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REPORT OF INVESTIGATIONS—NO. 86

EFFECT OF OXIDATION ON REACTIVITY AND SWELLING OF ILLINOIS COALS

BY

O. W. REES AND W. F. WAGNER

REPRINTED FROM INDUSTRIAL AND ENGINEERING CHEMISTRY,
Vol. 35, No. 3, 1943

PRINTED BY AUTHORITY OF THE STATE OF ILLINOIS

URBANA, ILLINOIS
1943

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Topographic Mapping in Cooperation with the United States Geological Survey.
This Report is a Contribution of the Analytical Division.

February 22, 1943

EFFECT OF OXIDATION ON REACTIVITY AND SWELLING OF ILLINOIS COALS

BY

O. W. Rees and W. F. Wagner

ABSTRACT

The effect of oxidation of laboratory samples on reactivity and British swelling index numbers has been studied. Samples were exposed up to 8-9 months and tests were made at various intervals. Agglutinating values were determined occasionally to demonstrate that oxidation did take place. Three sets of whole coal and banded ingredients representing high-volatile bituminous ranks A and C were studied.

IN ADDITION to the usual proximate and ultimate analyses, several special laboratory tests have been developed for determining the charactersitics and behavior of coals under different conditions. In general, these special tests measure over-all characteristics under specified conditions rather than specific properties or constituents such as those measured in the proximate and ultimate analyses. Some of these special tests are the C. R. L. reactivity test (*6*, *7*), British Standards swelling test (*3*, *4*), agglutinating value test (*1*), and various means for determining plastic properties and swelling characteristics of coal when heated.

The fact that Illinois coals are distinctly banded makes it possible to study the characteristics not only of the whole coal but also of the constituent bands vitrain, clarain, durain, and fusain. This report deals with the effect of oxidation of whole coal and constituent bands on ignition temperatures as measured by the C. R. L. reactivity test and swelling as measured by the British swelling index test. The agglutinating value test was used to indicate oxidation of samples during exposure.

SAMPLES AND PROCEDURES

Three sets of samples, including whole coal and banded constituents, from Henry, Macoupin, and Gallatin Counties were used for this work. These samples represented coals with approximate moisture levels of 19, 13, and 5 per cent and high-volatile bituminous ranks C, C, and A, respectively. The whole coals were about 2-inch channel samples cut down, crushed to pass a 4 mesh sieve, and sealed in friction-top cans in the mines. The banded ingredient samples were hand-picked, crushed to −4 mesh size, and sealed in friction-top cans in the mines. All samples were further crushed to desired sizes in the laboratory.

The samples were air-dried and crushed in a Braun type 6 CP pulverizer to −20 mesh and to finer sizes in a ball mill. The −40 +60 mesh samples were used for reactivity determinations, −60 mesh samples for British swelling index (B. S. I.) tests and −200 mesh samples for the agglutinating value tests. Proximate constituents, total sulfur, and calorific values were determined on −60 mesh samples according to standard A. S. T. M. procedures (*2*). The various special

Table I. Analyses of Coal Samples

Lab. No.	Description	Basis[a]	Moisture, %	Ash, %	Volatile, %	Fixed C, %	Total S, %	B.T.U.
		Henry County						
C-2425	Whole coal	A. R.	19.3	8.9	31.6	40.2	3.85	10,332
		Dry	....	11.0	39.2	49.8	4.77	12,802
C-2427	Clarain	A. R.	18.2	9.0	32.7	40.1	3.53	10,453
		Dry	....	11.0	40.0	49.0	4.32	12,772
C-2428	Durain	A. R.	16.2	6.3	33.8	43.7	3.23	11,252
		Dry	....	7.5	40.3	52.2	3.85	13,426
C-2429	Fusain	A. R.	25.2	10.5	17.3	47.0	7.82	9,161
		Dry	....	14.1	23.1	62.8	10.46	12,255
		Macoupin County						
C-2405	Whole coal	A. R.	14.0	10.2	34.1	41.7	4.50	10,624
		Dry	....	11.9	39.7	48.4	5.23	12,354
C-2406	Vitrain	A. R.	15.3	2.9	35.4	46.4	2.81	11,503
		Dry	....	3.4	41.8	54.8	3.32	13,579
C-2407	Clarain	A. R.	13.4	3.7	39.0	43.9	3.60	11,696
		Dry	....	4.3	45.0	50.7	4.16	13,502
C-2408	Durain[b]	A. R.	13.3	11.9	34.5	40.3	2.88	10,514
		Dry	....	13.7	39.8	46.5	3.33	12,130
C-2409	Fusain	A. R.	20.3	8.0	14.0	57.7	4.95	10,379
		Dry	....	10.0	17.6	72.4	6.20	13,017
		Gallatin County						
C-2416	Whole coal	A. R.	5.1	9.4	36.8	48.7	3.03	12,584
		Dry	....	10.0	38.8	51.2	3.19	13,267
C-2417	Vitrain	A. R.	4.4	7.7	37.6	50.3	2.31	13,109
		Dry	....	8.1	39.3	52.6	2.42	13,706
C-2418	Clarain	A. R.	3.6	9.9	35.6	50.9	2.65	12,749
		Dry	....	10.3	36.9	52.8	2.74	13,221
C-2419	Fusain	A. R.	17.8	15.6	19.0	47.6	3.50	9,693
		Dry	....	18.9	23.2	57.9	4.26	11,789

[a]A. R. = as received. [b]Probably dull clarain.

tests were made on the samples soon after receipt from the mines and again after various intervals of exposure. All samples were kept in stoppered bottles for 2 to 4 weeks following the first determination, and were then exposed to the laboratory atmosphere in shallow dishes with frequent stirring for the duration of the studies. The mesh size of exposed samples was that used in each particular test—that is, −40+60 mesh for ignition temperature samples, −60 mesh for B. S. I. test samples, and −200 mesh for agglutinating test samples. U. S. Standard sieves were used for sizing all samples.

The C. R. L. reactivity test was that described by Sebastian and Mayers (*6*) and by Sherman, Pilcher, and Ostborg (*7*). The equipment used was almost a duplicate of that described by Sherman and co-authors with the exception that split, hinged type refractories were provided to permit more rapid cooling of the furnaces between runs. Determinations were made in both oxygen and air and these results are indicated as T_{15} and T_{75}, respectively.

The B. S. I. test was essentially that of the British Standards Institution (*3*), the principal modification being the substitution of a Fisher high-temperature burner for the Teclu burner specified in the British standard method.

Agglutinating value determinations were made according to the "Proposed Method of Test for Agglutinating Value

of Coal" as published by the American Society for Testing Materials (*1*). Silicon carbide was the inert material used in these tests, the ratio being 15 silicon carbide to 1 coal. The apparatus for crushing test buttons was designed and built in this laboratory.

Discussion of Results

Analyses of the coals are given in Table I. The results of special tests and the effects of oxidation on the characteristics measured by these tests for samples from Henry, Macoupin, and Gallatin Counties are listed in Table II. Table III presents data on a Franklin County whole coal in which exposure was made on −40 +60 mesh coal for both reactivity and agglutinating tests. The main purpose of this study was to determine the effect of oxidation of laboratory samples during storage on the characteristic ignition temperatures and B. S. I. values. Although the agglutinating value data are not complete, they serve their purpose of demonstrating that oxidation did take place. This test was used because it has been found quite sensitive to oxidation of samples.

The sample from Macoupin County designated as durain (Table II) appeared to be durain when picked in the mine, but later examination in the laboratory indicated that it was mainly a dull clarain. The other hand-picked banded ingredients were probably not pure ingredients but, it is believed, were concentrates of sufficient purity to show the predominating characteristics.

A considerable amount of data is presented for comparison of the characteristics of different rank coals and banded ingredients. However, only the effect of oxidation on these characteristics is considered here. Comparisons based on these data and other information not included in this paper will be considered in another report.

In order to judge whether the rather small differences in characteristic ignition temperatures as related to various times of laboratory exposure were significant, it was necessary to have some basis of interpretation. For this purpose standard deviations for duplicate and triplicate values of T_{15} and T_{75} were calculated. Values three times these standard deviations were used as limits. They were found to be 12° and 11.5° C., respectively. It was considered that, if variations exceeding these limits were found, they would indicate a significant trend. Table II shows that the differences in characteristic ignition temperatures, determined after various times of exposure compared to the original values are, in general, considerably smaller than these limiting values. In some cases higher values are reported for samples exposed for longer times. This is more noticeable for the lower rank coals from Henry and Macoupin Counties than for the higher rank coal from Gallatin County. However, it is not possible to estimate the significance of these small increases on the basis of the limited data included in this report. It is concluded that laboratory exposure up to 8 or 9 months does not affect significantly the characteristic ignition temperatures of the samples studied. At the same time the agglutinating value data indicate that oxidation of samples did take place. This is evidenced by the decreasing values as exposure time increases.

It was mentioned earlier that exposure of samples was made in the size used for the various tests. Exposure of reactivity samples was in −40 +60 mesh size and of agglutinating value samples in −200 mesh size. The question arose as to whether oxidation of −40 +60 mesh coal might be negligible even though quite appreciable for −200 mesh samples. To answer this question a whole coal from Franklin County, Ill., was exposed in −40 +60 mesh size for both ignition temperature and agglutinating value tests. For the agglutinating value tests the −40 +60 mesh material was ground to −200 mesh at the time of the tests. The results obtained (Table III) indicate that oxidation did take place in the −40 +60 mesh size as evidenced by the decreasing agglutinating values with increased time of exposure. At the same time characteristic ignition temperatures remained practically the same.

In the interpretation of the effect of oxidation on the British swelling index, a value three times the standard deviation was taken as a limit. This limit was found to be 0.9. A difference of one unit,

TABLE II. RESULTS OF SPECIAL TESTS AND OF OXIDATION

Lab. No.	Description	Ignition Temperature			British Swelling Index		Agglutinating Value	
		T_{15}, °C.	T_{75}, °C.	Days stored	Index No.	Days stored	Kg.	Days stored
		Henry County Samples						
C-2425	Whole coal	164	186	0	4.5	4	...	...
		163	188	8	3.5	11	...	...
		167	187	14	3.0	21	4.6	14
		165	192	42	2.5	45	3.3	47
		168	192	95	1.0	95	3.4	99
		172	194	222	1.0	210	2.8	221
C-2427	Clarain	163	185	1	3.5	4	...	...
		167	188	10	4.0	11	4.8	15
		162	187	19	4.0	21	...	...
		163	188	45	4.0	45	3.1	55
		168	188	96	3.5	95	2.6	94
		172	189	223	2.5	210	2.6	221
C-2428	Durain	175	193	4	3.5	4	...	...
		170	191	7	...	...	...	...
		167	192	12	3.0	11	4.9	13
		167	190	21	2.5	21	...	...
		172	192	46	1.5	45	2.5	49
		175	194	109	1.0	95	3.3	102
		176	195	224	1.0	210	...	...
C-2429	Fusain	170	208	6	1+	4	...	...
		172	204	13	1+	11	...	...
		180	203	24	1+	21	...	...
		178	205	47	1+	45	...	...
		179	207	110	1.0	95	...	...
		181	204	224	1.0	210	...	...
		Macoupin County Samples						
C-2405	Whole coal	160	188	2	4.5	1	...	...
		165	191	5	4.5	5	...	...
		167	192	12	...	...	4.7	14
		164	192	18	...	...	...	...
		161	192	21	4.5	20	...	...
		166	196	47	3.5	61	3.5	48
		170	194	112	2.0	113	2.5	114
		169	196	271	1.5	266	1.7	270
C-2406	Vitrain	160	189	2	3.5	1	...	...
		156	184	5	4.0	5	...	...
		159	190	19	3.5	20	...	...
		162	192	22	...	...	...	...
		164	190	48	4.5	61	...	...
		162	192	113	4.5	113	...	...
		158	193	268	3.5	266	...	...
C-2407	Clarain	159	190	3	3.5	1	...	...
		160	191	6	4.0	5	...	...
		163	194	20	4.0	20	...	...
		165	195	23	...	...	5.5	27
		166	194	49	4.5	61	4.1	51
		163	196	114	4.5	113	3.7	116
		165	196	271	3.5	266	3.3	271

TABLE II. *Concluded*

Lab. No.	Description	Ignition Temperature			British Swelling Index		Agglutinating Value	
		T_{15}, °C.	T_{75}, °C.	Days stored	Index No.	Days stored	Kg.	Days stored
				Macoupin County Samples (Cont'd.)				
C-2408	Durain[a]	161	189	3	3.5	1	...	...
		165	191	9	3.5	5	...	...
		169	192	11	...	...	...	...
		165	190	20	3.5	20	4.1	23
		166	189	25	...	...	...	...
		...	...	...	...	...	3.2	55
		...	...	...	3.0	61	...	...
		173	195	116	1.5	113	2.6	116
		168	198	272	1.0	266	2.3	271
C-2409	Fusain	179	222	4	2.0	1	...	...
		...	...	...	1.5	5	...	...
		183	230	11	...	...	...	...
		186	220	21	1.5	20	...	...
		177	224	26	...	...	...	...
		191	224	50	...	...	...	...
		190	222	118	...	...	...	...
		185	234	272	...	...	...	...
				Gallatin County Samples				
C-2416	Whole coal	207	254	0	7.0	2	8.3	2
		207	252	3	7.0	4	...	...
		202	253	10	7.0	11	...	...
		209	254	56	7.0	59	...	...
		206	256	63	...	...	5.5	64
		207	254	102	6.5	106	...	...
		212	260	241	6.0	235	5.5	239
C-2417	Vitrain	210	258	1	6.5	2	...	...
		215	272	4	6.5	4	8.7	3
		211	267	7	...	...	...	...
		208	265	9	...	...	...	...
		209	270	10	6.5	11	...	...
		208	261	59	6.0	59	...	...
		206	270	65	...	...	5.7	65
		213	257	106	5.0	106	...	...
		210	265	109	...	...	5.7	114
		213	262	245	5.0	235	5.0	240
C-2418	Clarain	211	254	2	6.5	2	...	...
		204	260	5	6.5	4	...	...
		208	255	8	...	...	...	...
		207	259	11	6.5	11	8.3	16
		209	255	59	6.5	59	5.6	65
		211	254	107	6.5	106	5.4	114
		209	255	246	6.0	235	...	...
C-2419	Fusain	211	258	3	1+	2	...	...
		212	254	7	1+	4	...	...
		214	256	11	1+	11	...	...
		210	255	60	...	...	...	...
		209	252	112	...	...	...	...
		214	257	247	...	...	...	...

[a]Probably dull clarain.

Table III. Data for Franklin County Whole-Coal Sample C-2445

Ignition Temperature			Agglutinating Value	
T_{15}, °C.	T_{75}, °C.	Days stored	Kg.	Days stored
182	229	1	5.1	0
184	232	14	3.9	15
182	228	37	3.1	39
184	230	127	...	...
182	228	135	2.0[a]	132

[a]Based on average of eleven buttons.

therefore, was considered significant. The data in Table II, in general agreement with previous experience (*5*), indicate that the B. S. I. numbers decrease with increased exposure time of samples. The values for the higher rank Gallatin County samples show a smaller decrease than those for the lower rank coals from Henry and Macoupin Counties. Values for clarain from Henry and Gallatin Counties show the smallest decrease of the banded ingredients from these two sources. Fusain does not swell, and the values greater than 1 in Table II indicate the presence of other swelling coal material in these samples.

Attention is called to the fact that original B. S. I. numbers for whole coals are higher than those for individual petrographic constituents. Although no definite reason can be given for this at present, the authors have recently noted that the addition of small amounts of fusain to clarain and vitrain results in small increases in B. S. I. numbers for these two constituents. Additional studies are being made.

Conclusions

1. Exposure of −40 +60 mesh samples of whole coal and banded ingredients up to 8 or 9 months to the laboratory atmosphere does not appear to influence significantly the characteristic ignition temperatures of these samples.

2. B. S. I. numbers decrease with increased time of exposure of samples.

3. B. S. I. numbers of the higher rank Gallatin County coal show less decrease with increased time of exposure than do the lower rank coals.

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Presented before the Division of Gas and Fuel Chemistry at the 104th Meeting of the American Chemical Society, Buffalo, N. Y.

ILLINOIS STATE GEOLOGICAL SURVEY
REPORT OF INVESTIGATIONS 86
1943