

Kinetics of Coal Combustion: The Influence of Oxygen Concentration
on the Burning-Out Times of Single Particles

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1. INTRODUCTION

Single particles of coal burn in two stages. The first is a volatile combustion stage; and the second, with which this paper is primarily concerned, is burn-out of the solid carbon residue left after generation and combustion of the volatiles. Kinetically, this second, burn-out stage is a heterogeneous process in which oxygen reacts directly, at-and-with the solid surface. It has, therefore, been studied extensively, but as it were by proxy using relatively pure carbon in place of the coal; and the validity of extrapolating such results directly to coal residues has generally been then taken very much for granted. Direct work on coal has, of course, been done in the past but the results (1-3) have generally been too few, and the conditions too imprecise, for kinetic studies. Further experiments have, therefore, been carried out on single particles of coal under more precisely specified conditions, to check proposed kinetic equations. The first results obtained, described elsewhere (4,5), were concerned with the variation of burning time as a function of particle diameter and coal rank, with other parameters such as oxygen concentration, temperature, and ambient velocity, kept constant. The success of those first experiments then encouraged extension of the work to investigate the influence of other parameters, and the next one chosen for investigation, was the oxygen concentration; this is of particular interest as it also has direct bearing on the assumed but disputed (6) order of reaction at the solid surface. The object of this present paper is, therefore, to report the results of these further experiments on the influence of oxygen concentration on the burn-out times of the coal particle residues, together with a comparison between the experimental and predicted behaviour.

2. THEORY

As the general theory has been covered extensively in previous reviews and papers (4, 5, 7-9) only the salient points will be quoted in summary here.

The theory is based on the original analysis by Nusselt (10) in which he assumed that the rate controlling process in the reaction was the rate of diffusion of oxygen from the mainstream to the solid particle surface, through a boundary diffusion layer. Reaction at the solid surface was assumed to be instantaneous, or effectively so, and it was also assumed to be first order with respect to the oxygen partial pressure adjacent to the solid surface. With these limiting conditions, Nusselt's prediction was that the total burning time (t_b) of a solid carbon sphere would be proportional to the square of the initial particle diameter (d_0):

thus

$$t_b = K_D d_o^2 \quad (1)$$

where K_D is a predictable burning constant that is a function of temperature and oxygen partial pressure.

This equation was tested in the previous experiments; in the first instance (4) just as it stands using only a few coals, but subsequently (5) the number and rank range of coals was increased to determine the influence of coal rank. To do this, burning times (of both volatiles and residues) of particles in the size range 4000 to 300 microns were measured as a function of diameter. The particles were burned in air, between two small heating coils of resistance wire, under relatively quiescent ambient conditions, at an effectively constant temperature of about 1000°C. In all, 10 coals were ultimately tested, ranging in volatile percentage from 5 to 40, and all were found to obey the eqn. (1). The value of the burning constant K_D had, of course, to be adjusted to allow for the effects of volatile loss and swelling, but the experimental values of the burning constant, K , were found (5) to be in good agreement with the following predicted relationship between K and K_D

$$K = [(C_f/100)/f] \cdot K_D \quad (2)$$

where C_f is the fixed carbon percentage; and f is a swelling factor whose values were found by measurement to be: unity for coals of V.M. less than 5%; and 1.5 for coals of V.M. greater than 10%. The basis of this prediction was the assumption that the coals first lost volatiles at constant diameter, and then swelled by the linear factor f . This then provided a correction factor to the solid density σ that appears in the theoretical relation for K_D

$$K_D = \sigma/3\rho_o D_o (T/T_o)^{0.75} \cdot \ln(1 - p_o) \quad (3a)$$

$$= k/\ln(1 - p_o) \quad (3b)$$

$$\approx k/p_o \quad (\text{for small } p_o) \quad (3c)$$

where ρ_o is the s.t.p. density of air; D_o is the s.t.p. diffusion coefficient of oxygen through nitrogen; T is the absolute temperature; and p_o is the ambient fractional oxygen concentration (of value 0.21 for air).

This set of equations therefore provided us with relations between burning time, or burning constant, and the two additional variables: oxygen concentration and temperature, to be checked by comparison with measurement in further experiments. Our choice of variable for the experiments reported here was the first: that of oxygen concentration, for the reasons outlined in the Introduction. In doing this, we only used a single coal since, as the quantitative influence of rank is given by eqn. (2), we assumed that validation on a single coal chosen at random should be satisfactory. The experiments were then carried out by measuring burning time as a function of oxygen partial pressure, using five different sizes of particle taken from the single coal, as described in the section following.

3. EXPERIMENTAL

To burn the particles in variable but controllable oxygen atmospheres, a small combustion unit was used inside a large perspex (plexiglass) box so that the ambient atmosphere could be controlled at will. The combustion unit was, in principle, that used in the previous experiments (4, 5), in which the coal particles were cemented to silica fibres and held by these, cantilever-fashion, mid-way between two horizontal heating elements of electrical resistance wire.

The coal used was Winter (ex-Grimethorpe); a medium bituminous coal, No. 7 of the set prepared and used previously (5), of analysis: Ultimate - 84.0%C, 5.5%H, 8.3%O, 1.8%N, 0.4%S (d.m.f. by Fereday and Flint (11) equation); Proximate - 36.0% V.M., 2.6% H₂O, 1.7% ash, 0.77% CO₂.

The perspex box used to house the combustion unit had dimensions: 1.5 by 1.5 ft. in plan section, by 3 ft. high, (6.75 cu. ft.). The oxygen atmosphere inside could be adjusted as required over the range 3% to 70% O₂. To make up the required atmosphere, oxygen or nitrogen was metered in as required, and the analysis then checked by Orsat. The reason for making the box so large was that the oxygen depletion during combustion of a particle would not then be significant, and the atmosphere could, therefore, be taken as being effectively infinite, with the 'main stream' or ambient oxygen concentration constant during the reaction thus meeting the specified boundary conditions required by the theoretical analysis. In fact, the box was large enough for a number of particles to be burned without having to open and recharge the box for each particle, and without significant change in the box's atmosphere. To take advantage of this, a rail carrying 18 carriages was mounted in the box, and to each carriage could be attached one silica thread with its coal particle cemented on ready for burning. After burning, the oxygen concentration was checked by Orsat analysis.

The particles on the carriages were moved into position between the coils, as and when required, by means of a control rod extending outside the box. The heating coils were larger than those used previously. In place of the 2-cm diameter flat spirals, wound from 18-gauge Nichrome resistance wire, the new heaters were square elements, of face area about 5 x 5 cm., made of Nichrome strip. This strip was about 1/2 cm. wide, and was wound with 1/2 cm. spacing on a 1/2 cm. thick former; the gaps between the set of strips on one side of the former were, of course, substantially covered by the return strips on the other side so that the whole face area was radiant. The two heating units were then mounted horizontally with their faces about 1.5 cm. apart. The heating was electrical, as before, controlled by a variable transformer unit; with this, element temperatures of up to 1060°C, as measured by an optical pyrometer, could be reached.

Burning times were measured with a stop watch in place of the photocell and pen recorder units used previously. The photocell was abandoned because of light shielding and other difficulties experienced with the larger heating elements. This meant that very short volatile burning times could not be measured - those larger ones that were, were measured with a second stop watch. However, the volatile measurements that were made were found to exhibit such considerable variability in the different oxygen atmospheres that their value was greatly reduced. Most measurements were therefore restricted to the residue burning times alone, using the stop watch which was found to be perfectly satisfactory for these.

With this apparatus, the residue burning times were measured at different levels of oxygen concentration as the principal variable, for each of the following particle sizes: 1870; 1300; 928; 649; and 388 microns. The particles were derived from the single coal, and burned in effectively infinite atmosphere, at an approximately constant temperature of 1000°C.

4. RESULTS

4.1 Qualitative Behaviour - The general behaviour of the particles was as observed before (4, 5): the particles burned in two stages, with the volatiles (when they burned at all) igniting first and burning with the characteristic luminous, flickering flame; and this was then followed in the burn-out stage by the much steadier glow of residue combustion. The particles did not ignite immediately; they required time to heat up to ignition, and this ignition time increased with decrease of oxygen concentration. This may indicate that preliminary, but significant oxidation may be occurring, with significant heat generation, before the volatiles ignited in flaming combustion. This requires closer investigation.

As in the previous experiments, the volatiles of the smaller particles often failed to ignite because of their small quantity and this failure increased, as was to be expected, as the oxygen was reduced. Where the volatiles did ignite in reduced oxygen, the usual fractional lag in time between finish of the volatile flame and start of the residue combustion was occasionally increased to a long delay ranging from two to thirty seconds. At the low oxygen concentrations, below 6%, the burning times also started to become very scattered, and at 3%, the particles failed to ignite at all.

At the other end of the scale, at high oxygen concentrations, the particles showed increasing tendency to decrepitate or explode. This happened whether the coils were already up to temperature before the particle was inserted (as was the case in general in these experiments), or whether the particle was already in position (as in the previous experiments) before switching on the heating current. If decrepitation is due to too rapid generation of volatiles before the coal becomes sufficiently plastic, this suggests that the volatiles generation must be influenced, contrary to previous expectation, by the ambient oxygen concentration. This would seem to imply that the oxygen causes some significant and rapid change in the constitution of the potential volatile material before generation, though how it should do this is by no means clear; this also requires further detailed study. In contrast, the general residue behaviour is far better understood, and more predictable, as described in the sections following.

4.2 Influence of Oxygen Concentration - By combining eqns. (1) and (3b) we have for burn-out of the residues:

$$t_b = [(C_f/100)/f] \cdot kd_o^2 / \ln(1 - p_o) \quad (4)$$

To test this equation, the experimental data obtained have, therefore, been presented in two graphs, Figs. 1 and 2. Fig. 1 is a plot of: $\log_{10} t_b$ against $\log_{10} [\ln(1 - p_o)]$, to show that the slopes of the lines obtained are, within

reason, close to 45° , or -1 . Fig. 2 is the alternative plot of t_b against the reciprocal of $\ln(1 - p_o)$, to show that the plots obtained are again acceptably linear and passing through the origin. Within the limits of accuracy of the measurements, these plots are, therefore, considered to substantiate eqn. (4).

- 4.3 Influence of Particle Size - From eqn. (4) it is clear that the slopes of the lines in Fig. 2 (written as m) are related to particle size by

$$m = [(C_f/100)/f]k.d_o^2 \quad (5)$$

This equation has in turn been tested by plotting $(m)^{1/2}$ against d_o , as shown in Fig. 3. Here again the plot is reasonably linear, and also passes through the origin. Again within the limits of accuracy of this plot, it is considered to substantiate eqn. (5).

- 4.4 Comparison with Prediction - Now, the slope of Fig. 3 (written as M) is an experimental quantity whose value is predicted from the appropriate terms in eqns. (3a) and (5), thus:

$$M^2 = [(C_f/100)/f] \sigma/3\rho_o D_o (T/T_o)^{0.75} \quad (6)$$

For the Winter coal, C_f is 60.7; f is 1.5; and σ is 1.25. For air, ρ_o is 1.3×10^{-3} g/cc; D_o for oxygen diffusing through nitrogen at temperature T_o (273°K) is 0.181 sq.cm/sec. The particle temperature is taken as 1000°C (1273°K), as in the previous experiments. With these values, the calculated value of M^2 is 225; we therefore have for $M_{(calc.)}$ a value of 15, which is precisely the experimental value obtained from the slope of Fig. 3. This exact agreement is clearly fortuitous; but within an error of 5%, which is the estimate of the overall error in both experimental and calculated values, it is clear that agreement is still satisfactory. Since this agreement was obtained by using the rank-influence equation (2), this also supports the assumption made that validation of the tested equations using a single coal, but chosen at random, would probably be satisfactory.

- 4.5 Burning Constant - This is the constant K_D or K of eqn. (1). It is calculated almost universally from measurements made in air, so tabulated values (as in Reviews 7, 8) are given for $p_o = 0.21$. From eqns. (1), (2), (3), and (6), it is clear that

$$K = M^2/\ln(1 - p_o) \quad (7)$$

Hence with a value for: M^2 of 225; and for $\ln(1 - p_o)$ of 0.235 when p_o is 0.21; this gives a value for $K_{(calc.)}$ of 957 sec/sq.cm. This is lower than (though close to) the value obtained in the previous experiments (5) for this same coal (previous value: - 1095 sec/sq.cm) but this may be accounted for in part by the difference between the logarithmic term $[\ln(1 - p_o)]$ and the first term of its expansion $[p_o]$ since the former has been used in this paper, but the latter was used in the previous paper (5); the theoretical alternatives are compared in the two eqns. (3b) and (3c). Use of the first term expansion as in eqn. (3c) is very common, and is generally accepted as being valid for

air or vitiated air. Just how widely the two terms differ at enriched concentrations, is shown by Fig. 4 in which the logarithmic term is plotted against p_o . In vitiated air the two are clearly reasonably comparable, but even in air itself the difference amounts to nearly 12% (0.235 compared with 0.21). Use of the \ln term in place of p_o in the previous experiments would therefore, reduce the value of 1095 to 978. The further difference between this and the new value of 957 is well within 5% but can in any case be attributed to uncertainty in the precise temperature in the two cases. Agreement, however, is regarded as acceptable.

The significance of the logarithmic term also showed up in the graphs of Figs. 1, 2, and 3. To check their sensitivity to the first expansion term in place of the full expression, similar plots were prepared (not reproduced) with p_o in place of $\ln(1 - p_o)$. The plots were found to vary significantly as follows: in equivalent Fig. 1, the plots showed slight but detectable curvature in spite of the fairly considerable scatter; in equivalent Fig. 2, convincingly straight lines could be run through the points, but the plots then showed marked intercepts on the oxygen-function axis, and the displacements from the origin were found to be statistically significant; finally, in equivalent Fig. 3, a straight line could again be run through the points, but again only with a statistically significant intercept. This agreement with the logarithmic term thus provides by far the best substantiation of the original Nusselt analysis in terms of the diffusion-film theory of reaction-rate control. What is yet undetermined, however, are the limits of applicability of the Nusselt equation and analysis; this is considered briefly in the next section.

5. DISCUSSION

- 5.1 Reaction Order - As stated in the Introduction, one of the principal reasons for carrying out the work described was to provide a more direct check on the assumed order of reaction at the solid surface. Now, because of the adjacent diffusion layer, that under these quiescent conditions is rate-controlling, it is only possible to check the surface order of reaction indirectly. To do this we assume some appropriate value for the surface order of reaction and then deduce what net, overall, or 'global' order of reaction should then follow. First of all, in choosing an order for the surface reaction, we have (7) two extreme limiting values: (1) zero, when the temperature is low enough for the surface chemisorption sites to be fully saturated at all times; and (2) unity, when the temperature is high enough for the sequence of chemisorption, followed by desorption, to be effectively instantaneous. At intermediate temperatures the reaction approximates to a fractional order. If internal or pore reaction also takes place, the lower limiting order is then raised from zero to 1/2.

Superimposed on this pattern is the oxygen supply by boundary layer diffusion. Now, in the first place, if the temperature is high enough for first order reaction to prevail then we get the burning time equation that combines both the diffusional and adsorption resistance (9),

$$t_b = K_c d_o + K_D d_o^2 \quad (8)$$

where K_c is the high-temperature chemical burning constant; and K_D is the

diffusional burning constant. This clearly has limits, respectively, of a linear equation, or a square law equation, according to whether diffusion is unimportant or dominant.

At the other extreme of low temperatures, when the reaction order is zero, the burning time equation is linear only (9)

$$t_b = K'_c d_o \quad (9)$$

where K'_c is the low-temperature chemical burning constant.

At intermediate temperatures the burning time is proportional to some intermediate power of the diameter, d_o^n , where n lies between 1 and 2. It is, therefore, obvious that determination of the power index n in any burning time experiments will give a clear guide to the relative importance of the three factors considered: (i) rate of diffusion; (ii) rate of chemisorption; and (iii) rate of oxide-film decomposition. In particular, a value of 2 for n is quite unambiguous in implying that the rate of boundary layer diffusion dominates the reaction control: and the significant corollary of this is that the surface rate-of-adsorption reaction must be first order.

In concluding that the reaction in our experiments was in the high temperature region, and diffusion controlled, we have altogether three confirmatory points provided by the experimental results. (1) The first is the 'square-law' agreement illustrated by the square-root plot of Fig. 3; if the additional chemical term in eqn. 8 — $K_c d_o$ — was also important, the line would be curved with a tendency to an intercept on the t axis. Other checks such as plotting (t_b/d_o) against d_o confirmed that K_c was negligible under the conditions of experiment. (2) We also have the agreement between t_b and the oxygen function $\ln(1 - p_o)$. Since K_c is inversely proportional to p_o (9), then if this was important the plot of t_b against $1/p_o$ would not have been so poor in comparison with the plot of Fig. 2 against $1/\ln(1 - p_o)$, as discussed in sec. 4.5. (3) There is finally the excellent agreement between the experimental and calculated values of the burning constant (written as K instead of K_D after correction for coal rank and swelling by eqn. 2) as described in sec. 4.5.

Since the reaction is evidently diffusion controlled, it follows that the surface reaction must be first order.

- 5.2 Boundary Layer Thickness - A secondary point of interest that also emerges from this is a reflexion on the question of boundary layer thickness. In calculating heat and mass transfer to spheres it is almost universal to use Nusselt's concept of the effective or 'fictitious' boundary layer thickness. If the oxygen concentration at the solid surface of the particle of radius, a , is zero, its value at any other radius, r , from the center of the particle is given approximately by

$$p/p_o = 1 - a/r \quad (10)$$

Since p rises to the main-stream value p_o only when r becomes infinite, the

real, physical boundary layer must clearly be of infinite thickness if it is defined as the distance required for p to reach the main-stream value. If, however, (following Nusselt (10)) the real behaviour indicated by eqn. (10) is replaced by an equivalent behaviour such that p is assumed to rise linearly to p_0 and then to remain constant with r , the main stream value is then reached at the Nusselt fictitious film thickness at $r = 2a$, or one radius out from the surface of the particle. It should be realised, however, that at $r = 2a$, $p = p_0/2$ (from eqn. 10); in other words, the actual, physical rise in p is only half the fictitious value. This can be represented in another way by relating the fictitious film thickness to a definable real, or physical film thickness. This can be defined with physical realism as the distance within which p rises to, say, 99% of the main stream value (this is a standard solution to continuum problems in which some relevant parameter reaches its limiting value only at infinity). We then have that, writing the boundary layer thickness as δ ,

$$\delta_{(\text{physical})} = 100a = 100\delta_{(\text{fictitious})}$$

The significance of this becomes immediately apparent when considering the behaviour of dust flames since the interparticle distance at a stoichiometric concentration is generally of the order only of 30 particle diameters. This means that in real, physical terms, as opposed to fictitious film-thickness terms, most of the oxygen is already well inside the boundary layer of one particle or another in the flame. It is clear, therefore, that direct extrapolation of the results in this paper to particles in dust flames should be made with caution.

Now, in addition to the points made above, a further complication that emerges is the additional inadequacy of the fictitious film concept, even for purposes of calculation, over the range of oxygen concentrations used in our experiments. Because our range was high, and the oxygen function $\ln(1 - p_0)$ could not be approximated by p_0 (as shown in Fig. 4), it means that eqn. (10) — which is also based on the same approximation of p_0 for $\ln(1 - p_0)$ — is also inadequate to describe the behaviour of the oxygen concentration over our full experimental range. An effective film thickness can still be defined, but the expression for it is so complex as to be valueless as it is then easier to solve the initial differential equation and not to bother about the 'short cut' of using an effective or fictitious film thickness.

- 5.3 Range of Applicability - The final point to be considered as a consequence of these results is their range of applicability: the temperature range of application is of particular importance.

Now, what we have established so far is that the solid surface reaction is first order at a temperature as low as 1000°C. This, however, was unexpected, being about 200° lower than the expected value of the 'higher critical temperature'. This was estimated, by assessment (7) of the available literature, at 1200°C. Below that, through the transition range down to 800°C, the reaction order was expected to drop progressively from unity to zero. The explanation for this apparent contradiction would appear to be a consequence of the influence of the

ambient gas velocity on the boundary layer thickness. The effect of ambient velocity is to promote such increased speed of mixing of the ambient gases that the thickness of the boundary layer, however defined, is progressively reduced. Initially, this must steepen the oxygen concentration gradient, with the result that both oxygen transfer, and therefore reaction rate, are increased. This increase can proceed just so far, up to the point that the rate of chemisorption exceeds the rate of reaction, and at that point the coverage of the solid surface by the chemisorbed film must increase; the surface reaction order then becomes fractional, and can drop progressively to zero as the ambient velocity also increases. This means that the transition range of temperature between the two extreme reaction conditions is velocity dependent. This dependence is shown very clearly by the single sphere experiments of Tu, Davis, and Hottel (12), but the analytical function relating the two is still unknown.

It would therefore seem that previously published experiments that were assessed as showing a first order reaction only down to 1200°C, cannot yet be directly correlated with the results of our experiments because the previous ones were carried out in flowing systems, whilst ours were in effectively quiescent systems. The increased film thickness that must have existed in our experiments can account qualitatively for the first order reaction down as low as 1000°C, but this inter-relation between temperature and velocity is now, in our opinion, the most outstanding problem of the combustion system requiring to be resolved by future work.

6. CONCLUSIONS

For coal particles inside the size range 350 microns to 2 m.m., burning in vitiated and enriched O₂ atmospheres (3% to 70%), at about 1000°C, under quiescent ambient conditions, the combustion behaviour was found to be as follows:

(1) Qualitative behaviour was, in general, similar to that observed previously with coal particles burning in air; for particles large enough, combustion proceeded in two sequential stages: (i) volatile evolution and combustion; followed by (ii) residue combustion.

(2) Particles below 1 m.m. tended to produce too small a quantity of volatiles for their combustion, because of low limit requirements. The frequency of this combustion failure tended to increase as the oxygen concentration was reduced. At 3% oxygen, even the residues failed to ignite. At high oxygen concentrations the particles ignited and burned satisfactorily, but they showed increasing tendency to deprecipitate or explode.

(3) Quantitatively, only the residue combustion was examined in detail. Burning times of the residues (t_b) were found experimentally to obey the relation predicted from the Nusselt diffusion theory of reaction control:

$$t_b = m / \ln(1 - p_o)$$

where p_o is the ambient oxygen concentration; and m is a predictable constant.

(4) The results also showed good agreement with the following predicted relation between the constant m and the initial particle diameter d_0 :

$$m = M^2 d_0^2$$

where M is another predictable constant.

(5) The experimental value of the second constant M was also found to be in good agreement with the predicted value, as calculated from the relation

$$M^2 = [(C_F/100)/f] \sigma / 3 \rho_0 D_0 (T/T_0)^{0.75}$$

where C_F is the fixed carbon of the coal; f is the swelling factor (of value 1.5 for bituminous coals); σ is the solid particle density; ρ_0 is the s.t.p. density of air; D_0 is the s.t.p. coefficient of oxygen diffusing through nitrogen; T is the absolute temperature and T_0 is the reference temperature of 273°K. The predicted and experimental values of M^2 were in fact identical, at 225; although this exact agreement was fortuitous it is still entirely satisfactory within the expected limits of error.

(6) The burning constant K in the Nusselt square-law equation

$$t_b = K d_0^2$$

was also calculated, being given by

$$K = M^2 / \ln(1 - p_0)$$

The value was 957 c.g.s. units, and this is in adequate agreement with values obtained previously for this same coal under similar (though not identical) experimental conditions.

(7) This general agreement with prediction therefore substantiates the primary assumption of the theoretical analysis: that the order of reaction at the solid particle surface with respect to oxygen concentration is unity. It also follows from the results that the rate of diffusion is the slow step that dominates the reaction rate, and that the importance of the chemisorption process is negligible under the conditions of experiment. There is background evidence from other previous experiments, however, indicating that this is true only for the fully quiescent system; and that, in a velocity field at these temperatures, the chemisorption process is likely to become significant, and increasingly so with increasing velocity. This is now the outstanding point requiring investigation in any subsequent work.

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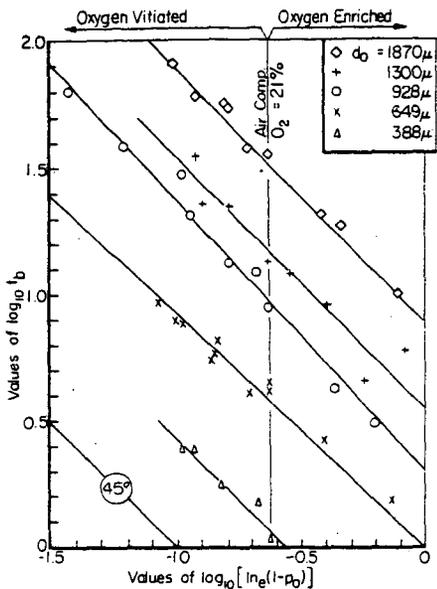


FIG. 1 - Double logarithmic plot of burning time, t_b , against the oxygen partial pressure function, $\ln_e(1-p_o)$.

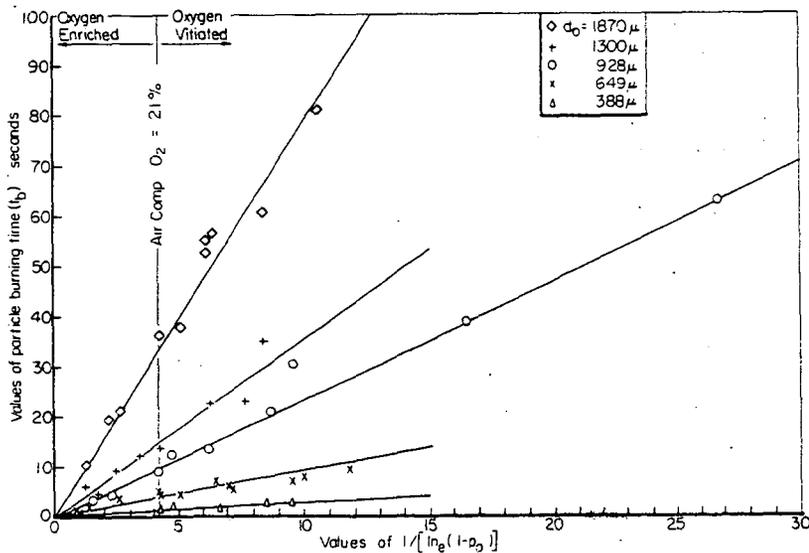


FIG. 2 - Linear plot of burning time, t_b , against the reciprocal of the oxygen partial pressure function: $\ln_e(1-p_o)$.

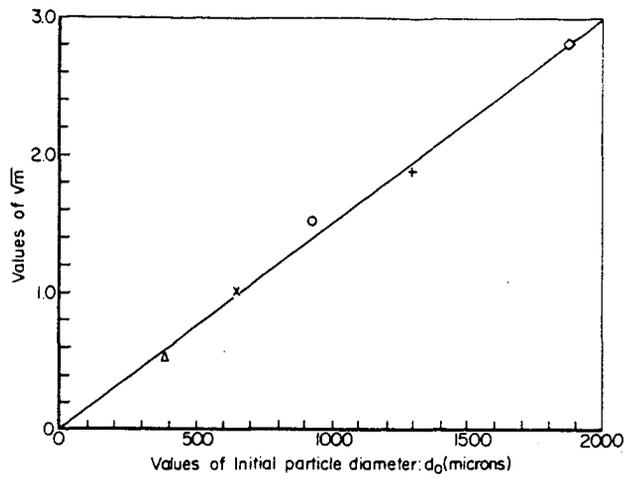


FIG. 3- Variation of square root of fig. 2 slopes (m) as a function of initial diameter of particle (d_0).

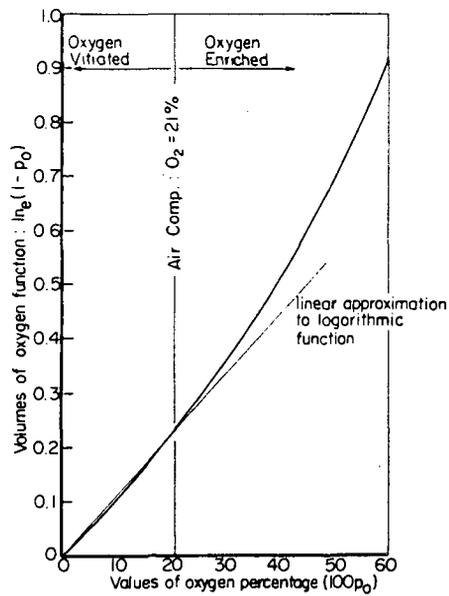


FIG. 4-Variation of oxygen function $\ln_e(1-p_0)$ with oxygen partial pressure p_0 .