## **277.** Studies in the Mechanism of Flame Movement. Part I. The Uniform Movement of Flame in Mixtures of Methane and Air, in Relation to Tube Diameter.

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NEARLY 50 years ago Mallard and Le Chatelier (Ann. Mines, 1883, viii, 4, 274) discovered that, when certain explosive mixtures of gases were fired at the open end of a horizontal tube, the other end being closed, the flame travelled for some distance at a uniform speed. Mason and Wheeler (J., 1917, 111, 1044) showed that the speed of "uniform movement" of flame in mixtures of methane and air increased progressively with increase in diameter of the containing tube, and Chapman and Wheeler (J., 1927, 38) suggested that the relation could be expressed, for the available measurements for a 10% methane-air mixture, by the equation  $V = cD^k$ , in which V is the speed of uniform movement of flame, D the diameter of the tube, and c and k are constants. Further investigation shows that the relation between tube diameter and speed of uniform movement of flame in mixtures of methane and air is not given exactly by this equation, but is more complex. A photographic analysis of the uniform movement has provided an explanation of the relationship.

Fig. 1 shows the speed of uniform movement of flame in mixtures of methane and air contained in horizontal tubes the diameters of which are indicated by figures placed above the corresponding curves. Five curves (2.5, 5, 9, 30.5, and 96.5 cm.), due to Mason and Wheeler, express the results of measurements of the time intervals between the fusion of thin screen-wires placed in succession in the path of the flame.

The higher of the two curves for the 5-cm. tube represents a redetermination by Mason (J., 1923, 123, 210), who photographed the flame by means of a revolving-drum camera. An independent determination of the curve for the 30.5-cm. tube was made by Coward and Greenwald (U.S.A. Bureau of Mines, Tech. Paper 427, 1928), by the screen-wire method. Their curve is that which indicates slightly higher speeds in the most explosive mixtures.

A study of the data of the seven curves suggested that, in order to elucidate the relation between tube diameter and flame speed, experiments with a tube of diameter between 9 and 30.5 cm. were necessary. The flame speeds were therefore determined in a tube of 20 cm. diameter, and, as a check on the older figures for a 9-cm. tube, a similar series of experiments was conducted in one of 10 cm. diameter, the nearest available size.

#### EXPERIMENTAL.

A metal tube of 20 cm. diameter was built in ten sections, each 1 m. long. One section was fitted with five quartz windows in line, each 18 cm. long and 0.5 cm. wide, so that the passage of flame along that section could be "seen"

### FIG. 1. Speed of uniform movement of flame in mixtures of methane and air, in tubes of various diameters.



by a camera through a slit which was continuous, except for the window frames, along the whole length of the section.

The tube of 10 cm. diameter was built of five sections, each 1 m. long. One section was fitted with quartz windows of the same size as those of the 20-cm. tube.

In each tube, the section provided with windows was fitted in succession at different positions in the tube, with the windows level with the axis of the tube

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and opposite the quartz lens of a motor-driven drum camera. The speed of the camera drum during the explosion was recorded by photographing break sparks produced with the aid of an electrically-maintained tuning fork of known frequency.

The CH<sub>4</sub> used was of 96-98% purity, the remainder being N<sub>2</sub> and air.

The gas mixtures to be exploded were made in the tube, after the products of a previous experiment had been displaced with air, by admitting a measured volume of CH4 and circulating the contents of the tube, through a by-pass, until the mixture was proved to be homogeneous by analysis of samples. The firing end of the tube was closed by a piece of oiled paper until the moment before ignition, the paper then being quickly and smoothly drawn up from the orifice. For most experiments, the gas was ignited by a small electric spark between electrodes fixed centrally 5 cm. within the tube, but in some preliminary experiments the position and nature of the means of ignition were varied. As in similar trials by Mason and Wheeler with narrower tubes, such changes were found to have no effect on the speed of uniform movement of flame, provided that they did not induce resonance in the column of gas and that the flame was allowed to travel a distance of 30 or 40 cm. before its speed was measured. According to Mason and Wheeler (loc. cit.) this is an essential precaution.

In order to obtain further information about the behaviour of flame during uniform movement, "snap-shot" photographs of explosions in glass tubes were taken on stationary plates.

The explosions of three mixtures in the 20-cm. tube were studied in detail by means of photographs taken on the revolving-drum camera. These mixtures contained (1) 10% of CH<sub>4</sub>, which is the most explosive of the series, (2) 7%, which contains excess of air, and (3) 12%, which contains excess of CH<sub>4</sub>. From about 0.5 m. beyond the point of ignition the speed of flame in successive portions of 0.25 m. was constant, within 5% of the mean, over a distance of about 2 m. in the 10% mixture and 3 m. in each of the others. Thereafter it was succeeded by a vibratory movement of the flame front.

When the length of the tube was halved, the uniform movement was set up, in a 10%  $CH_4$ -air mixture, at the same distance from the point of ignition, but was succeeded by the vibratory movement at a point only 60 cm. further. When the length of the tube was reduced to 2 m., the uniform movement was eliminated. That the tube should have a reasonably high ratio of length to diameter has long been recognised in work with narrow tubes.

In the tube of 5 m. length and 10 cm. diameter, the uniform movement was set up, in a 10% mixture, at 30 cm. from the point of ignition, and persisted over the next 75 cm.

Representative positions chosen for photographing the uniform movement in the whole range of inflammable mixtures of  $CH_4$  and air were the second metre of the 20 cm.-tube and the last quarter of the first metre of the 10-cm. tube. The results are given in Table I and in Fig. 1. (S.U.M. represents speed of uniform movement in cm./sec.)

## The Relation between Tube Diameter and the Speed of the Uniform Movement.

From the experimental observations recorded in Fig. 1, the curves of Figs. 2 and 3 are drawn. They show, for each unit % of CH<sub>4</sub>, the relation between

Tube diameter, 20 cm.				Tube diameter, 10 cm.			
CH4, %.	S.U.M.	CH4, %.	S.U.M.	CH₄, %.	S.U.M.	Сн₄, %.	S.U.M.
6.04	64	10.10	127	5.87	43	10.01	106
6.13	63	10.38	130	6.80	56	10.17	105
7.22	81	11.02	111	7.25	62	10.21	109
7.23	80	11.87	93	7.26	64	10.27	105
8.02	102	11.90	88	7.64	72	10.41	102
8.15	103	12.04	81	8.14	82	10.64	98
8.99	128	12.05	84	8.21	84	11.30	84
9.49	127	12.14	84	8.63	92	11.94	68
9.90	130	12.20	79	8.67	96	12.22	61
9.93	126	12.92	64	9.08	100	12.85	<b>48</b>
10.04	131	13.11	64	9.15	105	13.56	43
10.05	128			9.63	110		



## TABLE I.

tube diameter and speed of uniform movement. That the inflexions in the curves in the region of tube diameter between 10 and 20 cm. are real and not due to experimental error seems to be established by several considerations. (1) If any one, or indeed any two, series of observations are removed from the figures, it is not, as a rule, possible to draw curves without inflexions. (2) The differences of the two series for a 30.5-cm. tube, used by independent observers



employing almost the same methods, are relatively small. (3) The differences for a 5-cm. tube, when the photographic and the screen-wire method of registering flame speeds were used, are relatively small. (4) The results obtained by the screen-wire method with a 9-cm. tube are in accord with those obtained by the photographic method with a 10-cm. tube.

A possible extrapolation of the lower part of each of the curves in Figs. 2

## Moving-film Photographs of Explosions of 10% Methane-Air Mixtures, in Tubes of Different Diameters.

FIG. 4.—Diameter of tube, 2.5 cm. FIG. 5.—Diameter of tube, 20 cm.



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## PLATE II.

Snap-shot Photographs of Explosions of 10% Methane-Air Mixtures. FIG. 6.—Horizontal propagation of flame in a tube, 24 cm. in diameter.



FIG. 7.-Horizontal propagation in a tube, 5 cm. in diameter.



FIG. 8.—Downward propagation in a tube, 24 cm. in diameter.



FIG. 9.—Downward propagation in a tube, 24 cm. in diameter.



FIG. 10.-Horizontal propagation in a tube, 10 cm. in drameter.



and 3 is indicated by broken lines. These suggest that the speed of uniform movement of flame in any one mixture would be the same in all tubes wider than 20 cm. in the absence of some factor which becomes increasingly prominent in tubes of more than 15-cm. diameter. The nature of this factor was first indicated by an obvious contrast between the photographs of the uniform movement in narrow and in wide tubes. Figs. 4 and 5, representing explosions of 10% CH<sub>4</sub>-air mixtures, illustrate this. The former is a record of uniform movement over a 26-cm. length of a 2.5-cm. tube; it shows a straight-line front. The latter is a record over an equal length of the 20-cm. tube; it shows that, although the speed was grossly constant, small irregularities occurred in the flame front, and that in the horizontal section of the flame there was at every instant a complex geometrical pattern of luminous matter.

The mechanism of the flame movement became clearer when visual observation was made of explosions in wide glass tubes. Permanent records of the form of such flames were made with the aid of Ellis and Robinson's snap-shot camera (J., 1925, 127, 760). Fig. 6 is a print from a plate exposed momentarily after successive equal intervals of time, showing the form of flame travelling through a 10% CH<sub>4</sub>-air mixture contained in a glass tube 24 cm. in diameter and 2·13 m. long. The figure shows that, soon after ignition of the mixture, the flame had spread across the tube and had a smooth almost hemispherical front (Position 1). A moment later the flame-front was what may be described, for want of a better term, as nodular (Position 2). The third snapshot resembles the second, except that the flame is more tilted as a whole. The fourth shows only the lower part of the flame, the upper having passed beyond the field of view of the camera.

Fig. 6 shows that the movement of flame in the experiment which it illustrates is not due only to inflammation of successive layers of mixture, but is affected in two ways by convection; the flame is distorted and tilted forward, the upper part advancing faster than it would otherwise. Each involves an enlargement of the flame surface which presumably increases the amount of gas burnt in unit time, and in consequence the speed of flame increases correspondingly. It may be recalled that Gouy (*Ann. Chim. Phys.*, 1879, **18**, 5) postulated that the amount of gas burnt by the flame of a homogeneous gas mixture is proportional to the surface area of the flame, and that Ellis (*Fuel*, **1928**, **7**, **195**, etc.) consistently used this postulate.

The snap-shot photographs of explosions of the same mixture in a tube of 5-cm. diameter are strikingly different from those of the explosion in the wider tube. Fig. 7 is typical. The flame front is tilted, as before, but its shape is comparatively simple. In none of the many photographs taken did the "nodular" flame appear, and when occasionally the flame had two heads the next snap-shot showed the simple front.

The rapid increase in the speed of uniform movement with increase in tube diameter above 20 cm. is therefore to be explained by the disproportionately large increase in flame surface corresponding with the appearance of the nodular flame.

The increase in speed of uniform movement with increase in tube diameter between 2.5 and about 15 cm. (the lower parts of the curves in Figs. 2 and 3) is not accompanied by any such definite change in the shape of the flame front, and an explanation of the effect is less obvious. It does not seem to us reasonable to assume that the temperature of the flame front is affected by the walls, except in their immediate vicinity; nor does the cooling of the gaseous products, which is more rapid in the narrower tubes, cause an appreciable movement of the unburnt gases towards the flame and thus retard it, for we have found that the changes of pressure in the unburnt gases during this phase of CH<sub>4</sub>-air explosions are too small. It was clear from other photographs, however, that the flame front in the 2·5-cm. tube is steeper than that in the 5·0-cm. tube during the uniform movement, and that the flame front in the 5-cm. tube is steeper than that in the 10-cm. tube. Straight lines drawn, on the originals of the three photographs, from the points where each flame met the top of the tube to the corresponding point at the bottom were inclined to the axes at angles of  $39^{\circ}$ ,  $26\frac{1}{2}^{\circ}$ , and  $23\frac{1}{2}^{\circ}$ , respectively. The smaller the angle, the greater the surface represented per unit cross section (at right angles to the axis), and therefore the larger also the volume of gas burnt in unit time per unit cross-sectional area and the greater in consequence the speed of flame. We propose to discuss the quantitative aspect of this in a subsequent communication.

# An Attempt to Eliminate Convection Effects in the Propagation of Flame.

If it were possible to eliminate convection effects from the uniform movement of flame, the absolute speed of propagation through successive layers of the mixture could be measured. It seemed that in the downward propagation of flame in a vertical tube, the hot products being above the unburnt gas, convection should be eliminated and the flame front would be horizontal except, perhaps, close to the walls of the tube. In such circumstances each element of the flame would travel with the same speed, and the speed of uniform movement would be independent of the diameter of tube, provided that it were not too narrow. It had already been shown, however, by Mason and Wheeler (J., 1920, 117, 1227) that the speed of downward propagation of flame in a tube 23 cm. wide is considerably greater than in a tube 5-cm. wide for all  $CH_4$ -air mixtures, except those near the limits of inflammability. The fact that flame travels downward in the limit mixtures with equal speeds in these two tubes suggests that the cooling effect of the walls is negligible for these mixtures and, ipso facto, for faster-burning mixtures. Hence there was no obvious explanation of the difference in flame speeds of the faster-burning mixtures.

Visual observation and photographs (e.g., Fig. 8) showed that the downwardtravelling flame fronts of a 10% CH<sub>4</sub>-air mixture in a tube 24 cm. wide were tilted and nodular as in experiments with the tube horizontal. It seemed that the disturbance of the flame front might have its origin in an irregular outflow of hot gases from the open end of the tube. Attempts were made, therefore, to regulate the escape of the hot gases by covering the open end of the tube with wire gauze, and with wire gauze supporting a pad of glass wool, but in no experiment did this make any essential difference to the flame. Fig. 9 is typical of the results; the flame is more symmetrical than that of Fig. 8, but is otherwise unaltered. It appears that the source of the disturbance producing the nodular flame in wide tubes is in the vicinity of the flame itself, and is a convection effect. It nevertheless remains possible that the flame simultaneously over the whole area of the mouth of the tube; but while this may be accomplished approximately by drawing a small flame rapidly across the mouth of a narrow horizontal tube, it is impossible to do the same with a wide vertical tube without inducing strong convection currents during the operation.

The downward-moving flames of a 6% CH<sub>4</sub>-air mixture, which is near the lower limit of inflammability, developed horizontal fronts shortly after ignition in 5- and in 24-cm. tubes, and remained so for a period. Their speeds during this phase were equal, 10 cm./sec. It is reasonable to conclude that the movement was uncomplicated by convection and that its speed is the absolute speed of flame propagation, layer by layer, for that mixture. The difference between this and the speed of uniform movement for the same mixture in a horizontal tube, or in upward propagation, is a measure of the convection effect. Reference to the curve marked "6%" in Fig. 2 shows the predominance of the convection effect in determination of the speed of uniform movement in horizontal tubes.

The 12.7% CH<sub>4</sub>-air mixture, which is near the higher limit of inflammability, appears to have the same absolute speed of flame propagation, layer by layer, as the 6% mixture.

### A Note on the Initiation of Uniform Movement of Flame.

In an earlier paragraph it was stated that in the 20-cm. tube the uniform movement began after the flame had travelled some 0.5 m. from the point of ignition. In the interval, the speed of flame was somewhat less than that of the uniform movement. The explanation is obvious from a consideration of Fig. 10, a series of snap-shot photographs, at equal intervals of time, of the early stages of an explosion. These were taken in a horizontal glass tube, 10 cm. in diameter. In the first four photographs, starting from the point of ignition on the left, the flame is but little tilted and is clearly moving more slowly than in the succeeding photographs where it is more tilted, and, incidentally, is moving with uniform speed.

The uniform movement began about 30 cm. from the point of ignition in the 10-cm. tube, 15 cm. in the 5-cm. tube, and 10 cm. in the 2.5-cm. tube.

## The Term " Uniform Movement of Flame."

When Mallard and Le Chatelier measured the speed of "uniform movement" of flame, they believed that they were recording the normal speed of propagation of flame by conduction of heat. Mason and Wheeler (J., 1917, **111**, 1044) cast doubt on this, suggesting that the uniform movement should be regarded "simply as a particular phase in the propagation of flame that results when ignition is effected (in a quiescent mixture) at the open end of a straight, horizontal tube of any diameter closed at the other end; and not as resulting from a particular mode of heat transference." Later (J., 1920, **117**, 1227), they stated that "the latter is the preferable, if not the only correct, way of regarding the uniform movement." This conclusion is now strengthened by the demonstration that, if convection is ever absent in explosions of methane-air mixtures, initiated at the open end of a tube, it is only so when flame travels downwards in mixtures of composition near the limits of inflammability.

Mason and Wheeler showed that "a phase in the propagation of flame during which the speed is uniform is obtained when the flame travels from the open to the closed end of a tube, whether the direction of travel is horizontal, vertically upwards, or vertically downwards." Although they avoided the term "uniform movement" in writing of propagation in any direction but horizontal, we suggest, with the approval of Professor Wheeler, that there is no cogent reason to perpetuate that limitation, which doubtless has its origin in the accident that long tubes are more conveniently supported horizontally than vertically.

Furthermore, since Payman and Wheeler (J., 1923, 123, 1251), Georgeson and Hartwell (J., 1927, 265), and Bone, Frazer, and Winter (*Proc. Roy. Soc.*, 1927, A, 114, 402) have discovered several examples of non-uniform propagation of flame during the regime before the "vibratory movement," the definition should be qualified to allow for exceptions. We suggest, therefore, that the "uniform movement" may be described as "an early phase of sensibly uniform speed usually observed in the propagation of flame (through a quiescent gaseous mixture) from the open end of a straight tube towards the closed end."

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