

CCXLIII.—*The Propagation of Flame in Mixtures of Methane and Air. The "Uniform Movement."*

By RICHARD VERNON WHEELER.

IN the course of their well-known researches on the combustion of explosive gaseous mixtures, Mallard and Le Chatelier (*Ann. des Mines*, 1883, [viii], 4, 274) studied the propagation of flame in mixtures of methane and air contained in horizontal tubes. So far as such mixtures are concerned, the general conclusions drawn by them regarding the manner in which flame is propagated were as follows.

When the mixture contained in a horizontal tube closed at one end and open at the other is ignited at the open end, the flame travels for a short distance at a uniform speed. This "uniform movement" is followed by a "vibratory movement," in the course of which the flame travels backwards and forwards in an irregular manner, the mean speed from point to point along the tube being usually greater than that of the "uniform movement." These vibrations usually continue to the end of the tube, but sometimes during a particularly violent vibration the flame may be extinguished, owing to the mixing of burnt gases with the unburnt mixture.

When the mixture is ignited at the closed end of the tube the flame travels, in short tubes at all events, with increasing speed towards the open end.

In the course of investigations on mine explosions, carried out, in the first instance, for the Mining Association of Great Britain, and, latterly, at the Home Office Experimental Station, the necessity arose for repeating Mallard and Le Chatelier's experiments regarding mixtures of methane and air.

The present paper deals with the "uniform movement," the speed of which is the normal speed of propagation of flame by conduction of heat from layer to layer of the mixture, and is constant for a given mixture at a given temperature and pressure.

Mallard and Le Chatelier made a complete study of how far the diameter, length, and material of the tubes influenced the speed and duration of the uniform movement in many gaseous mixtures, with the object of determining the limiting dimensions requisite to ensure that the true speed—the speed that would be obtained in a mixture of indefinite extent—should be determined. Given the right dimensions of tubes, the material of which they were made did not appreciably affect the speeds. Repetition of Mallard

and Le Chatelier's experiments regarding these experimental conditions has confirmed their results.

The diameter of tube necessary to avoid cooling by the walls, and consequent retardation of the flame, was found to be greater the slower the speed of the flame. For the most slowly moving flames in mixtures of methane and air a tube of at least 5 cm. diameter is necessary. The speed of travel of flame in a tube 9 cm. in diameter is slightly greater than in a tube 5 cm. in diameter.

The duration of the uniform movement, which varies with each mixture, increases with the diameter and length of the tube up to a certain maximum; after which increase in length makes no appreciable difference. In a tube 5 cm. in diameter and 6 metres long the uniform movement in all mixtures of methane and air extends over a distance of about 150 cm., whereas in a tube of the same diameter and 2 metres long the distance travelled by the flame at a uniform speed may be less than 50 cm.

For their experiments Mallard and Le Chatelier used tubes 5 cm. in diameter and 1 metre long, and measured the speed of travel of flame over the first 50 cm. The length of the tube was insufficient to ensure that the measurements of the speed of the flame would not include part of the "vibratory movement," a fact which they themselves realised (*loc. cit.*, p. 317). Their measurements for the same mixture show, in consequence, rather wide variations. Their experiments were further vitiated by the fact that the methane used was prepared from sodium acetate ("il exhalait une forte odeur d'acetone"). Such "methane" may contain as much as 10 per cent. of nitrogen, 10 per cent. of unsaturated hydrocarbons, and 2 or 3 per cent. of hydrogen.*

The conclusions drawn by Mallard and Le Chatelier were:

(1) The speed of the uniform movement increases regularly with the percentage of methane up to a certain maximum, after which it decreases regularly. The curve obtained on plotting speeds as ordinates and percentages of methane as abscissæ is thus represented by two straight lines meeting at a point. Their curve (from Plate VIII of their paper) is reproduced in Fig. 1.

(2) The maximum speed is obtained, not with that mixture containing the quantity of methane required for complete combustion, namely, 9.4 per cent., but with a mixture containing about 12 per cent. of methane. Le Chatelier ("Leçons sur le carbone," Paris, 1908) explains this result by assuming that the speed of propagation of flame during the uniform movement depends, not only on the temperature of combustion of the mixture, but on its thermal conductivity, which is greater the greater the

* Compare Hauser, "Leçons sur le grisou," Madrid, 1908.

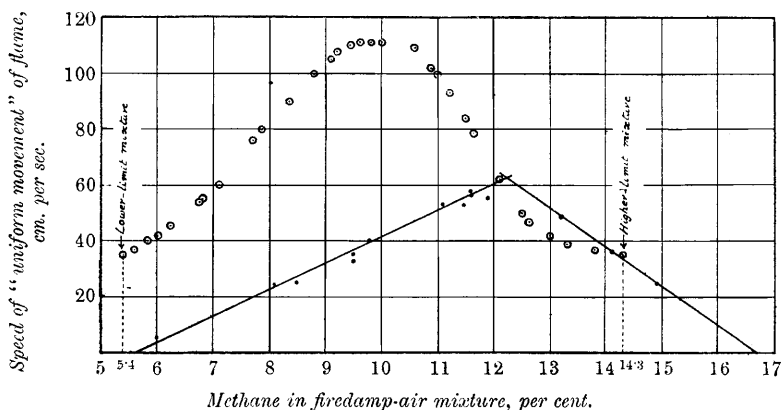
proportion of methane present. The thermal conductivities of air and of methane are 5.22×10^{-5} and 6.47×10^{-5} respectively.

Fresh determinations, made in the manner described in the experimental portion of this paper, do not bear out Mallard and Le Chatelier's results. The form of curve obtained on plotting speeds as ordinates and percentages of methane as abscissæ is shown in Fig. 1.

It will be seen that there is practically no difference between the speeds attained in mixtures containing from 9.45 to 10.55 per cent. of methane,* such differences as there are being probably within the limits of experimental error.

Near the lower- and higher-limits of inflammability, which, for horizontal propagation, are 5.4 and 14.3 per cent. respectively, the

FIG. 1.



curve flattens, more noticeably towards the higher limit, and becomes, ultimately, nearly horizontal. It will be understood, therefore, that a prolongation of either "limb" of the curve so as to cut the zero velocity ordinate, as done by Mallard and Le Chatelier to determine the theoretical limits of inflammability, is not justifiable.

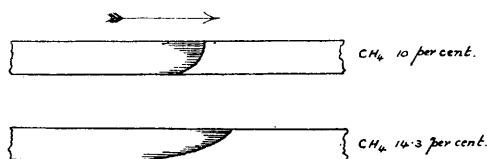
Vibrations were not developed by the flames in all the mixtures. In those containing more than 12.5 or less than 5.8 per cent. of methane the flame usually travelled at a uniform or slightly de-creas-

* This conclusion is confirmed by another series of experiments in which different mixtures of methane and air were ignited at the centre of a large spherical explosion vessel. The time that elapsed between the moment of ignition and the first indication of pressure on the sides of the vessel was less the higher the percentage of methane in the mixture up to 9.5 per cent. methane, after which it remained practically constant up to 11 per cent.

ing speed throughout the length of the tube, although sometimes slight vibrations were noticeable in all but the "limit-mixtures." In these latter the speed of travel of flame was quite uniform throughout, and was the same for both the higher- and lower-limit mixtures.

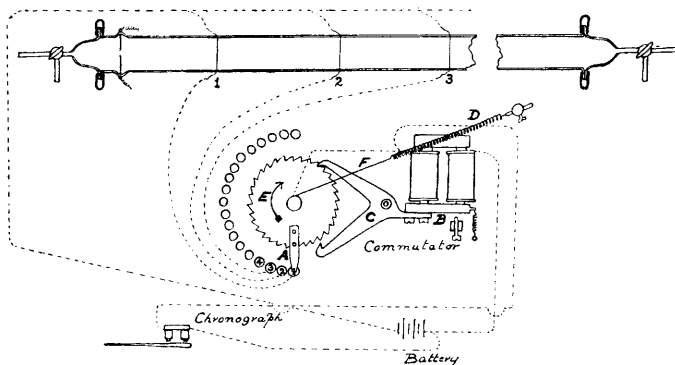
As noted in a previous paper (this vol., p. 2593), the flame

FIG. 2.



travelling horizontally in a 5.4 per cent. methane-air mixture, contained in a tube 5 cm. in diameter, occupies only the upper part of the tube. The flames in the other mixtures of methane and air, including the higher-limit mixture, completely filled the cross-section of the tube, the front of the flame (during the uniform

FIG. 3.



movement) being shaped as shown in Fig. 2. The faster the speed of the flame the blunter was its front.

EXPERIMENTAL.

The arrangement of glass tubes is shown in Fig. 3. Three lengths of tube of 5 cm. internal diameter, each 2 metres long, were joined together by broad pieces of stout rubber tubing, and supported horizontally in a straight line. Each end of the complete length

of 6 metres was flanged and ground to receive flanged end-pieces, which were held in position by metal clips. Each end-piece was fitted with a wide-bore three-way tap. Glass-covered platinum electrodes reaching to the centre of the tube, leaving a spark-gap of 3 mm., were fused 4 cm. from one end.

Another tube, similarly arranged, but of 9 cm. internal diameter, was used for a separate series of experiments.

"Screen-wires" of copper 0.025 mm. in diameter were threaded vertically across the tube through fine holes pierced through the walls at certain points, the holes being afterwards covered by adhesive plaster. In order to avoid including in the measurements of the speed of the flame any impetus that might be given by the igniting spark, the first screen-wire was fixed 40 cm. from the point of ignition. Other screen-wires were fixed 50, 100, 200, 300, and 400 cm. respectively from the first.

The method of recording the time of passage of flame along the tube was electrical. Each screen-wire carried a small electric current, the interruption of this current when the flame melted the wires being recorded by the movement of an electro-magnet.

It was important to avoid error due to latency or "time-lag" of individual electromagnets. An instrument, which can be termed an automatic commutator, was therefore designed to enable each successive break in circuit to be recorded by the same electromagnet. This instrument is operated in the following manner:

One terminal of the battery supplying the electric current is connected to the brush, *A*, of the commutator (Fig. 3), and a lead from the other terminal of the battery conducts the current to the electromagnet of the chronograph, so that its armature is attracted. The current then passes by a lead to the electromagnet on the commutator, and that armature is also attracted; the lead carrying the current then goes to one terminal of the screen-wires on the explosion-tube one after the other; the other terminal of each screen-wire is connected to the corresponding stud on the commutator by separate leads.

Supposing the brush, *A*, to be resting on No. 1 stud (the position that it occupies at the beginning of an experiment), the current is then flowing through the chronograph electromagnet, the commutator electromagnet, and No. 1 screen-wire; then through the brush, *A*, back to the battery. Suppose now that the flame passes along the tube and melts screen-wire No. 1; the chronograph electromagnet releases its armature, and the pen it carries makes a mark on the moving surface; at the same time the armature, *B*, of the commutator electromagnet is released, and the anchor-escapement, *C*, attached to the armature, is moved. This allows the

coiled spring, *D*, to pull the scape-wheel, *E*, round by the cord, *F*, which is wound on a drum attached to the axis of the scape-wheel. The brush, *A*, then moves on to stud No. 2, and the current at once begins to flow through screen-wire No. 2; the chronograph electromagnet and the commutator electromagnet, and the armatures of both these are again attracted; the pen on the chronograph is moved back to its former position, as also are the armature, *B*, and the escapement, *C*, whilst the brush, *A*, moves a little further on to stud No. 2. When the flame reaches No. 2 screen-wire the same cycle is repeated and so on for as many screens as may be required, all the interruptions of circuit being recorded by the one pen on the chronograph.*

The chronograph used was the laboratory chronograph of the Cambridge Scientific Instrument Company, the speed of travel of the moving surface (a spool of Morse paper) being recorded by a $\frac{1}{2}$ -second contact-clock.

Method of Conducting an Experiment.—The mixtures of methane and air were made in a 140-litre gas-holder over water rendered slightly alkaline by potassium hydroxide. A rapid current of the mixture was passed through the explosion-tube until the gases entering and leaving had the same composition, as shown by explosion-analyses of samples taken through the three-way taps. A volume of mixture equal to about six times the volume of the tube was found to be ample for sweeping out all the air contained in the tube.

All electrical connexions through the screen-wires and chronograph having been established, the left-hand end-piece of the explosion-tube was removed (by sliding it downwards) and the mixture ignited at the now open end by passing an induction-coil spark.

The methane used was a particularly pure supply of fire-damp from a "blower" at a colliery in South Wales, whence it was obtained compressed in cylinders. Analysis, after removal of 0.8 per cent. of carbon dioxide, showed it to contain 97.4 per cent. of methane, 2.3 per cent. of nitrogen, and 0.3 per cent. of other impurities (carbon monoxide and ethylene). It contained no hydrogen or ethane.

* A somewhat detailed description of this device has been given in the belief that it may prove of value to other workers. The author has found it adequate for measuring the speed of the rapidly moving flames of coal dust explosions and coal-gas and air explosions in large galleries. Its effectiveness depends essentially on the rapidity with which the brush of the commutator can be made to move from one stud to the next; by suitable proportioning and adjustment of the moving parts and regulation of the electric current passing through the magnets, the time taken for the brush to move from stud to stud can be made as little as $\frac{1}{100}$ th second.

Results of Experiments.—The results of all the determinations made of the speed of the uniform movement in different mixtures are given in the table that follows. As a general rule, the uniform movement extended for a distance of 150 cm. from the point of ignition, so that from each experiment with a particular mixture two determinations of the speed were obtained (between No. 1 and No. 2, and between No. 2 and No. 3 screen-wire respectively). Some of the more rapidly-moving flames, in mixtures containing between 9.5 and 11.0 per cent. of methane, began to vibrate just before reaching the third screen-wire; in such cases only the speed between screen-wires Nos. 1 and 2 was taken as being that of the uniform movement.

Methane in fire-damp-air mixture, per cent.	Speed of "uniform movement" of flame, cm. per second.
5.40	36.5, 36.0, 35.5, 35.5, 35.5, 35.5, 36.0, 36.0, 35.5.
5.60	37.0, 37.0.
5.85	40, 40.5, 40.5, 40.5.
6.25	46, 46, 45.5, 45, 45.5, 45, 45.5.
6.75	54, 54.
6.80	56, 55, 55, 55.
7.10	61, 59, 61, 59.
7.70	77, 77, 75, 75.
8.35	91, 90.
8.80	100, 100, 99, 100.
9.10	105, 104, 106, 104.
9.20	108, 109.
9.45	110, 110, 110, 110.
9.60	111, 111.
9.80	111, 112.
10.00	112, 110, 112, 113, 112, 110, 109, 113.
10.60	109, 109, 109.
10.90	102, 102, 101.
11.00	100, 98, 99, 99.
11.20	93, 92.
11.50	84, 84, 83, 83, 85, 84.
12.10	62.5, 62.
12.50	50, 50, 49.
12.65	49, 47, 48, 46.
13.00	42.5, 42, 42, 42.
13.05	41, 40.5.
13.30	39, 38, 39, 38, 38, 38, 38.
13.80	37, 36.5.
14.30	36.0, 35.5, 35.5.

For the determinations of the speed of travel of flame in the higher-limit mixture pure methane was used, since the 2.3 per cent. of nitrogen contained in the fire-damp slightly affected the higher limit, whereas it had no appreciable effect on the speed of travel of flame in the other mixtures (compare this vol., p. 2596).

A similar series of determinations was made, using an explosion-tube of 9 cm. internal diameter. The speeds were from 5 to 10 cm. per second greater than those of corresponding mixtures in the tube 5 cm. in diameter. The shapes of the curves connecting speeds

VOLATILE OIL FROM THE LEAVES OF BAROSMA VENUSTA. 2613

with percentages of methane, and the limits of inflammability, were the same in both series of experiments.

The propagation of flame in mixtures of methane and air, and in mixtures to which nitrogen has been added, has been further studied. An account of the work will be communicated later to the Society.

Addendum.

Since this paper was prepared an account has appeared of experiments on the same subject by A. Parker and A. V. Rhead (this vol., p. 2150). It is surprising to find that these authors are apparently unacquainted with Mallard and Le Chatelier's complete researches dealing with the "uniform" and "vibratory" movements during the propagation of flame in gaseous mixtures contained in glass tubes, as outlined in the present paper. Their results are interesting in that they emphasise the necessity, pointed out by Mallard and Le Chatelier, of employing tubes of ample diameter when conducting experiments of this nature; the tubes they used were of too small a diameter to enable them to determine either the true character of the speed-percentage curve or the limits of inflammability.

ESKMEALS,
CUMBERLAND.
