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THE OCCLUDED GASES IN COAL

BY

S. W. PARR

AND

PERRY BARKER



UNIVERSITY OF ILLINOIS
ENGINEERING EXPERIMENT STATION

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UNIVERSITY OF ILLINOIS
ENGINEERING EXPERIMENT STATION

BULLETIN No. 32

MARCH 1909

THE OCCLUDED GASES IN COAL

BY S. W. PARR, PROFESSOR OF APPLIED CHEMISTRY

AND

PERRY BARKER, RESEARCH ASSISTANT IN THE DIVISION OF APPLIED
CHEMISTRY

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I. INTRODUCTION

Experimental studies on coal have been carried on for some time in the Laboratory of Industrial Chemistry at the University of Illinois. A number of topics have been taken up, such as Weathering,¹ Distillation at Low Temperature,² Pure Coal,³ Classification,⁴ The Composition and Analysis of Illinois Coal,⁵ etc., etc.

These studies, while conducted as independent lines of work, are so interrelated that not infrequently the facts brought out under one division have aided very materially in the interpretation of results obtained under another heading. Especially, also, has it been found that each one of the original topics has subdivided so that the main subjects have grown in number. This is well illustrated in our studies on the weathering of coal. As the facts accumulated in that work, it very soon became evident that an initial loss of heat values occurred in the first few days after release of the coal from the seam, which could not be credited to oxidizing conditions. This naturally led to the study of the topic which we have for convenience designated the Deterioration of Coal, because it is not strictly a weathering process. A preliminary report on this phase was made in Bulletin No. 17 of the Engineering Experiment Station. Additional information on this topic was also published in an article contributed to the Journal of the American Chemical Society,⁶ showing conclusively the fact that a positive exudation of combustible gases occurs in freshly mined coal. From the development of this fact there naturally arose the necessity of inaugurating an independent series of experiments which has resulted in this special study of the occluded gases in coal.

Another subdivision of the work on weathering naturally arose in connection with the storage of coal. For a complete understanding of all phases of this topic it seemed essential that we add to our information concerning the causes underlying spontaneous combustion. As a means for promoting this latter end it was thought that a study of the occluded gases, their composition,

¹ Bulletin No. 17, Univ. of Ill. Eng. Exp. Sta.

² Bulletin No. 24, Univ. of Ill. Eng. Exp. Sta.

³ Trans. Am. Inst. Min. Engrs., July, 1907.

⁴ Jour. Am. Chem. Soc. Vol. 28, p. 1425.

⁵ University Studies, Vol. 1, No. 7.

⁶ Jour. Am. Chem. Soc., Vol. 30, p. 1027.

behavior, interreactions, etc., would contribute much needed information upon this very important point. Some of the results presented herewith bear upon this topic in quite an unexpected manner, and particular attention is called in the accompanying tables to the marked avidity of coal for oxygen.

Again, it is an interesting fact to note that in the studies as outlined in Bulletin No. 24 on the low temperature distillation of coal, a subdivision of that topic came about as a natural result of certain experiments which indicated an unexpectedly low temperature at which active oxidation was effected. A detailed study was therefore made of oxidation conditions, the temperatures at which chemical action was set up, and the relation between the external temperature and the speed of such reactions.¹ The fact was soon developed that oxidation took place at much lower temperatures than had been supposed. It is very evident, therefore, from a simple statement of these facts that we have developed still another independent topic as a result of the study of oxidation temperatures on one hand and the experiments connected with the occluded gases on the other. This work has, therefore, been taken up under the heading of Spontaneous Combustion. Here, it will be noted, the behavior of the occluded gases plays an important part, and it was believed that the facts which might develop from such a study would contribute materially to our understanding of oxidation in this more active form. Already the results of this line are beginning to give promise of information which will be of special value, and will at least contribute to a better understanding of this difficulty, if indeed there will not develop practical suggestions as to methods for avoiding the danger and loss attending the firing of coal in storage. It is hoped that this work will be sufficiently advanced to warrant at least a preliminary report during the current year.

Concerning the work here outlined on the occluded gases, it should be said that the study is not to be considered by any means complete. The facts obtained make a well rounded unit, as a preliminary study, and since a considerable time must elapse before the final round of work can be completed, it has seemed advisable to publish the results that have been thus far worked out with much skill and patience by Mr. Barker.

¹ Bulletin No. 24, Univ. of Ill. Eng. Exp. Sta.

Provision has been made by the Engineering Experiment Station for again taking up this work.

Thanks are due also to Dr. H. Foster Bain, Director of the Illinois State Geological Survey, for cooperation in this and the other investigations on Illinois coal. It is directly due to assistance from these two sources that the coal studies carried on in this laboratory have been made possible.

II. BIBLIOGRAPHY

All varieties of coal, as well as peat, are known to contain occluded or mechanically enclosed gases. Their nature and amount are dependent upon the structure and age of the coal and the conditions of weathering to which it has been exposed. A large part of these gases escapes when the coal is first exposed in the seam. At times the immense volume and explosive or poisonous nature of the escaping nitrogen, carbon dioxide, and methane derivatives, make it either dangerous or impossible to mine the coal. However, a portion is retained within the coal only to be given up by continued exposure and at increased temperatures.

Within the past thirty years numerous investigations of the character of the gases occluded in European coals have been made. The first investigation of this sort was conducted by Websky¹, who made an examination of the gases evolved by peat.

He found this gas to consist of nitrogen, 53.67; carbon dioxide, 2.97; and methane, 43.36. Later, Von Meyer² made quite an extensive examination of various German coals. The method of extraction in these experiments was as follows. The coal in nut-sized pieces was placed in a flask and covered with freshly boiled water. The flask was securely stoppered with a rubber cork, having an outlet to a bell for collecting the gas. On heating, the gas enclosed in the coal was given off and replaced a corresponding volume of water in the collector. The first series of experiments was upon Zwickan coal, both in the fresh state and after exposure, as shown in Table I.

¹ Jour. prakt. Chemie., Vol. 92, p. 76, (1864).

² Jour. prakt. Chemie., Vol. 5, p. 144, (1872).

Abs. Jour. Chem. Soc. Vol. 10, p. 798, (1872)

TABLE I
GASES IN ZWICKAN COAL

I, III, and V, coal fresh from seam.
 II same as I, weathered for 5 years.
 IV " " III, " " 1½ years.
 VI " " V, " " 5 years.

	I	II	III	IV	V	VI
	Fresh	Weathered	Fresh	Weathered	Fresh	Weathered
Vol. per 100 g.	38.00	18.20	25.50	18.60	52.80	13.60
CO ₂	2.42%	16.70%	4.02%	2.25%	.60%	7.62%
O	2.51	4.90	.62	.70	.0	2.44
C ₂ H ₄		1.47				.96
CH ₄	71.90	3.17	45.00	73.16	51.40	15.88
C ₂ H ₆		18.61				22.35
N	23.17	55.15	50.36	23.89	48.00	50.75

A series of coal from Bochum, both fresh and weathered, was also examined, with results as shown in Table II.

TABLE II
GASES IN BOCHUM COAL

	I	II	III	IV	V	VI
	Fresh	Weathered	Fresh	Weathered	Fresh	Weathered
Vol. per 100 g.	50.6	43.2	43.3	41.2	59.2	43.6
CO ₂	4.87%	11.12%	2.18%	15.84	4.82%	7.68%
O	2.66	2.88	2.12	3.06	1.99	2.24
CH ₄	16.65	7.40	25.19	6.57	31.57	3.31
N	75.82	78.60	70.51	74.53	60.62	86.77

	VII	VIII	IX	X	XI	XII
	Fresh	Weathered	Fresh	Weathered	Fresh	Weathered
Vol. per 100 g.	54.4	39.2	54.5	39.6	42.0	36.4
CO ₂	1.30%	4.35%	2.02%	2.15%	3.72%	8.49%
O	1.60	3.35	.90	3.14	.39	3.57
CH ₄	30.25	11.12	10.65	3.43	5.70	.0
N	66.85	81.18	86.43	91.28	90.19	87.94

The author notes the increase in the ratio of nitrogen content over that of ordinary air. He maintains that the nitrogen has been enclosed in the coal during its formation, that a part results

from the reactions in the formation, and that another part is due to the air taken into the material of the coal, this last portion being the residue from the air, the oxygen having been absorbed by the coal with the formation of water and CO_2 . He refers to the recent study upon the oxidation of coal by Richter,¹ who maintains that the resulting compound is principally water, while only comparatively small amounts of CO_2 were formed. This theory seems to be borne out by the above analyses in which the CO_2 has never completely replaced the oxygen of the air. It is also noted that one of these coals, very high in sulphur, gave high CO_2 , which is in accordance with Richter's² statement, that increase in the content of pyrite, facilitates the absorption of oxygen. The presence of higher hydrocarbons of the methane series, also ethylene, indicates a possible rise in temperature within the coal during exposure, due to the absorption of oxygen with a resulting destructive distillation.

Later, this same author³ has investigated a number of English coals. He found in none of these any gases absorbable by fuming sulphuric acid, and methane was the predominating combustible gas. O. Kolbe,⁴ in the same year, reported several analyses of lignites. Von Meyer has also made a second series of investigations on German coals.⁵

In 1875 Thomas⁶ began extensive tests upon the gases occluded in English coals. He devised an entirely different method for removing the gases. The coal was placed, in one large piece, in a glass tube which was sealed to a Sprengel mercury pump. When a vacuum had been established, the tube containing the coals was immersed in water at 100°C ., and the gases removed by the pump. The rapidity of evolution of the gas depends upon the hardness of the coal and the quantity contained. The following table gives the results of Thomas's first series of experiments.

¹ Chem. Centralblatt, 1870, p. 245, 543.

² Chem. Centralblatt, 1870, p. 543.

³ Jour. prakt. Chemie., Vol. 6, p. 407, (1872).

Abs. Jour. Chem. Soc., Vol. 10, p. 801.

⁴ Jour. prakt. Chemie., Vol. 6, p. 79, (1873).

⁵ Jour. prakt. Chemie., Vol. 6, p. 389, (1873).

⁶ Jour. Chem. Soc., Vol. 13, p. 793, (1875).

TABLE III
GASES IN ENGLISH COALS

		Cc. per 100 grams	CO ₂	O	CH ₄	N
1	Bituminous	55.9	36.42%	.80%	.00%	62.78%
2	“	61.2	16.77	2.72	.40	80.11
3	“	55.1	5.44	1.05	63.76	29.75
4	Semi-bituminous	73.6	12.34	.64	72.51	14.51
5	Steam coal	194.8	5.04	.33	87.30	7.33
6	“	250.1	13.21	.49	81.64	4.66
7	“	218.4	5.46	.44	84.22	9.88
8	“	147.4	18.90	1.02	67.47	12.61
9	“	375.4	9.25	.34	86.92	3.49
10	“	149.3	11.35	.56	73.47	14.62
11	“	215.4	5.64	.54	82.70	11.12
12	“	24.0	22.16	6.09	2.68	69.07
13	“	39.7	9.43	2.25	31.98	56.34
14	“	555.5	2.62		93.13	4.25
15	“	600.6	14.72		84.18	1.10

The second series of analyses of Thomas was on the gases evolved from fans or “blower gas”. Some previous work¹ has been done upon this class of gases and Thomas’s work confirmed these results. Most of these samples contained between 94 and 97 per cent of methane.

In another publication Thomas² gives the results of the examination of samples of lignites. He concludes, as in the case of a previous author,³ that lignites oxidize readily and always contain a small percentage of CO.

¹ Bischof, Edinburgh New Phil. Jour, Vol. 29, p. 39. Vol. 30. p. 127, (1840).

Bunsen, Petersburg Akad. Bull, Vol. 14, p. 59.

Playfair, Memoirs of the Chemical Soc., Vol. 2, p. 7.

Graham, Memoirs Geol. Sur. Gt. Britain, Vol. 1, p. 460.

² Jour. Chem. Soc., Vol. 17, p. 146, (1877).

³ Varretropp, Chem. Centralblatt, 1865, p. 953.

TABLE IV
GASES PRESENT IN LIGNITES

I Bohemian lignite III Bovey Heathfield lignite (at 100° C.)
II Earthly lignite IV Mineral resin

	I	II	III	IV
Vol. per 100 g.			59.9	21.4
CO ₂	96.41%	83.99%	89.53%	88.24%
O	.32	.65	-----	.23
CO	1.20	1.04	5.11	7.90
CH ₄	trace	-----	.33	.47
N	2.17 ₁	14.91	5.03	3.16

¹ Adds to 100.10 per cent in the original manuscript.

A series of experiments by Thomas on cannel coals and jets was reported² in the same year.

TABLE V
ANALYSES OF CONTAINED GASES IN CANNEL COALS AND JET

I Wigan cannel coal IV Scotch cannel coal
II " " " V Whitehill cannel shale
III Scotch " " VI Whitby jet

	I	II	III	IV	V	VI
Vol. per 100 g.	42.13	35.06	16.80	55.70	55.70	30.20
CO ₂	6.44%	9.05%	53.94%	84.55%	68.75%	10.93%
CH ₄	80.69	77.19				
C ₂ H ₆	4.75	7.80			2.67	
C ₄ H ₁₀				.91*		86.90
N	8.12	5.96	46.06	14.54	28.58	2.17

*C₃H₈

The noticeable feature in the gases extracted from these cannel coals is the high percentages of C₂H₆ and the absence of oxygen.

Bedson³ has reported the first analyses upon the gases extracted from coal dusts and found that they showed a general resemblance to those removed from coal and that all the combus-

² Jour. Chem. Soc., Vol. 15, p. 144, (1876).

³ Trans. North of Eng. Inst. Min. & Mech. Eng., Vol. 37, p. 245. (1888).

Abs. Jour. Soc. Chem. Ind., Vol. 7, p. 729, (1888).

tible constituents consisted of olefiant gases and methane derivatives.

Several years later Bedson¹ published results of amounts of gas given off when coals were heated to 100°C. for 100 hours. Fine coal gave about 300 cu. ft. of gas per ton and of this gas one-sixth was CH₄. Upon reducing these samples further to dust, more gas (20 cu. ft. per ton) was given off, of which 18 per cent was CH₄.

McConnell² investigated several seams of English coals as to their content of enclosed gases. Particular attention was paid to the nature of the paraffin hydrocarbons extracted from these coals. One of these analyses from a mine where an explosion had recently taken place showed CO₂, 1.7, 0, 1.0; methane, 88.7; nitrogen, 8.7. The author concludes that the deeper seams yield coals containing combustible gas and CO₂ while the younger ones contain more CO₂ and less combustible gases.

Broockmann³ criticizes the results of Bedson and McConnell, saying that explosions of dust are not dependent upon the gaseous content of the same. He also states that rubber stoppers will not stand a vacuum, thereby diluting the gas with the products of reactions at high temperatures between the air that has leaked in and the coal.

Bedson⁴ has replied to Broockmann's criticism and maintains that his apparatus was air-tight, as all rubber joints were coated with a vacuum-proof cement. He also states that the Austrian Fire-damp Commission has corroborated his statements in regard to the relation of enclosed gases to the inflammable nature of dusts. The author notes the absorption of oxygen by samples exposed in the laboratory, and in this way accounts for the high oxygen-nitrogen ratio in some of his samples.

The latest experimental work upon gases occluded in coal has been conducted by Trowbridge,⁵ who compared the gases in fresh coal, "mother of coal", surface dust, and dust from timbers. He concludes that his data show the absorption of gases from the

Jour. Soc. Chem. Ind. Vol. 11, p. 882, (1892).

² Jour. Soc. Chem. Ind. Vol. 13, p. 25, (1894).

³ Trans. North of Eng. Inst. Min. & Mech. Eng. Vol. 52, p. 16, (1902).

Abs. Jour. Soc. Chem. Ind., Vol. 22, p. 86, (1903).

⁴ Trans. North of Eng. Inst. Min. & Mech. Eng., Vol. 52, p. 25, (1902).

Abs. Jour. Soc. Chem. Ind., Vol. 22, p. 86, (1903).

⁵ Jour. Soc. Chem. Ind., Vol. 25, p. 1129, (1906).

air, preferably oxygen. Two analyses of fresh and exposed coal are given.

	Fresh	Exposed
CO ₂	1.65%	1.18%
O	8.79	23.80
CH ₄	44.60	3.58
N	44.76	71.44

Clark¹ upon summing up most of the data upon this subject comments upon the inability to make any comparisons of the results at hand, as all samples represented some loss in transit from mine to laboratory. Of course this loss is dependent upon the structure of the coal and upon the time occupied in transit.

In view of the fundamental relation it bears to a fuller understanding of the subject of alterations in coal, whether in transit or storage, this study of the composition, behavior, and reactions of the occluded gases has been undertaken.

III. SCOPE OF EXPERIMENTAL WORK

The experimental work may be conveniently described under the following headings:

1. The construction of apparatus for evacuating the samples.
2. The construction of apparatus for exact gas analysis.
3. The construction of apparatus for collecting samples and devising methods for making collections of coal and the occluded gases.
4. The analysis of the last portion of air and enclosed gases in several samples of coal that had been exposed outside for periods varying from two to fifteen months.
5. The analysis of the last portion of air and the enclosed gases in four sections of a vertical core cut from a pile of screenings that had been exposed outside for fifteen months.
6. The analysis of the gas that had surrounded samples kept in jars for a year; these samples had been previously kept sealed tight for one year, then were transferred to separate jars.
7. The analysis of gas that surrounded samples that had been sealed as soon as they came from the mine.

¹Bull. U. S. Geol. Surv., No. 330, p. 660.

8. The analysis of the last portion of air and the enclosed gases from a set of samples of drillings sealed at the mines.

9. The analysis of the last portion of air and the enclosed gases from a set of face mine samples that had been stored in the laboratory for two years. This set of samples corresponded as to mine location with the set described in (8).

10. Miscellaneous analyses of gases from samples used for experiments upon the absorption of oxygen by coal.

IV. DESCRIPTION AND MANIPULATION OF APPARATUS

The apparatus used for the removal of the occluded gases from the samples of coal consisted of a modified form of the Boltwood mercury air pump, as used by Cady and McFarland¹ in the determination of helium in natural gas, and is shown in Fig. 1. *A* is a side neck funnel for holding mercury. *B* is a stopcock to regulate the feed of the mercury flowing from *A* through *C* and producing a vacuum through *G*, *H*, etc. *D* is a 300 cc. bell jar supplied with a stopcock and is used to collect the gases brought down through *C* by the mercury column. *E* is the outlet of the tube for the return lift for the mercury. *L* is the overflow hole in the mercury tank *M* and keeps *E* from becoming sealed during the operation of the pump. *F* is the return lift to the reservoir *A*, which is connected to an ordinary filter pump through *K*. *G* is a drying U-tube of alternate lengths of phosphorus pentoxide (P_2O_5) and glass wool having ground glass joints to *C* and *H*, the latter being a drying tower of calcium chloride. At *I*, the outlet tube to *H*, a wide end is made in the tubing to admit the constricted tips of the coal sample containers of which *J* is a representation. This joint at *I* is made with rubber resin (Khotinsky) cement.² In operating the pump the stopcock *N* is closed and *O* is opened so that the train *G* and *H* can be completely evacuated before any gas from the coal container is admitted. Mercury is allowed to flow from *A* through *C* and out into *M*, the amount being regulated at the cock *B*. While the train is being evacuated, *D* is left empty with the stopcock open or is removed altogether. As *M* fills, the suction at *K* draws mercury from the surface at *E* and returns it to the reservoir *A*. Should the mercury come through *C* too fast,

¹ Jour. Amer. Chem. Soc., Vol. 29, p. 1523, (1907).

² Jour. Amer. Chem. Soc., Vol. 30, p. 20, (1908).

a seal will be formed at *E*. To prevent this an overflow is provided at *L*, and *E* is so regulated that it is on a level with the lowest point of *L*. In order to further prevent a stoppage at *E*, the mouth of this tube was filled with fine iron wire at first, but later a constriction was made at the bend *U* and this kept too much

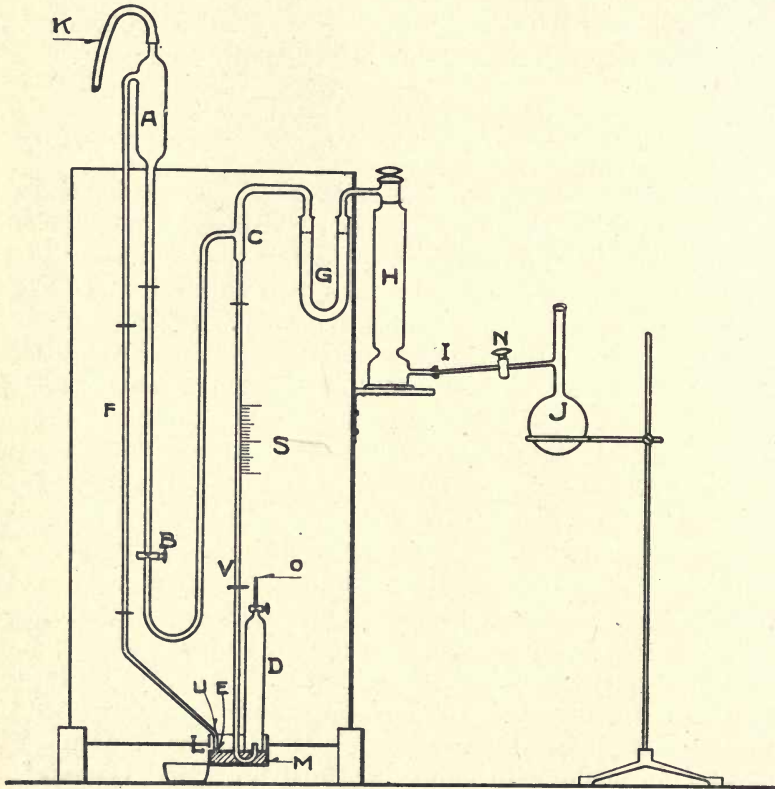


FIG. 1

mercury from passing up at a time. When a vacuum had been produced in the train, a solid column remained in *V* and after shutting off the pump connected to *K*, the column *V* should read within 1 or 2 millimeters of barometric height. The scale *S* facilitated the taking of barometer readings, as it was graduated in millimeters above the outlet into the tank *M*. The bell jar *D* is now filled with mercury by pouring into *M* and applying the filter pump to the outlet of *D* at *O*. After completely filling the

holder *D* the mercury pump is again started and *N* is opened to allow the gas from *J* to be removed. As this gas comes down *V*, it collects and displaces mercury in *D*. When the column shows approximately barometric height on *S*, the pump is stopped and the gases are transferred from *D* and analyzed.

The apparatus used for gas analysis consisted of a regular set of Hempel's pipettes using an explosion pipette for hydrocarbons. All volumes of gas were measured by means of the Hempel¹ compensating burette shown in Fig. 2. The arrangement and operation of this burette are described by Hempel as follows:

"The instrument consists of the graduated measuring tube *A*, the correction tube *B*, the manometer tube *F*, and the level bulb *G*. The measuring tube and leveling bulb are mounted in suitable iron feet. The measuring tube and the correction tube stand in the wide glass cylinders *C*, which are filled with water to insure that these two tubes are at all times at the same temperature.....

"The correction tube *B* and the manometer tube *F* are made from simple glass tubes fused together in the form shown in the cut; *g* is a small capillary tube. The manometer tube is U-shaped, and is somewhat widened at *k* and *i*, these two widened portions having marks scratched on the glass at exactly the same height. The manometer tube is joined to the measuring tube by means of a piece of rubber tubing connecting the end of the capillary *l* with the tube *a* of the stopcock. The reason for making the manometer tube so long lies in the fact that otherwise, if the apparatus is carelessly handled, the mercury might easily be driven from the manometer tube into the burette or the correction tube. With the arrangement shown in the figure this is almost impossible, since the difference in pressure must be more than half an atmosphere before the mercury can pass over into either tube.....

"Before proceeding with the analysis, the volume of the manometer tube from the mark *k* to the point *a* must be ascertained. To do this, draw over the mercury in the manometer until it reaches *a*, then turn the stopcock *D* to connect with the capillary *b*, and now draw any desired volume of air into the burette. Leaving the stopcock open, read off this volume of air on the scale of the burette, the air here being, of course, under the pre-

¹ Hempel, —Dennis, Gas Analysis, 3rd Edition, p. 60.

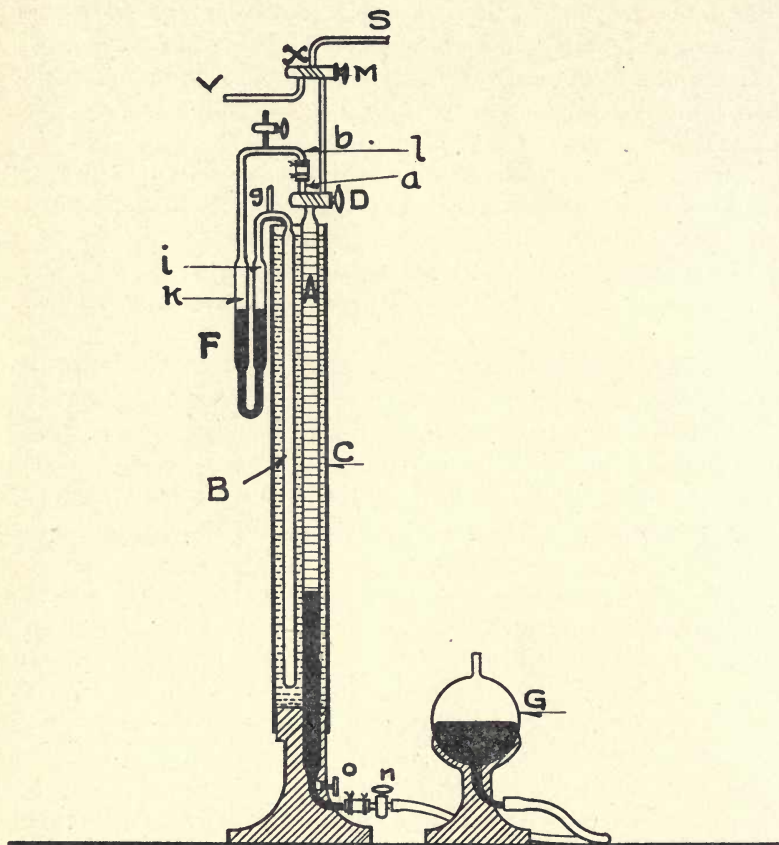


FIG. 2

vailing pressure of the atmosphere. Turn stopcock *D* so that the burette communicates with the manometer tube, and drive the air over into this latter tube until the mercury in it stands at equal height in its two branches; that is, at the marks on *k* and *i*. The difference between the two readings on the measuring tube, provided the tube *g* remains open, gives the volume of the manometer from the mark on *k* up to *a*.....

"In many cases it is highly desirable to arrange the apparatus so that the reading on the measuring tube *A* corresponds directly to volumes at 0° C. and 760 mm. pressure. To accomplish this, a piece of rubber tubing is slipped over the end of the capillary tube *g* and fastened firmly in place by a wire ligature. By

lowering the level-bulb, mercury is drawn over into the manometer tube until it reaches the capillary *l*, and the burette is then allowed to stand for two hours in a room of fairly constant temperature. The stopcock *D* is then opened so that the contents of the burette are in free communication with the atmospheric air. As soon as one is convinced that all parts of the apparatus are at the same temperature, the gas volume in the burette is read exactly, and the temperature and barometric pressure are noted. The thermometer and barometer should stand in the same room with the apparatus. The stopcock *D* is closed and the volume which the gas would occupy at 0° C. and 760 mm. barometric pressure is now calculated.

Example: The gas volume is 97 ccm., the barometric pressure 753.3 mm. and the temperature 8.75° C. The space from *k* to *a* in the manometer has previously been determined and found to be 1.8 ccm. The tension of the water vapor at 8.75° C. is 8.4 mm.

“If *b* represents the observed barometric pressure, *t* the temperature, *e* the tension of water vapor at that temperature, and *V* the observed volume, the volume *V*₀ which the gas would occupy under standard conditions may be calculated from the following formula:—

$$V_0 = V \frac{b - e}{760 (1 + 0.00367 t)}$$

In the above example this volume is 92.1 ccm.

Since, however, in making measurements with the correction tube, the gas fills the space from *k* to *a*, this volume must be subtracted from the above results:—

$$92.1 - 1.8 = 90.3 \text{ ccm.}$$

“In order now to adjust the gas volume in the correction tube so that readings of volumes in the burette will be reduced at once to standard conditions, the stopcock *D* is turned so that the burette communicates with the manometer tube, and the gas volume in the burette is compressed by raising the level-bulb *G* to the volume which it has been calculated that it would occupy at 0° C. and 760 mm. pressure. The mercury in the manometer tube is, of course, forced out of equilibrium by this operation. Air is now blown into the correction tube through the rubber tube at *g* until the mercury stands at the same height in the two branches

of the manometer tube, and the rubber tube is then closed by means of a strong pinchcock placed directly above the end of *g*."

In reading the volumes of gas in the burette, the mercury was always drawn over to *a* and left there while an absorption was being made. The extra stopcock *M* does away with any correction for the connection capillary tube *S*. Whatever absorbent is to be used is driven up from the pipette through *S* to *x*. *M* is then turned so that it connects with *b*, the gas being driven through *S* into the pipette. On returning, the absorbent is brought up to *x* and *M* and *D* are closed. The level at *k* and *i* is established and the volume read. To all readings the volume from *k* to *a* must be added to get the total gas in the burette. *M* is now turned so that the solution in *S* can be washed out through *Y* and the next absorbing pipette is attached.

The containers for samples consisted of the flasks such as *J*, Fig. 1, Putman fruit jars and Mason fruit jars. All the samples for removal of enclosed gases were placed in flasks (*J*). These were 300 cc. fractionating flasks with a stopcock at the outlet. As soon as the sample was placed in this flask, the stopcock was closed and a rubber stopper coated with the Khotinsky cement was sealed in at the mouth. The stopcocks on all the apparatus used in this were lubricated with a special mixture of 16 parts of vaseline, 1 part of paraffin, and 8 parts of pure rubber, maintained at a temperature of 350 to 400° for several hours.¹ The Putnam jars² were used for all samples from which the surrounding gas was to be removed by displacement. These were inverted and opened in a tank of water, saturated with CO₂ and illuminating gas, and all gas contained therein was removed by inserting a tube at the mouth and pulling the gas into a holder over mercury. The Mason jars were used for submerged samples. To remove the gas that adhered to the coal under water, the tops were removed and, as the gas was dislodged upon agitating the jars, it was collected in a bell jar, inverted over the top of the container.

V. EXPERIMENTAL DATA

The first set of experiments in connection with the study of the occluded gases in Illinois coal consisted of the examination of

¹ Jour. Amer. Chem. Soc. Vol. 29, p. 1725.

² Univ. of Ill. Eng. Exp. Sta. Bull. 17, p. 34.

a vertical section from a pile of 1½ inch screenings that had been exposed outside for 5 months. At the time of collection, this pile had been smoldering for two months. Five days before taking the samples it had been wetted down. A core 7 in. in diameter and about 85 in. long was cut vertically from the pile by means of the cylindrical sampler. As the sampler went through the pile it was removed after going down 20 in., and the contents were, in each case, sampled separately. The samples were transferred at once to flasks, as shown by *J*, Fig. 1, and left in contact with the enclosed air for varying lengths of time as shown in the tables. The analyses of the different sections are given in Table VI.

TABLE VI

GASES IN COAL FROM SMOLDERING SCREENINGS

I Christian County screenings First 20 inches of pile	III Christian County screenings Third 20 inches of pile
II Christian County screenings Second 20 inches of pile	IV Christian County screenings Fourth 20 inches of pile

Part 1 Last Portion of Air

	I	II	III	IV
Time of Standing, days	2	1	1	1
Weight of Coal, grams	200	200	200	200
Vol. of Gas at 0° C. 760 mm.	56.9	59.2	65.2	79.9
Cc. of Gas per 100 grams	28.45	29.6	32.6	38.95
CO ₂	.7	.7	3.2	3.15
O	5.6	6.1	4.0	7.60
CH ₄	.25	.05	0.	0.
N	21.90	22.75	25.4	28.20
Per cent by Volume				
CO ₂	2.46	2.37	9.32	7.90
O	19.69	20.61	12.27	19.02
CH ₄	.90	.17	0.	0.
N	76.95	76.85	78.41	73.08

Part 2 Gas Removed by Vacuum

Time of Standing, days	7	8	9	8
Weight of Coal, grams	200	200	200	200
Vol. of Gas at 0° C. 760 mm.	3.2	17.6	36.6	8.0
Cc. of Gas per 100 grams	1.6	8.8	18.3	4.0
CO ₂	.5	2.2	4.05	1.85
O	0.	1.05	1.45	.45
CH ₄	.1	.15	.75	.2
N	1.0	5.40	12.05	1.50
Per cent by Volume				
CO ₂	31.25	25.0	22.13	46.25
O	0.	11.93	7.93	11.25
CH ₄	6.25	1.70	4.10	5.00
N	62.50	61.37	65.84	37.50

TABLE VII

COMPOSITION OF GAS EXUDED FROM FRESH COAL

- I Lebanon, Lebanon City Coal Co., sealed, dry.
 II Lebanon, Lebanon City Coal Co., sealed, submerged.
 III Bennett, Bennett Mine, International Coal Mining Co., sealed, dry.
 IV Bennett, Bennett Mine, International Coal Mining Co., sealed, submerged.
 V O'Fallon, Mine No. 2, St. Louis & O'Fallon Coal Co., sealed, dry.
 VI O'Fallon, Mine No. 2, St. Louis & O'Fallon Coal Co., sealed, submerged.

	I Dry	II Submerged	III Dry	IV Submerged	V Dry	VI Submerged
Weight of Coal, grams	642	800	648	800	787	800
Volume of Gas cc. (a)	446(b)	38.8	442(b)	29.3	331(b)	97.8
Per cent by Volume						
CO ₂	0.	0.	5.88	0.	.88	0.
O	18.50	1.87	7.64	1.03	0.	1.08
CH ₄	0.	55.97	0.	35.39	11.83	90.28
N	81.50	42.16	86.48	63.58	87.29	8.64

(a) At normal temperature and pressure.

(b) Total gas from containers.

In order to study the composition of the gas found in certain experiments¹ to be liberated from Illinois coal and shown also to be combustible, several samples were collected from fresh seam

¹Jour. Am. Chem. Soc., Vol. 30, p. 1027.

faces and allowed to stand for seven months. They were then opened under water and the gas surrounding the coal in the containers was collected by displacement. A similar set of samples was collected in jars which were filled with water and whatever gas had been given off at the end of seven months was collected. The results of these two sets of analyses are given in Table VII.

It is evident that aside from the addition of methane and carbon dioxide to the ordinary constituents of air, a decrease in the percentage of oxygen originally contained in the air of the jars has taken place. In order to test the extent of this absorption of oxygen, a number of samples of coal were placed in jars with large volumes of air equal to about six to ten times the several volumes of coal. After being in contact with the coal for a year the air was collected by displacement and analyzed. The following results show the general nature of the changes which had taken place, Table VIII.

TABLE VIII

THE ABSORPTION OF OXYGEN BY COAL

- I Springfield, Sangamon Mine, Sangamon Coal Co.
- II Springfield, Sangamon Mine, Sangamon Coal Co.
- III Eldorado, Mine No. 8, O'Gara Coal Co.
- IV Marion, Chicago & Big Muddy Coal Co.
- V Herrin, Squirrel Ridge Mine, Chicago & Carterville Coal Co.
- VI DuQuoin, Greenwood, Davis Coal Co.
- VII Belleville, Suburban Coal Mining Co.
- VIII O'Fallon, Mine No. 2, St. Louis & O'Fallon Coal Co.

	I	II	III	IV	V	VI	VII	VIII
Weight of Coal in grams	109	139	180	183	146	134	138	153
Total Volume of Gas Enclosed in cc. at Normal Temperature and Pressure	873	849	816	814	843	853	850	837
Per cent by Volume								
CO ₂	.48	.94	.68	1.87	.25	1.23	1.11	1.62
O	.16	.13	0.	0.	.25	0.	0.	1.45
CH ₄	0.	0.	6.28	0.	2.17	0.	0.	0.
N	99.36	98.93	93.04	98.13	97.33	98.77	98.99	96.93

These two sets of analyses give some indication of the nature of the alterations that are going on when coal is exposed, but give no information as to the composition of the gas remaining in the coal. In addition, the samples were of various sizes of coal that had been broken from the face of the seam and they had been subjected to more or less exposure, even though but for short periods. In order to get coal closely representative of the material as it occurs in the seams, a set of samples of drill dust was collected in the following manner. As the drillings fell from the hole, they were collected in an ordinary half-liter fractionating flask fitted with a stopcock at the side tube. When the flask was filled it was sealed by a rubber stopper which was coated with the rubber resin vacuum cement. These flasks were taken to the laboratory as soon as possible, all the gases contained therein were removed by means of a mercury air pump and were collected over mercury. This portion of the gas is designated in the tables as *last air*. The flasks were then allowed to stand for several days after which they were again connected with the air pump and any gas that had been evolved was removed.

In order to have some extreme types of laboratory weathered samples to compare with the fresh drillings, a set of coals that had been used for some previous tests was evacuated in the above manner. These were portions of mine samples about two years old and had been quartered, reduced to buckwheat size, air-dried and enclosed in air-tight containers. These samples were transferred to flasks, as shown by *J* in Fig. 1, and evacuated by the mercury pump. The results are given in pairs, the fresh drillings being placed by the side of the samples two years in storage. These two series correspond as to location of the mines, so comparison of the changes in the occluded gases can be made by inspection of Table IX.

The striking feature of the above table is the large amount of combustible gases liberated by the fresh drillings. While this amounts to as much as 30 cc. per 100 grams, in the fresh samples, no such gases were detected in the old lots. However, it must be understood that the relative amounts of gas in coal from these various mines can not be critically judged from these analyses, as some of the working faces have been within short distances of long standing exposures¹. Some idea of the rapidity of transpira-

¹A universal shutdown in the Illinois coal mines during April and a part of May, 1908, made it impossible to get samples representative of continuous workings.

tion of occluded gases from exposed faces can be gathered from the following data.

As a drill hole was driven, the dust from the first 2½ feet was collected in one flask, while that from the last 3 feet was sealed in a separate container. As can be seen in Table X, the sample further from the exposed face contained more occluded gas and had less changes produced in what did remain.

TABLE IX

COMPARISON OF GASES FROM COALS OF DIFFERENT EXPOSURE PERIODS

- I Springfield, Sangamon Mine, Sangamon Coal Co., fresh drillings.
 II Springfield, Sangamon Mine, Sangamon Coal Co., face sample, 2 years old.
 III Herrin, Squirrel Ridge Mine, Chicago & Carterville Coal Co., fresh drillings.
 IV Herrin, Squirrel Ridge Mine, Chicago & Carterville Coal Co., face sample 2 years old.
 V Clifford, Mine No. 8, Big Muddy Coal & Iron Co., fresh drillings.
 VI Clifford, Mine No. 8, Big Muddy Coal & Iron Co., face sample 2 years old.
 VII Marion, Mine No. 3, Peabody Coal Co., fresh drillings.
 VIII Marion, Mine No. 3, Peabody Coal Co., face sample 2 years old.
 IX Westville, Mine No. 44, Dering Coal Co., fresh drillings.
 X Westville, Mine No. 44, Dering Coal Co., face sample 2 years old.

Part 1 Last Portion of Air

	I Fresh	II Old	III Fresh	IV Old	V Fresh	VI Old	VII Fresh	VIII Old	IX Fresh	X Old
Time of Standing, days	7	9	14	2	13	4	13	6	7	1
Weight of Coal, grams	261	209	220	205	244	204	217	204	231	108
Volume of Gas cc. (a)	141.2	96.6	192.1	33.5	287.4	40.6	160.6	63.3	197.2	38.4
Cc. of Gas per 100 g. (b)	54.21	46.2	87.4	16.37	117.8	19.78	74.2	30.97	85.50	35.66
CO ₂	2.12	1.91	3.37	1.27	6.65	1.23	3.27	.54	10.34	0.
O	2.87	2.06	.94	2.54	.58	2.75	.95	5.04	.95	7.90
CH ₄	12.22	0.	18.70	0.	28.22	0.	1.57	0.	19.81	0.
N	37.00	42.23	64.05	12.56	72.40	15.81	68.20	25.39	54.40	27.76
Per cent by Volume										
CO ₂	3.92	4.15	3.86	7.80	5.56	6.80	4.43	1.79	0.	12.09
O	5.30	4.46	1.04	15.50	.49	13.80	1.28	16.25	22.20	1.11
CH ₄	22.53	0.	21.79	0.	32.44	0.	2.12	0.	0.	23.17
N	68.65	91.39	73.31	76.70	61.51	79.4	92.17	81.96	77.80	63.63

Part 2 Gas Removed by Vacuum

	I Fresh	II Old	III Fresh	IV Old	V Fresh	VI Old	VII Fresh	VIII Old	IX Fresh	X Old
Time of Standing, days	13	8	12	10	13	12	13	11	13	10
Weight of Coal, grams	261	209	220	205	244	204	217	204	231	108
Volume of Gas cc. (a)	26.9	14.9	48.8	1.9	76.4	5.8	20.4	20.5	26.0	1.1
Cc. of Gas per 100 g. (b)	10.31	7.12	22.18	1.08	31.30	2.84	9.4	10.04	11.26	1.02
CO ₂	1.84	2.74	1.68	4.63	.69	3.18	2.01	3.51	.56
O	.50	.10	.1429	.15	0.	0.	.82	.10
CH ₄	6.14	0.	19.21	22.20	0.	.09	0.	2.17	0.
N	1.83	1.27	1.21	4.18	1.88	6.13	8.03	4.76	.36
Per cent by Volume										
CO ₂	17.85	80.50	7.58	14.79	24.20	33.84	20.0	32.09	54.60
O	4.83	1.30	.6192	5.90	0.	0.	7.32	9.90
CH ₄	59.59	0.	86.37	70.93	0.	1.00	0.	19.26	0.
N	17.73	18.20	5.44	13.36	69.90	65.16	80.0	41.33	35.50

(a) At 0° C and 760 mm. pressure. (b) Figured to coal as sampled.

TABLE X

GASES FROM COAL DRILLINGS TAKEN AT VARYING DISTANCES FROM THE FACE OPENING

I	Westville, drillings from first 2½ feet of hole, last air.
II	“ “ “ “ last 3 “ “ “ “ “ “
III	“ “ “ “ first 2½ “ “ “ gas by vacuum.
IV	“ “ “ “ last 3 “ “ “ “ “ “

	I	II	III	IV
Time of Standing, days	7	7	13	13
Weight of Coal	182	231	182	231
Volume of Gas at 0° C. 760 mm.	174	197.2	9.6	26.0
Cc. of Gas per 100 g. of coal	95.60	85.50	5.27	11.26
CO ₂	7.23	10.34	1.76	3.51
O	.59	.95	0.	.82
CH ₄	11.29	19.81	2.30	2.17
N	76.40	54.40	1.21	4.76
Per cent by Volume				
CO ₂	7.57	12.09	33.33	32.09
O	.62	1.11	0.	7.32
CH ₄	11.73	23.17	43.74	19.26
N	80.08	63.63	22.93	41.33

In addition to the loss of combustible gases, the fresh samples of drillings showed more extensive absorption than did the laboratory weathered ones. From this it may be concluded either that the oxygen has entered into some combination with the coal itself, or that a reaction has taken place, resulting in the formation of carbon dioxide. The presence of considerable amounts of carbon dioxide in the gases from the fresh samples seems to bear out the latter conclusion, although it does not completely replace the oxygen of the air. It may also be possible that the carbon dioxide formed and taking the place of the occluded gases is only given off at higher temperatures. That this is true to some extent is shown by the fact that 69% of the gases removed from one of these fresh samples at 100°C. consisted of carbon dioxide. This phase of the matter is receiving further study.

It is certainly true that this absorption of oxygen takes place either contemporaneously or as soon as the gases escape from the fresh coal. A study of some of the stages of this absorption or oxidation can be made from Table XI.

TABLE XI

AVIDITY OF OLD AND FRESH COAL FOR OXYGEN

- I Atmosphere surrounding old face sample in contact with large volume of air for 2 years.
 II Old face sample sealed in fresh air 2 days, then evacuated.
 III Drillings, sealed 14 days.
 IV " in vacuum 12 days.
 V " second air in contact with coal 7 days.

	I	II	III	IV	V
Weight of Coal, grams	146	205.5	220	220	220
Volume of Gas, cc.	843	33.5	192.1	48.8	130.4
Per cent by Volume					
CO ₂	.25	7.80	3.86	7.58	1.63
O	.25	15.50	1.04	.61	.37
CH ₄	2.17	0.	21.79	86.37	14.14
N	97.33	76.70	73.31	5.44	83.86

In this table all samples were from the same mine. No. I and II were partially air-dried face samples, which had been sealed in Putnam jars for some two years. From No. I the surrounding air in the container was collected by displacement and analyzed.

No. II was left in one of the sealed fractionating flasks for two days. At the end of that time both the surrounding air and some of the enclosed gases were removed by means of the air pump. No. III is a flask of fresh drillings from which the surrounding air and occluded gases were removed by vacuum, as above. No. IV is the analysis of the further gas given off after the surrounding air had been removed and the flask had stood in a vacuum for twelve days. No. V is the analysis of the air that had been readmitted to the evacuated flask and left in contact with the coal for seven days.

Further comparison of the rapidity of oxidation can be made from Table XII. All samples were from the same mine. No. I and II had been in laboratory containers for over two years. Two portions were transferred to the flasks and left in contact with normal air for two and six days respectively.

TABLE XII

RAPIDITY OF OXIDATION IN FRESH AND OLD SAMPLES OF COAL

- I Old face sample sealed 2 days, last air.
 II " " " second air in contact 6 days.
 III Fresh drillings, sealed 14 days, last air.
 IV Drillings, second air in contact 7 days.
 V Fresh drillings, sealed 13 days, last air.
 VI Drillings, second air in contact 8 days.

	I	II	III	IV	V	VI
Weight of Coal, grams	204	204	220	220	244	244
Volume of Gas at 0°C & 760 mm.	40.6	253.1	192.	130.4	287	157.
Cc. per 100 g. of Coal	19.78	124.2	87.40	59.28	117.7	64.4
CO ₂	1.23	1.95	3.37	.97	6.65	2.02
O	2.75	7.24	.94	.22	.58	.15
CH ₄	0.	0.	19.04	8.38	13.14	8.84
N	15.81	115.0	64.05	49.71	97.0	53.39
Per cent by Volume						
CO ₂	6.80	1.57	3.86	1.63	5.56	3.14
O	13.80	5.83	1.04	.37	.49	.23
CH ₄	0.	0.	21.79	14.14	11.14	13.70
N	79.40	92.60	73.39	83.86	82.81	82.93

The preceding results have thrown some light upon the changes produced by the deterioration of sealed laboratory samples but contain no data on samples subjected to outside exposure. Table XIII gives a comparison between samples of fresh

TABLE XIII

COMPARISON OF ACTIVITY AS BETWEEN SEALED AND EXPOSED SAMPLES OF COAL

- I Westville, Mine No. 44, Dering Coal Co., fresh drillings, sealed.
 II " " " " " " " " exposed screenings, 15 mo. old.
 III " " " " " " " " 2 " "
 IV Marion, Mine No. 3, Peabody Coal Co., fresh drillings.
 V " Binkley, Miles Coal Co., outcrop coal.
 VI Springfield, Sangamon Mine, Sangamon Coal Co., fresh drillings.
 VII " " " " " " " " screenings, 2 mo. old.

Part 1 Last Portion of Air

	I Fresh	II Ex- posed	III Ex- posed	IV Fresh	V Ex- posed	VI Fresh	VII Ex- posed
Time of Standing, days	7	3	6	13	19	7	3
Weight of Coal, grams	231	200	200	217	224	261	200
Volume of Gas, 0°C. 760 mm.	197.2	44.2	55.8	160.6	134.3	141.2	240.5
Cc of Gas per 100 g.	85.50	22.10	27.9	74.2	60.0	54.21	120.3
CO ₂	10.34	1.40	.25	3.27	3.38	2.12	3.03
O	.95	3.25	5.75	.95	.38	2.87	22.09
CH ₄	19.81	0.	0.	1.57	.54	12.22	.66
N	54.40	17.45	21.90	68.20	55.70	37.00	94.47
Per cent by Volume							
CO ₂	12.09	6.34	.90	4.43	5.64	3.92	2.51
O	1.11	14.71	20.61	1.28	.64	5.30	18.36
CH ₄	23.17	0.	0.	2.12	.54	22.53	.55
N	63.63	78.95	78.49	92.17	93.18	68.65	78.58

Part 2 Gas Removed by Vacuum

	I Fresh	II Ex- posed	III Ex- posed	IV Fresh	V Ex- posed	VI Fresh	VII Ex- posed
Time of Standing, days	13	14	13	13	7	13	13
Weight of Coal, grams	231	200	200	217	217	261	200
Volume of Gas, 0°C. 760 mm.	26.0	26.9	16.8	20.4	19.9	26.9	23.9
Cc of Gas per 100 g.	11.26	13.45	8.4	9.40	8.88	10.31	11.95
CO ₂	3.51	1.4	2.85	3.18	2.99	1.84	2.39
O	.82	2.8	.7	0.	.36	.50	.81
CH ₄	2.17	.15	.6	.09	.10	6.14	2.39
N	4.76	9.10	4.25	6.13	5.43	1.83	6.36
Per cent by Volume							
CO ₂	32.09	10.41	33.93	33.84	33.67	17.85	20.00
O	7.32	20.82	8.33	0.	4.02	4.83	6.78
CH ₄	19.26	1.12	7.15	1.00	1.05	59.59	20.00
N	41.33	67.65	50.59	65.16	61.26	17.73	53.22

drillings and samples exposed to the weather. No. I is a sample of fresh drillings from Westville, while No. II was collected from the surface of a pile of $1\frac{1}{2}$ inch screenings from the same mine and had been stored outside for fifteen months. No. III is from the surface of a pile of the same screenings that had been stored outside for two months. No. IV is a sample of fresh drillings from Marion, while No. V is from an outcrop of the same seam one mile from the place where No. IV was taken. This outcrop had been exposed for one year. No. VI is a sample of drillings from Springfield, while No. VII was collected from the surface of a pile of $1\frac{1}{2}$ inch screenings from the same mine. These screenings had been stored outside for two months. Samples of each type were sealed in the evacuating flasks, *J*, of Fig. 1, together with normal air for periods of from three to nineteen days, indicated in the following table. Upon applying the vacuum, the first portion of air, to about 150 mm. pressure, was discarded. The last portion secured by carrying the vacuum to the limit gives essentially the composition of the gases contained in the coal proper, as shown in Table XIII, Part 1. Without the readmission of air the flasks were set aside for varying periods and a second application of the vacuum gave results as shown in Part 2.

VI SUMMARY

It seems evident from a study of the results presented in the foregoing pages, that two active processes are set up immediately upon the liberation of coal from the vein. The first is an exudation of hydrocarbons, mainly consisting of marsh gas (CH_4); the second is an absorption of oxygen. There can be little question, moreover, that these alterations proceed simultaneously. In Tables IX and X, for example, on pages 21 and 22, there are present in the gases from all the samples of fresh drillings, notable quantities of methane, ranging from 18% to 86% of the various gas volumes. At the same time the oxygen present has dropped down in a very positive manner, in some cases even reaching the vanishing point. That this transpiration of gases is interdependent and is of the nature of an osmotic exchange can hardly be affirmed as an explanation of the phenomenon. On the contrary, there seems to be evidence that the gases operate independently of each other. Take for example the figures

for marsh gas. In the case of the samples listed in Table IX, the exudation of CH_4 seems to have spent itself in those samples held in laboratory containers for two years. In no case is there evidence of further liberation of this gas, even with thorough application of the vacuum. Table XI shows a similar condition, in that an evacuation of the gases from the two year old sample, as in No. II, shows no marsh gas present. Similar results are seen in samples No. I and II in Table XII. In Table XIII the completion of this exudation would seem to be reached after two months, though it is well to note that by forcing, as with a vacuum in the second part of this table, the two months old samples may be made to yield more methane, though in relatively small quantities.

On the other hand, the avidity of the coal for oxygen seems to be pronounced at the very beginning of the exposure of the freshly mined material, and while there are a number of cases where a certain agreement seems to exist between the in-going and the out going marsh gas, still there are more cases where the absorption of oxygen is pronounced without any evidence of marsh gas being present. For example, attention may be called especially to samples No. II, IV, VI, and VIII, in Table IX. In all of these cases the oxygen-nitrogen ratio shows a positive diminution of the oxygen from the normal ratio of approximately 1:4 with practically no evidence of marsh gas being present. While the statement, therefore, may be modified by further study, it seems fair to conclude, for the present, that there is no necessary connection, at least of a strictly chemical nature, between the exudation of marsh gas and the absorption of oxygen.

Again, the liberation of CH_4 , while very active in the first few days after removal of the coal from the ground, diminishes in amount quite rapidly till, after the second month, there is very little of this gas in evidence. The activity of the coal for oxygen, on the contrary, seems to be of longer duration. In Table XII, samples No. I and II were collected June 1, 1906. The tests for the table were made in May and June, 1908. There is marked absorption of oxygen in sample No. I after two days' exposure in the flask to normal air, while No. II, with five days' exposure, shows still further reduction in the oxygen ratio without accompanying evidence, also, it should be noted, of marsh gas. Similarly, sam-

ples No. II, IV, VI, and VIII, of Table IX, show a marked avidity for oxygen after two years from time of collecting.

These facts have a direct bearing on the topic of deterioration, as substantially defining the limit as to time of that form of alteration. While varying somewhat in different coals, the loss of hydrocarbons for the most part is practically complete at the end of two months. These facts have a bearing also upon the matter of weathering, and indirectly upon the matter of spontaneous combustion. The absorption of oxygen is undoubtedly closely associated with both of these phenomena. Our studies upon the weathering processes coincide with these studies in gases, namely, that in all probability this low type of oxidation extends over an indefinite length of time. Moreover, while under normal conditions there is effected but a very slight oxidation and loss of fuel values, the conditions are favorable, as, for example, upon an increase of temperature, for bringing about a very rapid combination with oxygen even to the point of combustion.

How far this absorption of oxygen is a chemical reaction, or low combustion resulting in CO_2 and H_2O , and how far it is an absorption into the molecular structure and composition of the coal must be left for further study.



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