Coal proximate analyses correlation with airborne respirable dust

S. J. Page, J. A. Organiscak and J. Quattro
Pittsburgh Research Center, US Bureau of Mines, PO Box 18070, Pittsburgh, PA 15236-0070, USA
SSI Services, Inc. PO Box 10826, Pittsburgh, PA 15236-0826, USA
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Laboratory tests have been performed on ten coal channel samples from Eastern, Midwestern and Western United States coal provinces to investigate the relationship of coal proximate analysis characteristics with the quantity of airborne respirable dust (ARD) generated. In general, strong correlations were obtained with fixed carbon, inherent moisture and volatile content. In particular, the singular composite variable (fixed carbon/volatile)/moisture provided a significant negative exponential correlation with ARD. It is shown that this relationship is exponential and not an inverse function. The initial objective of the study was to examine the relationship between coal characteristics and ARD and the subsequent implications for underground coal mining dust control technology to protect the health of coal miners. However, it appears clear that these results may be useful for other purposes such as fundamental investigations of coal strength, environmental issues of fugitive dust from coal preparation and processing, and coal cleaning by pulverization to micrometre-sized particles.

(Keywords: airborne respirable dust; coal proximate analyses; coal characteristics)

A previous study[1] by the authors investigated the following question: is bituminous coal seam type an influential factor in airborne respirable dust (ARD) generation? The conclusions of that study can be summarized as:

1. Seam characteristics were found to be an influential factor in the amount of ARD generated at longwalls.
2. High volatile, low ash bituminous seams tend to have higher ARD levels when mining.
3. The ash parameter could be described by non-linear models for both ARD and specific dust.
4. Additional research studies should strive to verify the ash and volatility association with ARD under strictly controlled conditions.

The authors have reviewed ASTM standard D 547-41, entitled 'Standard Test Method for Index of Dustiness of Coal and Coke'. This standard provides an empirical, although somewhat arbitrary, measure of the dust produced when handling coal or coke. However, dust is arbitrarily classified as either coarse or float based on specified settling times and in no way yields information on ARD or coal characteristics. Previous researchers[2] have related various coal cutting parameters to volatile matter content of coals. These cutting parameters are typically compressive strength and mean peak grooving force. Hanson and Roepke[3] have related ARD to cutting parameters such as bit angle of attack, bit tip radius, and depth of cut for four coal seams. The general result of this work shows that the Pocahontas coal produced less ARD than the other three coal seams tested by more than an order of magnitude. Coincidentally, the Pocahontas seam is known for its high rank, low volatile nature. However, no information could be found that attempts to relate ARD to coal proximate analysis for a representative number of coal types. To this end, laboratory tests have been performed. This paper describes the results of these tests.

It should be stated that the initial objective of the study was to examine the relationship between coal characteristics and ARD and the subsequent implications for underground coal mining dust control technology to protect the health of coal miners. However, it appears clear that these results may be useful for other purposes such as investigations of coal strength as well as environmental issues of fugitive dust from coal preparation and processing.

EXPERIMENTAL

Sample collection/preparation

Channel samples were collected from active faces at ten coal mines in nine seams from the Eastern and Rocky Mountain United States provinces. The Eastern province samples represent coals from both Northern and Southern regions. Each channel sample was initially sieved to obtain 4.75–5.66 mm (0.187–0.223 in) samples already present in the material. To obtain additional material for the tests, lumps of channel coal were selected, hand-broken and resieved. Rock pieces were intentionally rejected from the prepared samples for two reasons. First, the constructed breaker could not break rock suitably. Second, rock-generated dust should be treated separately from coal-generated dust because of the likely difference in generation characteristics.

Residual moisture from the prepared channel samples

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was removed by baking at 37.4°C (100°F) until no weight change occurred after 1 h baking time. The purpose of this procedure was to reduce the effect of dust agglomeration due to residual surface moisture during the coal breaking process.

Respirable dust generation

Fresh respirable dust for each sample was continuously generated for a period of 1.5 h by passing the sample through a breaker consisting of two counter-rotating knurled cylindrical rollers. This process was chosen for several reasons. First, underground coal cutting is quite representative of a breaking process. Second, using an actual cutting process would require much larger sample sizes than were obtainable. The samples were gravity-fed from a hopper into the 3.17 mm (1/8 in) clearance between the rollers. The breaker was powered by a silicon-controlled rectifier (SCR) variable speed d.c. motor. The motor current was monitored and maintained constant for all tests. Thus, for all tests the power input was eliminated as a variable in dust generation.

The breaker was placed in a sealed environmental chamber with relative humidity and temperature monitored and maintained at approximately constant values of 21–37% and 20.9–24.8°C (70–77°F) during the series of tests.

Dust samples were drawn from the chamber at 2.0 l min^{-1} and gravimetrically collected on standard 37 mm filter cassettes for a period of 1.5 h. Cyclone preseparation was not used for two reasons. First, it is felt that any correlations between airborne dust and coal characteristics need not follow man-made classification criteria. Second, the airborne dust generated and sampled was of respirable size naturally (no airborne particles larger than 7 μm, as verified by impactor sampling), although not in the same proportion as defined by cyclone preclassification.

At the end of the test, the material passing through the breaker was removed and weighed. The breaker, feed and collection hoppers, and dust sampling line were vacuumed and flushed clean. This procedure was also followed for replicate tests of the same coal sample.

The material that passed through the breaker was collected for each test (replicate or not) and analysed for ash, moisture, volatile matter and fixed carbon content. *Table 1* summarizes the proximate results for the tests as well as the dust concentrations obtained during the tests. Since it was not the purpose of this study to characterize accurately by proximate analysis the coal seam itself, it was not necessary to follow ASTM standard sample preparation techniques. Rather, it was hoped that variations in proximate analyses between the replicate samples from a given coal seam would manifest themselves in the quantities of airborne dust generated. ASTM standards for the proximate analysis were used.

Table 1 Proximate analyses of broken coal and dust data

	As-determined percentages					
Sample	Moisture	Ash	Volatile matter	Fixed carbon	Dust (mg m^{-3})	HGI
1	1.1	6.8	36.3	55.8	1.13	59
2	1.1	5.8	36.6	56.5	1.13	59
3	0.9	4.0	30.6	64.5	0.46	79
4	0.9	4.7	30.1	64.4	0.66	79
5	0.5	7.4	22.5	69.7	0.12	104
6	0.5	7.0	21.6	70.9	0.16	104
7	0.6	2.1	31.9	65.4	0.28	65
8	0.4	2.1	32.9	64.6	0.34	65
9	0.6	4.6	31.6	63.2	0.34	53
10	0.6	4.5	31.1	63.7	0.18	53
11	0.5	5.3	29.2	65.1	0.16	101
12	0.5	5.5	29.7	64.3	0.57	101
13	1.4	13.3	30.9	54.3	1.36	54
14	1.5	13.4	30.8	54.2	2.03	54
15	3.8	3.1	45.5	47.6	2.26	51
16	2.3	5.5	44.8	47.5	2.37	50
17	2.9	7.9	42.0	47.2	3.69	45
18	2.9	8.0	42.1	47.0	2.66	45

RESULTS

In general, strong correlations were obtained between fresh-generated ARD and fixed carbon, volatile matter and moisture content. No correlation was found with ash content, contrary to the result of the previous underground study. The method of analysis performed was the Marquardt–Levenberg method which systematically adjusts the parameters until a least-squares solution is reached.

Fixed carbon and volatile matter

A negative exponential fit to the ARD–fixed carbon data gave a coefficient of determination (r^2) of 0.95 with the high fixed carbon samples yielding dust concentrations lower by more than an order of magnitude.

A positive exponential fit to the ARD–volatile matter data gave an r^2 value of 0.88 with high volatile samples yielding dust concentrations higher by more than an order of magnitude. These results are remarkably similar over the same volatile range in comparison to data taken from Pomeroy and Foote[2]; their plots of impact strength of coals versus dry, ash-free (daf) volatile matter show the same functional form over the parameter range of 20–50% volatile matter. This indicates that, in this volatile range, ARD is directly proportional to coal impact strength.

Ash content

Figure 1 plots the dust concentration (in mg m^{-3}) versus per cent ash on a dry basis. It is quite apparent that there is no correlation. It is quite reasonable to question why the underground data from the previous study showed a strong correlation when the controlled laboratory study did not. Perhaps one of the most logical explanations is due to cleat frequency. It has been postulated by others[4–8] (although to the authors' knowledge it has not been tested) that the cleat lines of coal may contain a disproportionately higher amount of ash than is in the coal itself. These theories are based on geochemical considerations of the processes occurring in the syngenetic (at the time of bed formation) and epigenetic (after bed formation) pyrite formation. Cleating of coal seams does not occur at the time of bed formation. Epigenetic deposits include the category of deposition within the cleats. If this is so, then coals that are more heavily cleated would have higher ash contents than less cleated coals. Underground longwall shearers have bits that quite easily penetrate numerous cleats. Because cleats are planes of weakness in the coal, the coal will tend to break more along the cleats, in addition to breaking more easily. As a result, more ash would be liberated from the heavily cleated coals with presumably

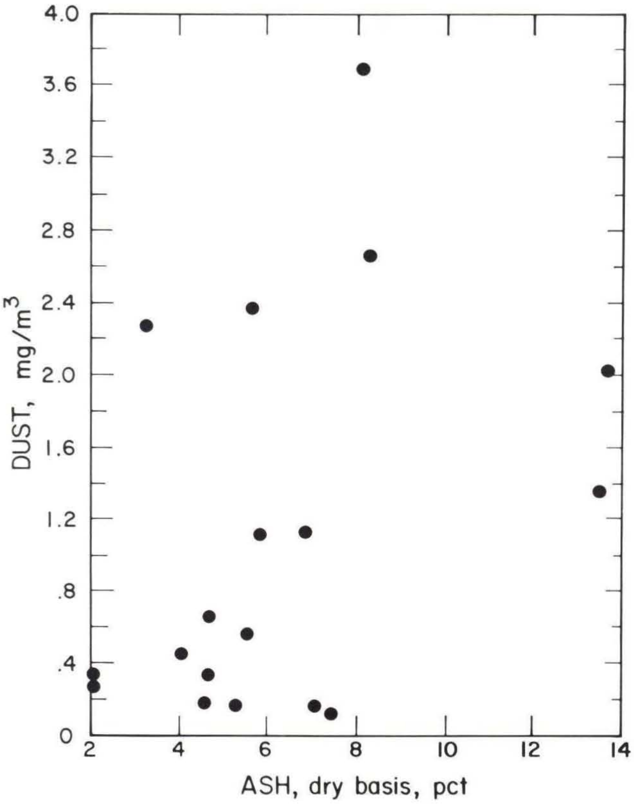

Figure 1 Airborne respirable dust concentration versus ash content

less dust make. Pomeroy and Foote[2] found a significant inverse linear correlation between mean peak grooving force and cleat frequency over a frequency range of 0 to 259 cleats m^{-1} (0 to 79 cleats ft^{-1}). They gave no explanation for the relationship other than mechanical weakness due to cleating.

In contrast, the laboratory tests were performed on minus 6.35 mm (1/4 in) coals. At this level of material size, cleat structure is insignificant or non-existent and therefore would not show a correlation with airborne dust make. Although there did not appear to be a correlation between airborne dust and ash content of the coals, the data suggest that there may be some fundamental relationship between dust and fixed carbon, volatile matter and inherent moisture. Elimination of the cleat structure variable, although differing from real mining conditions, may provide a clearer understanding of this relationship.

Moisture content

Figure 2 shows the scatter plot of per cent moisture versus dust concentration (in mg m^{-3}). The data of *Figure 2* show a definite relationship between ARD and moisture. This relationship would reasonably suggest a linear model with the exception of the unpaired data point at 3.8%. If this point is an anomaly it would be quite reasonable to exclude it from the analysis. However, since the data point does not have a replicate for comparison, it cannot be excluded. If this data point is accurate, a non-linear model may be reasonable. A case will be made later for an exponential relationship.

The exponential function in *Figure 2* provides a visually good fit and plausible explanation to the data with an r^2 of 0.96. The model assumes that the dust concentration maintains a constant value after a certain moisture content is achieved. Based on a peculiar result of the tests, this may be a valid assumption. Further speculation on this point will not be made until further testing at moisture values above 3% is performed.

Fuel ratio

The fuel ratio is defined as the ratio of fixed carbon to volatile matter and may be a very important measure of coal strength[9]. Use of the fuel ratio dates back to at least 1903[10]. At this point it is important to clarify the use of the term 'strength'. Although there are many strength indices, that which is appropriate for mechanical cutting (mining) is not clear. For the purpose of discussion here, strength is assumed to be a measure of the ease or difficulty with which a coal can be mined. Generally speaking, this strength is frequently considered synonymous with coal friability.

A negative exponential fit to the ARD–fuel ratio data gave an r^2 value of 0.90 with the high fuel ratio coals yielding dust levels lower than those of the low fuel ratio coals by more than an order of magnitude. This trend is in agreement with the previous results obtained by Hanson and Roepke[3].

There are several notable points to be made as to the significance of the fuel ratio with regard to future application of this data to mining. First, it is important to note that the fuel ratio is independent of the manner in which the fixed carbon and volatile matter are determined. For example, both could be determined on a dry, mineral matter free basis or on an as-determined basis. In other words, the fuel ratio is a constant for a given coal sample since it is an absolute measure of the total combustibles within the coal. This is particularly

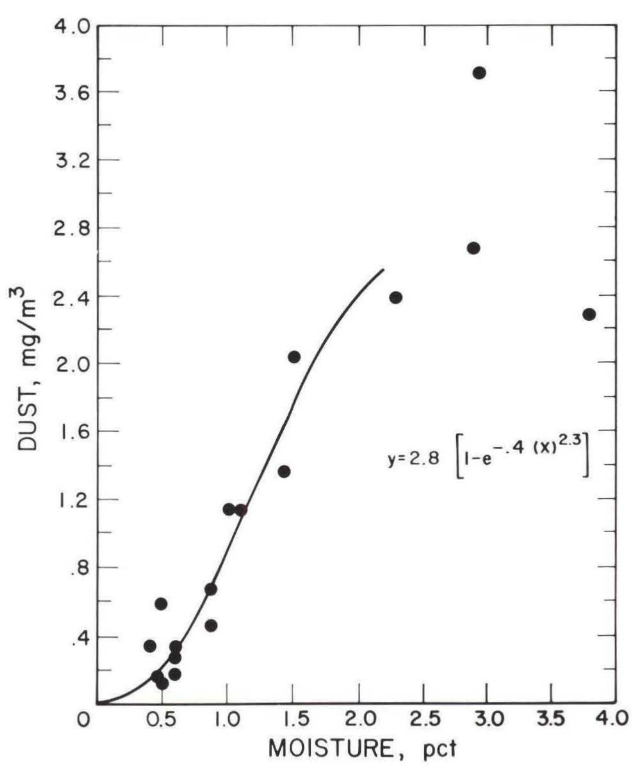

Figure 2 Airborne respirable dust concentration versus inherent moisture content with exponential fit

true over the subbituminous/bituminous range of samples used for this study.

Second, specifying the fixed carbon and volatile content values on a dry basis automatically accounts for the ash content of the sample. The utility of the fuel ratio becomes apparent when the question is asked, 'If fixed carbon, volatile content and moisture all correlate with ARD, which coal characteristic is most appropriate to use?' The use of the fuel ratio on a dry basis eliminates the need to specifically relate ash content to dust. Any changes in fixed carbon or volatile matter necessarily result in a change in the ash value.

In application to respirable dust concerns in mining, it is noted that virtually all mines use proximate analyses for coal quality determination. However, they do not all use the same analysis basis (dry, as-determined, etc.). Therefore, although there appears to be a reasonable correlation of dust with fixed carbon alone, the parameters of the correlation will change as the basis of analysis changes. Because of the constancy of the fuel ratio regardless of analysis basis, mine personnel can more easily draw meaningful conclusions concerning their particular coal.

Ratio of fuel ratio/moisture

In 1903, Collier[10] used the ratio of moisture to fuel ratio to classify Alaskan lignites. The reasoning behind this classification was based on the desire to account for moisture in low rank coals. In similar fashion, the reciprocal of Collier's ratio was used to plot the present data with quite interesting results. *Figure 3* shows the plot of respirable dust versus the 'moist' fuel ratio. If the exponential model were valid, it would be expected that the data would be linear on a log–linear plot. On such a plot, the deviation beyond a moist fuel ratio of 4 is

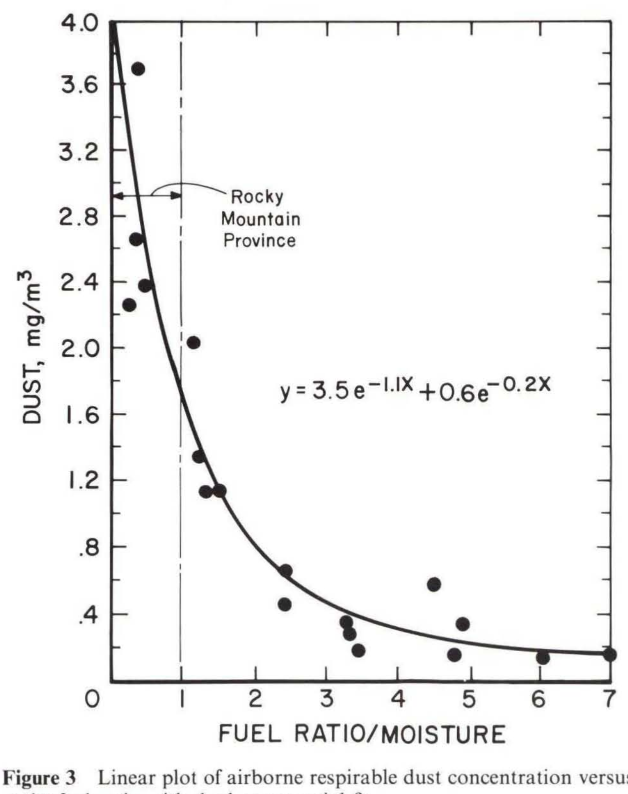

Figure 3 Linear plot of airborne respirable dust concentration versus moist fuel ratio with dual exponential fit

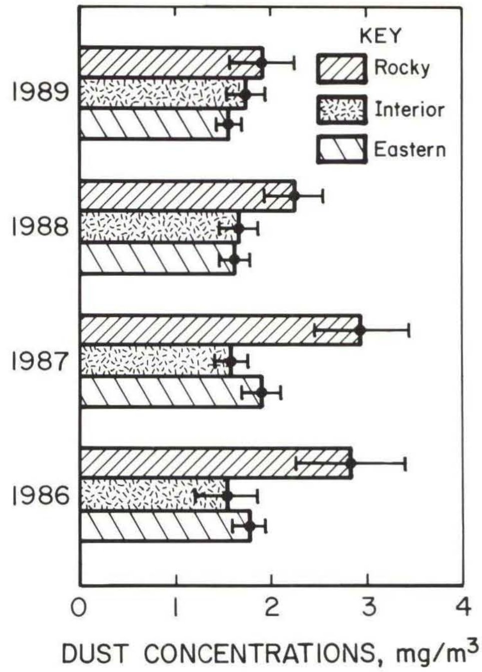

Figure 4 Average dust compliance concentrations of the coal provinces from which coal samples were analysed

suggestive of a second exponential which has a smaller decay constant and coefficient. The two-exponential model accounts for the deviation and provided an r^2 value of 0.94.

Figure 3 shows the samples from the Rocky Mountain Province to be in a group of the lowest moist fuel ratio and of the highest dust make. Comparison with *Figure 4* shows that indeed the Rocky Mountain Province has the highest dust compliance concentrations. Coal mines are required by law to maintain personal dust exposures at or below a 2.0 mg m^{-3} average concentration. Compliance samples are those required by the Mine Safety and Health Administration (MSHA) to determine the actual personal exposure. *Figure 3* is in agreement with the previous study of underground longwall mines[1] which concluded that Western high volatile bituminous coals tend to have higher ARD levels when mining.

It has been suggested that many of the data, and in particular those of *Figures 2* and *3*, appear to follow an exponential decay model. A graphically similar function is the inverse function $1/x$. Plotting the data of *Figure 3* on log–log scales clearly shows that the data mimic dual-exponential behaviour and not $1/x$. Having addressed the moist fuel ratio behaviour, the data of *Figure 2* can be readdressed. As stated earlier, the singular data point at moisture content of 3.8% cannot be eliminated. The question remains as to whether or not the proper functional relationship should be linear or exponential. *Figure 5* plots the ARD versus the reciprocal moist fuel ratio with an exponential fit. The similarity between *Figures 2* and *5* is striking. Having made a reasonable case for the ARD–moist fuel ratio exponential behaviour, it may be suggested that the data of *Figure 2* also exhibit exponential behaviour. In order to relate *Figures 2* and *5* more fully, *Figure 6* plots negative exponential values versus $1/x$. This plot provides evidence that the exponential effect of moisture on ARD is a reasonable possibility.

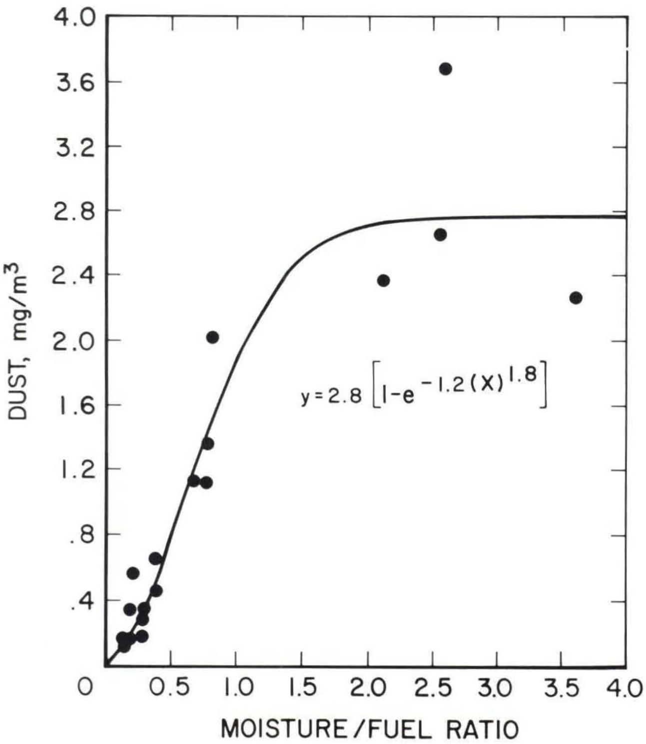

Figure 5 Linear plot of airborne respirable dust versus reciprocal moist fuel ratio

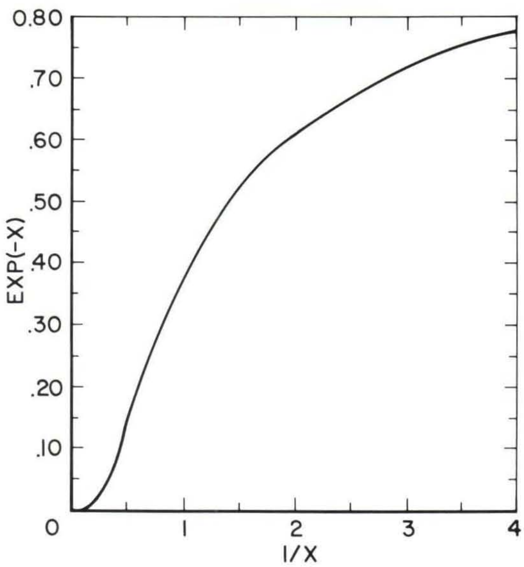

Figure 6 Theoretical plot of exponential function versus hyperbolic function

DISCUSSION

This discussion will present evidence which suggests that volatile matter is positively correlated with some weaker phase of the coal and that low volatile coals are inherently stronger than high volatile coals. By extension to the results of this study, which show that low volatile coals produce lower amounts of ARD, it can be reasoned that high strength coals produce less ARD. To accomplish this, the effect of cleating on coal strength will be examined as well as evidence available in the literature concerning solvent extraction of coals.

In general, two factors are important in characterizing the coal strength. First, is the weakness imposed on the coal by cleating. It has been shown in the literature[2] that very low volatile coals (less than 20% volatile matter, daf) have high macroscopic strength (as determined by impact strength index and compressive strength). Up to a certain point, the increasing volatile content and the increased cleating are additive in weakening the coal. However, a minimum in strength is reached at approximately 20–25% volatile matter (daf), and further increases in volatile matter show increasing strength. This increasing volatile matter is progressing towards the subbituminous and lignite coals, of which the lignites are characteristically strong coals (macroscopically).

The procedures followed for this work eliminated, to a large extent, the cleating effect on the strength of coal. Therefore, a plausible case can be presented for relating the moist fuel ratio to some measure of 'inherent strength' of the coals, exclusive of fracture weakness and cleating. This strength appears to be correlated with the fuel ratio. According to Schobert[11], coal can be considered to be a cellular solid. A property which is used to characterize cellular solids is the relative density. This relative density is the ratio of the density of the cellular substance to the density of the original solid from which it is made. The relative density of a coal can be approximated by the ratio of the apparent density in some liquid that does not penetrate the pores, to the helium density. The helium density is considered equivalent to the true density of the coal substance. It is seen that as the relative density of the cellular material increases, the cell walls thicken and the pore volume decreases. If it is accepted that inherent moisture is some representation of the pore volume, it could be said that the lower the inherent moisture, the higher the relative density. There is evidence that, for cellular solids, the strength (when strength is expressed as Young's modulus[12]) increases as relative density increases. Applying this reasoning to coals, it follows that as inherent moisture (and therefore pore volume) decreases, the strength increases.

If it is accepted for purposes of this discussion that coals are size-reduced (disregarding cleat structure) by cell wall breakage, then a factor other than pore volume is required to estimate the strength of the cell wall material. This is, in effect, some measure of the inherent strength of the carbonaceous substance itself. There is some concurrence that coals can be considered as three-dimensionally cross-linked polymers containing various amounts of plasticizers. By definition, a plasticizer is a substance added to or a part of the polymer which imparts flexibility or distensibility. If in a crude sense the fixed carbon represents the cross-linked polymer and the volatile matter represents the plasticizer (a structurally weaker phase of the coal), then decreasing the amount of volatile matter (the plasticizer) results in decreasing flexibility and hence, increasing strength. If a given inherent moisture (pore volume) is assumed, then strength will increase directly as the ratio of fixed carbon to volatile matter (fuel ratio) increases.

To examine whether or not volatile matter correlates with a structurally weak phase in coal at ambient temperatures, consider the evidence available in the literature concerning solvent extraction of coals. Two avenues will be examined. First, consider the solvents that extract coal on a relatively non-reactive basis. This class of solvents would significantly disrupt hydrogen bonds in coals but not covalent bonds to any extent. Second, consider the solvents that are very reactive with coal and may produce colloidal extracts consisting of large coal molecules cleaved from the parent coal. Herod *et al.*[13] present fuel analyses on 16 fuels ranging from peat to anthracite. The 14 coals excluding peat and anthracite range from approximately 65 to 88% total carbon (daf) and 35 to 48% volatile matter (daf). Linear regression analysis of the data shows that volatile matter is inversely related to total carbon. Dormanns and van Krevelen[14] and Wynne-Jones *et al.*[15] present data on the extraction yield of pyridine versus rank of the parent coal at 115°C. In general, the extraction yield is positively correlated with rank up to approximately 87% carbon (daf). Combining these two sources of information, it follows that, for pyridine, extraction yield is negatively correlated with increasing volatile matter. Pyridine may be classed as a mildly reactive, although good, solvent of coals up to 87% carbon (daf). Indeed, Iino *et al.*[16] also present direct evidence of this negative correlation. Iino *et al.* extracted coals with a solvent mixture of CS_2–*N*-methyl-2-pyrrolidinone (NMP) at room temperature. Their results also showed that in addition to excellent solubilities, there was little retention of CS_2–NMP in the coals, suggesting that the solvent did not react significantly with the coals. This solvent would also be classed as mildly reactive like pyridine. Both of these solvents generate a negative correlation of extraction yield with increasing volatile matter.

It is very important to consider the reactivity of the solvent and the extraction yield when comparing to volatile matter of coals. The ASTM standard test for volatile matter requires heating the sample to 950°C. At these temperatures, significant disruption of hydrogen bonds as well as covalent bonds in coals is known to occur. It is this disruption of covalent bonds which can produce significant chemical changes in the coal. It makes sense that in order to look for relations between solvent extract and volatile matter, the proper solvents should be examined. The reason is simply that devolatilizing coals is a very reactive process. Solvents that are not very reactive, hence not producing significant disruption of covalent bonds, are not appropriate for this comparison. Pyridine and CS_2–NMP are of this type. Interestingly, van Krevelen[17] presents a graphical illustration of extraction yield versus per cent carbon for five solvents: pyridine, ethylenediamine, benzylamine, diethylenetriamine and monoethanolamine. Only pyridine shows a positive correlation of yield with carbon (up to 88%) and, therefore, a negative correlation with increasing volatile matter. The other four solvents are quite reactive, especially ethylenediamine. Larsen and Green[18] state that there is evidence that ethylenediamine cleaves bonds in coal, suggesting a highly reactive nature. These highly reactive solvents are more representative of the chemical effects of the ASTM test for volatile matter on the coal than non-reactive solvents and give solvent extract yields which are positively correlated with volatile matter. This suggests that volatile matter is positively correlated with some weaker phase of the coal.

CONCLUSIONS

Given the model of coal as a cellular solid with inherent strength determined inversely by pore volume (inherent moisture) and directly by cell wall material strength (fuel ratio), it follows that for differing coals there is some macroscopic strength value which varies directly with the moist fuel ratio. It has been reasoned that high inherent strength coals are typically low volatile and low moisture (disregarding cleating effects) coals. In view of the data of this study, these coals would produce less dust than low inherent strength coals during a breaking process as used in this work. It should be stressed that this may not be the case for size reduction processes which are more closely related to actual pulverization than to one-, two-, or three-pass breakage.

REFERENCES

1 Organiscak, J. A., Page, S. J. and Jankowski, R. A. *US Bur. Mines* RI 9425, 1992
2 Pomeroy, C. D. and Foote, P. *Coll. Eng.* 1960, **37**, 146
3 Hanson, B. D. and Roepke, W. W. *US Bur. Mines* RI 8395, 1979
4 Allen, R. M. and VanderSande, J. B. *Fuel* 1984, **63**, 24
5 Casagrande, D. J. In 'Coal and Coal-bearing Strata: Recent Advances' (Ed. A. C. Scott), Geological Society Special Publication No. 32, Blackwell Scientific Publications, Oxford, 1987, pp. 87–105
6 Spears, D. A. In 'Coal and Coal-bearing Strata: Recent Advances' (Ed. A. C. Scott), Geological Society Special Publication No. 32, Blackwell Scientific Publications, Oxford, 1987, pp. 171–185
7 Yancey, H. F. and Fraser, T. *US Bur. Mines Bull.* 1929, No. 300
8 Chapman, W. R. and Mott, R. A. 'The Cleaning of Coal', Chapman and Hall, London, 1928
9 Lawall, C. E. and Holland, C. T. *Trans. AIMME, Coal Div.* 1932, **101**, 106
10 Collier, A. J. *US Geol. Surv., Bull.* 1903, **218**, 71
11 Schobert, H. personal communication, 1992
12 Gibson, L. J. and Ashby, M. F. 'Cellular Solids', Pergamon Press, Oxford, 1988
13 Herod, A. A., Stokes, B. J. and Radeck, D. *Fuel* 1991, **70**, 329
14 Dormanns, H. N. M. and van Krevelen, D. W. *Fuel* 1960, **39**, 273
15 Wynne-Jones, W. F. K., Blayden, H. E. and Shaw, F. *Brennst. Chem.* 1952, **33**, 201
16 Iino, M., Takanohashi, T., Ohsuga, H. and Toda, K. *Fuel* 1988, **67**, 1639
17 van Krevelen, D. W. 'Coal', Elsevier, Amsterdam, 1981, p. 192
18 Larsen, J. W. and Green, T. K. Dept. of Energy report DE-FG22-80PC30224-5, March 1984