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THE INFLUENCE OF COAL STRUCTURE ON HANDLING AND PROCESSING CHARACTERISTICS

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# THE INFLUENCE OF COAL STRUCTURE ON HANDLING AND PROCESSING CHARACTERISTICS

by

#### R.J. Mikula\*, M.W. Mikhail\*\* and V.A. Munoz\*\*\*

#### ABSTRACT

Laboratory studies were carried out to evaluate the influence of structure on coal surface oxidation and coke strength. Pore size distribution and the surface area of different coals were measured by mercury porosimetry and nitrogen adsorption. The rate of gas release from coal determined from the  $\Delta P$  index was used to measure the degree of fissuration and its relation to the degree of oxidation and the coking quality of various coals. The total sorption capacity of coal for methane gas, as measured by the gravimetric method, was used to evaluate coal surface characteristics by correlating the sorption capacity with the degree of surface oxidation. The  $\Delta P$  index has been correlated with natural levels of oxidation (on samples of different depths from an out-cropping coal seam) and with artificially oxidized samples.

Natural and laboratory (artificial) oxidation of coal samples produced a decrease in surface area and  $\Delta P$  index with increasing degree of oxidation. The decrease in surface area is possibly due to the sealing of micro-pores as a result of increasing oxidation, up to a limit, after which little change occurs in surface area. The gradual decrease in  $\Delta P$  values with increasing oxidation reflects changes in the characteristics of the micro-fissures rather than the sealing of micro-pores. The degree of oxidation alters the internal surface characteristics and changes the coal structure's affinity for methane sorption or desorption. The relationship between oxidation,  $\Delta P$  index and total sorption capacity can be of value in characterizing degree of oxidation and further work is underway to confirm these relationships.

A linear relationship between coke fissuration as measured by  $\Delta P$ and its stability and hardness is reported and is similar to a relationship between coal friability and  $\Delta P$  values observed earlier.

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# L'INFLUENCE DE LA STRUCTURE DU CHARBON SUR LES CARACTÉRISTIQUES DE MANUTENTION ET DE TRAITEMENT

par

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### RÉSUMÉ

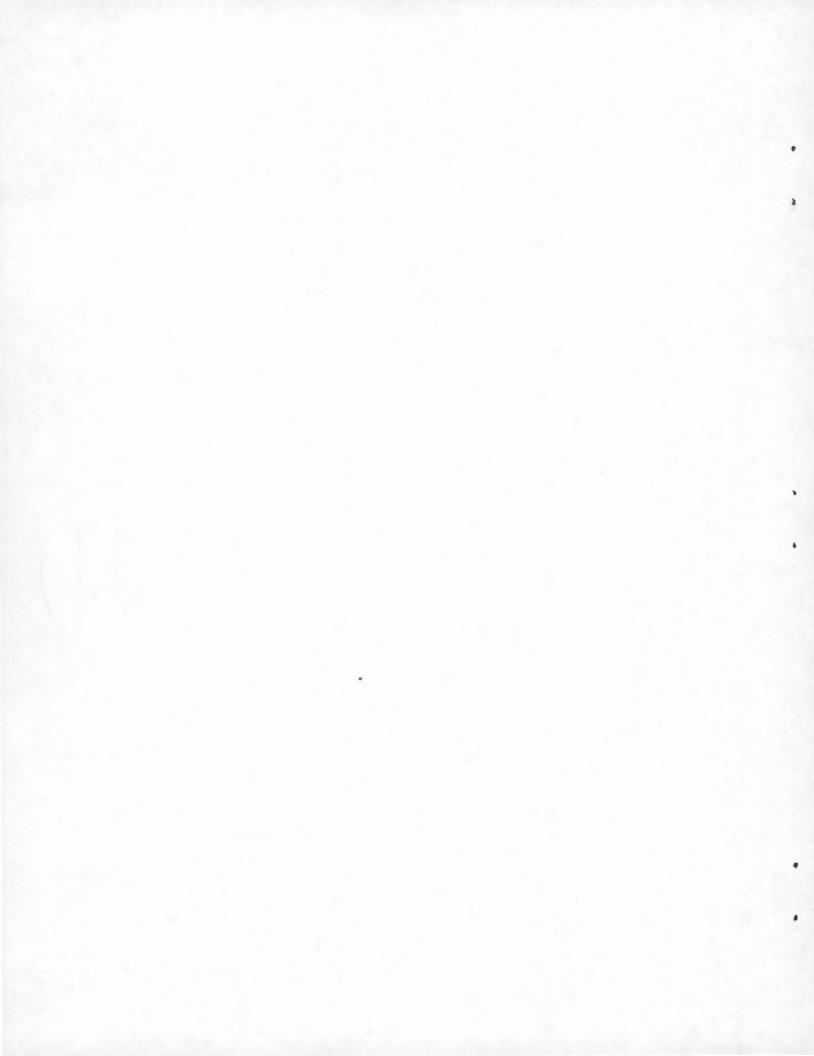
On a mené des études en laboratoire dans le but d'évaluer l'influence de la structure du charbon sur l'oxydation de la surface du charbon et la résistance du coke. La distribution de la grandeur des pores et la surface de différents charbons ont été mesurées au moyen de porosimétrie au mercure et d'adsorption d'azote. La vitesse de libération du gaz libéré par le charbon et déterminé par l'indice  $\Delta P$ , a été utilisée pour mesurer le degré de fissuration et son rapport avec le degré d'oxydation et la qualité de cokéfaction de divers charbons. On a utilisé la capacité totale d'absorption du charbon pour le gaz de méthane, mesurée par la méthode gravimétrique, pour évaluer les caractéristiques de la surface du charbon en faisant la corrélation entre la capacité d'absorption et le degré d'oxydation de la surface. On a mis en corrélation l'indice  $\Delta P$  et les niveaux naturels d'oxydation (dans le cas d'échantillons de différents profondeurs provenant d'une veine de charbon en affleurement) ainsi que des échantillons oxydés artificiellement.

L'oxydation naturelle et en laboratoire (artificielle) a produit une diminution dans la région de la surface et un indice AP possédant un degré croissant d'oxydation. La diminution dans la région de la surface est attribuée possiblement au scellage de micro-pores, en résultat de l'oxydation croissante, jusqu'à un certain point; après quoi pratiquement aucun changement ne se produit dans la région de la surface. La réduction graduelle des valeurs AP avec l'oxydation croissante reflète les changements des caracté-

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ristiques des micro-fissures plutôt que le scellage des micro-pores. Le degré d'oxydation altère les caractéristiques de la surface interne et change l'affinité de la structure du charbon pour l'absorption ou la désorption du méthane. Le rapport entre l'oxydation, l'indice AP et la capacité totale d'absorption peut s'avérer valable pour la caractérisation du degré d'oxydation; des travaux se poursuivant visant à confirmer ces rapports.

On rapporte un rapport linéaire entre la fissuration du coke telle que mesurée avec l'indice AP et sa stabilité et sa dureté. Ce rapport est semblable à une relation entre la friabilité du charbon et les valeurs AP observées antérieurement.



#### INTRODUCTION

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The structure of coal includes pores and fissures of various sizes which can influence many of its physical, mechanical and chemical properties such as friability, gas sorption, moisture capacity, oxidation and coking properties. All of these may influence coal properties important in mining, handling, beneficiation and processing. The degree of oxidation of weathered coal is related to the number of fissures and pores which present a larger coal surface to the atmosphere. Oxidation reduces the amenability of fine coal to upgrading by froth flotation and its usefulness for coking. The pore size distribution and degree of fissuration can influence coal reactions to external loads and chemical reagents. These factors influence coal behaviour as a sorbent or filtering medium and thus can have an important effect on the dewatering characteristics of a given coal. In utilization, this structure affects comminution characteristics of the coal, dust generation, coking characteristics and conversion (gasification and liquification) properties.

Pores are thought to be of natural origin and to be unaffected in size by stresses produced during mining. They vary in size from less than 0.01 to 100µm and are present in a wide range of coals with porosities varying from 4.1% in bituminous coals up to 23.2% in low rank coals and anthracite (1). While pores are generally considered to govern adsorption capacity, fissures can close under stress and thus they are assumed to control the gas permeability of coal. Fissures can be natural or occur later during mining, handling and processing. They range in size from less than 1 to 100µm or more. The redistribution of stresses during mining operations often leads to fracturing and fissuring. These man-made fissures along with natural fissures control the rate of release of gas in the mine atmosphere. Mining methods, types of explosives and mechanization can cause changes in the degree of fissuration that influence the generation of fines. Particle size degradation during handling and beneficiation are thus affected by the degree of fissuration that exists in the coal structure.

This paper discusses experimental techniques that are used in our laboratory to characterize coal structure and its relationship to coal

oxidation. These techniques include the index of speed of desorption ( $\Delta P$ ), total sorption capacity, B.E.T. surface area, and mercury porosimetry. Preliminary results from these different methods are correlated with characteristics of coal and coke to help establish relationships useful for predicting coal behaviour during handling, processing and utilization.

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#### **AP INDEX MEASUREMENT**

As part of its structure coal contains pores and fissures of a few Angstroms to several millimeters in size. Pores function as gas reservoirs whereas fissures act as transport arteries. The initial rate of desorption of gas from coal is very great and depends on the degree of fissuration. As the gas continues to flow out from the very fine pores  $(0.01\mu m)$  and fissures the rate of desorption decreases and the emission follows the laws of diffusion. Khodot et al (2) showed that coal friability is associated with the presence of a large number of fissures

Sum in thickness. A useful indication of fissuration is the  $\Delta P$  index, a measure of the initial rate of desorption which depends on micro-fissuration and perhaps also on the size and distribution of pores in the coal (3). The  $\Delta P$  index was developed by Ettinger, modified slightly by CERCHAR (Centre d'etudes et recherches des charbonnages de France) and has been used in France, Belgium and other countries to obtain a measure of the rate of desorption of methane from coal and as an indirect means of classifying coal structure (4). The AP index apparatus used in the present study was built to the same dimensions as the one designed and used by CERCHAR to characterize coals for susceptibility to gas outbursts. Their experimental work showed that  $\Delta P$  indices of various sizes of fragments stay nearly constant as long as the size of the coal particles is greater than the distance between fissures. The  $\Delta P$  index increases if particle sizes are less than this distance because comminution develops more new surfaces than were originally available; i.e., the particle size for which the  $\Delta P$ index begins to increase abruptly corresponds closely to the spacing of the fissures.

The  $\Delta P$  indices are empirical values which depend on the test being carried out in accordance with standard procedures and using

equipment of standard size. The apparatus and technique used have been described in detail elsewhere (3). As such, the  $\Delta P$  measurement provides a means of comparing the rates at which gas can be released from different coals. The movement of gas out of coal particles probably occurs as a two-stage process, the gas first desorbing and diffusing through the fine pore structure and then flowing more rapidly through any fissures present (5).

#### TOTAL SORPTION CAPACITY

The process of taking up a gas by a solid is known as sorption, a term which is used to include both adsorption and absorption. Adsorption describes the existence of a higher concentration of a particular substance at the surface of a solid than is present in the bulk of the solid; absorption describes a more or less uniform penetration of a given substance into the molecular structure of the solid.

Coal is readily able to sorb gases including carbon dioxide, methane and nitrogen. The amount of sorbed gas is proportional to the area of the surface of contact of the coal and is affected by many variables such as the nature of the gas, the nature of the coal, the presence of moisture and the temperature of the sample. The sorption properties of coals depend mainly on their fine structure (pores and fissures) and surface characteristics. This structure in turn is related to the degree of maturation and determines the filtering properties of the coal (1).

The sorption capacity was measured using a gravimetric determination apparatus built to similar specifications as the one developed by Daines (6) and which can be used for measurement at different pressures and temperatures. For the present study, the sorption capacities were measured at 30°C and 4.22 MPa (600 psig).

### POROSIMETRY AND B.E.T. SURFACE AREA MEASUREMENT

The porosity measurements of the various coal samples were done on a mercury intrusion porosimeter (Micrometerics model 9300). Each measurement was equilibrated for 20 s and in all cases the maximum pressure

reached was 200 MPa. This was done to avoid the corrections generally required when using this technique on compressible solids such as coal (7,8). Since this was a comparative study, these corrections could be neglected. However, work is continuing to obtain absolute values for pore size distribution and surface area of these samples taking into account the compressible nature of the coal.

Similar considerations arise when using nitrogen adsorption to measure B.E.T. surface areas. For this study both nitrogen and krypton adsorption (at liquid nitrogen temperature) was used to measure the relative surface areas of oxidized and unoxidized coal samples. The surface areas measured with N<sub>2</sub> and Kr were in very good agreement and only the Kr data is reported. It is well known that for reasons related to the pore size distribution in coal, B.E.T. surface areas measured by  $CO_2$ adsorption are many times larger than those obtained using N<sub>2</sub> (9). For this preliminary study however, it was the relative and not the absolute surface areas that were considered important.

#### DISCUSSION OF RESULTS

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Figure 1 shows an increase in surface area of the coal with an increase in the sampling depth from an outcropping seam (coal A). Coal obtained nearer to the surface and known to be oxidized showed a smaller surface area than the less oxidized samples taken deeper in the seam. This may occur because of the higher pressures deeper in the seam which can increase the degree of fracturing and fissuration thus resulting in a . larger surface area for the deeper coal. Surface area measurements on coal B which was oxidized under controlled conditions also showed a decrease with incrasing degree of oxidation (Fig. 2). The results showed some evidence suggesting that the increase in surface area with seam depth for the natural sample could have been due to a combination of a change in degree of oxidation and increased micro-fissuration due to tectonic movements rather than to just oxidation alone. This indirect evidence is given in Fig. 2 where the B.E.T. surface area for the laboratory oxidized coal show a decrease, then a levelling off whereas the surface area for the coal from the seam shows a more continuous decrease. If the change in

surface area of the coal (as a function of depth in the seam) were due to oxidation only then we might expect a similar levelling off as shown by the laboratory sample. Figure 2 shows decreases in surface area,  $\Delta P$  index and total sorption capacity as oxidation increases. The plots indicate that surface area drops off quite rapidly then levels off while the  $\Delta P$  index continues to decrease with rise in oxidation. The initial sharp drop in surface area could be explained as being the result of sealing of micro-pores since these make the largest contribution (per volume) to the surface area. The  $\Delta P$  index, on the other hand, is a measure of the rate of gas release as related to the degree of micro-fissuration rather than to micro-porosity. The fissures close or change only gradually during oxidation which could explain the slower but steadier decrease in  $\Delta P$ . The change in the larger pores and fissures would lead to a much smaller change in the total surface area. The sorption capacity is probably more sensitive to the chemical properties of the coal surface along with the accessibility of the micro-pores. The degree of oxidation alters the internal surface characteristics of the coal and changes it s affinity for the sorption of methane.

The relationship between micro-fissuration as measure by  $\Delta P$  and oxidation was evaluated for both the natural (coal A) and the artificially oxidized (coal B) samples. Figures 3 and 4 show that in both cases the  $\Delta P$ values decreased with increasing oxidation. This could have been due to oxidation causing the closing of fine pores and fissures that make up the micro-structure of coal. Figure 5 shows that the correspondence between the degree of oxidation (oxidation time) and  $\Delta P$  index is linear when the oxidation time is plotted on a log scale. Since oxygen was in a large excess, this may be just an indication of the sample pseudo-first-order reaction kinetics of the coal oxidation process.

The relationships between coke fissuration and its stability and hardness are shown in Fig. 6. These preliminary results indicate a linear relationship between the degree of fissuration as measured by  $\Delta P$  index and the stability and hardness of the coke. This is similar to a correlation found between coal degradation and micro-fissuration described previously (3).

### CONCLUSIONS

The types of correlation suggested by these preliminary data could be significant for characterizing the degree of oxidation of a coal seam. If the oxidation characteristics of a coal deposit can be determined at an early stage of development of a property, the information will help determine how the coal is ultimately to be beneficiated and utilized. Further work is needed and is underway to explain and confirm the relationships between  $\Delta P$ , sorption capacity, surface area and degree of oxidation.

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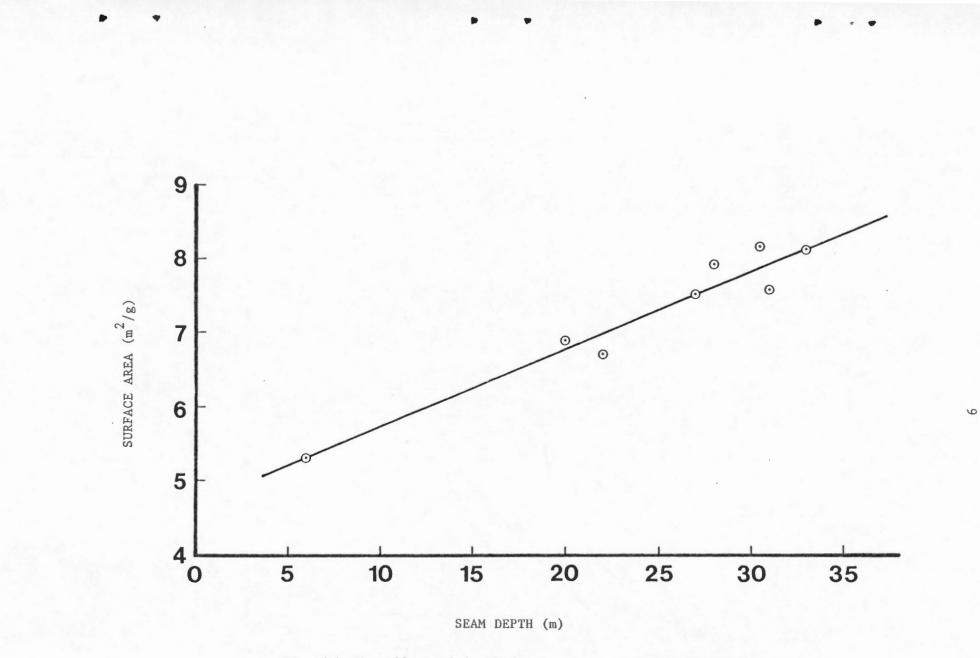
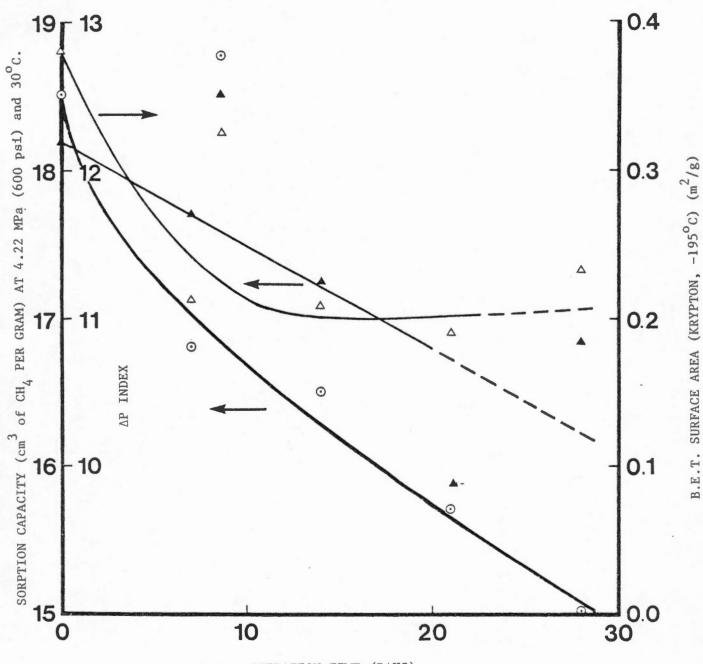
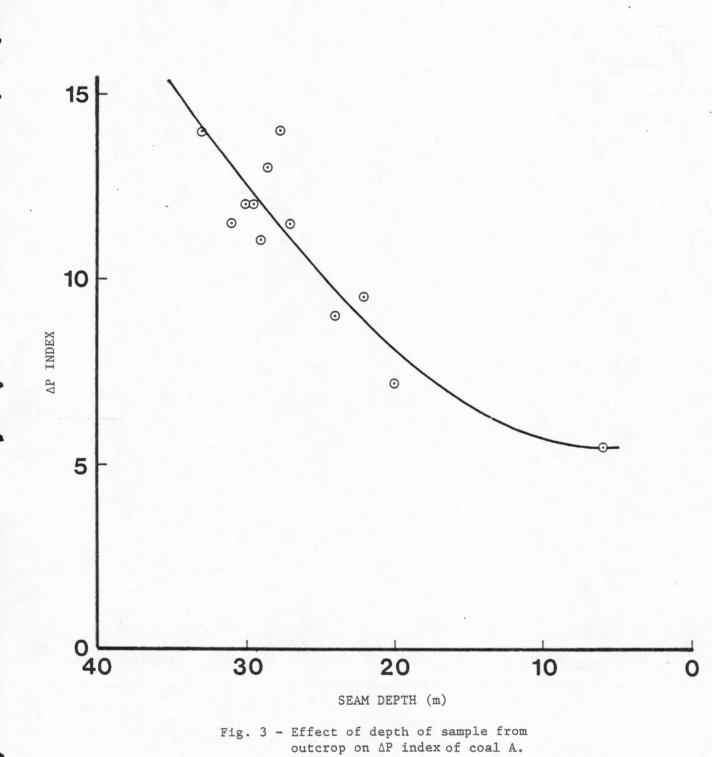


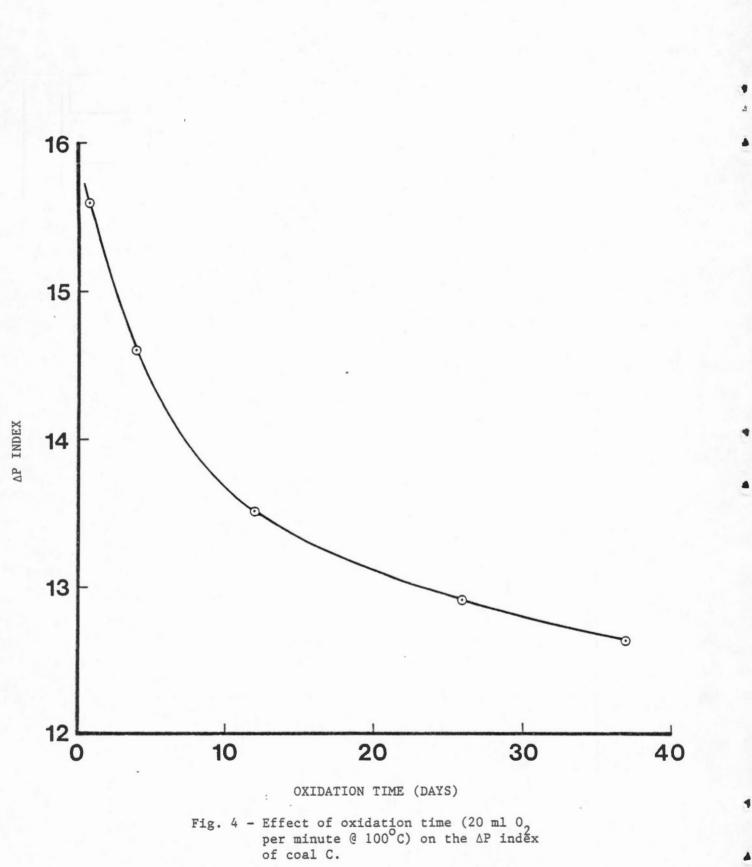
Fig. 1 '- The effect of depth from outcrop on the surface area of coal A (as measured by mercury porosimetry at 200 MPa).



OXIDATION TIME (DAYS)

Fig. 2 - The effect of oxidation time (45 ml 0 per minute @ 100°C) sorption capacity and B.E.T. surface area of coal B.





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