

*Effect of Solid Surfaces on the Propagation of Flame through  
Ethylene-Air Mixtures.*

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Three types of experiment have been carried out to investigate the effect of solid surfaces on the propagation of flame through various ethylene-air mixtures. These are: (A) Measurements of minimum tube diameters for flame propagation. (B) Measurements of critical flow rates for flash-back down cylindrical burner tubes; from these critical velocity gradients at the wall and penetration distances have been calculated. (C) Observations of the products of combustion after explosion in a small combustion chamber; from the results it was concluded that a portion near the wall was not consumed by the flame. The results have been discussed separately and in relation both to one another and to some measurements by Holm and by Lewis and von Elbe.

THERE is much evidence that the ability of a flame to propagate is considerably affected by the presence of a surface. For instance, it has been found that a mixture which will carry a flame down a wide tube will not do so down tubes the diameters of which are less than a certain critical value; *i.e.*, when all points in the tube are within a certain distance of the wall no propagation of flame is possible. Holm (*Phil. Mag.*, 1932, **14**, 18; 1933, **15**, 329) and Garside, Hall, and Townend (*Trans. Inst. Gas Eng.*, 1941—42, **91**, 80), among others, investigated this phenomenon; and Friedman ("Third Symposium on Combustion Flame and Explosion Phenomena," Williams & Wilkins, 1949, p. 110) and others (Harris,

Grumer, von Elbe, and Lewis, *ibid.*, p. 80) have studied the similar limiting separation for propagation of flame in a space bounded by plane surfaces. Another effect that can be used to obtain information about the effect of adjacent surfaces is that of "strike-back" or "flash-back" in a burner. This has been treated theoretically by Lewis and von Elbe who deduce from the results a quantity which they call the "penetration distance" ("Combustion, Flames, and Explosion of Gases," Academic Press, 1951, p. 285; cf. Wohl, Kapp, and Gazley, "Third Symposium, etc.," *op. cit.*, p. 3), which, in their treatment, measures the distance from the surface to the point where the burning velocity is equal to that in free space, it being supposed that it is lower in the region near the surface than it is in free space. Similar ideas have also been put forward by Forsyth, Davies, and Townend (*Trans. Inst. Gas Eng.*, 1941—42, **91**, 47; see also, Garside, Forsyth, and Townend, *J. Inst. Fuel*, 1944—45, **18**, 175, and Forsyth and Garside, "Third Symposium, etc.," *op. cit.*, p. 99). These authors relate the distances between the base of the visible inner cone and the top of the burner tube, which they call the "dead space," with the distance to which a surface can affect flame propagation (*i.e.*, Lewis and von Elbe's "penetration distance"). Whether this is justified seems uncertain. A flame cannot be treated as a single surface, and light emission is late in the succession of events constituting a flame, as is shown, for instance, by the separation between the visible and Schlieren shadow cones (van de Poll and Westerdijk, *Z. tech. Physik*, 1941, **22**, 29). This "dead space" above the burner port is not observed if Schlieren shadow photography is used.

In the present paper some results that have been obtained for ethylene-air mixtures are presented. They are of three types: (A) Measurements of minimum tube diameters for flame propagation for a range of mixtures; (B) measurements of limiting flow rates for flash-back down cylindrical tubes; (C) observations of the products of combustion of ethylene-air mixtures after explosion in a small combustion chamber. These will be described and discussed separately in three sections, and a general discussion is given at the end.

#### SECTION A

*Experimental.*—The apparatus used is shown in Fig. 1. The volume above the mercury in *F* was filled with a known mixture by drawing it in from a flow line, the mixture having been made up by adjusting calibrated flow-meters. The ethylene was prepared by dehydrating ethyl alcohol with metaphosphoric acid, and the air was obtained from the compressed-air supply. The gas was forced out of *F* into *G* and through the test capillary *H*, the diameter of which had been obtained by weighing a mercury pellet of measured length. A flame was lit and then the flow stopped to see whether the flame would travel back down the tube or not. A side tube from *G* dipping into mercury acted as a safety valve when inflammation reached *G*, and a gauze sealed into *G* eliminated the danger of the flame's passing into *F*.

*Results.*—Six tubes were tested, and the limits of ethylene content in which they allowed passage of flame were as follows:

Radius of tube, mm. ....	0.880	0.948	0.968	1.033	1.126	1.197
Limits of C <sub>2</sub> H <sub>4</sub> (%) .....	(None)	7.0—8.15	6.65—8.40	6.0—8.65	5.55—9.0	5.1—9.5

These results agree well with those given by Garside, Hall, and Townend (*loc. cit.*). This is interesting, for the present results were obtained by using glass tubes, whereas their experiments were carried out with holes in metal plates only 0.43 mm. thick.

*Discussion.*—The results are shown graphically in Fig. 2, from which it will be seen that the minimum tube radius must be about 0.93 or 0.94 mm. for a mixture containing about 7½% of ethylene, which is the mixture having the maximum burning velocity as determined by the burner method (Schlieren) (Conan and Linnett, *Trans. Faraday Soc.*, 1951, **47**, 981) and the soap-bubble method (Pickering and Linnett, *ibid.*, p. 989). However, there is no simple relation between the burning velocity and limiting diameter. For instance for mixtures containing both 5 and 9.5% of ethylene the limiting radius is 1.22 mm., but the burning velocity for the former mixture is 43.5 cm./sec. as against 48 for the latter. Also the burning velocities of mixtures containing 6 and 9% of ethylene are both 56 cm./sec.; yet the limiting radius for the former mixture is 1.03 mm. and for

the latter 1.12 mm. That is, for mixtures having the same burning velocity the rich mixture requires wider tubes for propagation than does the lean mixture.

SECTION B

*Experimental.*—The ethylene and air flow-rates were measured on capillary flow-meters. The gases were mixed and passed up the cylindrical tube in which the flash-back limits were to be determined. Three tubes were used : two of glass of internal radii 0.398 and 0.484 cm., and one of steel of internal radius 0.494 cm. The procedure for determining the critical volume flow rate for flash-back for a known mixture in a given tube was as follows : A mixture was first passed through the tube at such a flow rate that it would produce a stable flame. The flow rate of the air was reduced until the flow rate and composition had changed to those of a mixture which just struck back. The flow rate was then made just a little greater than that for strike-back, and the flow rate of air again reduced. The flow rates of both gases at strike-back were again noted, and the process repeated. It was possible to fix the limiting flow rate for strike-back for a given mixture with an accuracy of  $\pm 1$  c.c./sec. in a total flow rate of the order of 100 c.c./sec.

*Results.*—The results are presented in terms of the critical velocity gradient in  $\text{sec.}^{-1}$  at the wall when flash-back occurs. The velocity gradient at the wall in a tube of radius  $R$  in which

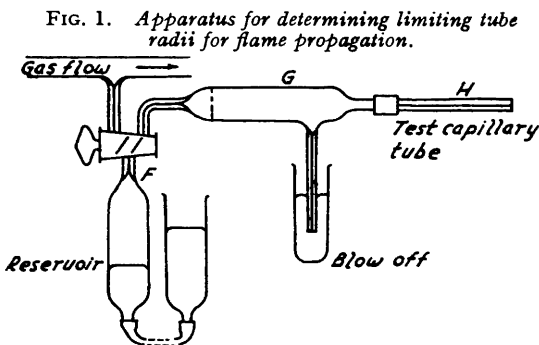
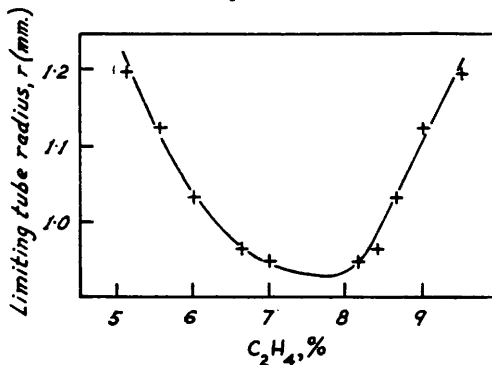


FIG. 2. Variation of limiting tube radii with composition.



the volume flow rate is  $V$  is  $4V/\pi R^3$ . This will be called  $v_g$ . The graph of the limiting value of  $v_g$ , which will be called  $v_g'$ , against the percentage of ethylene is shown for all three tubes in Fig. 3. It will be seen that  $v_g'$  is dependent only on the composition of the mixture and that it is a maximum for a mixture containing about 7½% of ethylene, which is the mixture having the maximum burning velocity. These results are consistent with those presented by Forsyth, Davies, and Townend (*loc. cit.*), whose experiments were carried out below atmospheric pressure and are given for a single burner tube.

*Discussion.*—In a tube through which gas is flowing in a stream-lined manner the linear flow rate is zero at the wall. Therefore, near the wall, the flow rate is always less than the burning velocity in free space. This led Lewis and von Elbe to suppose that the burning velocity was reduced by the presence of a nearby surface so that, when a stable flame was burning at the mouth of a cylindrical tube, the flow rate at any point exceeded the effective burning velocity there. When flash-back occurred, they supposed that the effective burning velocity exceeded the flow rate at some point. By following up this idea they were able to show that it was reasonable to suppose that the surface reduced the burning velocity below the value it had in free space (*i.e.*, affected the flame propagation to some extent) to a distance equal to  $V_b/v_g'$  from the surface, where  $V_b$  is the burning velocity. The values obtained for this quantity for different ethylene percentages (see Fig. 4) were : 5%, 0.78 mm.; 5½%, 0.63 mm.; 6%, 0.53 mm.; 7%, 0.46 mm.; 8%, 0.46 mm.; 9%, 0.49 mm.; 10%, 0.51 mm. The minimum penetration distance, as the above quantity is called, is about 0.46 mm. for a mixture containing about 7½% of ethylene. The curve is

very flat near the minimum, so it is difficult to place accurately the percentage at which the minimum lies.

Two facts may be noted about Fig. 4 in relation to the results of Section A. (i) For two mixtures having the same burning velocity, the penetration distance is less for the rich

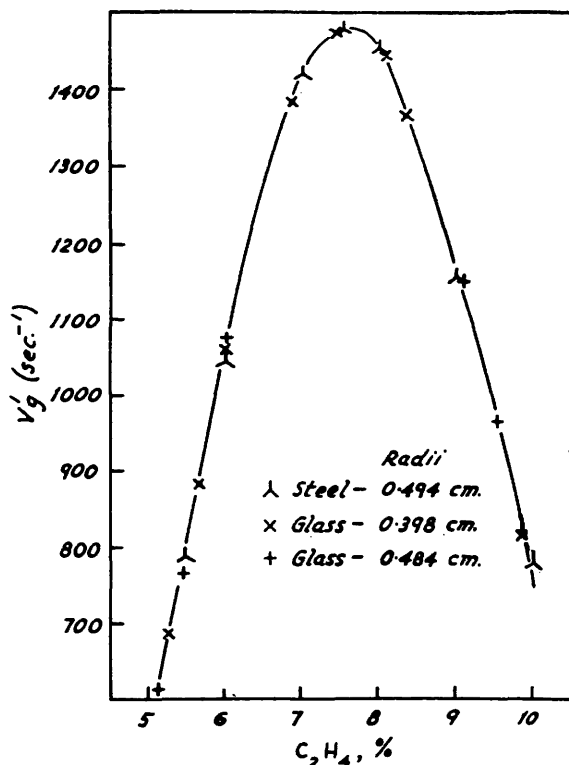
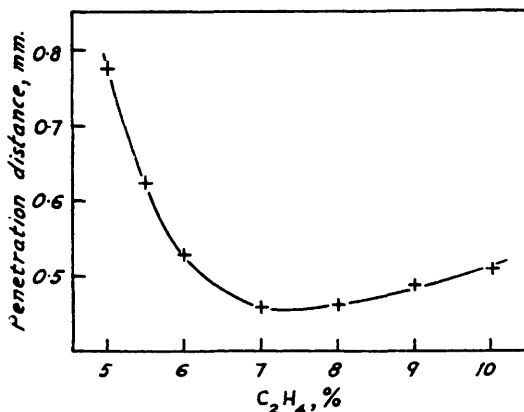


FIG. 3. Variation of limiting velocity gradient at the wall ( $v_g'$ ) with composition for three different tubes.

FIG. 4. Variation of penetration distance with composition.



mixture than for the corresponding lean one; this is the reverse of what would have been expected from Section A, for there it was found that, for two mixtures having the same burning velocity, the lean one could propagate flame in a tube the radius of which was less than the critical value for the rich one. (ii) The penetration distances of this section are less than the minimum radii of Section A. Since the penetration distance measures the distance to which some effect of the wall is experienced by the flame, whereas, in Section A, measurements were made of the tube radii necessary to extinguish the flame completely, this is perhaps surprising. The result is probably due, in part, to the fact that the

curvatures of the surfaces were greater in Section A than those used in the experiments described in this section. In the experiments of this section the radii of curvature of the surfaces were large compared with the penetration distances. It may also be due, in part, to the fact that the flame that travels down the narrowest tube possible must have itself a finite diameter and yet cannot approach too closely to the wall. A similar view has been expressed by Garside, Forsyth, and Townend (*loc. cit.*), who say, "the opinion has been formed that the limiting diameter in any particular case approximates to twice the dead space plus an allowance for the actual passage of the flame."

SECTION C

*Experimental.*—Various mixtures of ethylene and air of known composition were enclosed in a tube of radius 5 mm. over mercury and ignited by means of a spark. The volume of the

FIG. 5. Observed variation of the percentage composition of the products with the initial percentage of ethylene.

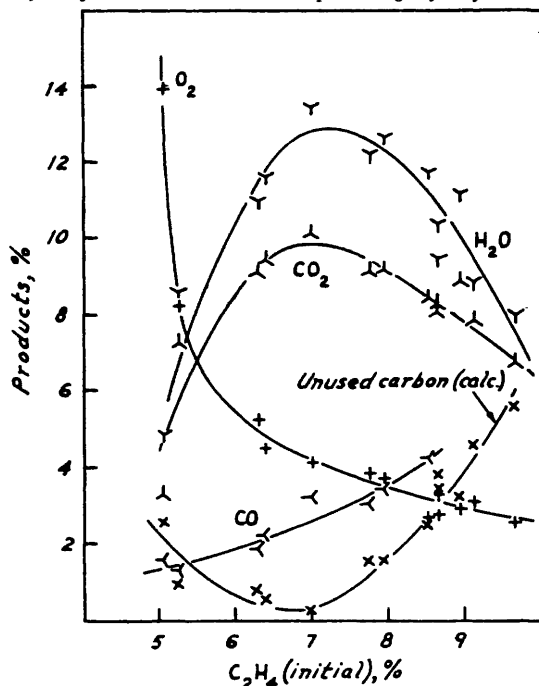
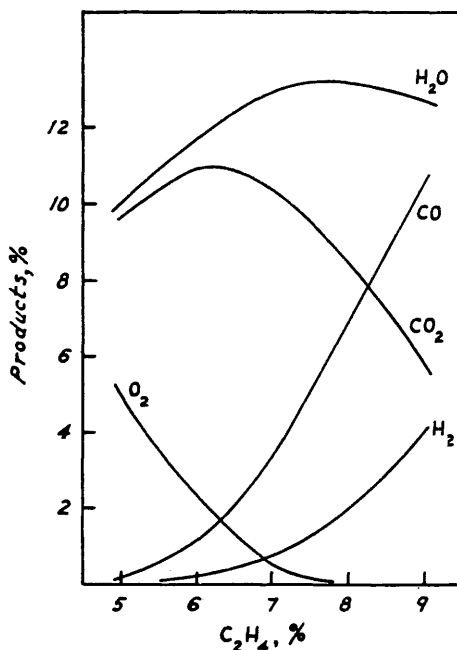
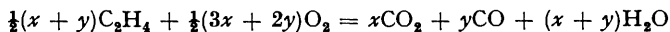


FIG. 6. Calculated variation of the percentage composition of the products with the initial percentage of ethylene.



mixture had been measured before ignition and the volume remaining was also measured. Both these measurements were made at room temperature and at the same humidity. The volumes of carbon dioxide and oxygen in the mixtures resulting from the explosions were measured by absorbing the former in potassium hydroxide and the latter in alkaline pyrogallol.

It was presumed that the combustion of the ethylene could be represented by :



where  $x$  and  $y$  represent volumes to be determined. The contraction on explosion,  $\frac{1}{2}(2x + y)$ , was measured as well as the volume,  $x$ , of carbon dioxide; so  $y$  could be deduced. In this it is assumed that all the hydrogen goes to water. This is a reasonable assumption for lean mixtures but is less good for rich ones, so our conclusions regarding the composition of the gases will be more satisfactory for lean than for rich mixtures.

*Results.*—The results are presented in Fig. 5. The percentages of carbon dioxide and monoxide in the products were determined as above (the percentages plotted in Fig. 5 are those that would be formed if the water remained gaseous—they can be readily calculated from those measured by assuming the above equation to be true). The percentage of water formed was deduced from the values of  $x$  and  $y$  which were measured. The percentage of oxygen remaining

was determined. The value calculated for this from  $x$  and  $y$  was found to be in good agreement with that observed. The amount of residual ethylene was calculated on the assumption that all that was consumed went to form the carbon dioxide and monoxide which had been determined. The experimental results in Fig. 5 may be compared with those in Fig. 6 calculated on the assumption that the products existed in equilibrium at an equilibrium flame temperature. The calculated and experimental curves for water, carbon dioxide, and oxygen agree in general form with one another, though the experimentally observed changes in the percentages with initial composition are greater than those calculated (*e.g.*, the maxima are sharper). The experimental curve for carbon monoxide is similar to the theoretical one from 5 to 7% but beyond this the percentage of carbon monoxide inexplicably decreases somewhat (points not shown). There is no theoretical curve for unburnt ethylene, for, by assuming equilibrium among the products, it is, as a consequence, assumed that all the ethylene is consumed.

*Discussion.*—The percentage of unburnt ethylene is a minimum for a mixture containing initially 7% of ethylene, which is the one having the highest calculated and the highest observed flame temperature. At both higher and lower initial percentages of ethylene the

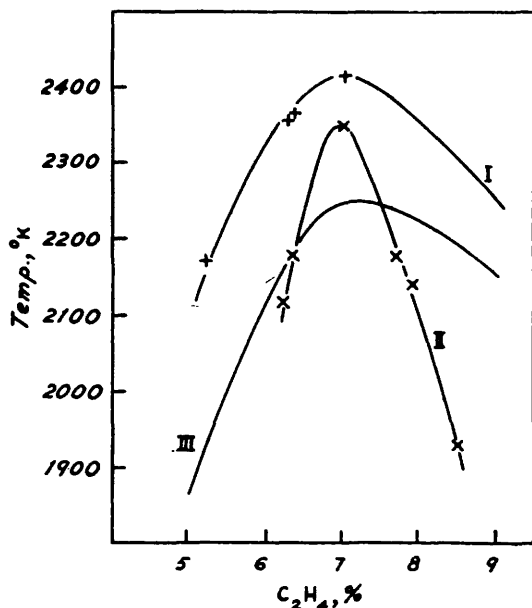


FIG. 7. *Variation of flame temperature with composition.*

The experimental curve (III) obtained by the sodium-line reversal method is shown together with curves calculated for equilibrium (I) and calculated from the analysis results (II). The temperatures calculated when allowance is made for edge correction are shown by +.

percentage of unburnt ethylene in the products appears to increase. That there should be unburnt ethylene in the rich mixtures is understandable, but the reason for its presence in the lean mixtures is not so obvious: the most reasonable explanation of this (and for part of the unburnt ethylene in the products from rich mixtures) is that there is a region near the wall which is not reached by the flame and that, in this space, the ethylene remains unburnt. This would explain why the percentage of unburnt ethylene is a minimum for the hottest flame.

Let us suppose that there is a region near the wall in which the ethylene is not burnt at all, while in the rest of the vessel it is all burnt. Knowing the amount of ethylene present originally, it is possible, from the calculated amount of ethylene remaining unburnt, to deduce the proportion of the volume of the combustion chamber which is unaffected by the passage of the flame. Since the sides of the tube account for the majority of the surface area (the diameter being much less than the length) this proportion may be put equal to  $2\pi r \Delta r / \pi r^2 = 2\Delta r / r$ , where  $r$  is the radius of the tube (5 mm.) and  $\Delta r$  is the distance from the wall to which the flame is supposed to be quenched. For the experiment with 6.97% of ethylene,  $\Delta r$  was calculated to be 0.09 mm.; for 6.37%, 0.23 mm.; for 6.28%, 0.33 mm.; for 5.24%, 0.48 mm.; and for 5.01%, 1.2 mm. The last figure is probably abnormally large, for the results in Fig. 5 indicate that, in this case, there was a particularly incomplete

combustion compared with those for higher ethylene percentages. The figure for 5.01% of ethylene being omitted, therefore, it will be seen that the values of  $\Delta r$  are less than the corresponding values for the quenching radius and penetration distance. This is not surprising, for it would be expected that after-burning, following the passage of the flame, would consume gas near the wall which had not been burnt during the actual passage of the flame. The ratio of  $r$  to the penetration distance varies from one-half to one-fifth on passing from 5 to 7% of ethylene.

Before concluding this section some calculations of the flame temperatures will be presented. In Fig. 7 are shown the flame temperatures calculated by assuming the products to be at equilibrium, and also the flame temperatures observed by the sodium-line reversal method (Jones, Lewis, Friauf, and Perrott, *J. Amer. Chem. Soc.*, 1931, 53, 869). The third curve is a flame-temperature curve calculated from the analysis results presented in Fig. 5. This was calculated from a knowledge of the heat liberated on going from the initial mixtures to the product mixtures and also the heat capacities of the various substances. This curve was obtained by considering the overall composition of the product mixture, no allowance being made for any wall effect. The four points marked by crosses (+) in Fig. 7 were calculated from the results for 5.24, 6.28, 6.37, and 6.97% of ethylene on the assumption that the part within  $\Delta r$  of the wall was unaffected by the flame (see above) and also that the composition of the products in the remainder of the combustion tube could be obtained by subtracting the initial contents of this volume from the products found (see Fig. 5) to obtain the contents, after explosion, of that part of the vessel which was not within  $\Delta r$  of the wall. The calculation of the final temperature was then made from these results in the manner outlined above. It will be seen that these points lie very close to the theoretical curve. This provides support for the simplified picture we have formulated of the effect of the walls on the gases during the explosion.

#### GENERAL DISCUSSION

Two further matters will be discussed here. The first is the relation between our results and some obtained by Holm (*Phil. Mag.*, 1932, 14, 18; 1933, 15, 329). If the limiting velocity gradient at the wall ( $v_g' = 4V/\pi R^3$ ) is independent of the tube radius then, for two tubes of radius  $R_1$  and  $R_2$ , the limiting flow rates for flash-back,  $V_1$  and  $V_2$ , must satisfy the relation  $V_1/V_2 = R_1^3/R_2^3$ . Since, for several ethylene-air mixtures, the limiting volume flow rates for a tube of a particular radius are known, and since, for the same mixtures, the limiting tube radius is also known, it is possible to calculate the limiting volume flow rate for flash-back in the tube of limiting radius. Holm used this to calculate "the velocity of propagation" of the flame in the tube of limiting diameter from the above formula and  $v_f = V/\pi R^2$ , where  $v_f$  is the velocity of propagation and  $V$  is the volume flow. The values of  $v_f$ , deduced in the above way, are plotted against the percentage of ethylene in Fig. 8, and the values of  $v_f$  for 5, 6, 7, 8, and 9% of ethylene are plotted in Fig. 9 against the limiting tube radii for these mixtures, together with some results obtained for other mixtures by Holm. It will be seen that, despite the variety of substances studied, most of the points lie close to a straight line, showing an unexpected regularity in behaviour. This, in effect, suggests that there is a smooth relationship between the critical velocity gradient for flash-back and the limiting tube radius for flame propagation which is independent of the combustible mixture, the relation being such that, as one becomes bigger the other becomes smaller.

A further point arises out of one already mentioned. In Section B it was pointed out that the limiting tube diameter could be regarded as being made up of two parts: (a) a flame diameter which was the minimum for which flame could propagate, and (b) the space between this and the wall in which the flame could not affect the gas because of the quenching effect of the wall. The fact that twice the penetration distance, as determined from the critical velocity gradient for flash back, was always less than the minimum tube diameter supports this concept.

Now, for various methane-oxygen-nitrogen mixtures, Lewis and von Elbe (*loc. cit.*, pp. 418, 426) have recorded the minimum tube diameter ( $d_0$ ) and minimum distance

between parallel plates ( $d_{11}$ ) for flame propagation. They have also recorded the penetration distances ( $d_p$ ) calculated from the critical velocity gradients and, in addition, have calculated from minimum energies for spark ignition values for the diameter ( $d_f$ ) of the smallest sphere of flame that will carry inflammation into the next layer. Values for these quantities, as given by Lewis and von Elbe, are listed in the following Table. In addition, two columns list the values of  $\frac{1}{2}(d_0 - d_f)$  and  $\frac{1}{2}(d_{11} - d_f)$ . These figures may be compared with  $d_p$ , the penetration distance, for they may be regarded as measuring the distance within which the quenching effect of the walls is operative. It will be seen that, in most cases,  $d_p$  lies between  $\frac{1}{2}(d_0 - d_f)$  and  $\frac{1}{2}(d_{11} - d_f)$ , which is reasonable, for the radius of curvature of the surface used when measuring  $d_p$  must have been intermediate between

FIG. 8. Variation of the calculated velocity of flame propagation in tubes of the limiting radius with composition.

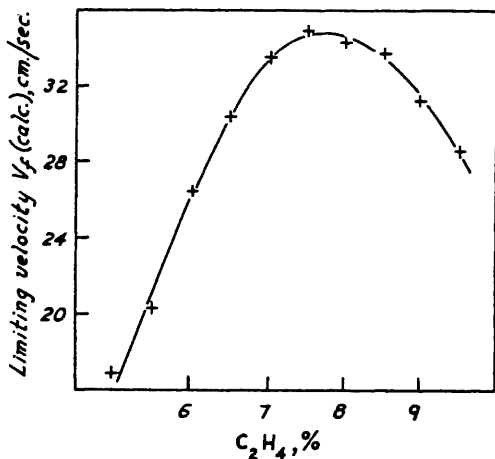
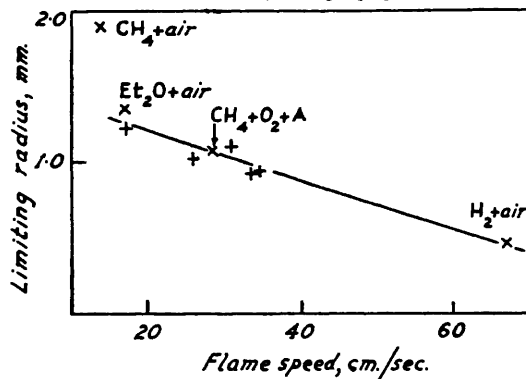


FIG. 9. Variation of the limiting radius with the limiting velocity of flame propagation.



Holm's results are shown as  $\times$ , ours as  $+$ .

the radius of the surface of the tube ( $d_0/2$ ) and that of the plane surface ( $\infty$ ). Moreover, except in one case for which  $d_f$  appears to be abnormally large, when it is outside the limits it is only just outside. The general idea of a small flame going down the centre of the tube cushioned from the wall by an annular ring of gas seems to be satisfactory. The results in the Table moreover lend support to the values of  $d_f$  calculated by Lewis and von Elbe.

*Lewis and von Elbe's values for minimum tube diameters ( $d_0$ ), minimum separation between plates ( $d_{11}$ ), minimum flame diameters ( $d_f$ ), and penetration distances ( $d_p$ )—in mm.—for methane-oxygen-nitrogen mixtures.*

Percentages of:			$d_0$	$d_{11}$	$d_f$	$d_p$	$\frac{1}{2}(d_0 - d_f)$	$\frac{1}{2}(d_{11} - d_f)$
CH <sub>4</sub>	O <sub>2</sub>	N <sub>2</sub>						
10	90	—	1.18	0.86	0.38	0.40	0.40	0.24
15	85	—	0.68	0.46	0.17	0.14	0.26	0.15
25	75	—	0.48	0.28	0.16	0.066	0.16	0.06
40	60	—	0.69	0.43	0.32	0.11	0.19	0.06
50	50	—	2.15	1.5	1.06	0.41	0.55	0.22
52.5	47.5	—	3.25	2.2	1.92	1.0	0.67	0.14
10	18.8	71.2	3.35	2.8	0.90	0.90	1.23	0.95
16.3	29.3	54.4	1.42	0.79	0.35	0.44	0.54	0.22
21.5	39.8	39.7	0.92	0.53	0.31	0.19	0.31	0.11
26.4	49.1	24.5	0.70	0.45	0.30	0.11	0.20	0.08

However, it should be remembered that the values of the burning velocity used by Lewis and von Elbe to calculate  $d_p$  and  $d_f$  were not very reliable for they were obtained by using the Gouy total area burner method and the visible cone. This method is known to give low values.