

Second—The helium content of the gas tends to increase as the nitrogen increases, though no direct relation is observed.

Third—In Kansas, lines of iso-helium and iso-paraffin content can be traced across the state and these follow approximately the lines of outcrop of the various geological strata.

Fourth—A very wide variation in composition is noted in the gases examined; hydrocarbons varying from 15.00 per cent. to 98.00 per cent., and helium from a trace up to 1.84 per cent.

LAWRENCE, KANSAS.
August 25, 1907.

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THE IGNITION TEMPERATURES OF GASEOUS MIXTURES.

SECOND PAPER.

BY K. GEORGE FALK.

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Introduction.

In a paper published about a year ago¹ a new method was described for determining the ignition temperatures of gaseous mixtures and the results obtained for mixtures of hydrogen and oxygen were given in detail. The ignition temperature was defined as the temperature at which a gaseous mass would ignite if left entirely to itself for some time, the catalytic action of the walls of the vessel in which the gas was enclosed being eliminated. The new method for determining these temperatures consisted of instantaneously compressing the mixtures enclosed in a suitable vessel. This compression raises the temperature of the gas, and, if carried out with sufficient rapidity, is adiabatic, no heat being lost by radiation or conduction. The rise in temperature of the mixture may then be calculated from the initial and final volumes of the gas, its initial temperature, and the ratio of the specific heats of the gas at constant pressure and constant volume. In compressing the mixtures, as soon as the temperature of ignition is reached there is a sudden reaction (explosion), enough force being developed by the reaction to make the determination of the final volume of the mixture a comparatively simple matter as was shown in the first paper.

In the present paper, the results obtained by this method for mixtures made up of carbon monoxide and oxygen; hydrogen, oxygen and nitrogen; carbon monoxide, oxygen and nitrogen; and carbon monoxide, oxygen and hydrogen are given, and the results obtained before for mixtures of hydrogen and oxygen confirmed. In the results given for mixtures of hydrogen and oxygen in the first paper a small correction was overlooked, but this has been introduced here and will be discussed later.

¹This Journal, 28, 1517.

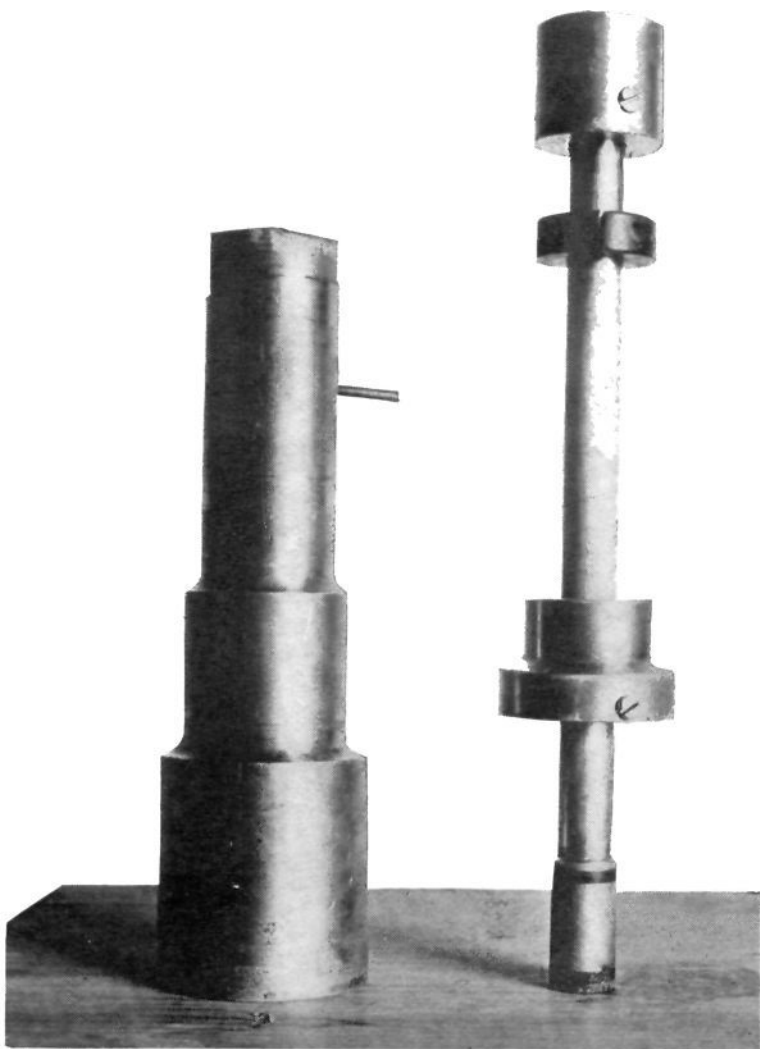


FIG. 1

Although there has been a considerable amount of work done on the determination of the ignition temperatures of mixtures of hydrogen and oxygen, there has been comparatively little with mixtures of other gases. V. Meyer and Krause¹ found the ignition temperature of the mixture $2\text{CO} + \text{O}_2$ to lie between 791° and 879° absolute, using the method of enclosing the mixture in sealed bulbs which were plunged into baths heated to definite temperatures. They found the same ignition temperature for the mixture $2\text{H}_2 + \text{O}_2$. (For a discussion of this method, cf. first paper, p. 1531.) Mallard and Le Chatelier,² by passing the gas into a tube heated to a definite temperature, found the ignition temperature of the mixture containing 70 per cent. carbon monoxide and 30 per cent. oxygen to be about 923° . They also found that diluting this mixture, as well as the detonating gas mixture, with one of the active constituents or with an inert gas, modified the ignition temperature only slightly.

Description of Apparatus and Method of Experimenting.

In the determination of the ignition temperatures of mixtures containing hydrogen and oxygen, two pieces of apparatus (I and II) of different sizes were used. In the work to be described here, two new pieces of apparatus (III and IV) entirely of steel, were constructed. Apparatus III is shown in Figure 1.

Apparatus IV is of exactly the same construction, only larger. The general plan of all four is the same, and since I and II were described in detail before, the cut of III and the dimensions of III and IV will suffice here :

	Apparatus III.	Apparatus IV.
Inner length of cylinder	25.460 cm.	24.142 cm.
Inner diameter of cylinder, and diameter of piston head	2.580 cm.	5.078 cm.
Thickness of walls of upper portion of cylinder	1.325 cm.	1.278 cm.
Thickness of walls of lower (thickest) portion of cylinder	2.823 cm.	3.276 cm.
Distance from lower inner edge of inlet tube to bottom of cylinder	19.500 cm.	18.640 cm.
Thickness of steel at bottom of cylinder	3.514 cm.	5.163 cm.
Length of piston	37.688 cm.	36.405 cm.
Volume of gas measured at atmospheric pressure used in each experiment	101.7 cc.	377.5 cc.

With Apparatus I, 69.8 cc. gas were used, with II, 283.9 cc.

The method of experimenting was essentially the same as used before. The mixture was passed into the cylinder through a thin rubber tube passing through the inlet tube, E, the piston being clamped in position so that the lower edge of the piston head was just above the opening of the inlet tube. The air displaced passed out through the inlet tube around the rubber tubing. As soon as sufficient of the mixture had been

¹ Ann., 264, 85.

² Annales des Mines (8), 4, 274.

run in to ensure of the air being completely displaced, the rubber tube was rapidly withdrawn, and the piston pushed down past the opening of the inlet tube. The height of the piston head from the base was then measured, from which the initial volume of the mixture was calculated. To cause adiabatic compression, a weight was dropped on the piston. The device for accomplishing this is shown in Figure 2, which requires no further explanation.

Apparatus IV is shown here ready for an experiment.

By the compression brought about in this way, the piston was forced down until the heat developed by the compression was sufficient to ignite the gas, when an explosion occurred, the entire mass of gas reacting at once, and the downward motion of the piston stopped. From the position of the movable ring, A, which was forced up as the piston descended, the final volume (*i. e.*, the volume at the instant of ignition), was calculated. The ring was made of fiber board and was clamped to the piston rod by a screw by means of which it could be tightened as much as was desired. The piston head was furnished with two leather rings (as shown in Fig. 1), which were renewed when necessary. Lanoline was again used as lubricant, and the two pieces of apparatus were as air-tight as Apparatus II had been found to be. The initial temperature of the mixture was taken to be the room temperature. The initial pressure was always greater than one atmosphere, and was calculated by dividing the distance from the lower edge of the inlet tube to the bottom of the cylinder by h_1 , the initial height of the enclosed mixture.

The hydrogen used was obtained in cylinders from Eimer & Amend, and was made from zinc and sulphuric acid. The oxygen was prepared by the S. S. White Dental Manufacturing Company for medicinal purposes, and was the purest obtainable. The carbon monoxide was prepared by dropping formic acid (90 per cent.) into concentrated sulphuric acid, pure carbon monoxide being evolved in large amounts very conveniently by this method. When an inert gas (nitrogen) was used, air was taken and the mixtures calculated on the assumption that it consisted of 80 per cent. nitrogen and 20 per cent. oxygen by volume. Although this is not exactly true, the ignition temperature of a mixture whose composition varies at most 2-3 per cent., as is the case here, is practically constant. This is shown most clearly by the curves for the ignition temperatures of mixtures of hydrogen and oxygen and of carbon monoxide and oxygen. The gases were mixed over water in a two-liter graduated glass gasometer, from which they were run directly into the cylinder, passing through a small wash bottle containing water. In this way the gas was always saturated with water at the ordinary temperature.

Apparatus III was first constructed and used, then Apparatus IV was

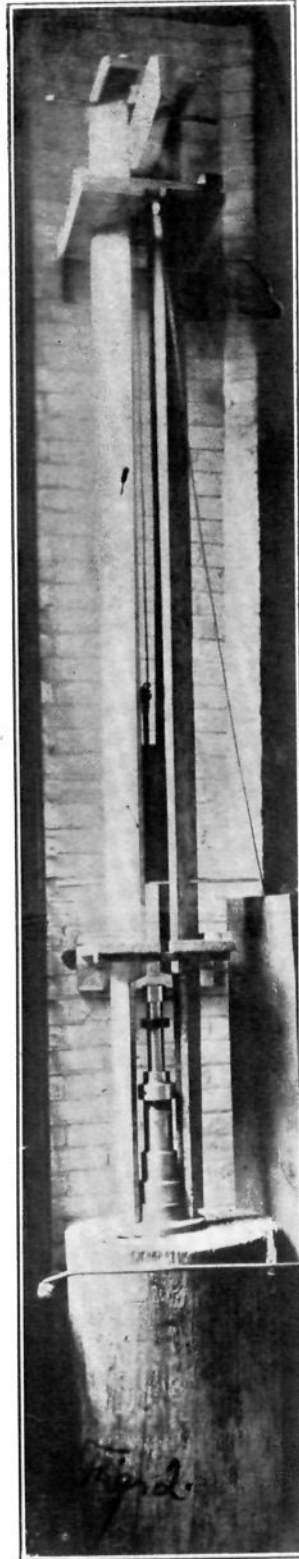


FIG. 2.

constructed in order to have as greatly differing amounts of gases to work with as possible. It was found, however, that the mechanical difficulties, which need not be detailed here, connected with the use of the larger apparatus, due to the large mass of exploding gas and the consequent violence of the explosion, resulted in so much time being taken up for repairs, etc., that after the results shown in Tables 10 and 16 were obtained, after working two months, this apparatus was abandoned temporarily, since the same results were obtained with Apparatus III, which could be handled much more easily. With Apparatus III, a 22.7 kg. weight was used, with Apparatus IV, a 31.8 kg. weight.

The distances were measured with a steel measuring rod graduated directly to 0.5 millimeters, and carrying a circular vernier with which readings to 0.01 millimeter could be made. The initial height was read to 0.1 millimeter, and the final height to 0.01 millimeter. Although the readings may appear to be more accurate than the method apparently justifies, it was thought best to make the actual experimental measurements as exact as possible.

The experimental work was carried out in the laboratory of Professor Charles E. Lucke, of the Mechanical Engineering Department of Columbia University, and it is due to his aid in assisting to overcome the mechanical difficulties, and in having the repairs which were constantly required made, that it was possible to proceed with this investigation as rapidly as has been the case.

Calculation of Results.

The method of calculating is the same as already given. The formulas used are:

$$\frac{T_2}{T_1} = \left(\frac{h_1}{h_2} \right)^{k-1} \quad \text{and} \quad \frac{p_2}{p_1} = \left(\frac{h_1}{h_2} \right)^k$$

in which

T_1 represents the initial (absolute) temperature of the mixture.

T_2 , the ignition temperature (absolute).

h_1 , the height in centimeters of the initial volume of the mixture (heights may be used in place of volumes since the diameter is constant).

h_2 , the final height in centimeters of the mixture (at the ignition temperature).

p_1 , the initial pressure of the gas in atmospheres.

p_2 , the pressure of the gas in atmospheres at the ignition temperature.

k , the ratio of the specific heats of the gases at constant pressure and constant volume, taken as equal to 1.4 for the gases (H_2 , O_2 , CO , N_2) used.¹ Since these are diatomic gases, k is the same for all. This brings up the question of a small error which was overlooked in the first

¹ The possible source of error introduced in this way will be discussed later.

paper. The gases are saturated with water vapor at the ordinary temperature and are then compressed adiabatically. The rise in temperature produced in this way is the same for the diatomic gases, but water vapor being triatomic would not be heated to such a temperature and would consequently lower the temperature of the mixture. Since the gases were saturated at the ordinary temperature (17° - 23°) the amount of water vapor present is perfectly definite and may be corrected for. Since $k=1.29$ for water vapor, the rise in temperature of the water vapor as well as of the other gases present may be calculated, and since their specific heats are known by applying the method of mixtures, the temperature of the mixture at the ignition point may be calculated. This ignition temperature would be lower than the one calculated on the assumption that the water vapor had no effect on the heating, but the difference is small. The correction will be applied in detail to one case. The initial temperature being 293° (20° C.) the temperatures produced by the same compression in a diatomic ($k = 1.4$) and in a triatomic ($k = 1.29$) gas are :

$k = 1.4$	$k = 1.29$
736°	571°
842°	630°
931°	678°
1009°	718°

For the mixture $4\text{H}_2 + \text{O}_2$, Table 1, the average ignition temperature, omitting the correction for water vapor, was calculated to be 874° . The temperature to which water vapor would be raised by the same compression is 647° . One cubic centimeter of the mixture contains 0.072 mg. H_2 , 0.286 mg. O_2 , and 0.0173 mg. H_2O . The specific heat at constant volume of hydrogen at 874° in thousandths of calories is 2.75, of oxygen at 874° , 0.17, and of water vapor at 647° , 0.38.¹ By the method of mixtures², the corrected ignition temperature of the mixture $4\text{H}_2 + \text{O}_2$ is :

$$\frac{(0.0173 \times 0.38 \times 647) + (0.72 \times 2.75 \times 874) + (0.286 \times 0.17 \times 874)}{(0.0173 \times 0.38) + (0.072 \times 2.75) + (0.286 \times 0.17)} = 868^{\circ}$$

The method is exactly the same for the other mixtures. The corrected final average results will only be given, however. The corrections are small and of the same order of magnitude throughout.

Experimental Results.

Only the corrected results for Tables 1-8 are given. The complete results will be taken up and discussed in groups after the possible sources

¹ Mallard and Le Chatelier, *Compt. rend.*, 96, 1358.

² The cooling effect of the water vapor on the gases is of course exerted continuously from the beginning of the compression, but the final result is essentially the same if the total effect is assumed to take place at the final volume, as has been done in the calculations.

of error have been spoken of. The corrected values of T_2 refer to the correction due to water vapor just explained.

TABLE 1.

$4H_2 + O_2$. Apparatus I.
Corrected T_2 — 868.

TABLE 2.

$2H_2 + O_2$. Apparatus I.
Corrected T_2 — 806.

TABLE 3.

$H_2 + O_2$. Apparatus I.
Corrected T_2 — 781.

TABLE 4.

$4H_2 + O_2$. Apparatus II.
Corrected T_2 — 887.

TABLE 5.

$2H_2 + O_2$. Apparatus II.
Corrected T_2 — 814.

TABLE 6.

$H_2 + O_2$. Apparatus II.
Corrected T_2 — 791.

TABLE 7.

$H_2 + 2O_2$. Apparatus II.
Corrected T_2 — 803.

TABLE 8.

$H_2 + 4 O_2$. Apparatus II.
Corrected T_2 — 844.

TABLE 9.

$2H_2 + O_2$. Apparatus III.

h_1	h_2	T_1	T_2	p_1	p_2
14.78	1.076	290.	827.	1.32	51.7
13.48	1.004	291.	822.	1.45	55.0
13.31	0.994	290.	819.	1.47	55.6
13.29	0.979	291.	826.	1.47	56.6
Average		823			
		Corrected T_2 — 818.			

TABLE 10.

$H_2 + O_2$. Apparatus IV.

h_1	h_2	T_1	T_2	p_1	p_2
14.50	1.185	291.	793.	1.29	43.0
		Corrected T_2 — 788.			

TABLE 11.

$H_2 + O_2$. Apparatus III.

h_1	h_2	T_1	T_2	p_1	p_2
14.12	0.852	293.	901.	1.38	70.4
13.92	0.906	294.	877.	1.40	64.2
13.75	0.903	294.	874.	1.42	64.2
Average		884.			
		Corrected T_2 — 878.			

TABLE 12.
6CO + O₂. Apparatus III.

h_1	h_2	T_1	T_2	p_1	p_2
14.88	0.669	289.	1000.	1.31	100.8
14.48	0.651	291.	1006.	1.35	103.8
14.31	0.669	291.	991.	1.36	99.1
13.86	0.575	290.	1036.	1.41	121.4
13.33	0.637	290.	979.	1.46	103.1

Average.....1002.

Corrected T_2 — 994.

TABLE 13.
4CO + O₂. Apparatus III.

h_1	h_2	T_1	T_2	p_1	p_2
15.70	0.900	290.	910.	1.24	67.9
14.86	0.833	289.	915.	1.31	74.0
14.83	0.837	290.	916.	1.31	73.3
14.09	0.869	292.	890.	1.38	68.1
12.73	0.740	289.	902.	1.52	81.6

Average.....907.

Corrected T_2 — 901.

TABLE 14.
2 CO + O₂. Apparatus III.

h_1	h_2	T_1	T_2	p_1	p_2
15.13	0.975	290.	869.	1.29	59.9
14.71	0.891	292.	896.	1.33	67.4
14.63	0.960	290.	862.	1.33	60.3
14.40	0.878	290.	888.	1.36	68.3
13.99	0.908	291.	869.	1.39	67.0
13.80	0.851	293.	893.	1.41	69.8
13.55	0.807	292.	903.	1.44	74.7
13.47	0.887	292.	867.	1.45	65.4
12.37	0.785	291.	877.	1.58	75.0

Average.....880.

Corrected T_2 — 874.

TABLE 15.
CO + O₂. Apparatus III.

h_1	h_2	T_1	T_2	p_1	p_2
15.39	0.860	288.	913.	1.27	72.1
15.35	0.938	292.	893.	1.27	63.6
14.68	0.857	289.	901.	1.33	71.0
14.49	0.861	290.	897.	1.35	70.3
13.87	0.825	290.	897.	1.41	73.3
13.85	0.782	290.	916.	1.41	78.8

Average.....903.

Corrected T_2 — 897.

TABLE 16.
CO + O₂. Apparatus IV.

h_1	h_2	T_1	T_2	p_1	p_2
14.88	0.811	289.	925.	1.25	73.4
14.25	0.827	292.	912.	1.31	70.5

Average.....918.

Corrected T_2 — 912.

TABLE 17.
 $H_2 + O_2 + N_2$. Apparatus III.

h_1	h_2	T_1	T_2	p_1	p_2
13.90	1.083	294.	816.	1.40	50.0
13.89	1.046	293.	824.	1.40	52.5
13.78	1.041	294.	826.	1.42	52.6
13.64	1.003	293.	832.	1.43	55.2
Average.....825.					
Corrected T_2 — 820.					

TABLE 18.
 $H_2 + O_2 + 2N_2$. Apparatus III.

h_1	h_2	T_1	T_2	p_1	p_2
14.60	0.982	292.	860.	1.34	58.5
14.16	0.913	293.	877.	1.38	63.9
14.14	1.007	294.	846.	1.38	55.7
13.80	0.971	293.	847.	1.41	58.1
13.75	0.962	293.	849.	1.42	58.7
Average.....856.					
Corrected T_2 — 851.					

TABLE 19.
 $H_2 + O_2 + 4N_2$. Apparatus III

h_1	h_2	T_1	T_2	p_1	p_2
14.18	0.863	294.	901.	1.38	69.2
14.15	0.808	294.	924.	1.38	75.8
13.92	0.802	294.	921.	1.40	76.1
13.85	0.786	294.	926.	1.41	78.2
13.67	0.815	294.	908.	1.43	73.9
Average.....916.					
Corrected T_2 — 910.					

TABLE 20.
 $2H_2 + O_2 + N_2$. Apparatus III.

h_1	h_2	T_1	T_2	p_1	p_2
14.65	1.061	294.	840.	1.33	52.5
14.22	1.035	294.	839.	1.37	53.7
14.00	0.957	294.	860.	1.39	59.6
13.84	0.895	292.	873.	1.41	65.2
13.58	0.953	294.	849.	1.44	59.2
Average.....852.					
Corrected T_2 — 846.					

TABLE 21.
 $2H_2 + O_2 + 4N_2$. Apparatus III.

h_1	h_2	T_1	T_2	p_1	p_2
14.27	0.797	294.	930.	1.37	77.5
14.16	0.829	294.	915.	1.38	73.2
14.10	0.813	294.	921.	1.38	75.1
13.95	0.793	294.	926.	1.40	77.4
13.90	0.736	294.	952.	1.44	87.3
Average.....929					
Corrected T_2 — 922.					

TABLE 22.
 $H_2 + 2 O_2 \rightarrow N_2$, Apparatus III.

h_1	h_2	T_1	T_2	p_1	p_2
14.10	1.009	294.	844.	1.38	55.5
14.08	0.974	294.	856.	1.38	58.3
14.06	1.051	294.	830.	1.39	52.4
14.04	1.025	294.	838.	1.39	54.8
Average.....842.					
Corrected $T_2 = 837$.					

 TABLE 23.
 $H_2 + 2 O_2 \rightarrow 4 N_2$, Apparatus III.

h_1	h_2	T_1	T_2	p_1	p_2
14.26	0.833	293.	913.	1.37	72.9
14.18	0.849	293.	904.	1.38	70.8
13.90	0.753	294.	944.	1.40	83.1
13.85	0.824	294.	909.	1.41	73.2
13.20	0.734	293.	931.	1.48	84.4
Average.....920.					
Corrected $T_2 = 914$.					

 TABLE 24.
 $2 CO + O_2 \rightarrow N_2$, Apparatus III.

h_1	h_2	T_1	T_2	p_1	p_2
14.03	0.806	293.	919.	1.39	75.9
13.90	0.819	293.	910.	1.40	73.9
13.78	0.786	293.	921.	1.42	78.0
13.28	0.719	293.	941.	1.47	87.1
Average.....923.					
Corrected $T_2 = 917$.					

 TABLE 25.
 $2 CO + O_2 \rightarrow 2 N_2$, Apparatus III.

h_1	h_2	T_1	T_2	p_1	p_2
14.05	0.708	293.	968.	1.39	91.0
13.72	0.691	293.	968.	1.42	93.3
13.22	0.712	293.	943.	1.48	88.1
12.95	0.631	293.	981.	1.51	103.4
Average.....965.					
Corrected $T_2 = 958$.					

 TABLE 26.
 $CO + O_2 \rightarrow N_2$, Apparatus III.

h_1	h_2	T_1	T_2	p_1	p_2
14.12	0.691	293.	980.	1.38	94.3
13.96	0.689	293.	976.	1.40	94.3
13.63	0.621	293.	1008.	1.43	108.0
13.37	0.656	293.	979.	1.46	99.3
Average.....986.					
Corrected $T_2 = 979$.					

 TABLE 27.
 $CO + O_2 \rightarrow 2 N_2$, Apparatus III.

h_1	h_2	T_1	T_2	p_1	p_2
14.96	0.544	293.	1103.	1.30	134.9
14.38	0.496	291.	1119.	1.36	151.2
13.98	0.581	292.	1042.	1.39	119.8
12.48	0.441	292.	1112.	1.56	168.4
Average.....1094.					
Corrected $T_2 = 1085$.					

TABLE 28.

4CO + O₂ + N₂. Apparatus III.

h_1	h_2	T ₁	T ₂	ρ_1	ρ_2
14.53	0.773	292.	944.	1.34	81.6
13.96	0.771	292.	930.	1.40	80.6
13.94	0.789	292.	921.	1.40	78.0
13.34	0.735	292.	931.	1.46	84.6

Average.....932.

Corrected T₂ — 925.

TABLE 29.

4 CO + O₂ + 2N₂. Apparatus III.

h_1	h_2	T ₁	T ₂	ρ_1	ρ_2
15.22	0.819	292.	940.	1.28	76.6
14.58	0.787	292.	939.	1.34	79.6
13.90	0.733	292.	947.	1.40	86.1
13.63	0.692	292.	962.	1.43	92.8

Average.....947.

Corrected T₂ — 940.

TABLE 30.

H₂ + O₂ + CO. Apparatus III.

h_1	h_2	T ₁	T ₂	ρ_1	ρ_2
14.30	1.081	294.	826.	1.36	50.7
14.13	1.075	294.	824.	1.38	50.8
14.02	1.104	294.	813.	1.39	48.8
13.98	1.093	294.	815.	1.39	49.5
13.76	1.075	294.	815.	1.42	50.3
13.74	1.101	294.	807.	1.42	48.6

Average.....817

Corrected T₂ — 812

TABLE 31.

H₂ + O₂ + 2CO. Apparatus III.

h_1	h_2	T ₁	T ₂	ρ_1	ρ_2
14.19	0.974	294.	858.	1.37	58.5
14.16	1.019	294.	842.	1.38	54.8
14.03	0.965	294.	858.	1.39	58.9
13.94	0.923	294.	871.	1.40	62.6
13.63	0.955	294.	851.	1.43	59.1

Average.....856.

Corrected T₂ — 851.

TABLE 32.

H₂ + O₂ + 4 CO. Apparatus III.

h_1	h_2	T ₁	T ₂	ρ_1	ρ_2
14.24	0.891	294.	891.	1.37	66.3
13.93	0.831	294.	908.	1.40	72.5
13.90	0.822	294.	911.	1.40	73.5
13.84	0.815	294.	913.	1.41	74.3
13.78	0.852	294.	895.	1.42	69.7

Average.....904.

Corrected T₂ — 898.

TABLE 33.

2H ₂ + O ₂ + 2 CO. Apparatus III.						
<i>h</i> ₁	<i>h</i> ₂	T ₁	T ₂	<i>p</i> ₁	<i>p</i> ₂	
14.63	0.883	294.	904.	1.33		67.9
14.18	0.921	293.	875.	1.38		63.2
13.91	0.862	293.	891.	1.40		68.8.
13.86	0.901	293.	874.	1.43		64.6
13.54	0.893	294.	872.	1.44		64.8
Average.....883.						
Corrected T ₂ — 877.						

TABLE 34.

2H ₂ + O ₂ + 4 CO. Apparatus III.						
<i>h</i> ₁	<i>h</i> ₂	T	T ₂	<i>p</i> ₁	<i>p</i> ₂	
14.19	0.749	294.	954.	1.37		84.5
14.09	0.756	293.	944.	1.38		83.1
13.78	0.771	294.	932.	1.42		80.1
13.76	0.716	294.	959.	1.42		88.8
13.66	0.750	293.	936.	1.43		83.0
Average.....945.						
Corrected T ₂ — 938.						

TABLE 35.

H ₂ + 2O ₂ + 2 CO. Apparatus III.						
<i>h</i> ₁	<i>h</i> ₂	T ₁	T ₂	<i>p</i> ₁	<i>p</i> ₂	
14.02	0.942	293.	863.	1.39		61.0
13.98	0.857	293.	895.	1.39		69.6
13.87	0.947	293.	857.	1.41		60.2
13.86	0.919	293.	867.	1.41		62.3
13.83	0.855	293.	893.	1.41		69.5
Average.....875.						
Corrected T ₂ — 869.						

TABLE 36.

H ₂ + 2 O ₂ + 4 CO. Apparatus III.						
<i>h</i> ₁	<i>h</i> ₂	T ₁	T ₂	<i>p</i> ₁	<i>p</i> ₂	
14.42	0.845	292.	908.	1.35		71.8
13.98	0.895	292.	877.	1.39		65.4
13.91	0.844	292.	896.	1.40		70.9
Average.....894.						
Corrected T ₂ — 888.						

TABLE 37.

2 H ₂ + 3 O ₂ + 2 CO. Apparatus III.						
<i>h</i> ₁	<i>h</i> ₂	T ₁	T ₂	<i>p</i> ₁	<i>p</i> ₂	
14.59	1.051	291.	834.	1.34		53.1
13.75	1.039	291.	818.	1.42		52.7
13.17	0.947	290.	831.	1.48		59.0
12.61	0.887	289.	836.	1.55		63.6
Average.....830.						
Corrected T ₂ — 825.						

The initial temperatures of the mixtures can have no effect on their ignition temperatures. This was shown in the first paper and the proof will therefore not be repeated. The effect of the pressure of the gas in any mixture on the ignition temperature of this mixture is so slight, if it exists

at all, that it could not be determined from the experiments. (This, of course, refers directly only to the high pressures at which the ignition temperatures were determined, but theoretically, as shown in the first paper, the pressure should cause only small differences in these temperatures).

Possible Sources of Error.

In discussing the possible sources of error, the method of calculating is the first to suggest itself, as to whether it is justifiable to assume k equal to 1.4 between the ordinary temperature and the temperature of ignition. Although the earlier work on this subject showed a considerable decrease in k with rising temperature, more recent work especially by Kalähne¹ indicated a different result. Kalähne found, working with air by the "resonance method" that at 723° (450° C.) no decrease in k could be observed, and that at 1173° (900° C.) $k=1.39$, the decrease being so small that only the most accurate measurements would show it. There has been no recent work on hydrogen and carbon monoxide at high temperatures, but the results obtained for air can doubtless be applied to them as well. It appears then that k varies from a value slightly greater than 1.40 to a value of about 1.395 for the temperatures used here. Increase in pressure, however, increases the value of k slightly, so that 1.4 differs from the true value of k as used in this work by not more than 0.1 per cent. The correction introduced by the water vapor has already been spoken of and applied to the results. The ignition temperatures are all affected equally by it.

In the compressions used in this work, there is a possibility of waves of compression and reflection being set up in the cylinder during the downward motion of the piston.² These waves would cause the temperature of the mixture to be higher than that calculated by the ordinary adiabatic formula. There is no expression which takes into account these waves, which may be considerable, and if so, would invalidate the results. The experimental results show, however, that this is not the case. The initial height, h_1 , varied for the different mixtures and even for the same mixture. The effect caused by the waves would differ greatly for the different heights, if this effect was in any way large. The experimental work, however, shows that no regular differences can be perceived with different values of h_1 , so that this effect must be very small. Whatever error is introduced in this way, would make the calculated temperatures slightly too low.

In dropping the weight on the piston, there is a possibility of the explosion not developing sufficient force to stop the downward motion of

¹ Ann. Phys., 11, 225 (1903).

² The possibility of this and the following error were also pointed out to the writer by Professor E. Jouguet.

the piston immediately. This has already been spoken of (cf. first paper), and it was shown (in detail in Table 2), that dropping the weight from different heights made no practical difference in the ignition temperatures. There remains the possibility of a small constant error, however. This would tend to make the calculated temperatures slightly higher than the actual ignition temperatures, that is, the error would lie in the opposite direction from the former and would tend to neutralize it. These two sources of error, if they exist at all, must be small and act in opposite directions, so that the effect on the ignition temperatures as calculated is entirely negligible.

That there is no radiation or loss of heat in any other way during compression is true because the four pieces of apparatus differing so much in size give essentially the same results.

Discussion of Results.

In the following discussion of results, when the ignition temperature of a mixture was determined in more than one apparatus, the average result obtained with the different sizes was taken. It is impossible to state with certainty that one piece of apparatus gave more accurate results than another, and in any case the results differ by such small quantities that any other way of averaging would show no appreciable difference. The numbers in parentheses refer to the tables which give the details of the experiments.

TABLE A.
HYDROGEN AND OXYGEN.

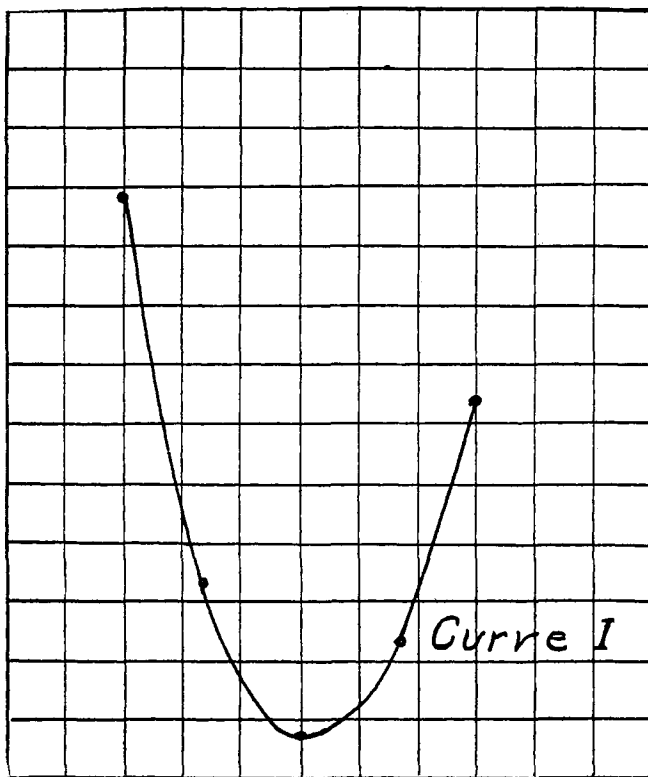
	T_1
$4\text{H}_2 + \text{O}_2$ (1, 4, 11)	878
$2\text{H}_2 + \text{O}_2$ (2, 5, 9)	813
$\text{H}_2 + \text{O}_2$ (3, 6, 10)	787
$\text{H}_2 + 2\text{O}_2$ (7)	803
$\text{H}_2 + 4\text{O}_2$ (8)	844

The following curve (I) shows the ignition temperatures of the different mixtures of hydrogen and oxygen.

The fact that the maximum affinity is shown by the mixture $\text{H}_2 + \text{O}_2$, proves that H_2O_2 must be the first product of the reaction between hydrogen and oxygen. This was spoken of in detail in the first paper.

TABLE B.
CARBON MONOXIDE AND OXYGEN.

	T_1
$6\text{CO} + \text{O}_2$ (12)	994
$4\text{CO} + \text{O}_2$ (13)	901
$2\text{CO} + \text{O}_2$ (14)	874
$\text{CO} + \text{O}_2$ (15, 16)	904



From the curve (II) and by extrapolation as shown in the dotted portion, the ignition temperatures of different mixtures of carbon monoxide and oxygen can be determined.

The maximum affinity is shown by the mixture $2\text{CO} + \text{O}_2$, doubling the carbon monoxide or the oxygen, causing the same rise in the ignition temperatures of the resulting mixtures.

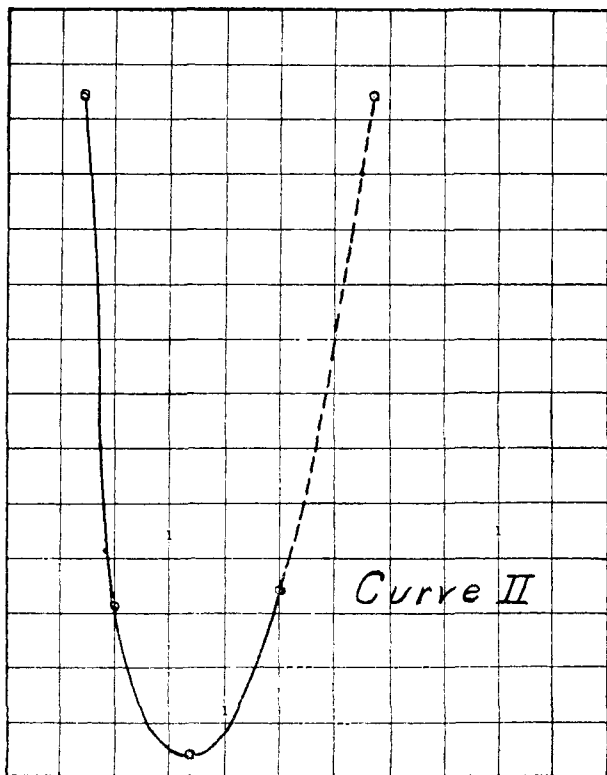
TABLE C.
HYDROGEN, OXYGEN AND NITROGEN.

	T_2 (obs.)	T (calc.)
$\text{H}_2 + \text{O}_2 + \text{N}_2$ (17)	820	817
$\text{H}_2 + \text{O}_2 + 2\text{N}_2$ (18)	851	847
$\text{H}_2 + \text{O}_2 + 4\text{N}_2$ (19)	910	907
$2\text{H}_2 + \text{O}_2 + \text{N}_2$ (20)	846	843
$2\text{H}_2 + \text{O}_2 + 4\text{N}_2$ (21)	922	933
$\text{H}_2 + 2\text{O}_2 + \text{N}_2$ (22)	837	833
$\text{H}_2 + 2\text{O}_2 + 4\text{N}_2$ (23)	914	923

The values of T (calc.) are obtained by means of the empirical equation :

$$(a) \quad T = T_c + 3cn,$$

T denoting the calculated ignition temperatures, T_c the ignition temperatures of the pure hydrogen-oxygen mixtures as obtained from Curve I,



and n , the volume of the inert gas divided by the lesser volume, of the two active constituents.

$$\left(n = \frac{\text{volume inert gas}}{\text{volume of H}_2 \text{ or O}_2 \text{ (whichever is present in smaller amount)}} \right)$$

Since nitrogen acts only as an inert gas, the above equation (a) taken in conjunction with curve I, allows the ignition temperature of any mixture of hydrogen and oxygen, alone or in the presence of an inert gas to be calculated very simply. This equation does not say anything about the limit of inflammability, but such a limit must exist for the mixtures containing a large excess of one of the constituents.

TABLE D.
CARBON MONOXIDE, OXYGEN AND NITROGEN.

	T_c (obs.)	T (calc.)
2 CO + O ₂ + N ₂ (24).....	917	914
2 CO + O ₂ + 2N ₂ (25).....	958	954
CO + O ₂ + N ₂ (26).....	979	984
CO + O ₂ + 2N ₂ (27).....	1085	1064
4 CO + O ₂ + N ₂ (28).....	925	921
4 CO + O ₂ + 2N ₂ (29).....	940	941

The values T (calc.) are obtained by means of the empirical equation

$$(b) \quad T = T_c + 80n'$$

in which T denotes the calculated ignition temperatures, T_c , the ignition temperatures of the mixtures without the inert gas as obtained from curve II, and $n' = \frac{\text{volume inert gas.}}{\text{volume carbon monoxide.}}$ With the aid of

curve II and equation (b), the ignition temperature of any mixture of carbon monoxide and oxygen, alone and in the presence of an inert gas, may be calculated up to the point where ignition is impossible.

TABLE E.

HYDROGEN, CARBON MONOXIDE AND OXYGEN.

	T (obs.)	T_a (calc.)	T_b (calc.)
$H_2 + O_2 + CO$ (30)	812	817	984
$H_2 + O_2 + 2CO$ (31)	851	847	914
$H_2 + O_2 + 4CO$ (32)	898	907	921
$2H_2 + O_2 + 2CO$ (33)	877	873	954
$2H_2 + O_2 + 4CO$ (34)	938	933	941
$H_2 + 2O_2 + 2CO$ (35)	869	863	944
$H_2 + 2O_2 + 4CO$ (36)	888	923	894
$2H_2 + 3O_2 + 2CO$ (37)	825	822	1030

The values under T_a (calc.) were obtained from equation (a) and curve I, assuming carbon monoxide to act as a neutral gas, under T_b (calc.) from equation (b) and curve II, assuming hydrogen to be neutral. As seen from the table, the experimentally determined value agrees in every case with the lower of the calculated values, indicating that the ignition temperature is fixed by two of the constituents, the third simply acting as an inert gas. This evidently holds also if nitrogen, carbon dioxide, or any other inert gas is present, so that to calculate the ignition temperature of any mixture of hydrogen, carbon monoxide, oxygen, and an inert gas, it is simply necessary to calculate first the ignition temperature of the hydrogen and oxygen, assuming the rest of the mixture to be inert (curve I, equation (a)), and then, to do the same with the carbon monoxide and oxygen with the other constituents inert (curve II, equation (b)). The lower of these two temperatures will be the required ignition temperature of the mixture.

A number of experiments were tried with hydrogen and nitrogen, nitrogen and oxygen, and carbon monoxide and hydrogen mixtures, but in no case could any sign of a reaction having taken place under the conditions described, be perceived.

Producer gas which is used in gas engines consists essentially of hydrogen, carbon monoxide, oxygen, nitrogen, and carbon dioxide. The results given in this paper show how the ignition temperature of any such mixture may be calculated. A general discussion of this phase of the subject would not be of much value without a large number of

analyses of the gas and a complete account of the working of the engine.

Some light may also be thrown on the causes of explosions in mines by the results given here.

Theoretical.

Consider a bimolecular reaction taking place in a homogeneous gaseous system. Let r denote a definite volume of the mixture at the ignition temperature, x the part by volume of one of the constituents, y that of the other ($x + y = r$). Then, if v denotes the velocity of the reaction at the ignition temperature, and k the corresponding velocity constant,

$$v = kxy = kx(r - x) \quad \therefore \frac{dv}{dx} = -kx + k(r - x) = 0 \quad \therefore x = \frac{r}{2} \quad \text{and} \quad y = \frac{r}{2};$$

or, for the velocity at the ignition temperature to be a maximum, the two constituents must be present in equal volumes or parts by volume. The maximum velocity of reaction evidently corresponds to the greatest affinity, or to the lowest ignition temperature of a mixture containing these two constituents. Conversely, if a mixture of two gases shows the lowest ignition temperature when these gases are present in equal volumes, the reaction between them must be bimolecular. The lowest ignition temperature for mixtures containing oxygen and hydrogen was found for the mixture containing equal volumes of the two gases. The reaction between them is therefore one of the second order. Bodenstein¹ measured directly the velocity of the reaction between hydrogen and oxygen and concluded it to be trimolecular, but Rowe² on recalculating Bodenstein's results, found the constant to agree just as well for a bimolecular as for a trimolecular reaction. He assumes, however, that since hydrogen peroxide is decomposed with very great rapidity at higher temperatures³ it is incapable of being formed at all at these temperatures, and that the reaction is therefore trimolecular. The results obtained by determining the ignition temperatures of the different mixtures of hydrogen and oxygen, show that the reaction must be bimolecular and that hydrogen peroxide is the first product formed, although it may decompose again immediately.

The ignition temperature corresponds to a certain definite reaction velocity. If an inert gas is present, the velocity of the reaction between the two gases will be less at the former ignition temperature, and a higher temperature will be required to reach the required velocity and cause ignition.

With the bimolecular reaction just considered, $v = kx(r - x)$ or with

¹ Z. physik. Chem., 29, 665.

² Z. physik. Chem., 59, 41. In the review of this paper in Chemical Abstracts 1907, 1662, owing to a printer's error, "bimolecular" appears in place of "trimolecular."

³ Nernst, Z. physik. Chem., 46, 720.

equal volumes of the gases present, $x = \frac{r}{2}$, and $v = \frac{kr^2}{4}$. Assume an inert gas, occupying one-third of the volume r to be present. Then $x_1 + y_1 = \frac{2r}{3}$, and for the maximum velocity v_1 , $x_1 = y_1 = \frac{r}{3}$. Then $v_1 = k_1 x_1 y_1 = k_1 x_1 \left(\frac{2r}{3} - x_1 \right)$. At the ignition temperature of the mixture containing no inert gas, $k_1 = k$, and $v_1 = \frac{kr^2}{9}$, or the velocity of the

reaction with the inert gas present will be $\frac{4}{9}$ thus $\left(\text{since } \frac{v_1}{v} = \frac{\frac{9}{kr^2}}{\frac{kr^2}{4}} = \frac{4}{9} \right)$

of the reaction velocity of the pure mixture at the ignition temperature of the latter. To increase the velocity of the former until the velocity of the latter (or ignition) is attained will require a rise in temperature. Experimentally this was found to be 30° (equation (a)). A rise in temperature of 10° for this mixture increases the velocity of the reaction by, $\sqrt[3]{\frac{9}{4}}$, or the velocity coefficient is equal to 1.31 between the limits 787° and 820° (the ignition temperatures of the mixtures $\text{H}_2 + \text{O}_2$ and $\text{H}_2 + \text{O}_2 + \text{N}_2$).

The effect of the pressure on the ignition temperature, which must also be considered, should be small according to the theoretical view developed by Nernst.¹ This is born out by the experiments on any one mixture, but it must be remembered that the final pressures, p_2 given in the tables represent only an upper limit, since whatever leakage took place between the time of enclosing the gas in the cylinder and of igniting it, tended to decrease the value of p_1 , the initial pressure, and therefore also of p_2 . The effect of these pressures on comparing the reaction velocities of different mixtures has been entirely disregarded in the theoretical treatment, but this effect would decrease the velocity coefficient for a 10° rise only slightly, as the pressures at which the ignition temperatures were determined were high and with the exception of a few mixtures, of approximately the same magnitude.

Returning to the temperature coefficient of the reaction velocity, the following table shows some of the values calculated, together with the ignition temperatures of the mixtures which show within what temperature limits these values hold :

	Temperature coefficient per 10° rise	Between the temperature limits
$(\text{H}_2 + \text{O}_2 \text{ and } \text{H}_2 + \text{O}_2 + \text{N}_2)$	1.31	$787. - 820.$
$(\text{H}_2 + \text{O}_2 + \text{N}_2 \text{ and } \text{H}_2 + \text{O}_2 + 2\text{N}_2)$	1.21	$820. - 851.$
$(\text{H}_2 + \text{O}_2 + 2\text{N}_2 \text{ and } \text{H}_2 + \text{O}_2 + 3\text{N}_2)$	1.16	$851. - 880. (\text{approx.})$
$(\text{H}_2 + \text{O}_2 + 3\text{N}_2 \text{ and } \text{H}_2 + \text{O}_2 + 4\text{N}_2)$	1.13	$880. - 910.$

¹ This Journal, 28, :519.

The temperature coefficient decreases from 1.31 at about 800° to 1.13 at about 900°. A list of the ratios of the velocities for a number of reactions at intervals of 10° is given by van't Hoff.¹ These ratios do not differ much for different reactions, and are almost all between 2. and 3. for temperatures not far removed from the ordinary temperature. For two reactions, however, the ratios are given for higher temperatures; for the decomposition of phosphine between the temperatures 583° and 785°, the ratio per 10° rise is 1.2, and for the decomposition of arsine, the ratio between 529° and 640° per 10° rise is 1.23. These temperature coefficients (or ratios) are very nearly the same as those found for the reaction between hydrogen and oxygen.

The effect of adding an excess of one of the active constituents will now be considered. If one of these (x) is present in double the quantity

of the other (y), then $x + y = r$, but $x = \frac{2y}{3}$ and $y = \frac{r}{3}$ and v_2

$k_2 x y = \frac{2k_2 y^2}{9}$ (since the reaction is bimolecular). Applied to the

hydrogen-oxygen mixtures for equal volumes of hydrogen and oxygen

$v = \frac{k r^2}{4}$, and therefore at the same temperature ($k = k_2$), $\frac{v_2}{v} = \frac{8}{9}$

For the mixtures $H_2 + O_2$ and $H_2 + O_2 + N_2$ as shown before $\frac{v_1}{v} =$

$\frac{k r^2}{4}$

$\frac{9}{k r^2} = \frac{4}{9}$, a rise of 30° being sufficient to increase the velocity of v_1 so

that it becomes equal to v . The ratio of $\frac{v_2}{v}$ is half as great as the ratio

$\frac{v_1}{v}$ so that half the rise in temperature, or 15°, should equalize the velocities.

With the mixture $4H_2 + O_2$ or $H_2 + 4O_2$, similarly $v_3 = k_3$.

$\frac{4r}{5} \cdot \frac{r}{5} = \frac{4k_3 r^2}{25}$, and when $k_3 = k$, $\frac{v_3}{v} = \frac{16}{25}$. Since 15° corresponds to

an increase in velocity of $\frac{1}{9}$, to correspond to an increase of $\frac{9}{25}$, $15 \times$

$\frac{9}{25}$

$\frac{25}{1} = 49^\circ$ should be required. The calculated and experimentally

$\frac{1}{9}$

determined temperatures are as follows:

¹ Chemical Dynamics, Translation by Lehfeldt, p. 228.

Experimental		Calculated
H ₂ + O ₂	787.	} 802.
H ₂ + 2O ₂	803.	
2H ₂ + O ₂	813.	} 836.
H ₂ + 4O ₂	844.	
4H ₂ + O ₂	878.	

The ignition temperatures of the mixtures containing an excess of oxygen agree fairly well with the calculated ignition temperatures, those containing an excess of hydrogen do not agree at all. In fact the mixtures containing an excess of hydrogen agree better if the hydrogen in excess of that required to react with the oxygen is assumed to be inert, the mixture H₂ + O₂ + 3H₂ corresponding to the mixture H₂ + O₂ + 3N₂, etc. There is at present no explanation for this abnormality (from the theoretical point of view) on the part of the hydrogen.¹ The only certain feature appears to be the fact that an excess of hydrogen or oxygen should affect the ignition temperature of the mixture H₂ + O₂ equally which is not the case, and to judge from theoretical considerations and from the experimental work, the cause of the abnormality is the hydrogen. It must be borne in mind, however, that the methods of calculating the ignition temperatures of different mixtures (Curve I, Equation (a)) based upon purely experimental work, hold true, even if the theoretical views at present are not sufficiently developed to support the conclusions arrived at from the experiments.

The same method of treatment may be applied to a trimolecular reaction. Since the experimental data are at hand for the reaction between carbon monoxide and oxygen, a reaction in which two molecules of one constituent react with one of another will only be considered, although, of course, the reasoning may be applied to other cases as well. Let x represent the amount of the former substance present and y of the latter, both measured as fractions of a definite volume r , then the velocity of the reaction at the ignition temperature will be (k being constant for this temperature):—

$$v = kx^2y = kx^2(r - x),$$

$$\frac{dv}{dx} = 2kx(r - x) - kx^2 = 0, \text{ and } x = 0, \text{ or } \frac{2}{3}r.$$

$x = 0$ has no physical significance, but for $x = \frac{2}{3}r$, v will be a maximum. One substance must therefore be present in twice the quantity (by volume) of the other for the reaction to proceed with the maximum velocity in a trimolecular reaction, and conversely, a reaction which proceeds with the maximum velocity at any given temperature when two volumes of one substance react with one of another must be one of the

¹ Greater radiation from the mixtures containing a large amount of hydrogen cannot be the cause as the mixture 2H₂ + O₂ + 4N₂ among others shows the same relative behavior.

third order. The reaction between carbon monoxide and oxygen where the maximum velocity (or lowest ignition temperature) is shown by the mixture $2\text{CO} + \text{O}_2$ is trimolecular, and the reaction between hydrogen and oxygen where the maximum velocity is shown not by the mixture $2\text{H}_2 + \text{O}_2$ but by the mixture $\text{H}_2 + \text{O}_2$ cannot be trimolecular.

The value of v at the ignition temperature for the mixture $2\text{CO} + \text{O}_2$ is $v = k\left(\frac{2r}{3}\right)^2 \frac{r}{3} = \frac{4kr^3}{27}$. If inert gas is added to take up one-fourth of the volume, the velocity will be $v_1 = k_1\left(\frac{r}{2}\right)^2 \frac{r}{4} = \frac{k_1 r^3}{16}$, or at the ignition temperature of the first mixture, $k_1 = k$, and $\frac{v_1}{v} = \frac{27}{64}$. At the same temperature the velocity of reaction of the mixture $2\text{CO} + \text{O}_2 + \text{N}_2$ will be $\frac{27}{64}$ as great as that of the mixture $2\text{CO} + \text{O}_2$. Experimentally (equation (b)) it was found that a rise in temperature of 40° would cause the same velocity for the former mixture as for the latter, ignition being brought about then in both cases. For a 10° rise, therefore, between the limits of the ignition temperatures of these mixtures, the temperature coefficient is $\sqrt[4]{\frac{64}{27}} = 1.24$. For other mixtures the following values are obtained:

	Temperature coefficient per 10° rise	Between the temperature limits
$(2\text{CO} + \text{O}_2 \text{ and } 2\text{CO} + \text{O}_2 + \text{N}_2)$	1.24	874. — 917.
$(2\text{CO} + \text{O}_1 + \text{N}_2 \text{ and } 2\text{CO} + \text{O}_2 + 2\text{N}_2)$	1.18	917. — 958.
$(2\text{CO} + \text{O}_2 + 2\text{N}_2 \text{ and } 2\text{CO} + \text{O}_2 + 3\text{N}_2)$	1.14	958. — 1000 (approx.)

The temperature coefficient decreases from 1.24 at about 900° to 1.14 at about 1000° , values which are very close to those obtained for the reaction between hydrogen and oxygen.

The attempt to apply these principles to mixtures of carbon monoxide and oxygen containing an excess of one of the active constituents will not be made here, as there are some apparently contradictory and inexplicable features in the ignition temperatures of these mixtures. For instance, the ignition temperature of the mixture $6\text{CO} + \text{O}_2$ is 994° , but replacing part of the carbon monoxide by nitrogen lowers the ignition temperature so that for the mixture $4\text{CO} + \text{O}_2 + 2\text{N}_2$ the value 940° is found. An excess of carbon monoxide above a certain amount has a negative catalyzing effect.

Bodenstein and Ohlmer¹ found the same effect on measuring the velocity of the reaction on quartz surfaces between carbon monoxide and oxygen, an excess of carbon monoxide retarding the reaction. As shown

¹ Z. phys. Chem., 53, 166.

above, hydrogen appears to exert a similar negative catalyzing effect, but not as pronounced, on the reaction between hydrogen and oxygen.

Conclusions.

The ignition temperatures of mixtures of:

Hydrogen and oxygen, carbon monoxide and oxygen, hydrogen, oxygen and nitrogen, carbon monoxide, oxygen and nitrogen, and hydrogen, carbon monoxide and oxygen, were determined by the "adiabatic compression method," and the possible sources of error fully discussed.

It is shown how the ignition temperature of any gaseous mixture containing hydrogen, carbon monoxide, oxygen, and an inert gas, may be calculated.

The reaction between hydrogen and oxygen is shown to be bimolecular and between carbon monoxide and oxygen, trimolecular.

The velocity coefficient per 10° rise in temperature is calculated to be 1.31 at about 800° and 1.13 at about 900° for the reaction between hydrogen and oxygen, and 1.24 at about 900° and 1.14 at about 1000° for the reaction between carbon monoxide and oxygen.

The reactions between hydrogen and oxygen in the presence of an excess of hydrogen and between carbon monoxide and oxygen in the presence of an excess of carbon monoxide do not follow the simple laws of chemical kinetics.

LABORATORY OF PHYSICAL CHEMISTRY,
July, 1907.

[CONTRIBUTIONS FROM THE RESEARCH LABORATORY OF PHYSICAL CHEMISTRY OF
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SOLUTIONS OF METALS IN NON-METALLIC SOLVENTS; I. GENERAL PROPERTIES OF SOLUTIONS OF METALS IN LIQUID AMMONIA.

By CHARLES A. KRAUS.
Received September 11th, 1907.

INTRODUCTION.

The present investigation was undertaken for the purpose of obtaining evidence regarding the nature of the process of electrical conduction in metals. In order to indicate in what respect the solutions of metals in non-metallic solvents are of especial importance in connection with the problem of metallic conduction, it will be necessary to consider, briefly, the experimental aspects of this problem, particularly in relation to the methods to be employed in its attack.

To account for the electromagnetic properties of metals, especially for the relation between electrical and thermal conduction, the electron theory of metallic conduction has been advanced. According to this theory, the passage of electricity through a metallic conductor is effected by the movement of charged carriers, the electrons, whose mass, relative to charge, is many times smaller than that of the hydrogen ion, and whose