in shape and differ markedly from the profiles found earlier, when uncorrected fringe orders were considered. The maximum temperature for any z-section lies near the central axis of the system. The maximum experimental scatter, along the temperature profiles away from the two known boundaries, was found to be less than 5%, with considerably better correlation in the lower temperature regions. This reflects the increasing dependence of  $T$  on  $S$  with temperature.

It was pointed out earlier that preliminary studies



PLATE 12.

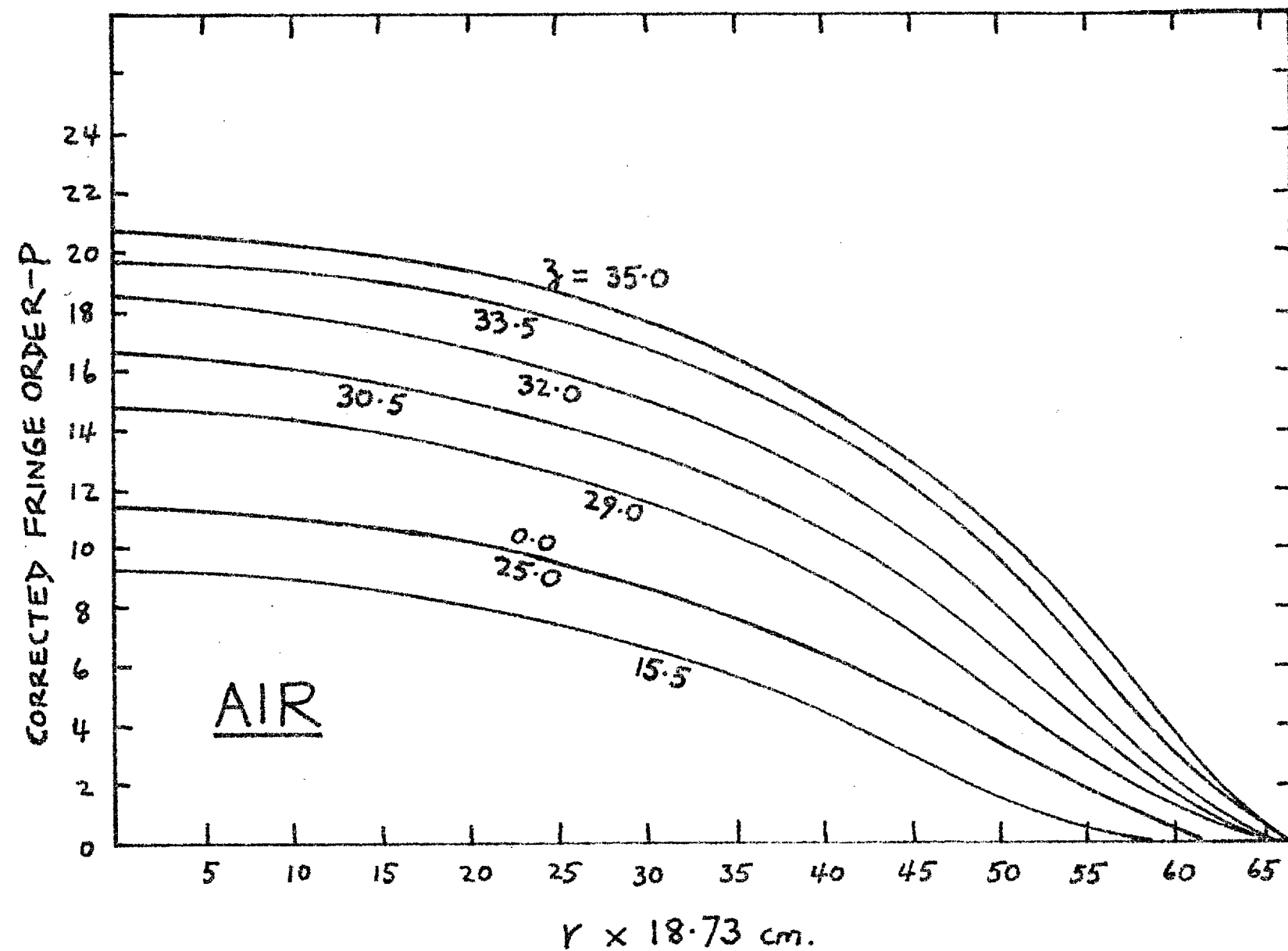


FIG. 9.1

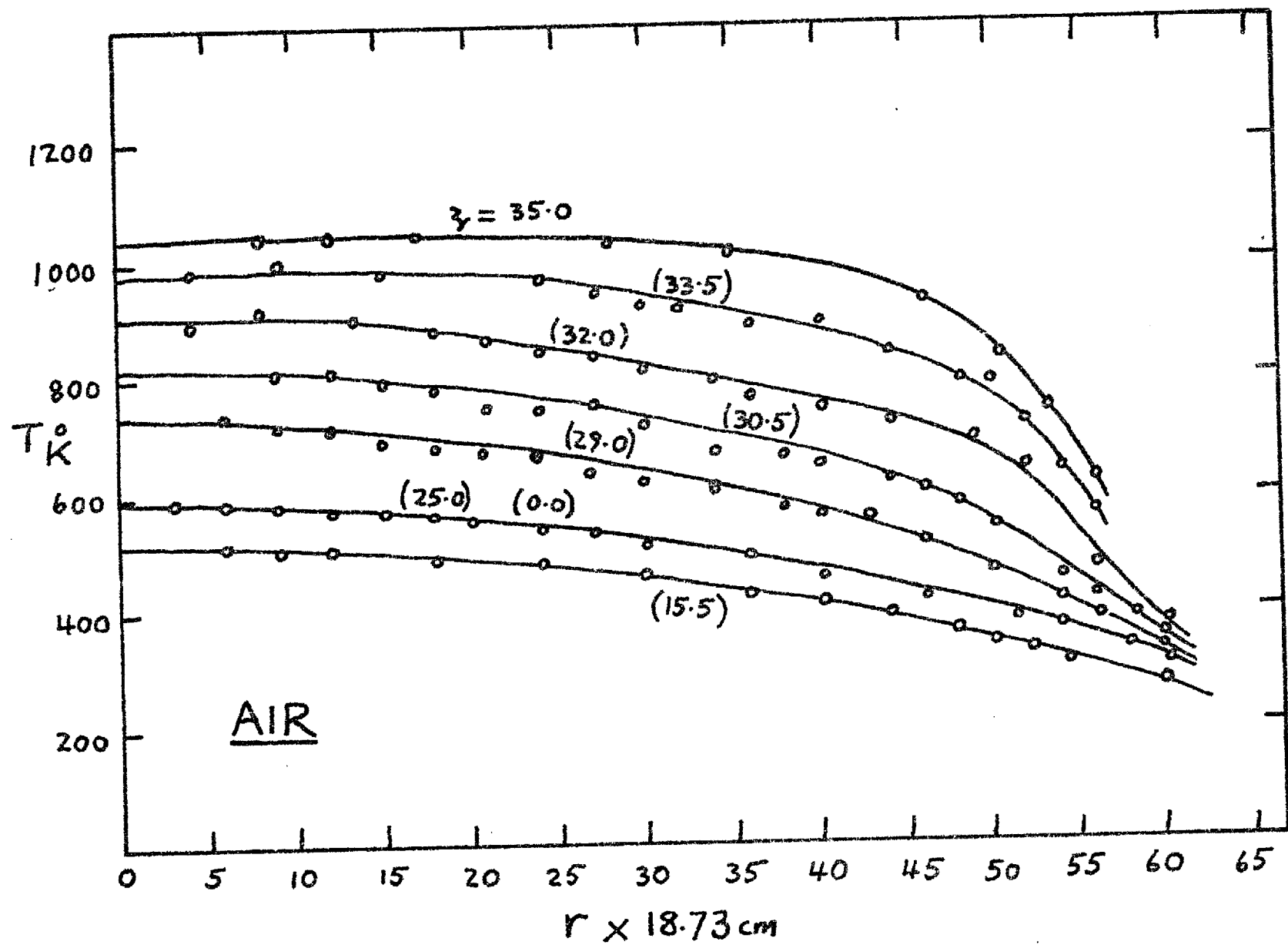


FIG.92

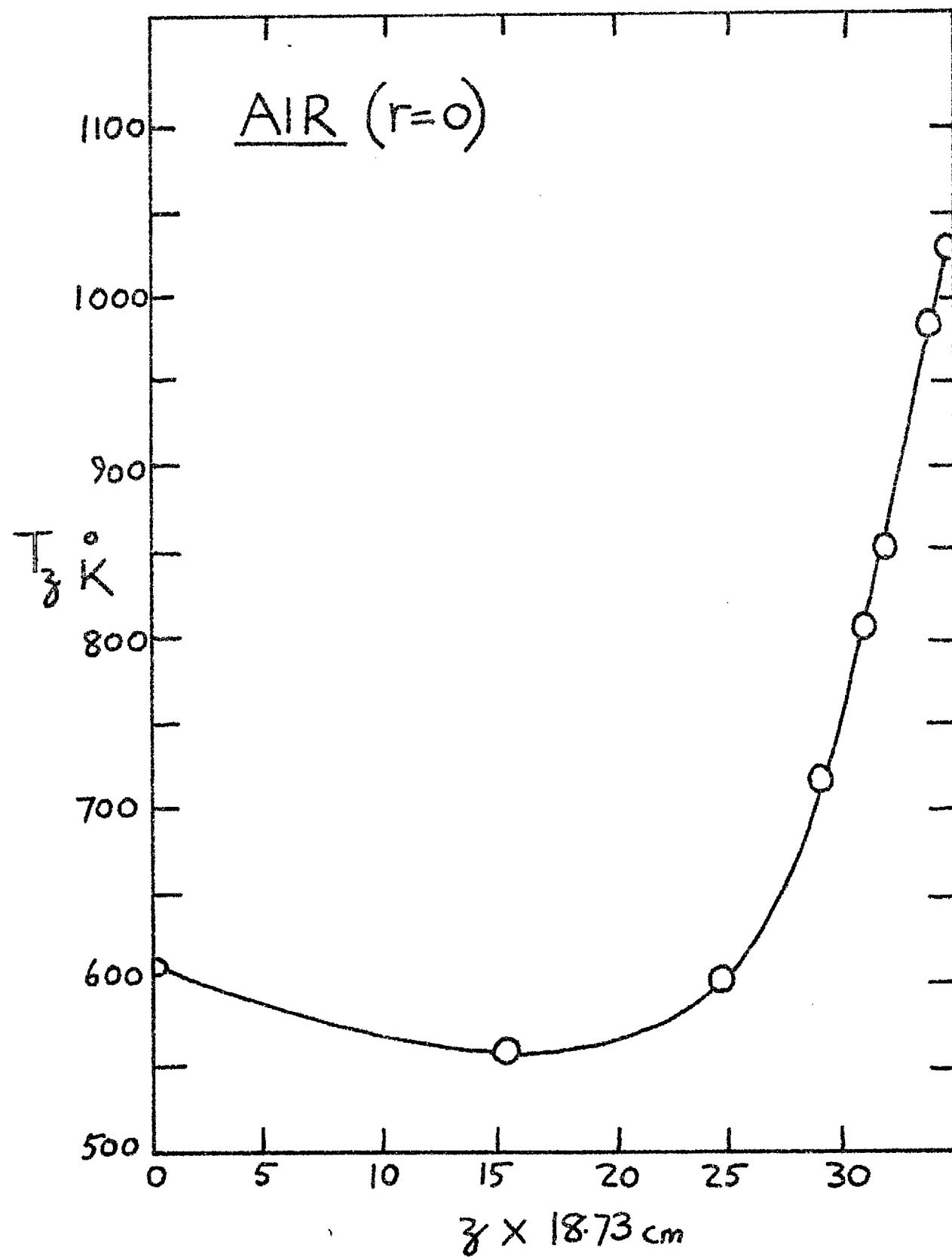
were confined to an elementary cylinder with the (z) axis as its axis of symmetry. Figure 9.3 shows the relevant temperature profile as a function of (z).

### 3. Velocity profiles.

Both radial and axial velocity distributions were determined from 5 particle-track photographs. The distributions of velocity for the system were shown in figures 7.5 and 7.7. In regions near to the disc surface, the particle tracks were not easily observed due to masking by light scattered at the surface. This difficulty was further aggravated by low particle density near to the central axis in this region. It was therefore necessary to extrapolate over the last  $10^{-1}$  cm or so to complete the profile.

### 4. Interpretation of the temperature and velocity profiles.

It had been established from preliminary work that the highest temperature region was at the centre of the system close to the disc surface. Since this region also coincided with a low velocity regime, it seemed probable that any pre-ignition reactions would be favoured to take place here. A preliminary analysis was therefore carried out along the central axis of the system.

FIG.9.3

Referring again to equation (9.1), it will be noticed that the first term involves the radial temperature gradient,  $\frac{\partial T}{\partial r}$ . The temperature profiles in figure 9.2 indicate that along the (z) axis,  $\frac{\partial T}{\partial r} \rightarrow 0$ . Simplification of equation (9.1) then gives

$$\frac{\partial}{\partial z} \left( -K \frac{\partial T}{\partial z} \right) + v_z \frac{\partial T}{\partial z} \left( f \frac{\partial H}{\partial T} + H \frac{\partial f}{\partial T} \right) + f H \frac{\partial v_z}{\partial z} + 2 \frac{v_r}{r} H = 0 \quad (9.2)$$

All necessary data on the temperature dependence of K,  $f$  and H for air were found in (73). The remaining temperature and velocity gradient terms were found by graphical differentiation of the relevant distribution curves. It was shown that the radial velocity ( $v_r$ ) was directly proportional to (r). Thus the last term in the equation was determined by finding the slope of the curves in figure 7.7, since in the limit  $r \rightarrow 0$ ,  $v_r/r = dv_r/dr$ .

Figures 9.4a and b show the distribution of  $v_r/r$  and  $dv_z/dz$  as a function of co-ordinate (z). All graphical differentiation was carried out on enlarged profiles. Equation (9.2) was applied to a number of points along the (z) axis and the residual heat release rates calculated. Tables 1 and 2 give the numerical data and the distribution of (q) so found is shown in figure 9.5 as a percentage of the conduction and convection heat flux.

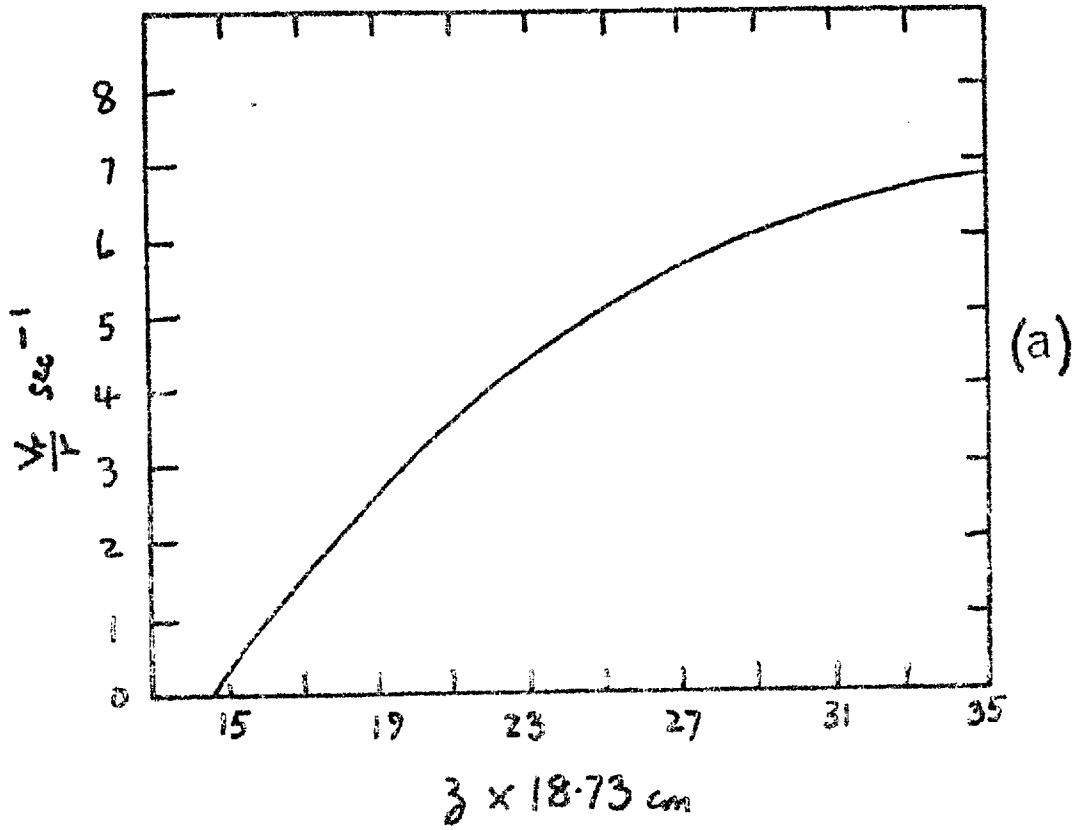
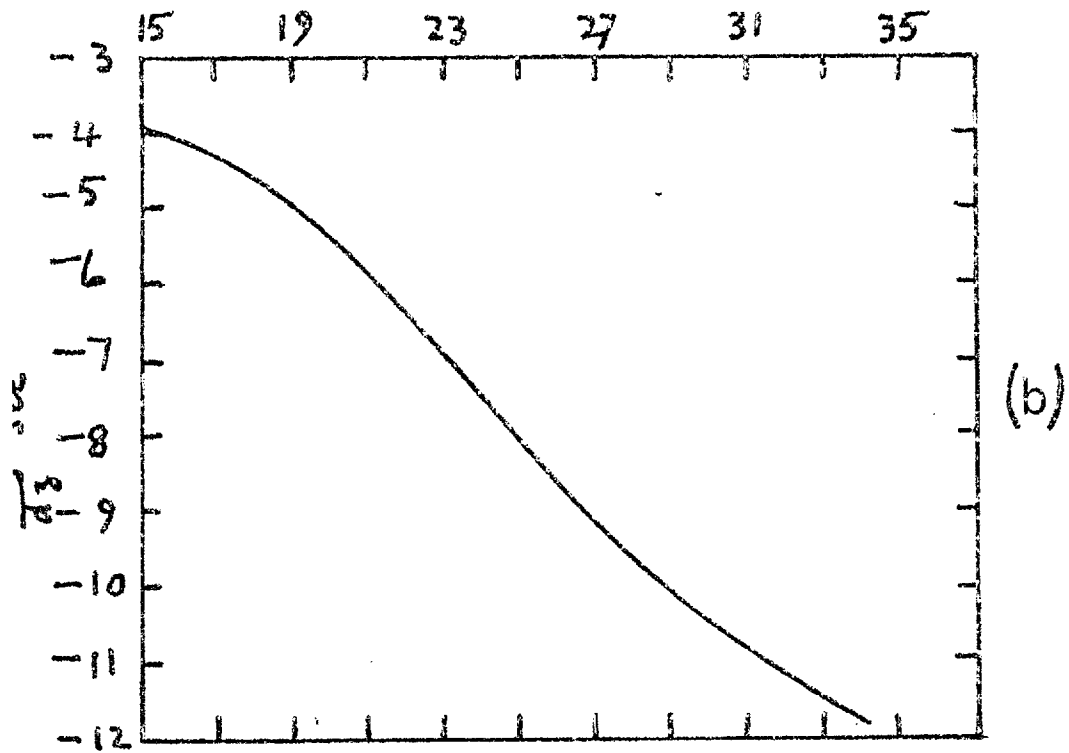


FIG.94



| $z \times 18.73$<br>(cm.) | $T$<br>(°K) | $\rho \times 10^4$<br>(gm.cm <sup>-3</sup> ) | $H$<br>(cal.gm <sup>-1</sup> ) | $\frac{V_r}{r}$<br>(sec <sup>-1</sup> ) | $-\frac{\partial \rho}{\partial T} \times 10^6$<br>(gm.cm <sup>-3</sup> °K <sup>-1</sup> ) | $\frac{\partial H}{\partial T}$<br>(cal.sec <sup>-1</sup> °K <sup>-1</sup> ) | $-\frac{\partial V_z}{\partial z}$<br>(sec <sup>-1</sup> ) | $\frac{\partial T}{\partial z}$<br>(°K.cm <sup>-1</sup> ) | $V_z$<br>(cm.sec <sup>-1</sup> ) |
|---------------------------|-------------|----------------------------------------------|--------------------------------|-----------------------------------------|--------------------------------------------------------------------------------------------|------------------------------------------------------------------------------|------------------------------------------------------------|-----------------------------------------------------------|----------------------------------|
| 15.0                      | 515         | 6.84                                         | 124.0                          | 0.20                                    | 1.26                                                                                       | 0.245                                                                        | 3.98                                                       | 0                                                         | 7.76                             |
| 19.0                      | 520         | 6.76                                         | 125.0                          | 2.60                                    | 1.19                                                                                       | 0.247                                                                        | 4.80                                                       | 74.92                                                     | 6.90                             |
| 21.0                      | 530         | 6.52                                         | 127.5                          | 3.40                                    | 1.15                                                                                       | 0.248                                                                        | 5.90                                                       | 123.62                                                    | 6.20                             |
| 23.0                      | 553         | 6.32                                         | 133.0                          | 4.30                                    | 1.07                                                                                       | 0.248                                                                        | 7.10                                                       | 269.71                                                    | 5.40                             |
| 25.0                      | 589         | 6.00                                         | 142.0                          | 5.30                                    | 0.97                                                                                       | 0.253                                                                        | 8.10                                                       | 423.30                                                    | 4.70                             |
| 27.0                      | 649         | 5.44                                         | 157.0                          | 5.80                                    | 0.83                                                                                       | 0.254                                                                        | 8.80                                                       | 696.76                                                    | 4.25                             |
| 29.0                      | 729         | 4.84                                         | 178.0                          | 6.06                                    | 0.68                                                                                       | 0.256                                                                        | 9.79                                                       | 899.04                                                    | 3.25                             |
| 30.0                      | 779         | 4.52                                         | 191.0                          | 6.20                                    | 0.58                                                                                       | 0.260                                                                        | 10.21                                                      | 945.86                                                    | 2.80                             |
| 31.0                      | 829         | 4.26                                         | 204.0                          | 6.34                                    | 0.52                                                                                       | 0.267                                                                        | 10.80                                                      | 958.23                                                    | 2.25                             |
| 32.0                      | 881         | 4.02                                         | 218.0                          | 6.40                                    | 0.46                                                                                       | 0.268                                                                        | 11.10                                                      | 965.82                                                    | 1.80                             |
| 33.0                      | 933         | 3.80                                         | 232.0                          | 6.48                                    | 0.42                                                                                       | 0.275                                                                        | 11.50                                                      | 966.91                                                    | 1.20                             |
| 34.0                      | 983         | 3.60                                         | 246.0                          | 6.60                                    | 0.38                                                                                       | 0.287                                                                        | 11.80                                                      | 966.80                                                    | 0.60                             |

TABLE 1

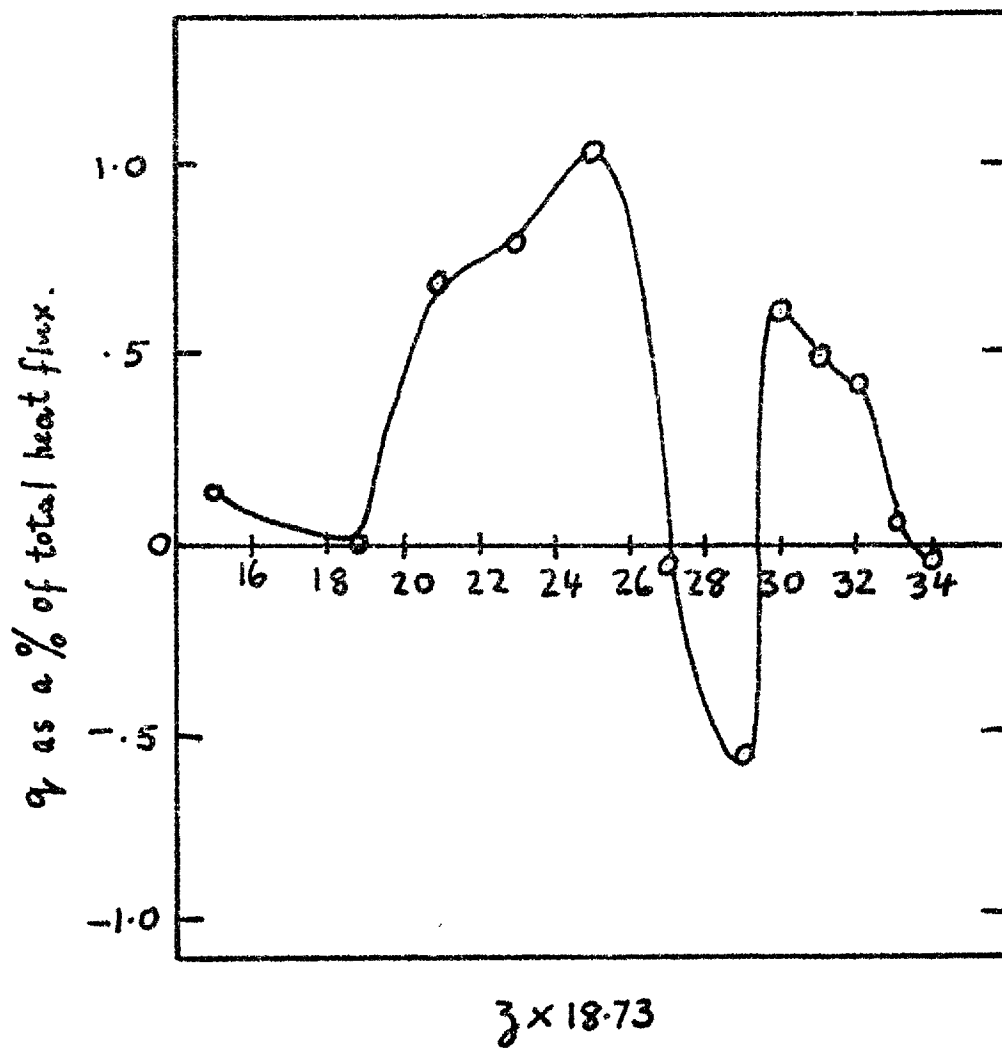
| $q$<br>cal.cm. <sup>-3</sup> .sec <sup>-1</sup> | $\frac{\partial}{\partial z}(-k\frac{\partial T}{\partial z})$ | $V_3 \frac{\partial T}{\partial z}$ | $\frac{2V}{T}(\rho H) \times 10^4$ | $H \frac{\partial \rho}{\partial T} \times 10^6$ | $V_3 \frac{\partial T}{\partial z} (\rho \frac{\partial H}{\partial T}) \times 10^3$ | $\rho H \times 10^4$ | $-V_3 \frac{\partial T}{\partial z} (H \frac{\partial \rho}{\partial T}) \times 10^3$ | $-\rho H \frac{\partial V_3}{\partial z} \times 10$ | $\rho \frac{\partial H}{\partial T} \times 10^4$ | $z \times 18.73$ |
|-------------------------------------------------|----------------------------------------------------------------|-------------------------------------|------------------------------------|--------------------------------------------------|--------------------------------------------------------------------------------------|----------------------|---------------------------------------------------------------------------------------|-----------------------------------------------------|--------------------------------------------------|------------------|
| 0.001                                           | —                                                              | —                                   | 3392                               | —                                                | —                                                                                    | 848.2                | —                                                                                     | 3.38                                                | —                                                | 15.0             |
| 0                                               | 0.044                                                          | 516.9                               | 4394                               | 148.8                                            | 8.66                                                                                 | 845.0                | 76.9                                                                                  | 4.06                                                | 1.669                                            | 19.0             |
| 0.01                                            | 0.085                                                          | 766.4                               | 5900                               | 146.6                                            | 12.39                                                                                | 831.3                | 112.4                                                                                 | 5.07                                                | 1.617                                            | 21.0             |
| 0.015                                           | 0.130                                                          | 1456.4                              | 7228                               | 142.3                                            | 22.83                                                                                | 840.6                | 207.3                                                                                 | 5.98                                                | 1.567                                            | 23.0             |
| 0.025                                           | 0.216                                                          | 1989.5                              | 9031                               | 137.7                                            | 30.20                                                                                | 852.0                | 274.0                                                                                 | 6.90                                                | 1.518                                            | 25.0             |
| -0.001                                          | 0.268                                                          | 2961.4                              | 9907                               | 130.3                                            | 41.37                                                                                | 854.1                | 385.9                                                                                 | 7.52                                                | 1.397                                            | 27.0             |
| -0.016                                          | 0.225                                                          | 2921.8                              | 10441                              | 121.0                                            | 36.20                                                                                | 861.5                | 353.7                                                                                 | 8.44                                                | 1.239                                            | 29.0             |
| 0.017                                           | 0.191                                                          | 2648.4                              | 10705                              | 110.8                                            | 31.12                                                                                | 863.3                | 293.4                                                                                 | 8.80                                                | 1.175                                            | 30.0             |
| 0.013                                           | 0.165                                                          | 2156.0                              | 11019                              | 106.9                                            | 24.54                                                                                | 869.0                | 230.5                                                                                 | 9.38                                                | 1.137                                            | 31.0             |
| 0.011                                           | 0.148                                                          | 1738.4                              | 11218                              | 101.1                                            | 18.72                                                                                | 876.4                | 176.5                                                                                 | 9.73                                                | 1.077                                            | 32.0             |
| 0.001                                           | 0.135                                                          | 1160.3                              | 11425                              | 97.4                                             | 12.12                                                                                | 881.6                | 113.0                                                                                 | 10.14                                               | 1.045                                            | 33.0             |
| -0.008                                          | 0.130                                                          | 676.7                               | 11610                              | 92.5                                             | 6.99                                                                                 | 885.6                | 62.6                                                                                  | 10.45                                               | 1.033                                            | 34.0             |

TABLE 2

## 5. Observations and conclusions.

It is apparent from figure 9.5 that no meaningful significance can be attached to the small heat release rates found. The points are scattered on both sides of the (z) axis and are probably due to small errors in differentiating graphically. Since there seems to be no set trend in the profile, no corrections for zero  $q$  were attempted. (i.e. for example, if small negative heat release rates had been found throughout it is possible that the neglect of a radiation term in the heat conservation equation would account for the observed rates of negative heat release.)

The magnitude of the heat release rates, as a percentage of the conduction and convection terms, was shown to be very small, not exceeding 1-2% in all cases. These effects were therefore considered negligible and it was concluded that the temperature and velocity distributions in the 2-dimensional system had been interpreted successfully.

FIG.9.5

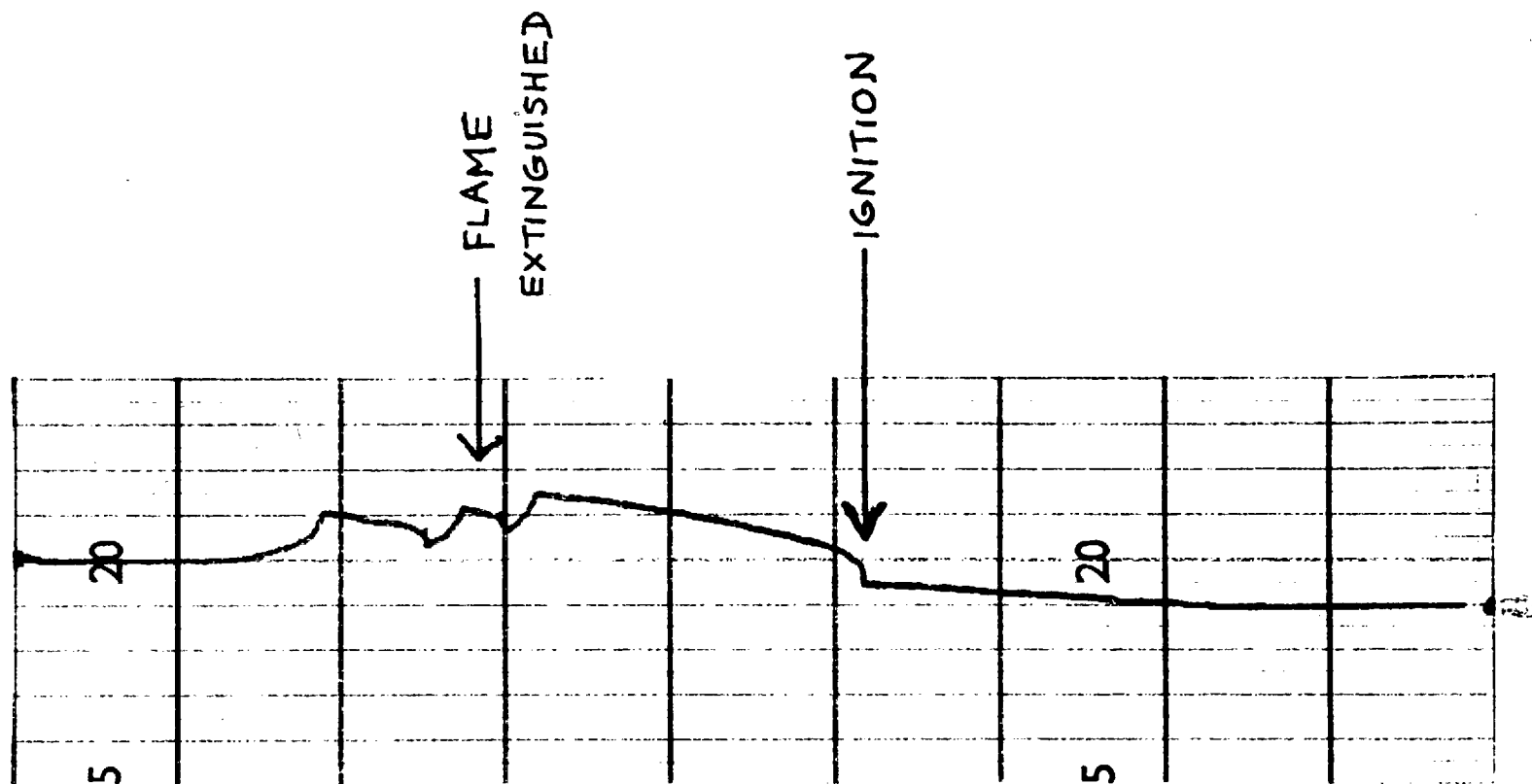


PLATE 13.

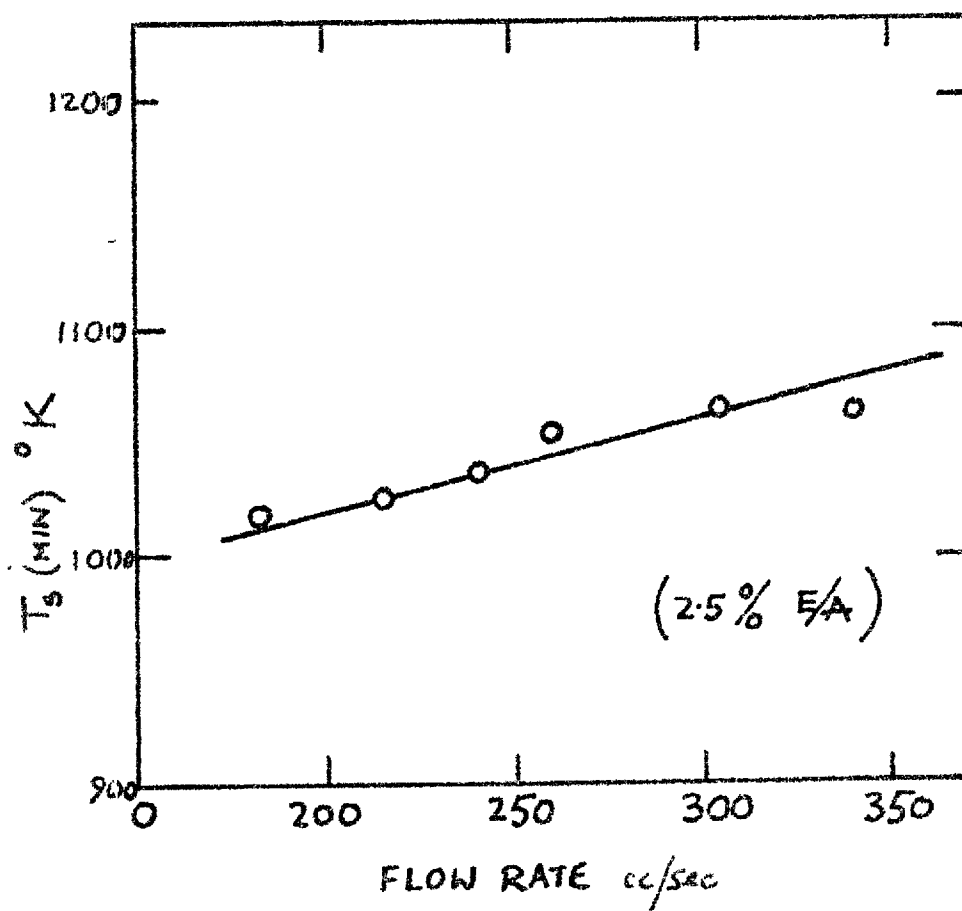
## CHAPTER 10.

EXPERIMENTS WITH ETHYLENE - AIR MIXTURES.

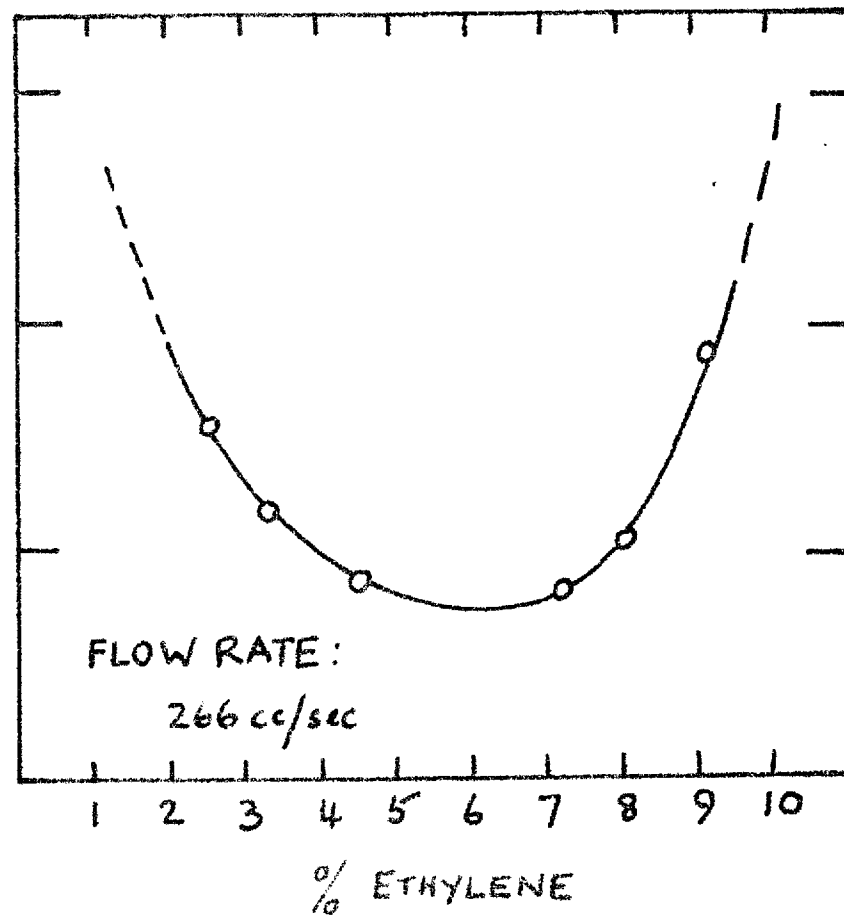
1. The range of conditions over which the system might be used was first investigated. Both fuel concentration and stream velocity were found to influence the minimum surface temperature necessary to ignite the flowing gas. Results, which are characteristic of all the experiments carried out, are shown in Figure 10.1. The experimental procedure was to specify first the flow rate and ethylene concentration of the gas mixture. While keeping the flow conditions constant, the surface temperature of the disc was slowly raised by increasing the heater current until ignition occurred. By repeating the experiment and adjusting the heater current, a minimum surface ignition temperature could be found for various fuel concentrations and flow stream velocities. During these experiments the temperature history of the disc surface was registered automatically on the single pen recorder. Ignition onset accompanied by flame formation was clearly recorded by a spike in the output of the pyrometer as shown in Plate 13.

The main conclusions drawn from the results are

- (1) the minimum ignition temperature shows an approximately linear dependence on the approach flow stream velocity;
- (2) for a given flow rate, the minimum ignition temperature



(a)



(b)

FIG.10.1

corresponds to an approximately stoichiometric ethylene/air mixture, as in Paterson's work with quiescent gas mixtures and hot, moving spheres.

The extent of the study over a wide range of flow rate and mixture strength was restricted by the maximum surface temperature to which the surface could safely be raised. The temperature limit approached  $1100^{\circ}\text{K}$ . Above this value, the Kanthal wire elements rapidly oxidised and burned out.

## 2. Experimental conditions.

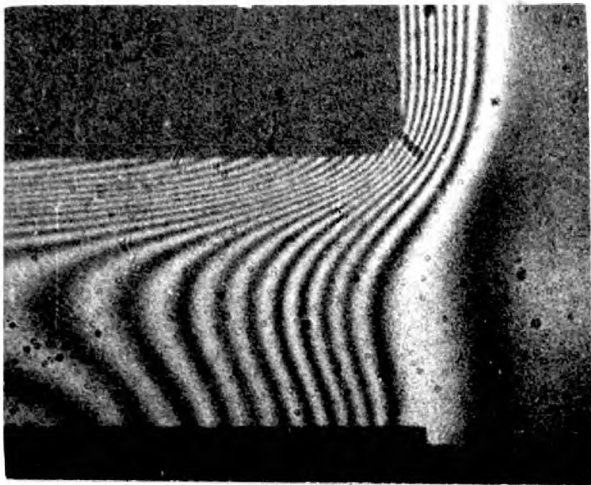
The choice of flow rate proved to be important for two reasons. Under very slow flow conditions, the symmetry of the flow pattern was perturbed by small local disturbances in the laboratory atmosphere. With increasing flow rate, these effects became imperceptible. At very high flow rates, however, the temperature field became compressed against the disc surface, until the steepness of the consequent refractive index gradients made interferometry unsuitable. (Though this would not, of course, preclude the use of deflection mapping). An overall flow rate of 11.83 litres/minute was chosen, for which both flow symmetry and fringe resolution in the interferograms were satisfactory. In order to avoid complications due to

appreciable composition changes, a lean, 3.5% ethylene/air mixture was used. Under the above flow conditions, the minimum surface ignition temperature for the 3.5% mixture was found to be about  $1040^{\circ}\text{K}$ , and was readily reproducible.

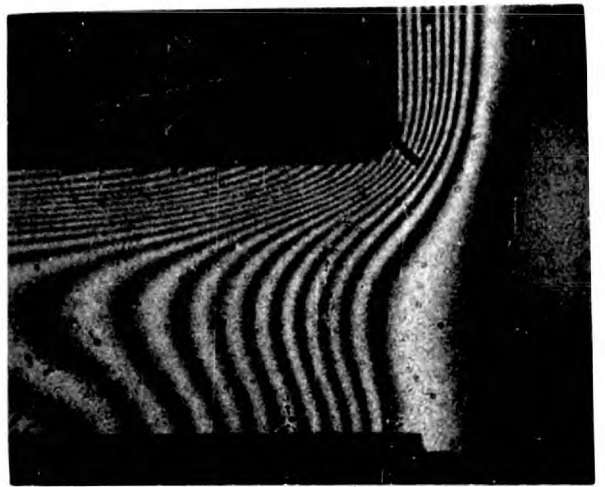
### 3. Interpretation of the interferogram.

The experimental procedure outlined in Chapter 7 was strictly followed. Plate 14 shows the ignition sequence photographed under the conditions stated above. The frame preceding that showing complete break up of the fringe pattern contains the relevant information about the temperature distribution prior to ignition onset. After pegging the interferogram and applying the necessary corrections, as in the case for air, smoothed order distributions were obtained as shown in figure 10.2. Each z-section was then analysed to yield the radial temperature profiles in figure 10.3. It will be noticed that the radial temperature gradients  $(\frac{\partial T}{\partial r})$  at the axis tend to zero, enabling the heat conservation equation anywhere along the axis to be simplified as in the case for air.

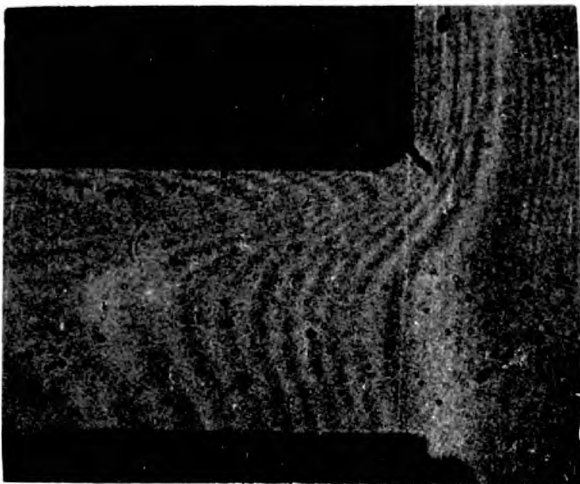
Using these temperature distributions, isothermals were mapped out over the entire test region, and these are shown in figure 10.4. From this figure it is noticed that the steepest temperature gradients lie near the periphery



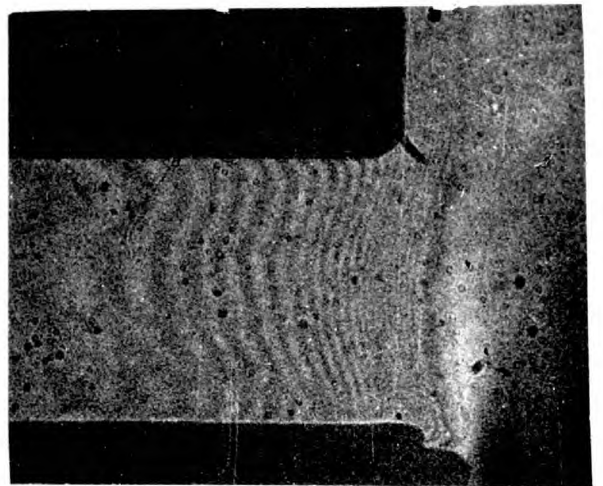
1



2



3



4

PLATE 14

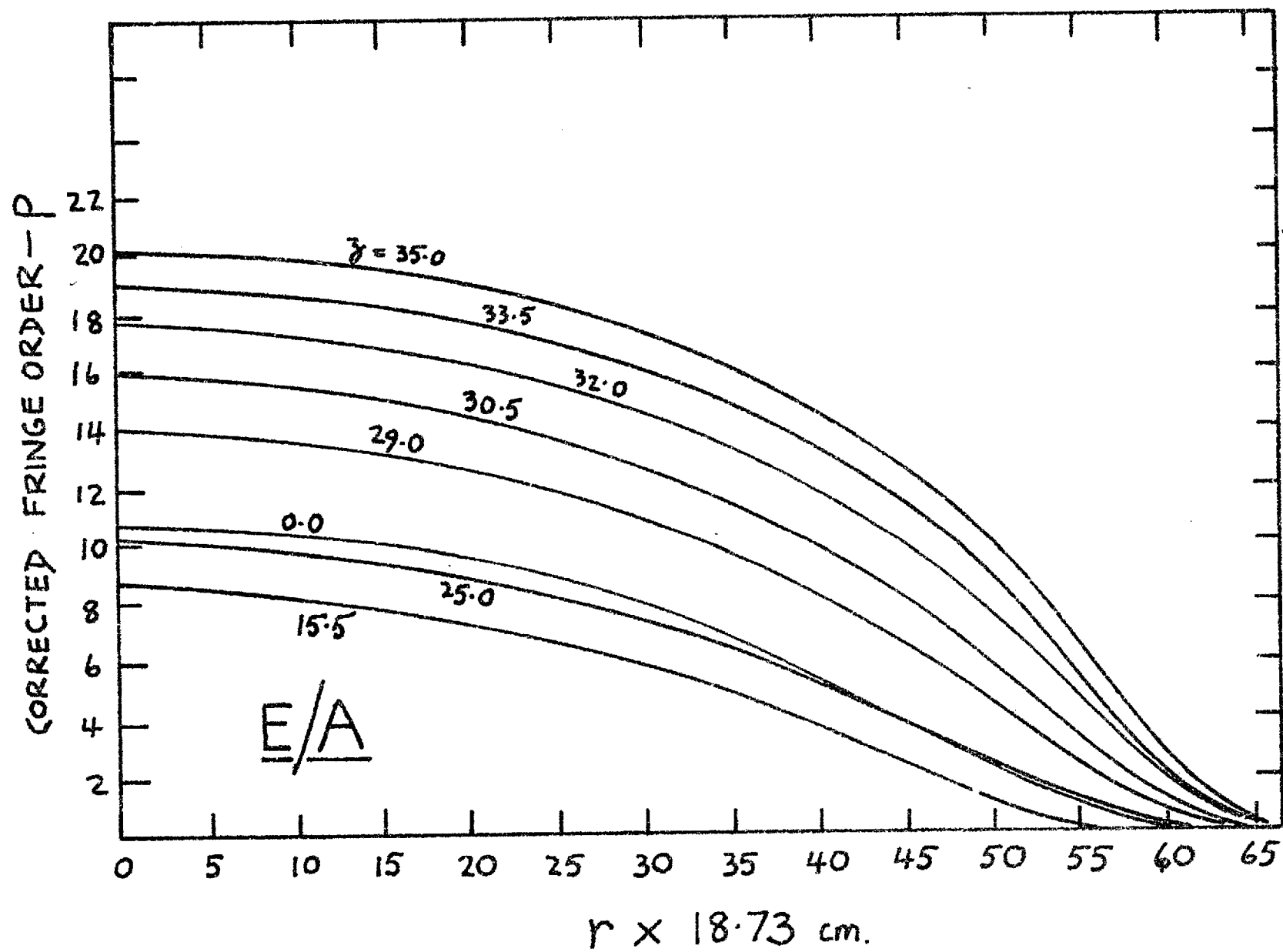


FIG.10.2

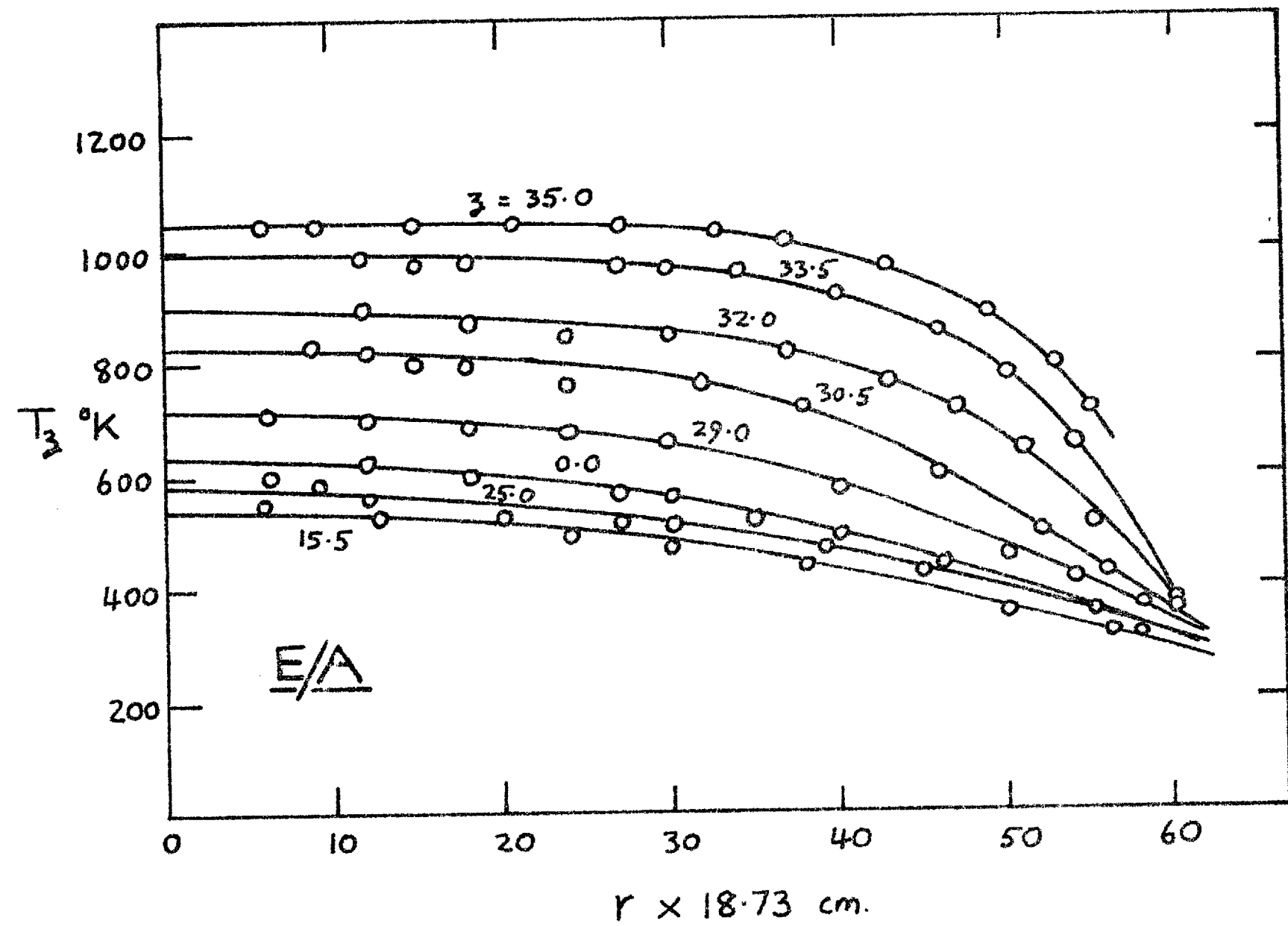


FIG.10.3

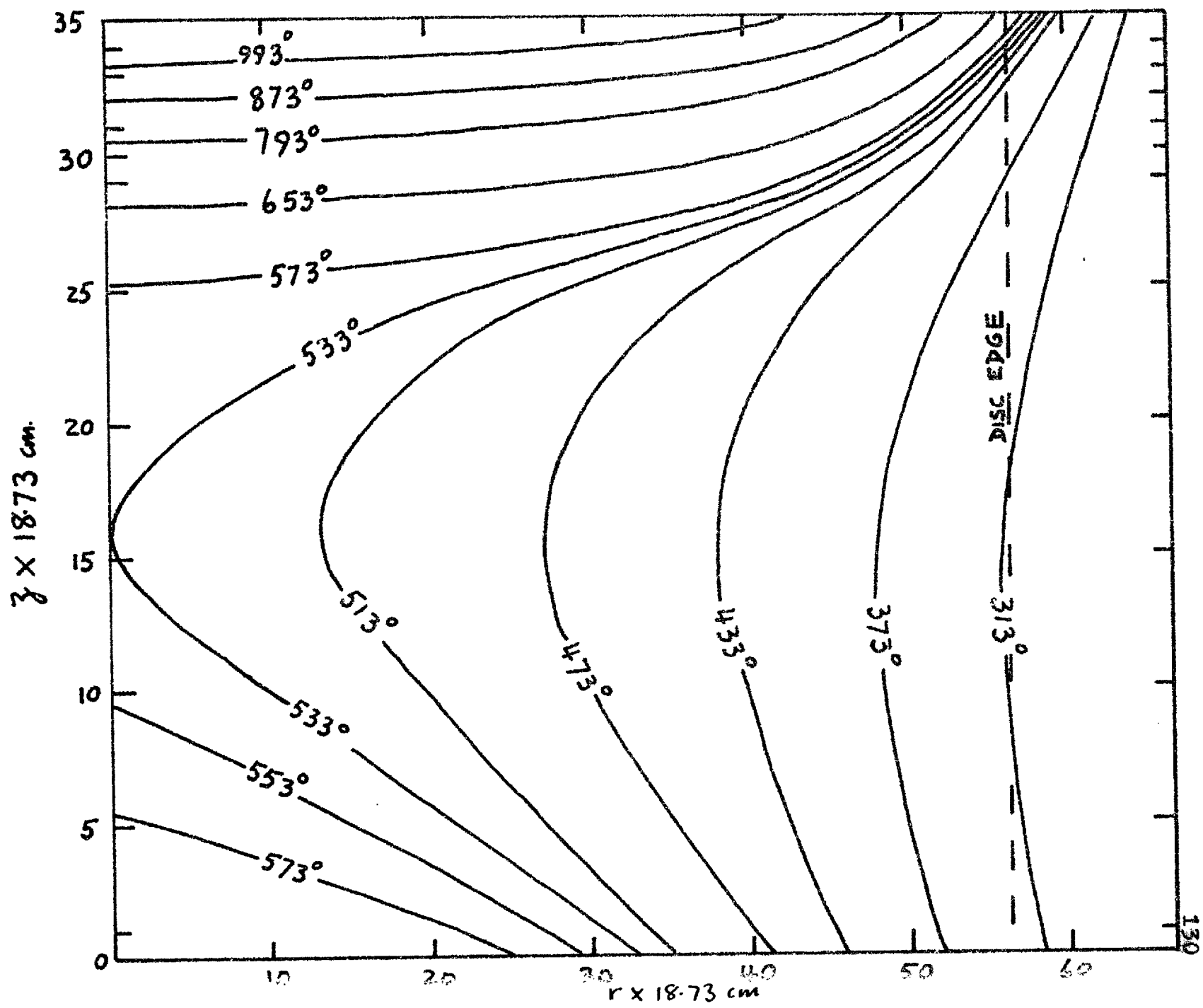


FIG.10.4

of the disc where the hot stream meets cold entrained gas. The isothermals near the matrix surface indicate that the gas loses heat initially, after passing through the matrix, probably by lateral conduction to the colder entrained gas. Fig 10.5 shows the axial temperature profile which was used for subsequent calculations.

#### 4. Velocity distributions.

Velocity data for this experiment was obtained from the measurement of 6 particle-track photographs, taken on H.P.3 plates at a camera setting of  $f/8$  and time exposure of  $1/10$  the second. It was obviously essential that the particle-track photographs should be taken under conditions directly relevant to the interferometric sequence recorded.

In practice, photographing the particle-tracks demanded patience because, with the system adjusted to operate at the ignition threshold, there was a constant risk of ignition-onset occurring as the result of minor perturbations while the particles were being photographed. By repeating the experiment many times, however, a sufficient number of particle track photographs were obtained just before spontaneous ignition occurred. The relevant profiles of axial and radial velocities are given in figures 10.6 and 10.7.

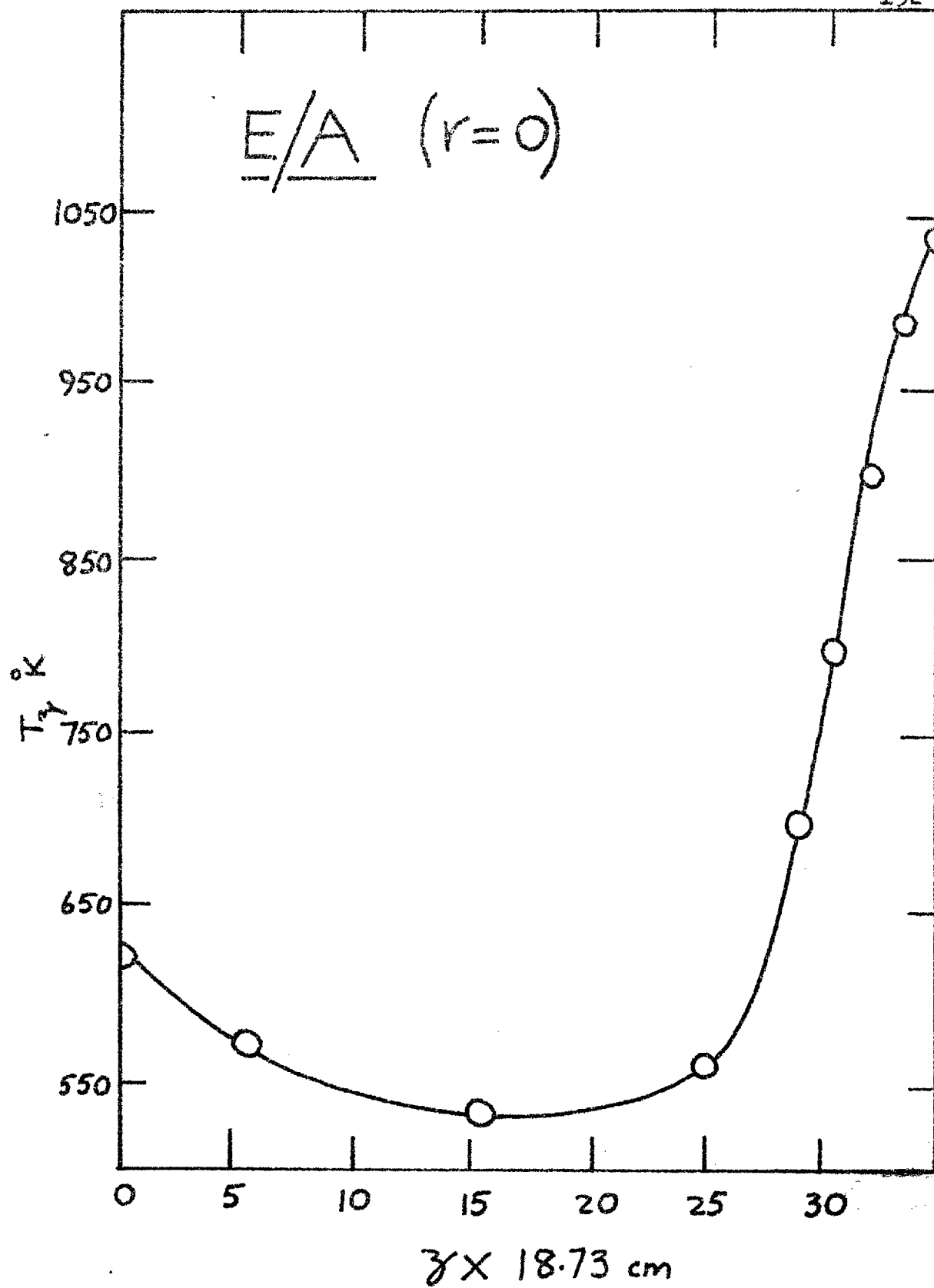


FIG.10.5

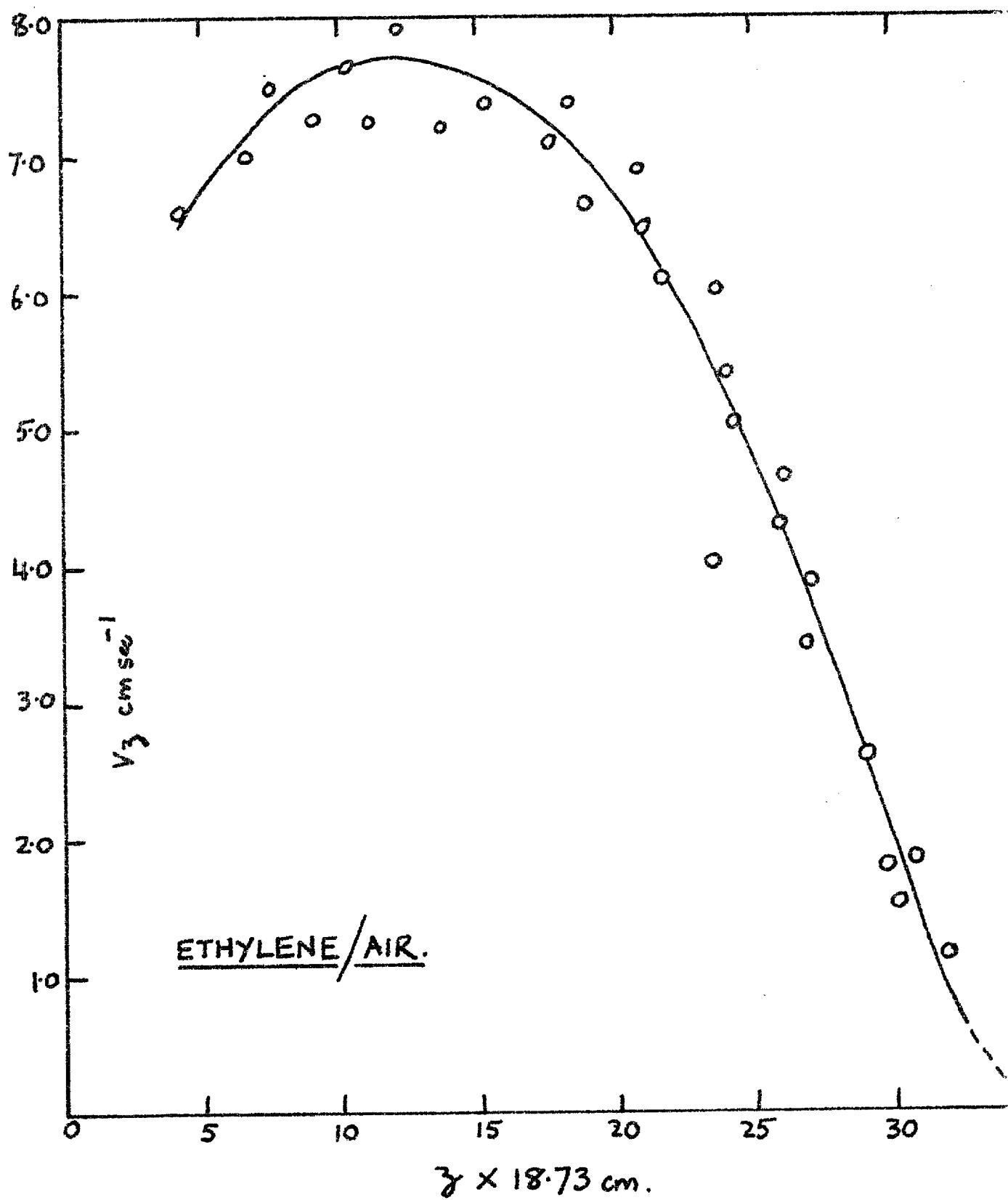


FIG.10.6

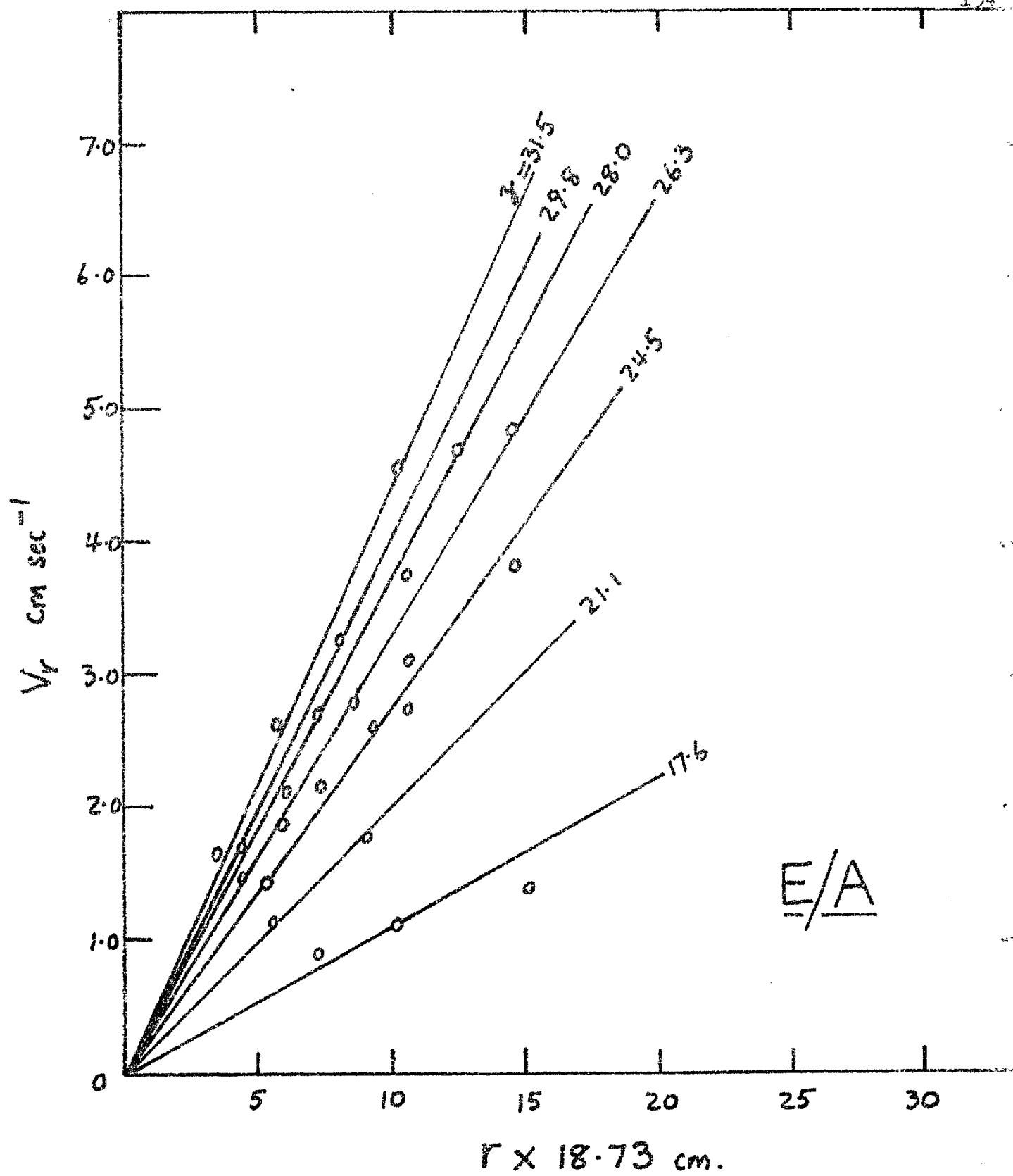


FIG.10.7

## 5. Derivation of heat release profile.

Enlarged profiles of the temperature and velocity distributions were used for the graphical differentiation needed to solve the heat conservation equation for (q). The curves so found for  $dv_z/dz$  and  $dv_r/dr$  as functions of co-ordinate (z) are shown in figures 10.8a and b respectively.

The conduction term  $K \frac{dT}{dz}$  was plotted against (z) as in figure 10.8c. These smoothed distribution curves, together with data found in (73) on the temperature dependence of enthalpy and density, were used to determine values of local heat release rates. Numerical values for the relevant terms in the heat conservation equation are given in Tables 3 and 4. Calculated values of local H, K, and  $\rho$  for a 3.5% ethylene/air mixture were used in the analysis. Under the conditions of very lean mixture strength and small extent of reaction, the composition of the gas was considered to remain constant throughout the system. Using the method in (78), the variation of the arithmetic mean conductivity of the gas mixture with temperature was found (see figure 10.9). As had been anticipated, the difference in conductivity between pure air and the weak mixture is marginal. The local heat release rates were plotted against (z) giving the profile shown in fig.10.10 and will be discussed in the next chapter.

$\lambda \times 18.73 \text{ cm.}$

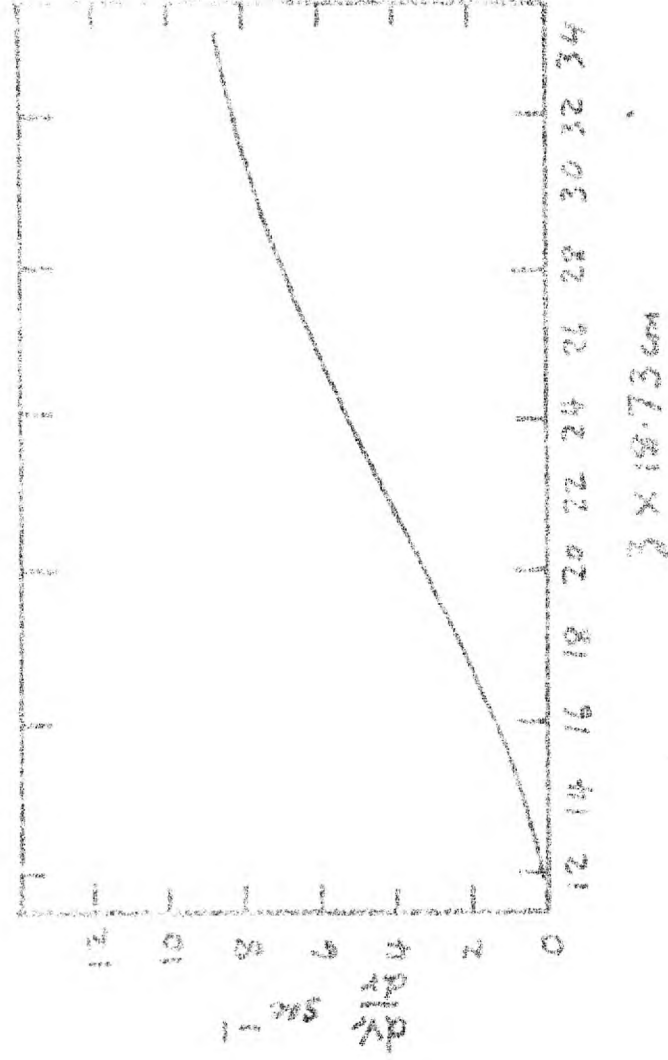
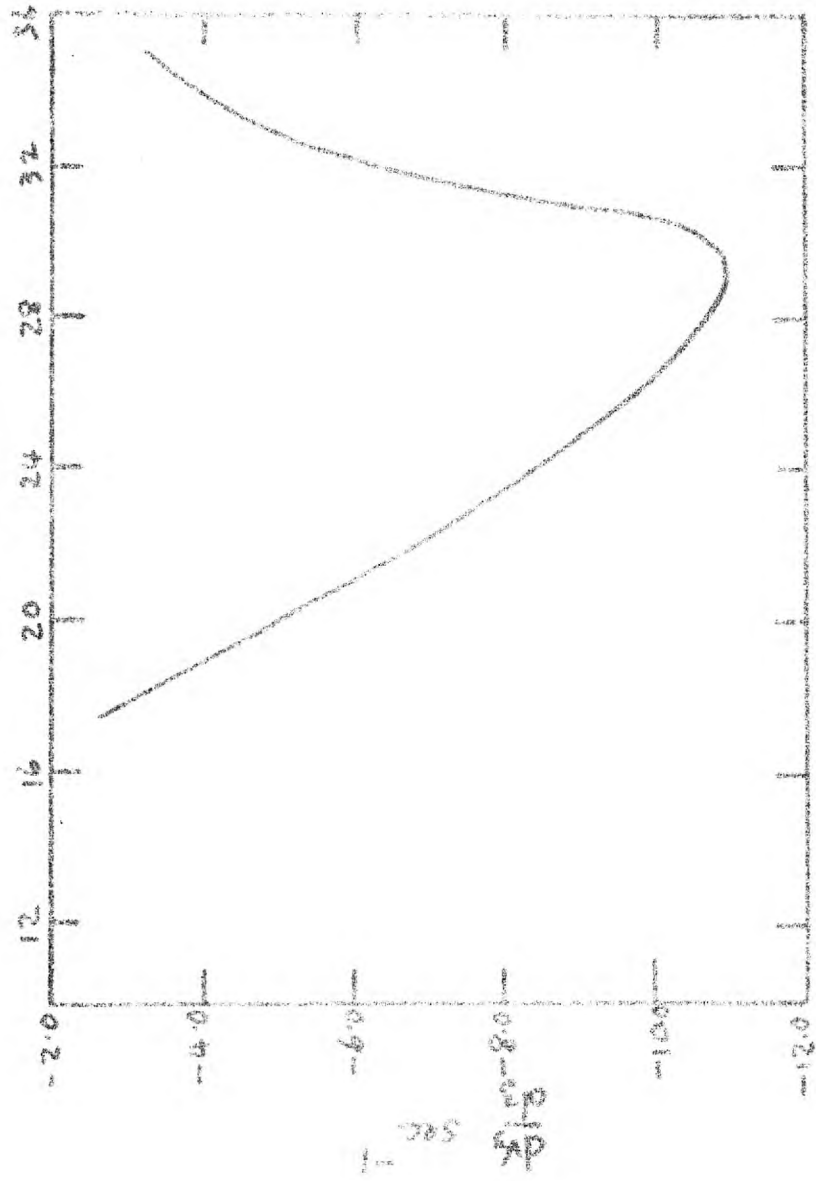


FIG. 10.8

| $z \times 18.73$<br>(cm.) | $T$<br>(°K) | $\rho \times 10^4$<br>(gm.cm <sup>-3</sup> ) | $H$<br>(cal.gm <sup>-1</sup> ) | $\frac{V_r}{r}$<br>(sec <sup>-1</sup> ) | $-\frac{\partial \rho}{\partial T} \times 10^6$<br>(gm.cm <sup>-3</sup> .°K <sup>-1</sup> ) | $\frac{\partial H}{\partial T}$<br>(cal.sec <sup>-1</sup> .°K <sup>-1</sup> ) | $-\frac{\partial V_z}{\partial z}$<br>(sec <sup>-1</sup> ) | $\frac{\partial T}{\partial z}$<br>(°K.cm <sup>-1</sup> ) | $V_z$<br>(cm.sec <sup>-1</sup> ) |
|---------------------------|-------------|----------------------------------------------|--------------------------------|-----------------------------------------|---------------------------------------------------------------------------------------------|-------------------------------------------------------------------------------|------------------------------------------------------------|-----------------------------------------------------------|----------------------------------|
| 34.0                      | 1012        | 3.50                                         | 270                            | 8.85                                    | 0.388                                                                                       | 0.310                                                                         | 4.08                                                       | 706.1                                                     | 0.30                             |
| 33.0                      | 966         | 3.68                                         | 257                            | 8.45                                    | 0.422                                                                                       | 0.306                                                                         | 5.15                                                       | 926.6                                                     | 0.60                             |
| 32.0                      | 910         | 3.88                                         | 240                            | 8.15                                    | 0.472                                                                                       | 0.300                                                                         | 8.17                                                       | 1161.3                                                    | 1.00                             |
| 31.0                      | 840         | 4.22                                         | 220                            | 7.90                                    | 0.552                                                                                       | 0.293                                                                         | 8.63                                                       | 1404.7                                                    | 1.50                             |
| 30.0                      | 764         | 4.64                                         | 198                            | 7.75                                    | 0.654                                                                                       | 0.285                                                                         | 10.80                                                      | 1412.2                                                    | 2.00                             |
| 28.0                      | 644         | 5.50                                         | 164                            | 7.00                                    | 0.866                                                                                       | 0.273                                                                         | 10.44                                                      | 690.6                                                     | 3.10                             |
| 26.0                      | 585         | 6.04                                         | 149                            | 6.15                                    | 1.000                                                                                       | 0.267                                                                         | 9.70                                                       | 420.7                                                     | 4.50                             |
| 24.0                      | 553         | 6.38                                         | 140                            | 5.12                                    | 1.100                                                                                       | 0.263                                                                         | 8.20                                                       | 198.7                                                     | 5.60                             |
| 20.0                      | 530         | 6.64                                         | 134                            | 3.20                                    | 1.170                                                                                       | 0.261                                                                         | 4.79                                                       | 28.5                                                      | 7.00                             |
| 10.0                      | 549         | 6.40                                         | 139                            |                                         | 1.108                                                                                       | 0.263                                                                         | -1.20                                                      | 74.2                                                      | 7.56                             |

TABLE 3

| $q$<br>$\text{cal. cm. sec}^{-1}$ | $-\frac{\partial}{\partial z}(-\kappa \frac{\partial T}{\partial z})$ | $V \frac{\partial T}{\partial z}$ | $\frac{2M}{r}(\rho H) \times 10^3$ | $H \frac{\partial \rho}{\partial T} \times 10^6$ | $V \frac{\partial T}{\partial z} (\rho \frac{\partial H}{\partial T}) \times 10^2$ | $\rho H \times 10^4$ | $-V \frac{\partial T}{\partial z} (H \frac{\partial \rho}{\partial T}) \times 10^3$ | $-\rho H \frac{\partial V}{\partial z} \times 10$ | $\rho \frac{\partial H}{\partial T} \times 10^4$ | $z \times 18.73$ |
|-----------------------------------|-----------------------------------------------------------------------|-----------------------------------|------------------------------------|--------------------------------------------------|------------------------------------------------------------------------------------|----------------------|-------------------------------------------------------------------------------------|---------------------------------------------------|--------------------------------------------------|------------------|
| 2.080                             | -0.780                                                                | 211.8                             | 1673                               | 104.76                                           | 2.29                                                                               | 945                  | 22.2                                                                                | 385                                               | 1.08                                             | 34.0             |
| 1.680                             | -0.541                                                                | 555.9                             | 1600                               | 108.45                                           | 6.28                                                                               | 946                  | 60.3                                                                                | 487                                               | 1.13                                             | 33.0             |
| 1.240                             | -0.455                                                                | 1161.3                            | 1517                               | 113.28                                           | 13.47                                                                              | 931                  | 131.5                                                                               | 761                                               | 1.16                                             | 32.0             |
| 0.720                             | 0                                                                     | 2107.0                            | 1467                               | 121.44                                           | 26.13                                                                              | 928                  | 255.9                                                                               | 8.01                                              | 1.24                                             | 31.0             |
| 0.060                             | +0.560                                                                | 2824.4                            | 1424                               | 129.49                                           | 37.28                                                                              | 918                  | 365.7                                                                               | 9.92                                              | 1.32                                             | 30.0             |
| 0.142                             | +0.350                                                                | 2140.8                            | 1263                               | 142.02                                           | 32.11                                                                              | 902                  | 304.0                                                                               | 9.41                                              | 1.50                                             | 28.0             |
| 0.155                             | +0.295                                                                | 1893.1                            | 1107                               | 149.60                                           | 30.48                                                                              | 900                  | 283.2                                                                               | 8.73                                              | 1.61                                             | 26.0             |
| 0.222                             | +0.113                                                                | 1112.7                            | 914                                | 154.00                                           | 18.69                                                                              | 893                  | 171.3                                                                               | 7.32                                              | 1.68                                             | 24.0             |
| 0.134                             | +0.023                                                                | 199.3                             | 570                                | 156.78                                           | 3.45                                                                               | 890                  | 31.3                                                                                | 4.45                                              | 1.73                                             | 20.0             |
| 0.039                             | -0.011                                                                | 560.7                             | 0                                  | 154.01                                           | -9.42                                                                              | 890                  | -86.3                                                                               | -1.07                                             | 1.68                                             | 10.0             |

TABLE 4

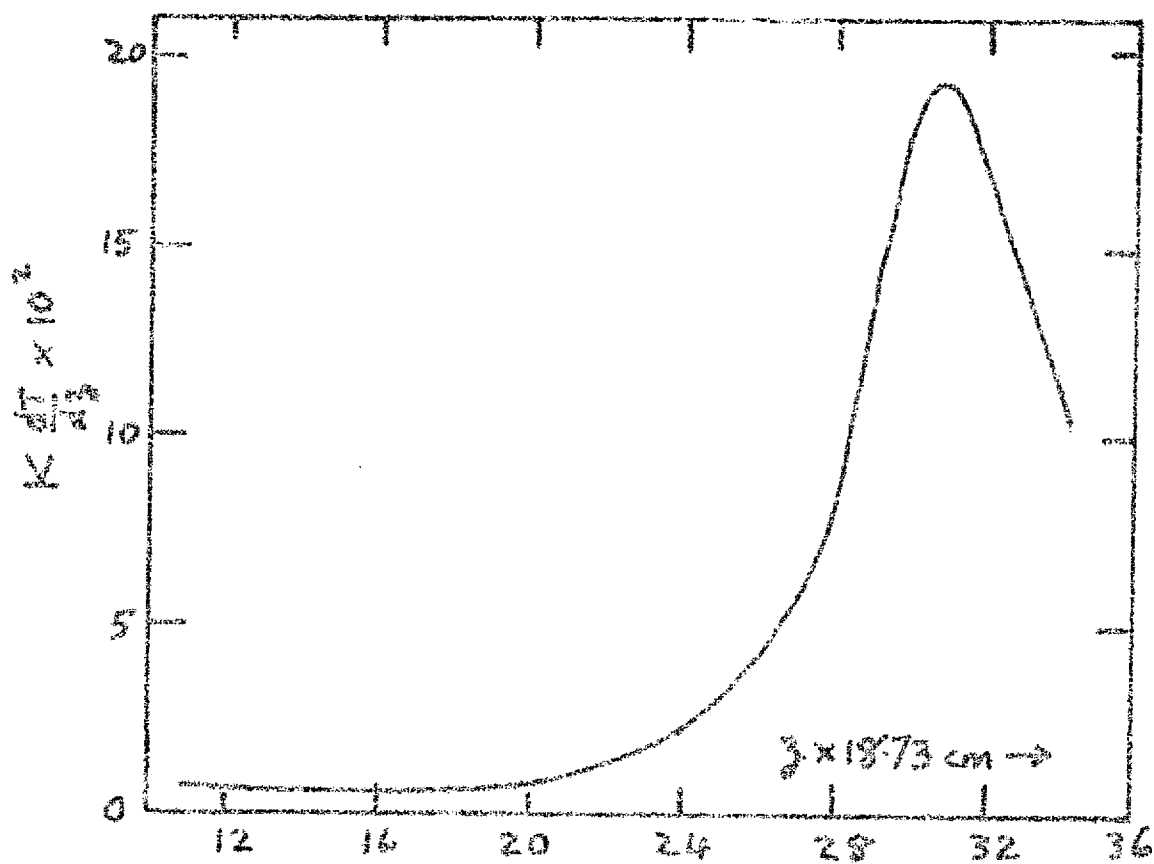


FIG.10.8 (c)

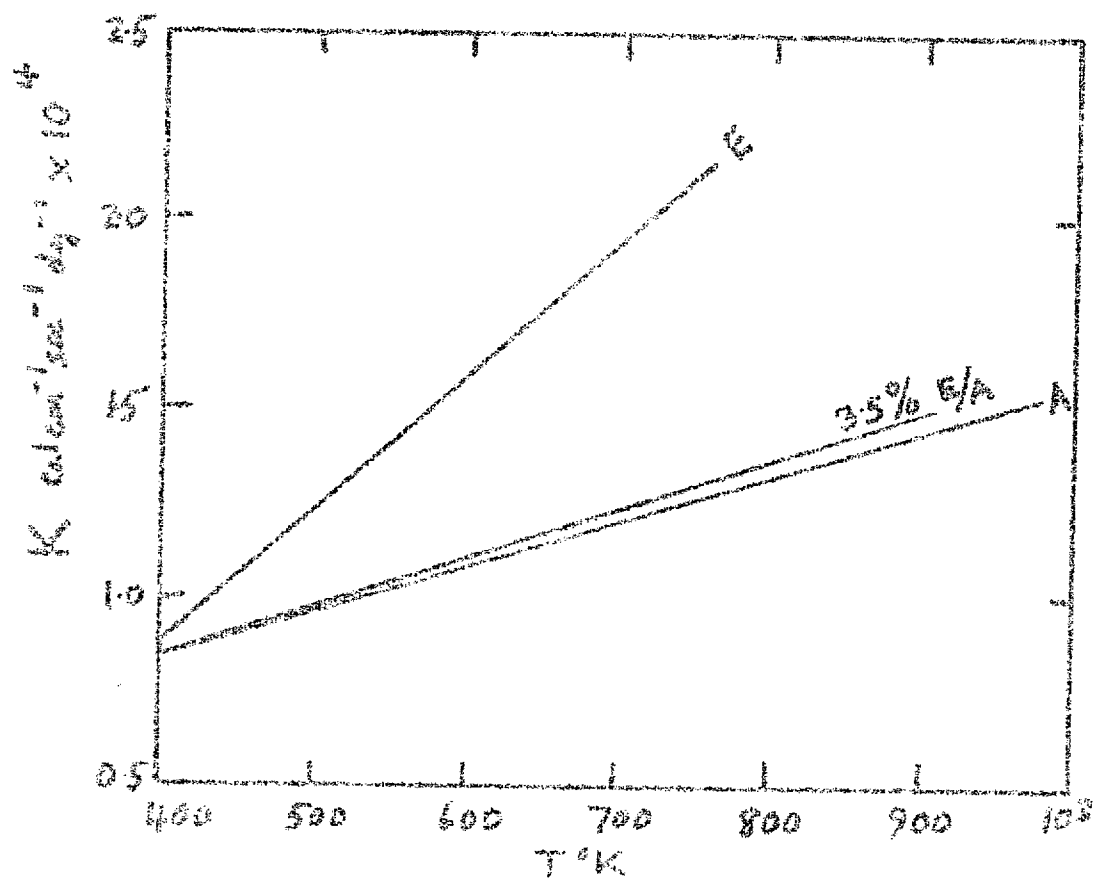
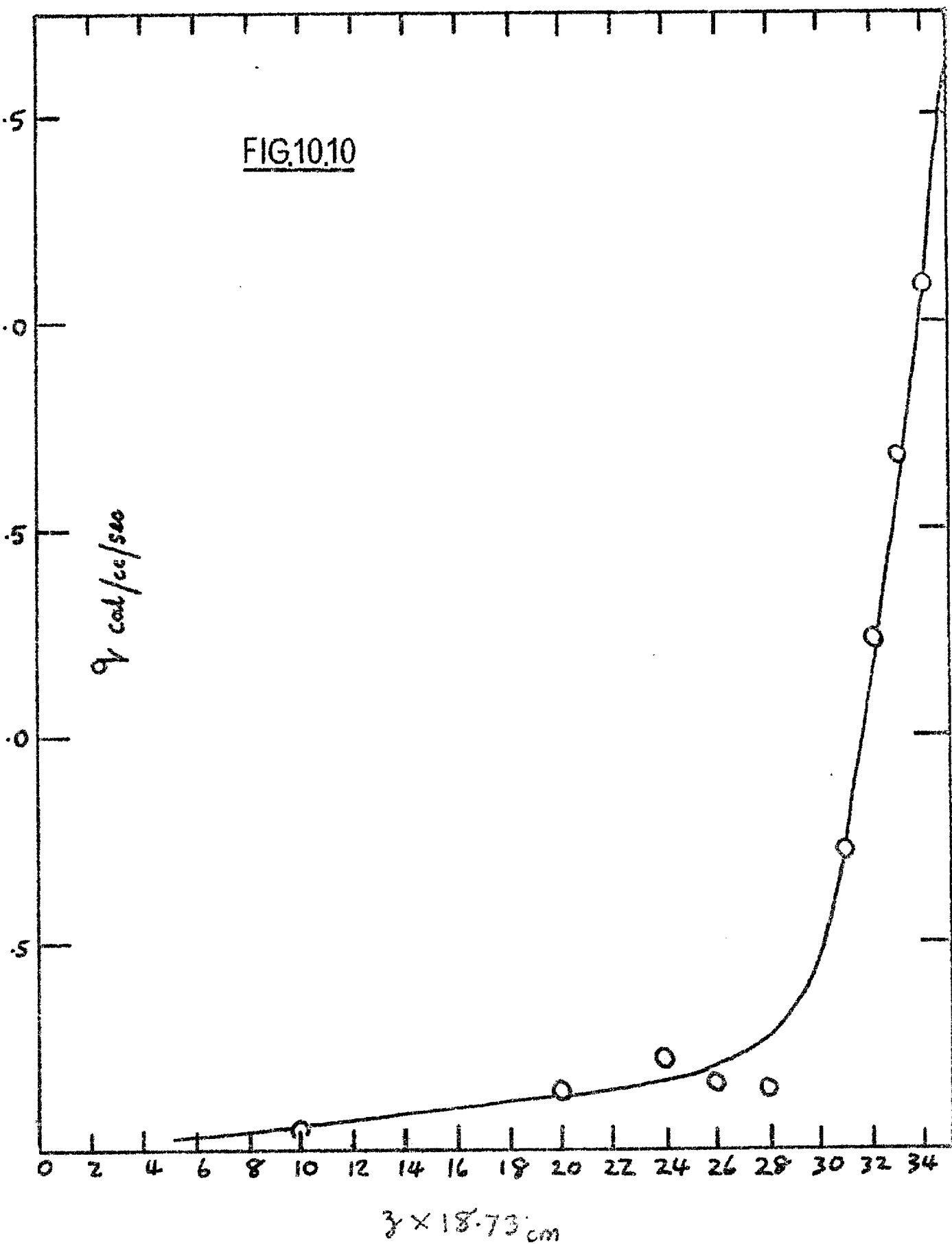


FIG.10.9

FIG.10.10

## 6. Spectroscopic studies.

Work carried out by Thomas et al <sup>79</sup> suggested a possible physical method for investigating the concentration of active intermediates during the critical pre-ignition stages occurring in the present work which might yield information about the chemical kinetics involved. In experiments with cyclic and branched hydrocarbons premixed with air, Thomas et al observed absorption peaks at about 2600 Å just prior to ignition. Similar phenomena had been recorded previously <sup>80,81</sup>. Attempts by Thomas to establish the identity of the species responsible for the strong absorption were unsuccessful, though branching intermediates or peroxy radicals were suggested as possible candidates.

Absorption measurements under flame conditions have been well established <sup>82</sup>. The work of Wolfhard and Parker <sup>83</sup> is particularly interesting, because radical concentrations were determined by absorption spectroscopy for ethylene/oxygen diffusion flames which were stabilised on a Wolfhard-Parker burner.

The experimental arrangement employed in the present work is shown in figure 10.11.. A good background continuum source in the U.V. is required for absorption experiments, and a low pressure hydrogen discharge lamp was used for this purpose. Radiation from the hydrogen

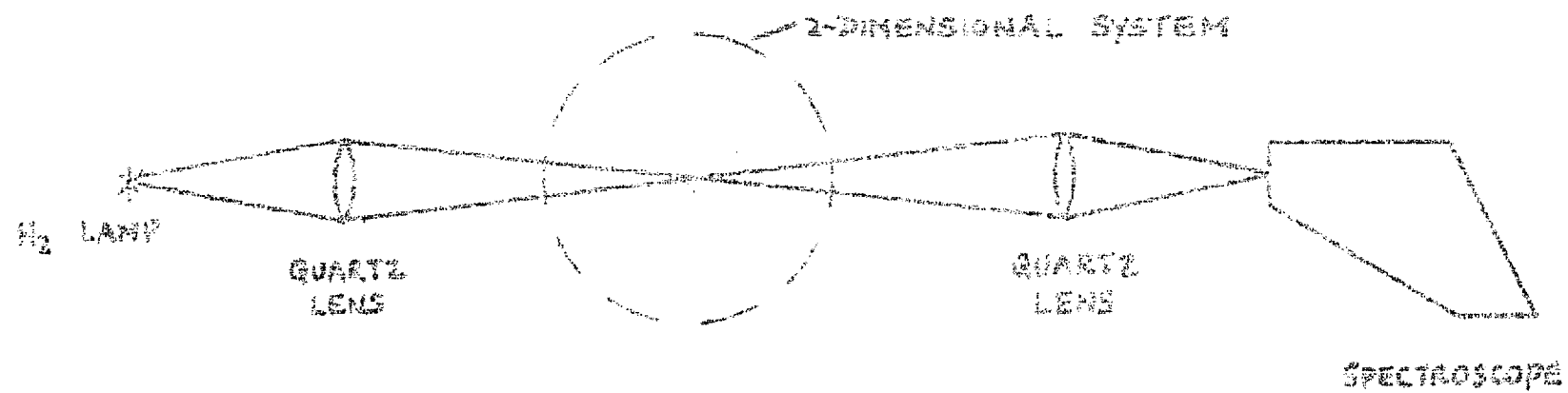


FIG. 10.11

lamp was focused by means of a quartz lens across a diameter of the system, just upstream of the disc surface. The source image at the centre of the gas stream covered the entire region over which appreciable heat release had been observed under the pre-ignition conditions discussed in the preceding chapter. After passing through the test space, the light was focused by a second lens onto the slit of a Hilger Medium Quartz spectroscope. Continuum spectra for a non-reactive gas flow were photographed and compared with those found for various ethylene/air mixtures at the ignition threshold. Under the quasi-steady state conditions of the experiment, exposures of 2 to 3 minutes were used, but careful analysis failed to reveal the presence of absorption. It seemed possible that a test path length of about 6 cm was insufficient for absorption to be detectable on the photographic plate, especially in view of the small ethylene concentrations involved. The optical path length through the test region was therefore increased six-fold by introducing two concave mirrors, which enabled multiple traversals of the test region to be made by the incident light beam. It was appreciated that the reflectivity coefficient for the concave mirrors in the U.V. was low and that, due to multiple reflections, the overall intensity of the traversing beam would be decreased

appreciably. No definite absorption bands were observed for the ethylene/air system, or when traces of liquid paraffin were introduced into the gas stream.

From these experiments it was concluded that no significant absorption occurred, and that the type of peak absorption observed by Thomas et al was probably specific to the group of hydrocarbons studied in (79). Unfortunately, there did not appear to be any alternative method whereby possible intermediate species in the 2-dimensional system could be investigated, which would not seriously disturb the critical flow and temperature conditions preceding ignition.

## CHAPTER 11.

DISCUSSION OF RESULTS AND CONCLUSIONS.1. Some General Observations.

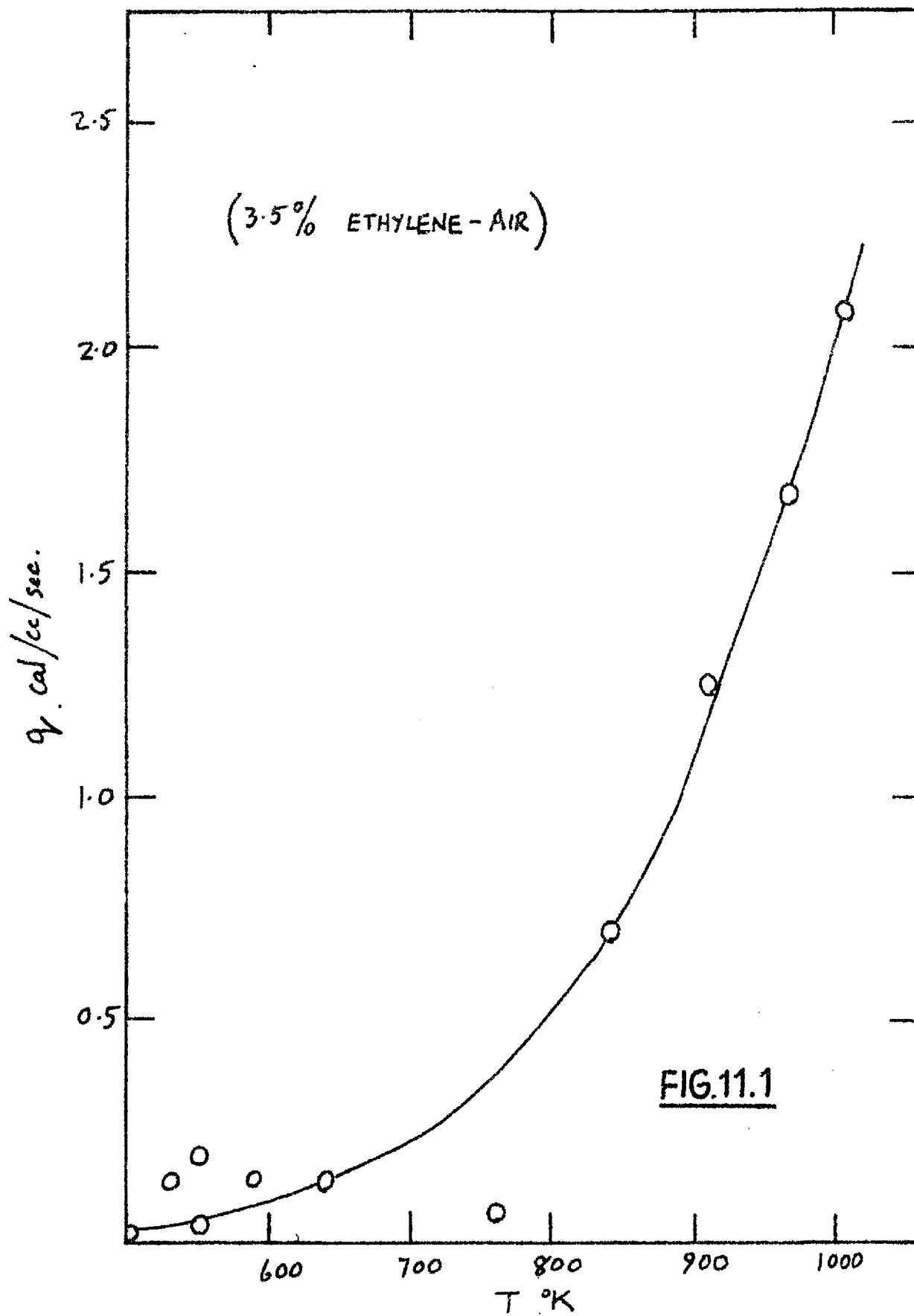
For a given ethylene/air mixture, the dependence of surface temperature on flow stream velocity at the ignition threshold, figure 10.1(a), was found to be slight over the flow range studied. The significance of the small temperature dependence can probably be explained in terms of increased heat loss from the reacting gas. Ignition may be regarded essentially as an imbalance in the fundamental heat conservation equation. Under steady-state conditions, the heat released is normally balanced by a conduction term and a convection term. The conduction term is proportional to  $\nabla^2 T$  and the convection term to  $\nabla T$ , so that, when changes occur as the result of changes in flow, it is an indication of relative changes that occur as between  $\nabla^2 T$  and  $\nabla T$ . It has been shown that  $q$  is an extremely steep function of  $T$ , so even quite a small temperature variation would be expected to produce an appreciable change in the heat release rate. This change is taken up by relative adjustments in  $\nabla^2 T$  and  $\nabla T$ , controlling the conduction and convection contributions to the total heat flux, and thereby re-establishes a steady-state condition.

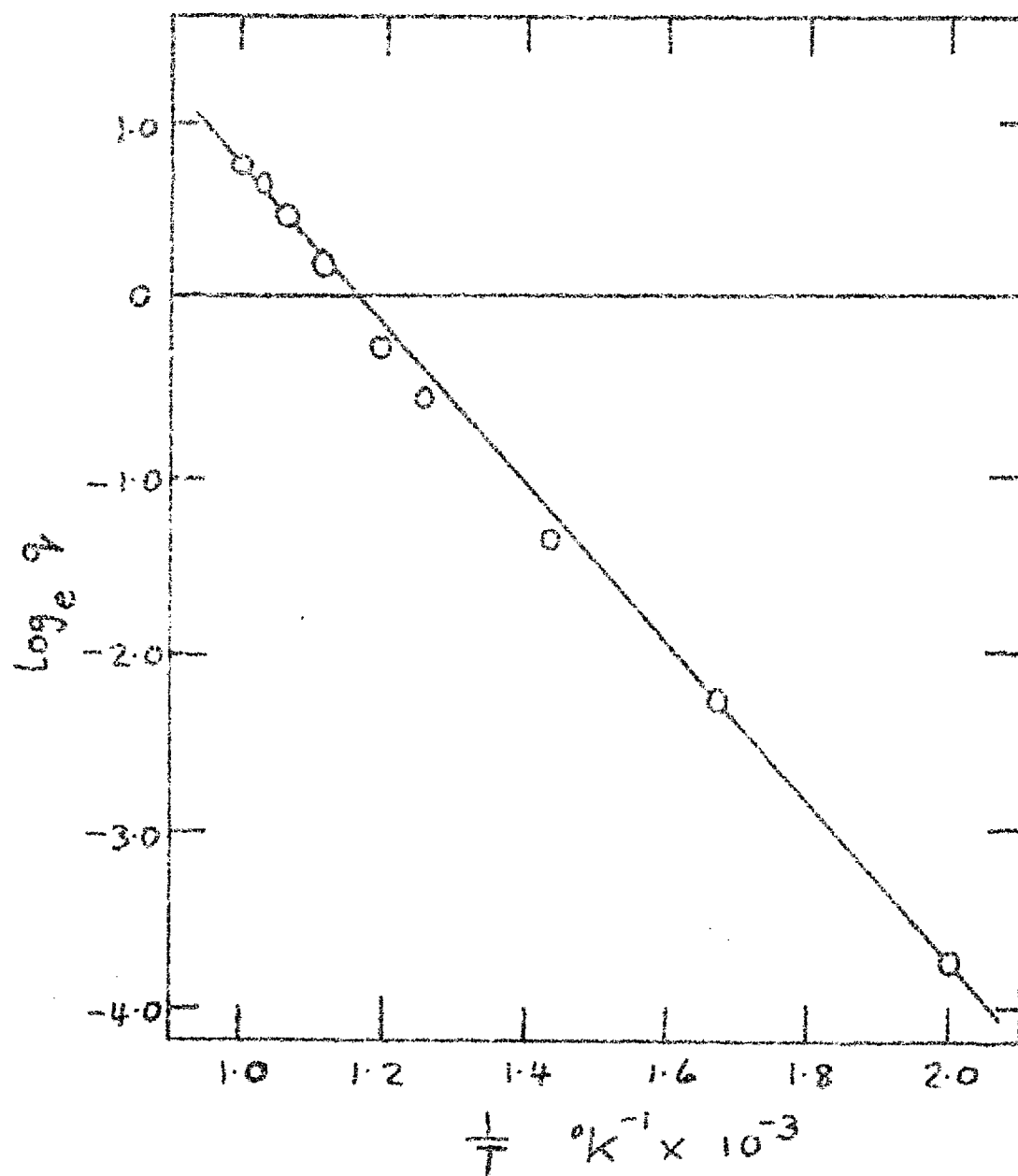
## 2. Deduction of an overall activation energy.

From figure 10.10, it is evident that exothermic reaction is negligible until the gas temperature approaches 600°K, at a distance of about 0.6 cm from the hot surface of the disc. Downstream of this point, the rate of heat release increases rapidly to over 2 cal./cc./sec., 0.05 cm from the disc. The experimental scatter in the lower part of the curve includes errors in differentiating graphically which, however, amount to less than 8% of the local convection terms. Figure 11.1 shows the heat release rate/cc, ( $q$ ), plotted as a function of temperature.

An overall activation energy for the reacting gas mixture was computed from the heat release rates determined, by assuming a single step reaction rate with an Arrhenius type temperature dependence, i.e.  $q = A \exp\left(\frac{-E}{RT}\right)$  - where  $E$  is the activation energy for the single step reaction and  $A$  is a constant. When  $\log_e q$  was plotted against  $\frac{1}{T}$  a straight line was indeed obtained. Its slope gives  $E$ . Figure 11.2 shows the graph obtained using the data in figure 11.1. This gave a value of 8.8 Kcal/mole for the effective activation energy.

It is uncertain how much significance can be attached to the activation energy calculated, because of the basic assumptions made; i.e. that the course of the chemical



FIG.11.2

kinetics involved can be described by a single reaction mechanism. In reality, the situation is probably more complex, with two or more competitive reactions taking place simultaneously. However, it is interesting to compare the activation energy found here with certain well established values <sup>61</sup> for a number of elementary radical reactions met with in flames. A few of these are given below.

|                                    |                                                  |               |
|------------------------------------|--------------------------------------------------|---------------|
| OH + CH <sub>4</sub>               | CH <sub>3</sub> + H <sub>2</sub> O               | 9.9 Kcal/mole |
| O + H <sub>2</sub>                 | OH + H                                           | 9.4 " "       |
| H + C <sub>2</sub> H <sub>6</sub>  | C <sub>3</sub> H <sub>5</sub> + H <sub>2</sub>   | 9.7 " "       |
| H + C <sub>3</sub> H <sub>8</sub>  | C <sub>3</sub> H <sub>7</sub> + H <sub>2</sub>   | 8.2 " "       |
| H + C <sub>2</sub> H <sub>4</sub>  | C <sub>2</sub> H <sub>5</sub>                    | 6.1 " "       |
| OH + C <sub>2</sub> H <sub>4</sub> | C <sub>2</sub> H <sub>3</sub> + H <sub>2</sub> O | 6.5 " "       |

The activation energy found in the present work is of the same order as that for such radical reactions, but only about 1/5<sup>th</sup> of those typifying effective flame propagating reactions. However one would hardly be justified, on the basis of a single overall activation energy value alone, in proposing a reaction mechanism which could not be distinguished from a number of elementary flame reactions. Additional data on the chemical kinetics associated with the present experimental conditions would therefore seem desirable, and in fact it was this which

prompted the absorption spectroscopy studies described in the previous chapters.

### 3. Aerodynamic temperature measurements.

In the work described, pre-ignition conditions in a flowing gas in contact with a heated surface were investigated, using interferometry to measure the gas temperature distributions upstream of the heated surface. The dependence of heat release rates on fuel concentration, as well as any variation with flow stream velocity which might indicate dependence on radical build up, appeared an interesting next step in the development of the present investigation. As will have been apparent, accurate interpretation for a 2-dimensional refractive index field entailed a very lengthy and tedious analysis. For this reason an alternative and less intricate experimental technique for measuring the temperature distribution was sought, which would reduce the amount of work involved in calculating the heat release data. Suitable methods of gas temperature measurement were therefore carefully re-considered.

Gas temperature measurements based on a particle tracer technique have been made in certain flame structure studies<sup>58,59</sup>. They were not very successful in regions of steep temperature gradients found in premixed atmospheric flames. The main reasons for not seriously considering the

method at an earlier stage of the research, in preference to interferometry or deflection mapping, were as follows.

From previous work <sup>58,61,69</sup> it was clear that the experimental errors inherent in particle-track measurements were large enough to make consequent temperature measurements considerably less accurate than could be expected using optical methods. In addition there was uncertainty about the physical and possible chemical effects the presence of solid tracer particles might have on critical pre-ignition conditions. Under the circumstances, it seemed conceivable that a method of temperature measurement less accurate than interferometry could lead to erroneous conclusions as to the feasibility of measuring pre-ignition heat release rates at all.

Theoretically, the principle of mass conservation is applied along a flow tube, which has been defined by means of a particle-track photograph. Figure 11.3(a) shows schematically an elemental flow tube in relation to a 2-dimensional axi-symmetric system.

Applying the principle of mass conservation to the flow tube gives

$$\rho_1 v_1 A_1 = \rho_0 v_0 A_0 = m \quad (11.1)$$

$$\text{i.e. } \rho_1 v_1 r_1 d_1 = \rho_0 v_0 r_0 d_0 \quad (\text{Figure 11})$$

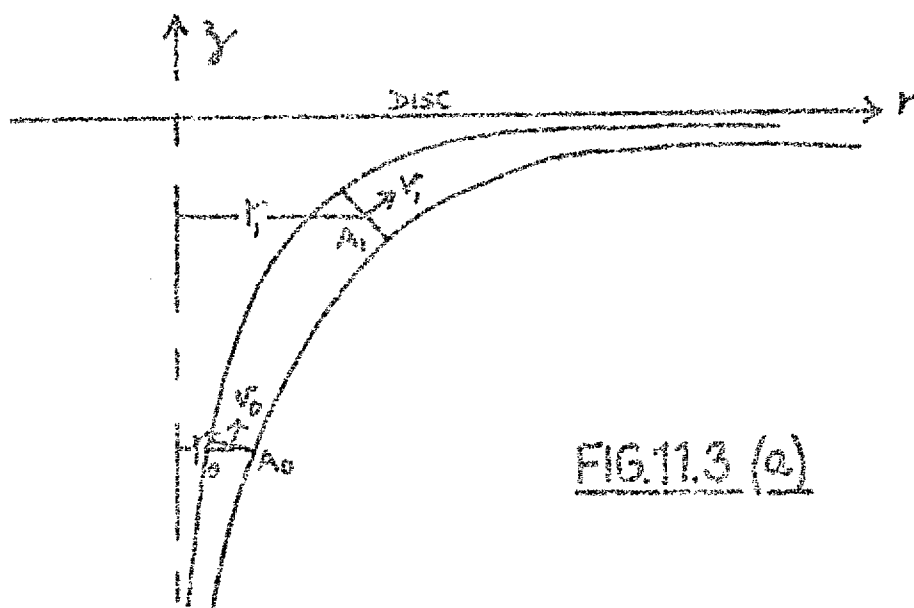


FIG.11.3 (a)

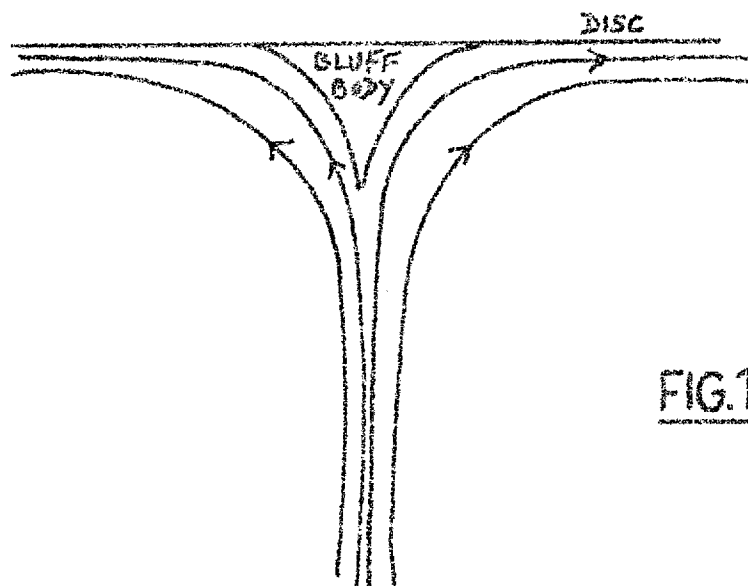


FIG.11.3 (b)

where  $A_1$ ,  $A_0$  are the flow tube areas, and subscript zero refers to an arbitrary reference state.

Using the perfect gas relationship

$$\rho = \frac{P \bar{M}}{R T} \quad (11.2)$$

in the present work, where very small concentrations of ethylene were used and composition changes could be considered negligible prior to ignition, i.e.  $\sum_i X M_i \rightarrow M_{AIR}$  — and combining (11.1) and (11.2), the temperature anywhere along the flow tube is given by :

$$T = \frac{d r v}{d_o r_o v_o} \quad T_o = R_A \frac{v}{v_o} T_o \quad (11.3)$$

where  $R_A$  is the area ratio function.

From equation (11.3) it is clear that the local gas temperature can be found by measuring the gas velocity and flow tube area at that point.

The accuracy of the method depends on the errors in measuring the local flow stream areas and velocities. In flame structure studies, expansion of the flow tubes is sometimes negligible<sup>58</sup> and the chief error in determining the gas temperature depends on the accuracy to which the local gas velocities can be measured. Under the conditions of steep temperature gradients found in certain flames, thermo-mechanical and velocity lag effects become important correction sources, but in the present work these were shown

to be negligible (Appendix 2).

Several particle-track photographs taken with the 2-dimensional system were analysed in the way just described, in order to determine the temperature distribution between the disc surface and lower matrix. Relatively long exposures of 1 - 2 seconds were required for the flow rates used, in order that the flow tubes could be observed over appreciable distances across the test space. Two mercury vapour lamps were used to increase the extent of the Tyndall beam, which was focused across a diameter of the experimental system. A typical particle-track photograph taken under these conditions is shown in Plate 11(a). From the photograph it was apparent that the gas along the central region of the system expanded rapidly on approaching close to the heated disc and flowed radially outwards to the periphery of the disc.

It had been established from previous heat release analysis, Figure 10.10, that appreciable exothermic reaction only set in the vicinity of the disc surface. Particle-track photographs were therefore selected, in which adjacent pairs of tracks followed paths along the central axis of the system. Analysis along the chosen flow tubes yielded impossible profiles, with maximum temperatures several hundred degrees higher than the disc surface temperature. It was necessary to take the matrix temperature as the

reference state, which naturally made the analysis less accurate than if the surface temperature of the disc had been used. This arose due to difficulty in measuring the flow tube areas and local velocities close to the disc surface with any certainty. Lateral expansion of the gas in this region was great and the tracer particles frequently appeared to follow zig-zag paths as the surface of the disc was approached (see plate 11(a)).

From this it was calculated that, for the experimental system used, an aerodynamic method of temperature measurement was not suitable, because the zone in which pre-ignition reaction occurred happened to coincide with a region near the forward stagnation point. In this important region the tracer particles were retarded to very low velocities, which meant that the ratio of error factors, such as thermo-mechanical and free-fall velocity effects, to actual velocities, tended to infinity.

The same situation applied, of course, in the previous work, as regards the velocity distributions measurements. But, as was noted previously, the velocity profiles along the z-axis were always extrapolated to zero over the difficult region just upstream of the heated surface. The errors introduced into the thermal analysis as a result of such extrapolations are minor, because the convective terms in the appropriate heat conservation

equation becomes increasingly smaller than the corresponding conduction and heat release terms, as the surface of the disc is approached. Thus the important parameter appears to be the gas temperature close to the hot surface, and interferometry enabled its distribution to be measured extremely accurately.

Work currently in progress is designed to overcome the difficulties referred to, which are associated with the forward stagnation zone in the 2-dimensional experimental system. When a stagnation point is approached, so that the convective velocity falls to zero, the particle-tracer method is obviously impossible, because the errors remain finite while the quantity measured falls to zero.

It was therefore decided to block-off the forward stagnation zone, by introducing a suitable bluff body at the base of the heated disc. This is indicated schematically in Figure 11.3b. The approximate shape and size of the bluff body required was deduced from particle-track photographs showing the general divergence of flow lines approaching the stagnation point. By blocking off the stagnation zone, the gas stream incident on the heated surface was induced to impinge obliquely onto the outer annulus surrounding the central conic section of the bluff body.

In the modified system, the surface temperature of the outer annulus exceeds that of the bluff body, thus

prohibiting the initiation of ignition near the central region as before. An obvious way of testing the modified system, to see if the aerodynamic method of temperature measurement could now be incorporated, would be to carry out a mass conservation analysis along selected flow tubes under isothermal conditions. An investigation of this kind is currently being undertaken.

#### 4. Conclusions.

The main conclusions of the present research may be summarized as follows.

1. An optical method has been established, whereby critical conditions preceding ignition of a flowing gas in contact with a heated surface can be studied quantitatively.
2. The assumption of a purely thermal model enables pre-ignition heat release rates to be determined for a flowing gas, as a function of temperature.
3. The concept of a series of quasi-steady states prior to ignition onset appears to be valid, judging by the optical records obtained, and permits a time invariant system to be considered for the purpose of calculating the critical heat release rates.
4. Under the present experimental conditions, where the relevant refractive index gradients are mild, interferometry

is ideal for determining the steady-state temperature distributions over the entire test region.

5. Pegging the test interferograms to known temperature boundaries is essential, otherwise very large errors arise in the analysis.

6. Some form of optical pyrometry is convenient for the purpose of determining the temperature distribution across the heated surface, and hence providing the necessary data for pegging the interferograms.

7. The graphical method used for the analysis is arduous and time consuming. A system suitable for programming a computer would, therefore, be desirable.

8. Application of the aerodynamic method of temperature measurement is not practical in the case of the experimental system used, because ignition occurs near the stagnation region, where flow velocities cannot be determined with any accuracy. However, a modified system has since been proposed which would, in principle, be free of such complications as arise in the presence of stagnation zones.

9. A method has been established, even though it is arduous, which enables pre-ignition heat release rates as a function of temperature to be measured very accurately for a flowing, explosive gas in contact with a heated surface.

If, for any particular configuration, the distributions of temperature and flow velocity were known, it would be possible, using the data obtained, to determine whether the temperature at a point in the system would rise or not and therefore whether the conditions in question are hazardous.

## 6. Suggestions for future work.

The difficulties encountered in applying the particle-track method to the 2-dimensional system could be overcome, as has been suggested, by blocking off the forward stagnation zone with a surface at a lower temperature than the remaining flat surface. Thus, by modifying the present system, the gas flow upstream of the disc approaches the heated surface obliquely. This seems the obvious method of minimizing ignition hazards in general, since ignition will occur in zones of low flow velocity and high temperature. The general streamlining of all bluff bodies is bound to increase the tolerance of the system to ignition.

On the other hand, for the purpose of the present work, it may prove more profitable, in practice, to use a system in which the gas flows parallel to a heated surface. The flow regime would then be relatively simple. In addition, numerous theoretical papers <sup>33,34,35</sup> dealing with similar systems could be referred to with advantage.

At the outset of this research it was suggested

that, unlike previous ignition data, which usually relate minimum ignition temperatures of a gas to a specific experimental system, critical heat release rates at the ignition threshold could be employed to establish criteria for systems in which the distribution of temperature is specified. The assumption implied in this proposal is that the heat release rate within the gas is a unique function of temperature. There seems to be no obvious reason why the method described should not be applied to more general systems in which the gas phase reactions are time dependent. This would only require the introduction of relevant time derivative terms into the generalised heat conservation equation (1.2). The question of available light flux through the optical system used may prove an obstacle, especially when studying phenomena which vary rapidly with time. With the advent of powerful laser sources, however, this need not pose a major problem<sup>84,85</sup>.

In addition to a programmed experimental system, it would also be advantageous to use an interferometer which was virtually free from optical defects. This would obviate the need to correct test interferograms for residual fringe components. Pegging at known temperature boundaries would still be desirable for accurate analysis, of course, but in the absence of residual fringe components the pegging procedure is trivial. If a laser source is available, the

problem of residual fringe components in an optical system can, in theory, be eliminated altogether. The method involved requires the use of holography and is dealt with in (86). The long exposure times needed, however, implies that the method would only be suitable for studying steady-state phenomena.

Finally, accurate quantitative measurements of rate constants and activation energies for elementary reactions, whose reaction mechanisms are fairly well established, could be achieved by means of interferometry along the lines described. Experimental kinetic data of this kind is sadly lacking at present, and would be invaluable in reaching a fuller understanding of ignition and related combustion phenomena.

## APPENDIX 1.

DEFLECTION EFFECTS.

A ray of light is deviated from its incident path when passing through a refractive index field. The local radius of curvature (R) along the path of the ray is given by the relationship

$$\frac{1}{R} = - \frac{\text{grad } n \sin \phi}{n} \quad (1)$$

where n is the local refractive index and grad n is the perpendicular gradient to local surfaces of equal n.  $\phi$  is the angle between the incident ray and grad n.

If a simple two-dimensional case is considered, the radius of curvature can be expressed in terms of cartesian co-ordinates x and y as :

$$R = - [1 + (\frac{dy}{dx})^2]^{-3/2} / \frac{d^2y}{dx^2} \quad (2)$$

where x is the direction coordinate of the incident ray and y is parallel to the direction of grad n.

For simplicity a one-dimensional refractive index field will be considered where  $\text{grad } n = \frac{dn}{dy}$ , say and  $\phi = \frac{\pi}{2}$ .

Combining (1) and (2)

$$\frac{d^2y}{dx^2} = - [1 + (\frac{dy}{dx})^2]^{1/2} \frac{1}{n} \frac{dn}{dy} \quad (3)$$

Equation (3) may be simplified by making approximations

(i)  $n \approx 1$ , which in the present work was valid since lean ethylene/air mixtures were employed; (ii)  $(\frac{dy}{dx})^2 \ll 1$ , because very small deviations are involved.

Hence, equation (3) simplifies to

$$\frac{d^2y}{dx^2} = - \frac{dn}{dy} \quad (4)$$

which on integrating gives

$$\left[ \frac{dy}{dx} \right]_1^2 = - \int_1^2 \frac{dn}{dy} dx \quad (5)$$

For an incident ray parallel to the plane  $\frac{dn}{dx} = 0$

$$\left[ \frac{dy}{dx} \right]_1 = 0$$

Applying this boundary condition to equation (5), the deflection,  $\theta$ , suffered by the ray after passing through the refractive index field is given by :

$$\theta = - \int_1^2 \frac{dn}{dy} dx$$

where the integral is performed along the path of the ray. For small deviations, however, it is permissible to integrate along the x-coordinate which defines the path of the incident ray,

$$\text{i.e.} \quad \theta \approx \tan \theta = \frac{dy}{dx} = - \int_0^X \frac{dn}{dy} dx \quad (6)$$

where  $X$  is the distance covered by the ray through the refractive index field.

The deviation  $y-y_0$  experienced by the ray is therefore given by

$$y = -\frac{1}{2} \frac{dn}{dy} x^2 + y_0 \quad (7)$$

In the present work an estimate of the deflection was obtained by considering the steepest refractive index gradients in the direction normal to the hot zone in the 2-dimensional system. For this calculation  $\frac{dn}{dy} \approx 2.2 \times 10^{-4}$  over a maximum distance  $X \approx 5.0$  cm., giving a value for  $z-z_0$  of .003 cm.

Since the minimum distance between the (z)-sections in the analysis of the interferograms was .08 cm, the approximation of zero ray deflection was considered reasonable, and no parabolic corrections were attempted in calculating the optical path lengths for the test rays.

## APPENDIX 2.

SOURCES OF ERROR IN PARTICLE-TRACK ANALYSIS.

Three main sources of error in determining gas velocity measurements using the particle-track method were considered.

1. Aerodynamic drag effect.

When in an accelerating or decelerating field, a particle experiences a force due to viscous drag which can be expressed in terms of Stokes' law.

$$F_{\text{drag}} = 6\pi \bar{\eta} a \Delta v \quad (1)$$

where  $\bar{\eta}$  is the mean viscosity of the gas and  $a$  is the radius of the particle.

Equating the viscous drag to the accelerating force  $m\bar{\ddot{x}}$ , gives :-

$$v = \frac{m \bar{\ddot{x}}}{6\pi \bar{\eta} a} \quad (2)$$

where  $m$  is the particle mass and  $\bar{\ddot{x}}$  is the average acceleration experienced by the particle.

It can be shown that

$$\bar{\ddot{x}} \approx \frac{v_{\infty}^2 - v_0^2}{2L} = \frac{[(T_{\infty}/T_0)^2 - 1]}{2L} v_0^2 \quad (3)$$

where  $v_{\infty}$ ,  $T_{\infty}$ ;  $v_0$ ,  $T_0$  refer to the final and initial states of the particle.  $L$  is the extent of the accelerating field.

Using some typical values ( $T_{\infty} = 10^3 K$ ,  $T_0 = 300^\circ K$ ,  $L = 0.5 \text{ cm.}$ ,  $v_0 = 8 \text{ cm. sec}^{-1}$ ,  $\bar{\eta} = 5 \times 10^{-4}$ ) relevant to the present work, substitutions into equations (3) and (2) gave :-

$v \approx .03 \text{ cm/sec.}$  - which was considered negligible in view of the large experimental scatter in the particle velocities observed.

## 2. Thermo-mechanical effect.

A particle surrounded by gas experiences unequal molecular bombardment over its surface when placed in a region of high temperature gradient. This non-equilibrium effect results in a nett force which acts on the particle in a direction towards the colder gas region. Theoretical treatments of the phenomenon include 27-28%, while a comprehensive experimental analysis has been carried out by Schmidt. It is found that the thermo-mechanical effect falls off sharply as the particle size increases and become large compared with the mean free path. Schmidt has shown that for large particles, greater than  $2-3 \mu$ , errors of less than 4% are introduced under temperature gradient conditions relevant to flames.

The following expressions for the thermo-mechanical effect in terms of velocity corrections have been proposed.

$$v = \left( -17.9 \frac{K_g}{2K_g + K_p} \right) P \frac{L^2}{T} \frac{dT}{dz} \frac{(1 + LA/a)}{6\pi\eta} \quad (87)$$

$$v = -1.5 \left( 1 + \frac{AL}{a} \right) \frac{\rho}{\rho T} \frac{dT}{dz} \left( \frac{K_g}{2K_g + K_p} \right) \quad (88)$$

$$v = \frac{K_g}{P(5 + \frac{\pi a'}{8})} \frac{dT}{dz} \quad (89)$$

where  $L$  is the mean free path length,  
 $P$  is the pressure,  
 $K_p, K_g$  are the particle and gas conductivities,  
 $a'$  is the thermal accommodation coefficient for  
molecular collisions.

For the purpose of the present work the last expression was used to estimate the probable error arising due to the thermo-mechanical effect. The values for  $K_g \approx 15.7 \times 10^{-5}$ ,  $a' \approx 0.75$  and  $\frac{dT}{dz} \approx 9.36 \times 10^2$  gave a value for  $v$  of  $2.8 \times 10^{-4}$  cm/sec. This confirmed what had been anticipated: i.e. that under the relatively mild temperature gradients encountered in the present investigation, errors due to the effect described could be neglected.

### 3. Gravitational effects.

The terminal velocity acquired by a particle of given size can be estimated on the basis of Stokes's law, as used in section (1) which dealt with aerodynamic drag

considerations, by equating the accelerating force due to gravity with the viscous drag on the particle

$$\text{i.e.} \quad v_T = \frac{0.22 (\sigma - \rho) g a^2}{\bar{\eta}}$$

where  $v_T$  is the terminal velocity,

$\sigma$  is the density of the particle,

$\rho$  is the density of the gas.

Substitution for  $\rho$ ,  $g$ ,  $a$  and  $\bar{\eta}$  using values relevant to the present investigation gave maximum terminal velocities of approximately 0.15 cm/sec. and, again, was considered negligible.

$$z = 0; \quad T_{\text{ref}} = 295^{\circ}\text{K.}$$

| $r_i$ | $T_i$ | $\delta_i$ | $(\times 18.73) \left( 60 \sum_1^n x_i - \sum_1^{n-1} x_i \right)$ | P      |
|-------|-------|------------|--------------------------------------------------------------------|--------|
| 56    | 347   | 2.1124     | 5.2044                                                             | 1.0174 |
| 55    | 355   | 2.0648     | 7.0851                                                             | 1.3851 |
| 54    | 363   | 2.0193     | 8.8277                                                             | 1.7258 |
| 53    | 371   | 1.9757     | 10.1776                                                            | 1.98   |
| 52    | 378   | 1.9340     | 12.2736                                                            | 2.40   |
| 51    | 388   | 1.8843     | 14.0670                                                            | 2.75   |
| 50    | 394   | 1.8557     | 15.6955                                                            | 3.07   |
| 49    | 402   | 1.8188     | 17.3447                                                            | 3.39   |
| 48    | 410   | 1.7834     | 18.9930                                                            | 3.71   |
| 47    | 419   | 1.7494     | 20.6189                                                            | 4.03   |
| 46    | 427   | 1.7166     | 22.2440                                                            | 4.34   |
| 45    | 433   | 1.6928     | 23.7662                                                            | 4.65   |
| 44    | 441   | 1.6621     | 25.3199                                                            | 4.95   |
| 43    | 449   | 1.6325     | 26.8482                                                            | 5.25   |
| 42    | 455   | 1.6110     | 28.3546                                                            | 5.54   |
| 41    | 461   | 1.5900     | 29.7164                                                            | 5.80   |
| 40    | 468   | 1.5662     | 31.1173                                                            | 6.08   |
| 39    | 473   | 1.5497     | 32.4647                                                            | 6.35   |
| 38    | 480   | 1.5271     | 33.7855                                                            | 6.60   |
| 37    | 485   | 1.5113     | 35.0605                                                            | 6.85   |
| 36    | 490   | 1.4959     | 36.2728                                                            | 7.09   |
| 35    | 496   | 1.4778     | 37.4889                                                            | 7.33   |
| 34    | 501   | 1.4631     | 38.6545                                                            | 7.55   |
| 33    | 505   | 1.4515     | 39.7591                                                            | 7.77   |
| 32    | 509   | 1.4401     | 40.8185                                                            | 7.98   |
| 30    | 517   | 1.4178     | 42.8729                                                            | 8.38   |
| 27    | 529   | 1.3856     | 45.7294                                                            | 8.94   |
| 24    | 540   | 1.3574     | 48.2461                                                            | 9.43   |
| 21    | 550   | 1.3327     | 50.4959                                                            | 9.87   |
| 18    | 560   | 1.3089     | 52.4072                                                            | 10.24  |

| $r_i$ | $T_i$ | $S_i$  | $S_0 \sum_{j=1}^n x_j - \sum_{j=1}^{n-1} x_j$ | P     |
|-------|-------|--------|-----------------------------------------------|-------|
| 15    | 566   | 1.2950 | 53.9907                                       | 10.55 |
| 12    | 573   | 1.2792 | 55.3119                                       | 10.81 |
| 9     | 580   | 1.2638 | 56.2712                                       | 11.02 |
| 6     | 585   | 1.2530 | 57.1138                                       | 11.16 |
| 3     | 589   | 1.2445 | 57.5914                                       | 11.26 |

| $r_i$ | $x_i$ | $\sum_{i=1}^n \text{AIR}$<br>$\sum_{i=1}^n x_i$ | $z = 3.0$ (x 18.73)<br>9.366P | $\sum_{i=1}^n x_i^2$<br>$\sum_{i=1}^n x_i$ | $\delta_i$ | $T_i$ |
|-------|-------|-------------------------------------------------|-------------------------------|--------------------------------------------|------------|-------|
| 59    | 11.0  | 27.3229                                         | 0.51151                       | 26.8114                                    | 2.4374     | 301   |
| 58    | 10.9  | 38.2521                                         | 1.0230                        | 26.2608                                    | 2.4092     | 304   |
| 57    | 10.9  | 46.4973                                         | 1.5345                        | 25.0056                                    | 2.2941     | 319   |
| 56    | 10.8  | 53.6522                                         | 2.5575                        | 25.4972                                    | 2.3608     | 310   |
| 55    | 10.6  | 59.8620                                         | 3.0691                        | 24.8017                                    | 2.3398     | 312   |
| 54    | 10.5  | 65.0782                                         | 4.6036                        | 23.3841                                    | 2.2271     | 329   |
| 53    | 10.5  | 70.0460                                         | 6.6496                        | 21.6799                                    | 2.0647     | 355   |
| 52    | 10.4  | 74.5170                                         | 8.6967                        | 21.2034                                    | 2.0388     | 359   |
| 51    | 10.3  | 78.7396                                         | 10.2302                       | 20.6548                                    | 2.0053     | 265   |
| 50    | 10.2  | 82.7139                                         | 12.2762                       | 19.5414                                    | 1.9558     | 383   |
| 49    | 10.1  | 86.1913                                         | 14.3223                       | 18.6675                                    | 1.8483     | 397   |
| 48    | 10.0  | 89.6688                                         | 16.3683                       | 17.8952                                    | 1.7895     | 407   |
| 47    | 9.9   | 92.8979                                         | 17.9028                       | 17.6601                                    | 1.7838     | 411   |
| 46    | 9.8   | 95.8785                                         | 19.9489                       | 16.7402                                    | 1.7082     | 429   |
| 45    | 9.6   | 98.8592                                         | 21.9949                       | 15.8539                                    | 1.6514     | 444   |
| 44    | 9.6   | 101.5915                                        | 23.5295                       | 15.8699                                    | 1.6531     | 443   |
| 43    | 9.4   | 104.0754                                        | 25.5755                       | 14.8218                                    | 1.5768     | 465   |
| 42    | 9.1   | 106.8077                                        | 26.5985                       | 14.7028                                    | 1.6377     | 448   |
| 41    | 9.1   | 109.0432                                        | 28.1330                       | 14.4266                                    | 1.5853     | 462   |
| 40    | 8.9   | 111.2787                                        | 29.6676                       | 13.8262                                    | 1.5535     | 472   |
| 39    | 8.9   | 113.5142                                        | 30.6906                       | 14.0859                                    | 1.5827     | 463   |
| 38    | 8.7   | 115.5013                                        | 32.2251                       | 13.2647                                    | 1.5247     | 481   |
| 37    | 8.7   | 117.4885                                        | 33.7597                       | 12.9248                                    | 1.4856     | 493   |
| 36    | 8.6   | 119.4756                                        | 34.7827                       | 12.9671                                    | 1.5078     | 486   |
| 35    | 8.5   | 121.2143                                        | 35.8057                       | 12.8263                                    | 1.5090     | 486   |
| 34    | 8.4   | 122.9530                                        | 36.8387                       | 12.5960                                    | 1.4995     | 489   |
| 33    | 8.2   | 124.6918                                        | 37.8517                       | 12.2241                                    | 1.4907     | 492   |
| 32    | 8.0   | 126.4305                                        | 38.8748                       | 11.8087                                    | 1.4761     | 496   |
| 30    | 11.1  | 129.4112                                        | 40.4093                       | 16.5852                                    | 1.4942     | 491   |
| 27    | 13.1  | 133.3854                                        | 42.9668                       | 19.1158                                    | 1.4592     | 502   |
| 24    | 12.4  | 136.8629                                        | 45.0129                       | 17.9041                                    | 1.4439     | 508   |
| 21    | 11.6  | 140.0920                                        | 46.5474                       | 17.0223                                    | 1.4674     | 499   |
| 18    | 10.9  | 142.5759                                        | 48.0819                       | 15.7546                                    | 1.4454     | 507   |
| 15    | 10.0  | 144.8114                                        | 49.6165                       | 14.0896                                    | 1.4090     | 520   |
| 12    | 9.0   | 146.3017                                        | 50.6395                       | 12.6965                                    | 1.4107     | 520   |
| 9     | 7.9   | 147.7920                                        | 51.8671                       | 11.0131                                    | 1.3941     | 526   |

|       |       | AIR                |         | $z = 15.5 \quad (x \ 18.73)$              |            |       |
|-------|-------|--------------------|---------|-------------------------------------------|------------|-------|
| $r_i$ | $x_i$ | $\sum_{i=1}^n x_i$ | 9.366P  | $\sum_{i=1}^n x_i - \sum_{i=1}^{n-1} x_i$ | $\delta_i$ | $T_i$ |
| 58    | 10.9  | 27.0745            | 0.25575 |                                           | 2.4604     | 298   |
| 57    | 10.9  | 37.755             | 0.81842 | 26.3569                                   | 2.4181     | 303   |
| 56    | 10.8  | 46.2005            | 1.5345  | 25.6610                                   | 2.3760     | 308   |
| 55    | 10.6  | 53.1555            | 2.3018  | 24.8050                                   | 2.3401     | 313   |
| 54    | 10.5  | 59.1168            | 3.0691  | 24.2598                                   | 2.3104     | 317   |
| 53    | 10.3  | 64.5814            | 3.9898  | 23.2614                                   | 2.2584     | 325   |
| 52    | 10.4  | 69.3008            | 4.8593  | 23.4007                                   | 2.2500     | 326   |
| 51    | 10.3  | 73.7718            | 6.0358  | 22.5775                                   | 2.1920     | 334   |
| 50    | 10.2  | 77.7461            | 7.2123  | 21.9168                                   | 2.1487     | 341   |
| 49    | 10.1  | 81.7203            | 8.6957  | 21.0283                                   | 2.0820     | 352   |
| 48    | 10.0  | 85.4462            | 10.4348 | 20.0449                                   | 2.0045     | 366   |
| 47    | 9.9   | 88.6752            | 12.2762 | 19.1291                                   | 1.9322     | 379   |
| 46    | 9.8   | 91.9043            | 13.9642 | 18.4881                                   | 1.8865     | 389   |
| 45    | 9.6   | 94.8850            | 15.6010 | 17.7310                                   | 1.8470     | 397   |
| 44    | 9.6   | 97.8657            | 17.2379 | 17.3510                                   | 1.8074     | 406   |
| 43    | 9.4   | 100.5979           | 18.7724 | 16.6948                                   | 1.7760     | 413   |
| 42    | 9.1   | 103.0818           | 19.9489 | 16.2093                                   | 1.7812     | 411   |
| 41    | 9.1   | 105.5657           | 21.4322 | 15.7340                                   | 1.7290     | 424   |
| 40    | 8.9   | 108.0496           | 22.5064 | 15.5663                                   | 1.7490     | 419   |
| 39    | 8.9   | 110.2852           | 23.8364 | 15.2353                                   | 1.7118     | 428   |
| 38    | 8.7   | 112.2723           | 25.0640 | 14.7008                                   | 1.6897     | 434   |
| 37    | 8.7   | 114.5078           | 26.1382 | 14.6626                                   | 1.6854     | 435   |
| 36    | 8.6   | 116.2465           | 27.3658 | 14.2268                                   | 1.6543     | 443   |
| 35    | 8.5   | 118.2336           | 28.4911 | 13.8802                                   | 1.6330     | 451   |
| 34    | 8.4   | 119.9724           | 29.5653 | 13.6636                                   | 1.6266     | 451   |
| 33    | 8.2   | 121.7111           | 30.5371 | 13.2839                                   | 1.6200     | 452   |
| 32    | 8.0   | 123.4498           | 31.6113 | 12.7425                                   | 1.5540     | 472   |
| 30    | 11.1  | 126.4305           | 33.5039 | 17.5974                                   | 1.5853     | 462   |
| 27    | 13.1  | 130.6531           | 36.0614 | 20.3713                                   | 1.5550     | 471   |

| $r_i$ | $x_i$ | AIR $z = 15.5$ ( $x = 18.73$ ) |                    |                      |                      | $T_i$ |
|-------|-------|--------------------------------|--------------------|----------------------|----------------------|-------|
|       |       | 9.366P                         | $\sum_{i=1}^n x_i$ | $\sum_{i=1}^n x_i^2$ | $\sum_{i=1}^n x_i^3$ |       |
| 24    | 12.4  | 134.1306                       | 38.3121            | 18.9617              | 1.5292               | 479   |
| 21    | 11.6  | 137.3597                       | 40.3070            | 17.4468              | 1.5040               | 487   |
| 18    | 10.9  | 139.8436                       | 41.944             | 16.2802              | 1.4936               | 491   |
| 15    | 10.0  | 142.0791                       | 43.4783            | 14.6550              | 1.4655               | 500   |
| 12    | 9.0   | 143.8178                       | 44.8083            | 12.9265              | 1.4363               | 510   |
| 9     | 7.9   | 145.3081                       | 45.8313            | 11.2545              | 1.4246               | 514   |
| 6     | 6.7   | 146.0533                       | 46.5474            | 9.4354               | 1.4083               | 520   |

| $r_i$ | $x_i$ | AIR                |         | $z = 25.0$ (x 18.73) |                                       | $T_i$ |
|-------|-------|--------------------|---------|----------------------|---------------------------------------|-------|
|       |       | $\sum_{i=0}^n x_i$ | $x_i$   | 9.366P               | $\sum_{i=0}^n x_i - \sum_{i=0}^n x_i$ |       |
| 61    | 11.0  | 27.3229            |         | 0.5115               | 26.8114                               | 301   |
| 60    | 11.1  | 38.7488            |         | 2.5575               | 25.2230                               | 322   |
| 59    | 11.0  | 47.4425            |         | 3.5806               | 24.8620                               | 324   |
| 58    | 10.9  | 54.3974            |         | 4.8593               | 24.1020                               | 331   |
| 57    | 10.9  | 60.6072            |         | 6.1381               | 23.6701                               | 337   |
| 56    | 10.8  | 66.0717            |         | 7.4680               | 22.9917                               | 344   |
| 55    | 10.6  | 71.2879            |         | 8.9514               | 22.0258                               | 353   |
| 54    | 10.5  | 75.7589            | 10.5371 |                      | 21.2469                               | 362   |
| 53    | 10.3  | 80.2300            | 12.1228 |                      | 20.3973                               | 370   |
| 52    | 10.4  | 83.9558            | 13.8108 |                      | 20.0610                               | 380   |
| 51    | 10.3  | 87.6817            | 15.3453 |                      | 19.5594                               | 386   |
| 50    | 10.2  | 91.1591            | 16.9310 |                      | 18.9578                               | 394   |
| 49    | 10.1  | 94.6366            | 18.4144 |                      | 18.5214                               | 400   |
| 48    | 10.0  | 97.6173            | 20.2046 |                      | 17.7376                               | 413   |
| 47    | 9.9   | 100.5977           | 21.7392 |                      | 17.3342                               | 419   |
| 46    | 9.8   | 103.3302           | 23.2737 |                      | 16.8741                               | 426   |
| 45    | 9.6   | 106.0625           | 24.8082 |                      | 16.2392                               | 433   |
| 44    | 9.6   | 108.7948           | 26.1893 |                      | 16.1160                               | 437   |
| 43    | 9.4   | 111.0303           | 27.6215 |                      | 15.5048                               | 444   |
| 42    | 9.1   | 113.5142           | 29.1561 |                      | 14.6425                               | 455   |
| 41    | 9.1   | 115.7497           | 30.4348 |                      | 14.6140                               | 456   |
| 40    | 8.9   | 117.9852           | 31.7136 |                      | 14.1426                               | 461   |
| 39    | 8.9   | 119.9724           | 33.0947 |                      | 13.8669                               | 470   |
| 38    | 8.7   | 121.9595           | 34.3223 |                      | 13.4380                               | 475   |
| 37    | 8.7   | 123.6982           | 35.4476 |                      | 13.3940                               | 476   |
| 36    | 8.6   | 125.6853           | 36.7776 |                      | 12.9241                               | 488   |
| 35    | 8.5   | 127.4241           | 37.8517 |                      | 12.7693                               | 489   |
| 34    | 8.4   | 128.9144           | 38.8748 |                      | 12.5655                               | 490   |
| 33    | 8.2   | 130.6531           | 39.9489 |                      | 12.1169                               | 496   |
| 32    | 8.0   | 132.1435           | 40.9208 |                      | 11.7303                               | 498   |

| $r_i$ | $x_i$ | AIR            | $z = 25.0 \quad (x \ 18.73)$ |                                   |              |       |
|-------|-------|----------------|------------------------------|-----------------------------------|--------------|-------|
|       |       | $\sum_0^N x_i$ | 9.366P                       | $\sum_0^N y_i - \sum_0^{N-1} x_i$ | $\epsilon_i$ | $T_i$ |
| 30    | 11.1  | 135.1242       | 43.0180                      | 15.9415                           | 1.4362       | 510   |
| 27    | 13.1  | 138.8500       | 45.8824                      | 18.3398                           | 1.4000       | 524   |
| 24    | 12.4  | 142.3275       | 48.3377                      | 17.0274                           | 1.3732       | 534   |
| 21    | 11.6  | 145.3081       | 50.6395                      | 15.5371                           | 1.3394       | 549   |
| 18    | 10.9  | 147.7920       | 52.6855                      | 14.2450                           | 1.3069       | 561   |
| 15    | 10.0  | 149.7792       | 54.2201                      | 12.9902                           | 1.2990       | 564   |
| 12    | 9.0   | 151.5179       | 55.7546                      | 11.3425                           | 1.2603       | 582   |
| 9     | 7.9   | 152.7598       | 56.7776                      | 9.9461                            | 1.2590       | 582   |
| 6     | 6.7   | 153.7534       | 58.3121                      | 8.4804                            | 1.2657       | 579   |

| $r_i$ | $x_i$ | AIR            |         | $z = 29.0$ (x 18.73)                  |            | $T_i$ |
|-------|-------|----------------|---------|---------------------------------------|------------|-------|
|       |       | $\sum_0^i x_i$ | 9.366P  | $\sum_0^i x_i^2 - \sum_0^{i-1} x_i^2$ | $\delta_i$ |       |
| 63    | 11.3  | 28.0681        | 1.0742  | 26.9939                               | 2.3888     | 307   |
| 62    | 11.1  | 39.4940        | 2.6087  | 25.4191                               | 2.2900     | 320   |
| 61    | 11.0  | 48.1877        | 4.3478  | 24.2383                               | 2.2035     | 333   |
| 60    | 11.1  | 55.6394        | 6.1381  | 23.6974                               | 2.1349     | 343   |
| 59    | 11.0  | 61.8491        | 8.1842  | 22.5667                               | 2.0515     | 357   |
| 58    | 10.9  | 67.3137        | 10.2302 | 21.6142                               | 1.9829     | 360   |
| 57    | 10.9  | 72.5299        | 12.5320 | 20.7330                               | 1.9021     | 385   |
| 56    | 10.8  | 77.2493        | 14.6803 | 19.9419                               | 1.8465     | 397   |
| 55    | 10.6  | 81.4719        | 16.7775 | 19.0638                               | 1.7985     | 408   |
| 54    | 10.5  | 85.4462        | 18.9259 | 18.2866                               | 1.7416     | 421   |
| 53    | 10.3  | 89.1720        | 20.9719 | 17.5345                               | 1.7024     | 431   |
| 52    | 10.4  | 92.8979        | 23.3248 | 16.9698                               | 1.6317     | 449   |
| 51    | 10.3  | 96.3753        | 25.4732 | 16.3988                               | 1.5921     | 460   |
| 50    | 10.2  | 99.3560        | 27.5704 | 15.8130                               | 1.5503     | 473   |
| 49    | 10.1  | 102.5851       | 29.6676 | 15.2643                               | 1.5113     | 485   |
| 48    | 10.0  | 105.3174       | 31.7136 | 14.7365                               | 1.4736     | 487   |
| 47    | 9.9   | 108.0496       | 33.7597 | 14.2006                               | 1.4344     | 511   |
| 46    | 9.8   | 110.7819       | 35.8057 | 13.6670                               | 1.3946     | 520   |
| 45    | 9.6   | 113.2658       | 37.8006 | 13.0706                               | 1.3615     | 538   |
| 44    | 9.6   | 115.7497       | 39.3863 | 13.1471                               | 1.3995     | 535   |
| 43    | 9.4   | 117.9852       | 41.1765 | 12.5155                               | 1.3314     | 550   |
| 42    | 9.1   | 120.2208       | 42.7108 | 12.0834                               | 1.3278     | 552   |
| 41    | 9.1   | 122.2079       | 44.3479 | 11.8578                               | 1.3030     | 562   |
| 40    | 8.9   | 124.4434       | 46.0359 | 11.3198                               | 1.2719     | 576   |
| 39    | 8.9   | 126.4305       | 47.4704 | 11.2048                               | 1.2590     | 582   |
| 38    | 8.7   | 128.1692       | 48.8492 | 10.9853                               | 1.2627     | 580   |
| 37    | 8.7   | 129.9080       | 50.1791 | 10.8930                               | 1.2521     | 585   |
| 36    | 8.6   | 131.6467       | 51.6114 | 10.5278                               | 1.2242     | 599   |
| 35    | 8.5   | 133.3854       | 52.6855 | 10.2968                               | 1.2114     | 605   |
| 34    | 8.4   | 134.8758       | 54.2201 | 10.0547                               | 1.1970     | 612   |

| $r_i$ | $x_i$ | AIR                    |         | $z = 29.0 \quad (x = 18.73)$                  |        | $T_i$ |
|-------|-------|------------------------|---------|-----------------------------------------------|--------|-------|
|       |       | $S_0 \sum_{i=1}^n x_i$ | 9.366P  | $S_0 \sum_{i=1}^n x_i - \sum_{i=1}^n S_i x_i$ | $S_i$  |       |
| 33    | 8.2   | 136.6145               | 55.2431 | 9.9439                                        | 1.2127 | 604   |
| 32    | 8.0   | 138.1048               | 56.7776 | 9.1800                                        | 1.1475 | 639   |
| 30    | 11.1  | 140.8371               | 58.8236 | 12.9302                                       | 1.1649 | 629   |
| 27    | 13.1  | 144.5630               | 61.8927 | 14.8857                                       | 1.1363 | 645   |
| 24    | 12.4  | 147.7920               | 64.9618 | 13.4186                                       | 1.0821 | 677   |
| 21    | 11.6  | 150.5243               | 67.2636 | 12.5383                                       | 1.0809 | 678   |
| 18    | 10.9  | 153.0082               | 69.5654 | 11.3213                                       | 1.0386 | 706   |
| 15    | 10.0  | 154.9954               | 71.0999 | 10.5245                                       | 1.0524 | 696   |
| 12    | 9.0   | 156.4857               | 72.6344 | 9.1882                                        | 1.0209 | 718   |
| 9     | 7.9   | 157.7276               | 73.7597 | 8.0067                                        | 1.0135 | 723   |
| 6     | 6.7   | 158.7212               | 74.6805 | 6.6153                                        | 0.9874 | 742   |

| $r_i$ | $x_i$ | AIR                |         | $z = 30.5 \quad (x \ 18.73)$                   |            |       |
|-------|-------|--------------------|---------|------------------------------------------------|------------|-------|
|       |       | $S_0 \sum_i^n x_i$ | 9.366P  | $S_0 \sum_i^n x_i - \sum_i^{n-1} \delta_i x_i$ | $\delta_i$ | $T_i$ |
| 64    | 11.3  | 28.0681            | 0.9207  | 27.1474                                        | 2.4024     | 305   |
| 63    | 11.3  | 39.7424            | 2.0460  | 26.2727                                        | 2.3250     | 315   |
| 62    | 11.1  | 48.4360            | 3.9898  | 24.5976                                        | 2.2160     | 331   |
| 61    | 11.0  | 55.8877            | 6.1381  | 23.1221                                        | 2.1020     | 349   |
| 60    | 11.1  | 62.0975            | 8.6957  | 22.2790                                        | 2.0071     | 365   |
| 59    | 11.0  | 67.8105            | 10.9975 | 21.2901                                        | 1.9355     | 379   |
| 58    | 10.9  | 73.0267            | 13.5550 | 20.1100                                        | 1.8449     | 397   |
| 57    | 10.9  | 77.7461            | 16.3683 | 19.0578                                        | 1.7884     | 419   |
| 56    | 10.8  | 82.2171            | 18.6701 | 18.4949                                        | 1.7125     | 428   |
| 55    | 10.6  | 86.1913            | 21.3811 | 17.2643                                        | 1.6287     | 450   |
| 54    | 10.5  | 89.9172            | 23.6829 | 16.7213                                        | 1.5925     | 460   |
| 53    | 10.3  | 93.6430            | 26.2405 | 15.7351                                        | 1.5277     | 480   |
| 52    | 10.4  | 97.1205            | 28.6446 | 15.4415                                        | 1.4848     | 494   |
| 51    | 10.3  | 100.3496           | 31.4579 | 14.4062                                        | 1.3987     | 524   |
| 50    | 10.2  | 103.3302           | 33.9643 | 13.7947                                        | 1.3524     | 542   |
| 49    | 10.1  | 106.3109           | 36.3172 | 13.3617                                        | 1.3229     | 554   |
| 48    | 10.0  | 109.0432           | 28.8748 | 12.6074                                        | 1.2607     | 581   |
| 47    | 9.9   | 111.7755           | 40.9208 | 12.4454                                        | 1.2571     | 583   |
| 46    | 9.8   | 114.2594           | 43.3760 | 11.6841                                        | 1.1923     | 615   |
| 45    | 9.6   | 116.7433           | 45.4732 | 11.3037                                        | 1.1775     | 622   |
| 44    | 9.6   | 118.9788           | 47.3147 | 11.3039                                        | 1.1775     | 622   |
| 43    | 9.4   | 121.2143           | 49.1050 | 10.9193                                        | 1.1616     | 631   |
| 42    | 9.1   | 123.4498           | 50.8441 | 10.4461                                        | 1.1479     | 639   |
| 41    | 9.1   | 125.4369           | 52.6344 | 10.2317                                        | 1.1244     | 652   |
| 40    | 8.9   | 127.4241           | 54.2201 | 9.9862                                         | 1.1220     | 653   |
| 39    | 8.9   | 129.4112           | 55.7546 | 9.8098                                         | 1.1022     | 665   |
| 38    | 8.7   | 131.1499           | 57.2891 | 9.5417                                         | 1.0967     | 668   |
| 37    | 8.7   | 132.8886           | 58.5679 | 9.6202                                         | 1.1057     | 663   |
| 36    | 8.6   | 134.6274           | 59.8467 | 9.4733                                         | 1.1015     | 665   |

| $r_i$ | $x_i$ | AIR                |          | $z = 30.5 \quad (x \ 18.73)$                       |            | $T_i$ |
|-------|-------|--------------------|----------|----------------------------------------------------|------------|-------|
|       |       | $\sum_{i=1}^n x_i$ | $9.366P$ | $\sum_{i=1}^n x_i - \sum_{i=1}^{n-1} \delta_i x_i$ | $\delta_i$ |       |
| 35    | 8.5   | 136.3661           | 61.3300  | 9.0796                                             | 1.0682     | 686   |
| 34    | 8.4   | 137.8564           | 62.4042  | 9.1235                                             | 1.0861     | 675   |
| 33    | 8.2   | 139.3468           | 63.8876  | 8.5249                                             | 1.0396     | 705   |
| 32    | 8.0   | 140.8371           | 65.0129  | 8.4033                                             | 1.0504     | 698   |
| 30    | 11.1  | 143.5694           | 67.4682  | 11.2876                                            | 1.0169     | 721   |
| 27    | 13.1  | 147.2953           | 70.9664  | 12.7851                                            | 0.9760     | 751   |
| 24    | 12.4  | 150.5243           | 73.4528  | 12.2335                                            | 0.9866     | 743   |
| 21    | 11.6  | 153.2566           | 75.7035  | 11.3907                                            | 0.9819     | 746   |
| 18    | 10.9  | 155.4921           | 78.0053  | 10.2154                                            | 0.9372     | 782   |
| 15    | 10.0  | 157.4793           | 79.7956  | 9.2590                                             | 0.9259     | 792   |
| 12    | 9.0   | 158.9636           | 81.3301  | 8.1256                                             | 0.9039     | 811   |
| 9     | 7.9   | 160.2115           | 82.4043  | 7.1806                                             | 0.9089     | 806   |

ALL

z = 32.0 (x 18.73)

| $r_i$ | $x_i$ | $\sum_0^n x_i$ | 9.366P  | $\sum_0^n x_i - \sum_1^{n-1} x_i$ | $\delta_i$ | $T_i$ |
|-------|-------|----------------|---------|-----------------------------------|------------|-------|
| 64    | 11.3  | 28.0681        | 1.7903  | 26.2778                           | 2.3255     | 315   |
| 63    | 11.3  | 39.7424        | 3.5806  | 25.2320                           | 2.2329     | 328   |
| 62    | 11.1  | 48.4360        | 5.6778  | 23.6685                           | 2.1223     | 344   |
| 61    | 11.0  | 55.8877        | 8.1842  | 22.2110                           | 2.0192     | 363   |
| 60    | 11.1  | 62.0975        | 10.9975 | 21.1457                           | 1.9050     | 385   |
| 59    | 11.0  | 67.8105        | 13.9642 | 19.7745                           | 1.7977     | 408   |
| 58    | 10.9  | 73.0267        | 17.0333 | 18.4907                           | 1.6964     | 432   |
| 57    | 10.9  | 77.7461        | 20.3069 | 17.3582                           | 1.5925     | 460   |
| 56    | 10.8  | 82.2171        | 23.0179 | 16.7645                           | 1.5523     | 472   |
| 55    | 10.6  | 86.1913        | 26.5985 | 15.0319                           | 1.4181     | 517   |
| 54    | 10.5  | 89.9172        | 29.6676 | 14.2516                           | 1.3573     | 540   |
| 53    | 10.3  | 93.6430        | 32.7366 | 13.3549                           | 1.2966     | 565   |
| 52    | 10.4  | 97.1205        | 35.7034 | 12.9351                           | 1.2437     | 589   |
| 51    | 10.3  | 100.3496       | 38.8748 | 12.0222                           | 1.1672     | 628   |
| 50    | 10.2  | 103.3302       | 41.9438 | 11.2372                           | 1.1017     | 665   |
| 49    | 10.1  | 106.3109       | 44.5014 | 11.0483                           | 1.0939     | 670   |
| 48    | 10.0  | 109.0432       | 47.0589 | 10.6107                           | 1.0611     | 691   |
| 47    | 9.9   | 111.7755       | 49.6165 | 10.1302                           | 1.0232     | 716   |
| 46    | 9.8   | 114.2594       | 51.6625 | 10.1222                           | 1.0328     | 710   |
| 45    | 9.6   | 116.7433       | 53.7085 | 9.8291                            | 1.0239     | 716   |
| 44    | 9.6   | 118.9788       | 55.7034 | 9.7051                            | 1.0109     | 725   |
| 43    | 9.4   | 121.2143       | 57.4337 | 9.4484                            | 1.0051     | 729   |
| 42    | 9.1   | 123.4498       | 59.3352 | 8.9957                            | 0.9885     | 741   |
| 41    | 9.1   | 125.4369       | 61.1254 | 8.8699                            | 0.9747     | 752   |
| 40    | 8.9   | 127.4241       | 62.7111 | 8.7086                            | 0.9786     | 749   |
| 39    | 8.9   | 129.4112       | 64.5479 | 8.5827                            | 0.9643     | 760   |
| 38    | 8.7   | 131.1499       | 65.7290 | 8.4154                            | 0.9673     | 758   |
| 37    | 8.7   | 132.8886       | 67.2636 | 8.2541                            | 0.9487     | 773   |
| 36    | 8.6   | 134.6274       | 68.5935 | 8.1999                            | 0.9535     | 769   |
| 35    | 8.5   | 136.3661       | 70.0769 | 7.8905                            | 0.9283     | 789   |

|       |       | S.M.           |         | $z = 32.0 \quad (x \ 18.73)$      |            |       |
|-------|-------|----------------|---------|-----------------------------------|------------|-------|
| $r_i$ | $x_i$ | $\sum_0^i x_i$ | 9.366P  | $\sum_0^i x_i - \sum_0^{n-1} x_i$ | $\delta_i$ | $T_i$ |
| 34    | 8.4   | 137.8554       | 71.3556 | 7.7923                            | 0.9276     | 790   |
| 33    | 8.2   | 139.5468       | 72.7567 | 7.4355                            | 0.9068     | 808   |
| 32    | 8.0   | 140.8371       | 74.1639 | 7.0602                            | 0.8825     | 830   |
| 30    | 11.1  | 143.5694       | 76.3634 | 9.9398                            | 0.8955     | 818   |
| 27    | 13.1  | 147.2953       | 79.7956 | 11.2547                           | 0.8591     | 853   |
| 24    | 12.4  | 150.5245       | 82.3531 | 10.7677                           | 0.8684     | 844   |
| 21    | 11.6  | 153.2566       | 85.0130 | 9.8625                            | 0.8502     | 869   |
| 18    | 10.9  | 155.4921       | 86.9567 | 9.0495                            | 0.8302     | 883   |
| 15    | 10.0  | 157.4793       | 89.0027 | 8.1440                            | 0.8144     | 900   |
| 12    | 9.0   | 158.9696       | 90.5373 | 7.2099                            | 0.8011     | 915   |

| $r_i$ | $x_i$ | ATR                |          | $z = 33.5$ ( $x = 18.73$ )               |        | $T_i$ |
|-------|-------|--------------------|----------|------------------------------------------|--------|-------|
|       |       | $\sum_{j=0}^i x_j$ | $9.366P$ | $\sum_{j=0}^i x_j - \sum_{j=0}^i 9.366P$ | $S_i$  |       |
| 64    | 11.3  | 28.0681            | 3.0691   | 24.9990                                  | 2.2123 | 331   |
| 63    | 11.3  | 39.7424            | 5.1151   | 24.2295                                  | 2.1442 | 342   |
| 62    | 11.1  | 48.4360            | 8.1842   | 21.9953                                  | 1.9815 | 370   |
| 61    | 11.0  | 55.8877            | 10.7417  | 21.0414                                  | 1.9128 | 383   |
| 60    | 11.1  | 62.0975            | 14.3223  | 20.4208                                  | 1.8397 | 398   |
| 59    | 11.0  | 67.8105            | 17.3913  | 17.9967                                  | 1.6361 | 448   |
| 58    | 10.9  | 73.0267            | 21.4834  | 16.2048                                  | 1.4867 | 493   |
| 57    | 10.9  | 77.7461            | 25.0640  | 15.3757                                  | 1.4106 | 520   |
| 56    | 10.8  | 82.2171            | 29.1561  | 13.8182                                  | 1.2795 | 573   |
| 55    | 10.6  | 86.1913            | 32.7366  | 12.8885                                  | 1.2159 | 603   |
| 54    | 10.5  | 89.9172            | 35.8057  | 11.9256                                  | 1.1358 | 645   |
| 53    | 10.3  | 93.6430            | 39.6932  | 11.1815                                  | 1.0855 | 675   |
| 52    | 10.4  | 97.1205            | 43.2737  | 10.4415                                  | 1.0040 | 730   |
| 51    | 10.3  | 100.3496           | 46.5474  | 9.8794                                   | 0.9592 | 764   |
| 50    | 10.2  | 103.3302           | 49.6165  | 9.3758                                   | 0.9192 | 797   |
| 49    | 10.1  | 106.3109           | 52.0717  | 9.4463                                   | 0.9353 | 784   |
| 48    | 10.0  | 109.0432           | 54.4758  | 9.2575                                   | 0.9257 | 792   |
| 47    | 9.9   | 111.7755           | 56.7776  | 9.0433                                   | 0.9135 | 802   |
| 46    | 9.8   | 114.2594           | 59.2328  | 8.5865                                   | 0.8762 | 836   |
| 45    | 9.6   | 116.7436           | 61.3812  | 8.3902                                   | 0.8740 | 839   |
| 44    | 9.6   | 118.9788           | 63.4272  | 8.3121                                   | 0.8658 | 847   |
| 43    | 9.4   | 121.2143           | 65.4733  | 7.9283                                   | 0.8434 | 869   |
| 42    | 9.1   | 123.4498           | 67.5193  | 7.4699                                   | 0.8209 | 893   |
| 41    | 9.1   | 125.4369           | 69.3096  | 7.6126                                   | 0.8256 | 888   |
| 40    | 8.9   | 127.4241           | 71.0399  | 7.2530                                   | 0.8149 | 899   |
| 39    | 8.9   | 129.4112           | 72.6344  | 7.3736                                   | 0.8285 | 885   |
| 38    | 8.7   | 131.1499           | 73.9132  | 7.3628                                   | 0.8463 | 860   |
| 37    | 8.7   | 132.8886           | 75.2431  | 7.4058                                   | 0.8512 | 861   |
| 36    | 8.6   | 134.6274           | 76.7265  | 7.1198                                   | 0.8279 | 885   |
| 35    | 8.5   | 136.3661           | 77.9030  | 7.2003                                   | 0.8471 | 865   |
| 34    | 8.4   | 137.8564           | 79.2840  | 6.9030                                   | 0.8218 | 892   |

| $r_i$ | $x_i$ | AIR                |                      | $z = 33.5 \quad (x \ 18.73)$ |                      |       |
|-------|-------|--------------------|----------------------|------------------------------|----------------------|-------|
|       |       | $\sum_{i=1}^n x_i$ | $\sum_{i=1}^n x_i^2$ | $\sum_{i=1}^n x_i$           | $\sum_{i=1}^n x_i^2$ | $T_i$ |
| 33    | 8.2   | 139.3468           | 80.7163              | 6.5394                       | 0.7975               | 919   |
| 32    | 8.0   | 140.8371           | 81.8416              | 6.5082                       | 0.8135               | 901   |
| 30    | 11.1  | 143.5694           | 84.2457              | 8.7963                       | 0.7925               | 925   |
| 27    | 13.1  | 147.2953           | 87.4682              | 10.1778                      | 0.7769               | 943   |
| 24    | 12.4  | 150.5243           | 90.4350              | 9.3006                       | 0.7500               | 977   |
| 21    | 11.6  | 153.2566           | 93.0948              | 8.6756                       | 0.7419               | 980   |
| 18    | 10.9  | 155.4921           | 95.1409              | 8.143                        | 0.7470               | 981   |
| 15    | 10.0  | 157.4793           | 96.6754              | 7.442                        | 0.7442               | 985   |
| 12    | 9.0   | 158.9696           | 98.2099              | 6.664                        | 0.7404               | 990   |
| 9     | 7.9   | 160.2115           | 99.2329              | 5.791                        | 0.733                | 1000  |

| AIR $z = 35.0$ ( $\bar{x} = 18.73$ ) |                        |            |                                                |       |
|--------------------------------------|------------------------|------------|------------------------------------------------|-------|
| $r_i$                                | $S_0 \sum_{j=1}^n x_j$ | $\delta_j$ | $\sum_{j=1}^n x_j - \sum_{j=1}^n \delta_j x_j$ | P.    |
| 55.6                                 | 91.9044                | 1.0827     | 36.2215                                        | 7.08  |
| 55.2                                 | 93.1463                | 1.0486     | 37.7547                                        | 7.38  |
| 54.8                                 | 94.6367                | 1.0166     | 39.2433                                        | 7.67  |
| 54.4                                 | 96.1270                | 0.9892     | 40.7547                                        | 7.97  |
| 54.0                                 | 97.3690                | 0.9734     | 42.1742                                        | 8.24  |
| 53.6                                 | 98.8573                | 0.9582     | 43.5510                                        | 8.51  |
| 53.2                                 | 100.1013               | 0.9434     | 44.9130                                        | 8.78  |
| 52.8                                 | 101.3432               | 0.9290     | 46.2236                                        | 9.04  |
| 52.4                                 | 102.8336               | 0.9174     | 47.5112                                        | 9.29  |
| 52.0                                 | 104.0755               | 0.9060     | 48.6630                                        | 9.51  |
| 51.6                                 | 105.0691               | 0.8906     | 49.9975                                        | 9.77  |
| 51.2                                 | 106.3111               | 0.8799     | 51.1988                                        | 10.01 |
| 50.8                                 | 107.5530               | 0.8716     | 52.2804                                        | 10.24 |
| 50.4                                 | 108.7949               | 0.8613     | 53.5541                                        | 10.47 |
| 50.0                                 | 109.7885               | 0.8533     | 54.6416                                        | 10.68 |
| 49.6                                 | 111.0305               | 0.8454     | 55.7566                                        | 10.90 |
| 49.2                                 | 112.0240               | 0.8396     | 56.8084                                        | 11.11 |
| 48.8                                 | 113.2660               | 0.8301     | 57.8942                                        | 11.32 |
| 48.4                                 | 114.2595               | 0.8227     | 58.8618                                        | 11.51 |
| 48.0                                 | 115.2531               | 0.8171     | 59.8908                                        | 11.71 |
| 47.6                                 | 116.2466               | 0.8117     | 60.9141                                        | 11.91 |
| 47.2                                 | 117.2402               | 0.8064     | 61.8729                                        | 12.10 |
| 46.8                                 | 118.2338               | 0.8011     | 62.8263                                        | 12.28 |
| 46.4                                 | 119.2273               | 0.7976     | 63.6658                                        | 12.45 |
| 46.0                                 | 120.2209               | 0.7916     | 64.9615                                        | 12.70 |
| 45.6                                 | 121.2145               | 0.7856     | 65.5383                                        | 12.81 |
| 45.2                                 | 121.9596               | 0.7806     | 66.5122                                        | 13.00 |
| 44.8                                 | 122.9532               | 0.7773     | 67.3118                                        | 13.16 |
| 44.4                                 | 123.9468               | 0.7740     | 68.1526                                        | 13.32 |
| 44.0                                 | 124.6919               | 0.7707     | 68.7877                                        | 13.49 |
| 43.6                                 | 125.6855               | 0.7675     | 69.7971                                        | 13.64 |

| $r_i$ | AIR $z = 35.0$ ( $\times 18.73$ ) |        |                                           |       |
|-------|-----------------------------------|--------|-------------------------------------------|-------|
|       | $S_0 \sum_1^n x_i$                | $S_1$  | $S_0 \sum_1^n x_i - \sum_1^{n-1} S_i x_i$ | P     |
| 43.2  | 126.4307                          | 0.7643 | 70.5906                                   | 13.80 |
| 42.8  | 127.4242                          | 0.7612 | 71.6549                                   | 14.01 |
| 42.4  | 128.1694                          | 0.7580 | 72.1190                                   | 14.10 |
| 41.2  | 130.6533                          | 0.7502 | 74.3782                                   | 14.54 |
| 40.4  | 132.1436                          | 0.7472 | 75.7616                                   | 14.81 |
| 39.6  | 133.3856                          | 0.7411 | 77.1443                                   | 15.08 |
| 38.8  | 134.8758                          | 0.7382 | 78.4412                                   | 15.33 |
| 38.0  | 136.3663                          | 0.7337 | 79.7616                                   | 15.59 |
| 37.2  | 137.6082                          | 0.7380 | 80.9480                                   | 15.82 |
| 34.8  | 141.5825                          | 0.7222 | 84.3943                                   | 16.50 |
| 30.8  | 147.2954                          | 0.7137 | 89.3979                                   | 17.48 |
| 24.6  | 154.2999                          | 0.7085 | 95.6435                                   | 18.70 |
| 15.5  | 161.5055                          | 0.7085 | 101.9539                                  | 19.73 |

| ETHYLENE/AIR $z = 0 \quad (x = 18.73)$ |       |                    |          |                    |            |       |
|----------------------------------------|-------|--------------------|----------|--------------------|------------|-------|
| $r_i$                                  | $x_i$ | $\sum_{j=0}^i x_j$ | $9.366P$ | $\sum_{j=0}^i x_j$ | $\delta_i$ | $T_i$ |
| 59                                     | 11.0  | 27.3229            | 0.5115   | 26.8114            | 2.4374     | 315   |
| 58                                     | 10.9  | 38.2521            | 0.6138   | 26.6700            | 2.4468     | 314   |
| 57                                     | 10.9  | 46.6973            | 1.0230   | 26.2497            | 2.4082     | 319   |
| 56                                     | 10.8  | 53.6522            | 1.5345   | 25.8904            | 2.3973     | 320   |
| 55                                     | 10.6  | 59.8620            | 2.2506   | 24.9590            | 2.3546     | 326   |
| 54                                     | 10.5  | 65.0782            | 3.2225   | 24.1451            | 2.2995     | 334   |
| 53                                     | 10.5  | 70.0460            | 4.6036   | 22.8720            | 2.1783     | 343   |
| 52                                     | 10.4  | 74.5170            | 6.1893   | 22.4981            | 2.1633     | 355   |
| 51                                     | 10.3  | 78.7396            | 8.1842   | 21.1459            | 2.0530     | 374   |
| 50                                     | 10.2  | 82.7139            | 10.2302  | 20.0645            | 1.9671     | 390   |
| 49                                     | 10.1  | 86.1913            | 12.2762  | 19.1680            | 1.8978     | 405   |
| 48                                     | 10.0  | 89.6688            | 14.0665  | 18.6107            | 1.8611     | 413   |
| 47                                     | 9.9   | 92.8979            | 15.8568  | 17.9933            | 1.8175     | 422   |
| 46                                     | 9.8   | 95.8785            | 17.4425  | 17.5733            | 1.7932     | 428   |
| 45                                     | 9.6   | 98.8592            | 19.3351  | 16.6329            | 1.7326     | 443   |
| 44                                     | 9.6   | 101.5915           | 20.9719  | 16.4111            | 1.7095     | 449   |
| 43                                     | 9.4   | 104.0754           | 22.5064  | 15.8377            | 1.6849     | 456   |
| 42                                     | 9.1   | 106.8077           | 24.0921  | 15.0893            | 1.6582     | 463   |
| 41                                     | 9.1   | 109.0432           | 25.5755  | 14.8238            | 1.6290     | 471   |
| 40                                     | 8.9   | 111.2787           | 27.1100  | 14.2071            | 1.5963     | 481   |
| 39                                     | 8.9   | 113.5142           | 28.6446  | 13.9530            | 1.5677     | 490   |
| 38                                     | 8.7   | 115.5013           | 29.9233  | 13.5940            | 1.5625     | 491   |
| 37                                     | 8.7   | 117.4885           | 31.4579  | 13.2070            | 1.5180     | 506   |
| 36                                     | 8.6   | 119.4756           | 32.7366  | 12.9924            | 1.5107     | 508   |
| 35                                     | 8.5   | 121.2143           | 34.2200  | 12.4823            | 1.4685     | 523   |
| 34                                     | 8.4   | 122.9530           | 35.2942  | 12.4406            | 1.4810     | 519   |
| 33                                     | 8.2   | 124.6918           | 36.7776  | 11.6997            | 1.4268     | 538   |
| 32                                     | 8.0   | 126.4305           | 37.8517  | 11.4687            | 1.4336     | 536   |
| 30                                     | 11.1  | 129.4112           | 40.3070  | 15.4191            | 1.3891     | 553   |
| 27                                     | 13.1  | 133.3854           | 43.2737  | 17.9811            | 1.3726     | 559   |
| 24                                     | 12.4  | 136.8629           | 46.0359  | 16.5662            | 1.3360     | 575   |
| 21                                     | 11.6  | 140.0920           | 48.4915  | 15.1547            | 1.3064     | 588   |
| 18                                     | 10.9  | 142.5759           | 50.1280  | 14.3992            | 1.3213     | 581   |
| 15                                     | 10.0  | 144.8114           | 51.4579  | 12.2736            | 1.2736     | 603   |
| 12                                     | 9.0   | 146.3017           | 52.4230  | 11.2401            | 1.2489     | 615   |

| ETHYLENE/AIR $z = 3.0$ (x 18.73) |       |                |         |                                        |            |       |
|----------------------------------|-------|----------------|---------|----------------------------------------|------------|-------|
| $r_i$                            | $x_i$ | $\sum_0^i x_i$ | 9.366P  | $\sum_0^i x_i - \sum_0^i \delta_i x_i$ | $\delta_i$ | $T_i$ |
| 59                               | 11.0  | 27.3229        | 0.2557  | 27.0672                                | 2.4606     | 312   |
| 58                               | 10.9  | 38.2521        | 0.6138  | 26.5656                                | 2.4372     | 315   |
| 57                               | 10.9  | 46.6973        | 1.0230  | 26.3361                                | 2.4161     | 318   |
| 56                               | 10.8  | 53.6522        | 1.5345  | 25.8186                                | 2.3906     | 321   |
| 55                               | 10.6  | 59.8620        | 2.5575  | 24.6206                                | 2.3227     | 331   |
| 54                               | 10.5  | 65.0782        | 3.4783  | 23.9964                                | 2.2854     | 336   |
| 53                               | 10.5  | 70.0460        | 4.6036  | 23.0148                                | 2.1919     | 350   |
| 52                               | 10.4  | 74.5170        | 5.9847  | 22.7527                                | 2.1878     | 351   |
| 51                               | 10.3  | 78.7396        | 7.6726  | 21.6011                                | 2.0972     | 366   |
| 50                               | 10.2  | 82.7139        | 9.2072  | 20.8673                                | 2.0458     | 375   |
| 49                               | 10.1  | 86.1913        | 11.0998 | 19.8475                                | 1.9651     | 391   |
| 48                               | 10.0  | 89.6688        | 12.7877 | 19.3132                                | 1.9213     | 400   |
| 47                               | 9.9   | 92.8979        | 14.8338 | 18.2119                                | 1.8396     | 417   |
| 46                               | 9.8   | 95.8785        | 16.8287 | 17.4218                                | 1.7777     | 432   |
| 45                               | 9.6   | 98.8592        | 16.5167 | 16.8419                                | 1.7544     | 438   |
| 44                               | 9.6   | 101.5915       | 20.4092 | 16.3253                                | 1.7005     | 452   |
| 43                               | 9.4   | 104.0754       | 21.9949 | 15.8014                                | 1.6809     | 457   |
| 42                               | 9.1   | 106.8077       | 23.5295 | 15.1479                                | 1.6646     | 461   |
| 41                               | 9.1   | 109.0432       | 25.0640 | 14.8358                                | 1.6303     | 471   |
| 40                               | 8.9   | 111.2787       | 26.5985 | 14.2450                                | 1.6006     | 480   |
| 39                               | 8.9   | 113.5142       | 28.1330 | 13.9928                                | 1.5722     | 488   |
| 38                               | 8.7   | 115.5013       | 29.4630 | 13.5878                                | 1.5618     | 492   |
| 37                               | 8.7   | 117.4885       | 30.6906 | 13.5345                                | 1.5557     | 491   |
| 36                               | 8.6   | 119.4756       | 32.0717 | 13.0983                                | 1.5231     | 504   |
| 35                               | 8.5   | 121.2143       | 33.2481 | 12.9015                                | 1.5178     | 506   |
| 34                               | 8.4   | 122.9530       | 34.5269 | 12.5132                                | 1.4895     | 516   |
| 33                               | 8.2   | 124.6918       | 35.8057 | 12.0718                                | 1.4656     | 524   |
| 32                               | 8.0   | 126.4305       | 37.0845 | 11.4916                                | 1.4364     | 535   |
| 30                               | 11.1  | 129.4112       | 38.9771 | 16.0943                                | 1.4499     | 530   |
| 27                               | 13.1  | 133.3854       | 41.8415 | 18.5896                                | 1.4190     | 541   |
| 24                               | 12.4  | 136.8629       | 43.7341 | 17.8835                                | 1.4422     | 532   |
| 21                               | 11.6  | 140.0920       | 46.0679 | 16.1124                                | 1.3890     | 553   |
| 18                               | 10.9  | 142.5759       | 46.5474 | 15.9523                                | 1.4635     | 525   |
| 15                               | 10.0  | 144.8114       | 47.5704 | 14.9903                                | 1.4990     | 512   |

| ETHYLENE/AIR $z = 15.5$ (x 18.73) |       |                |         |                                   |        |       |
|-----------------------------------|-------|----------------|---------|-----------------------------------|--------|-------|
| $r_i$                             | $x_i$ | $\sum_0^i x_i$ | 9.366P  | $\sum_0^i x_i - \sum_0^i S_i x_i$ | $S_i$  | $T_i$ |
| 56                                | 10.8  | 26.8261        | 0.5115  | 26.3146                           | 2.4365 | 315   |
| 55                                | 10.6  | 37.5069        | 1.0230  | 25.5197                           | 2.4075 | 319   |
| 54                                | 10.5  | 45.4554        | 1.5345  | 25.0410                           | 2.3848 | 322   |
| 53                                | 10.3  | 52.4103        | 2.5064  | 23.9158                           | 2.3219 | 331   |
| 52                                | 10.4  | 58.1233        | 3.0691  | 24.1180                           | 2.3190 | 331   |
| 51                                | 10.3  | 63.3394        | 4.0921  | 23.3067                           | 2.2628 | 339   |
| 50                                | 10.2  | 68.3072        | 5.1151  | 22.6626                           | 2.2218 | 346   |
| 49                                | 10.1  | 72.5299        | 6.8031  | 21.4339                           | 2.1222 | 362   |
| 48                                | 10.0  | 76.5041        | 8.3888  | 20.6147                           | 2.0615 | 372   |
| 47                                | 9.9   | 80.2300        | 9.7698  | 20.0799                           | 2.0283 | 379   |
| 46                                | 9.8   | 83.9558        | 11.4067 | 19.2608                           | 1.9654 | 391   |
| 45                                | 9.6   | 87.1849        | 12.7877 | 18.6277                           | 1.9404 | 396   |
| 44                                | 9.6   | 90.1656        | 14.3223 | 18.1833                           | 1.8941 | 405   |
| 43                                | 9.4   | 93.1462        | 15.8568 | 17.3930                           | 1.8530 | 415   |
| 42                                | 9.1   | 95.8785        | 16.8798 | 16.9720                           | 1.8650 | 412   |
| 41                                | 9.1   | 98.6108        | 18.2097 | 16.6214                           | 1.8265 | 420   |
| 40                                | 8.9   | 101.0947       | 19.4374 | 16.0624                           | 1.8048 | 425   |
| 39                                | 8.9   | 103.5786       | 20.4604 | 16.0604                           | 1.8045 | 426   |
| 38                                | 8.7   | 105.8141       | 21.5857 | 15.4872                           | 1.7801 | 431   |
| 37                                | 8.7   | 108.0496       | 22.7622 | 15.2379                           | 1.7515 | 438   |
| 36                                | 8.6   | 110.0363       | 23.6318 | 15.1428                           | 1.7608 | 436   |
| 35                                | 8.5   | 112.0239       | 24.6448 | 14.7744                           | 1.7382 | 442   |
| 34                                | 8.4   | 114.0110       | 25.6266 | 14.4759                           | 1.7233 | 446   |
| 33                                | 8.2   | 115.7497       | 26.5985 | 13.9943                           | 1.7066 | 450   |
| 32                                | 8.0   | 117.4885       | 27.6727 | 13.3836                           | 1.6729 | 459   |
| 30                                | 11.1  | 120.7175       | 29.5653 | 18.3216                           | 1.6506 | 465   |
| 27                                | 13.1  | 124.9402       | 32.3274 | 21.0224                           | 1.6048 | 479   |
| 24                                | 12.4  | 128.6660       | 34.4758 | 19.7215                           | 1.5904 | 483   |
| 21                                | 11.6  | 132.1435       | 36.3172 | 18.2916                           | 1.5769 | 487   |
| 18                                | 10.9  | 134.6274       | 38.1075 | 16.8076                           | 1.5420 | 498   |
| 15                                | 10.0  | 136.8629       | 39.5397 | 15.2612                           | 1.5261 | 503   |
| 12                                | 9.0   | 138.8500       | 40.9208 | 13.3964                           | 1.4835 | 516   |
| 9                                 | 7.9   | 140.0920       | 41.9438 | 11.6339                           | 1.4726 | 521   |
| 6                                 | 6.7   | 141.0855       | 42.9668 | 9.4968                            | 1.4174 | 542   |

| ETHYLENE/AIR $z = 25.0$ (x 18.73) |       |                    |         |                                           |            |       |
|-----------------------------------|-------|--------------------|---------|-------------------------------------------|------------|-------|
| $r_i$                             | $x_i$ | $\sum_{j=1}^i x_j$ | 9.366P  | $\sum_{j=1}^i x_j - \sum_{j=1}^{i-1} x_j$ | $\delta_i$ | $T_i$ |
| 61.0                              | 11.0  | 27.3229            |         | 27.3229                                   | 2.4839     | 309   |
| 60.0                              | 11.1  | 38.7488            | 1.0230  | 26.5483                                   | 2.3917     | 321   |
| 59.0                              | 11.0  | 47.4425            | 1.5857  | 26.1522                                   | 2.3775     | 323   |
| 58.0                              | 10.9  | 54.3974            | 2.5575  | 25.3186                                   | 2.3228     | 331   |
| 57.0                              | 10.9  | 60.6072            | 3.2737  | 25.1533                                   | 2.3076     | 333   |
| 56.0                              | 10.8  | 66.0717            | 4.5524  | 24.1561                                   | 2.2367     | 343   |
| 55.0                              | 10.6  | 71.2879            | 5.7289  | 23.2787                                   | 2.1961     | 350   |
| 54.0                              | 10.5  | 75.7589            | 7.1611  | 22.4100                                   | 2.1344     | 360   |
| 53.0                              | 10.3  | 80.2300            | 8.6957  | 21.4015                                   | 2.0778     | 370   |
| 52.0                              | 10.4  | 83.9558            | 9.9744  | 21.3699                                   | 2.0548     | 376   |
| 51.0                              | 10.3  | 87.6817            | 11.2532 | 20.8731                                   | 2.0265     | 379   |
| 50.0                              | 10.2  | 91.1591            | 12.7877 | 20.1034                                   | 1.9709     | 390   |
| 49.0                              | 10.1  | 94.6366            | 14.0665 | 19.7201                                   | 1.9575     | 393   |
| 48.0                              | 10.0  | 97.6173            | 15.3453 | 19.2871                                   | 1.9282     | 398   |
| 47.0                              | 9.9   | 100.5979           | 16.8798 | 18.5664                                   | 1.8754     | 408   |
| 46.0                              | 9.8   | 103.3302           | 18.3632 | 18.0182                                   | 1.8386     | 418   |
| 45.0                              | 9.6   | 106.0625           | 19.5908 | 17.5635                                   | 1.8295     | 420   |
| 44.0                              | 9.6   | 108.7948           | 20.9719 | 17.2419                                   | 1.7960     | 428   |
| 43.0                              | 9.4   | 111.0303           | 22.4553 | 16.4704                                   | 1.7522     | 438   |
| 42.0                              | 9.1   | 113.5142           | 23.5806 | 15.9510                                   | 1.7528     | 438   |
| 41.0                              | 9.1   | 115.7497           | 25.0640 | 15.5020                                   | 1.7035     | 451   |
| 40.0                              | 8.9   | 117.9852           | 26.1893 | 15.1525                                   | 1.7025     | 451   |
| 39.0                              | 8.9   | 119.9724           | 27.6215 | 14.7441                                   | 1.6566     | 464   |
| 38.0                              | 8.7   | 121.9595           | 28.7469 | 14.3702                                   | 1.6517     | 465   |
| 37.0                              | 8.7   | 123.6982           | 29.9745 | 14.1559                                   | 1.6271     | 472   |
| 36.0                              | 8.6   | 125.6853           | 31.0998 | 13.8810                                   | 1.6141     | 476   |
| 35.0                              | 8.5   | 127.4241           | 32.2251 | 13.5614                                   | 1.5955     | 481   |
| 34.0                              | 8.4   | 128.9144           | 33.2993 | 13.2790                                   | 1.5805     | 486   |
| 33.0                              | 8.2   | 130.6531           | 34.3223 | 12.8701                                   | 1.5695     | 497   |
| 32.0                              | 8.0   | 132.1435           | 35.3965 | 12.3714                                   | 1.5464     | 497   |

| ETHYLENE/AIR |       |                | z = 25.0 (x 18.73) |                               |        |       |
|--------------|-------|----------------|--------------------|-------------------------------|--------|-------|
| $r_i$        | $x_i$ | $\sum_0^i x_i$ | 9.366P             | $\sum_0^i x_i - \sum_0^i x_i$ | $G_i$  | $T_i$ |
| 30.0         | 11.1  | 135.1242       | 37.2891            | 17.0022                       | 1.5317 | 501   |
| 27.0         | 13.1  | 138.8500       | 39.6932            | 19.9147                       | 1.5202 | 505   |
| 24.0         | 12.4  | 142.3275       | 41.6881            | 18.7516                       | 1.5122 | 508   |
| 21.0         | 11.6  | 145.3081       | 43.4783            | 17.3904                       | 1.4992 | 512   |
| 18.0         | 10.9  | 147.7920       | 45.0640            | 16.1434                       | 1.4810 | 519   |
| 15.0         | 10.0  | 149.7792       | 46.7009            | 14.3713                       | 1.4371 | 534   |
| 12.0         | 9.0   | 151.5179       | 48.1331            | 12.5897                       | 1.3989 | 549   |
| 9.0          | 7.9   | 152.7598       | 49.6165            | 10.5094                       | 1.3303 | 577   |
| 6.0          | 6.7   | 153.7534       | 50.6395            | 8.6757                        | 1.2949 | 593   |

| ETHYLENE/AIR |       |                    |         | z = 29.0 (x 18.73)                            |            |       |
|--------------|-------|--------------------|---------|-----------------------------------------------|------------|-------|
| $r_i$        | $x_i$ | $\sum_{i=1}^n x_i$ | 9.366P  | $\sum_{i=1}^n x_i^2 - \sum_{i=1}^{n-1} x_i^2$ | $\delta_i$ | $T_i$ |
| 63           | 11.3  | 28.0681            | 0.2557  | 27.8124                                       | 2.4613     | 312   |
| 62           | 11.1  | 39.4940            | 1.0742  | 26.6056                                       | 2.3969     | 329   |
| 61           | 11.0  | 48.1877            | 2.0972  | 25.7183                                       | 2.3380     | 328   |
| 60           | 11.1  | 55.6394            | 3.5806  | 25.0228                                       | 2.2552     | 340   |
| 59           | 11.0  | 61.8491            | 5.1151  | 24.0865                                       | 2.1897     | 351   |
| 58           | 10.9  | 67.3137            | 6.6496  | 23.2785                                       | 2.1356     | 360   |
| 57           | 10.9  | 72.5299            | 8.3376  | 22.6287                                       | 2.0760     | 370   |
| 56           | 10.8  | 77.2493            | 10.2302 | 21.6521                                       | 2.0048     | 383   |
| 55           | 10.6  | 81.4719            | 12.3785 | 20.2875                                       | 1.9233     | 399   |
| 54           | 10.5  | 85.4462            | 14.3244 | 19.6670                                       | 1.8730     | 410   |
| 53           | 10.3  | 89.1720            | 16.6752 | 18.4068                                       | 1.7871     | 430   |
| 52           | 10.4  | 92.8979            | 18.5167 | 18.3804                                       | 1.7673     | 435   |
| 51           | 10.3  | 96.3753            | 20.6138 | 17.5748                                       | 1.7063     | 450   |
| 50           | 10.2  | 99.3560            | 22.7622 | 16.8616                                       | 1.6531     | 465   |
| 49           | 10.1  | 102.5851           | 25.0640 | 16.0202                                       | 1.5862     | 484   |
| 48           | 10.0  | 105.3174           | 27.0589 | 15.5747                                       | 1.5575     | 493   |
| 47           | 9.9   | 108.0496           | 29.0538 | 15.0522                                       | 1.5204     | 505   |
| 46           | 9.8   | 110.7819           | 30.8440 | 14.7186                                       | 1.5019     | 511   |
| 45           | 9.6   | 113.2658           | 32.7366 | 14.0676                                       | 1.4654     | 524   |
| 44           | 9.6   | 115.7497           | 34.7315 | 13.6313                                       | 1.4199     | 541   |
| 43           | 9.4   | 117.9852           | 36.3684 | 13.2540                                       | 1.4100     | 545   |
| 42           | 9.1   | 120.2208           | 38.3121 | 12.3448                                       | 1.3566     | 566   |
| 41           | 9.1   | 122.2079           | 39.8978 | 12.3281                                       | 1.3447     | 567   |
| 40           | 8.9   | 124.4434           | 41.4323 | 11.9421                                       | 1.3418     | 572   |
| 39           | 8.9   | 126.4305           | 42.9668 | 11.7772                                       | 1.3233     | 580   |
| 38           | 8.7   | 128.1692           | 44.5014 | 11.2750                                       | 1.2960     | 593   |
| 37           | 8.7   | 129.9080           | 45.9336 | 11.1735                                       | 1.2843     | 598   |
| 36           | 8.6   | 131.6467           | 47.2635 | 10.9807                                       | 1.2768     | 601   |
| 35           | 8.5   | 133.3854           | 48.7469 | 10.5683                                       | 1.2433     | 618   |
| 34           | 8.4   | 134.8758           | 50.1791 | 10.2412                                       | 1.2192     | 630   |

## ETHYLENE/AIR

z = 29.0 (x 18.73)

| $r_i$ | $x_i$ | $\sum_0^n x_i$ | 9.366P  | $\sum_0^n x_i - \sum_1^{n-1} x_i$ | $\xi_i$ | $T_i$ |
|-------|-------|----------------|---------|-----------------------------------|---------|-------|
| 33    | 8.2   | 136.6145       | 51.6625 | 9.7274                            | 1.1863  | 647   |
| 32    | 8.0   | 138.1048       | 52.9413 | 9.4433                            | 1.1804  | 651   |
| 30    | 11.1  | 140.8371       | 55.2431 | 13.0113                           | 1.1722  | 655   |
| 27    | 13.1  | 144.5630       | 58.4144 | 15.0573                           | 1.1494  | 668   |
| 24    | 12.4  | 147.7920       | 61.0231 | 14.1244                           | 1.1391  | 674   |
| 21    | 11.6  | 150.5243       | 63.3249 | 13.0689                           | 1.1266  | 682   |
| 18    | 10.9  | 153.0082       | 65.1664 | 12.2407                           | 1.1230  | 684   |
| 15    | 10.0  | 154.9954       | 66.5986 | 11.2838                           | 1.1284  | 681   |
| 12    | 9.0   | 156.4857       | 67.9797 | 9.9380                            | 1.1042  | 695   |
| 9     | 7.9   | 157.7276       | 69.0027 | 8.6815                            | 1.0989  | 699   |
| 6     | 6.7   | 158.7212       | 70.0769 | 6.9689                            | 1.0890  | 705   |

| ETHYLENE/AIR |       |                |         | z = 30.5 (x 18.73)                |         |       |
|--------------|-------|----------------|---------|-----------------------------------|---------|-------|
| $r_i$        | $x_i$ | $\sum_0^n x_i$ | 9.366P  | $\sum_0^n x_i - \sum_0^{i-1} x_i$ | $\xi_i$ | $T_i$ |
| 64           | 11.3  | 28.0681        | 0.0511  | 28.0170                           | 2.4794  | 310   |
| 63           | 11.3  | 39.7424        | 0.7673  | 27.3219                           | 2.4178  | 318   |
| 62           | 11.1  | 48.4360        | 1.9437  | 25.9611                           | 2.3388  | 328   |
| 61           | 11.0  | 55.8877        | 3.5906  | 24.6827                           | 2.2439  | 342   |
| 60           | 11.1  | 62.0975        | 5.1151  | 24.2818                           | 2.1875  | 351   |
| 59           | 11.0  | 67.8105        | 7.1100  | 23.0420                           | 2.0947  | 367   |
| 58           | 10.9  | 73.0267        | 9.3606  | 21.7902                           | 1.9991  | 384   |
| 57           | 10.9  | 77.7461        | 11.8159 | 20.7820                           | 1.9066  | 403   |
| 56           | 10.8  | 82.2171        | 14.3223 | 19.6947                           | 1.8236  | 421   |
| 55           | 10.6  | 86.1913        | 16.7775 | 18.6119                           | 1.7558  | 437   |
| 54           | 10.5  | 89.9172        | 19.4374 | 17.5342                           | 1.6699  | 460   |
| 53           | 10.3  | 93.6430        | 21.9949 | 16.5717                           | 1.6089  | 477   |
| 52           | 10.4  | 97.1205        | 24.6036 | 16.0606                           | 1.5443  | 497   |
| 51           | 10.3  | 100.3496       | 27.5192 | 14.9684                           | 1.4532  | 528   |
| 50           | 10.2  | 103.3302       | 29.9233 | 14.5100                           | 1.4225  | 540   |
| 49           | 10.1  | 106.3109       | 32.4809 | 13.8147                           | 1.3678  | 561   |
| 48           | 10.0  | 109.0432       | 34.7827 | 13.3818                           | 1.3382  | 574   |
| 47           | 9.9   | 111.7755       | 36.9822 | 12.9682                           | 1.3099  | 586   |
| 46           | 9.8   | 114.2594       | 39.0282 | 12.6385                           | 1.2896  | 595   |
| 45           | 9.6   | 116.7433       | 41.0742 | 12.1232                           | 1.2628  | 608   |
| 44           | 9.6   | 118.9788       | 43.0691 | 11.8889                           | 1.2384  | 620   |
| 43           | 9.4   | 121.2143       | 45.1663 | 11.1972                           | 1.1912  | 645   |
| 42           | 9.1   | 123.4498       | 47.0589 | 10.6753                           | 1.1731  | 655   |
| 41           | 9.1   | 125.4369       | 48.6957 | 10.7139                           | 1.1773  | 652   |
| 40           | 8.9   | 127.4241       | 50.6395 | 10.0582                           | 1.1301  | 680   |
| 39           | 8.9   | 129.4112       | 52.2763 | 10.0312                           | 1.1271  | 681   |
| 38           | 8.7   | 131.1499       | 54.1178 | 9.4064                            | 1.0812  | 710   |
| 37           | 8.7   | 132.8886       | 55.7546 | 9.3121                            | 1.0704  | 717   |
| 36           | 8.6   | 134.6274       | 57.2891 | 9.1182                            | 1.0603  | 724   |
| 35           | 8.5   | 136.3661       | 58.7725 | 8.9154                            | 1.0489  | 732   |

| ETHYLENE/AIR |       |                | z = 30.5 |                                       | (x 18.73) |       |
|--------------|-------|----------------|----------|---------------------------------------|-----------|-------|
| $r_i$        | $x_i$ | $\sum_0^n x_i$ | 9.366P   | $\sum_0^n x_i \cdot \sum_0^{n-1} x_i$ | $\phi_i$  | $T_i$ |
| 34           | 8.4   | 137.8564       | 60.0542  | 8.8440                                | 1.0529    | 729   |
| 33           | 8.2   | 139.3468       | 61.3812  | 8.5449                                | 1.0421    | 737   |
| 32           | 8.0   | 140.8371       | 62.7623  | 8.1525                                | 1.0191    | 754   |
| 30           | 11.1  | 143.5694       | 64.9106  | 11.4724                               | 1.0335    | 743   |
| 27           | 13.1  | 147.2953       | 67.9797  | 13.3509                               | 1.0191    | 754   |
| 24           | 12.4  | 150.5243       | 70.5372  | 12.5920                               | 1.0155    | 756   |
| 21           | 11.6  | 153.2566       | 72.7367  | 11.7522                               | 1.0131    | 758   |
| 18           | 10.9  | 155.4921       | 74.8339  | 10.7252                               | 0.9840    | 780   |
| 15           | 10.0  | 157.4792       | 76.6242  | 9.6392                                | 0.9639    | 797   |
| 12           | 9.0   | 158.9696       | 78.1587  | 8.4651                                | 0.9406    | 816   |
| 9            | 7.9   | 160.2115       | 79.3863  | 7.3019                                | 0.9243    | 831   |

| ETHYLENE/AIR |       |                |         | z = 32.0 (x 18.73)                         |            |       |
|--------------|-------|----------------|---------|--------------------------------------------|------------|-------|
| $r_i$        | $x_i$ | $\sum_0^i x_i$ | 9.366P  | $\sum_0^n x_i - \sum_1^{n-1} \delta_i x_i$ | $\delta_i$ | $T_i$ |
| 64           | 11.3  | 28.0681        | 0.6038  | 27.4543                                    | 2.4296     | 316   |
| 63           | 11.3  | 39.7424        | 1.7903  | 26.5330                                    | 2.3481     | 327   |
| 62           | 11.1  | 48.4360        | 3.0691  | 25.3494                                    | 2.2837     | 336   |
| 61           | 11.0  | 55.8877        | 5.1151  | 23.8194                                    | 2.1654     | 355   |
| 60           | 11.1  | 62.0975        | 7.1611  | 23.1399                                    | 2.0847     | 368   |
| 59           | 11.0  | 67.8105        | 10.2302 | 21.1470                                    | 1.9224     | 399   |
| 58           | 10.9  | 73.0267        | 12.7877 | 20.1713                                    | 1.8506     | 415   |
| 57           | 10.9  | 77.7461        | 15.8568 | 18.9150                                    | 1.7353     | 443   |
| 56           | 10.8  | 82.2171        | 19.4374 | 17.1905                                    | 1.5917     | 482   |
| 55           | 10.6  | 86.1913        | 22.7110 | 15.9771                                    | 1.5073     | 509   |
| 54           | 10.5  | 89.9172        | 26.0358 | 14.8356                                    | 1.4129     | 544   |
| 53           | 10.3  | 93.6430        | 29.1561 | 13.9302                                    | 1.3524     | 568   |
| 52           | 10.4  | 97.1205        | 32.5320 | 12.1230                                    | 1.2618     | 609   |
| 51           | 10.3  | 100.3496       | 35.7034 | 12.3615                                    | 1.2001     | 640   |
| 50           | 10.2  | 103.3302       | 38.3632 | 12.0556                                    | 1.1819     | 650   |
| 49           | 10.1  | 106.3109       | 41.4323 | 11.1773                                    | 1.1067     | 694   |
| 48           | 10.0  | 109.0432       | 43.9899 | 10.8979                                    | 1.0898     | 705   |
| 47           | 9.9   | 111.7755       | 46.3428 | 10.6663                                    | 1.0774     | 713   |
| 46           | 9.8   | 114.2594       | 48.5934 | 10.3909                                    | 1.0603     | 724   |
| 45           | 9.6   | 116.7433       | 50.6395 | 10.1457                                    | 1.0568     | 727   |
| 44           | 9.6   | 118.9788       | 52.8901 | 9.7810                                     | 1.0188     | 754   |
| 43           | 9.4   | 121.2143       | 54.8850 | 9.4264                                     | 1.0028     | 766   |
| 42           | 9.1   | 123.4498       | 56.8799 | 8.9306                                     | 0.9814     | 783   |
| 41           | 9.1   | 125.4369       | 58.5679 | 9.0301                                     | 0.9923     | 774   |
| 40           | 8.9   | 127.4241       | 60.3582 | 8.6550                                     | 0.9725     | 790   |
| 39           | 8.9   | 129.4112       | 61.9950 | 8.6268                                     | 0.9693     | 792   |
| 38           | 8.7   | 131.1499       | 63.7341 | 8.1565                                     | 0.9375     | 819   |
| 37           | 8.7   | 132.3886       | 65.2175 | 8.2021                                     | 0.9428     | 815   |
| 36           | 8.6   | 134.6274       | 66.7009 | 8.0334                                     | 0.9341     | 822   |
| 35           | 8.5   | 136.3661       | 68.0820 | 7.9170                                     | 0.9314     | 825   |

| ETHYLENE/AIR |       |                |         | z = 32.0 (x 18.73)                    |            |       |
|--------------|-------|----------------|---------|---------------------------------------|------------|-------|
| $r_i$        | $x_i$ | $\sum_0^n x_i$ | 9.366P  | $\sum_0^n x_i - \sum_{i=1}^{n-1} x_i$ | $\delta_i$ | $T_i$ |
| 34           | 8.4   | 137.8564       | 69.3608 | 7.8225                                | 0.9212     | 825   |
| 33           | 8.2   | 139.3468       | 70.7418 | 7.4829                                | 0.9125     | 842   |
| 32           | 8.0   | 140.8371       | 72.1229 | 7.1525                                | 0.8941     | 859   |
| 30           | 11.1  | 143.5694       | 74.3224 | 10.0556                               | 0.9059     | 848   |
| 27           | 13.1  | 147.2953       | 77.2380 | 11.8241                               | 0.0926     | 851   |
| 24           | 12.4  | 150.5243       | 79.7956 | 11.2447                               | 0.9068     | 847   |
| 21           | 11.6  | 153.2566       | 81.8416 | 10.6530                               | 0.9184     | 836   |
| 18           | 10.9  | 155.4921       | 83.8365 | 9.7456                                | 0.8941     | 859   |
| 15           | 10.0  | 157.4793       | 85.4222 | 8.9029                                | 0.8903     | 863   |
| 12           | 9.0   | 158.9696       | 86.9567 | 7.7258                                | 0.8584     | 895   |
| 9            | 7.9   | 160.2115       | 88.3889 | 6.7411                                | 0.8533     | 900   |

| ETHYLENE/AIR $z = 33.5$ (x 18.73) |       |                             |         |                                                             |            |       |
|-----------------------------------|-------|-----------------------------|---------|-------------------------------------------------------------|------------|-------|
| $r_i$                             | $x_i$ | $\delta_0 \sum_{i=1}^n x_i$ | 9.366P  | $\delta_0 \sum_{i=1}^n x_i - \sum_{i=1}^{n-1} \delta_i x_i$ | $\delta_i$ | $T_i$ |
| 64                                | 11.3  | 28.0681                     | 1.5345  | 26.5336                                                     | 2.3481     | 327   |
| 63                                | 11.3  | 39.7424                     | 3.0691  | 25.6372                                                     | 2.2688     | 338   |
| 62                                | 11.1  | 48.4360                     | 5.1151  | 23.9775                                                     | 2.1601     | 355   |
| 61                                | 11.0  | 55.8877                     | 7.6726  | 22.2889                                                     | 2.0353     | 377   |
| 60                                | 11.1  | 62.0975                     | 10.2302 | 21.5668                                                     | 1.9429     | 395   |
| 59                                | 11.0  | 67.8105                     | 13.2993 | 19.9728                                                     | 1.8157     | 423   |
| 58                                | 10.9  | 73.0267                     | 17.1356 | 17.9096                                                     | 1.6431     | 467   |
| 57                                | 10.9  | 77.7461                     | 20.9719 | 16.5025                                                     | 1.5140     | 507   |
| 56                                | 10.8  | 82.2171                     | 25.5755 | 14.3544                                                     | 1.3291     | 578   |
| 55                                | 10.6  | 86.1913                     | 29.6676 | 13.0427                                                     | 1.2304     | 624   |
| 54                                | 10.5  | 89.9172                     | 33.2481 | 12.3002                                                     | 1.1714     | 656   |
| 53                                | 10.3  | 93.6430                     | 36.8287 | 11.4047                                                     | 1.1072     | 694   |
| 52                                | 10.4  | 97.1205                     | 39.8978 | 11.2787                                                     | 1.0845     | 708   |
| 51                                | 10.3  | 100.3496                    | 42.9668 | 10.7227                                                     | 1.0420     | 737   |
| 50                                | 10.2  | 103.3302                    | 46.0359 | 10.1044                                                     | 0.9906     | 775   |
| 49                                | 10.1  | 106.3109                    | 48.8492 | 9.7398                                                      | 0.9643     | 796   |
| 48                                | 10.0  | 109.0432                    | 51.4579 | 9.4402                                                      | 0.9440     | 813   |
| 47                                | 9.9   | 111.7755                    | 53.7085 | 9.3881                                                      | 0.9483     | 810   |
| 46                                | 9.8   | 114.2694                    | 56.2661 | 8.8449                                                      | 0.9025     | 851   |
| 45                                | 9.6   | 116.7433                    | 58.3121 | 8.7817                                                      | 0.9148     | 839   |
| 44                                | 9.6   | 118.9788                    | 60.3582 | 8.6883                                                      | 0.9050     | 849   |
| 43                                | 9.4   | 121.2143                    | 62.4042 | 8.2869                                                      | 0.8816     | 871   |
| 42                                | 9.1   | 123.4498                    | 64.3991 | 7.8618                                                      | 0.8639     | 889   |
| 41                                | 9.1   | 125.4369                    | 66.0871 | 7.9803                                                      | 0.8769     | 876   |
| 40                                | 8.9   | 127.4241                    | 68.0308 | 7.5124                                                      | 0.8441     | 910   |
| 39                                | 8.9   | 129.4112                    | 69.5654 | 7.6709                                                      | 0.8618     | 891   |
| 38                                | 8.7   | 131.1499                    | 71.2533 | 7.2526                                                      | 0.8336     | 921   |
| 37                                | 8.7   | 132.8886                    | 72.8902 | 7.1591                                                      | 0.8229     | 933   |
| 36                                | 8.6   | 134.6274                    | 74.4247 | 7.0046                                                      | 0.8145     | 943   |
| 35                                | 8.5   | 136.3661                    | 75.8058 | 6.9745                                                      | 0.8205     | 936   |

ETHYLENE/AIR       $z = 33.5$       ( $\times 18.73$ )

| $r_i$ | $x_i$ | $S_0 \sum_{i=1}^n x_i$ | 9.366P  | $S_0 \sum_{i=1}^n x_i - \sum_{i=1}^{n-1} \delta_i x_i$ | $\delta_i$ | $T_i$ |
|-------|-------|------------------------|---------|--------------------------------------------------------|------------|-------|
| 34    | 8.4   | 137.8564               | 77.2380 | 6.7509                                                 | 0.8037     | 955   |
| 33    | 8.2   | 139.3468               | 78.5168 | 6.6251                                                 | 0.8079     | 951   |
| 32    | 8.0   | 140.8371               | 79.7956 | 6.4232                                                 | 0.8079     | 956   |
| 30    | 11.1  | 143.5694               | 82.1485 | 8.8419                                                 | 0.7966     | 969   |
| 27    | 13.1  | 147.2953               | 85.2687 | 10.3690                                                | 0.7915     | 970   |
| 24    | 12.4  | 150.5243               | 87.7240 | 9.9791                                                 | 0.8048     | 954   |
| 21    | 11.6  | 153.2566               | 90.0258 | 9.2713                                                 | 0.7992     | 961   |
| 18    | 10.9  | 155.4921               | 91.9183 | 8.6514                                                 | 0.7937     | 968   |
| 15    | 10.0  | 157.4793               | 93.3506 | 7.9180                                                 | 0.7918     | 970   |
| 12    | 9.0   | 158.9696               | 94.6293 | 7.0461                                                 | 0.7829     | 981   |

## ANALYSIS OF INTERFEROGRAMS FOR

ETHYLENE - AIR.

Fringe order distribution correction.

| Ethylene/Air                 |       |            |                                                             |       |
|------------------------------|-------|------------|-------------------------------------------------------------|-------|
| $z = 35.0 \quad (x \ 18.73)$ |       |            |                                                             |       |
| $r_i$                        | $T_i$ | $\delta_i$ | $\delta_0 \sum_{i=1}^n x_i - \sum_{i=1}^{n-1} x_i \delta_i$ | P     |
| 55                           | 717   | 1.0711     | 35.5444                                                     | 6.95  |
| 54                           | 759   | 1.0118     | 39.2622                                                     | 7.67  |
| 53                           | 793   | 0.9685     | 42.8131                                                     | 8.37  |
| 52                           | 819   | 0.9377     | 46.0607                                                     | 9.00  |
| 51                           | 845   | 0.9089     | 49.1438                                                     | 9.61  |
| 50                           | 867   | 0.8858     | 52.0022                                                     | 10.17 |
| 49                           | 885   | 0.8678     | 54.7161                                                     | 10.7  |
| 48                           | 907   | 0.8467     | 57.3227                                                     | 11.2  |
| 47                           | 923   | 0.8320     | 59.7873                                                     | 11.7  |
| 46                           | 939   | 0.8179     | 62.1325                                                     | 12.15 |
| 45                           | 948   | 0.8101     | 64.3400                                                     | 12.6  |
| 44                           | 961   | 0.7992     | 66.4651                                                     | 13.0  |
| 43                           | 971   | 0.7909     | 68.4384                                                     | 13.38 |
| 42                           | 981   | 0.7829     | 70.3499                                                     | 13.75 |
| 41                           | 989   | 0.7765     | 72.2026                                                     | 14.12 |
| 40                           | 995   | 0.7718     | 73.9609                                                     | 14.46 |
| 39                           | 1001  | 0.7672     | 75.6507                                                     | 14.79 |
| 38                           | 1007  | 0.7627     |                                                             |       |
| 35                           | 1023  | 0.7507     | 81.6275                                                     | 15.96 |
| 30                           | 1038  | 0.7399     | 87.7824                                                     | 17.16 |
| 21                           | 1043  | 0.7363     | 95.7354                                                     | 18.72 |
| 9                            | 1043  |            | 101.3985                                                    | 19.82 |

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