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PREDICTION OF COAL HYDROPHOBICITY

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ABSTRACT

A statistical analysis of previously published data, as well as newly acquired data, shows that coal hydrophobicity correlates better with moisture content than with carbon content, and better with the moisture/carbon molar ratio than with the hydrogen/carbon or oxygen/carbon atomic ratios. These findings indicate that there is a strong association between hydrophobicity and coal moisture content.

INTRODUCTION

Coals are physically heterogeneous and chemically complex solids which, based on the continuous evolution of the starting plant material towards a pure carbon structure, are ranked as lignite, subbituminous coal, bituminous coal and anthracite. The classification according to rank is probably the most often used and most generally useful classification, and it is thus essentially a means of designating the stage of metamorphism that a coal has reached in its slow, progressive transformation from oxygen-rich plant debris in peat to carbon-rich, oxygen-poor anthracite coal. As this transformation takes place, the following general changes in properties are noted: moisture content decreases, more in the early stages, less in the later stages; carbon content increases; oxygen content decreases; hydrogen content changes very little until later stages are reached, when it decreases markedly; volatile matter content generally decreases; calorific value increases, levels off, and then drops slightly as the anthracite stage is reached; and total void volume (porosity) decreases, levels off among coals with 87-89% carbon before increasing again among anthracites (Berkowitz, 1979; Elliott and Yohe, 1981).

Carbon content is most often used for characterizing rank over the entire range of coal metamorphism as it is the most suitable criterion, although other parameters can also be used for indicating rank for some stages of metamorphism. Carbon is furthermore the major constituent of coal and its determination is a direct and simple procedure. Other constituents are present in smaller amounts, and the determination of some of these constituents is often associated with specific problems. For example, the determination of moisture and oxygen contents is affected by drying and oxidation, respectively; precautions are therefore necessary to obtain reliable values that reflect the true composition of the coal in its fresh unoxidized state. Unfortunately, the bulk composition of the coal may not reflect accurately the chemical composition of the coal on or near the surface which determines the hydrophobic/hydrophilic nature of the material.

Many coals exhibit a certain degree of native hydrophobicity. The more hydrophobic coals (the higher-rank coals) are easily beneficiated by froth flotation or oil agglomeration, while the more hydrophilic coals (the lower-rank coals) are floated or agglomerated with difficulty. Coals of different ranks and often even of the same rank sometimes differ greatly in hydrophobicity as measured by contact angle or natural floatability. Although the degree of hydrophobicity of a coal is related to its rank (Horsley and Smith, 1951; Brown, 1962) and has been correlated with other surface properties of the coal (Fuerstenau et al. 1983; Aplan, 1983; Laskowski, 1986), the known information is still not sufficient to allow a good estimation to be made of the hydrophobicity of a given coal and does not explain the variation of coal hydrophobicity as a function of rank. One reason for this dilemma is the heterogeneity of coal which makes it difficult to experimentally relate hydrophobicity to the contribution of a single chemical component on the coal surface.

Traditionally, various hydrophobic indicators such as natural floatability and contact angle have been plotted against carbon content to illustrate the relationship between hydrophobicity and coal rank. These plots have shown that hydrophobicity increases with an increase in rank, reaches a maximum for medium-volatile bituminous and low-volatile bituminous coals (coals with approximately 88-90% carbon content on a dry and ash-free basis), and decreases for anthracite (Brady and Gauger, 1940; Horsley and Smith, 1951; Brown, 1962; Gutierrez-Rodriguez et al., 1984). Because carbon content has been used in illustrating the variation of coal hydrophobicity with rank, the assumption has been made that the increase in hydrophobicity from lignite to low-volatile bituminous coal is due mainly to an increase in carbon content. This led to numerous explanations to account

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for the "anomalous" behavior of anthracite coal, which is generally less hydrophobic than medium-volatile and low-volatile bituminous coals, even though anthracite has a higher carbon content.

Aplan (1983) noted the close relationship between carbon content and contact angle for a specific range of coals that does not include anthracite coals. According to Aplan, the increase in floatability from lignite to semi-anthracite is related to the increase in carbon content and the decrease in oxygen content. Fuerstenau *et al.* (1983) showed the close relationships between flotation response and carbon content, moisture content, phenolic oxygen content and carboxylic oxygen content for bituminous coals and coals on the borderline between bituminous and subbituminous. Gutierrez-Rodriguez *et al.* (1984) found various relationships between contact angle and fixed carbon, carbon content, oxygen content, and hydroxyl content, with anthracite coals occupying "anomalous" positions in all of these correlations. Rosenbaum and Fuerstenau (1984) concluded that it still remains puzzling how coal hydrophobicity varies with surface composition as rank goes from the high-volatile bituminous coals to anthracite, and explained the variation in coal hydrophobicity in terms of a semi-quantitative model. Keller (1987) explored the relationship of the contact angle with rank in terms of a mathematical relationship in which coal porosity, which is related to the equilibrium moisture content of coal, is an important variable in controlling the degree of hydrophobicity.

From the preceding discussion it appears that the hydrophobicity of unoxidized coals could be due to a number of parameters that are associated with coal rank. Since coal hydrophobicity and coal composition vary with coal rank, it is not unusual to illustrate various relationships between coal hydrophobicity and coal composition. However, this does not prove that the specific rank parameter that is correlated with coal hydrophobicity, is responsible for the change in hydrophobicity. The fact that some of these rank parameters increase, as others decrease, also makes it difficult to demonstrate, for instance, that the change in hydrophobicity is due to an increase in one component rather than due to a decrease in another component. To state that it is due to an increase of one component and a decrease of another component also does not provide information on the quantitative contributions of both components to coal hydrophobicity. The fact that anthracites occupy "anomalous" positions in the hydrophobicity-carbon relationship further illustrates the difficulty in explaining hydrophobicity in terms of compositional changes in the organic matrix of coal.

In an earlier paper, Labuschagne *et al.* (1988) showed by means of a statistical analysis of previously published data that the correlation between coal hydrophobicity and moisture content is better than that between hydrophobicity and carbon content and that the correlation between hydrophobicity and the moisture/carbon ratio is better than that between hydrophobicity and either the hydrogen/carbon or the oxygen/carbon ratio. Unfortunately, the charts presented as Figures 5 and 6 of that paper were interchanged inadvertently. While this did not affect the conclusions, it would be confusing to anyone attempting to repeat the analysis.

For the present paper, the preceding statistical analysis has been extended by including two additional data sets and another indicator of hydrophobicity. Thus, in addition to contact angle and floatability, oil agglomerability is used as an indicator of hydrophobicity. The agglomerability is taken to be the average recovery rate when coal is agglomerated with tetralin under standard conditions. Recent work by the authors indicates a close correlation between coal recovery and other indicators of hydrophobicity. In addition, the present analysis is applied to a greater number of parameters associated with coal rank. These parameters are divided into the following two categories:

- (i) elemental composition parameters reported on a dry and ash-free (daf) basis including carbon content, hydrogen content, oxygen content, hydrogen/carbon atomic ratio, and oxygen/carbon atomic ratio; and
- (ii) other parameters reported on an as-received (ar) basis including carbon content, moisture content, and moisture/carbon mole ratio.

The overall purpose of the present analysis is to evaluate the relationship between the different indicators of coal hydrophobicity and the parameters associated with coal rank in order to show which of these parameters are the better predictors of hydrophobicity. The analysis is based on evaluating the well-known Spearman correlation coefficient and the R-squared multiple correlation coefficient over two different ranges of coal rank. The Spearman correlation coefficient is a nonparametric measure of the association between two variables and it is employed as an index of monotonicity. The R-squared value is used as an index of the degree of conformance of the experimental data to a quadratic function. These indices are evaluated over Range I which include data for coals ranging from lignite to low-volatile bituminous coal (88-90% carbon content) and over Range II which include data for all coals ranging from lignite to anthracite.

SOURCES OF EXPERIMENTAL DATA

Data from several published sources have been utilized for the present analysis, as well as data generated by the first author of this paper by agglomerating South African coals. The various sources of data for coals in both Range I and Range II are listed in Table I. The various indicators of hydrophobicity are also listed including contact angle measured by the captive bubble (CB) technique and contact angle measured by the sessile drop (SD) method. Agglomerability is based on the average rate of organic recovery during an agglomeration test.

Agglomerability data were collected for the South African coals listed in Table II. These coals were crushed to approximately 5 mm size and ballmilled in water to approximately 10 μ m size. For each agglomeration test, 20 g coal and sufficient water to form 200 ml of slurry were placed in a 500 ml beaker which was fitted with a Morat R20S digitronic agitator. The coal slurry was stirred at 1000 rpm and agglomerated with 15 ml tetralin (1,2,3,4-tetrahydronaphthalene) as the bridging liquid. The average rate of organic recovery (k) was calculated using the formula:

$$k = R/t$$

where R = percentage organic recovery obtained in time t; and
t = agglomeration time in seconds

The organic recovery on an oil-free basis (% R) was determined using the formula:

$$\% R = 100 [(y_3 - y_1)(100 - y_2)] / [(y_3 - y_2)(100 - y_1)]$$

where y_1 = % ash in the feed;
 y_2 = % ash in the agglomerated coal; and
 y_3 = % ash in the tailings.

These percentages (y) were determined by ashing the fractions (feed, agglomerates, reject) at 825°C for 10 h.

Agglomeration time (t), measured in seconds, is the minimum time needed for complete agglomeration. However, when agglomeration was incomplete after 3000 s, the process was stopped, and the organic recovery was measured at the end of this period. The method used to determine agglomeration time was previously described in detail (Labuschagne, 1986).

STATISTICAL METHODS

The eight parameters associated with coal rank which are listed above were statistically compared to see how well they correlate with hydrophobicity. The degree of correlation between each parameter and an indicator of hydrophobicity for the ten data sets (Table I) was evaluated by two different statistical methods.

The first statistical method is based on the value of the Spearman correlation coefficient (ρ):

$$\rho = \frac{\sum_{i=1}^n \left(R_i - \frac{n+1}{2} \right) \left(S_i - \frac{n+1}{2} \right)}{\sum_{i=1}^n \left(i - \frac{n+1}{2} \right)^2}$$

where n = the number of coal samples (data pairs X_i, Y_i);
 X_i = the value of the coal rank parameter of the i th sample;
 Y_i = the value of the hydrophobicity indicator of the i th sample;
 R_i = the relative rank of X_i among the n values of X; and
 S_i = the relative rank of Y_i among the n values of Y.

The absolute value of ρ measures the degree of monotonicity of the observed (X,Y) relationship. It equals one when the relationship is entirely increasing monotonically (when the smallest X and smallest Y belong to the same coal sample, as do the second-smallest X and the second-smallest Y, etc.) or entirely decreasing monotonically (when the smallest X and the largest Y belong to the same coal sample, as do the second-smallest X and the second-largest Y, etc.). It equals zero when the relation is completely disordered.

The ρ -based competitions among the rank parameters as predictors of hydrophobicity are especially relevant for the range from lignite to low-volatile bituminous coal (Range I), since hydrophobicity tends generally to be monotonic in all the rank parameters over this range of coal ranks. In other words, ρ is an indicator of the degree of monotonicity of the various rank parameters, all of which tend to be monotonic in this range. The ρ -based competitions among the rank parameters as predictors of hydrophobicity are also relevant for the range lignite to anthracite (Range II), even though hydrophobicity does not tend to be monotonic for all rank parameters in this range (e.g., the hydrophobicity-carbon relationship does not appear monotonic when anthracite data are included). Thus, ρ again is an indicator of the degree of monotonicity of the various rank parameters, not all of which tend to be monotonic in Range II.

The second statistical method is based on the multiple correlation coefficient about quadratic regression:

$$R^2 = \frac{\sum_{i=1}^n (Y_i - \bar{Y})^2 - \sum_{i=1}^n (Y_i - (a + bX_i + cX_i^2))^2}{\sum_{i=1}^n (Y_i - \bar{Y})^2}$$

where $Y = a + bX + cX^2$ is the least squares quadratic regression function of Y on X .

R-squared measures the degree of conformance of the (X,Y) relation to the best-fitting quadratic function. It equals one when all (X_i, Y_i) points fall precisely on the best-fitting (least squares) quadratic function. It equals zero when the best-fitting quadratic function is a horizontal straight line, so that the dispersion of the Y values about the best-fitting quadratic function is no smaller than their dispersion as measured by the relation-insensitive variance numerator

$$\sum_{i=1}^n (Y_i - \bar{Y})^2.$$

The R-squared based competitions among all the rank parameters as predictors of hydrophobicity are relevant for the range lignite to low-volatile bituminous coal (Range I) and for the range lignite to anthracite (Range II) since quadratic functions allow for monotone quadratics.

DISCUSSION

To quantify the degree of correlation between various indicators of coal hydrophobicity and different parameters associated with coal rank, the Spearman correlation coefficient (ρ) and the multiple correlation coefficient (R-squared) were calculated for the eight coal rank parameters using each of the 10 data sets listed in Table I. The calculated coefficients are listed in Tables III and IV. An insight into the information in Tables III and IV is given in Tables V through VIII. These tables compare the different rank parameters as predictors of coal hydrophobicity and indicate the number of times any given rank parameter is ranked first, second, third, etc., on the basis of the values presented in Tables III and IV.

Table V indicates the number of times the Spearman correlation coefficient is ranked first, second, third, etc., on the basis of ρ -values calculated for Range I. This is the range in which all rank parameters show a monotone relationship with hydrophobicity. In this table there are five different groupings of rank parameters so that comparisons can be made among a few or among all of the parameters as predictors of hydrophobicity.

In Table V, group (1) consists of coal rank parameters which reflect the chemical composition of the organic matrix on a dry and ash-free basis. The data show that oxygen content is associated with the highest value of ρ in four of the six data sets; that the oxygen/carbon ratio is associated with the highest value of ρ in three of the six data sets (oxygen content and oxygen/carbon ratio share the highest value in three data sets); and that carbon content is associated with the highest rank of ρ in two of the six data sets. The oxygen/carbon ratio is also associated with the second-highest value of ρ in three data sets, having been associated with the highest value of ρ in the other three data sets. Therefore among the parameters in group (1), coal hydrophobicity is primarily associated with carbon content, oxygen content or the oxygen/carbon atomic ratio, as has often been suggested. However, when these rank parameters are compared with other rank parameters that reflect more than elemental composition, a different conclusion is reached.

In Table V, group (2) consists of rank parameters that were calculated on an as-received basis. The data show that moisture content or the moisture/carbon ratio is associated with the highest value of ρ in four of the six data sets (moisture content and moisture/carbon ratio share the highest value of ρ in three data sets). On the other hand, carbon content is associated with the highest value of ρ in only one data set, and with the lowest value of ρ in five of the six data sets. Thus moisture content or the moisture/carbon ratio is a better predictor of hydrophobicity than carbon content.

In Table V, group (3) consists of the two carbon content parameters (dry and ash-free and as-received) and moisture content (as-received). Moisture content is associated with the highest value of ρ in four of the six data sets, and with the second-highest value of ρ in the remaining two data sets. On the other hand, each of the carbon content parameters is associated with the highest value of ρ only once, and three times each with the lowest value of ρ . Therefore, the relationship between hydrophobicity and moisture content is generally much better than that between hydrophobicity and either of the carbon content parameters.

In Table V, group (4) consists of the hydrogen/carbon atomic ratio, oxygen/carbon atomic ratio and moisture/carbon mole ratio. The moisture/carbon ratio is associated with the highest value of ρ in four of the six data sets, and with the second-highest value of ρ in the remaining two data sets. The oxygen/carbon ratio is associated with the highest value of ρ in two data sets, and with the second-highest value of ρ in the remaining four data sets. The hydrogen/carbon ratio is associated with the lowest value of ρ in all six data sets. Therefore, the moisture/carbon ratio is generally a better predictor of hydrophobicity than either the oxygen/carbon ratio or the hydrogen/carbon ratio.

In Table V all the rank parameters are combined in group (5). Carbon content (daf), oxygen content (daf), oxygen/carbon ratio (daf) and carbon content (ar) are associated with the highest value of ρ once (oxygen content and oxygen carbon ratio share the highest value of ρ in one data set), while moisture content is associated with the highest value of ρ in three data sets and the moisture/carbon ratio with the highest value of ρ in two data sets (moisture content and moisture/carbon ratio share the highest value of ρ in two data sets).

The results in Table V show that moisture-based rank parameters are generally better predictors of hydrophobicity than the other rank parameters, suggesting that the elemental composition of the organic coal matrix is not the primary factor which determines the degree of hydrophobicity.

Table VI indicates the number of times the Spearman correlation coefficient is ranked first, second, third, etc., on the basis of ρ -values calculated for Range II. This is the range which includes anthracite, and therefore not all rank parameters show a monotone relationship with hydrophobicity. In this table the rank parameters are placed in similar groups as in Table V. Table VI indicates that moisture-based parameters (moisture content and moisture/carbon ratio) always rank highest as predictors of hydrophobicity when compared to other coal rank parameters.

The ρ -results also show that the moisture based parameters to be as monotonic over Range II as over Range I. This can be seen when the ρ -values listed in Table III for data sets A, B, C and D are compared with those for data sets G, H, I and J, respectively (these data sets are identical except for the exclusion/inclusion of anthracite samples). The other rank parameters are less monotonic over Range II than over Range I.

Tables VII and VIII indicate the number of times R-squared is ranked first, second, third, etc., on the basis of R-squared calculated for Ranges I and II, respectively. The R-squared results show that, for Range I, the correlation between hydrophobicity and moisture-based rank parameters compares very favorably to that with other rank parameters. Values of R-squared listed for the moisture-based parameters in Table IV are also generally higher compared to values of R-squared for the other parameters. Table VIII indicates that, for Range II, the correlation between hydrophobicity and moisture-based rank parameters is always better than the correlation between hydrophobicity and other rank parameters. The ranking of R-squared values thus follows a pattern very much like that of the ρ -values.

The ρ -values and R-squared results identified moisture-based parameters as the best predictors of hydrophobicity, while the ρ -results also showed moisture based parameters to be as monotonic over Range II as over Range I. These findings imply that the variation in inherent moisture content, and not the variation in carbon content (as has often been assumed), predicts more accurately the variation in the degree of coal hydrophobicity. This is in agreement with the mathematical models of Rosenbaum and Fuerstenau (1984) and Keller (1987) concerning the variation of contact angle with rank.

The model of Fuerstenau and Rosenbaum (when analyzed) shows that the variation in number of hydrophilic sites is primarily responsible for the variation in the degree of hydrophobicity (i.e., that a decrease in number of hydrophilic sites is primarily responsible for the increase in hydrophobicity from lignite to medium-volatile and low-volatile bituminous coals, and that an increase in number of hydrophilic sites is primarily responsible for the decrease in hydrophobicity from medium-volatile and low-volatile bituminous coals to anthracite). The model of Keller (when analyzed) specifically indicates that changes in the equilibrium moisture content of coal is responsible for the classical variation in hydrophobicity with rank (i.e., responsible for the increase in hydrophobicity from lignite to medium-volatile and low-volatile bituminous coals, and the decrease in hydrophobicity from medium-volatile and low-volatile bituminous coals to anthracite). Anthracite samples should thus occupy "normal" or expected positions on hydrophobicity-moisture curves. This has been experimentally demonstrated (Labuschagne, 1987).

The high degree of correlation between hydrophobicity and the moisture-based parameters (moisture content and moisture/carbon ratio) is remarkable as the discovery is based on results which arose from studies in which the moisture content may not have been specially controlled for the purpose. Therefore, the "as-received" moisture content of the coal samples may not have reflected accurately the natural coal bed or inherent moisture content. Since the latter is a more fundamental property of the coal than "as-received" moisture content, it could provide a better correlation with hydrophobicity. By conducting measurements in which moisture content is carefully controlled and by correlating hydrophobicity with the natural coal bed moisture content, it may be possible to demonstrate an even tighter correlation between hydrophobicity and moisture content than the ones revealed in this study.

CONCLUSIONS

Based on the results of this study, it can be concluded that:

- (i) the variation in hydrophobicity with coal rank appears not to be controlled solely by the coal content of any single element such as carbon or by the ratio of the concentrations of two different elements (e.g., hydrogen/carbon); and
- (ii) the variation in hydrophobicity with coal rank appears to be determined by factors which control the moisture content of the coal.

Consequently, moisture-based parameters associated with coal rank appear to be better predictors of hydrophobicity than the other parameters which were investigated.

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Table I. Sources of experimental data.

Range	Set No.	Indicator of hydrophobicity	No. points	Data source
I	A	contact angle (CB)	14	Sun, 1954
I	B	contact angle (CB)	17	Gutierrez-Rodriguez <u>et al.</u> , 1984
I	C	contact angle (SD)	17	Gutierrez-Rodriguez <u>et al.</u> , 1984
I	D	floatability	19	Sun, 1954
I	E	floatability	9	Fuerstenau <u>et al.</u> , 1983
I	F	agglomerability	26	Table II
II	G	contact angle (CB)	20	Sun, 1954
II	H	contact angle (CB)	19	Gutierrez-Rodriguez <u>et al.</u> , 1984
II	I	contact angle (SD)	19	Gutierrez-Rodriguez <u>et al.</u> , 1984
II	J	floatability	27	Sun, 1954

Table II. Rate of organic recovery and coal composition on as-received (ar) and dry and ash-free (daf) bases.

Coal No.	Recovery rate k(%/s)	Parameter (ar)		Parameter (daf)				
		H ₂ O(%)	ash(%)	C(%)	H(%)	O(%)	N(%)	S(%)
1	1.90	1.7	11.8	89.75	3.78	3.55	2.38	0.54
2	0.63	2.7	17.8	83.77	4.96	8.33	2.21	0.73
3	0.49	2.5	11.1	82.69	4.21	10.59	2.06	0.45
4	0.38	2.4	20.3	81.46	4.57	9.48	2.12	2.37
5	0.34	2.8	14.6	84.39	4.41	7.71	2.13	1.36
6	0.32	2.8	8.2	81.34	4.49	11.80	1.97	0.40
7	0.24	2.8	13.3	83.13	4.83	9.13	2.10	0.81
8	0.23	3.3	10.2	83.12	4.77	9.75	1.95	0.41
9	0.17	3.4	16.9	81.86	4.58	11.30	2.03	0.23
10	0.16	3.5	9.9	80.50	4.56	12.17	1.90	0.87
11	0.07	4.0	16.6	79.52	4.69	13.61	1.94	0.24
12	0.05	4.0	18.9	80.49	4.22	12.56	1.87	0.86
13	0.04	4.6	14.2	82.61	4.09	10.81	1.97	0.52
14	0.04	4.4	15.1	80.36	3.86	12.27	1.93	1.58
15	2.38	1.6	11.7	89.10	3.60	4.02	2.24	1.04
16	0.50	2.5	13.3	84.22	4.64	7.69	2.05	1.40
17	2.71	1.6	14.1	89.95	3.77	3.01	2.29	0.98
18	0.33	3.7	11.4	82.34	4.90	10.09	1.81	0.86
19	0.04	4.6	18.4	79.10	4.52	13.38	1.79	1.21
20	0.28	3.7	18.6	82.97	4.34	10.11	1.83	0.75
21	0	5.1	21.5	78.05	4.25	14.50	1.91	1.29
22	0	5.2	19.2	78.68	4.33	14.54	1.93	0.52
23	0	5.9	30.9	81.74	4.53	11.25	2.06	0.42
24	0	8.1	26.0	77.18	4.05	16.31	2.06	0.40
25	0	7.6	28.4	76.47	4.00	17.08	2.00	0.45
26	0.32	3.0	16.1	83.18	4.30	8.54	2.15	1.83

Table III. Spearman correlation coefficients for the relationship between coal hydrophobicity and coal rank parameters.

Set No.	Parameter (daf basis)					Parameter (ar basis)		
	C	H	O	H/C	O/C	C	H ₂ O	H ₂ O/C
A	0.750	0.382	0.817	0.306	0.817	0.899	0.913	0.913
B	0.852	0.234	0.818	0.575	0.839	0.795	0.869	0.869
C	0.792	0.133	0.771	0.478	0.778	0.689	0.727	0.738
D	0.765	0.293	0.840	0.351	0.840	0.932	0.927	0.928
E	0.717	0.600	0.800	0.083	0.800	0.767	0.783	0.783
F	0.847	0.059	0.882	0.155	0.880	0.813	0.959	0.946
G	0.386	0.274	0.600	0.087	0.600	0.769	0.911	0.892
H	0.805	0.294	0.774	0.583	0.792	0.765	0.900	0.900
I	0.676	0.133	0.678	0.415	0.682	0.593	0.758	0.764
J	0.214	0.416	0.549	0.060	0.549	0.697	0.862	0.855

Table IV. R-squared values for the relationship between coal hydrophobicity and coal rank parameters.

Set No.	Parameter (daf basis)					Parameter (ar basis)		
	C	H	O	H/C	O/C	C	H ₂ O	H ₂ O/C
A	0.585	0.143	0.669	0.056	0.659	0.886	0.872	0.856
B	0.805	0.135	0.765	0.313	0.786	0.821	0.890	0.877
C	0.840	0.107	0.806	0.247	0.819	0.682	0.777	0.702
D	0.067	0.744	0.052	0.593	0.050	0.448	0.959	0.962
E	0.795	0.283	0.737	0.428	0.746	0.887	0.717	0.725
F	0.942	0.647	0.929	0.881	0.920	0.723	0.739	0.665
G	0.349	0.044	0.411	0.045	0.398	0.596	0.608	0.581
H	0.642	0.041	0.651	0.290	0.664	0.634	0.879	0.863
I	0.493	0.054	0.504	0.266	0.515	0.439	0.734	0.622
J	0.045	0.071	0.076	0.022	0.073	0.435	0.952	0.957

Table V. Spearman correlation coefficients ranked according to order for Range I (data sets A to F).

Groups of parameters associated with coal rank	Number of times ρ ranked ^a							
	1st	2nd	3rd	4th	5th	6th	7th	8th
(1) carbon (daf)	2	0	4	0	0	-	-	-
hydrogen (daf)	0	0	0	2	4	-	-	-
oxygen (daf)	4	0	2	0	0	-	-	-
hydrogen/carbon (daf)	0	0	0	4	2	-	-	-
oxygen/carbon (daf)	3	3	0	0	0	-	-	-
(2) carbon (ar)	1	0	5	-	-	-	-	-
moisture (ar)	4	1	1	-	-	-	-	-
moisture/carbon (ar)	4	2	0	-	-	-	-	-
(3) carbon (daf)	1	2	3	-	-	-	-	-
carbon (ar)	1	2	3	-	-	-	-	-
moisture (ar)	4	2	0	-	-	-	-	-
(4) hydrogen/carbon (daf)	0	0	6	-	-	-	-	-
oxygen/carbon (daf)	2	4	0	-	-	-	-	-
moisture/carbon (ar)	4	2	0	-	-	-	-	-
(5) carbon (daf)	1	0	1	0	1	3	0	0
hydrogen (daf)	0	0	0	0	0	0	2	4
oxygen (daf)	1	0	2	2	1	0	0	0
hydrogen/carbon (daf)	0	0	0	0	0	0	4	2
oxygen/carbon (daf)	1	1	0	4	0	0	0	0
carbon (ar)	1	0	1	0	1	3	0	0
moisture (ar)	3	0	2	0	1	0	0	0
moisture/carbon (ar)	2	2	1	1	0	0	0	0

^aNot all columns add up to 6 due to positions shared (some parameters have the same ρ -value in a specific data set); all rows add up to 6.

Table VI. Spearman correlation coefficients ranked according to order for Range II (data sets G to J).

Groups of parameters associated with coal rank	Number of times ρ ranked ^a							
	1st	2nd	3rd	4th	5th	6th	7th	8th
(1) carbon (daf)	1	0	2	1	0	-	-	-
hydrogen (daf)	0	0	1	1	2	-	-	-
oxygen (daf)	2	1	1	0	0	-	-	-
hydrogen/carbon (daf)	0	0	0	2	2	-	-	-
oxygen/carbon (daf)	3	1	0	0	0	-	-	-
(2) carbon (ar)	0	0	4	-	-	-	-	-
moisture (ar)	3	1	0	-	-	-	-	-
moisture/carbon (ar)	2	2	0	-	-	-	-	-
(3) carbon (daf)	0	2	2	-	-	-	-	-
carbon (ar)	0	2	2	-	-	-	-	-
moisture (ar)	4	0	0	-	-	-	-	-
(4) hydrogen/carbon (daf)	0	0	4	-	-	-	-	-
oxygen/carbon (daf)	0	4	0	-	-	-	-	-
moisture/carbon (ar)	4	0	0	-	-	-	-	-
(5) carbon (daf)	0	0	1	0	1	1	1	0
hydrogen (daf)	0	0	0	0	0	1	1	2
oxygen (daf)	0	0	0	3	1	0	0	0
hydrogen/carbon (daf)	0	0	0	0	0	0	2	2
oxygen/carbon (daf)	0	0	1	3	0	0	0	0
carbon (ar)	0	0	2	0	0	2	0	0
moisture (ar)	3	1	0	0	0	0	0	0
moisture/carbon (ar)	2	2	0	0	0	0	0	0

^aNot all columns add up to 4 due to positions shared (some parameters have the same ρ -value in a specific data set); all rows add up to 4.

Table VII. R-squared values ranked according to order for Range I (data sets A to F).

Groups of parameters associated with coal rank	Number of times R^2 ranked							
	1st	2nd	3rd	4th	5th	6th	7th	8th
(1) carbon (daf)	4	0	2	0	0	-	-	-
hydrogen (daf)	1	0	0	1	4	-	-	-
oxygen (daf)	1	1	3	1	0	-	-	-
hydrogen/carbon (daf)	0	1	0	4	1	-	-	-
oxygen/carbon (daf)	0	4	1	0	1	-	-	-
(2) carbon (ar)	2	1	3	-	-	-	-	-
moisture (ar)	3	2	1	-	-	-	-	-
moisture/carbon (ar)	1	3	2	-	-	-	-	-
(3) carbon (daf)	2	1	3	-	-	-	-	-
carbon (ar)	2	2	2	-	-	-	-	-
moisture (ar)	2	3	1	-	-	-	-	-
(4) hydrogen/carbon (daf)	0	2	4	-	-	-	-	-
oxygen/carbon (daf)	3	2	1	-	-	-	-	-
moisture/carbon (ar)	3	2	1	-	-	-	-	-
(5) carbon (daf)	2	1	0	1	0	2	0	0
hydrogen (daf)	0	0	1	0	0	0	1	4
oxygen (daf)	0	1	1	2	0	1	1	0
hydrogen/carbon (daf)	0	0	0	2	0	0	3	1
oxygen/carbon (daf)	0	1	2	0	2	0	0	1
carbon (ar)	2	0	1	0	1	2	0	0
moisture (ar)	1	2	0	1	1	1	0	0
moisture/carbon (ar)	1	1	1	0	2	0	1	0

Table VIII. R-squared values ranked according to order for Range II (data sets G to J).

Groups of parameters associated with coal rank	Number of times R^2 ranked							
	1st	2nd	3rd	4th	5th	6th	7th	8th
(1) carbon (daf)	0	0	3	1	0	-	-	-
hydrogen (daf)	0	0	1	0	3	-	-	-
oxygen (daf)	2	2	0	0	0	-	-	-
hydrogen/carbon (daf)	0	0	0	3	1	-	-	-
oxygen/carbon (daf)	2	2	0	0	0	-	-	-
(2) carbon (ar)	0	1	3	-	-	-	-	-
moisture (ar)	3	1	0	-	-	-	-	-
moisture/carbon (ar)	1	2	1	-	-	-	-	-
(3) carbon (daf)	0	2	2	-	-	-	-	-
carbon (ar)	0	2	2	-	-	-	-	-
moisture (ar)	4	0	0	-	-	-	-	-
(4) hydrogen/carbon (daf)	0	0	4	-	-	-	-	-
oxygen/carbon (daf)	0	4	0	-	-	-	-	-
moisture/carbon (ar)	4	0	0	-	-	-	-	-
(5) carbon (daf)	0	0	0	0	2	1	1	0
hydrogen (daf)	0	0	0	0	0	1	0	3
oxygen (daf)	0	0	0	4	0	0	0	0
hydrogen/carbon (daf)	0	0	0	0	0	0	3	1
oxygen/carbon (daf)	0	0	2	0	2	0	0	0
carbon (ar)	0	1	1	0	0	2	0	0
moisture (ar)	3	1	0	0	0	0	0	0
moisture/carbon (ar)	1	2	1	0	0	0	0	0

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