CXXXVII.—*The Propagation of Flame in Complex Gaseous Mixtures. Part III. The Uniform Movement of Flame in Mixtures of Air with Mixtures of Methane, Hydrogen and Carbon Monoxide, and with Industrial Inflammable Gases.*

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THE common industrial gases contain as their inflammable constituents methane, hydrogen, and carbon monoxide in various proportions.

The speed of the uniform movement of flame in mixtures of methane and air in a tube 2·5 cm. in diameter was given in Part II of this series of papers. The speeds with hydrogen and air in a similar tube (over the major portion of the range of inflammable mixtures) have been determined by Haward and Otagawa (T., 1916, **109**, 83). The speeds in mixtures of carbon monoxide and air are recorded in the present paper.

Mixtures of Carbon Monoxide and Air.—It is well known that the rate of combustion of carbon monoxide is dependent on the amount of water vapour present. Dixon, for example (*Phil. Trans.*, 1893, **184**, 97), has shown that the velocity of the detonation wave in a mixture of carbon monoxide and oxygen ($2CO + O_2$) increases with the percentage saturation of water vapour.

The present series of determinations of the speed of the uniform movement of flame in mixtures of carbon monoxide and air was carried out with mixtures saturated with water vapour at the ordinary temperature and pressure. Since the room temperature varied, it was not surprising to find that the speed in a given mixture did not remain constant from day to day. Identical results were, however, obtained in experiments made within a few minutes of each other at the same temperature and pressure. Table I illustrates the effect of change in the percentage saturation of water vapour on the speed of the uniform movement of flame in a mixture of carbon monoxide and air containing 50 per cent. of carbon monoxide.

TABLE I.

Speed of Uniform Movement of Flame in a Mixture of Carbon Monoxide and Air (50 *per cent.* CO) *in a Tube* 2·5 *cm. in Diameter.*

Temperature and pressure.	Cm. per sec.	
10° and 750 mm.	(1) 59·9	(2) 59·9
15° and 750 mm.	(1) 65·0	(2) 64·5
17° and 755 mm.	(1) 79·4	(2) 79·0

A series of determinations of speeds of flame over the whole range of inflammable mixtures was carried out during a period when the temperature of the laboratory did not alter appreciably (about 12°). The values obtained are given in table II.

TABLE II.

Speed of Uniform Movement of Flame in Mixtures of Carbon Monoxide and Air in a Tube 2·5 *cm. in Diameter at* 12° *and* 750 *mm.*

Per cent. of carbon monoxide.	Cm. per sec.	Per cent. of carbon monoxide.	Cm. per sec.
16·15	Tongue of flame only.	59·81	54·2
16·29	19·5	65·55	37·4
16·40	19·4	65·84	36·3
16·51	19·4	67·10	30·2
24·47	34·0	67·57	29·6
30·50	46·0	69·00	26·0
44·84	60·1	70·63	20·0
50·45	59·9	70·68	20·3
54·40	57·8	71·19	19·4
59·58	56·2	71·31	Trailing flame travelled 15 cm.

These values are of interest in themselves, apart from their connexion with the problem of the propagation of flame in complex gaseous mixtures, inasmuch as they disclose the fact that the maximum speed of flame is obtained with mixtures containing from 45 to 50 per cent. of carbon monoxide. The mixture for complete combustion contains 29·5 per cent. carbon monoxide, so that the "displacement" of the maximum-speed mixture is greater even than with hydrogen, despite the fact that the thermal conductivity of carbon monoxide is but little different from that of air.

Industrial gas mixtures may contain varying proportions of water vapour. There may therefore be some uncertainty as to the correct values to use for the speed of flame in mixtures of carbon monoxide and air when attempting to calculate the speed of flame in the mixed industrial gas. Such gases, however, contain hydrogen as well as carbon monoxide, and the presence of hydrogen affects the speed of flame in a similar degree to that of water vapour. With mixtures of gases containing fairly high proportions of hydrogen, it is therefore not unlikely that the effect of variation in the moisture content would be inappreciable. It should therefore be sufficient for our purpose to know the values for the speed of flame in mixtures of air with a mixture of hydrogen and carbon monoxide. Or the "effective" speeds for mixtures of carbon monoxide and air could be calculated from such values and these speeds used for further calculation.

In this connexion, it is interesting to note that Berthelot (*Ann. Chim. Phys.*, 1881, [v], **28**, 289) found the rate of detonation in mixtures of carbon monoxide and oxygen to be about half the calculated value. For mixtures of oxygen with carbon monoxide plus hydrogen, the calculated values were in good agreement with those found. Similarly, in the present research, the maximum speed of uniform movement of flame in mixtures of carbon monoxide and air is found to be about half the value calculated, making use of the values determined for hydrogen–air and hydrogen–carbon monoxide–air mixtures.

Mixtures of Hydrogen and Air.—As with tubes of smaller diameter (this vol., p. 36), it was not found possible to determine accurately the speed of the uniform movement of flame in the upper-limit mixture of hydrogen and air in a tube 2·5 cm. in diameter. A mixture containing 71·4 per cent. of hydrogen was found to be the richest which would propagate flame under the experimental conditions. The flame was not hot enough to melt "screen wires," but its speed, as measured by means of a tapping key in connexion with a chronograph, was found to be approximately 50 cm. per second.

A characteristic of the lower-limit mixture and of mixtures near to it is the formation on ignition of minute balls of flame, which pass steadily from the open to the closed end of the tube. These flames are propagated mainly by the influence of convection currents, and the speed–percentage curve at the lower-limit region is not continuous, but shows a definite break. Nevertheless, no definite distinction, at the point of break in the curve, could be drawn between the normal and the balls of flame, the latter increasing in size and gradually changing their form as the percentage of hydrogen increased.

The speeds of the flames in mixtures near the limits are given in table III, which completes the table given by Haward and

TABLE III.

Speed of the Uniform Movement of Flame in Mixtures of Hydrogen and Air in a Tube 2·5 cm. in Diameter.

Hydrogen. Per cent.	Speed, cm. per sec.
6·10	No flame observed.
6·19	10
6·31	12
6·52	15
14·71	120
71·39	50
71·51	Flame to open end only.

Otagawa (*loc. cit.*, p. 89). In only one instance was the flame hot enough to melt "screen wires," namely, with the mixture containing 14·71 per cent. of hydrogen; the remaining speeds were determined by means of a tapping key.

Mixtures of Methane, Hydrogen, and Air.—The speed of the uniform movement of flame in a tube 2·5 cm. in diameter was determined over a range of mixtures of air with two mixtures of methane and hydrogen. The first mixture contained equal volumes of methane and hydrogen (CH_4+H_2), the second three volumes of methane to one volume of hydrogen ($3CH_4+H_2$). The results are recorded in table IV. The lower-limit flames preserved the general character of the corresponding hydrogen flames, and their speeds were found to be lower than the speed in the limit mixture of methane and air.

TABLE IV.

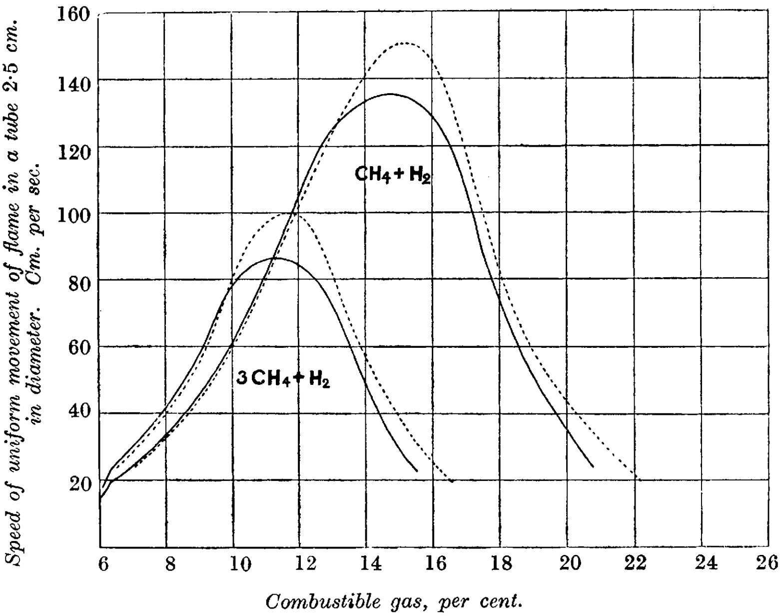
Speed of the Uniform Movement of Flame in Mixtures of Air with Hydrogen Methane Mixtures in a Tube 2·5 cm. in Diameter.

$CH_4 + H_2$.		$3CH_4 + H_2$.	
Combustible gas. Per cent.	Speed, cm. per sec.	Combustible gas. Per cent.	Speed, cm. per sec.
6·03	15·0	6·09	18·0
6·20	17·1	6·22	19·9
6·31	19·1	6·50	21·0
6·73	22·1	6·80	27·7
7·68	28·6	7·84	39·6
9·05	45·6	9·06	58·3
10·23	67·4	9·93	78·7
11·95	104·1	11·35	84·9
11·99	106·3	12·26	82·2
13·50	128·6	13·25	66·7
14·93	135·3	14·20	45·7
15·93	127·3	14·99	27·8
16·90	111·9	15·50	22·6
18·31	65·6		
19·96	35·5		
20·22	30·5		
20·32	28·5		
20·48	27·3		
20·80	24·3		

The results are plotted as curves in Fig. 1, the calculated curves being shown in dotted line. The maximum speeds calculated by the method given in Part II are 150 and 99 cm. per second respectively for the mixtures CH_4+H_2 and $3CH_4+H_2$. The values found were 135 and 85 cm. per second.

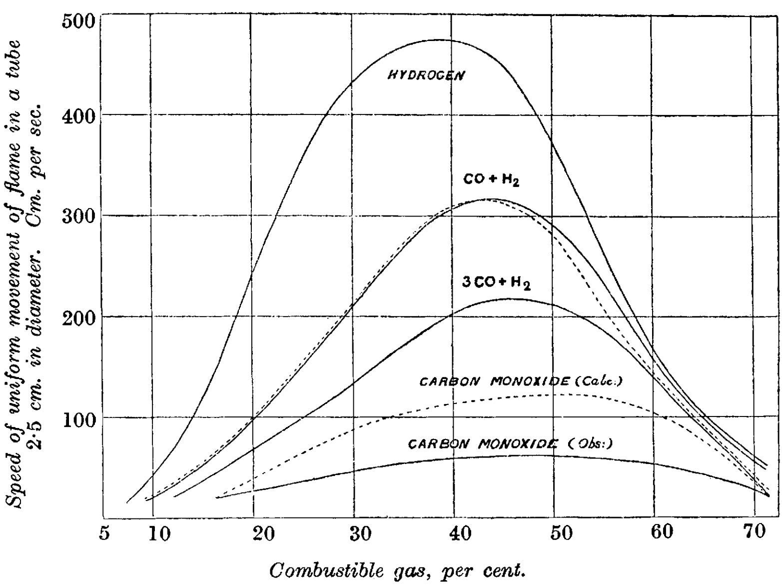
Mixtures of Carbon Monoxide, Hydrogen, and Air.—Two mixtures of carbon monoxide and hydrogen were employed, of composition $CO+H_2$ and $3CO+H_2$, corresponding with the methane–hydrogen mixtures. The results are given in table V, and are plotted as curves in Fig. 2.

FIG. 1.



Speed of uniform movement of flame in a tube 2·5 cm. in diameter. Cm. per sec.
160
140
120
100
80
60
40
20
$CH_4 + H_2$
$3CH_4 + H_2$
6
8
10
12
14
16
18
20
22
24
26
Combustible gas, per cent.

FIG. 2.



Speed of uniform movement of flame in a tube 2·5 cm. in diameter. Cm. per sec.
500
400
300
200
100
HYDROGEN
$CO + H_2$
$3CO + H_2$
CARBON MONOXIDE (Calc.)
CARBON MONOXIDE (Obs.)
5
10
20
30
40
50
60
70
Combustible gas, per cent.

From the values found for hydrogen and for the mixture $3CO+H_2$, the speeds of the flames in mixtures of air with $CO+H_2$ were calculated. The results are shown in dotted line in Fig. 2. The values for carbon monoxide and air were also calculated from these values, and the curve is given in the diagram for comparison. It will be seen that the values calculated in this manner are much higher than those found by experiment. These "effective" speeds have been used in subsequent calculations instead of the values as determined, which are dependent on the amount of water vapour present.

TABLE V.

Speed of Uniform Movement of Flame in Mixtures of Air with the Mixtures $CO+H_2$ *and* $3CO+H_2$ *in a Tube* 2·5 *cm. in Diameter.*

$CO + H_2$.		$3CO + H_2$.	
Combustible gas. Per cent.	Speed, cm. per sec.	Combustible gas. Per cent.	Speed, cm. per sec.
9·25	18·2	12·00	19·2
10·35	21·1	18·99	67·1
15·40	58·3	27·82	115·0
20·57	100·4	34·73	166·2
30·25	211·5	41·32	205·5
36·94	282·9	46·90	214·0
41·50	309·7	53·17	200·0
45·92	315·2	58·49	154·7
51·23	280·0	70·36	34·4
58·55	178·5	71·42	20·8
69·00	64·5		
70·75	50·1		
71·34	44·4		

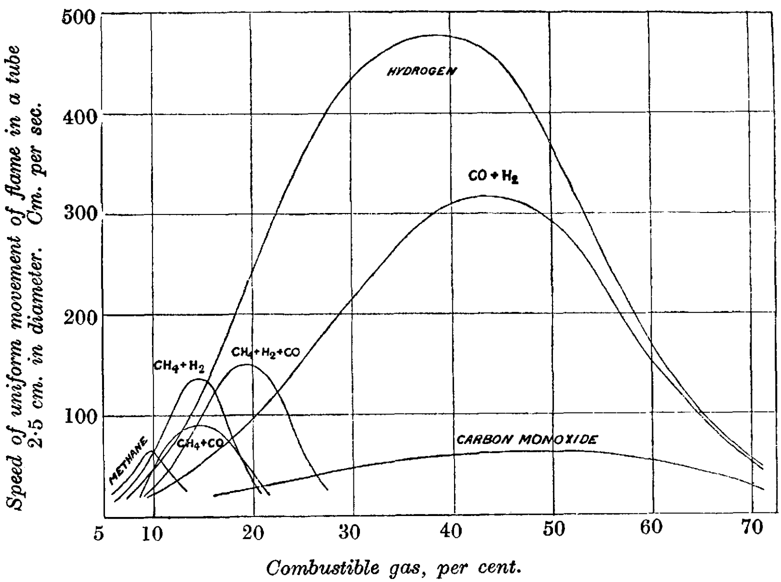
Mixtures of Methane and Carbon Monoxide, and Mixtures of Methane, Hydrogen, and Carbon Monoxide with Air.—Table VI records the results obtained with a mixture containing equal volumes of methane and carbon monoxide, and with one containing equal volumes of methane, hydrogen, and carbon monoxide. Methane, or any gas into the composition of which hydrogen enters, acts towards mixtures of carbon monoxide and air in a manner comparable with that of hydrogen and water vapour. The maximum speed of uniform movement of flame in mixtures of air with each of the mixtures CH_4+CO and CH_4+CO+H_2 was found to be 91 and 150 cm. per second respectively, whilst the corresponding calculated values are 78 and 145 cm. per second.

TABLE VI.

Speed of Uniform Movement of Flame in Mixtures of Air with the Mixtures $CH_4 + CO$ *and* $CH_4 + H_2 + CO$ *in a Tube* 2·5 *cm. in Diameter.*

$CH_4 + CO$.		$CH_4 + H_2 + CO$.	
Combustible gas. Per cent.	Speed, cm. per sec.	Combustible gas. Per cent.	Speed, cm. per sec.
9·45	21·9	7·70	21·2
9·88	36·2	10·01	36·5
12·07	62·5	14·01	83·3
13·73	85·7	15·80	109·4
15·95	91·3	18·92	150·0
18·06	68·9	20·42	148·7
19·32	52·3	22·43	118·5
21·55	19·8	25·05	57·8
		27·57	21·8

FIG. 3.



The speed–percentage curves for the equimolecular mixtures $CH_4 + H_2$, $H_2 + CO$, $CO + CH_4$, and $CH_4 + H_2 + CO$ are plotted in Fig. 3, the curves for the pure gases being included for comparison.

Mixtures of Industrial Gases with Air.—The equimolecular mixture of carbon monoxide and hydrogen correspond nearly with "water-gas." A coal-gas and a producer-gas were also examined, the compositions of these being:

	Coal-gas. Per cent.	Producer-gas. Per cent.
Benzene and higher olefines	1·1	—
Carbon dioxide	0·3	5·0
Ethylene	2·6	—
Carbon monoxide	9·6	21·3
Hydrogen	49·2	12·6
Methane and higher paraffins	33·9	3·1
Nitrogen (by difference)	3·3	58·0

The speeds of the uniform movement of flame in mixtures of air with each of these two gases are given in table VII.

TABLE VII.

Speed of Uniform Movement of Flame in Mixtures of Air with Coal-gas and with Producer-gas in a Tube 2·5 cm. in Diameter.

Coal-gas. Per cent.	Speed, cm. per sec.	Producer-gas. Per cent.	Speed, cm. per sec.
7·2	21·5	24·7	20·0
10·0	50·5	38·9	47·4
11·9	87·1	46·0	62·7
14·7	133·7	49·0	72·2
16·8	153·9	54·3	69·7
17·9	154·1	58·8	43·5
20·4	115·6	61·6	24·0
21·8	74·3		
24·3	22·0		

The principal constituents of the coal-gas are hydrogen, methane, and carbon monoxide. If all the hydrocarbons be reckoned as methane, the calculated maximum speed of uniform movement of flame in mixtures of air with this coal-gas is 164 cm. per second, with a mixture containing 18·4 per cent. of coal-gas. Since the content of inert gases (nitrogen and carbon dioxide) is low, they may be neglected when making the calculations.

Producer-gas, on the other hand, always contains a large proportion of inert gas; the sample used for these experiments contained only 37 per cent. of combustible gas. For this reason, a value for the maximum speed of uniform movement of flame in a mixture of producer-gas and air, calculated from the maximum speeds in mixtures of the pure gases with air, would be too high.

The speed of flame in mixtures of air with gas containing a large proportion of nitrogen can be calculated on the assumption that the cooling or retarding effect on the flame of excess of air or of nitrogen will be the same, since their specific heats are the same.* A mixture of carbon monoxide, hydrogen, and methane in the pro-

* This assumption is not quite correct, since the presence of reactive gas slightly opposes the retarding effect of air.

portions in which they are found in the sample of producer-gas used in these experiments will have as its "fastest-speed" mixture with air one containing 34·7 per cent. of combustible gases. If nitrogen is added to this mixture, so that the ratio of nitrogen to combustible gases is the same as in the producer-gas, the result is a mixture containing 21·7 per cent. of combustible gases. (The carbon dioxide content being low, it may be calculated as nitrogen.) The speed of flame in this mixture should, on the assumption given above, be but little different from the speed of flame in the same mixture of combustible gases with air. The latter speed is most easily determined by a graphical method, and is found to be 85 cm. per second.

The mixture of air and producer-gas with the fastest speed of uniform movement of flame contains slightly more inflammable gases than is required for complete combustion. A greater "displacement" of the maximum-speed mixture might be expected for the reason that the chief inflammable constituents are hydrogen and carbon monoxide, the individual displacements of which are considerable. The small displacement with producer-gas is due to the presence of inert gases, as will be explained in the succeeding section of this series of researches. The effect, in general, of inert gases on the speed of the uniform movement of flame in gaseous mixtures will also be considered.

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