

Summary

1. Phosphorus tri-iodide has been prepared, using red phosphorus and a solution of iodine in carbon disulfide from which the free sulfur has been removed.

2. Phosphorus tri-iodide has been prepared by the interaction of solutions of iodine and white phosphorus in purified carbon disulfide.

3. The crystals of phosphorus tri-iodide obtained by both methods of preparation were found to melt at 61.0° .

4. Phosphorus di-iodide was prepared by the interaction of solutions of iodine and white phosphorus in purified carbon disulfide.

5. The crystals of phosphorus di-iodide obtained gave a melting point of 124.5° . This value is 14.5° higher than the commonly accepted value for the melting point of this compound, and should be adopted to replace the old value of 110° .

6. The presence of a compound of phosphorus, sulfur and iodine, probably $P_4S_3I_2$, is suggested as the cause of the low melting point of the two iodides of phosphorus. This will be true whenever they are prepared or recrystallized from carbon disulfide which has not had the free sulfur, which is always present, removed immediately before using.

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THE DIFFUSION OF OXYGEN THROUGH SILVER

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In a previous paper¹ an apparatus was described for the investigation of the rate of diffusion of oxygen through silver. The rates of diffusion under various conditions of temperature and pressure as well as for different thicknesses of silver were given. The results were summed up in the equation $X = (1.71 \sqrt{p}/10^{43}h)T^{14.62}$, where X is the rate of diffusion in cc. per sq. meter per hour, p the pressure of oxygen in mm. of mercury, h the thickness of silver in mm. and T the absolute temperature. (NOTE.— \sqrt{p}/h was printed by error instead of \sqrt{p}/h on pp. 1387 and 1389 of Ref. 1).

This paper is a continuation of the previous one.

Further investigation has shown that a general equation can represent the results of the diffusion of gases through metals for all cases so far investigated.

Apparatus

The apparatus used to obtain the results of Table I was that described in the first paper. That for the other results was practically the same, ex-

¹ Johnson and Larose, *THIS JOURNAL*, **46**, 1377 (1924).

cept that the silver plate was welded to the silver block (Ref. 1, Fig. 1), instead of being fixed to it with silver sulfate. This was done with a view to reaching higher temperatures, but the method proved unsatisfactory for, after a few experiments, the plate always developed a leak. This explains why so many trials were made with the silver of 0.205 mm. thickness. The results given are those obtained before a leak occurred.

An apparatus was devised and used for obtaining gas mixtures of definite proportions. This apparatus is shown in Fig. 1. It consisted essentially of two small vacuum pumps *K* and *L*, a Maass control *A-V* enabling intermittent suction to be applied to the pumps, and a gas reservoir *R* to maintain a steady flow of gas to the diffusion apparatus.

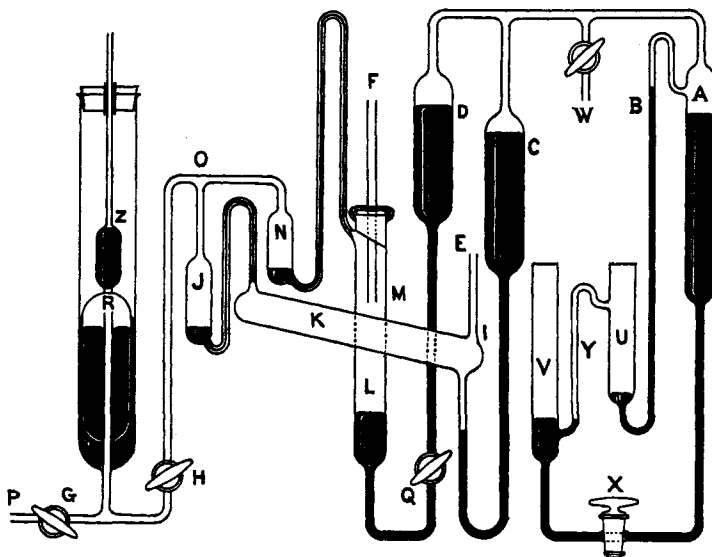


Fig. 1.

The apparatus functioned as follows. Suction produced by a water pump was applied at *W*, causing the mercury to rise in *A*, *B*, *C* and *D*. Tube *E* was connected to a cylinder of oxygen through a T-tube (not shown) the vertical branch of which dipped just below the surface of oil in a small dish, so that the pressure of the oxygen entering the pump at *E* could never greatly exceed atmospheric pressure. Similar connection was made between a cylinder of nitrogen and *F*. As the mercury rose in *C* and *D*, nitrogen entered at *F* and oxygen at *E*. After a short time, the mercury in *B* siphoning into *A* allowed air to enter the apparatus through *B*. The mercury consequently went down in *A*, *C* and *D*, and when it reached *I*, it cut off a definite volume of oxygen which it forced through the mercury seal *J* and similarly when it reached the bottom of the tube *F*, it forced a definite volume of nitrogen through the seal *N*. Oxygen and

nitrogen, therefore, came together in definite proportions at *O* and passed through the tube *P* which was connected to the diffusion apparatus through the sulfuric acid wash bottle. The volume of the reservoir was sufficiently large to take care of the total volume of gas coming over in one stroke of the pumps. The tap *G* was so adjusted that the flow into the diffusion apparatus was not too rapid and only a small part of the gas sent over by the pumps passed through it during the upstroke, the surplus being taken up by the reservoir. By means of *Q*, the rate of fall of the mercury in *D* could be so adjusted that the mercury reached the level *M* at the same time as that in *K* reached *I*. In this way oxygen and nitrogen passed into *O* at the same time. The rate of fall of the mercury in *A* could be regulated by *X*. The mercury was thus made to siphon from *Y* just as it reached the tops of Pumps *K* and *L*. In so doing it prevented access of air through *B* and the suction again acted, causing the cycle to repeat. During this suction stroke no gas was being forced into *J* and *N* and the flow through the diffusion apparatus was maintained from the supply in the reservoir *R* due to the cylinder weighted by the mercury in *Z* descending slowly. The weight of the mercury was such as to cause the cylinder almost to reach the bottom when gas was again forced through *J* and *N*. The stream of gas through *P* was greater in the upstroke

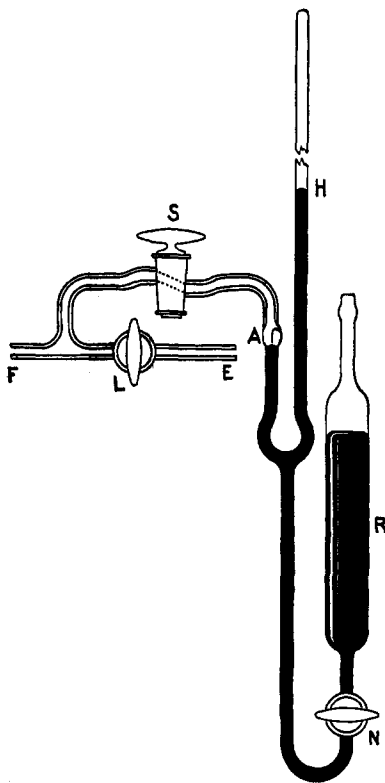


Fig. 2.

of the pumps than at the end of the cycle but by a proper adjustment of the taps *G* and *H* the difference in the rate of flow could be made very small.

The volume of gas sent over by the pump *L* was adjusted by lowering or raising the tube *F* and the composition of the mixture thus was varied. By this alteration of the height of Tube *F*, or by interchanging the oxygen and nitrogen at *E* and *F*, or by using air instead of one of the gases, almost any desired mixture was obtained.

The volume of the pumps was obtained by collecting the gas sent over in a known number of strokes and measuring its volume in a calibrated flask, necessary corrections being made for temperature and water vapor.

The apparatus shown in Fig. 2 was used by one of us in previous experi-

ments in which the pressure on the low-pressure side of the plate increased gradually as the oxygen diffused through.

The apparatus was interposed between the silver capillary tube and the pump. The capillary tube was connected at *F* and the end *E* to the pump. The volume from *F* to *L* and to *A* was made small by using capillary tubing. *H* is a manometer which indicated the pressure existing in the apparatus. Before starting an experiment, the apparatus was evacuated and then *L* was closed. Owing to the small volume into which the gas diffused, the pressure increased rapidly in the apparatus. The volume was kept constant by maintaining the level of the mercury to the pointer *A* by means of Tap *N*. The pressures as shown by the mercury in *H* were read at known times. The rate of increase of pressure gives the rate of diffusion, since the volume from the silver plate to *L* and *A* is known.

Experimental Results

TABLE I

RATE OF DIFFUSION OF OXYGEN THROUGH A SILVER PLATE					
Silver plate thickness, 0.0787 mm.			Pressure of oxygen, 760 mm.		
Temp., °C.	Rate ^a	Temp., °C.	Rate	Temp., °C.	Rate
352	5.1	463	56	528	179
375	7.8	472	62	539	219
404	17.8	481	85	550	264
416	22.5	484	84	550	271
428	28.6	495	99	561	311
439	35.7	503	142	584	442
448	43	515	150	607	617
460	58				

^a Rate of diffusion is always expressed as cc. per sq. meter per hour.

These results are plotted on Curve 1, Fig. 3. Other results were also obtained with plates of 0.135 mm. and 0.205 mm. thickness, and are represented by Curves 1, 2 and 3, Fig. 3. The following results were obtained with a plate of 0.30 mm., but are not plotted. The pressure of oxygen is 760 mm.

Temp., °C.	520	608	754	580
Rate	46.3	189	1330	124

It was mentioned in the first paper² that the variation in some of the results may have been due to the fact that the silver sulfate did not quite limit the area of the plate to that of the depression in the block. The following observation makes this explanation a most probable one. It was noticed that with one of the plates which was at first simply fixed with silver sulfate, the rate of diffusion was higher than expected. The plate was then welded to the block. The new rate of diffusion was lower and agreed with that expected. The results obtained with the welded

² Ref. 1, p. 1381.

plates (those of Curves 1, 2 and 3 and those obtained with the 0.30mm. plate) should then be given more weight than is given to the others (those of Table I plotted as Curve 4 and the results of the first paper).

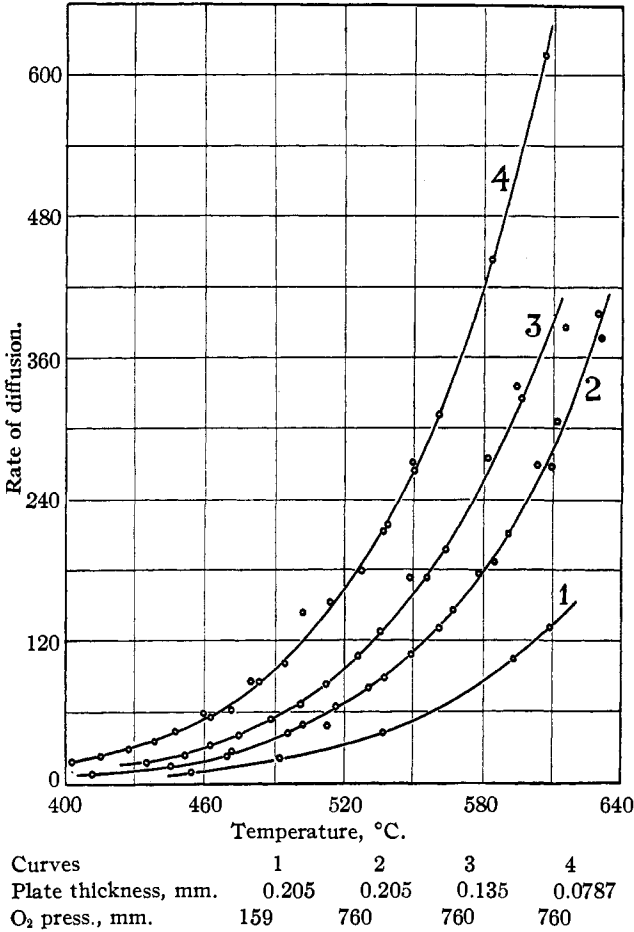


Fig. 3.

Fig. 4 shows the relation between the rate of diffusion and the square root of the pressure as was done in Fig. 6 of the first paper but the curves have been extended to show that they pass through the zero point as they should.

Fig. 5 shows the results obtained when the pressure was allowed to increase on the low-pressure side of the plate, the apparatus being that of Fig. 2. A plate of 0.0889 mm. thickness was used, the area of which was 2.5 sq. cm. The volume of the apparatus into which the gas diffused was 0.985 cc.

Comparison of Results with Those of Other Investigators

In our first paper, reference was made to the work of Troost and others in which it was found that the rate of diffusion was proportional to the square root of the pressure.

This relation has been found to hold for the diffusion of hydrogen through nickel by Lombard.³ The results of Spencer⁴ on the diffusion of oxygen through silver, though only approximate, show this relation to a certain extent.

The preliminary experiments carried out with the apparatus of Fig. 2 gave results which are also in accord with the same relation as can be shown by the following reasoning.

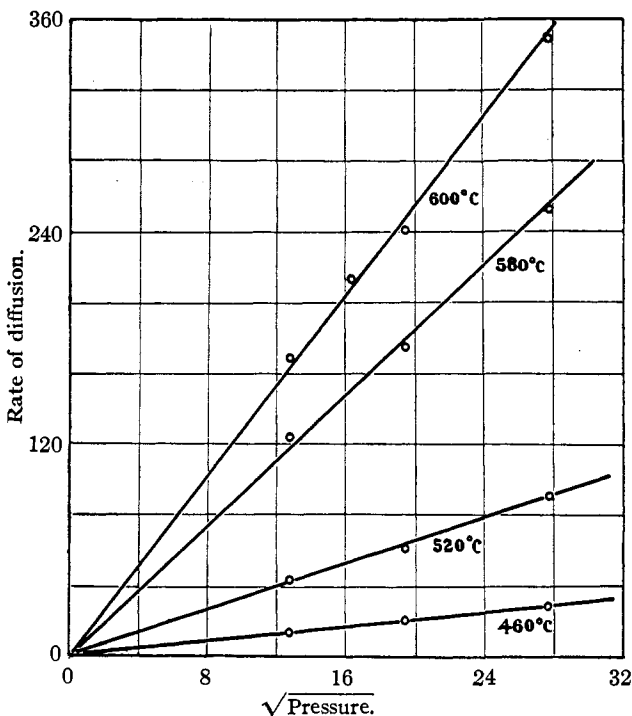


Fig. 4.

Let P_o be the pressure of oxygen on the outside of the apparatus and P_i be that inside, at any time. In the experiments, P_o was maintained constant while P_i increased from zero to a maximum as diffusion proceeded at constant temperature. The pressure outside causes diffusion to proceed inward with a velocity $V_1 = K\sqrt{P_o}$, while the pressure inside produces

³ Lombard, *Compt. rend.*, **177**, 116 (1923).

⁴ Spencer, *Trans. Chem. Soc.*, **123**, 2124 (1923).

a counter-diffusion opposing that inward with a velocity $V_2 = K\sqrt{P_i}$, K being a constant. The net rate of diffusion is the difference between the two, that is, $K(\sqrt{P_o} - \sqrt{P_i})$ or, substituting for K , $dA/dt = V_1(\sqrt{P_o} - \sqrt{P_i})/\sqrt{P_o}$, where A represents the amount of gas in the apparatus at any time t .

Now $A = kP_i$, k being a constant defined by the equation $dA/dt = kdP_i/dt$, so $dP_i/dt = (V_1/k)(1 - \sqrt{P_i}/\sqrt{P_o})$. Integrating,

$$t = -\frac{2k^2P_o}{V_1} \left\{ \frac{\ln \left(\frac{V_1}{K} - \frac{V_1}{K} \sqrt{\frac{P_i}{P_o}} \right)}{\frac{1}{K} - \frac{1}{K} \sqrt{\frac{P_i}{P_o}}} + I \right\}$$

When $t = 0$, $P_i = 0$, $I = 1/k(1 - \ln(V_1/k))$ and

$$t = -\frac{2kP_o}{V_1} \left\{ \ln \left(1 - \sqrt{\frac{P_i}{P_o}} \right) + \sqrt{\frac{P_i}{P_o}} \right\}$$

By plotting t against $\log \left(1 - \sqrt{\frac{P_i}{P_o}} \right) + \sqrt{\frac{P_i}{P_o}}$, a straight line should be obtained. Fig. 6 shows the results of Curve 2, Fig. 5, plotted in this way.

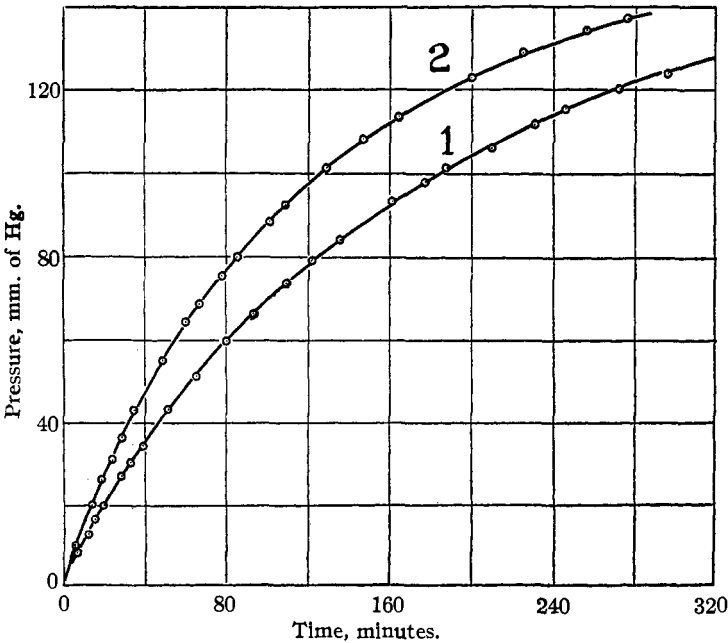


Fig. 5.—(1) For 572°; (2) for 600°.

The equation also enables one to calculate V . The following show the agreement between the values so obtained and those obtained in the later experiments, correction being made for the difference in thickness of silver plate.

Silver plate thickness, 0.0889 mm.		
Temp., °C.	Rate of diffusion calcd. from above eq. Cc./sq. meter/hr.	Later results
600	545	535
572	394	351
532	205	180

It was shown previously that the results could be expressed fairly well by the equation $X = (1.71\sqrt{p}/10^{43}h)T^{14.62}$, which is of the form $D = aT^b$, where a and b are constants for any one thickness and pressure, a form given by Ryder.⁵

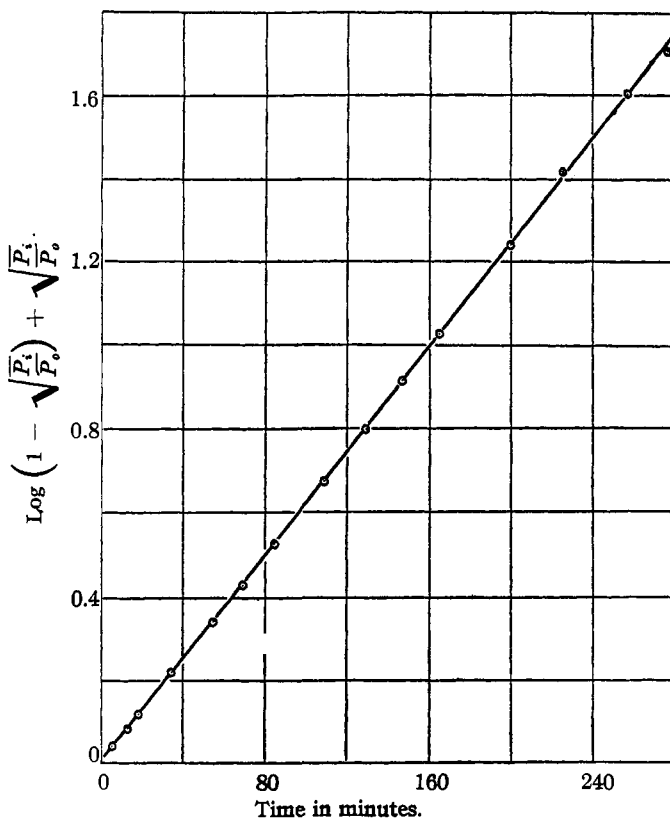


Fig. 6.

Lombard³ noted that his results were in agreement with an equation of the form $D = ab^T$.

Richardson⁶ has deduced on certain assumptions an equation different from these but in agreement with his results obtained with Nicol and Parnell⁷ on the diffusion of hydrogen through platinum.

⁵ Ryder, *Elect. J.*, 17, 161 (1920).

⁶ Richardson, *Phil. Mag.*, [6] 7, 266 (1904).

⁷ Richardson, Nicol and Parnell, *ibid.*, 8, 1 (1904).

Other investigators have not given any relation between temperature and rate of diffusion for their results but we have applied the equation of Richardson as well as the other two suggested above to all the results available on diffusion of gases through metals and it has been found that Richardson's equation represents the results of the experiments more accurately than do the others.

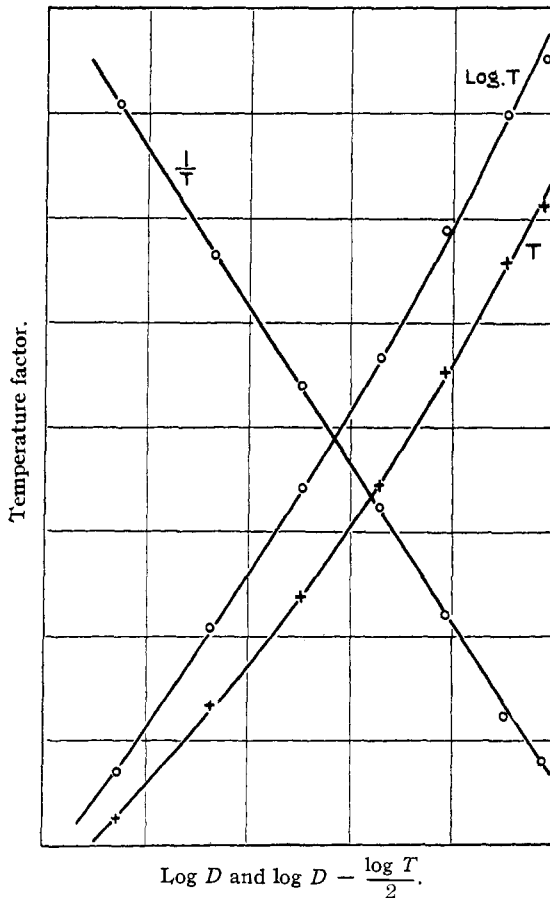


Fig. 7.—Lombard. Hydrogen through nickel.

The equation given by Richardson is of the form $Q = (C/d) \left(P^{1/2} T^{1/2} e^{-\frac{q}{4T}} \right)$, where Q is the rate of diffusion, d the thickness of metal, P the pressure of gas outside the metal, T the absolute temperature, q the heat of dissociation of the gas *inside* the metal and C is a constant.

The results of two investigations have been plotted in three different ways in Figs. 7 and 8.

The logarithm of the rate of diffusion has been plotted as abscissas

while the ordinates represent the temperature factor, in one case T (according to Lombard's equation), in another $\log T$ (according to Ryder's equation and that given in our first paper). From Richardson's equation $\log D - \log T/2$ should vary linearly with $1/T$ so that in the third case $\log D - \log T/2$ was chosen as abscissas while the ordinates represent $1/T$. It is seen that the $\log D - \log T/2$ relation gives straight lines

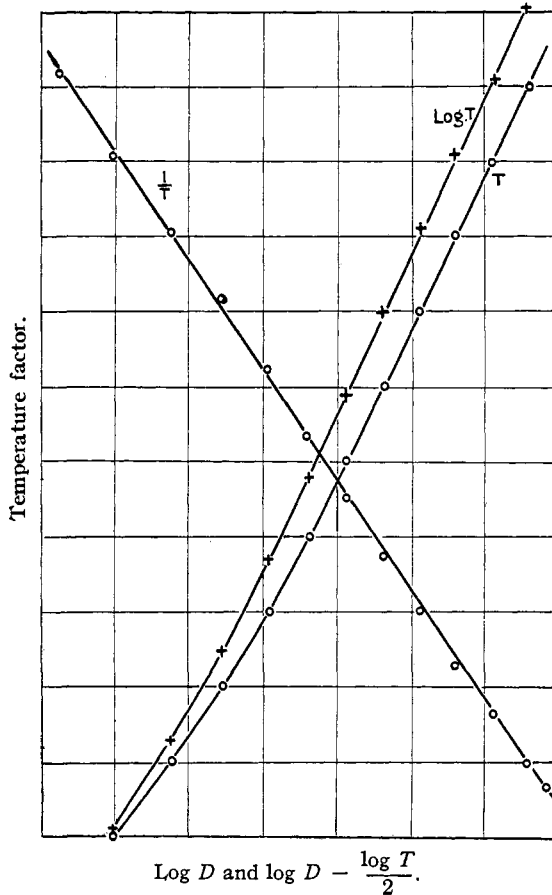


Fig. 8.—Results of Fig. 5 for 760 mm. pressure.

while the others do not. Similar curves are obtained by plotting the results of other investigations; for example, those of Deming and Hendricks⁸ on the diffusion of hydrogen through nickel, Spencer⁴ on the diffusion of oxygen through silver, Richardson, Nicol and Parnell⁷ on the diffusion of hydrogen through platinum.

In Fig. 9, our results have been plotted (according to Richardson's

⁸ Deming and Hendricks, *THIS JOURNAL*, 45, 2857 (1923).

equation) for various conditions of pressure and thickness. The lines have very nearly the same slope which shows that q in the equation is independent of the pressure of gas outside or of the thickness of silver.

The following figures show that the rates of diffusion calculated by means of Richardson's equation agree much better with the observed values than do those obtained by making use of our former equation where $\log D$ was supposed proportional to $\log T$. That this last relation was only approximate was foreseen as shown by our remarks on p. 1385 of the first paper,¹ where it was pointed out that the points seemed to lie on slightly curved lines instead of on straight lines as drawn.

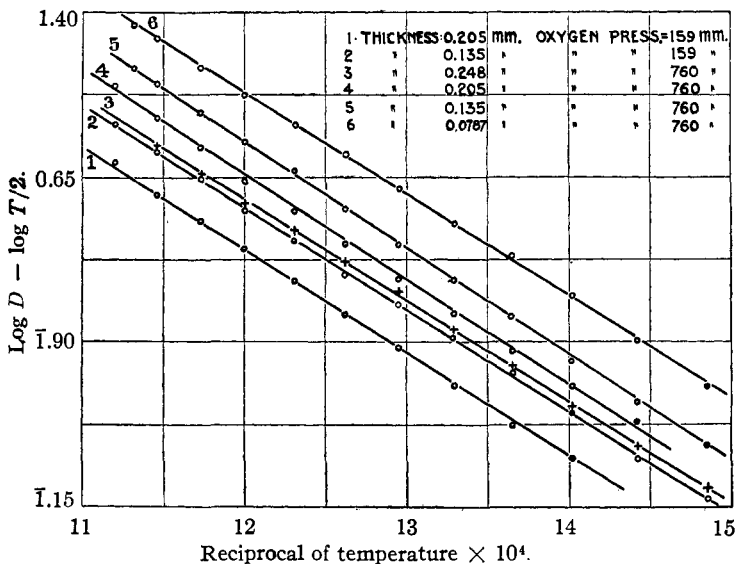


Fig. 9.

The average difference between observed and calculated values, taking a large number of observations, was found to be 7.7% for the first relation, but only 4.4% with Richardson's equation.

General Discussion

No satisfactory explanation of the mechanism of diffusion has yet been put forward.

There is no doubt that diffusion is preceded by absorption and that the properties go hand in hand. This is well illustrated by the following. Argon and helium are not absorbed by, nor do they diffuse through, iron. Hydrogen and nitrogen are absorbed by iron and diffuse through it. Oxygen is not absorbed by platinum and does not diffuse through it, while hydrogen diffuses readily through this metal and is also absorbed by it. Palladium behaves similarly towards hydrogen but it does not absorb

TABLE II
COMPARISON OF CALCULATED RATES OF DIFFUSION WITH THOSE OBSERVED

Thickness of silver	Press. of O ₂ , mm.	Temp., °C.	Obs.	Diffusion	
				Calcd. from Richardson's eq. ^a	Calcd. from log T eq. ^b
0.0787 mm.	760	420	21	21.1	20.4
		480	77	76.4	68.6
		500	111	112	101
		610	651	686	704
.135	159	420	6	5.7	5.4
		500	32	30.0	26.8
		560	91	85.1	80.0
.135	372	420	9.0	8.6	8.3
		460	20	20.8	18.9
		600	239	242	243
.135	760	460	28	29.7	27
		540	134	134	123
		580	260	256	248
.205	159	460	9	8.9	8.1
		540	43	40.3	36.9
		580	83	77.0	74.6
.205	760	440	13	12.7	11.9
		500	43	43.1	38.6
		580	173	168.6	163
.248	760	460	16.5	16.2	14.7
		540	73	72.7	66.7
		620	252	251	263

$$^a Q = \frac{43,054}{d} P^{1/2} T^{1/2} e^{-\frac{43,232}{4T}}$$

$$^b Q = \frac{1.71}{10^{43}} \frac{P^{1/2}}{d} T^{14.62}$$

helium and does not let it diffuse through. Nickel and copper both allow the diffusion of hydrogen through them and also absorb the gas. Copper does not absorb nitrogen nor carbon monoxide and these gases have been found not to diffuse through this metal.

It has been found also that absorption is generally proportional to the square root of the pressure, which shows again the close connection between diffusion and absorption. The above was determined by studies on the absorption of oxygen by silver,⁹ of sulfur dioxide by alloys of copper,¹⁰ of hydrogen by platinum,^{7,11} of hydrogen by nickel,¹² of hydrogen by copper¹³ and of hydrogen by iron.¹⁴

It is noteworthy that the rare gases so far investigated show no absorption or diffusion in metals. Nitrogen, which is considered a very inert

⁹ Steacie and Johnson, *Proc. Roy. Soc.*, **112A**, 542 (1926).

¹⁰ Sieverts and Bergner, *Z. physik. Chem.*, **82**, 257 (1913).

¹¹ Sieverts and Jurisch, *Ber.*, **45**, 221 (1912).

¹² Sieverts and Hagenacker, *Ber.*, **42**, 338 (1909).

¹³ Sieverts and Krumbhaar, *Z. physik. Chem.*, **74**, 277 (1910).

¹⁴ Neumann, *Stahl Eisen*, **34**, 252 (1914).

gas, shows little tendency for absorption or diffusion. Chemical affinity must play a large part in deciding whether a gas is absorbed or not.

The equation deduced by Richardson assumes a solubility of the gas in the metal and the agreement with the experimental results obtained brings out the fact that it is mainly the atoms of the dissociated gas molecules which diffuse and suggests that if the gas were not dissociated no appreciable diffusion would take place. It is very probable that absorption also depends to a large extent on this dissociation.

Heald¹⁵ was of the opinion that when a metal absorbs hydrogen, the gas is first dissociated by the metal. Ramsay¹⁶ and Winkelmann¹⁷ both suggested that hydrogen undergoes dissociation in passing through hot metals.

Berthelot¹⁸ and Sieverts¹⁹ have referred to the possible formation of hydrides when hydrogen is absorbed by a metal.

Newbery²⁰ assumes that hydrogen is in the atomic form in the metal, at least in the case of palladium. X-ray spectra taken by Yamada are in agreement with this view.

Many other facts point to a dissociation of gases in the presence of a metal or even to a formation of gaseous ions, for example, the charge that gold takes in hydrogen or oxygen,²¹ the dissociation of hydrogen in the presence of a heated filament,²² the activity of hydrogen in the diffusion experiments of Ramsay,¹⁶ work on electromotive force of gas cells,²³ the fact that hydrogen evolved by a fresh cathode in a vacuum tube obeys Faraday's laws,²⁴ the transparency of thin sheets of silver when heated in an atmosphere of oxygen,²⁵ the latter fact being easily explained if one assumes that the oxygen takes up electrons on absorption.

The fact that the equation expressing the electron emission or evaporation with temperature is of the same form as that given by Richardson for diffusion is perhaps not without significance. Wilson has shown that negative ionization produced by hot platinum is increased tremendously by the presence of hydrogen and he assumed that this effect was produced by collisions of hydrogen atoms inside the metal. It appears then that the more readily the metal gives off electrons, the more readily diffusion takes place, since both vary in the same way with temperature.

¹⁵ Heald, *Phys. Rev.*, **24**, 269 (1907).

¹⁶ Ramsay, *Phil. Mag.*, [5] **38**, 206 (1894).

¹⁷ Winkelmann, *Ann. Physik*, [4] **6**, 104 (1901).

¹⁸ Berthelot, *Ann. chim. phys.*, [5] **30**, 519 (1883).

¹⁹ Sieverts, *Z. physik. Chem.*, **60**, 129 (1907).

²⁰ Newbery, *THIS JOURNAL*, **41**, 1887 (1919).

²¹ Hartley, *Proc. Roy. Soc.*, **90A**, 61 (1914).

²² Langmuir, *THIS JOURNAL*, **37**, 417 (1915).

²³ Bose, *Z. Physik*, **34**, 201 (1900).

²⁴ Skinner, *Phil. Mag.*, [6] **12**, 481 (1906).

²⁵ Turner, *Phys. Rev.*, [2] **12**, 251 (1918).

One may look upon the process of diffusion as taking place in the following way. When the gas molecules strike the metal, the affinity or attraction of the metal for the gas is such as to produce a dissociation of the latter. The necessary electrons which the atoms of the gas are always striving for in order to assume a more stable configuration of electrons are furnished by the metal but the compound so formed is not stable since it is above the dissociation pressure; it therefore breaks up. The atoms of the gas take up anew electrons from the metal farther in and subsequently give them up again, thus making their way through the metal.

In the case of oxygen and silver, for example, the unstable compound formed would be of the formula Ag_2O , each oxygen atom taking up two electrons and giving them up again, not necessarily to the same silver atoms, but to those farther in the metal. The silver can be disregarded since it is stationary and the process simply looked upon as a taking up and giving up of electrons by the oxygen atoms alternately taking place. There is a sort of dynamic equilibrium between oxygen atoms and ions.

In this connection it may be mentioned that Schenck²⁶ pointed out that when oxygen dissolves in silver the system may be considered as a solution of silver oxide in silver.

The explanation just given is in accord with the way in which diffusion varies with external pressure of the gas. The process in the case of silver and oxygen can be represented by the equations $\text{O}_2 + 4\text{Ag} \longrightarrow 2\text{Ag}_2\text{O}$; $2\text{Ag}_2\text{O} \longrightarrow 4\text{Ag} + 2\text{O}$, the net effect being equivalent to $\text{O}_2 \longrightarrow 2\text{O}$.

The dissociation pressure due to the second reaction is about 1 atmosphere at 185° and increases with temperature, so that during the experiments carried out it was always larger than the pressure of oxygen used.

If such an equilibrium exists it follows that the concentration of atoms inside the metal is proportional to the square root of the concentration outside, that is, $\text{concn. O} = k \sqrt{\text{concn. O}_2}$, and if we assume, as in Fick's law for diffusion through liquids, that the diffusion velocity is proportional to the concentration gradient, then the rate of diffusion must be proportional to the square root of the pressure and it follows also that it must vary inversely as the thickness of metal.

The relation between rate of diffusion and temperature is probably complicated by several factors: motion of electrons in the metal, affinity of the gas for the metal, tendency for the gas molecules to dissociate, diffusion of the dissociated gas molecules inside the metal. All these factors would vary with the temperature.

It seems very probable then that the process of diffusion consists in a breaking-up of the gaseous molecules into atoms, that these atoms become ions in the presence of the metal and go through it in this form, and are given up on the low-pressure side of the metal plate where they

²⁶ Schenck, "Physikalische Chemie der Metalle," 1909.

combine to form molecules, though they may also then react with other substances if these are present.

It may be pointed out that, since from Richardson's equation the heat of dissociation of the gas inside the metal can be calculated, one could obtain the heat of dissociation of the gas outside the metal if the heat of absorption of the gas were known.

Grateful acknowledgment is hereby made of a studentship granted to one of the authors by the Research Council of Canada, during the tenure of which part of this work was carried out.

Summary

1. An apparatus has been devised in which two or more gases can be continuously and automatically mixed in any proportions and a constant flow of any definite mixture can be maintained.
2. The rate of diffusion of oxygen through silver has been measured with welded plates of 0.135, 0.205 and 0.30 mm. thickness, and the results have been shown to agree with those previously obtained.
3. Results obtained previously by one of the authors under experimental conditions different from those described in the first paper showed an agreement with the square-root relation between rate of diffusion and pressure.
4. It has been shown (as far as the data allowed) that Richardson's equation represents, within the experimental error, the diffusion of gases through metals.
5. A possible explanation of how diffusion takes place has been given and it has been shown to be compatible with the results obtained.

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[CONTRIBUTION FROM THE CHEMICAL LABORATORY OF HARVARD UNIVERSITY]

THE PHOTOLYSIS OF POTASSIUM NITRATE

BY DONALD STATLER VILLARS¹

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Introduction

When potassium nitrate in aqueous solution is exposed to ultraviolet light it decomposes into potassium nitrite and oxygen. Many investigators have studied this reaction.² Warburg³ has found that the photolysis in polychromatic light proceeds much more rapidly in alkaline than in neutral or acid solutions. He has also determined in alkaline solutions the moles transformed per calorie absorbed for the wave lengths 207,

¹ National Research Fellow.

² Thiele, *Ber.*, **40**, 4914 (1907). Lombard, *Compt. rend.*, **150**, 228 (1910). Berthelot and Gaudechon, *ibid.*, **152**, 522 (1911). Baudisch, *Ber.*, **44**, 1009 (1911).

³ Warburg, *Sitzb. kgl. preuss. Akad. Wiss.*, **1918**, 1228; *Z. Elektrochem.*, **25**, 334 (1919).