401. *Studies in the Mechanism of Flame Movement. Part II. The Fundamental Speed of Flame in Mixtures of Methane and Air.*

By H. F. Coward and F. J. Hartwell.

The movement of flame in a gaseous explosion may be resolved into (i) the transmission of the zone of chemical reaction from layer to layer of the medium, and (ii) the mass movement of the medium itself at the flame front. Except, perhaps, during detonation, the latter component is almost always prominent, for over the whole period of inflammation the products of combustion create currents by their expansion during formation and by convection after formation. Hence the movement of flame into a medium free from mass movements is not, as a rule, directly observable. We are now able to show, however, that the speed of flame moving into a stationary mixture (the expansion and consequent movement of gas occurring behind the flame front) may be deduced from observations on the movement of flame in tubes.

In the early stages of a methane–air explosion started at the open end of a tube, closed at the far end, there is a phase of uniform movement during which, as our many photographs show, the shape and the size of the flame are constant. As the mixture ahead of the flame is at constant temperature and pressure, the volume of gas burnt by the flame in passing from one position to another is easily calculated. A series of snap-shot photographs of flame in uniform movement, taken on the same plate at known intervals of time, enabled the amount of gas burnt in unit time and the area of the flame to be calculated. The ratio between these quantities proves to be constant for any one mixture of methane and air.

EXPERIMENTAL.

Fig. 1 shows successive positions of the flame, at intervals of 0·036 sec., when a 10% CH_4–air mixture (dried by $CaCl_2$) was ignited at the open end of a tube, 2·5 cm. in diam. The flame was at first almost hemispherical and symmetrical about the axis of the tube, but, under the influence of convection, became tilted forward until it assumed a stable form, reproduced while it remained within the field of view of the camera. Its speed was then const., 71·5 cm./sec., in the direction of the axis of the tube, and it consumed 351 c.c. of the mixture per sec. at room temp. and press.

The area of the flame surface during the uniform movement was calc. on the assumption that it was approx. equal to that of a semi-ellipsoid, the axes of which were equal to (1) a line joining the points where the flame touched the top and bottom of the tube, (2) twice the perpendicular distance between that line and the point most remote from it on the photograph of the flame front, and (3) the diameter of the tube.

The area of the half-surface of the ellipsoid was calc. by the following formula, which was derived from the integral form given by Jellett (Williamson, "Integral Calculus," 1891, p. 283):

$$\pi b(a^2 - c^2)^{\frac{1}{2}} . E(k,\phi) + \pi bc^2(a^2 - c^2)^{-\frac{1}{2}} . F(k,\phi) + \pi c^2$$

where a, b, c, are the semi-axes, $a > b > c$; $k = e'/e$; $\phi = \sin^{-1}e$; e and e' are the eccentricities of the ellipses in the plane of the axes a and c, and b and c, respectively. The values of $E(k,\phi)$ and $F(k,\phi)$, the elliptic functions of the second and first kind respectively, are obtained from standard mathematical tables. (This formula was generally used for subsequent calculations, but simpler formulæ were used when, as often observed in vertical propagation, the flame surface was symmetrical about the axis of the tube.)

The area thus calculated from measurements on Fig. 1 was 12·56 sq. cm. Hence the vol. of gas burnt per sec. per sq. cm. of flame surface was 351/12·56 = 27·9 c.c. Similar determinations for the same mixture contained in tubes of two and four times the diam. (Figs. 2 and 3) gave approx. the same figure for the vol. of gas burnt per sec. per sq. cm. of flame surface, whereas the axial speeds in the three tubes were 71·5, 92, and 111 cm./sec. respectively, and the areas of the flames were 12·6, 66, and 300 sq. cm. As the ratios of wall surface to volume, for the three tubes, were as 4 : 2 : 1 respectively, it would appear that the flame itself was not sensibly cooled by the walls of the tube.

Further confirmation of the constancy of the ratio, R = *vol. burnt per sec./area of flame*, was obtained from Figs. 1—3. Shortly after ignition, and before the uniform movement was established, two successive flames can be chosen of nearly equal area. The distance between them is less than that during the uniform movement and their areas are less, but the ratio (see Table I) calc. for this " initial stage " is the same as that for the uniform movement (U.M. in table). Even more striking are the results calc. from Fig. 4, the record of an expt. in which the mixture was ignited low down at the open end of the 2·5-cm. tube. The flame was at first tilted back, " Initial (i) " in the table, then became upright (almost hemispherical), " Initial (ii)," but the ratio remained constant.

A critical test of the constancy of the ratio R was provided by observations in a vertical tube, with propagation of flame upwards or downwards (Figs. 5—8). In either direction it was possible to obtain either a symmetrical (Figs. 5 and 7) or an unsymmetrical flame (Figs. 6 and 8) of greater area and speed, but the vol. burnt per sec. per sq. cm. of flame surface was in each instance the same as for horizontal propagation, within the limits of experimental error (Table I).

TABLE I.

10% *Methane–air mixture.*

(U.M. = Uniform movement.)

Tube diam., cm.	Direction and stage of propagation of flame.		Axial speed of flame, cm./sec.	Area of flame, sq. cm.	R.
10	Horizontal	U.M.	111	300	29
10	,,	Initial	71	189	29
5	,,	U.M.	92	66	27
5	,,	Initial	61·5	48·5	25
2·5	,,	U.M.	71·5	12·6	28
2·5	,,	Initial (i)	63	11·0	28
2·5	,,	Initial (ii)	59	10·4	28
5	Upward	Symm.	68	48	28
5	,,	Unsymm.	92·5	66·5	27
5	Downward	Symm.	61	46	26
5	,,	Unsymm.	85	63·5	26
					Mean 27

In all the expts. recorded in Table I, the value of the ratio lies between 25 and 29 cm./sec. for a very wide range in the variables. Closer agreement is not to be expected, in view of the possible errors in the method of measuring the areas of the flames.

Flame areas of the 10% CH_4–air mixture in wider tubes could not be measured, for, as previously shown (Fig. 6, this vol., p. 1996), the flame-front was too complex. If, however, it be assumed that the ratio R remains the same for wider tubes, then the area can be calc. from the axial speed of flame. Thus the area of the complex flame surface in a 24-cm. diam. tube was calc. to be 40—50% greater than the area of the envelope surface, and some 5 times the area of the cross-section of the tube.

Snapshot photographs of explosions of 10% methane–air mixtures. (*Reproduction to scale.*)

'FIG. 1.—*Horizontal propagation* (*also Figs.* 2, 3, 4).

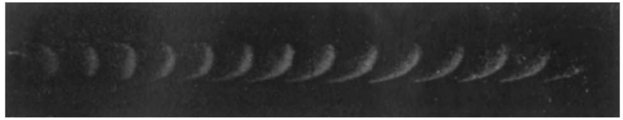

FIG. 2.

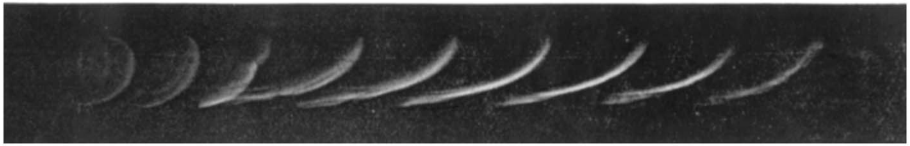

FIG. 3.

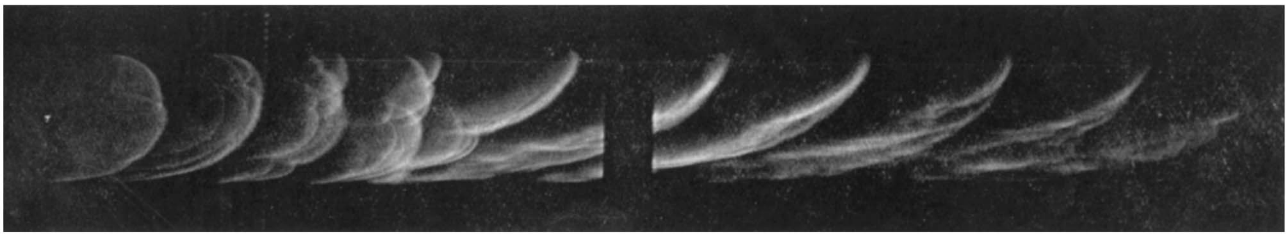

FIG. 4.

(See overpage for references.)

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Upward propagation of flame.

FIG. 5.

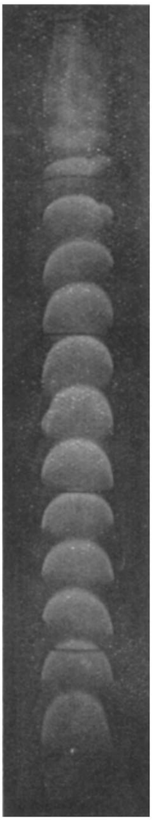


FIG. 6.

Downward propagation of flame.

FIG. 7.

FIG. 8.

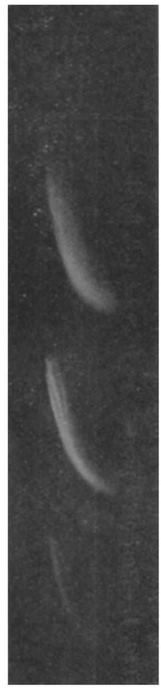


Figure	1	2	3	4	5	6	7	8
Diameter of tube, cm.	2·5	5	10	2·5	5	5	5	5
Time-interval between exposures, milliseconds	36	73	97	36	48	48	73	146

The Fundamental Speed of Flame.

The constancy of the ratio *R* for the 10% CH_4–air mixture at const. temp. and press. just ahead of the flame shows that each element of the flame surface consumes the same amount of mixture in unit time. This might have, and indeed has, been accepted without proof, but for a difficulty illustrated by Figs. 9 and 10. The former shows diagrammatically the successive positions of flame in uniform movement along a horizontal tube, and suggests that an element E at the nose of the flame-front consumes a greater vol. of mixture than an element E′. If each element consumes the same amount of mixture, the flame-front should take the successive positions shown in Fig. 10, and should ultimately become a flat vertical disc.

The difficulty disappears when it is recognised that convection due to the hot products of combustion not only tilts the flame but also causes a continued small movement of the gases in the tube in and about the flame: forward in the upper part of a horizontal tube, backward in the lower part. This effect is inevitable, and was made visible by charging the mixture with smoke just before an experiment. During the uniform movement of flame the shape of the flame-front is maintained constant by a balance between the two effects: (1) the tendency to straighten up, illustrated by Fig. 10, and (2) the tendency to tilt forward, due to convection.

FIG. 9.

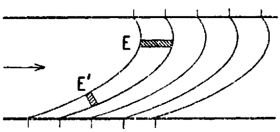
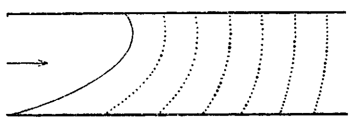


FIG. 10.



It is concluded, therefore, that the ratio *R* is the linear speed of flame, in a direction normal to its surface, through a gaseous mixture at rest and at const. temp. and press. just ahead of the flame. We suggest that this be described as the "fundamental speed of flame," for there are objections to the terms "normal speed" (Le Chatelier), "ignition speed" ("Entzündungsgeschwindigkeit"; Bunte *et al.*), and "absolute speed" (this vol., p. 1996), each of which has been used with the same meaning.

Measurements of the Fundamental Speed of Flame in the Range of Explosive Mixtures of Methane and Air.—The foregoing results were obtained with the fastest-burning mixture of CH_4 and air (10%). The flames of the slow-burning limit mixtures are low in actinic value, but fairly good photographs were ultimately obtained on Ilford "Golden Iso-Zenith" plates (H. & D. 1400), developed in complete darkness with a strongly alkaline developer to a point just short of fogging. The "fundamental speeds" deduced for two of the slowest-burning mixtures are given in Table II. For each mixture the speeds lie between about 6 and 8 cm./sec., which may be regarded as satisfactory in view of the wide range of the variables and the photographic difficulties. The result is lower than the former estimate, 10 cm./sec. (this vol., p. 1996), which, being based on the appearance of the flame, is less reliable than the present determinations.

Table III gives the measurements and the deduced fundamental speeds of flame for the whole range of inflammable mixtures of CH_4 and air. The expts. for this series were limited to horizontal propagation in a tube of

Table II.

Slow-burning mixtures.

(Tube diameter = 5·0 cm.)

CH_4, %.	Directn. of propagn. of flame.	Axial speed of flame, cm./sec.	Area of flame-front, cm.^{2}.	" Fundamental speed," cm./sec.
5·98	Upward	28·2	69·8	7·9
5·97	Horizontal	36·7	114·8	6·3
5·98	Downward	9·2	24·2	7·4
13·54	Upward	27·3	67·3	7·9
13·54	Horizontal	29·6	99·3	5·8
13·54	Downward	9·1	24·9	7·2

5-cm. diam., but *a priori* the " fundamental speeds " are independent of the direction of propagation and of the diameter of tube.

Table III.

Fundamental speeds of flame in methane–air mixtures.

CH_4, %.	A.S.F.,* cm./sec.	A.F.-F.,* sq. cm.	F.S.,* cm./sec.	CH_4, %.	A.S.F., cm./sec.	A.F.-F., sq. cm.	F.S., cm./sec.
5·89	35·3	117·5	5·9	9·95	95·6	70·4	26·7
5·97	36·7	114·8	6·3	10·00	91·8	66·2	27·2
6·55	43·4	81·4	10·5	10·27	94·0	68·9	26·8
6·71	48·1	85·2	11·1	10·68	87·8	69·2	24·9
6·80	47·9	86·9	10·8	10·95	81·9	68·0	23·6
7·77	65·5	70·0	18·4	11·27	75·2	65·4	22·6
8·53	79·1	68·9	22·6	11·77	60·9	62·3	19·2
8·61	84·3	66·6	24·9	12·32	48·9	67·2	14·3
9·23	87·0	69·1	24·7	12·64	39·7	71·2	10·9
9·31	91·0	65·6	27·2	12·92	35·9	77·2	9·1
9·61	92·9	68·0	26·9	13·54	29·6	99·3	5·8

* A.S.F. = Axial speed of flame; A.F.-F. = area of flame-front; F.S. = " fundamental speed."

Fig. 11 shows that, although the fundamental speeds of flame for this series of mixtures are smaller than the speeds of propagation along tubes, the curve is similar to those for tubes (this vol., p. 1996). It has a flat max. at about 9·5—10% CH_4, the mixture for complete combustion containing 9·46% CH_4. The least speed at which the slowest-burning mixtures can propagate flame from layer to layer is about 6 cm./sec.

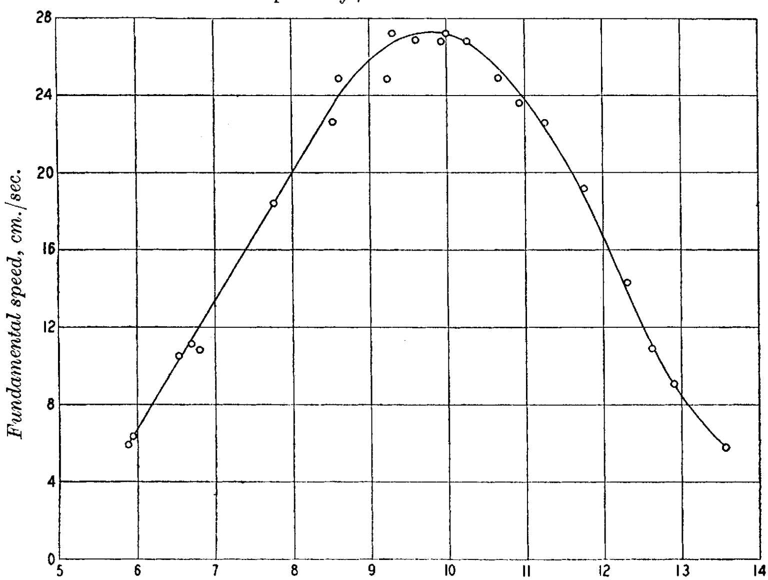
The left limb of the curve is apparently a straight line nearly as far as the mixture for complete combustion. It shows that the speed of flame, in mixtures containing excess of air, is not proportional to the CH_4 content of the mixture or to the heat produced on combustion; but is proportional to the excess of CH_4 above about 5%, and is therefore proportional to $t - t'$, where t is the temp. of the products of combustion and t' is a const. which may be the instantaneous ignition temp. in the conditions of the expts. (Le Chatelier).

Gouy's Method of determining the Fundamental Speed of Flame by Observations on the Inner Cone of the Bunsen Burner.

Hitherto only one method has been applied, with any success, to determine the fundamental speed of flame. Gouy (*Ann. Chim. Phys.*, 1879, **18**, 5) argued that the speed, V, of a flame, normal to its surface and relative to the gaseous mixture in which it was propagated, could be calculated from the speed of flow of the mixture and the dimensions of the inner cone of a Bunsen burner. When the cone is stationary, the speed V at any point is equal to the normal component to the surface of the speed v of the gas striking it at an angle a, so that $V = v \sin a$; and furthermore, V is equal to the total vol. of gas issuing from the tube in unit time, divided by the area of the cone. Michelson (*Ann. Phys. Chem.*, 1889, **37**, 1), Mache (*Sitz. Akad. Wiss. Wien*, 1904, **113**, 341), Bunte *et al.* (*J. Gasbel.*, 1913, **56**, 1225; 1914, **57**, 733; 1916, **59**, 49; 1928, **71**, 673; 1930, **73**, 837), Passauer (*ibid.*, 1930, **73**, 313),

FIG. 11.

Fundamental speeds of flame in methane–air mixtures.

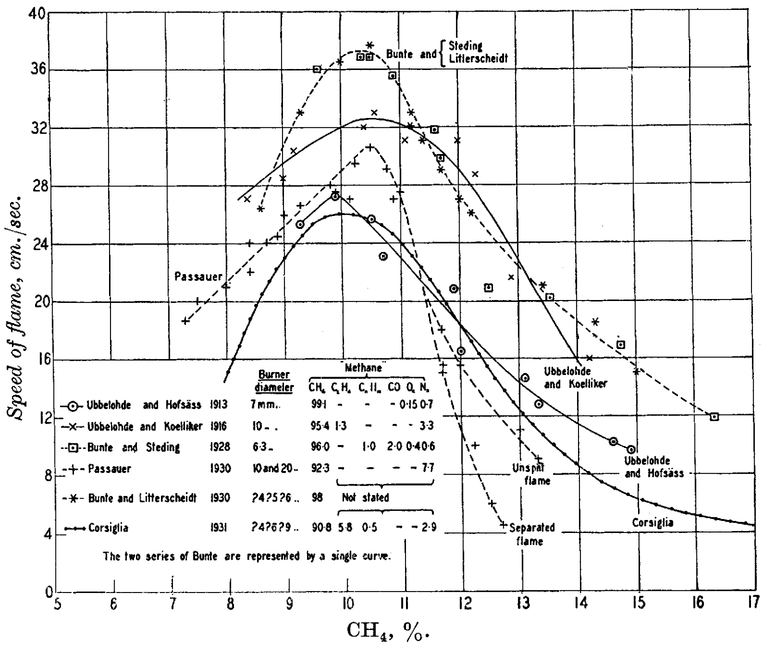


and Corsiglia (*Amer. Gas Assoc. Monthly*, 1931, **13**, 437) have discussed and somewhat elaborated Gouy's argument, but have retained his conclusions.

The various results obtained for CH_4–air mixtures by this means are shown in Fig. 12, for comparison with Fig. 11. The considerable differences between the curves of Fig. 12 are obviously not due solely to differences in the comp. of the "methane" used. Without attempting to discover the errors of experiment or interpretation which give rise to such differences, we may make a rough comparison between the results of Fig. 12 and those of Fig. 11, as follows. (1) The speed of the fastest-burning mixture is variously given between 26 and 37 cm./sec. by the Gouy method; 27 cm./sec. by our method. (2) The composition of the fastest-burning mixture is 10—10·5% CH_4 (Gouy), which is somewhat higher than our figure. (3) The left limbs of the curves end abruptly at some mixture much above the lower limit. (4) The right limbs of four curves are extended beyond the higher limit of inflammability.

Those who have used the Gouy method are well aware of an imperfection which explains (3) and (4) above, *viz.*, the impossibility of avoiding the entry of some of the external atmosphere at the base of the flame cone. In consequence of this it has been found impossible to maintain a flame on a Bunsen burner fed with mixtures containing less than about 7·3% of CH_4, even with an unusually wide burner; and mixtures which in themselves are too rich to propagate flame become diluted with air and can burn, or, if air is excluded, the burner gases take in some of the products of combustion and the flame speeds are too low. The reason for (2) above is also to be found in the entry of air at the base of the cone.

FIG. 12.



Michelson (*loc. cit.*) gave five reasons for not accepting the results of tube experiments as capable of giving the fundamental speed of flame. These need not be repeated; the method now used counters his difficulties and gives results that (1) are of the same order as those obtained by Gouy's method where comparison is possible, (2) are extended to mixtures which cannot be examined by Gouy's method, and (3) are accurately related to the composition of the mixture, thereby incidentally avoiding the error of ascribing a flame speed to a range of mixtures above the higher limit of inflammability.

SUMMARY AND DISCUSSION.

The Fundamental Speed of Flame Propagation.—During the uniform movement of flame in mixtures of methane and air, the volume of gas burnt in unit time is proportional to the area of the flame-front. This is true for explosions in tubes of diameter from 2·5 to 10 cm., and also probably outside these limits; it holds for any direction of propagation of flame. The area of the flame-front is determined by the diameter of the tube and by convection currents set up by the difference in densities between the burnt and unburnt gases. The flame is rarely, if ever, a flat disc at right angles to the axis of the tube; hence the axial speed of propagation is usually much greater than the normal speed of propagation (at right angles to the flame-front) of a plane flame-front in a stationary mixture. Values for this fundamental constant have, however, been deduced (Fig. 11) and are available for a more intimate analysis of the physicochemical processes of flame.

Nature of the Uniform Movement.—The uniform movement of flame, established soon after ignition at the open end of a tube closed at the other end, is characterised by constant dimensions of the flame-front; these are maintained so by a balance between the effects of a constant speed of propagation of flame normal to the flame-front, and a steady operation of the convection effect.

Relation between Tube-diameter and Speed of Uniform Movement.—The analysis given provides a quantitative proof of the explanation previously advanced (this vol., p. 1996) for the variation in speed of uniform movement of methane–air flames with tube-diameter.

Vibratory Movement of Flame.—The uniform movement develops into a vibratory movement as the flame travels. It is not improbable that the volume of gas burnt by unit area of flame in unit time is the same in the vibratory phase as in uniform movement; for when the amplitude of the vibrations is small, the flame area and its speed are both less than during uniform movement, and when the amplitude is large both flame area and mean (algebraic) speed are greater.

Turbulence.—As the effect of gentle turbulence due to convection is to increase the rate of consumption of the mixture solely by increasing the area of the flame-front, the effect of violent turbulence, as in the cylinder of an internal-combustion engine, is probably due entirely to the same cause, as suggested by previous investigators.

General Aspect.—If the results obtained with methane–air mixtures are representative of non-detonating gaseous explosions in general, it follows that the rate at which flame consumes a gaseous mixture is directly proportional to (*a*) the fundamental speed of propagation of the flame in that mixture, layer by layer, at the temperature and pressure just in front of the flame, and (*b*) the area of the flame-front. These can be determined, for flames of sufficient actinic value, from measurements of snap-shot photographs of flame during the phase of uniform movement.

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