THE EFFECT OF SURFACE PROPERTIES OF FINE COAL ON THE BULK COAL HANDLEABILITY

by

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ABSTRACT

Handleability of the coal, which describes the ability of coal to pass through the handling system without causing major obstructions, has been studied for over 50 years. In the extensive research projects, a number of factors have been found to influence coal handleability. Among them the amount of fines (-0.5 mm) and moisture content were shown to be the most critical. Ash content in coal, size distribution of the bulk sample (spectrum of sizes), bulk density and content of clay materials were also found important. All these factors related only some physical properties of coal samples to the behavior of coal particles in the presence of water. None of the studies addressed the effect of surface properties of fine coal in spite of the fact that the behavior of fine particles in the presence of water sufficient of the set particles.

In this project, the effect of surface properties of coal on its handleability was studied with the emphasis on coal wettability. Characterization of surface properties, in terms of specific surface area and porosity, was carried out to supplement coal wettability studies. Furthermore, pelletization was used as a method to test behavior of particles in presence of water, as this process involves movement of particles and to some extent imitates possible handling conditions for coal samples. Moreover, the ability of fine particles to aggregate was found to have a deteriorating effect on the handleability of the bulk coal. The pellet's strength was used as a measure of interparticle forces. The wettability-aggregation model was proposed to link the wettability characteristics with the ability of particles to aggregate.

It was found that the strength of pellets produced from hydrophobic coals is only dependant on the ash content, while the strength of the pellets made of hydrophilic coal particles is related to the porosity, microporosity, total surface area and wettability of these particles. The more hydrophilic particles are, the easier they are to pelletize. Therefore, easy to pelletize fines tend to aggregate and thus become more difficult to handle.

The relationship between the pelletization behavior of coal fines and the bulk coal handleability was examined using Durham Cone method. It was found that the effect of wettability of fine coal on the bulk sample handleability was significant. For hydrophobic coals, only the mineral matter affects the handleability of these coals; the flow rates

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deteriorate in presence of high amount of clays. An increase in moisture content affects flow rates only to a certain level, and even at high moisture contents, these coals do not cease to flow. It was concluded that because the aggregation of hydrophobic coal particles does not strongly affect the flow, the bridges that are built by these particles are breakable, even at high moisture content these samples continue to flow. This was confirmed by pelletization results of these coals. For hydrophilic coals, the pattern of flow rates change with the moisture increase was shown to be quite different; past the equilibrium moisture, the handleability of these coals drastically deteriorates, leading to non-flow conditions.

The bulk tests confirmed further that the amount of fines (-0.5 mm) is a significant factor. The critical moisture as derived in this study, turned out to be a very important parameter, indicating the moisture content at which handling characteristics of the bulk sample sharply deteriorate. The validity of this finding is confirmed by the correlation between the surface moisture and ash fraction of fines (AF_{0.50}), which was found to have best fit at the critical moisture for tested samples. The surface moisture, which is the amount of moisture in excess of equilibrium moisture, along with the ash content is the cause for particles' aggregation, as predicted from the wettability-aggregation model.

Bulk density measurements were carried out to supplement the handleability studies. It was found that for hydrophobic coals, the effect of increasing moisture on the bulk density was less pronounced than for hydrophilic coals; additionally, the range between the critical moisture and lowest bulk density moisture (LBD) was much narrower for hydrophilic coals, confirming that these coals reach non-flowing conditions much more quickly than hydrophobic ones. The fact that the moisture content at which the lowest bulk density LBD is reached coincides with the critical moisture prompted to develop a simplified procedure for predicting handleability behavior of a bulk coal from the bulk density tests.

The comparisons between the Handleability Monitor and Durham Cone method were made, providing information on how these methods are influenced by changing conditions and possibly how they can be improved. The results of testing the same coal samples using both methods have shown that the effects of wettability on coal handleability can be used to explain behavior of coals with different characteristics.

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LIST OF SYMBOLS

Α	cross-sectional area of the column
Ac	ash content in coal
A _{0.038}	ash content in 0.038 mm size fraction
A _{0.50}	ash content in 0.50 mm size fraction
A _{cs}	cross-sectional area of the adsorbate molecule
AF _{0.038}	ash fraction of the 0.038 mm size fraction
AF _{0.5}	ash fraction of the 0.5 mm size fraction
AMP	average maximum pressure
A _{tot}	ash content in the total (bulk) sample
Ash _{feed}	ash of the feed – total (bulk) sample
Ash _{0.038}	ash of the 0.038 mm size fraction
BD	bulk density
bd	bulk density
BET	Brunauer-Emmet-Teller, N ₂ adsorption method
С	BET constant
c	cohesion
Cr	parameter related to the roughness of coal particles
СМ	critical moisture
D	fractional exponent
DCI	handleability index (Durham Cone Index)
DFT	Density Functional Theory method
d_{min}/d_{max}	two dimensional aspect ratio

d _p	particle diameter
EYL	effective yield loci
F.C.	fixed carbon in coal
£	unconfined wield stress (unconfined wield strength
I _C	uncommed yield stress/ uncommed yield strength
FF	flow functions
GAI	generalized adsorption isotherm
hı	height of the settled fraction
h ₂	height of the settled plus suspended fraction
Но	height corresponding to the loosely-packed coal sample in the bulk density measurement
Hı	height corresponding to the tightly packed (after consolidation) coal sample in the bulk density measurement
HBD	highest bulk density
HGI	Hargrove grindability index
HI	handleability index (f _c /bd)
К	constant
LBD	lowest bulk density
Μ	moisture
MBV	methylene blue value
M_N	molecular weight of the N ₂
МР	microporosity method
M _w	water requirement for coal pelletization
Ν	Avogardo's number (6.023×10^{23} molecules/mol)

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NK	Neimark-Kiselev method to determine the fractal dimension
p / p ₀	the ratio of the water pressure to the saturation vapor pressure of water
P ₀ /P	N ₂ , relative pressure
Pmax	maximum pressure
Ps	pellet strength
R	rugousity
Rc	correlation coefficient
S	specific surface area
S _{BET}	surface area of pores up to 8 nm (80 Å), calculated from BET method
S _{DFT}	surface areas of mesopores 2 - 8 nm (20-80 Å), calculated from DFT method
SE	sand equivalent values
S _p	surface properties parameter
S _{ext}	external surface area of the particle
S _{sph}	surface area equivalent to the sphere surface area
St	total surface area
t	statistical thickness of the adsorbed layer
V _{liq}	volume of the liquid adsorbed
V.M.	volatile matter in coal
W	weight of gas adsorbed
W	weight
WII	the work of cohesion of the liquid
W _m	weight of the adsorbate (N_2) forming the monolayer on the surface

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$W_{n,}W_{n-1}$	sequential pairs of weights	
W _{sl}	work of adhesion of liquid to solid	
X ^D	fractal dimension	`
YL	yield loci	
γ0.038	yield of 0.038 mm size fraction	
γ0.50	yield of 0.50 mm size fraction	
δ	effective angle of internal friction	
3	bed porosity	
η	dynamic liquid viscosity	
θ	contact angle	
ν	kinematic viscosity	
ρ	density of the liquid	
σ	the normal stress acting on the sample	
σ_{c}	normal stress in a continuous flow	
τ	shear stress to initiate the flow	
τ _c	shear stress in continuous flow	
φ	the angle of internal friction	

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adhesion	attraction exerted by one phase across the interface on the other; the work of adhesion is regarded as a measure of the attraction existing between two phases
aggregation	process in which fine particles attach to each other and form aggregates, e.g. in the presence of liquid
angle of repose	the angle to the horizontal plane assumed by the free surface of the heap formed by particles at rest
arching	the flow condition usually associated with building of the strength within the mass of bulk solid in bins or hoppers; arching develops in the vicinity of the opening; only the material near the gate is able to flow out, leaving the cavity in form of the arch at the bottom of the hopper
ash	solid residue after burning coal under standardized conditions; the ash is a relative measure of the amount of mineral matter in coal
bridging	the flow condition usually associated with the arching at the bottom of the hopper, however, in bridging, the actual flow is much more restricted than in the arching
bulk coal	the sample composed of coal particles of different sizes; these particles are in contact with each other; their behavior is affected by the presence of other particles
bulk density	mass of the solid particles per unit of volume they occupy, including voids between particles
coal	organic sedimentary rock, composed of macerals intermixed with minerals
coal blend	in the context of this research refers to the clean coal product; a blend of different size fractions
coarse fractions	in the context of this research, a material coarser than 0.50 mm; according to powder mechanics, this material is a combination of granular solids (0.10 to 3 mm) and broken solid (+ 3 mm)
cohesion	in the context of the granular material; is the attraction of similar particles to each other

coned-and- -quartered method	the practical method for splitting and mixing bulk coal sample
contact angle	angle formed by a drop of liquid resting on the surface of a solid (measured through the liquid)
critical moisture	the moisture content at which deterioration of flowing properties of the bulk sample is observed
fines	in the context of this research, the material below 0.50 mm size; this is consistent with the coal preparation terminology; according to powder science terminology, this type of material is classified as mixture of powder material (0 up to 0.1 mm) and granular solid (0.1 to 3 mm)
flowability	ability of coal to flow freely through the handling system; the term flowability is usually related to the gravity flow through the handling system
fractal dimension	the topography of the real surface, "roughness exponent"; known as fractal dimension
handleability	the ability of coal to pass through the handling system without causing obstruction
handling of coal	operations of transferring the coal product or sample by mechanical means; usually operations involving transport from one place to the another, e.g. by trains; or transfers within the distribution system, e.g. hoppers or during the processing of coal from one coal preparation circuit to the other
hydrophilic coal	coal that can be wetted by water; water spreads on the surface of coal particles
hydrophobic coal	coal that is not wetted by water
macerals	the smallest distinguishable components of organic matter, different macerals vary in physical and chemical properties
macropores	the pores with the diameters exceeding 50 nm
mesopores	the pores with the diameters between 2 to 50 nm
metallurgical coal	coal used to make coke; it has special plastic properties, metallurgical coals are hydrophobic, unless oxidized.

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micropores	the pores with the diameters not exceeding 2 nm
mineral matter	inorganic crystalline minerals associated with coal
Mohr`s circle	diagram representing the stresses' distribution in the flowing powder
oxidation	process in which oxygen adsorbs on to the surface and reacts with the coal surface resulting in physical and chemical deterioration of the coal surface
pelletization	process leading to the formation of pellets using rotating tumbling devices such as disks or drums
pellets	pellets are round-shaped aggregates produced in pelletizing devices
rank of coal	refers to the coalification degree of organic matter; chemical and physical changes are associated with the increase in rank in coals
ratholing	the flow condition associated with flowing out of the hopper or bin; formation of the tunnel type of the material discharge from the hopper
raw coal	untreated coal, usually run-of-mine coal
shear strength	the magnitude of resistance, at the point of sliding in a powder or fine solids mixture
shear stress	the force required to initiate the sliding in a powder or fine solids mixture; for maintaining a flow (sliding between the particles in the powder), the shear stress must always be greater than its shear strength developed under normal stress acting on the surface of a powder
thermal coal	coal used for power generation, thermal coals are generally oxidized metallurgical coals or lower rank coals; thermal coals are hydrophilic coals
wettability	refers to the ability of water to spread on the surface of coal; wettable-hydrophilic; non-wettable-hydrophobic

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CHAPTER 1. INTRODUCTION

Coal is the most abundant energy resource in comparison to oil or even natural gas. It is estimated that the world's coal reserves will last for 1500 years, whereas crude oil will remain as a major source of energy for the next 40 to 60 years and reserves of natural gas will be depleted in 70 to 120 years. It is expected that both population growth and industrial advancement will further intensify coal use; furthermore, it is projected that by 2020, coal could overtake crude oil as the world's most important energy source. In Canada alone, coal reserves are estimated at 8 billion tones.

Coal production in British Columbia is over 27 million tones annually, comprising over 40% of the total solid mineral production, with the value of over \$ 2 billion Canadian dollars (British Columbia Mineral Exploration Review, 2006; Price Waterhouse Coopers, 2004). All of the produced clean coal is exported outside of the province, and transported by trains across the continent for the use in eastern Canada or the United States, and to other overseas destinations by cargo ships.

As important as it is for a coal mine to produce high quality coal, whether thermal or metallurgical, it is also quite essential to produce coal characterized by a good handleability at the same time. The coal must flow by gravity in each operation of the distribution system, in order to keep many transitional operations running smoothly, and most importantly to ease the transportation process. Handleability is commonly defined as the ability of the coal to pass through a handling system without causing blockages and hold-ups (Brown, 1997).

Handling systems consist of many operations which, include transferring of coal within the coal preparation plant, from the preparation plant to mine surge bins, to the trains, from the trains into a rotary dumper and into the receiving hopper. Further on, the coal is transferred to stockpiles, from stockpiles to a reclaimer, to the terminals surge bins and finally into the ship's holds, if transported by ship. This multistage coal handling operation is especially critical for western Canadian coals, as they take three time longer to unload from trains compared to coals from elsewhere (Transport Canada Development, 1977).

The western Canadian coals exhibit poor flow (handleability) properties. Usually they have large amounts of fines and a high surface moisture at the point of load, which ' affects their handleability characteristics. They are also very friable and the content in the consignment of very fine particles continues to increase during transportation. The amount of fines in metallurgical coals is usually around 30-35 % (material below 0.5 mm). In part, the high fines content in bituminous coals produced in British Columbia is related to the rank of these coals. The medium volatile coals are characterized by the highest friability. Coals from the Rocky Mountain region of the province had been subjected to severe geological disturbance, which resulted in extensive shearing of coal seams in the region. The strongly sheared coals tend to be very friable and produce high amounts of fines during mining, handling and processing (Holuszko, 1994). In tectonically sheared coal seams of southeastern British Columbia, fine coal fractions are especially enriched in vitrinite, which is a very valuable component for coke making (Bustin et al., 1983).

A large amount of fines (-0.5 mm) in run-of-mine coal translates into high amounts of fines being processed and included in the final clean coal product. All of the coal cleaning operations are performed in water, and therefore the final coal product contains a considerable amount of moisture. High moisture content and large amounts of fines contribute to coal handleability problems.

Preparation plants produce coal as blends of various size fractions. Metallurgical coal fines are treated by flotation and fine coal products are recovered from flotation circuits by filtration. The wet filter cake commonly contains more than 20% moisture. This in combination with the rest of the blend, can result in handling problems. In the preparation of thermal coals, untreated fines are usually recombined with the cleaned coarse fractions from wet gravity separators, which also have considerable moisture content, leading to poor handling characteristics as well.

Typical problems for western Canadian coals are arching, bridging and ratholing. In addition, transported coal compacts and gains strength in rail cars and in the cargo ships. The problems with difficult to handle coals are commonly overcome by special designs and modifications of the handling system at various points to avoid or reduce the probability of blockages. In any case, blockages of chutes, transfer points, and hoppers at the discharge stations are costly and very inconvenient to the customers; the most difficult to overcome

are problems associated with transportation in the rail cars where modifications are rather limited. Although the handleability of western Canadian coals has been a major problem, only a few studies throughout the years have intended to solve this problem (Transport Canada, 1977; 1978; CANMET, 1996).

The coal handleability problems have been encountered in other coal producing countries throughout the world. As a result, a number of techniques have been developed over the years to assess the handleability; for example using a specially designed cone to measure the flow of coal referred to as the Durham Cone (Hall and Cutress, 1960; Vickers, 1982; Brown et al., 1997). Other methods focused on either measuring the shear strength of fines (Jenike, 1961; Arnold, 1992; Barois-Cazenave et al.,1999) or tensile strength of fines, as in the method developed in Poland (Polish Norms, 1982; Wawrzynkiewicz, 2003). The extrusion Through Index method that was developed by Brown at the University of Nottingham (Brown, 1997; Brown and Atkin, 2000) aimed at measuring the compressive strength of the bulk sample. Blondin et al., (1988) showed that handleability is strongly affected by the content of clays and settling properties of fine particles. They developed the handleability classification system based on these two parameters.

A number of factors have been shown to influence handleability of coal. These include: moisture, ash and fines content. In general, as the moisture content of coal increases, the handleability becomes more difficult until it reaches a point where coal is so moist that it behaves as a fluid. Similarly, with an increase in fines content, handleability deteriorates. Apparently there are two type of limits; one being the amount of fines and the other moisture content. As a result, past a certain moisture content, deterioration of the handleability intensifies, while below a certain level of fines content, the effects of moisture on bulk coal are negligible. Widening the spectrum of sizes in bulk coal improves handleability; the larger particles counteracting moisture effects (Mikka and Smitham, 1985). It has also been shown that the effect of moisture content is greater on fines (-0.5 mm) than on coarse coal (Wawrzynkiewicz, 2003). Mineral matter content has the strongest effect on handleability when clays are present because clays swell in the presence of water and glue the particles together to form aggregates. Mikka and Smitham (1985) showed that the effect of bentonite on fine coal handleability is much larger than the effect of kaolin. In

general, a low-ash coal always appears to have better handling properties than a high-ash coal.

While there is an abundance of literature on handleability relating physical factors, as discussed above, to the ability of coal to flow through different handling systems, the understanding of the basic phenomena behind the flow of wet coal is poor. The moisture tends to affect mostly fines, whilst the behavior of fine coal particles in the presence of water is dictated by their wettability, none of the research projects on handleability examined this effect.

No studies have been undertaken to examine the effect of the surface properties of fine coal particles on the handleability of the whole blend of coal, albeit the fines are predominately responsible for the deterioration of handling characteristics of coal blends. Therefore, characterization of surface properties of fines in terms of wettability was the essential aspect of this study. The characterization of other surface properties such as surface area and porosity were carried out to supplement the wettability study.

There are apparent similarities between coal handling properties and the behavior of fine coal in the pelletization process. Pelletization is the process of forming pellets by tumbling fine particles sprayed with water in devices specially designed for this purpose. Pelletization of coal particles is controlled by interfacial and capillary forces resulting from the presence of a liquid phase (Kapur and Fuerstenau , 1966; Sastry and Fuerstenau 1977; Sastry and Mehrotra, 1981; Sastry and Fuerstenau, 1982). In order for pelletization to take place, first a liquid has to wet the surface of coal particles, then liquid bridges between particles have to be formed, and only then, the capillary forces become responsible for forming bonds between particles. Mechanical forces are required to bring individual wetted particles into contact with each other, so the pellets can form. The process commonly involves tumbling in a pelletizing disk.

When the particles aggregate, they tend to stick together and flow en-mass, which results in a deterioration of the handleability of the bulk coal. In handling, fines and coarse coal are tumbled together in the presence of water, thus, ability of fine particles to aggregate becomes an important characteristic in the handleability determination.

Since pelletization depends strictly on wettability of the particles, this method was used to test the effect of coal surface properties on a tendency of coal particles to aggregate

when subjected to mechanical motion. The understanding of the behavior of fines in pelletization was also used to investigate the particles interactions in the presence of water. Pelletization has been used to pelletize iron ore concentrates for years, but only recently it has been concluded (Gustafsson and Adolfsson, 1997) that the presence of a flotation collector in the iron concentrate contributes to lowering the strength of the formed pellets.



Figure 1.1-1 Contracting capillary forces in a green pellet by an encased hydrophobic surface layer (Gustafsson and Adolfsson, 1997); reproduced by permission of Gesellschaft fur Bergbau, Metallurgie, Rohstoff- und Umwelttechnik.

This is why the iron ore industry uses reverse flotation when processing iron ores and pelletizes the flotation tailings (not the concentrate). Figure 1.1-1 shows that capillary forces in the pellet are weakened by an encased hydrophobic area in otherwise hydrophilic particles. In this thesis, the pellet strength was used as a measure of forces acting between interacting particles. This novel approach provided an understanding of the effects of coal surface properties on coal handleability. The wettability-aggregation model for hydrophobic and hydrophilic particles is also proposed, which links wettability of particles with their ability to aggregate.

CHAPTER 2. OBJECTIVES

2.1 Research objectives

The main objective of this thesis is to study the effect of coal surface properties (with the emphasis on wettability of fines) on the handleability of the bulk coal sample and to develop a practical procedure to assess handleability behavior of coal using bulk density data in conjunction with wettability characteristics of coal fines.

2.2 Research strategy

The research program included:

- 1. Characterization of coal fines:
 - Evaluation and selection of the suitable method for measuring the wettability of coal fines, and the assessment of the wettability of tested coal fines;
 - b. Studying the surface properties: surface area, porosity, particle shape, roughness and their effect on particles aggregation ability, as the aggregation of particles leads to deterioration in handling properties of the bulk coal;
 - c. Studying the mineral matter characteristics;
 - d. An assessment of the flowing behavior of coal fines using the angle of repose method.
- 2. Studying the aggregation of coal fines:
 - a. A model of aggregation for hydrophilic and hydrophobic particles is proposed and referred to as wettability-aggregation model;
 - b. Pelletization is used as a method to test fine particles ability to aggregate;
 - c. The pellet strength is used as an evidence of forces acting on these particles owed to their surface properties;
 - d. The correlation between the wettability, surface area, porosity, ash content of coal fines and pellets strength is established based on the wettability-aggregation model.
- 3. Bulk sample testing using Durham Cone and Handleability Monitor methods:

- a. Investigating the effects of moisture on the handleability of coals with different wettability characteristics;
- b. A comparison between Durham Cone and Handleability Monitor tests;
- c. Determination of bulk density to supplement the handleability results.
- 4. Development of a practical procedure to assess bulk handleability of coal.

CHAPTER 3. LITERATURE REVIEW

3.1 Coal handleability studies

Handling properties of coal are important for coal producers as well as coal users. Delays and stoppages in the coal handling systems are always inconvenient and costly. Throughout the years several methods have been developed to measure the handling properties of coal samples, and to predict their behavior in defined situations. The coal distribution system is composed of many components that are virtually independent of each other. The handling process begins at the coal preparation plant from where the coal product is being transferred to its final usage. Each component of this handling system is different in the way it handles the bulk material. For example, the handling of coal during the loading into the hoppers may be different from the handling of coal while transporting in rail cars. Measuring techniques have usually been designed to evaluate the handling (flow) properties at a particular point of the handling system, therefore resulting in different emphasis on coal behavior.

For producers, it is important that coal does not cause any problems during the loading or processing at the plant. However, many problems are caused by bad handleability of coal during its transportation and unloading at its destination. This has prompted researchers to look for more methods to address this issue.

An empirical method developed by British Coal scientists in 1950's (Cutress et al., 1960) was introduced to provide means to assess the ease of discharge of washed coals and blends from the hopper at the bottom of rail wagons. It was referred to as Durham Cone test, and offered a quick and relatively simple way to determine coal flowability. The vibrating cone was designed to imitate train movement; as a result the behavior of coal during the transport by trains could be reproduced using the Durham Cone.

Over the years the method became used as the standard test to examine handleability and was employed in many studies with varied success. A first comprehensive study on handleability of coal using Durham Cone was published by Hall and Cutress (1960) and was followed by many others (Jenike, 1961; Vickers, 1982; Arnold, 1982; Brown et al., 1997). It was also pointed out that mixing of the sample prior to the Durham

Cone test was extremely important and that any mixing involving rolling produced a balling effect and altered the flow properties of the mixture as measured by the Durham Cone.

In this test, a representative coal sample is passed through the cone and its flow rate is measured and used as a handleability index referred to as Durham Cone Index (DCI). Different sizes of cones can be used; a cone with the original outlet opening of 100 mm for the top coal size of 25 mm and sample size between 8 to 24 kg, or 150 mm for coal blends with maximum particle size of 50 mm and much larger one with the opening of 500 mm that can be used for even larger top sizes and sample size between 30-34 kg. According to the handleability assessment by Durham Cone, a blend of coal having DCI below 1.6 kg/s is identified as a difficult to handle material. The Durham Cone was specifically designed and tested to simulate behavior of coal sample in one type of railway wagons; recently new railway cars have been introduced and apparently the low 1.6 kg/s flow rate is quite acceptable for the new-design rail wagon (Brown and Miles, 2004).

Another well established method used frequently to test handleability of fine coal is Jenike's shear cell test. This method was developed to predict the flow of solids in hoppers. Information obtained from this test, such a slope angles and outlet diameters, can be used to design hoppers. This technique has been found to be more precise than the Durham Cone, however, testing procedure is lengthy and uses only small amounts of fine coal (Jenike, 1961). In this test, about 0.6 kg coal sample (top size of 1 mm) is placed in the shear cell, a load is applied, and the force needed to shear the compressed sample is measured. Figure 3.1-1 presents schematic diagram of the Jenike shear cell. From the shear test results the flow functions are plotted and the stresses inside the coal mass are calculated. The correlation between the shear stress needed to initiate the flow and corresponding normal stress acting on the surface of coal mass is referred to as yield loci (YL) and follows the Mohr-Coulomb relationship:

$$\tau = \sigma \tan \varphi + c \tag{3-1}$$

where: τ is the shear stress needed to initiate the flow, σ is the normal stress acting on the sample, ϕ is the angle of internal friction and c is the cohesion.



Figure 3.1-1 Schematic diagram of Jenike Shear Cell (Brown and Atkin, 2000); reproduced by permission of Taylor & Francis Group.

The yield loci describes the normal stress (x axis) vs. shear stress (y axis) relationship and usually is represented by a straight line intercepting the y axis at the value equivalent to the cohesion at normal stress equal to zero.

For maintaining gravity flow, the shear stress in the coal, τ_{c} , must always be larger than its shear strength developed under the normal stress acting on its surface, σ_{c} . The equation that satisfies these conditions can be described as follows:

$$\tau_{\rm c} = \sigma_{\rm c} \tan \delta \tag{3-2}$$

where: δ is the effective angle of internal friction, τ_c shear stress in continuous flow; σ_c normal stress in a continuous flow

The relationship between the shear stress required to maintain a flow and the normal stress, is referred to as effective yield loci (EYL). Constructing the Mohr's circles tangent to the EYL, defines the major and minor principal consolidation stresses. The YL and EYL are identical for cohesionless powders. When the shear stress acting tangent to the surface and the normal stress are zero, the greatest stresses that can exist in a free surface are represented by Mohr's circle. The circle is constructed in such a way that it passes through the origin and is tangent to the yield locus for the given sample and the corresponding

major principal stress is referred as unconfined stress f_c . Since the samples gain the strength with consolidation, the unconfined yield stress increases; for Columb-type of solids (the relationship between normal stress and shear stress is represented by a straight line) the value of unconfined yield stress is equal to unconfined yield strength of the particular sample.

Jenike (1964) used the flow function for the classification of bulk materials, which was described as:

$$FF = \sigma_c / f_c \tag{3-3}$$

Where: σ_c is the major principal stress acting in the bulk and f_c is the unconfined yield strength (unconfined yield stress). The flow behavior according to Jenike is classified into:

1 < FF < 2 very cohesive, non-flowing material

2 < FF < 4 cohesive, hardly flowing material

4 < FF < 10 slightly cohesive, easy flowing material

10 < FF cohesionless free-flowing material.

The major concern with the shear test was that it required well-trained personnel in order to obtain reproducible results. According to the initial procedure (Jenike, 1964), the tests are conducted with fine coal fraction and not with the bulk sample representing the whole coal blend. Car and Walker (1967) extended the scale of the shear cell up to 500 mm in diameter, which enabled researchers to measure flow properties of coal blends with the top size of up to 25 mm. Still, the data interpretation can be very complex, and the tests can not be used as a quick method for assessing handleability.

Schonlebe and Seewald (1994) examined the flow properties of narrow size fractions of coal using Jenike shear cell, and found a strong dependence of the flow characteristics on the particle size. According to their findings, the reciprocal value of the flow function 1/FF, and thus the unconfined yield strength increases exponentially and attains a state of hardly flowing material when the particles diameter is less than 0.1 mm. At the value of 1/FF=0 material is free flowing, at the other limit, the value approaches 1, which means that the unconfined yield strength is very high and the material is not capable of flowing.

The function can be described as follows:

$$1/FF = Kd_n^{Cr}$$
(3-4)

where: K is the constant, d_p is the particle diameter, C_r is a parameter related to the roughness of coal particles

Recently Munjack and Hogg (2004) studied the flow characteristics of blends of coarse (-2.36 mm) and fine (-44 μ m) coal with the use of the Jenike shear cell. Their objective was to examine the effects of addition of fines and coarse fractions on flowability as determined from the shear cell tests. They concluded that the stresses required to maintain steady flow in a consolidated fine coal appear to be relatively insensitive to mixture composition and moisture content. The addition of the coarse coal (-2.36 mm +44 μ m) to the fine fraction (-44 μ m) led to the modest increase in the frictional resistance to flow, while moisture addition reduced the friction between the particles. The moisture increase in their case-study (from 5 to 25%) was rather significant; which is probably why the observed lubricating effect is so obvious.

A measure of stresses to initiate the flow in a consolidated powder revealed that blends of the fine and coarse coal fractions tend to be more cohesive than either of the components of this mixture tested separately. This effect has been attributed to better packing in the blends, and to the fact that blending fine coal with the coarse coal at a certain critical ratio may actually deteriorate the flowability of the blend. The mixtures tested by Munjack and Hogg were prepared as follows: 100 % fines (-44 μ m), 100 % coarse (-2.36 mm to +44 μ m), and 20 % coarse and 80 % coarse. They have also shown that increased moisture enhances the cohesion between the particles as shown by the increased shear stress values used during the tests. The increase in the cohesion value, while not being very large, may indicate a combination of the opposing effects; one causing lubrication due to high moisture (25 %) and the other one increasing cohesion between particles.

This also implies that due to the bimodal size distributions, the coarse/fine blends pack to a higher bulk density than the separate components. It was concluded that the unconfined yield stress of the coarse coal and fine coal tested separately are relatively insensitive to the consolidation stress, while for the blends the unconfined yield stress increases as consolidation increases. This means that blends are more compressible especially when fines are added, leading to a deterioration in flowability. With the increase

in bulk density, particles are packed closer packed resulting in more cohesive flow. However, when the coarse material is added to fines (20 % coarse), a deterioration in flowability is observed, with bulk densities increasing slightly. It appears that in this case; particles are packed more closely than in the coarse sample alone, and this is sufficient cause for an increase in an unconfined yield stress. The authors suggested that fines in the blend are disproportionately more affected by consolidation stresses than coarse coal particles, therefore adding the small quantities of coarse coal to the mixture could improve handleability of the blend.

In summary, their results indicate that conditions which support continuous flow are less affected by the blending of coarse coal and fine coal fractions; whereas the conditions for initiating the flow or collapse of the stable obstruction such as bridge formed during the flow of bulk coal are dependant on the amounts of fines and coarse coal in the mixture. The flow characteristics are governed by friction between adjacent particles (internal friction), by attractive forces between the particles (cohesion), as well as wall friction (material related). The latter can be excluded if the tests are carried out in the same type of cell. Flow can take place when the applied stress (shear stress) is sufficient to overcome the frictional and cohesion forces. From the presented rheological data, it appears that a blend consisting 80 % of coarse material displays the highest cohesion value along with the highest angle of internal friction although it attains the lowest bulk density amongst all tested samples. Therefore an increase in the unconfined yield stress is the combined effect of both of these factors acting simultaneously, conversely, for the mixture composed of 20 % coarse and 80 % fines, cohesion is the same as for 100 % fine coal. The only reason for the increased unconfined yield stress in the sample with 20 % added coarse coal is due to larger value of angle of internal friction coupled with increased bulk density. This can imply that geometry of packing between coarse and fine particles can be an important factor.

Arnold (1990; Arnold et al., 1992; Arnold, 2004) based her work on the concept of using the ratio of unconfined yield strength (unconfined yield stress) to bulk density as a measure of handleability and defined it as handleability index (HI) for testing eastern US coals. For her research, she used a triaxal shear tester, which can measure directly the unconfined yield strength rather than using experimental values to calculate it, as in the Jenike shear test. In this tester larger samples could be evaluated and with the larger top

sizes (up to 6.35 mm) than in the Jenike shear cell. This could be considered a major advantage, because it allows examination of the effect of fines content and larger samples are more representative of the tested bulk coal.

Subsequently to the implementation of the HI, researchers developed a much simpler device to measure unconfined yield strength (Arnold, 2004). This apparatus was designed in such a way that could measure low-yield stresses and at the same time be used as a loading mechanism for sample preparation before testing. For these tests, a coal sample was consolidated in the special mold and then transferred carefully for testing as described by Arnold (2004). Then the prepared column of coal was subjected to failure by loading the weight on the top of the specimen in a controlled manner. A comparison between triaxial tester and the new tester was made by testing the same coal samples simultaneously. A regression analysis of the data yielded a slope of 0.89 with a correlation coefficient of 0.8589. The HI values obtained by both methods showed very good correlation over the tested range of coal samples.

To further validate her data (Arnold, 2004) conducted the series of tests on a pilotplant scale. Samples of the coal discharging from the actual size bin were collected and used for the HI determination with the use of triaxial shear tester. Flow rates from the industrial bin were compared to the measured HI index to determine the applicability of the HI in predicting coal flow problems. For this particular bin it was found that a HI critical value of less than 0.35 and moisture content below 5.5 % would ensure good flowability from the bin. Further industrial testing resulted in the development of coal handleability classification system for eastern US bituminous coals. In this classification ranges of HI are plotted to correlate moisture content wt % with amount of 0.5 mm size fraction. Although, this classification could be considered as a method of predicting handleability, it still shows a significant degree of inconsistency amongst the tested coals. A reason for such a wide scatter of the data may be the fact that these coals varied significantly in their quality characteristics. Figure 3.1-2 and Figure 3.1-3 show the proposed coal handleability classification by Arnold (2004).



Figure 3.1-2 Combined effects of moisture content and fines content on handleability index (HI) according to Arnold (2004);); reproduced by permission of Taylor & Francis Group.

In the work on handleability reported by Arnold (2004), the focus was on a methodology to assess handling characteristics of coals for power utilities and addressed their concerns in predicting and anticipating problems with coals being stored in bins (bunkers) or being transferred with the use of hoppers. Therefore, the shear tester appeared to be very appropriate. The parameter derived from the shear tests can be used in design of these storage vessels. In her work, Arnold derived the correlation between the moisture in the coal and critical arching diameter of the hopper. She concluded that at 10% moisture there was a significant increase in arching diameter from 0.7 m to 1.8 m for the tested coal hopper.



Figure 3.1-3 Simplified classification for handleability of eastern US bituminous coals (Arnold, 2004);); reproduced by permission of Taylor & Francis Group.

Extrusion Through Handleability Index is another parameter that was derived by Brown (1997) at the University of Nottingham. This index was further developed by testing the coal blend samples in the device designed by Brown, (Brown and Atkin, 2000) and referred to as Handleability Monitor. The monitor consists of a hopper, control panel, presentation unit, hydraulic ram and narrowing venturi extrusion section as illustrated in Figure 5.3-4 in Chapter 5. In this test, approximately 50 kg of coal sample is loaded into the hopper. With the full hopper, a hydraulic ram is operated extending and retracting three times in pre-run cycles (three cycles), while coal sample is pushed through the narrowing channel. With a good handling coal sample, coal particles do not adhere to each other and a minimal force is required to push them through the channel, resulting in minimal pressure used, while for poor-handling sample, a significant force is required to move it in the system. Pressure measurements are taken during five cycles vs. time. The pressure exerted by the ram is recorded at intervals of 0.2 seconds. For each coal sample a series of five graphs (traces) of pressure vs. time were obtained and the average values are produced. The
average value of the maximum pressure P (max) obtained from "Pmax-Time" traces is considered to be a measure of coal handleability and is referred as the Extrusion Through Handleability Index as shown in Figure 5.3-6. The value of this Handleability Index is expressed (in bars) as the Average Maximum Pressure, the term AMP is also used to describe handleability.

Brown and Atkin (2000) carried out extensive research, and tested different coal blends with this device and established classification for metallurgical and thermal coals. According to this classification, coal blends producing an AMP of less than 10 bars were the blends with high moisture contents. Due to the excessive moisture content, these blends acted more like fluids; hence their handling was classified as poor for the handling systems designed for solids. The good handling coals were the ones with the AMP values higher than 10 bars but lower than 35 bars for thermal coals and lower than 20 bars for metallurgical coals. The wider spectrum of the handleability for thermal coals results from the fact that for power generation a much wider range of coals are used, therefore, handling systems for thermal coals are more diversified. For metallurgical coals on the other hand, only a narrow range of coals are used with the more stringent handling facilities.

This new method developed at the University of Nottingham, was found to be quick and reliable for assessing coal handleability, as discussed by Brown et al. (1997) and Brown and Atkin (2000). The advantage of this method over other existing methods is that it can be used continuously, and for large samples, taking only 8 to 10 minutes to determine handling characteristics of a coal blend.

In another approach the avalanching behavior of a bulk coal was used to predict its handleability characteristics (Brown and Miles, 1996; Brown and Miles, 2004). In this study the avalanching behavior was monitored of the coal blends as they descend and formed a heap on an inclined surface. For the purpose of the study a special apparatus was constructed. It was designed in such a way that coal was fed to hopper, and then was transferred to horizontal conveyor and then advanced to the inclined avalanche ramp. As the sample was fed continuously onto the avalanche ramp, a heap was shaped and past the critical point avalanche proceeded; coal then slid down onto the recording balance. The procedure is continued until the mass of collected coal reached 6 kg. The experimental data was analyzed by producing the strange attractor plot of the avalanche weights. The strange

attractors represents the sequentially of the "time –history" of a chaotic system. In the above application, the strange attractor was plotted as the mass (the only measured variable) plotted against itself, but delayed by time interval equal to the recorded sampling time (time the avalanche occurred).





The weight map in which sequential pairs of weights W_n , W_{n-1} are plotted and linked by joining successive points with lines defines strange attractor for the tested coal sample. The patterns of behavior from these strange attractor plots were used to differentiate between good and poor handling samples. The erratic avalanching behavior was found typical for poor handling coals and resulted in the larger values and wider spread of the data points in the strange attractors plot. On the other hand, good handling coals would have patterns less scattered and more concentrated around the "nest" created from strange attractor lines.

Figure 3.1-4 illustrates plots obtained for poor and good handling coals using the avalanche-method. To further analyze such data, statistical methods for calculating mean centroid, first, second, and third order moments of a population were employed as described by Kaye (1993). These in depth analyses showed that reasonable correlation can be found between the avalanching behavior of the material and its handleability (Brown and Miles, 2004).

Although this research had been initiated in 1996 at the University of Nottingham and resumed in recent years, reported results reflect rather developing stage for this procedure. This method however, does offer several advantages over other methods, as a bulk material testing procedure. It is quick, simple, and reliable as it can be easily adjusted to the varying handling parameters (e.g. feed rate, angle of inclination for the avalanche ramp). As well as there is no limitation on the top size for the tested coal samples. It needs to be tested commercially before can be accepted as a method to assess handleability.

In Poland, yet another method for handleability assessment was developed in 1982 (PN-82/G-04544, Polish Standard). This method relies on measuring the tensile strength of a specifically formed cylinder from coal particles. The cylinder-column is formed at two different heights, and the results are plotted on a logarithmic scale correlating height of the sample with the force required to destroy the column made from that sample. According to this procedure (Wierzchowski and Wawrzynkiewicz, 2003; Wawrzynkiewicz, 2003; Wawrzynkiewicz, 2004) the handleability index is found as a number expressing the tensile strength of a theoretical coal column of 0 mm in height; it is obtained from plotting the test results as described above. The force required to destroy the theoretical column of 0 mm is extrapolated from the plot line. The key principle behind this is that the formation and consolidation of the sample under given standard conditions and then subjecting the sample to tensile loads imitates to some extent physical phenomena during transportation and unloading from trains.

The obvious constraint in this particular method is the fact that the size of particles should not exceed 14 mm and also the size of the tested samples is limited to 3 kg. It can

only be used to test fine coal. It was shown to be relatively reliable and it was tested commercially throughout the years as a measure of handleability for Polish coals.

It is known that surface moisture and fines content have a strong influence on coal handling behavior, however, this is not sufficient to explain bad handleability. According to a study done by Blondin et al. (1988), dry screening at a low aperture (6 mm) is also a good tool for assessing the handleability of wet coal fines. In their tests, coals with various ash, moisture and fines contents were used. Difficult to handle coals always plugged the screens, while easy to handle coals were easily screened without difficulties.

To evaluate the effect of clays on handleability of coals, Blondin et al. (1988) used the Methylene Blue test. From this test MBV (methylene blue value) is determined. The MBV is related both to the amount of clays and to their specific surface area. Therefore, the higher the MBV value, the worse the handling characteristics of the whole coal sample. When the results from dry screening and MBV were compared, a good correlation was found. However, there were still some discrepancies for the intermediate values of the MBV. For example, coals with similar values of MBV displayed quite different handling behaviors.

To clarify some of the discrepancies, Blondin et al. (1988) used the sand equivalent test. In this procedure, the tested coal is placed in a cylinder with water containing dispersant. After a given time, the settled fraction is related to the suspended fraction. The sand equivalent values, (SE) are calculated from the following equation:

$$SE=100 - h_1/h_2$$
 (3-5)

where:

 h_1 = height of the settled fraction

 h_2 = height of the settled plus suspended fraction

High values of the SE indicate coals easy to handle, while low values are always associated with difficult to handle coals.

The results of the sand equivalent tests for various coals were plotted on a two dimensional net combined with methylene blue values MBV for the same coals (Figure 3.1-5). The graph area can be divided into three zones: difficult to handle coals with high MBV and low sand equivalent values, intermediate zone, and easy to handle coals zone with low MBV and high sand equivalent values. Difficulty in handling these coals is due to the presence of clays. Clays are suspected to act as a glue between coal particles, and the large surface area of clays tends to retain moisture and deteriorate handling behavior of the whole coal blend. From the series of the above-mentioned tests, it became obvious that handleability must depend on coal surface properties and the composition of the mineral matter associated with it. The method proposed here showed that it was possible to correlate to some extent the handleability behavior of different coals, to mineral matter characteristics, and to the settling patterns of fine coal. Therefore, taking into account only mechanical factors, such as particle size, yield of fine fraction and moisture, apparently is not sufficient.

All of the above-discussed approaches provided ways of assessing handleability properties of various coals. Some were more successful than others in industrial applications due to the technical constrains or requirements imposed by the size of the bulk sample. The only two methods suitable for the use with bulk samples are Durham Cone and Handleability Monitor. The Durham Cone which has been used over the years with reasonable success for quick assessments during the handling procedures involving vibrations, mechanical shaking or intensive mixing. The Handleability Monitor, however, appears to be more suited for the use in situations where coal is pushed through the channels and without considerable shaking or vibrations. These two methods were chosen as the most appropriate for the bulk handleability testing in this project.

3.2 Factors influencing handleability

Factors that were shown in previous publications to influence the handleability of coal are moisture, ash, size distribution and fines content. An increase in the fines and moisture content was found to have the most significant effect on the handleability of coal. In the early study on handleability with the use of Durham Cone, Hall and Cutress, (1960) concluded that the time to empty the cone increases to a maximum as the moisture content increases, and then decreases. The maximum time to empty was the measure of handleability, as they used same-weight samples for testing. The maximum time to empty was much shorter for lower rank coals with the same fines content than for higher ranks; the maxima generally occurred at higher moisture content as the rank decreased. This suggests that lower rank coals were easier to handle over the studied moisture range (5-25 % moisture).



Figure 3.1-5 Coal handleability classification (Blondin et al., 1988); reproduced by permission of the American Institute of Mining, Metallurgical and Petroleum Engineers.

They also pointed out that when the moisture was greater than 6 %, the time to empty increased with the proportion of fines. Conversely, below 6 % moisture there was no effect of increased amount of fines on time to empty. When the fines content (-0.6 mm fraction) was less than 10 % the increase in handling difficulties with moisture increase was not significant. For example: at 10 % fines content the maximum time to empty was around 12 % moisture; at 20 % fines content the maximum time to empty moved to 17 % moisture; while at 30 % fines content was at the same 17 % moisture level, but with considerably longer time to empty. Apparently, with the addition of fines more moisture is required to affect handling properties of a particular blend. The moisture level associated with the maximum time to empty could be perceived as the critical value, beyond which handleability drastically changes as the moisture increases. For some coals past this critical moisture value, the handleability would improve, while for others will lead to non-flowing conditions (Mikka and Smitham, 1985).

The effect of moisture on coal handleability was tested by Vickers (1982). For studied coal (-25 mm in top size), it was shown that an increase in total moisture content

from 10 % to 12 %, changes the Durham Cone handleability from being "easy" to "difficult".

In the study by Mikka and Smitham (1985), the handleability behavior of some Australian coals was assessed using Durham Cone. The following was established: the size distribution was the most significant variable affecting handleability of washed coals; in coals with little fines content (-0.5 mm) handleability was found to be insensitive to moisture content, while at the high level of fines it was found to be very sensitive to moisture content. The change in size distribution, i.e. increasing the coarse fraction content, was found to improve handleability, indicating that whole size distribution has an influence on the handling behavior of a blend of coal. The effect of added fines on handleability of coal samples, as derived from Durham Cone test, was shown to be significant only above 7% moisture for the tested coals.

Regarding the effect of moisture on fines, the following conclusions were reached by Wawrzynkiewicz (2003). He tested handleability of Polish coals using a tensile tester and reported that the behavior of the -0.5 mm size fraction was greatly affected by the increasing moisture content. The fraction -3.0 + 0.5 mm was affected only slightly, while the handleability of the coarsest fraction (-14.0 +3.0 mm) was practically not influenced at all by the increase in moisture. The effect of moisture increase on the fines was always more pronounced in the samples when clays were present.

In an extensive study from the 1990's Arnold et al. (1992; Arnold, 2004) tested over 60 coal samples ranging from lignite to anthracite covering a wide range of coal properties. She used a Triaxial shear tester for her study as described in the previous section of this chapter and showed that various coal properties influence the value of handleability index (HI= f $_c$ /bd; unconfined yield strength/bulk density) as derived from her method Arnold (1990, 1992). She reported that an increase in the content of very fine particles (-44 μ m) has a greater effect on coal handleability than an increase in the moisture content.

The effect of consolidation pressure on the HI was tested on coals with varying amounts of fine fraction (-44 μ m) and moisture content (Arnold, 1990). The samples were selected in such a way that they represented different cases: low moisture-low fines; medium moisture-low fines; medium moisture-high fines. In all cases an increase in either moisture or fines content led to an increase in the magnitude of HI, however, the samples

with high fines content exhibited the greatest HI value at all consolidating pressures. This indicates that above all, the amount of very fine particles (-44 μ m) is the most significant factor. The other important conclusion drawn from this study was that for every coal tested, the unconfined yield strength and HI decreased, with increasing void ratio (worse packing). Therefore, the packing has a large effect on handling characteristics of the sample. Similar conclusions were reached in the study by Munjack and Hogg (2004), when they tested coal fines with the Jenike shear cell.

Arnold et al. (1992) reported that for samples of lignite with different ash contents, an increase in moisture content followed different handleability trends. The higher the ash content of the sample, the more pronounced was the effect of increased moisture on deteriorating handleability. The handleability in this study was correlated to the calculated Critical Arching Diameter derived from the tests with Triaxial shear tester as discussed in the previous section. For more difficult to handle coals, wider critical arching diameter is required. In the same study, coals of different origin were tested, and a correlation between surface moisture and critical arching diameter was obtained. It was found that with increasing surface moisture content, the critical arching diameter increases; higher moisture content also increased the scatter of the data.

For the western Canadian coals it was shown that handleability was not a problem provided that moisture was kept below 7-8 % and the amount of -0.6 mm size fraction was minimized as reported by CANMET (1996). They also concluded that improvement in handleability could be achieved by increasing the particle size of the coal by pelletization or agglomeration. The excessive amount of fines in the product coal appears to be the greatest disadvantage for these coals.

Coal is a mixture of organic carbonaceous matter associated with inorganic crystalline minerals. The most common minerals associated with coal are clays, on average they account for 60-80 % of total mineral matter in coal. Next the most abundant type are carbonates, quartz and sulfides. Among the sulfides, pyrite is the most common mineral found in coal. These minerals in coal can be present in different forms depending on their origin, they can either be deposited in cavities of coal macerals, in cracks or fissures (Gaudin, 1957; Klassen, 1966; O'Gorman, 1971) or intergrown with the coals organic

matter. The intergrowth of mineral matter with coal, influences the physical properties of coal particles.

The minerals associated with coal have significantly different physical and chemical properties than coal. The physical properties such as specific gravity, for example are taken advantage of in coal preparation to separate these minerals from coal. Since most of the minerals in coal are hydrophilic, flotation can be used successfully to recover hydrophobic coal particles from hydrophilic mineral matter associated with coal.

Mineral matter (ash) content has a pronounced effect on coal handleability, especially when clays are present. It has been observed that low ash coals have better handling properties than high-ash coals. In the study by Bennett et al. (1987) as described by Arnold et al. (1992), it was shown that the clay content estimated by measuring percentage of silica in coal correlated well with the shear test results. In addition to the silica content, the surface moisture has been found to be a critical parameter as well. The greater the percentage of the silica and moisture content, the higher the unconfined yield strength of the fines. In research on the effect of clays on coal, Mikka and Smitham (1985) concluded that kaolinite has much less effect on handleability of coal than that of bentonite.

The negative effect of bentonite on coal handleability of Walloon coals from Australia had been discussed in the work by Crisafulli et al. (1985). The increase in amount of the -0.125 mm fraction after wet tumbling was found to be indicative of clays' presence. The analysis of this fraction gave an accurate estimation of the type of clays in a particular coal. Typically, bentonite absorbs water and starts to swell, becomes sticky and adheres to coal particles forming an impermeable layer around them and acting as a bonding paste between these particles. The study by Blondin et al. (1988), as discussed in the previous section, also demonstrated that by detecting the presence of clays and measuring the surface area and amount of fines, it is possible to predict difficulties in handling for coals.

Correlation between the ash content of the whole blend of coal and the handleability index (DCI) derived from the Durham Cone test was found rather insignificant (Brown et al. 1996). The resulting correlation factor $R_c = 0.32$, was very low as compared to $R_c = 0.94$ for the product of the moisture and ash fraction (M x AF) of the fines (-38 µm) for these coals. The correlation between the (AF) ash fraction of the -38 µm fraction (AF = $\gamma_{0.038}$ Ash $0_{.038}/$ 100Ash_{feed}) and handleability index (DCI) was found to be $R_c = 0.46$. The

significant correlation between the product of the moisture and ash fraction of the fines (-38 μ m) and the handleability index may indicate strong effects of clays in the presence of water. This correlation did hold for some coals tested using the Handleability Monitor but not for all (Brown, 2000).

It is generally accepted that particle size distribution influences the flow behavior of a dry material so that a dry coal made up of coarse particles, e.g. -1 mm, generally is free flowing. In comparison, a material made up of particles -100 µm in size leads to handling problems as described in the CANMET report (1996). The increase in amount of fines in coal blends is considered a critical factor. The size of a particle is directly related to its surface area (Carr, 1969), the relative surface area increases with decrease in particle size. The larger the surface area of particles, the greater will be the effect of the surface properties on particles behavior resulting in the non-free-flow (Adamson, 1967).

The particle size, at which surface forces will predominate, depends on the properties of a particulate solid. It has been shown by Carr (1969) that a coal sample with 95 % of particles being on average of 3 mm in size was classified as a granular material with a good flowability/handleability. The same sample with 80 % of particles below 74 μ m had poor flowability and acted like cohesive powders.

In the reviewed literature on coal handleability, various sizes of fines were examined. Typically, the fine fraction of -3.3 mm or -2.3 mm, or -1.2 mm were used for testing with the use of the Jenike Shear Cell. Therefore, only behavior of these size fractions was analyzed. Arnold (1990) for example, used the amount of -44 μ m fraction of coal to correlate with the handleability index (HI). She found a relatively good relationship between increasing amounts of fines in the presence of moisture and handleability index for tested coals. In the work by Brown et al., (1996) and Brown, (2000), the yield of -38 μ m fraction was correlated relatively well with the handleability indices derived from Durham Cone and Handleability Monitor.

These correlations were obtained for the yield of fractions where typically clays are found. Thus, the effect seems to be very much related to the influence of clays on coal particles in presence of water. In all of these studies where the finest fractions were linked with the handleability behavior, the resulting relationships were holding very well, possibly due to the fact that the presence of clays was a significant factor in these correlations. This

also might be the explanation for the inconsistencies for other samples which did not have high amounts of clay material in the finest fractions and therefore did not conform to the relationship.

In coal preparation, the fines that are of significant importance are in the -0.5 mm fraction. The correlation between the amount -0.5 mm fraction and handleability as derived from bulk testing methods, always indicated that an increased amount of fines led to the deterioration of handling characteristics of coal blend. On the other hand, adding the coarse material to the bulk coal was shown to improve handleability; the larger particles most likely offset the moisture effects (Mikka and Smitham, 1985). From previous studies appears that there are two types of limits for deterioration in handleability of bulk coal: one being moisture content in the sample and the other amount of fines (-0.5 mm). It was difficult to infer whether there is any meaningful correlation between these two variables from the existing literature on coal handleability.

In a number of studies it was indicated that at certain moisture levels (6 to 10 %) significant deterioration in handleability occurred. This roughly coincides with a minimum in bulk density of a loosely packed coal which is observed at 8 % moisture, as described by Leonard et al. (1993) or between 5 and 9 % for coals ranging from low to high rank as tested by Hall et al. (1960). Correlation between the bulk density and handleability of coals of different ranks was studied by Hall and Cutress (1960). It was concluded that a minimum in the bulk density always preceded the sudden deterioration in handleability of the tested coals.

Major factors influencing the bulk density of coal (Leonard et al. 1992) are: moisture content, particle surface properties, particle shape, particle size distribution, and particle density. The bulk density of the solid is the weight of the solid particles per unit of volume they occupy, and is expressed in kg/m³. The bulk density of the granular solid is less than its specific density. At least two bulk densities can be determined for any given granular solid: aerated bulk density (loose) and packed bulk density (Carr, 1965; 1969).

Due to the hydrophobic nature of higher rank coals, the bulk density of coal rapidly decreases with increase in moisture, as the water stays on the surface of coal in between the solid particles increasing volume of the bulk solid (decreasing bulk density). Further increase in moisture leads to the minimum bulk density attained at about 6 to 8 % moisture

content (Leonard et al., 1992). Beyond this level of moisture, more water penetrates the spaces in between the particles making possible aggregation of particles which are packed into smaller volume leading to an increase in bulk density of the sample. It was shown that small additions of chemical reagents resulted in increased bulk density. The reagents were added to the coal to reduce the surface tension of water and to enhance hydrophilicity of coal particles (Leonard et al. 1992). These experiments showed that increase of 13 % to 15 % in bulk density could be achieved using such additives.

Adding hydrocarbon oil to high rank coals increased the bulk density and at the same time considerably improved handleability of these coals (Hall and Cutress, 1960) over a wide range of moisture contents. This effect of bulk density increase is associated with the flattening of the bulk density curve, making it much less influenced by moisture increase over the range from 5 % to 18 %. The addition of the same oil to the lower rank coals had no effect on the bulk density, while the addition of water-soluble oils to these coals led to the change in the bulk density with the increase of moisture content. This phenomenon is explained by the fact that hydrocarbon oils have strong affinity to hydrophobic surfaces such as those encountered in higher rank coals. Thus, it is possible for oil to displace water from high rank coal and spread on particles surface, while for low rank coals, the oil cannot displace water from the surface of coal due to its strong hydrophilic character. An improvement in handleability of oiled coals seems to be attributed to further enhancement of hydrophobicity of coal particles, although the bulk density was increased (more packing), the effects of increasing moisture were less pronounced. This can also indicate that perhaps the hydrophobicity of coal surface is somewhat more important than increased packing between these particles.

From the cited literature, the amount of fines, moisture and ash content are the most important factors. There may be some interrelation between the amount of fines and moisture content. At least in two independent studies it was shown that once certain level of moisture is reached the correlation with other factors, e.g. the amount of fines, becomes less predictable, whereas samples with low fines content are practically insensitive to moisture increases (Hall and Cutress, 1960; Mikka and Smitham, 1985). The size distribution was also shown to be a significant factor. The way the bulk sample packs into a confined space is reflected by its bulk density and is related to the size distribution as well

as the effects of moisture on the coal particles. Mineral matter content has the greatest effect on handleability when clays are present. A high content of clays in the fines contribute to major problems in handling as their presence leads to buildup of cohesiveness within fines as well as coarse particles.

Other surface properties of particles that influence handleability of fine coal apart from wettability are: surface roughness (particle shape), electrical charge and interparticle friction as described by Leonard et al., (1992). These factors influence the agglomerating properties of the particles and affect the flowability of fine coal. The finer the coal particle size, the more pronounced the effect of surface properties on flowability.

It has been shown that the shape of a particle may influence flow with the roundish particles to have the best flow due to the fewer interparticle contacts. Particles with random and irregular shapes will have more interparticle contacts through the edges, corners and uneven surfaces. According to Carr (1969), fibrous, dendritic, plate-like, irregular, and angular particles tend to be less flowable. The irregular shape of these particles increases resistance to sliding against each other, and is referred to as friction.

For example, particle shape is an important variable affecting packing of particles in the mass of bulk solids, influencing bulk density. Cubic particles can be potentially packed to a minimum porosity of 0 %, while spherical particles of the same size can be packed to the minimum porosity of 26 % (Leonard et al., 1992). Angular particles can be frequently packed closer than rounded particles due to the fitting of the projections into voids. Fine powders pack less closely than coarse powders, larger size fractions of material will have greater bulk density than will smaller size fractions. There will always be such a mixture of fine and coarse particles at which the bulk density will assume the highest value, higher than assumed by any of these fractions when packed separately. According to Wakeman (1975), as the spectrum of sizes of particles is increased, the bulk density is also increasing.

Roughness has also an effect on the ability for fine particles to aggregate; according to Coehlo and Harnby (1978), the surface roughness will cause a distance to be maintained between the particles of about the average peak-to-trough height of surface irregularities. The surface roughness will also reduce the effective layer thickness of the adsorbed liquid, and decrease the binding force between the particles. However, once an adsorbed liquid reaches certain thickness, it becomes thick enough to cover most peaks of roughness

causing the attraction between the particles will increase greatly. Therefore, roughness will be a critical factor in aggregation of particles, when there is no significant amount of moisture present in the sample.

3.3 Coal and its surface properties

3.3.1 Wettability of coal

Solid wettability is determined by equilibrium between the energy of solid/liquid adhesion and liquid cohesion as described by Harkins (1952). For a solid to be hydrophobic-non wettable, the work of adhesion of liquid to solid (W_{sl}) has to be smaller than the work of cohesion of the liquid (W_{ll}). Work of adhesion (W_{sl}) can be split into Lifshits-van der Waals dispersive forces contribution and acid-based contributions. Therefore, freshly formed solid surface remain naturally hydrophobic only if its fracture or cleavage occurs without rupture of inter-atomic bonds other than residual ones (Gaudin, 1957). These surfaces can interact with an aqueous environment through the weak dispersion forces. Hydrophobicity arises essentially from the weakness of adhesion of water to the solid (Fowkes, 1963; Laskowski and Kitchener, 1969).

Coal is considered to be naturally hydrophobic. This is a result of its hydrocarbon structure. Coal chemical structure is described as highly cross-linked polymer, where a number of stable fragments are connected by weakly cross-links. At lower ranks, coal structure is predominately aliphatic and becoming increasingly aromatic in higher ranks. Aromaticity in coal, reaches its maximum at about 94 % carbon content (van Krevelen, 1961; Lowry, 1963; Ignasiak and Gawlak, 1977; Larsen and Kovac, 1978; Given, 1984). Coal also contains heteroatoms and functional groups attached to the organic matrix as depicted by Wiser's model (Laskowski, 2001). The presence of these groups strongly affects the wettability of coal. The oxygen functional groups such as hydroxyl, carbonyl or carboxyl occur either as a result of oxidation, or can be a part of the coal structure. The amount and type of these groups change with the rank of coal or with degree of oxidation. The oxygen functional groups' type and content, and their type was frequently used to stress the change of coal surface properties and wettability with rank (Ihnatowicz,1952; Bloom et al. 1957; van Krevelen, 1961). The hydrophobicity/wettability of coal was shown to be a function of rank. Lower rank coals are usually less hydrophobic (more wettable) than higher rank coals due to the presence of a large number of oxygen functional groups in the coal structure (Klassen 1966).

Oxidation is the process of deterioration of the coal surface. A hydrophobic coal can become hydrophilic when oxidized. The oxygen reacts with the coal surface and functional groups such as -OH, -CO, -COOH and OCH₃ are formed. These functional groups are preferential sites for water adsorption on to the coal and as a result the degree of oxidation of coal significantly affects the wettability of coal. Many sophisticated methods and techniques have been used to study and detect oxidation in coal. Among these are pyrolysis, mass spectrometry, gas chromatography, FTIR technique (Meuzelaar, 1987), along with simpler testing procedures such as titrations of acidic groups (Ihnatowicz, 1952, Bloom, 1957, Schafer, 1970; Ignasiak and Ignasiak, 1970) or very practical ones, frequently used in the industry such as the alkali solubility test (Gray et al. 1976; Gray and Lowenhaupt, 1989), which became the ASTM standard for detection of oxidation in coal.

Coal is an organic sedimentary rock composed of two basic components: inorganic crystalline minerals and organic carbonaceous components recognized as macerals. The macerals are a product of the parent organic matter, and change with the degree of coalification. Coalification is the progressive enrichment of the coal substance in organically bound carbon. Since most of the coal minerals are hydrophilic, the presence of mineral matter on the coal particles has an effect on their wettability as well, as these mineral patches on coal particle become sites for water attraction. As a result, the type and amount of mineral matter associated with coal becomes an important factor in the wettability of coal.

As shown by Kelebek et al. (1981), coal becomes more hydrophilic with increasing mineral matter (ash) content. However the ash content characterizes only the amount of mineral matter and not its distribution or the mineral matter grain size distribution; thus it is obvious that the mineral grain size and distribution must have a very important effect on coal surface hydrophobicity/wettability. The experimental evidence that the mineral grain size affects values of a contact angle was provided by Drelich et al. (2002).

For a given rank and controlled level of oxidation, the hydrophobicity of coal is primarily dependant on the surface properties of the coal organic matrix, represented by the maceral makeup and the association of mineral matter with these macerals (Laskowski et al., 2002). There is a limiting value of mineral matter (ash) content in coal below which,

depending on the degree of surface hydrophobicity/wettability of the coal matrix, mineral matter content does not have any significant effect. It was found that for one bituminous coal, the limiting value was at 15 % ash content (Holuszko, 1991; Holuszko and Laskowski, 1995) and for another bituminous coal it was at 20 % ash content resulting from unliberated mineral matter content (Laskowski et al., 2002). For very hydrophobic coal, even a high amount of ash content may not affect its wettability significantly. For each coal, depending on its degree of hydrophobicity and mode of mineral matter association within the coal matrix there will be a limiting value of ash. In general, large mineral matter grains have a more pronounced effect than small grains on the degree of hydrophobicity for the same coal. In hydrophilic coal, however the same amount of mineral matter (ash) could have more significant effect on the wettability than on hydrophobic one (Bustamante and Warren, 1983).

The contact angle has long been the only measure of hydrophobicity of coal. Brady and Gauger (1940), and Elyashevitch (1941) reported the earliest results on contact angles of different coals. In studies by Horsley and Smith (1951), Klassen (1966) and Sun (1954) it was shown that bituminous coals always had higher contact angle than either anthracite or lignite. The hydrophobicity of coal was shown to be a function of rank (Ihnatowicz, 1952; Bloom et al., 1957). Aplan, (1983), Guetierriez-Rodriquez et al. (1984), and Onlin and Aplan (1984) related hydrophobicity of coal to various rank parameters, such as % C content, fixed carbon, oxygen, OH/carbon ratio, and vitrinite reflectance.

Characterization of coal wettability by contact angle is quite difficult not only because of natural coal heterogeneity but also due to coal surface preparation related problems. In recent years, the experimental procedure for contact angle measurements using captive-bubble and sessile drop methods have been significantly improved (Drelich, 1997). Many effects were eliminated by improving the sample preparation stage (Drelich, 1997; Drelich et al. 2000).

All these improvements in contact angle techniques are important steps in surface wettability assessments. The measurement of a large population of contact angles on polished coal specimens, however, is time consuming and depends largely on the quality of the polished surfaces. Also, a randomly selected block of coal hardly represents fine coal particles usually processed in a coal preparation plant. While measuring contact angles

directly on powders is attainable as shown by Eissler and van Holde (1962), it can also become a very tedious task to accomplish. In another method, where fine particles are compressed into a disc and the contact angle is measured directly on fine powder, it was shown that such measurements become complicated by the effects related to porosity of the prepared surface as discussed by He and Laskowski, (1992).

Other indirect techniques of characterizing the hydrophobicity/wettability have been developed over the years. The following methods are applicable for measuring wettability of fine particles and powders: the rate of disappearance of coal particles deposited on the surfactant solutions (Garshva et al., 1978; Glanville and Whigtman, 1980), a measure of immersion time for particles to sink into the liquids of different surface tensions (Hornsby and Leja, 1980; Fuerstenau, et al. 1986; Fuerstenau and Williams, 1987), induction time (Ye and Miller, 1988) or suction potential method (Clark and Mason, 1968; Qiu and Wheelock, 1994). The experimental technique that can provide a distribution of contact angle values measured directly on fine particles was developed by Fuerstenau et al. (1990) and referred to as film flotation. This method, when used along with Neuman's equation of state (Li and Neumann 1992), becomes very practical in evaluating surface energy of many low energy materials including those having polar components (Drelich, 1997).

Another practical method of quantifying wettability of fine particles as an alternative to contact angle measurements is the assessment of the capillary rise in a bed of particles packed into the column. Washburn in 1921 and later Crowl and Wooldridge (1967), followed by White (1982) formulated an equation to describe the rate of penetration of liquid into a bed of powder. This technique became widely accepted for assessing wettability of fine particles. In a number of studies (Ku et al. 1984; Kilau and Voltz, 1991; Diggins and Ralston, 1992; van Oss et al. 1992; Zhmud et al. 2000; Yildirim and Yoon, 2002) the capillary rise method was used to determine wettability for a variety of solids.

The liquid penetration rate into the bed of particles, depends on the surface tension of the penetrating liquid γ_{lv} , contact angle between the liquid and the particles and the effective pore radius in the bed of fine particles. As the effective pore radius is influenced by the mode of packing in the powder beds, this approach requires consistency in sample preparation in order to give reproducible results. In studies by Ku et al. (1985) or van Oss et al. (1992) brief descriptions of packing techniques were included. In the paper by Wolfrom

et al. (2002) a more precise attempt was made to formulate a standardized procedure for packing the powder into the column. The consistency in the bed packing was described as critical and adding the powder to the column has to be always performed in the same manner for every sample tested (Tampy et al., 1988; Siebold et al., 1997).

The rate with which liquid penetrates the bed made of fine particles is either reported as the correlation between the rising heights of the progressing wetting front or as an increase in weight measured automatically over the time. It has been shown that the automatic weight measurement has greater precision than the visual height observation (Siebold et al., 1997). The shape of the penetration rate curves can be indicative of the differences in wetting between various samples (Laskowski et al., 2003). The contact angle or adhesion tension of liquids in contact with powders can also be assessed from such measurements.

This, however, requires a knowledge of the effective or hypothetical pore radius that has to be estimated along with the other constants related to the geometry of the pore system. This is usually done in separate calibration steps, in which a liquid that completely wets solid particles is used (Crowl and Wooldridge, 1967; Bruil and von Aasten, 1974; Crawford et al., 1987).

The penetration rate measurements are of practical importance to handleability, because the test resembles the conditions during handling of fine coal. Although, during handling, coarse and fine particles are tumbled and compacted together and the fines are the most affected by the moisture content. Thus, the water (moisture) is likely to penetrate a bed of particles formed by the mechanical movements associated with the transportation, storage or other handling procedures. This technique was found to be the most relevant for assessing wettability of fines for handleability studies, along with the transmittance method to measure the content of humic acids extracted from coal.

The transmittance tests are commercially used as the method to detect oxidation in coal. Lowenhaupt and Gray (1980), in a study using bituminous coals, correlated results of the alkali-extraction test with petrographic, infrared and oxygen analyses. They showed that the alkali-extraction test was in agreement with the results obtained from petrographic procedures as well as with the data obtained from the oxygen content study using neutron activation and FTIR analysis. This method has been accepted widely throughout the coal

industry as a quick and inexpensive test for the determination of the relative degree of oxidation in bituminous coals. Oxidized coal produces a yellow to brown solution when leached with sodium hydroxide solution.

The transmittance value measured at 520 nm UV is proportional to the degree of oxidation. From the industrial testing, Gray and Lowenhaupt (1989) determined, for coking purposes, the transmittance must be at 90 % value for coal to be acceptable for coke production. This coincides well with the hydrophobic characteristics of coal.

In local preparation plants a 90 % transmittance value is also the cut-off point for coal to be acceptable for flotation (personal communication¹). It is a common practice to use transmittance for assessing the relative degree of oxidation and wettability of bituminous coals.

3.3-2 Coal moisture and porosity

Moisture content in coal is a rank dependant property. While lower rank coals have high moisture contents, bituminous coals have moisture contents in the range of a few percent. There are many different forms of moisture that can be measured in coal. When the moisture is determined in the samples as received according to standardized procedure (ASTM D 3302-91) it is referred to as the total moisture in coal. When the coal is air-dried, the moisture remaining in the sample is called residual and the difference between the total and residual moisture is the surface moisture. Equilibrium moisture (ASTM D 1412-93) and moisture holding capacity (ISO) represent moisture in coal when at equilibrium with an atmosphere of 97 % humidity and at 30^oC. Equilibrium moisture, or bed moisture (inherent) as it is often referred to, is a reliable classification parameter along with the calorific value for lower rank coals.

A steady decrease in moisture content over the range from low rank lignite to medium volatile coals results from a decrease in porosity and the content of functional groups. These oxygen containing groups such as -OH, -COOH, -OCH₃ and --CO, are mainly responsible for water adsorption and water holding capacity within the coal structure. In the range between high volatile coals and anthracite, the moisture content falls to a minimum value at a level of medium volatile, and increases slightly to an anthracite level (Teichmuller and Teichmuller , 1982). Similar trends are observed for porosity,

surface area and wettability for coals. This is the result of the coalification process, which is associated with chemical changes. Coal becomes increasingly aromatic as the rank progresses. Porosity and surface area change with the rank of coal accordingly, and this determines water adsorption onto the coal structure.

One method of assessing the extent of porosity in coal is to determine its equilibrium moisture content. Depending on the volume (size) of the pores in the coal, the amount of water adsorbed onto the coal surface is influenced by the topology of pore networks within the coal as well as the content of oxygen functional groups in coal (Mahajan and Walker, 1971; Silbernagel, 1988; Laskowski, 2001). According to Gan et al. (1972) and Parkash et al. (1984) in lower-rank coals porosity is primarily due to the presence of macropores (30-200 nm), while in coals of intermediate rank (76 to 84 %C), most of the pores are transitional pores (1.2-30 nm). In higher rank coals micropores predominate.

Mahajan and Walker (1971) plotted water vapor adsorption onto coals of different ranks as a function of relative pressure (Figure 3.3-3). The moisture sorption isotherms were measured at 20^{0} C (p/p₀ is the ratio of the water pressure to the saturation vapor pressure of water). It is evident that the amount of water adsorbed onto coal at high humidities (close to the saturation pressure p/p₀=1) is consistent with coal porosity.

The sorption isotherms, however, are quite different for coals of varying ranks (Laskowski, 2001). The shape of the sorption isotherm depends on porosity as well as the functional group content; this is especially true at the low values of p/p₀. The correlation between wettability and moisture content is complicated by the capillary condensation phenomena, which influences coal moisture content. The oxygen functional groups are the primary sites for water molecule adsorption on coal. Kaji et al. (1986) showed that there is a linear correlation between coal-water holding capacity (equilibrium moisture) and the product of oxygen content and specific surface area of coal. He also observed that for some coals, adsorbed water occupies only 30-75% of the total pore volume, while for other coals the adsorbed water exceeds 2 to 3 times their pore volume. Mineral matter, however, was found not to affect the water holding capacity of studied coals and this was also in agreement with the observation by Murata (1981).

Kaji et al. (1986) proposed the model adsorption where water adsorption onto the coal surface occurs in three steps: monolayer sorption, multilayer condensation and capillary condensation. It has been found that the monolayer capacity for water on coal correlated well with the number of hydrophilic functional groups. The water in the monolayer was believed to be attached to the coal surface through hydrogen bonding, and the further sorption of water was considered to occur by clustering around the hydrophilic sites on top of the monolayer (Shafer, 1972). This could probably explain that for very hydrophilic coals, the water adsorption was exceeding pore volume, or even the total surface area of tested coals as measured by Kaji et al, (1986).

In the recent study by Fuji et al. (2000) microscopic and macroscopic wettabilities of various silica surfaces (hydrophilic and modified-hydrophobic) were examined. Microscopic (at nano-level including mesopores) wettability was determined by water adsorption, while macroscopic (at macro-level) wettability by measuring the contact angle. It was shown that microscopic wettability leads to the formation of a continuous twodimensional water adsorption layer, whereas macroscopic wettability essentially arises from microscopic wettability. Figure 3.3-4 and Figure 3.3-5 illustrate wettability concept for hydrophobic and hydrophilic solid. In summary, the solid is strongly hydrophilic when the water adsorbs into micropores, this results in the formation of the firm layer on the outside surface due to capillary condensation in these micropores. In hydrophobic samples, only the outer surface structure influences the wettability of the particles.

This is significant because, when the surface of coal is hydrophilic, pores are filled with water and multiple layers of adsorbed water form a very strong and stable film around each particle. Coal cannot adsorb more than its surface can hold, due to the porosity, degree of wettability and total surface available for adsorption. Therefore, the water holding capacity (equilibrium moisture) can be considered as the limiting value beyond which an excess of water appears on the surface of coal. It is this liquid layer which will take a part in building the bridges between particles, leading to their aggregation.



Figure 3.3-1 Water vapor adsorption onto coals as a function of relative pressure for coals of different ranks. Values above the lines indicate % C in studied coals (Majahan and Walker, 1971); reproduced by permission of Elsevier.

Hydrophobic surface



Figure 3.3-2 Macro and Micro (nano-level) wettability model for porous hydrophobic silica adapted from Fuji et al., (2000) by permission of The Society of Powder Technology of Japan.

3.4 Aggregation and Pelletization

3.4-1 Aggregation

Cohesion of fine particles is due to the presence of liquid bridges between these particles. Bridges are formed as a result of liquid mobility between the particles. At low liquid levels or when the surface of solid particles is hydrophobic, only a few contacts between particles through the liquid phase are made. This is referred to as the pendular state, that persists until liquid drops begin to coalesce. Once the liquid occupies more than 20% of the volume in the aggregate of particles, the funicular state is reached. This is when a liquid forms an almost continuous network with a few spaces filled with air.

Hydrophilic surface



Figure 3.3-3 Macro and Micro (nano-level) wettability model for porous hydrophilic silica adapted from Fuji et al., (2000) by permission of The Society of Powder Technology of Japan.

When all the spaces between particles become filled with liquid, the capillary state prevails. Figure 3.4.1-1 shows the aggregation stages as described by Capes, 1980. Strength of the bonding between the particles in the pendular state is one third of that in the capillary state, while the funicular state yields intermediate values (Capes, 1980). For aggregation in presence of liquid, the liquid has to wet the surface of coal particles, the liquid bridges between particles must form, and the capillary forces must create the bonds between the particles.

This indicates that the wettability of particles can have a significant effect on the bond strength developed between particles. The aggregation of fine particles has a negative effect on the flowability of the whole sample. When the fine particles aggregate they stick together and form a mass that prevents flow of the whole sample. The stronger the aggregates are, the less handleable the whole sample becomes. The tendency for fine particles to aggregate has very damaging consequence for the handleability.

3.4-2 Pelletization

Pelletization is the particle-enlargement process, in which the particles are deliberately forced to aggregate to form pellets. This is accomplished by tumbling the moist fine particles in drums, discs, or in conical devices. Physical forces, such as interfacial attraction, surface tension, van der Waals interactions, and particle interlocking effects, combined with the applied mechanical energy of tumbling, bring particles together to initiate the pelletization process. In an extensive study on pelletization of coals of various ranks by Sastry and Fuerstenau (1982), the fundamental principles of coal pelletization have been delineated.



Figure 3.4-1 Liquid bonding; states of liquid content in an aggregate A- pendular, Bfunicular, C- capillary, D – particle in liquid (Capes, 1980); reproduced by permission of Elsevier. The important conclusions arising from this research were:

- 1. Coals of all ranks and ash contents can be pelletized;
- 2. Moisture addition for successful pelletization is critical, and lies in a narrow range for each coal. It is directly dependent on coal ash content, and varies inversely with ash;
- 3. It was found that the moisture requirement for pelletization, M_w (weight percent of dry coal), and the percent ash in the coal fines, A_c, can be described by the following relationship:

 $M_w = 43.96 \exp(-0.017 A_c),$ (3-6)

This equation is applicable for all types of coals except lignites irrespective of their rank and origin. Lignites require a much higher moisture addition for pelletization, than expected from the correlation. This may be attributed to the large internal pore volume of lower rank coals.

4. Clean coal fines of the size range minus 0.6 mm can be pelletized successfully. Pellet strength, however, decreases with decreasing ash content indicating that while these fines can be pelletized, pellets do not have acceptable strength.

According to these authors, the driving force for pelletization is the lowering of the total surface free energy of the system through a reduction of the effective air water interfacial area (Sastry and Fuerstenau, 1982). The surface forces that can play a role in reducing the surface free energy are:

- 1. Interfacial effects giving rise to capillary and surface tensions forces;
- 2. van der Waal's interactions;
- 3. Electro- and magnetic interactions;
- 4. Interlocking effects between the particles;
- 5. Immovable bridging bonds.

Mechanical forces such as tumbling motion bring the individual wetted particles into proximity with each other so that surface forces become operative.

According to Kapur and Fuerstenau (1966) and Sastry and Fuerstenau (1977), the pellet formation is achieved in three stages: first the formation of nuclei agglomerates, then the transitional stage, and finally, ball growth stage. The mechanical forces of rolling bring the particles into proximity with each other. Then, the physical forces become operative and

cause the particles to rearrange in such a way that a reduction in surface tension facilitates nuclei formation through the bridges of wetting liquid. Therefore, the aggregation of particles due to capillary forces depends on the wettability of the particles by bridging liquid (Schubert, 1984).

Due to the heterogeneous character of coal (patches of mineral matter and different proportions of macerals with varying hydrophobicity), water does not spread over the whole coal particle surface to form a film. On a relatively hydrophobic surface, water will only be attracted to the hydrophilic sites, represented by mineral matter patches, and macropores related to the physical roughness of particles. This is probably why, in the pelletization study by Fuerstenau et al. (1982), the ash content was found to be the most significant property determining the moisture requirement. Once there is a contact between particles through the water bridges, nuclei are formed, and the aggregation proceeds into the further stages of pellet formation. In order to obtain a strong pellet, it is necessary for the bond between colliding particles in a pelletization device to be strong enough to withstand the shearing forces in the device.

The strength of these pellets, however, is related to the capillary and surface forces (Rumpf, 1962). It has been shown that weaker capillary forces were observed in the pellets made from iron ore when they were weakened by the hydrophobic surface layer of flotation collector (Gustafsson and Adolfson, 1997). Therefore, the hydrophobicity of coal particles becomes an important parameter as well. Other researchers also showed that bond strength decreases with an increasing contact angle (Naidich et al. 1965a,b; Mehrotra and Sastry, 1980), that is the more hydrophobic particles the less stable the pellets. This perhaps explains why coal fines with high ash content (supposedly more hydrophilic) can be pelletized at low moisture levels while coals with low ash content need a greater amount of water for pelletization. This implies that adding more water can compensate for a reduction in pellet strength due to hydrophobicity (Sastry and Fuerstenau, 1982).

On the other hand, pelletization tests performed on clean coal indicated that clean coals would need 3 to 4 percentage units less moisture to pelletize than respective raw coals (Sastry and Fuerstenau, 1982). The only difference between the clean and raw coal samples tested in this study was the distribution of ash bearing particles in coal fines. In the case of raw coals, apart from the mineral matter associated with coal, there is usually liberated

mineral matter which is hydrophilic and may represent the first sites for water adsorption. An increased amount of water consumption can be observed, especially when clays are present. It has also been concluded, that pellets prepared from clean coal were not strong enough to withstand the shearing in the pelletizing device. This may imply that while it is possible to pelletize clean coal, due to the strong hydrophobicity of particles these aggregates lack the bond strength as discussed previously. Furthermore, it was found that coals with a higher ash content can produce stronger pellets than the coals with lower ash content. The effect of the ash content of coal on the strength of pellets can be attributed to the fact that some ash forming minerals might be acting as binders, e.g. clays. The other explanation is that liberated mineral matter or coal particles contaminated with mineral matter become the nuclei around which pellets are built during the pelletization.

The effect of the feed fineness (increased surface area by grinding) on the strength of coal pellets was determined for coal samples varying in rank (Sastry and Fuerstenau, 1982). It was concluded that the pellet strength increases with the increase in the feed fineness for all coals, except for anthracite. The anthracite pellets were very weak even with finer feeds. According to Sastry and Fuerstenau (1982), the effect of feed fineness on the strength of pellets can be explained in terms of an increase in the interparticle friction as the surface area of the feed increases. As the feed particles become finer, the frictional forces, which are proportional to the contact area between surfaces, also increase. If the coal particles are less hydrophobic, bonds between particles are stronger due to the formation of more stable water film around them. Therefore, the effect of frictional forces on pellet strength is also much more pronounced for these type of particles than for anthracite coal particles, which are more hydrophobic.

It is not clear, however, what was the degree of hydrophobicity of these studied coal particles. The hydrophobicity of a particle is described only on the basis of its mineral matter content (ash in the coal). It was also assumed that the contact angle measured on the block of coal represented the hydrophobicity of the fine coal particles, and was used to explain certain hydrophobic vs. hydrophilic effects. This is a questionable approach, as the random piece of coal cannot fully represent the surface of the fine coal particles.

There are apparent similarities between particles in pelletization and in free flow that determine handleability. In both processes, coal particles are subjected to mechanical

forces; therefore, the same phenomena should be responsible for their behavior. It can be assumed that in the system where coal particles are subjected to mechanical movement due to rotation (pelletization) or flowing (handleability), the particles collide with each other and as the moist particles encounter each other, the aggregation of particles may take place. The same surface properties which lead to aggregation in pelletization will cause particles to stick together and build cohesiveness of the fines affecting handleability of the bulk sample. As a result, pelletization can be used as the method to elucidate the fine coal particles behavior that determines handleability. The pelletization can be used as a procedure for testing ability of fine particles to aggregate. This will be the first such approach to use pelletization as a method to assess the handleability of fine coal.

CHAPTER 4. COAL SAMPLES

4.1 Sample types

Two types of samples were used for the handleability tests: raw coal samples and coal products (clean coal). Raw coal samples were obtained from the mine site, they were collected as run-of -mine samples, and clean coal product samples were acquired from the coal preparation plant of this same mine.

Five raw coal samples were chosen for this study. Three coals were of metallurgical grade (medium volatile; LC 3, LC 10BC, LC 10B) and two of thermal grade; one was a lower rank coal (high volatile; LS 20) and one was higher rank (medium volatile-oxidized; LC 8U). In addition, one of the raw coals was oxidized by heating at 180°C in the oven for eight weeks and used for testing (LC 30XY). Coal analysis data for these samples is provided in Table 4.1-1.

The clean coal samples are represented by metallurgical and thermal coal products. Two coal products were from British Columbia; one referred to as "LC met" and the other one is "LC thermal". Other coal products were also used in the study: two metallurgical coals were from Australia (coal #11 and #12) and three thermal coals were from the UK (coal #2, #4 and #5). The size distributions of the original samples, and with the added coarse fractions are presented in Table 4.1-2, along with their ash content.

In addition to the above-discussed samples, seven product coal samples were randomly chosen for simultaneous testing with the Handleability Monitor and the Durham Cone. These seven coals were tested on "as received" basis. There was no physical or chemical data provided for these samples.

4.2 Sample preparation

For bulk testing, raw coal samples were prepared in two top sizes. One set of samples was run-of-mine (ROM) as originally received from the mine. Large lumps found in the run-of-mine samples were removed and only the coal particles 100 % passing 53 mm screen were included in samples for bulk coal testing.

Coal	Moisture %*	Ash % (AR)**	V.M. (daf)***	FC (daf)***	HGI (Hardgrove Grindability Index)
LC 3	0.57	30.12	26.4 3	73.57	83
LC 3 OXY	1.91	30.12	32.2 7	67.73	-
LC 10BC	0.60	37.45	30.1 7	69.83	80
LC 10B	1.29	24.30	29.9 8	70.02	83
LC 8U	2.26	23.63	29.0 1	70.99	74
LS 20	4.42	33.38	42.5 4	57.46	50
LC met	0.52	9.77	25.4 8	74.52	83
LC thermal	1.58	15.15	25.5 1	74.49	80

Table 4.1-1 Proximate analyses of studied coals

* -total moisture **(AR)- as received ***(daf) - dry ash free basis

The second set of samples was prepared by crushing to 100 % passing 6.3 mm screen. The crushed samples were also used in the handleability tests and in the study on the effect of size distribution on handleability. The third set of samples was prepared by adding 20 % (by weight of the whole sample) of the -0.5 mm fraction to the ROM samples. The size analyses of these samples are shown in Table 4.2-1. Coal product samples were used for handleability study on as received basis. The top size in these bulk samples was 50 mm, the amount of fines (-0.5 mm) was 44 % wt in LC met, and 45 %wt in LC thermal coal. Normally, coal products from the local mines have from 30-35 % wt of fines, so these particular samples represented much higher levels of fines.

				Pres			
Sample	Coal 11 cumul.	Coal 11 (added	Coal 12 cumul.	Coal 12 (added	Coal 2 cumul.	Coal 4 Cumul	Coal 5
Size (mm)	remain. wt%	coarse)	remain. wt%	coarse)	remain. wt%	remain. wt%	remain.
Size (mm)		cumul.		cumul.			wt%
		remain.		remain.			
		wt%		wt%			
12.5-9.5	24.19	34.19	28.58	39.99	38.48	17.66	24.57
9.5-0.5	59.08	64.48	72:69	77.06	91.53	83.91	87.53
0.5-0.15	83.19	85.41	91.86	93.16	97.36	95.20	95.25
0.15-0038	99.30	99.39	99.70	99.75	99.84	99.46	99.48
-0.0038	100.0	100.0	100.0	100.0	100.0	100.0	100.0
						:	
Ash % in bulk	9.67	-	10.46		11.70	13.49	11.50
sample Yield of -0.5mm	40.92	35.52	27.31		8.47	16.09	12.47
Ash % in - 0.5mm	8.95	8.95	15.13		14.87	34.1	23.6

Table 4.1-2 Size distributions of U.K. and Australian coal products.

4.2.1 Bulk sample preparation

For the handleability study with the Durham Cone, samples were prepared in batches of 10 kg. These samples were mixed immediately prior to testing. When the moisture was added, special caution was taken to mix the coal sample thoroughly. Mixing was done by vigorous shaking in plastic bags. Samples were always coned-and- quartered before each moisture addition. The conditioning method was uniform for all the tested samples.

For testing with the Handleability Monitor, larger samples (50 kg), were used asreceived. The samples were mixed done by the cone-and-quarter method, after which the sample was loaded into the Handleability Monitor and tested. The testing was performed at the University of Nottingham where the Handleability Monitor is located.

4.2.2 Preparation of fines

The fines (-0.5 mm) were sieved from each tested sample and kept in plastic bags under N_2 in the fridge to avoid oxidation. These fines were used for wettability and other experiments. The size distributions of fine fractions for LC 3, LC 10B, LC 10BC, LC8U, LS 20, LC met and LC thermal are shown in Figure 4.2-1. The size distribution data for all studied fines is included in Appendix A.

For pelletization and surface properties characterization, the -0.5 mm size fraction was used. The clean coal products were used as-received and a fine fraction of -0.5 mm was also isolated from the bulk sample for surface properties characterization.



Figure 4.2-1. Particle size distribution of -0.5 mm size fractions from left to right: LC thermal, LC8U, LC 10B, LC 10BC, LC met and LS 20.

Size(mm)	LC 3	LC 3	LC 10B	LC 10B	LC 8U	LC 8U	LC 10BC	LC 10BC
	ROM	crushed	ROM	crushed	ROM	crushed	ROM	crushed
	Cumulative							
	wt%							
53-50	14.0		5.0		0.0			
33-30	14.0	-	5.0	-	9.0	-	4.0	-
50-12.5	25.9	-	18.0	-	21.0	-	18.0	-
12.5-6.3	39.1	-	35.0	-	35.0	-	33.0	-
6.3-0.5	78.2	64.0	77.0	55.0	80.0	63.0	79.0	61.0
0.5-0.25	90.5	84.0	88.0	78.0	91.0	84.0	90.0	83.0
0.25-0.15	93.0	88.0	91.0	82.0	93.0	88.0	92.0	87.0
0.15- 0.075	96.8	95.0	94.0	90.0	97.0	93.0	96.0	93.0
0.075- .045	99.6	99.0	97.0	95.0	99.0	97.0	98.0	97.0
0.045- .038	99.7	100.0	99.0	97.0	99.0	98.0	98.0	98.0
-0.038	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0

.

Table 4.2-1 Size distribution of raw coal samples (cumulative - wt % retained)

CHAPTER 5. METHODS

5.1 Wettability assessment of fines

5.1.1 Transmittance

Transmittance values were determined following the ASTM D5263-93 method. This method allows determination of the relative degree of oxidation of coals by alkali extraction. As the oxygen bonds with the coal surface during oxidation, it forms complex hydrocarbons commonly called humic acids. Because they are acids they are soluble by alkali solutions. Coal oxidation adversely affects many technological properties, such as coking quality of coal and plastic properties of bituminous coals, it also makes coal hydrophilic (wettable by water). Transmittance measurement allows to differentiate between the degree of oxidation of coal samples. It determines the concentration of humic acids, which are extracted to NaOH solution, by measuring the transmittance at 520 nm UV wavelength. The intensity of the color produced by the humic acids is dependant on the degree of oxidation. The darker the color, the higher the concentration of humic substances. The high value of transmittance (>90 %) indicates unoxidized coal, while 80 to 90 % indicates some oxidation; and value below 80 % is a signs of oxidation in coal. This simple test is very quick and reliable for assessing oxidation in coals.

The fines (-0.5 mm) from each tested coal were used to determine the transmittance following the standard procedure. The spectrophotometer was set up at 520 nm UV wavelength. Transmittance was measured for the solutions obtained by boiling coal fines in NaOH solution (1N) for 3 minutes. After boiling, the sample was transferred to the Micropore filtration unit and the leachate was used for transmittance determination. The tests were performed following the ASTM D 5263-93 procedure.

5.1.2 Penetration rate

Penetration rate tests were used to characterize wettability of fines obtained from the studied coals. The gravimetric version of the method based on the Washburn equation (Washburn, 1921) was used as described by Laskowski (2001). In this method, 3 grams of coal was packed in a tube and compacted by a column-packing device under a load of 5 kg and 11 kg. The tests were also performed with the loose packed particles; this procedure

involved compacting the fines by tapping (50 times). The tube with the compacted material was then attached to a balance, contacted with water and the total weight of the column was monitored on-line using an electronic balance (Figure 5.1.2-1). The patterns of penetration for the studied coals were plotted as the squared mass versus time.

Wettability of the fine particles by liquid can be characterized by measuring the penetration rate of water into a bed of packed fine particles. The method is based on the Washburn equation:

$$\frac{h^2}{t} = \frac{r\gamma_{LV}\cos\Theta}{2\eta}$$
(5-1)

where: h is the height of the wetted column of fine particles, t is the time, r is hypothetical mean radius of the capillaries between solid particles (depends on particle size and packing), γ_{LV} is the liquid-vapor interfacial tension, η is the liquid dynamic viscosity and Θ is the contact angle.



Figure 5.1.2-1 A penetration rate set-up in which the weight of the column is monitored on-line versus time (Laskowski, 2001); reproduced by permission of Elsevier.

Since the wetting front is not always visible, and in most cases does not reflect the inner progression of the liquid in the bed of packed particles accurately, it is more
convenient to follow the weight of the penetrating liquid. This weight, w, is interrelated with h by the following relationship

$$w = \varepsilon A h \rho \tag{5-2}$$

where ε is the bed porosity, A is the cross-sectional area of the column, and ρ is the density of the liquid.

This leads to the following relationship

$$\frac{w^2}{t} = \frac{\varepsilon^2 A^2 \rho^2 r \gamma_{LV}}{2\eta} \cos\Theta$$
(5-3)

and after rearranging

$$\frac{w^2}{t} = \left(\frac{\varepsilon^2 A^2 r}{2}\right) \frac{\rho^2 \gamma_{LV}}{\eta} \cos\Theta$$
(5-4)

and since kinematic viscosity $v = \frac{\eta}{\rho}$

$$\frac{w^2}{t} = \left(\frac{\varepsilon^2 A^2 r}{2}\right) \frac{\rho \gamma_{LV}}{\nu} \cos\Theta$$
(5-5)

Because density of water is 1, it can be assumed that $\rho = \text{const}$ and

$$\frac{w^2}{t} = \left(\frac{\varepsilon^2 A^2 r \rho}{2}\right) \frac{\gamma_{LV}}{\nu} \cos\Theta$$
(5-6)

Therefore

$$\frac{w^2}{t} = K \frac{\gamma_{LV}}{v} \cos\Theta$$
(5-7)

The above equation (5-7) shows that an increase in weight due to the capillary raise is proportionally related to the contact angle, when the samples are tested using the same conditions and the same wetting liquid. Therefore, plots of increasing mass of the sample versus time are considered a good indication of wettability characteristics for tested samples.

5.2 Equilibrium moisture

Equilibrium moisture indicates the water-holding capacity of the coal sample and was determined following ASTM D1412-93. Equilibrium moisture was determined on fines (-0.5 mm) for each tested coal.

An air-dried coal sample (20 g) was placed in a 250 ml Erlenmeyer flask and 100 ml of boiled distilled water was added to the sample. Then the coal was mechanically shaken for 30 minutes, after which the flask was placed in the constant-temperature bath. In this procedure, a vacuum oven was used instead of water bath and the temperature was set at 30° C. The samples remained in the oven for a wetting period of 3 hours. After the wetting stage, samples were taken from the oven and the excess of water was removed from the coal by filtering through a Micropore filtration unit connected to a water pump. After filtration, the sample was mixed thoroughly and about 5 grams of coal was transferred to the designated filter paper and placed in the glass weighing dish of known weight. The wet coal sample had to be spread evenly to form a uniform layer in the weighing dish. Prepared in this way, the sample was placed uncovered in the vacuum oven containing a saturated K₂SO₄ solution to maintain the relative humidity of 96-97 %. The vacuum oven was evacuated to about 30 mm Hg and the samples were left there for 48 hours to equilibrate.

After equilibrating, the coal samples were removed from the vacuum oven and immediately covered and weighed to the nearest 0.2 mg. Immediately after uncovered weighing dishes were placed in the oven to determine their moisture content. The coal samples were dried at 105 °C for $1^{1}/_{2}$ hour, removed and cooled down over H₂SO₄ vapors in a desiccator, and then weighed. The difference between the mass values of the wet coal and the dried samples gives the value of moisture referred to as the equilibrium moisture. For each tested coal two samples were prepared to have the duplicate values. The reproducibility between duplicates was very good and did not exceed 0.3 % for all the tested samples.

5.3 Specific surface area and porosity

Surface area was determined by the Quantachrome Autosorb Automated Gas Sorption System using the BET (Brunauer-Emmett-Teller) method with N_2 as an adsorbate at 77.35° K. The samples were dried at 120°C overnight in a vacuum oven and then outgassed at 50 °C. The BET isotherms were obtained from 21 point adsorption-desorption analysis, the BET surface area was calculated according to the BET equation as follows:

$$1/W((P_0/P) - 1) = 1/W_mC + (C - 1)/W_mC(P/P_0)$$
(5-8)

where: W is the weight of gas adsorbed at a relative pressure, P/P_{0} , and W_{m} is the weight of the adsorbate (N₂) forming the monolayer on the surface of coal (adsorbent). The C is the BET constant, and is related to the energy of adsorption between the adsorbant and adsorbate and it indicates the magnitude of their interactions.

The BET plot is linear in the range of 0.05 to 0.35 of relative pressures and its slope and intercept are used to calculate the weight of the monolayer W_m using the Equation (5-8). The intercept *i* is $1/W_mC$ and slope *s* is (C-1)/ W_m , while both of these parameters can be read from the BET plot, the W_m can be calculated from the correlation between *i* and *s*. Hence, the surface area is computed according to the formula:

$$S_{t} = W_{m} N A_{cs} / M$$
(5-9)

where: S_t is the total surface area; N is the Avogardo's number $(6.023 \times 10^{23} \text{ molecules/mol})$; A_{cs} is the cross-sectional area of the adsorbate molecule and M is the molecular weight of the N₂. The A_{cs} for liquid nitrogen (at 77K) is 16.2 Å. The specific surface area is obtained as the ratio of S_t to the sample weight (*w*).

$$S = S_t / w \tag{5-10}$$

Porosity can be obtained from gas adsorption studies, the total pore volume is derived from the amount of vapor adsorbed at a relative pressure close to unity, by assuming that the pores are filled with liquid adsorbate. The pore size distribution is the distribution of pore volume with respect to pore size. The desorption branch of the isotherm is used for deriving the porosity calculations.

The total surface area of pores (pore volume) in this study was derived using calculations from the Density Functional Theory (DFT). Micropore volume, with its equivalent surface area was computed from the Brauner MP method (Lowell and Shields, 1998). According to Lowell and Shields (1998), DFT method provides a much more accurate approach for pore size analysis. This method apparently is the closest one in bridging the gap between the molecular level and macroscopic approaches. In this method, adsorption isotherms are determined based on the intermolecular potentials of the fluid-fluid and solid-fluid interactions. The relationship between isotherms is determined by these microscopic approaches and the experimental isotherm on a porous solid is interpreted in terms of a Generalized Adsorption Isotherm (GAI). The GAI equation assumes that the total isotherm consists of a number of individual single-pore isotherms multiplied by their relative distribution, over a range of pore sizes. It is also the most appropriate approach for the range of pores found in coals with the use of N₂ at 77K for the adsorption analysis.

The Brunauer MP method is an extension of de Boer's *t*-method for micropore analysis in the presence of mesopores (Mikhail, et al., 1968). In this technique, the measurement of N_2 adsorbed by the sample is extended to the higher relative pressures to permit the calculation of the surface area in the non-microporous part of the material. The *t*method uses a *t*-curve obtained from the analysis isotherm. The standard *t*-curve is expressed by the empirical de Boer's equation (de Boer et al., 1966):

$$t = [13.99/(\log P_0/P + 0.034)]^{1/2}$$
(5-11)

where: *t* is the statistical thickness of the adsorbed layer in Angstroms (Å), P and P₀ are the gas pressures initial and at each point of measurement during the analysis.

The calculation of t from this equation at various relative pressures is used to replot the analysis isotherm as a *t*-curve, a plot of the volume of gas adsorbed versus t. The *t*-plots are the straight lines intercepting the adsorption axis (adsorbed volume) at the equivalent to the micropore volumes. In the MP method, volumes of adsorbed gas are converted to N₂ liquid volumes from which t is calculated according to the equation:

$$t = V_{\text{liq}} / S_{\text{BET}} \ge 10^4 \,(\text{\AA})$$
 (5-12)

where: V_{liq} volume of the liquid adsorbed; S_{BET} surface area from BET measurement.

The V-*t* plot is constructed by plotting V_{liq} versus *t* (Å); where *t* is taken from *t* versus P/P₀ plot, in the relative pressure intervals of 0.05. The calculation of the surface area of pores is accomplished through the drawing tangents to the incremental points on the V-*t* curve between the *t* values corresponding to the pore sizes (e.g. 0 to 4Å, 4 to 4.5 Å, 4.5 to 5Å etc). For the microporosity, the *t* thickness of the adsorbed monolayer of gas is the actual measure of the pore size. The surface area values are obtained from the slope of these tangent lines for these pore size intervals. The calculations are continued until no further decrease is observed in the slope of the V-*t*, which means that all the pores are filled. From the pore surface area, the pore volume is calculated and distribution is obtained in a similar manner for each point on the V-*t* plot. The advantage of the Brunauer MP method is that it has the ability to obtain the micropore volume, surface area and their distributions from one experimental isotherm. The MP method is applicable to surfaces (adsorbents) containing macro-, transitional (mesopores) as well as micropores. The Brunauer's and de Boer's method, both assume that the N₂ BET measured surface area is valid for all micropores present in the absorbent.

5.4 Pelletization as a method to assess particle to particle interaction

Pelletization was used as a method of characterization of the particle-particle interaction in fine coal (-0.5 mm). The same operating conditions were used for testing all samples. Approximately 1 kg of air dried coal was placed in pelletizing disc (Syndron) rotating at 25 rpm and sprayed with water at a rate of 10 ml/min. The pelletizing dish is shown in Figure 5.4-1. The amount of water added to each sample was calculated from the equation given by Sastry and Fuerstanau (1982):

$$M_{\rm w} = 44.0 \, \exp(-0.017 A_{\rm c}) \tag{5-13}$$

where: M_w is the moisture required for pelletization, A_c is the ash content on dry basis. This equation is applicable for all types of coals except lignite, irrespective of their rank and origin. Since the addition of water for each coal depends on the ash content, the duration of pelletization run was adjusted accordingly. The pelletization tests were carried out until suitable pellets were formed; for most coals, the pelletization was completed within 30 to 35 minutes. The pellets were then collected and their strength was measured using a Tritester tensiometer. The compressive strength of a single pellet was measured by crushing the pellet between flat parallel surfaces; the loads at which failure occurred was recorded. The rate of loading was digitally set at 0.75 mm per minute. The pellets' strength for all tested coals and the associated statistics of these calculations are given in Appendix D.

5.5 Particle shape and roughness estimation

Particle shape was determined using the optical microscope Olympus BX60 with a total magnification of 500x. To analyze particle shape characteristics of fine coal particles (-0.5 mm fraction), the two-dimensional ratio of d_{min}/d_{max} was used to calculate the aspect ratio. In this method, coal pellets were prepared for microscopic examination and coal particles were randomly chosen while scanning the pellet for measurements. Due to the random position of these coal particles, the ratio of minimal to maximal diameter (d min/d max) was used to compute the aspect ratios.

The BET total surface area and pore surface area, calculated from pore volume using the DFT method (Lowell, 1998), were used to estimate roughness. For each sample, the BET specific surface area and calculated pore surface were compared. The difference between these values indicates the external surface beyond the pores, thus representing roughness and the macropore contribution to the total surface area of fine particles. In these calculations, the pores are considered as the cavities where depth is greater than width. Therefore everything else is considered to be the roughness of the particle surface. The calculations for each of the samples are presented and discussed in the result section. The DFT method calculates pore volume for pores which do not exceed 8 nm (80 Å) in diameter.



Figure 5.4-1 Syndron pelletizing dish.

Another measure of the particle roughness is the ratio of external surface area of the particle to its equivalent sphere surface area (Bikerman, 1970). It is called rugousity and is given by:

$$R = S_{ext} / S_{sph}$$
(5-14)

The larger the R number, the more irregular, non-uniform the particle surface. The S_{ext} is the surface area, calculated as the difference between the total surface area derived from BET (S_{BET}), and surface area of pores (S_{DFT}) obtained from the DFT method. The S_{sph} is the surface area equivalent to the surface of particles of the same volume, but assumed to be spheres. The sphere always assumes the smallest surface area of the particles for the same volume. Malvern 2000 size analyses were used to estimate the external surface area of particles, which are assumed to be spherical. Rugousity can be influenced, however, by both the shape and roughness of the particle. To account for these effects, aspect ratio and fractal dimension were compared for these particles.

The surface roughness topography is known as fractal dimension, D. Real surfaces are irregular and these irregularities are referred to as fractals because their magnitude is proportional to X^D ; where X is dimension and D is a fractional exponent that generally assumes the value between D=2 for smooth surfaces and D=3 for very rough surfaces.

The Neimark-Kiselev (NK) method was used to determine the fractal dimension of studied coal fines. This method uses a single gas sorption isotherm for its calculations, and combines the thermodynamic and fractal arguments (Neimark, 1991). It assumes that the cumulative surface area is equivalent to the measurable adsorbent area, with a size proportional to the mean radius of curvature of the adsorbate-vapor interface within the multilayer region of isotherm. The fractal dimension values were calculated for all the coal samples from the N₂ adsorption isotherm using the Quantasorb software.

5.6 Mineral matter characterization

Microscopic examination of the coal fines was performed using the optical microscope Olympus BX60 with the total magnification of 500x. Fines (-0.5 mm) from each coal were used to prepare pellets. The pellets were made using epoxy, polished and used for microscopic examination. Examinations were carried out to find out what type of mineral matter is present in the coal and to assess the association of mineral matter with coal particles. The results of the microscopic examination are presented in the Table 6-6 in the Result and Discussion (Chapter 6).

A low temperature ashing method was used to obtain mineral matter in the form as it originally exists in coal. In this method, as discussed by Gluskoter (1965), Rao and Gluskoter (1973), the oxygen gas is activated through the use of a radio-frequency generator which reacts exothermally with the organic matter, ashing the coal at a temperature between 130° and 170°C. At this temperature, most mineral species in the coal (representing ash) are unaffected. For the low-temperature ashing, the coal samples were ground to -0.177 mm, and dried at 105°C for 30 minutes. About 5 grams of coal was spread on a shallow glass dish and placed in the asher. Samples were ashed using the Low Temperature Asher LTA-504 by LFE corporation. The samples were left for ashing for at least one week. The samples were stirred twice during the course of the ashing period. When the ashing was completed, the samples were removed and ground to -0.074 mm and

stored in glass vials. Each sample was prepared in duplicate to ensure a sufficient amount for XRD analysis.

For X-ray Powder Diffraction analysis, samples were ground into a fine powder to the grain size range less than 10 microns. Step-scan X-ray data was collected over a range $3-70^{\circ}2\theta$ with CuK α radiation on a standard Siemens (Bruker) D5000 Bragg-Brentano diffractometer equipped with diffracted-beam graphite monochromic crystal, 2mm (1°) divergence and antiscatter slits, 0.6 mm receiving lit and incident-beam Soller slit. The long fine-focus Cu X-ray tube operating at 40 kV and 40 mA, using a take-off angle of 6°.

The X-ray diffractograms were analyzed by the International Centre for Diffraction Database PDF-4 using Search-Match software by Siemens (Bruker). The X-ray powderdiffraction data was refined with Rietveld Topas 2.1 (Bruker AXS). The results of quantitative phase analysis by Rietveld refinement represent the relative amounts of crystalline phases normalized to 100 %.

Liberation studies were carried out to determine the amount of liberated mineral matter in coal fines of the tested samples. Approximately 3 grams of air dry coal (-0.5 mm) was put into each of 100 ml vials filled with a heavy organic liquid (Perchlorethylene) prepared at 1.8 g/ml density and inserted into the centrifuge. Samples were centrifuged for 10 minutes at 7000 rpm in the Sorval-Biofuge Primo centrifuge. After the centrifuging was completed the vials with coal samples were removed and were let to stand for 30 minutes to stabilize. After that, the floating fraction was gently removed with a small pipette, so that the separation between the phases not to be disturbed. The liquid was then slowly decanted from the vials and the sink material was filtered.

Removed floats and sinks were then washed with acetone, to remove traces of organic liquid and then dried in an oven at 105°C. After drying, the float and sink fractions were weighed and their weights recorded for calculations. The amount of liberated mineral matter in each of the coal samples was then calculated on the weight percentage basis.

5.7 Assessment of handling properties

5.7.1 Angle of repose

The angle of repose is a simple method used in powder mechanics to estimate the flow property of the dry solid. It is defined as the angle to the horizontal plane assumed by a cone-like pile of the material dropped from the constant height (Carr, 1969; Brown and Richards, 1970). When a pile of fines is formed, the angle of inclination to the horizontal of its free surface may assume a value up to a maximum angle. The pile is carefully built by dropping the material from a point above the horizontal plane using the setup as shown in Figure 5.7.1-1.

In this test approximately 30 grams of -0.5 mm fine coal was used to build a cone shaped pile for each tested coal. The samples were tested at increasing moisture content. Tests were carried out in duplicate for each moisture level. First tests were performed on a dry basis, then the moisture content was increased incrementally by 2 to 5 % up to 20 % moisture in each coal sample.





In each run, four measurements were taken on each side of the formed pile. After the test a small amount of coal was taken for moisture determination. Duplicate runs were performed immediately after the first run. The average value of the two measurements was used for plotting the results. The standard deviation of the duplicate values varied from 1° to a max 4° . The standard deviation of the 4 measurements for each test was in the range from 1° to the 5° max. The highest standard deviations were always for the tests at the intermediate moisture content (5 %-15 %).

5.7.2 Bulk density

The bulk density was determined for three samples. These were bulk coal samples: LC 8U and LC 3 and LC3 OXY. The LC 3 and LC 8U samples were tested with different amounts of fines (10 %, 20 % and 30 %), and at increasing moisture levels. LC3 OXY was tested at 33 % fines content. Both packed and aerated bulk densities were determined for each sample at each moisture.

The container used for bulk density determination was made of Plexiglas in the form of a large cylinder. The cylinder dimensions were: height H = 48.8 cm; Diameter D =25cm; L= H₀ or H₁; where: H₁, H₀ are the heights corresponding to the packed and loose (aerated) level of the coal sample. Figure 5.7.2-1 illustrates the container used to measure the bulk density for the selected samples



Figure 5.7.2-1 Bulk density container with the lid. The dimensions are: H=48.8 cm, r=12.5 cm.

The Durham Cone container was used for compacting the coal sample before the bulk density determination. For zero conditioning, the coal sample was placed into the Durham Cone container and vibrated, flowing out of the container into the cylinder. Once in the designated cylinder, the sample was leveled off using a specially designed lid and the heights of the coal in the cylinder were recorded. At least 8 measurements of height were taken around the cylinder in equal spaces. Their values were then averaged and the BD_o value was assigned as the aerated bulk density.

For packed density, coal sample was placed in the Durham Cone and vibrated for 3 minutes, after which the coal was allowed to flow out of the cone into the Plexiglas container. The heights were measured in the same manner as for the aerated density. The packed bulk density was referred to as BD₁.

5.7.3 Durham Cone tests

The handleability of bulk samples was tested with the use of a Durham Cone as described in the original work by Cutress et al., (1960). For each test 10 kg of coal was prepared and placed into the cone-shaped container. The cone was vibrated for 30 seconds and then the coal was allowed to flow out through an opening of 100 mm in diameter. The time required to discharge the coal from the vibrating cone was measured, to calculate the flow rate in kg/s. In these experiments the handleability index was expressed as the flow rate in kg/s and referred to as the Durham Cone Index (DCI). The test was repeated at least 8 times and the average value of flow rate was calculated.

Each of the coal samples used in the study was thoroughly mixed prior to the testing. Mixing was performed by the "cone and quarter" method followed by shaking in rotational movement by hand for about 20 times in the sealed plastic bag, prior to loading the sample into the cone. After each 30 second test, the sample was re-loaded into the cone and runs were repeated until the completion of the series. Segregation of coal particles was observed as the cone was vibrated. There was no mixing in between runs, to preserve the segregation patterns for each series. The testing continued for at least 300 seconds while repeating runs every 30 seconds. In each series 8 to 16 runs were performed. The flow rates were plotted versus cumulative time of the whole series of tests. Each point on the curve represented a separate run carried out every 30 seconds. As a result, the patterns of flow rates could be obtained for varying moisture content or other testing conditions.

Samples were tested using at least three different moisture contents. Every single point representing flow rate at a particular moisture level was calculated as an average

value for at least 8 separate runs. Selected samples were tested at varying amounts of fines. The Durham Cone tests were performed at two different locations: initially at the University of Nottingham and the second series of tests were performed at the University of British Columbia, (UBC) Mining Department. Figure 5.7.3-1 presents Durham Cone built at UBC Mining Department.



Figure 5.7.3-1 Durham Cone used at UBC Mining Department for handleability tests.

5.7.4 Handleability Monitor tests

The Handleability Monitor as described in detail by Brown and Atkin (2000) has been used to measure the compressive strength of selected samples. In each test, approximately 50 kg of coal was loaded into the hopper. With the full hopper, a hydraulic ram was operated; extending and retracting three times in pre-run cycles (three cycles).

Measurements were taken during the following five cycles vs. time. The pressure exerted by the ram was recorded at intervals of 0.2 seconds. For each coal sample, five graphs (traces) of pressure vs. time were obtained, and subsequently combined producing a single pressure-time graph. The average value of the maximum pressure P (max) obtained from "Pmax-Time" trace is considered to be a measure of coal handleability. Since the value of the Handleability Monitor Index is expressed (in bars) as the Average Maximum Pressure, the AMP is used as the handleability parameter. An example of the Handleability Monitor data is shown in Appendix G. Figure 5.7.4-1 shows the schematic representation of the instrument. The photograph in Figure 5.7.4-2 demonstrates discharging coal from the Handleability Monitor. Figure 5.7.4-3 presents a typical trace graph obtained for the Handleability Monitor run.

The handleability tests using the Handleability Monitor were performed at the University of Nottingham, UK. The tests were carried out on some random coal samples, as well as specially preselected coal samples. When the comparisons between the Durham Cone and the Handleability Monitor tests were made, the Handleability Monitor tests were always carried out prior to the tests using the Durham Cone. In such tests the same coal samples were subjected to both tests.



Figure 5.7.4-1 Schematic sketch of Handleability Monitor (Brown and Atkin, 2000); reproduced by permission of Taylor and Francis Group.



Figure 5.7.4-2 Handleability Monitor discharge



Figure 5.7.4-3 Typical trace plot of the measurements obtained using Handleability Monitor (Brown and Atkin, 2000); reproduced by permission of Taylor and Francis Group.

CHAPTER 6. RESULTS AND DISCUSSION

6.1 Coal fines characterization

The fines of five raw coal samples and two clean coal products were used in the pelletization tests. The metallurgical coals were: LC3, LC 10BC and LC 10B; LS20 and LC 8U were thermal coals. The LC 3 coal was oxidized under laboratory conditions and used for testing as LC 3OXY. Two clean coal products were LC met and LC thermal.

One set of raw coal samples was crushed to 100 % passing 6.3 mm, as discussed in Chapter 4, section 4.2. For surface properties characterization, a -0.5 mm size fraction was sieved out of the crushed sample and used for evaluations. The clean coal products were used as-received and a fine fraction of -0.5 mm was also separated from the bulk sample for the surface properties characterization.

Another set of samples included coals used in the series of tests with the Handleability Monitor as well as with the Durham Cone, and the surface properties of fines from these coals were also studied in terms of wettability. The metallurgical coals were from Australia (coals #11 and #12), and the thermal coals were from the UK (coals #2 #4 and #5). While surface properties such as penetration rate and equilibrium moisture were determined for these samples, the N₂ adsorption and porosity were not measured, as these samples were not used in the pelletization tests.

The coal analyses are given in Tables 6.1-1 and 6.1-2 for both sets of tested coals. The equilibrium moisture and transmittance values are included to demonstrate different hydrophobicity (wettability) characteristics of the selected coal fines.

6.1.1 Wettability assessment

6.1.1.1. Penetration rate

The results of the penetration rate for the two sets of coals are presented in Figure 6.1.1-1, depicting penetration rates for the following coals: LC 3, LC 10B, LC 10BC, LC 8U, LS 20 and LC met, LC thermal. Figure 6.1.1-2 presents the penetration trends for product coals; Coal #2, 4, 5. 11 and 12. The mass increase is expressed in the form of squared mass, as provided in equation (5-7). Each graph is plotted as the squared mass vs.

time, the tested coal samples were compacted under the same 5.5 kg load in the preparation stage.

From the results, it is clear that hydrophobic samples are characterized by a very small mass increase versus time and equilibrate after a relatively short time, while for hydrophilic coals the mass increase is significant, especially in the first few seconds. The most hydrophobic coals are: LC 3, LC 10BC followed by LC met and LC 10B being somewhat less hydrophobic and more wettable, respectively. In Figure 6.1.1-2 the penetration rates are presented for Coal #11 and #12 (the most hydrophobic), followed by Coal #4, Coal #2 and Coal #5 (the most hydrophilic).

Coal	Yield of (-0.5 mm) (wt %)	Ash in fines (AR*, wt %)	Ash in bulk sample (wt %)	Equilibrium moisture %	Transmittance %
•LC 3	36.0	12.61	30.12	1.30	95.25
•LC 10BC	39.0	24.41	37.45	1.65	80.90
■LC 10B	45.0	12.94	24.30	2.96	56.24
■LC 8U	37.0	15.37	23.63	7.34	26.06
∎LS 20	22.0	33.7	35.00	8.02	34.80
•LC met	44.0	12.66	9.77	1.56	99.02
■LC thermal	45.0	15.86	15.15	2.09	50.63
LC 3 OXY	36.0	12.61	30.12	4.38	57.80

Table 6.1-1 Characterization of fines from; LC 3, LC 10BC, LC 10B, LC 8U, LS 20, LC thermal and LC met, and LC 30XY coals.

•- hydrophobic coal; • – hydrophilic coal; * AR as received

Coal	Yield of (-0.5 mm) (wt %)	Ash in fines (wt %)	Ash in bulk sample (wt %)	Equilibrium moisture (%)
■Coal # 2	8.5	14.87	11.70	4.43
∎Coal # 4	19.3	34.1	20.8	5.23
∎Coal # 5	12.5	23.6	11.5	1.98
●Coal # 11	40.9	8.95	9.67	1.51
●Coal # 12	27.3	15.13	10.46	1.91

Table 6.1-2. Characterization of fines from coal products; Coal #2, #4, #5, #11 and #12.

●- hydrophobic coal; ■ – hydrophilic coal

In a separate series of experiments, each coal sample was packed in a tube and compressed by a column-packing device under different loads (5.5 kg and 11 kg) and an additional sample was always packed without any load to show the packing effect on the wettability of these samples. The two samples which showed a significant effect of packing on the penetration rates were LS20 (very hydrophilic) and LC 3 (very hydrophobic). For both these samples, an increase in load (greater compaction) led to a decrease of wettability, as more compaction meant smaller voids between particles thereby reducing the ability for water to penetrate through the material. The exceptions were that for hydrophilic coal LS20, loosely packed and packed under a 5.5 kg load sample, both samples having the same high penetration rates; while loosely packed LC3 coal sample having the highest penetration rate, and LC 3 coal packed under 5.5 and 11 kg had similar, much lower penetration rates. For all remaining samples, the changing of the penetration rate with an increased load either had no effect or followed patterns observed for LS20 and LC3. Figure 6.1.1-3 and 6.1.1-4 illustrate penetration rates for LS20 and LC3 respectively, under increasing compacting load. Penetration rates for other tested coals are included in Appendix B.



Figure 6.1.1-1 Penetration rates for LC3, LC10B, LC10BC, LC8U, LS20, LC thermal and LCmet coals.



Figure 6.1.1-2 Penetration rates for Coals #12, #11, and Coals #4, #2, #5.



Figure 6.1.1-3 Penetration rates for LS20. The effect of increasing compacting load.



Figure 6.1.1-4 Penetration rates for LC 3. The effect of increasing compacting load.

The fact that LS20 was found to be affected by packing only at very high load may be related to its highest grindability index HGI (Table 4.1-1). Consequently, harder

particles would require more pressure to compact them. All the other samples were characterized by much higher but very similar HGI values ranging from 74 to 83 (50 for LS 20; 80 for LC 10BC; 74 for LC 8U; 83 for LC 3 and LC 10B). For the LC 3 sample, it may be reasonable to assume that less packing among hydrophobic particles leads to slightly higher penetration rates, as the water is only able to penetrate through the channels in between the particles. For other tested coals (#2, #4, #5, #11 and #12), the observed trends were as follows: for hydrophobic coals there was no effect on wettability with increased compaction pressures, while for hydrophilic coals greater compaction led to lower penetration rates.

An important conclusion from this is that consolidation of particles may be a significant factor affecting wettability characteristics of a handled coal sample and therefore it is critical that preparation procedures are uniform for all tests, especially in the sample preparation part. However, the way a sample packs into certain confined volume may be related to its physical characteristics, such as size distribution of particle, and particle shape.

6.1.1.2 Transmittance

Transmittance was determined on coal fines (-0.5 mm). The transmittance values for tested coals are presented in Table 6.1-1 along with other sample characteristics. According to transmittance values, the most hydrophobic or the freshest (non-oxidized) coal appear to be LC met, metallurgical coal product sample; followed by LC 3 then LC 10BC coal. Significantly oxidized are LC 3 OXY (57.8 %) followed by LC 10B (56.24 %), then LC thermal (50.63 %) and finally the two most oxidized and least hydrophobic are LC 8U and LS20 with the transmittance values of 26.06 % and 34.80 %, respectively.

The correlation between equilibrium moisture and transmittance is presented in Figure 6.1.1-5. The equilibrium moisture is strongly correlated with the oxygen functional groups content and surface area as described by Kaji et al., (1986) and this aspect of the study will be discussed in greater detail in the following sections of this chapter.



Figure 6.1.1-5 The correlation between equilibrium moisture and transmittance values for studied coals.

6.1.2 Specific Surface area, porosity and equilibrium moisture

Specific surface area was determined on coal fines (-0.5 mm), which were used in the pelletization study. The surface properties of the studied coals are summarized in Table 6.1-3. The BET surface area and porosity data for the studied coals are similar for most of the samples and in agreement with the rank of these coals. An increase in the total surface area and pore surface area (recalculated pore volume) are reported for LC 8U-heavily oxidized sample and LS 20-lower rank coal.

The adsorption and desorption isotherms were obtained for all the tested samples. The shape of the adsorption isotherm indicates the porosity of the adsorbent. All of the tested samples conformed to a Type II isotherm. This type of isotherm is characteristic of the macro- mesoporous adsorbent, with a minimal contribution from micropores to the total surface area, representing an unrestricted monolayer-multilayer adsorption. Coal samples LC 3, LC 10BC, LC 10B, LC 30XY, LC met and LC thermal displayed almost identical characteristics in terms of N₂ adsorption onto their surfaces, with minimal hysteresis. Figure 6.1.2-1 and Figure 6.1.2-2 illustrate the isotherms for LS20 and LC8U. The LS20 isotherm shows considerable hysteresis as compared to other samples, the hysteresis was also observed for LC 8U sample. When comparing these two samples, one can presume that

desorption from the LS20 coal surface is much greater due to the larger microporosity contribution to its total surface area (11.8 % of porosity derived from pores up to 2 nm (20 Å) when compared to the LC8U (2.8 % in pores up to 2 nm (20 Å)). The small capillaries are enhanced energy sites for adsorption, due to their overlapping wall potentials. According to Thommes (2005), pore filling by an adsorbate in the micropores less than 2 nm in size (20 Å) occurs continuously, whereas in the case of mesopores (2 to 50 nm (20 – 500 Å)), pore filling occurs by pore condensation.



Figure 6.1.2-1 The BET isotherm for N₂ adsorption on LS 20 coal.

Although the microporosity for LS20 is 11.8 % and 2.8 % for LC8U, with the mesopore contribution to the total surface area for both samples being almost the same, in the LS20 sample, however, pores spread out over a considerably larger surface area. They thus become a significant factor in the desorption mechanism for this sample.

The total surface area for each sample was obtained from BET plots, while the pore surface area was derived from the Density Functional Theory (DFT) method and the micropore surface area from the Brunauer MP method (Mikhail et al., 1968) as described in Chapter 5. The DFT method was used to calculate porosity because it is known to be the most suitable to measure the surface area for materials containing both micro- and mesopores (Lowell and Shields, 1998; Thommes, 2003). The method was shown to be highly reliable in the systems where N_2 is used as an adsorbate and applicable for pores with diameters in the range 0.35 nm to 8 nm (3.5-80 Å). The porosity results for tested coals, as derived from DFT method, are provided in Table 6.1-3.

The pore volumes, which are customarily used in porosity evaluations, were converted to the pore surface area, as it is easier to note the contribution of porosity to the total surface area expressed in m^2/g . The contribution of 8 nm pores (< 80 Å) to the total surface area is somewhat similar for all the samples (55 to 58 %) with the exception of the LS 20 sample, where the contribution is above 65 % (Table 6.1-3). In order to further discuss the porosity results, it is necessary to introduce the pore classification system. The classification of pores according to the pore sizes was described by Lowell and Shields (1998) as follows:

- 1. micropores are pores with diameters not exceeding 2 nm (20 Å);
- 2. pores of intermediate size are called mesopores 2 to 50 nm (20 to 500 Å)
- 3. pores greater than 50 nm (500 Å) in size are categorized as macropores



Figure 6.1.2-2 The BET isotherm for N₂ adsorption on LC 8U coal.

In line with this classification, the pores which are measured by the DFT method include micropores and to some extent the mesopores, while the microporosity derived from the MP method assessed only pores up to 2 nm (20 Å) in size. The difference between surface area of pores from the DFT method and the MP method provides the surface area contribution from the mesopores ranging from 2 to 8 nm in size (20-80 Å), as presented in Table 6.1-3.

An attempt was made to calculate the macropore surface area from the difference between the BET surface area (S_{BET}) and the surface area obtained from the DFT method (S_{DFT}). According to the type of isotherms derived from the N₂ adsorption analysis, the total surface area for these samples is not limited to the pores; part of the surface area is associated with the external surface area of individual particles. Thus a contribution of macropores is also interrelated to the external surface area of particles.

The coals examined here are of intermediate-higher rank (medium volatile), with the exception of LS 20, which is a high volatile, low rank coal. From the BET adsorption tests, it is evident that only two samples have a considerably higher BET surface; LS 20 being of lower rank coal and LC 8U, a heavily oxidized coal. Mesopores ranging from 2 to 8 nm (20-80Å) are major contributors to the total surface area for practically all of the samples, while the contribution from macropores may be implicated by the external surface area of the particles. The latter could not be determined from the present surface area study. It is assumed that for the higher rank coals, macroporosity is not a significant factor with the exception perhaps of LS 20 or LC 8U. For the four samples LC met, LC 3, LC 10BC and LS 20, microporosity appeared to be enhanced, while for LS 20, alone this could have a major impact on the overall surface properties, since its surface area is considerably greater than the surface area of all three remaining samples.

6.1.3 Summary and discussion

Water adsorption is controlled by surface area, porosity and wettability of the coal surface. The amount of water adsorbed onto the coal under saturation conditions determines the value of equilibrium moisture. In the work by Mahajan and Walker (1971), as discussed in Chapter 3, the moisture sorption isotherms were measured at 20^{0} C and at an increasing relative pressure (p/p₀; water pressure to the water saturation vapor pressure). It was shown

that the amount of water adsorbed onto the coal at a high humidity (close to the saturation pressure $p/p_0=1$) is consistent with coal porosity. The shape of the sorption isotherm was related to the porosity, as well as the functional groups content, which was especially true at the low values of p/p_0 (Laskowski, 2001).

Coal	BET total surface area (m ² /g)	BET total surface area (m²/g)Pore surface area in pores upto 8 nm (80Å) DFT method (m²/g)Micropore 		Mesopore surface area in pores 2 to 8 nm (20 to 80 Å) $S_{DFT} - S_{MP}$ (m ² /g)	Macropore and external surface area $S_{BET} - S_{DFT}$ (m ² /g)
•LC 3	0.390	0.222 (56.92) *	0.048 (12.3) [*]	0.174 (44.61)*	0.168 (43.1)*
•LC 10BC	0.652	0.360 (55.21)	0.069 (10.6)	0.291 (44.61)	0.292 (44.8)
∎LC 10B	0.376	0.207 (55.05)	0.013 (3.5)	0.194 (51.6)	0.169 (44.9)
■LC 8U	1.440	0.793 (55.07)	0.040 (2.8)	0.753 (52.3)	0.647 (44.9)
∎LS 20	4.144	2.712 (65.44)	0.491 (11.8)	2.221 (53.6)	1.432 (34.5)
■LC 3 OXY	0.274	0.158 (57.66)	0.019 (6.9)	0.139 (50.7)	0.116 (42.3)
•LC met	0.439	0.251 (57.18)	0.149 (33.9)	0.102 (23.2)	0.188 (42.8)
■LC thermal	0.661	0.367 (55.52)	0.051 (7.7)	0.316 (47.8)	0.294 (44.4)

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●- hydrophobic coal; ■ – hydrophilic coal

*(% contribution to the total surface area)

This means that an increased value of equilibrium moisture should be consistent with porosity, as well as with the wettability characteristics of coal samples. Table 6.1-1 presents results of equilibrium moisture and transmittance determinations on the studied samples. The correlation between equilibrium moisture and transmittance values are in relatively good agreement as shown in Figure 6.1.1-5. The greater the degree of oxidation, the more hydrophilic the surface, the higher the equilibrium moisture in the fines. The penetration rates for both type of coals (Figure 6.1.1-1) are also consistent with the equilibrium moisture and transmittance values.

The highest equilibrium moisture 8.02 % and 7.34 % for LS 20 and LC 8U samples respectively is linked with the largest measured specific surface area and considerable wettability for these samples. The specific surface area for LS 20 is much larger than for LC 8U and the microporosity of LS 20 is more extensive than that of LC 8U, therefore one should anticipate that equilibrium moisture for LS20 should be noticeably higher than 8.02 %. The fact that the equilibrium moisture and penetration rates are consistent indicates that under normal conditions (atmospheric pressure), micropores may be filled with water. This may be a result of water condensation in micropores, especially in the case of strongly hydrophilic surfaces. This type of water is referred to as a bound water and adheres strongly to the micropores, behaving almost as if it was chemically bound to these small capillaries. The existence of such capillary-bound water was discussed by many authors (Gauger, 1947; Bond et al., 1948; Sato and Suzuki, 1982; Colin and Gazbar, 1995). As a result, any additional physical adsorption of water occurs only into large pores and to the external surface of particles.

This capillary water is very difficult to remove, unless the sample is subjected to a high vacuum degassing or extreme thermal treatment. The coal samples which were used for porosity measurements (N₂ adsorption tests), were dried at 120° C and degassed at 10^{-6} mm of Hg vacuum pressure. It can be assumed that the porosity measured on such highly degassed surfaces, has had micropores voided of capillary-bound water as opposed to the surfaces of the samples which were used for the penetration test or the equilibrium moisture measurements. The micropore contribution to the total surface area of LS 20 is almost four times greater than that in LC 8U, and the total surface area of LS 20 is almost three time as large as in LC 8U. Hence, if such a large surface area (micropores) becomes unavailable in LS 20 for water adsorption under normal conditions, it is possible that the equilibrium moisture moisture for this sample is distorted and as a result its value may be much smaller.

For other samples, an increase in equilibrium moisture was not related to the porosity, unless it was linked with the increase in wettability, e.g. LC 3 and LC 10BC, or LC 10B and LC 8U, LC 3 and LC 3OXY, etc., as shown in Tables 6.1-1 and 6.1-3. Kaji et al. (1986), showed that there is a good correlation between equilibrium moisture, the product of oxygen content, and specific surface area of coal. This indicates that wettability is indeed a necessary condition for water adsorption on to the coal surface, and surface properties such as surface area and porosity become fully effective only when the coal is hydrophilic. Equilibrium moisture is an important factor, as it sets the boundary for each coal sample of how much water can be accommodated into the coal surface. The extent of this theory needs to be verified in terms of the behavior of coal particles of different wettability characteristics in the presence of water. For that purpose pelletization was chosen as the method to elucidate the behavior of coal particles in the presence of water, as pelletization relies on the surface properties of the pelletized particles.

6.1.4 Pelletization as a method of assessing surface properties of fine coal particles

The pelletization of coal fines was carried out to test the tendency of coal particles to aggregate. The differences in the behavior of various coal samples in the pelletization tests were visible in the manner the pelletization proceeded. The hydrophilic coal fines were very easy to pelletize while hydrophobic particles were very difficult to pelletize. The rate of pellet growth was also different for these two types of particles. Hydrophilic particles formed pellets in the first few minutes of the test and these pellets were strong enough to withstand destruction during the remaining time of the pelletization procedure. For hydrophobic particles it took a long time to form pellets and resultant pellets were not very strong.

The surface properties which are believed to have an effect on aggregation of fine coal are: surface area, porosity and wettability. In previous studies on pelletization, the effect of porosity and wettability of coals on the pellets' strength have not been studied. In this study, the emphasis is placed on relating the surface area, porosity and wettability of coal particles to pellet strength. The pellet strength is used here as the evidence of interparticle forces acting upon these particles in the presence of water. The pelletization results for five studied coals are presented in Table 6.1-4.

The pellets strength is calculated as the weighted average value of pellets strength according to their size distribution. Considering ash content and different degrees of wettability for the tested samples are important parameters in the strength of produced pellets, one should be able to expalin the pellets' strength values obtained from these pelletization tests. For example, comparing the pellets' strength for LC 3 and LC 3OXY coal; LCmet and LC thermal coal, one can see that stronger pellets are always produced from hydrophilic coals. A higher ash sample (LC 10BC), however, produced only slightly stronger pellets than hydrophilic coal with a lower ash (LC10B). This illustrates that the wettability and ash content of fines are not sufficient to explain the differences in pellet strength. Another example is a comparison between strength of the pellets produced from LS 20 and LC 8U coals. The wettability of both of these coals is essentially identical (Figure 6.1.1-1), the ash content in LS 20 is twice that of LC 8U, while the strength of pellets from LC 8U coal. This significant difference in the strength of the pellets from LS 20 cannot be explained on the basis of wettability nor ash content alone.

Similarly, the strength of pellets produced from LC 10BC and LC met (both hydrophobic coals with different porosity characteristics), could not be explained unless a model of aggregation of particles in terms of wettability, surface properties and porosity is proposed. This model should fill the gap between the generally accepted concepts of pelletization and wettability phenomena and the effect of coal porosity on them. The proposed model of wettability of coal surface and its effect on the coal particle aggregation will be discussed in the subsequent sections of this chapter.

The surface friction and interlocking of particles are also known to contribute to the strength of pellets. These forces depend on the shape and packing characteristics of the individual particles. The higher the irregularity in the shape of the particles, the better is the interlocking of the particles forming pellets, and thus the greater strength of the pellets. This contribution from the mechanical interlocking however is generally considered to be small in comparison to other mechanisms of forming bonds between the particles and building the strength of produced pellets (Capes, 1980). Although wettability is the major cause for water to adsorb onto the surface of a particle, the roughness of the particle may also be a contributing factor when two particles of similar wettability are aggregated. Therefore, to

fully characterize surface properties of fine coal, the particles shape and roughness were also estimated for the studied coal fines and this will be the focus of the next section of this chapter.

Coal	BET total surface area (m ² /g)	Ash in Fines, *(AR) wt %	Micropore surface area in pores upto 2 nm (20Å) MP method (m ² /g)	Mesopore surface area in pores 2 to 8 nm (20- 80 Å) $S_{DFT} - S_{MP}$ (m^2/g)	Macropre and external surface area $S_{BET} - S_{DFT}$ (m^2/g)	Pellets strength (kg/m ²) x10 ⁻⁶
•LC 3	0.390	12.61	0.048 (12.3)*	0.174 (44.61)**	0.168 (43.1)**	8.5
•LC 10BC	0.652	24.41	0.069 (10.6)	0.291 (44.61)	0.292 (44.8)	13.8
■LC 10B	0.376	12.94	0.013 (3.5)	0.194 (51.6)	0.169 (44.9)	11.6
■LC 8U	1.440	15.37	0.040 (2.8)	0.753 (52.3)	0.647 (44.9)	20.0
■LS 20	4.144	33.70	0.491 (11.8)	2.221 (53.6)	1.432 (34.5)	289.0
■LC 3 OXY	0.274	12.61	0.019 (6.9)	0.139 (50.7)	0.116 (42.3)	14.3
•LC met	0.439	12.66	0.149 (33.9)	0.102 (23.2)	0.188 (42.8)	7.5
■LC thermal	0.661	15.86	0.051 (7.7)	0.316 (47.8)	0.294 (44.4)	20.0

Table 6.1-4. Surface properties and pellets' strength of the studied coals.

•- hydrophobic coal; ■ – hydrophilic coal
* AR – as recieved; **(% contribution to the total surface area)

6.1.5 Particle shape and roughness estimation

Table 6.1-5 presents the set of the data pertaining to the external surface area of particles as derived from the BET analysis, from the Malvern particle size analysis and from the microscopic analysis of coal particles used in this study. Particle shape in terms of a two-dimensional aspect ratio of d_{min}/d_{max} and the fractal dimension were also determined.

To estimate roughness, the BET total surface area and pore surface area were used as derived from the DFT method (Lowell, 1998) and discussed in Chapter 5. The difference between the total surface area and surface area of pores up to 8nm (80Å; micropores and partly mesopores) indicates the external surface area beyond the mesopores, thus representing macropores and the contribution of the external surface area of fine particles to the total surface area. In the DFT calculation method, pores are considered to be only the cavities where depth is greater than width. Since the DFT method estimated surface area for pores up to 8 nm (80 Å) in diameter, for lower ranks or oxidized coals, which may contain larger pores, the external surface area (S_{ext}) may be overestimated. The external surface area (S_{ext}) of the particle to its equivalent sphere surface area (S_{sph}) is called rugousity (R) and is calculated using Equation (5-14) (as in Chapter 5).

The larger the R number, the more irregular, non-uniform the outer surface of the particle. The S_{ext} is the surface area calculated as the difference between the total surface area derived from BET (S_{BET}) and the surface area of pores up to 8 nm (80 Å) in size (S_{DFT}) obtained from the DFT method. The S_{sph} is the equivalent sphere surface area of the measured particle and is calculated from the size distribution as derived from the Malvern 2000 size analyzer. The sphere always assumes the smallest surface area for the particles of the same volume. Rugousity is the measure of how much the particle surface area exceeds its smallest possible surface area represented by the equivalent sphere. Rugousity can be influenced, however, by the shape and roughness of the particle and to clarify these effects, the aspect ratio and fractal dimension parameters were compared for these particles.

From Table 6.1-5, it is clear that the surface area of particles as derived from the Malvern size analysis representing surface area of spheres is much smaller than S_{BET} , since it does not include porosity.

Coals	Surface area from Malvern S _{spher} (m ² /g)	BET surface Area S _{BET} (m ² /g)	Pore surface area S _{DFT} in pores upto 8nm (<80Å) (m ² /g)	External surface area $S_{ext} = S_{BET} - S_{DFT}$ (m^2/g)	Rugousity S _{ext} /S _{sphere}	Aspect Ratio d _{min} /d _{max}	Fractal dimension D (range 2 to 3) 2-smooth; 3- very rough
•LC 3	0.124	0.390	0.222	0.168	1.35	0.77	2.76
●LC 10BC	0.123	0.652	0.360	0.292	2.37	0.78	2.68
■LC 10B	0.099	0.376	0.207	0.169	1.71	0.77	2.73
∎LC 8U	0.150	1.440	0.793	0.647	4.31	0.76	2.75
∎LS 20	0.159	4.14	2.70	1.440	9.06	0.77	2.93
■LC 3 OXY	0.050	0.265	0.158	0.107	2.14	0.78	2.75
•LC met	0.102	0.439	0.251	0.188	1.84	0.72	2.75
■LC thermal	0.176	0.661	0.367	0.294	1.67	0.79	2.70

Table 6.1-5. Surface properties, shape and roughness estimations of studied coal fines.

●- hydrophobic coal; ■ – hydrophilic coal

The external surface area of particles as calculated from the difference between the S_{BET} and S_{DFT} is in the range from 0.107 m²/g to 1.44 m²/g, while the highest values were obtained for LS 20 and LC 8U samples. As per the discussion in the previous section on porosity, one can assume that the external surface area for LS 20 and LC 8U coals includes surface area from macropores, therefore these values are highly exaggerated.

Since the contribution from the macropores in the remaining samples (higher rank coals) is not expected to be significant, it is assumed that the difference between S_{BET} and S_{DFT} represents mostly external surface area in the remaining samples.

The external surface area of these coals is in the range from $0.107 \text{ m}^2/\text{g}$ to 0.294 m²/g. The rugousity varies from lowest value of 1.37 for LC 3 coal to the highest at 2.37 for the LC 10BC coal sample. This indicates that particles of the LC 3 coal have the most regular surfaces whereas the particle of the LC 10BC coal are the most non-uniform.

The aspect ratios for these samples indicate that the shape of these particles is not very different. The aspect ratio changes from 0.72 to 0.79 at most for LC met and LC thermal, respectively, otherwise equals to 0.77 on average. When the aspect ratio is 1, it represents a sphere, whereas 0.5, ellipsoidal shape particles. The standard deviation of these measurements was in the 0.14 to 0.17 (+/-) range and was very consistent. The roughness was assessed for these coal particles from the fractal dimension analysis as described in Chapter 3. D is a parameter that generally assumes the value between D=2 for smooth surfaces and D=3 for very rough surfaces.

The Neimark-Kiselev (NK) method was used to determine the fractal dimension of studied coal fines. Their values are also included in Table 6.1-5. The fractal dimension values vary from the lowest 2.68 value for LC 10BC to 2.93 for LS 20, indicating that the smoothest surface is typical for the particles from the LC 10BC sample, while the roughest surface is characteristic for the particles from the LS 20 sample. Since the scale between the smoothest and roughest particle extends from D= 2 to D=3, the difference between the roughest and smoothest particles (LC 10BC and LS 20) is 17 %, and for all the other cases the difference does not exceed 10 %.

In summary, the external surface of fine particles of LS 20 and LC 8U samples was found to be overestimated due to the possible presence of macropores in these samples. Therefore, these two samples were excluded from further comparisons based on the external surface area. The aspect ratio and fractal dimension analysis, however, confirmed that these samples were similar in respect to the shape and roughness to other tested samples. When analyzing LC 10BC, one could conclude that this sample has the particles with the smoothest surfaces (fractal dimension parameter, D=2.68), with the most irregular topography (rugousity) and the extensive external surface area. The LC 3OXY particles

have the smallest external surface area, however, with highly irregular topography of the surface (rugousity) and is 7 % rougher (fractal dimension D=2.75 for LC 3 OXY compared to D=2.68 of LC 10 BC) than LC 10BC, while the shapes of these particles are identical. In general, the shape and roughness of particles in the tested samples appeared to be similar, therefore their effect on the aggregation ability should be negligible.

6.1.6 Mineral matter characteristics

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The coal fines were subjected to: microscopic examination, X-ray diffraction analysis of the low-temperature ash (LTA), and liberation studies in order to assess the amount of liberated mineral matter. The latter could play a potentially important role in the behavior of fine coal particles, especially in the presence of water. The results of the microscopic examinations are presented in Table 6.1-6.

The microscopic analyses of coal fines revealed that all of the samples contained considerable amounts of clay minerals and quartz as a second most abundant mineral. Only in the LC 10BC and LS 20 samples were carbonates visible amongst other mineral matter particles. The LC 3 coal fines appeared to be the only one with intermixed quartz and clay minerals, which perforated the surface of coal particles. As a result these particles appeared to be very contaminated with dispersed clays and small quartz grains. Both clean coal product samples LC thermal and LC met appeared to be very clean with no liberated mineral matter present in the sample.

The complete set of results from the XRD analysis along with the X-ray diffractograms are provided for all the samples in Appendix E. The summary of these results are given in Table 6.1-7. The total clays content for all the samples with the exception of LS 20 (49.2 %) is in the range from 57 % to around 63 %, while the quartz content is the highest for LS 20 (42.2 %) and lowest for clean coal products and LC 8U (below 30 %). Kaolinite was found as the major clay mineral, while the bentonite-type clays were not present in any of the studied samples. Gypsum was the third most abundant mineral occurring in the studied samples, however its presence in low-temperature-ash (LTA) samples is related to the oxidation of pyritic sulfur during low temperature ashing (Rao and Gluskoter, 1973).

Coal	Mineral matter characteristics
LC thermal	A lot of unliberated quartz and unliberated clays. Clays and quartz are found within coal grains.
LC metallurgical	Clean coal particles, very few free mineral matter particles
LC 10BC	Some carbonates and large liberated mineral matter particles
LC 8U	Shattered and crumbled coal particles, probably due to oxidation. Larger particles appear clean, unliberated clays present on coal
LC 10B	Clays mixed with quartz, more liberated mineral matter
LC 3	Abundance of clays and quartz mixed with clays. Dispersed clays appear to cover coal particles
LS 20	Larger particles with mineral matter in form of large quartz particles, some carbonates and clays present as lumps

Table 6.1-6. Microscopic examination of studied coal fines.

The ash content of fines was recalculated to represent mineral matter content (Table 6.1-7) according to Parr's formula (Ward, 1984) as given in equation 6-1 below:

 $MM_{wt\%} = 1.08 A_c + 0.55 S$ 6-1

where: $MM_{wt\%}$ is the weight percentage of mineral matter; A_c , is the ash content in the coal sample; S is the sulphur content (average sulfur content for LC coals, S = 0.44 was obtained from Grieve et al.,1996; for LS 20 S=0.8%).

Finally, the liberation studies were carried out to determine the amount of liberated mineral matter in coal fines of the tested samples. The data from this analysis is shown in Table 6.1-8. The amount of liberated mineral matter as obtained from sink-and-float tests is compared with the total clays content derived from the XRD analysis for all of the samples. As expected, the negligible amount of liberated minerals was found in the clean coal

samples (LC met and LC thermal), LC 8U sample had 14.08 % of liberated minerals, while the highest amount was found in the fines of the LC 3 coal and LC 10BC.

Coal	Mineral matter content, wt%*	Total clays content, wt %	Quartz content, wt %	Gypsum content, wt%	Other minerals content, wt%
LC thermal	17.37	60.6	29.5	6.7	3.2
LC met	13.86	57.6	26.8	4.3	11.4
LC 10BC	26.60	57.2	35.8	2.6	4.4
LC 8U	16.84	63.1	27.3	6.4	3.2
LC 10B	14.22	60.3	31.1	3.7	5.0
LC 3	13.86	62.2	33.7	2.4	1.7
LS 20	36.8	49.2	42.2	3.0	5.6

Table 6.1-7. Summary of the XRD analyses of mineral matter from studied coal fines.

*Mineral matter, wt % calculated from Parr's formula

In summary, one can conclude that all of the samples had a similar composition of the mineral matter with the exception of the LS 20 coal in which clays and quartz were present in almost equal amounts. The highest clays content was in LC 8U and LC 3, while the highest quartz content was found in the LC 10 BC and LC 3 sample. From microscopic examinations, the LC3 sample had the most particles contaminated with finely dispersed clays intermixed with quartz grains, while coal particles in the LC 10BC sample were observed to be cleaner. Fine clays were not covering coal particles, but rather appearing in separate bands.
Coal	Mineral matter content wt %*	Amount of liberated, mineral matter wt %	Total clays content wt %**	
LC thermal	17.37	0.80	60.6	
LC met	13.86	0.95	57.6	
LC 10BC	26.60	22.17	57.2.8	
LC 8U	16.84	14.08	63.1	
LC 10B	14.22	19.95	60.3	
LC 3	13.86	25.1	62.2	
LS 20	36.8	19.98	49.2	

Table 6.1-8. Results of mineral matter characterization

*Mineral matter, wt % calculated from Parr's formula; ** from XRD analysis;

6.1.7 Angle of repose

The angle of repose was measured for the fine fractions (-0.5 mm) of LC 3, LC 3OXY, LC 10B, LC 10BC, LC 8U and LS 20 coals. These samples were tested at increasing moisture contents. The tests were carried out on dry samples first, then increasing moisture of 2-5 % up to a maximum of 20 % moisture in each coal sample. The trends in changing flow characteristics with an increase in moisture content in these samples are shown in Figures 6.1.7-1 and 6.1.7-2. The results were divided into two categories; for hydrophobic and for hydrophilic coals, as these samples had distinctly different patterns of behavior.



Figure 6.1.7-1 The effect of moisture content on angle of repose for hydrophobic coals.



Figure 6.1.7-2 The effect of moisture content on angle of repose for hydrophilic coals.



Figure 6.1.7-3 The effect of moisture content on angle of repose for LC3 and LC3 OXY coals.

The hydrophobic coal fines (LC 3, LC 10BC and LC 10B) displayed a consistent increase in the angle of repose from 42 ° to 70 °, with increasing moisture content from dry to 10 %. In general, free flowing material should have an angle of repose in the range of 25° to 35° . The material is regarded as having fair to acceptable flow characteristics when angle of repose is between 38° and 45° . The cohesive fines will usually attain the values from 60° to 70° according to Carr, (1969). For LC 3 coal, initially the angle of repose increased steadily with the moisture increase, however at high moisture content (20 %), the angle of repose was reduced to 60° value. This indicates improved flowing properties for this coal at this high moisture content.

In the case of hydrophilic coals, a gradual increase in moisture content was accompanied by slow but steady increase in the angle of repose values, with no leveling off at high moisture content. This distinctive response to increasing moisture content for hydrophobic fines as compared to hydrophilic ones, is shown in Figure 6.1.7-3, the only difference between LC 3 and LC 30XY sample is wettability. LC 3 OXY is hydrophilic as discussed in earlier sections of this chapter. It is clear that for LC 3 sample, the flowability deteriorates up to a given moisture level and then improves (smaller angle of repose).

The flowability of the LC 3OXY fines continues to deteriorate steadily with no improvement at higher moisture content. This may indicate that for hydrophilic coals, the flowability deteriorates continuously until these coals eventually stop flowing entirely at high moisture content.

6.1.8 Summary and discussion

The handleability of any bulk material deteriorates with increasing amount of fines; these fines can represent material from -1 mm down to the micron size particles. In coal blends, material below 0.5 mm is referred as fines and an increase in the amount of this size fraction is critical for coal handling characteristics. Numerous handleability studies have established that the amount of fines is the most important factor, and the behavior of fines is affected disproportionately more by moisture increases than coarse fractions. As a result, the majority of the research related to handleability was aimed at studying properties of fines to predict handleability of the bulk sample. Therefore, the quality of fines was always an important factor in the studies on the handleability of coals. Usually, only the physical characteristics of fines were taken into account and related to the handling behavior of a particular coal blend. The approach taken in this study was to assess the quality of fines in terms of their surface properties with the emphasis on wettability of the studied coals.

The results of the detailed study on surface properties of tested coals were discussed in the previous sections of this chapter. Characterization of coal fines was accomplished through comprehensive analysis of surface properties of fines such as surface area, porosity and equilibrium moisture. Wettability was examined using the penetration rate technique and transmittance tests. An angle of repose method was utilized to assess the flowability of these fines. In addition, particle shape and roughness were addressed in the context of the surface properties of particles. Mineral matter associated with coal particles was analyzed using optical microscopy, an XRD of low-temperature ash analysis and liberation studies of mineral matter present in these samples. Pelletization was also used as a method to test the quality of fines in terms of their surface properties.

The wettability assessment revealed that the studied coals differ in wettability characteristics. The results from the applied methods were found to be in a good agreement

with each other, even though each of these methods relies on different coal characteristics to estimate the wettability of studied coals as discussed in Chapter 5.

The surface area and porosity measurements showed that specific surface area as derived from the BET method of the majority of studied samples was $< 1 \text{ m}^2/\text{g}$ and this is in accordance with the rank of these samples (Gan et al., 1972; Parkash et al., 1984). The surface area for the LS 20 and LC 8U coals was found to be higher due to the lower rank in the case of LS 20 and an extensive oxidation of the LC 8U coal. In terms of porosity, mesopores 2 to 8 nm (20-80 Å) were found as major contributors to the total surface area for almost all of the samples, while microporosity was increased for four out of eight of the studied samples, but the micropores were not generally predominant in these coals. The macroporosity was found to be implicated by the external surface area of the particles; however, due to the high rank of these coals, this should not have a significant effect on the overall porosity estimation.

The amount of water adsorbed onto the coal under saturation conditions represents the equilibrium moisture. The equilibrium moisture was found to be an important parameter, setting the boundary for the adsorption of water onto the coal surface. The coal surface cannot adsorb more water than its surface properties will allow; once the equilibrium moisture is exceeded, the water will start forming a film around the particles, enhancing aggregation of particles. This is where the ability of particles to aggregate becomes visible. Pelletization tests were carried out to test the interaction between coal particles. Pelletization results show that the strength of the pellets can be used as the evidence of particle to particle interactions, but neither surface area, porosity, wettability nor ash content alone were sufficient to explain the differences in strength of pellets formed from these coals.

In addition to the surface area and porosity measurements, particle shape and roughness were studied. These analyses can be an indication of physical characteristics relating shape, roughness and extent of the surface area for the studied samples. It was concluded that most of these coal particles were similar in shape and described as comparable in terms of roughness. Therefore, the differences between the physical surface properties of the tested coals are considered to be negligible. Mineral matter

characterization complemented the surface properties analysis of coal particles and became helpful in understanding the role of mineral matter in the aggregation of particles.

The estimation of the angle of repose provided flowing characteristics of the examined fines. The results revealed that hydrophobic fines behaved differently with the moisture content increase than hydrophilic fines. This was the first evidence that fine coal with different wettability characteristics could behave differently in the handleability tests.

Pelletization was confirmed to be an adequate method to test the tendency of fine coal to aggregate. However, there is a missing link between the wettability concept of coals, their specific surface area, and porosity and ability to aggregate. In the following section an attempt is made to provide a model of coal particles behavior in the presence of water, which is based on coal particles' wettability .

6.2 Proposed model of aggregation for hydrophobic and hydrophilic coals

The required condition for particles to aggregate is that the surface of the solid is wetted, since is only that the water film is then formed around the particle. There are three stages which lead to pellets formation these are: pendular, funicular and capillary stage. However, only the capillary stage is responsible for building the stable bridges between the particles and forming the pellets. In general, hydrophilic particles are easy to pelletize, while pelletization of hydrophobic particles is difficult.

In order for water film to appear on the surface, the coal surface has to be saturated with adsorbed water. To analyze this situation, two cases will be considered, one where coal is hydrophobic and the other one where coal is hydrophilic.

Wettability model for water adsorption on to hydrophilic and hydrophobic surfaces has been proposed by Fuji, et al., (2000) as discussed in Chapter 3. Previous studies into water vapor adsorption (sorption isotherms) (Mahajan and Walker 1971) can give us some indication of a water adsorption pattern with respect to the porosity and wettability of particles. Equilibrium moisture correlates well with the surface area and hydrophilic groups' content (Kaji et al., 1986), which means that the amount of water needed to saturate the coal surface is related to wettability. According to Kaji et al. (1986), for some coals the amount of water exceeded 2 to3 times the porosity, while for other coals, it only filled 30-70% of pore volume. Since in hydrophobic coals, water does not penetrate into the pores to

a significant extent, the water adsorption is limited, while in hydrophilic coals, water not only penetrates but also condensates in the pores, therefore the water adsorption onto such surfaces is very extensive, as shown in adsorption studies by Kaji et al. (1986).

In the pelletization, it is generally accepted that the thinner the film around the particles, the stronger are the bonds created between the aggregating particles in the capillary stage. According to Naidich et al., (1965a), Mehrotra and Sastry (1980), the more hydrophobic the particles, the weaker the pellets formed from such coals. If pelletization relies on the wettability of particles it could be assumed that the surface area and porosity have the same effect on the particles' ability to form stable pellets.

It is proposed in this thesis to use the pellet strength as a measure of surface forces acting between the particles. Therefore, the magnitude of the pellet strength is the consequence of particle wettability, its surface properties (surface area and porosity) as well as mineral matter content (ash). Since the proposed model links the wettability of particles with their ability to aggregate it will be referred to as the **wettability-aggregation model**.

In order to test this hypothesis, two separate cases of hydrophobic and hydrophilic coals were to be considered:

Hydrophobic case: wetting of hydrophobic coal by water is difficult. Once the coal is covered with coalesced droplets of water, the bonds formed between particles of such a solid are not strong due to a small adhesion energy of water to coal. Although the capillary state required for pellets to form may be reached, the layer of water around aggregating particles remains relatively thick and unstable. As a result, the strength of the formed pellets is low. In this case, the mineral matter will be the only factor contributing to the pellets strength formed from such particles. Figure 6.2-1 depicts the aggregation of hydrophobic particles in the presence of water.

Hydrophilic case: in the case of hydrophilic coal, water penetrates quickly into the pores creating micro-capillary pressure from within. Once an excess of liquid appears on the surface of the particles, (at the moisture content exceeding the equilibrium moisture), aggregation proceeds right into the capillary state. The layer of water on the surface of the particles is stable and therefore, the strength of the pellets is much greater due to the strong capillary pressures from within the coal structure. Figure 6.2-2 illustrates aggregation-pelletization of hydrophilic particles. In the case of hydrophilic coals, porosity,

microporosity as well as the mineral matter become important variables in building the strength of pellets.



Aggregation of hydrophobic particles



6.2.1 The effect of surface properties on the pellets' strength

To graphically illustrate the correlation between the pellet strength and surface properties and ash content, the diagram shown in Figure 6.2.1-1 is proposed. According to the proposed model, when the coal particles are very hydrophobic ($\cos\theta = 0$; $\theta = 90^{\circ}$), the only sites for water droplets attachment are the mineral matter inclusions (ash); for these particles pellet strength (P_s) is a function of ash content f(ash). For very hydrophilic coal particles, pellets' strength (P_s) depends on the ash content as well as the specific surface area, and porosity (surface properties parameter; S_p), therefore $P_s = f(ash, S_p)$.

The correlation between the measured values of transmittance versus pellets' strength is shown in Figure 6.2.1-2. It is evident that there is a valid relationship ($R^2 = 0.76$) between the wettability of these samples as expressed by the value of transmittance and pellets' strength. With the increase in degree of oxidation, the coal surface wettability increases, and so does the pellets' strength.



Aggregation of hydrophilic particles

Figure 6.2-2 Wettability- aggregation model for aggregation of hydrophilic particles in presence of water (a); interaction between water and coal surface at microscopic level(b)



Contribution to pellet strength



The correlation between the surface area and pellets' strength is shown to be consistent with the porosity and pellets' strength for all tested coals except LS 20 coal (Figures 6.2.1-3 and 6.2.1-4). The strength of pellets made from LS 20 coal was so high that it was found to skew the results for other coal samples and was excluded from this correlation. The correlation coefficient for both type of plots was shown to be almost identical $R^2 = 0.46$ for specific surface area vs. pellets' strength, and $R^2 = 0.45$ for porosity vs. pellets' strength, as the surface area is related to the coal porosity. The studied coals were of the same rank, therefore surface properties such as specific surface area and porosity are very similar, thus there is no significant change in surface area or porosity over the studied range with the exception of LC 8U as discussed earlier in this chapter. The effect of an increased specific surface area and porosity on pellets' strength is a dominating factor for hydrophilic coals, while hydrophobic coals have always lower pellets' strength with the exception when high amounts of clay material is present in the coal e.g. LC 10BC (Table 6.2-1). The scattered correlations between data points in Figures 6.2.1-3 and 6.2.1-4 reflect this effect.



Figure 6.2.1-2 Correlation between the transmittance and pellets' strength for tested coals.



Figure 6.2.1-3 Correlation between specific surface area and pellets' strength for studied coals.



Figure 6.2.1-4 Correlation between surface area in pores (porosity) and pellets' strength for studied coals.

6.2.2. The effect of mineral matter type on the pellets' strength

The ash content is a measurable parameter which characterizes the amount of mineral matter in coal. It represents quantity of minerals present in the coal sample, while the type of mineral matter may be important when aggregation of coal particles is considered. This is especially true, when clays are present in the coal. The effect of clays on handleability has been discussed in Chapter 3.

In the pelletization study by Sastry and Fuerstenau (1982), it was shown that stronger pellets were produced from high ash and high sulfur coals. This has been attributed to the fact that some ash forming minerals act as binders, especially when large proportions of clay minerals are present in the coal. In view of this, the strength of pellets depends not on the amount of ash but rather the type of mineral matter present in coal. For example, the bentonite clay is used as an additive to iron ore pelletization to improve the pellets' strength. However, in the study by Sastry and Fuerstenau (1982), the bentonite was shown not to be as effective in coal pelletization as in the case of iron ores. To examine the effect of type of mineral matter on the pellets' strength, the content of major ash forming minerals has been calculated to represent the contribution to the total mineral matter content (Table 6.2-1).

The plots were obtained correlating pellets strength with the mineral matter content for the studied coal fines. The correlation between data points from this relationship for tested coal fines is very poor (Figure 6.2.2-1). The LS 20 sample was excluded from the correlation, as explained earlier. The LS 20 sample is only used for comparisons on case-by case basis. The correlation shown in Figure 6.2.2-1 illustrates that for coals with similar level of mineral matter, the strength of the pellets can vary noticeably. Similar trends are found for quartz and clays.

Since the correlation between the wettability and pellets' strength was shown to be significant (Figure 6.2.1-1), and the mineral matter content is an important parameter in building the strength of the formed pellets, the results correlating mineral matter content with pellets' strength are replotted separately for hydrophobic and hydrophilic coals. Once these two cases are plotted separately, correlation for both type of coals became very good; for hydrophobic coals was found to be almost perfect fit ($R^2 = 0.98$) as shown in Figure 6.2.2-5. The correlations between content of clays and quartz on pellets' strength for both type of coals is included in Appendix E. There is no obvious difference between the effect of clays and the effect of quartz on pellets' strength in both type of samples (hydrophobic and hydrophilic). Similar trends for the correlations for clays and quartz minerals in hydrophobic and hydrophilic coals indicate that overall mineral matter effect on the pellets' strength is determined by the most dominant minerals present in these coals, such as clays and quartz. The effect of the mineral matter content on the pellets' strength is more pronounced for hydrophilic coals as compared to the hydrophobic ones, indicating that the same amount of mineral matter will have a greater effect on the pellets' strength for hydrophilic than for hydrophobic coal.

Coal	Mineral matter content, wt%*	Total clays content in mineral matter ,wt %	Quartz content in mineral matter, wt %	Gypsum content in mineral matter, wt%	Pellets' strength. kg/m ² x 10 ⁻⁶
■LC thermal	17.37	10.60	5.12	1.16	20.0
•LC met	13.86	8.04	3.71	0.60	7.5
•LC 10BC	26.60	15.16	9.52	0.69	13.8
■LC 8U	16.84	10.61	4.60	1.08	20.0
■LC 10B	14.22	8.53	4.42	0.53	11.6
•LC 3	13.86	8.59	4.67	0.33	8.5
∎LS 20	36.8	18.03	15.53	1.10	289.0

Table 6.2-1 The mineral matter composition and its effect on pellets' strength

•-hydrophobic coal; ■ – hydrophilic coal ;* from Parr's formula

The example of this is the LS 20 hydrophilic coal sample with very high clays content, and LC 10BC, hydrophobic sample with similarly high clays content, yet the pellets' strength is significantly greater for LS 20 coal than for LC 10 BC coal (Table 6.2-1). This is attributed to the strong hydrophilicity and extensive porosity of LS 20 sample, while in the LC 10BC hydrophobic coal sample, only the mineral matter contribution to the pellets strength is a significant factor. Likewise, for LC 3 and LC 3 OXY, and LC 3 and LC 10B samples, with similar clays content stronger pellets are always made from hydrophilic coals.

The fact that correlation between mineral matter content and pellets' strength was found only valid when hydrophobic and hydrophilic samples were considered separately, confirms that wettability is the most significant parameter affecting pellets' strength, while the effect of mineral matter type and content depends on the wettability of coal. This also confirms proposed model.



Figure 6.2.2-1 The correlation between the mineral matter content and pellets' strength for studied coals.



Figure 6.2.2-2 The correlation between total clays content in mineral matter and pellets' strength for studied coals.



Figure 6.2.2-3 The correlation between quartz content in mineral matter and pellets' strength for studied coals.



O hydrophobic coals □ hydrophilic coals

Figure 6.2.2-4 The correlation between the mineral matter and pellets' strength for hydrophobic and hydrophilic coals.

6.2.3 Pellets strength as evidence for particles interactions

To further evaluate the **wettability-aggregation model**, more detailed analyses of the following cases were used here to compare the strength of pellets made from samples of different surface properties and mineral matter content (Tables 6.1-4 and 6.2-1).

<u>Case 1. Comparison of the behavior of the LC 3 and LC 3OXY coal fines.</u> Since the LC 3OXY sample was prepared by oxidizing the LC 3 sample, the mineral matter content (ash) and type of both samples is identical. Although there is some change in the microporosity after oxidation, the strength of the pellets almost doubled for the oxidized sample (LC 3OXY). For "oxidized – hydrophilic coal", the strength of the pellets results from strong capillary forces which are due to surface hydrophilicity, porosity (surface area) as well as the mineral matter content. Therefore it is evident that a decrease in hydrophobicity has a larger overall effect on the strength of the pellets.

<u>Case 2. Comparison of the behavior of the LC met with LC 10B coal fines</u>. These coals showed a similar trend to LC 3 and LC 3OXY. Significantly stronger pellets were produced from the hydrophilic LC 10B coal than from the LC met coal. The surface area and porosity (Table 6.2-2) are similar for both of these coals, as well as the mineral matter and clays content. The strength of the pellets made from LC 10B coal fines is much greater than pellets made from LC met. The wetting water on the hydrophobic LC met coal was not able to penetrate the pores to create a significant force for aggregation.

<u>Case 3. Comparison of the behavior of the LC met with LC 10BC coal fines</u>. These two coals are of similar hydrophobicity (Figure 6.1.1-1), but LC 10BC produced 1.83 times stronger pellets than LC metallurgical. In both cases, contribution of porosity to the force pulling particles together was apparently negligible; the only difference was in the mineral matter content (LC met = 13.86%; LC 10BC = 26.60%) and amount of clay material in LC 10BC is also twice as much as in LC met sample. In this case, it appeared that the mineral matter in the LC 10BC sample contributed to the strength of the pellets, and this effect was almost exactly the same magnitude as the difference between the strength of the pellets formed for these two coals.

<u>Case 4. Comparison of the behavior of the LC 8U with LS 20 coal fines</u>. Both of these coals are hydrophilic. According to the penetration rate, both coals have a similar level of

hydrophilicity with a similar equilibrium moisture value, while pellets from LS 20 coal are 14 times stronger than pellets formed from LC 8U. The LS 20 coal has twice as much mineral matter (ash) content with also much higher clays content, as compared to LC 8U, hence the ash forming minerals contribute to the pellets' strength. The fact that LS 20 coal fines are much coarser than LC 8U, as found from particle size analysis (Figure 4.2-1), would rather decrease the pellets' strength of LS 20, as indicated by Sastry and Fuerstenau, 1982) in the study on the effect of increased feed fineness on pellets' strength. Furthermore, the LS 20 coal has much greater microporosity and over almost three times greater specific surface area than LC 8U and this could have a significant effect on an increase in pellets' strength for the LS 20 coal fines as compared to those made of the LC 8U coal fines, confirmed by experiments. Therefore, the surface properties' contribution to the pellets' strength of the LS 20 and LC 8U coal fines confirms the wettability-aggregation model for the hydrophilic coals case.

Case 5. Comparisons of the behavior of samples with liberated and unliberated mineral matter. The effect of liberated vs. unliberated mineral matter on pellets' strength is examined for LC thermal and LC 10B, and for LC met and LC 3 coals. The LC thermal and LC 10B are both hydrophilic coals, while LC thermal has 10.60 % clays in mineral matter, which is mostly unliberated as compared to 19.95% liberated mineral matter of the LC 10B with 8.53 % clays (Table 6.1.8). The pellets made from LC thermal are much stronger than the pellets made from LC 10B coal fines. This indicates that greater surface area and porosity, in combination with increased clays content in LC thermal (Tables 6.1-4 and 6.2-1), are contributing factors to the pellets' strength. Furthermore, the LC met-hydrophobic coal with 8.04 % clays content in mostly unliberated mineral matter, produces pellets with the same strength as hydrophobic LC 3 coal, with 25 % liberated mineral matter, which has a similar clays content (8.59 %).

All the other cases confirm the conclusion that the strength of the formed pellets is related to the hydrophilic/hydrophobic character of coal particles; i.e. their surface properties parameter (S_p) and mineral matter content as described in the earlier section of this chapter and illustrated in Figure 6.2.1-1. The liberation of mineral matter, apparently has no significant effect on pellets' strength, while the clays content contributes significantly to the increased strength of produced pellets. The effects of other parameters

contributing towards the pellets strength are difficult to separate, because it is impossible to isolate one parameter from the other, as they represent coal inherent properties (surface area, porosity), therefore a case-by-case analysis was shown to be the effective way of studying these effects.

Thus, another approach was taken to analyze the effects of these parameters on pellets' strength from studied coals. In this approach, the parameters-variables were categorized as low, medium and high according to their values and put into a table along with the resulting response values (pellets' strength) as shown in Table 6.2-2. The coal samples are not identified, except for their wettability character, there is no connection between the position of these samples in the table-matrix with regard to their other variable-characteristics. From these results it is evident that for all four variables at every level (low, medium, high), the effect of each variable on the pellets strength is always greater for hydrophilic coals. In other words, the wettability is the most significant factor, because it determines the effects of all other variables on the pellets' strength.

For hydrophilic coal particles as predicted from the model, surface area and porosity contribute towards the pellets' strength. However, they do not have any effect on the increase in pellets' strength for hydrophobic coal fines when considered at the same mineral matter or clays content. The stronger pellets are always produced from hydrophilic coals, and at all levels of clays contents. For hydrophobic samples however, only a high content of clays is able to increase the pellets' strength, as in the case of LC 10 BC hydrophobic coal with high clays content (15.16 %) and resulting pellets' strength of 13.8 kg/m²x 10⁻⁶.

6.2.4 Summary and discussion

The interaction between fine particles was examined using pellets strength as the evidence of forces acting upon these particles and leading to their aggregation.

Wettability-aggregation model was proposed and discussed in view of pelletization results. It was confirmed that the magnitude of the pellets' strength is a consequence of the particles' surface properties. The pellets' strength for hydrophobic coal fines was shown to be related only to the mineral matter (ash) content, while for hydrophilic coals, the surface properties such as surface area and porosity also become parameters affecting pellets' strength. This confirmed the validity of the proposed model. The

wettability was found to have the most significant effect on the pellets' strength (Figure 6.2.1-2), while surface area and porosity were found to be somewhat correlated with the pellets' strength from the studied coals (Figures 6.2.1-3 and 6.2.1-3). Surface area and porosity characteristics of studied coals were very similar, thus there was no significant difference in their effect on pellets' strength, with the exception of LC 8U and LS 20 coal.

There was no correlation between mineral matter content and pellets' strength, when all coals were plotted on the same graph, whilst a very strong correlation was found when data was plotted separately for hydrophobic and hydrophilic coals. This indicates that the effect of mineral matter content on the pellets' strength is surface-specific, the effect depends on the type of surface in terms of wettability.

Similarly, the effects of two most dominant minerals, clays and quartz, on pellets strength were shown to have the same correlation as the total mineral matter content on the pellets' strength, demonstrating that the overall effect of mineral matter content is dictated by the most dominant minerals present in the coal. Since the clays are the most abundant minerals in the studied coal fines, their effect was considered in the multivariable analysis.

The multivariable analysis (Table 6.2-2) has shown that at all three levels of the variables, the pellets' strength was always greater for pellets made from hydrophilic coals, indicating that all variables (clays content, surface area, porosity and microporosity) play a much greater role in increasing the pellets' strength for these coals, while for hydrophobic coals only clays content contributes to the pellets' strength.

In summary, the effectiveness of pelletization depends on wettability. Wettability is the most important factor because it decides whether the porosity can create a strong capillary force that affects particle-to-particle adhesion. In the case of hydrophobic coals, the strength of the pellets results only from the mineral matter (ash content) since water will not penetrates into the pores, to any significant extent, thereby not creating a strong pulling force from within coal pores. While for hydrophilic coals, water penetrates into the pores, producing a strong capillary force, which coupled with mineral matter, generates strong pellets.

The pelletization behavior of the studied coal fines therefore provides information on how these fines would behave in the presence of water. In general, easy to pelletize coal fines will tend to aggregate, and these aggregating particles will have a much greater

strength and as a result, should be difficult to handle. To verify the relevance of pelletization as the method of testing handleability of coal, testing of bulk samples followed and this part of the research will be discussed in the subsequent sections.

6.3 Assessment of bulk sample handling properties

In this part of the study, evaluation of the flow properties of the studied coals was performed using Durham Cone and Handleability Monitor. The flow behavior was assessed with an emphasis on the particle segregation mechanism and the wettability of the coal fines. The effect of increased fines content on the bulk sample handleability was examined. The critical moisture of the bulk sample was defined as the handleability parameter based on the Durham Cone tests.

Handleability tests of the coals from United Kingdom and Australia were carried out using Durham Cone and Handleability Monitor. Comparisons were made and the correlation between the respective handleability indices was derived from these two methods. The effect of coal wettability was discussed using the results obtained from random handleability tests with the use of Durham Cone and Handleability Monitor. The bulk density measurements were performed to supplement bulk testing data and also to shed the light on packing characteristics of the tested coals.

6.3.1 Bulk sample testing using Durham Cone

The Durham Cone was used to measure flow rates of the studied coal samples according to the procedure described in section 5.7.3 of Chapter 5. For bulk sample testing four raw coals (LC 3, LC 8U, LC 10B and LC 10BC) and two clean coal products (LC met and LC thermal) were used. The raw samples were prepared to give three different size distributions. One set of samples was run-of-mine (ROM) with top size 53 mm (similar to the product size distribution from the local mines) and the second set was crushed down to 100 % passing 6.3 mm top size. The third set was prepared by adding 20 % (by weight of the whole sample) of the -0.5 mm fraction to the ROM samples. The size analyses of these samples are shown in Table 4.2-1. The effect of adding fines and coarse fractions on the handleability was examined using Durham Cone method.

The Handleability Monitor was used to test several thermal and metallurgical coal blends with different particle size distributions at several moisture levels. After each series of tests, the same samples were subjected to flow tests using the Durham Cone. In addition to the above-discussed tests, seven product coal samples were randomly chosen for simultaneous testing with the Handleability Monitor and Durham Cone. These seven coals were tested "as received" and then the same amount of water was added to each sample and the samples were tested again. To reduce the moisture content of coal, a powdered zeolite was added to each sample as an absorbent.

6.3.1.1 Segregation effects

Segregation of coal particles was observed as the Durham Cone was vibrated. The sample was not mixed between runs to preserve the segregation patterns for the whole sample. The flow rates were plotted versus cumulative time of all runs for a particular sample (LC 8U in Figure 6.3.1-1). It was observed that at low moisture content, even though segregation took place, it did not affect the flow in a visible way. Usually the larger particles would predominantly appear on the sides of the vibrating cone, while the fines would be in the center. Once the flow had been initiated, the fines were the first to flow out from the cone into the collecting pail followed by the coarse particles. The photographs showing segregation sequence of coal particles during Durham Cone tests are included in Appendix G. For each subsequent run in a given test, the sample was transferred from the collecting pail to the cone, and as a result, the particles which were at the bottom of the pail, were placed on the top.

The range of high to low flow rates is noticed for the LC 8U sample (Figure 6.3.1-1). As the moisture content increased, the segregation effects became more pronounced. For the LC8U coal sample at 4 % moisture content, the flow rates for fines and coarse particles were not that different, while for 10 % moisture, the difference is considerable (Figure 6.3.1-1). At this level, moisture appears to affect the fines more than the coarse coal. At 15 % moisture, with the time progressing, more and more particles become affected, and at 17 % moisture content all the particles were affected in the same way, as they reached saturation level and no-flow condition, with little or no flow through the cone outlet.

Variables\	Total	Pellets'	Specific	Pellets'	Surface	Pellets'	Surface	Pellets'
values	clays	strength	surface	strength	area in	strength	area in	strength
	content	kg/m ² x 10 ⁻⁰	area	kg/m²x 10 ⁻⁰	pores	kg/m²x 10 ⁻⁰	micropores	kg/m ² x 10 ⁻⁰
T	<u>%</u>		<u>m'/g</u>	14.0	m^{-}/g	110	<u>m⁻/g</u>	11 /
Low	●8.04	7.5	■0.274	14.3	∎0.156	14.3	■0.013	11.6
Low	■8.53	11.6	■0.376	11.6	■0.207	11.6	■0.019	14.3
Low	•8.59	8.5	•0.390	8.5	•0.222	8.5	■0.040	20.0
Low	■8.59	14.3	•0.439	7.5	•0.253	7.5	•0.048	8.5
								
Medium	∎10.60	20.0	•0.652	13.8	•0.360	13.8	■0.051	20
Medium	∎10.61	20.0	■ 0.661	20.0	∎0.361	20.0	•0.069	13.8
High	•15.16	13.8	∎1.44	20.0	■0.792	20.0	•0.149	7.5
High	∎18.03	289.0	■4.14	289.0	■2.71	289.0	■0.489	289.0

Table 6.2-2 The effects of different variables on pellets' strength for studied coals.

●-hydrophobic coal; ■ – hydrophilic coal

The segregation was common to all tested coal samples; for hydrophobic samples, however, the deterioration of flow rate was not so pronounced. At high moisture content, these samples did not cease to flow, even though their flow rates were very low. Figure 6.3.1-2 presents the flow rate for LC 10BC hydrophobic coal at increasing moisture levels. Apparently all particles were affected by these moisture increases, but this did not lead to building strong cohesiveness amongst particles as they were able to continue to flow. There is a visible change in flow rates fluctuation from one moisture level to the other. While the degree of fluctuation may vary between the samples (Figures 6.3.1-1 and 6.3.1-2), there is always such a moisture content at which the variation is the greatest and this moisture represents a critical moisture value.



Figure 6.3.1-1 The flow rates for LC 8U at different moisture contents.



Figure 6.3.1-2 The flow rates for LC 10BC at different moisture contents.

6.3.1.2 Critical Moisture

The **critical moisture** is the moisture content at which the greatest fluctuations in flow rates are observed for a given coal sample. This is also the moisture, at which behavior of fines starts to be problematic. Hall and Cutress (1960) described the moisture that coincides with the maximum time to flow out of the cone (lowest flow rate) as the important parameter, whereas Mikka and Smitham (1985) called this moisture a "critical moisture". The moisture content which is described as "critical" in this study is actually the level prior to which the sample stops flowing from the cone. It is determined by plotting the standard deviation for measured flow rate versus the moisture content, and the critical moisture is determined by a maximum on the curve as the one shown for the hydrophilic LC 8U coal in Figure 6.3.1-3 and for the hydrophobic LC 10BC coal in Figure 6.3.1-4.

Figures 6.3.1-5 and 6.3.1-6 depict the critical moisture for LC 10B ROM and LC 10B crushed coal and LC 3 ROM and LC 3 crushed respectively. It can be observed that the critical moistures for ROM and crushed samples are very close for both samples; however, the effect of increasing the moisture for the coarser sample is less pronounced. The values

of the critical moisture for other samples are included in Table 6.3-1. For the LC 10 BC sample the standard deviations of flow rates at 6 % and 10 % moisture contents were almost identical, thus critical moisture was determined from the degree of fluctuation as compared to the average value, rather than from standard deviation value alone (Table G.1-1)

The critical moisture is a very important parameter that characterizes handling properties of a bulk sample because it precedes the non-flowing or significantly deteriorating conditions for the sample. It can vary from sample to sample, as it depends on the characteristics of the bulk coal.



Figure 6.3.1-3 The critical moisture determination for LC 8U coal



Figure 6.3.1-4 The critical moisture determination for LC 10 BC coal.



Figure 6.3.1-5 The critical moisture determination for LC 10B ROM and LC 10B crushed.





6.3.1.3 The effect of size distribution on handleability

The effect of adding fines to the coal blend has been examined by comparing the flow rates for the same samples but with various size distributions. In Figures 6.3.1-7 and 6.3.1-8, the flow rates of two coals (LC 3 and LC 10B) for ROM and crushed samples. The amount of fines in crushed samples is almost double as compared to the ROM samples. It is clear that an increasing amount of fines in each of these samples leads to the deterioration of the flow rates.

However, the deterioration in flow rates is not as pronounced as one would expect from the amount of fines present in the crushed sample (Table 4.2-1). It is believed that larger top sizes and the greater amount of coarse particles in ROM samples could hinder flow by blocking the opening of the cone. This could counteract the benefit of the small amount of fines contained in these samples.

To clarify this effect, samples were prepared in such a way that 20 % (by weight of the original sample) of fines was added to the ROM coal. The flow rates for the LC 10BC

sample with added fines were compared with flow rates of the original ROM sample (Figure 6.3.1-9). The effects of the fines addition were more pronounced when the top size was the same in both samples. When the two samples, which had approximately the same amounts of fines, but different size distributions (LC 8U ROM with added fines and LC 8U crushed) were tested it was found that the sample with the larger top size had higher flow rates (Figure 6.3.1-10).

It can be concluded that it is not just the amount of fines in the sample but the whole particle size distribution that plays an important role in the flow of bulk coal. The results showed the wider the spectrum of sizes, the better tolerance for an increased amount of fines. There may be a limit for top size in each blend, past which the presence of large particles with cohesive fines will cause a problem when passing through the handling system



Figure 6.3.1.-7 The flow rates for LC 3; ROM and crushed sample



Figure 6.3.1-8 The flow rates for LC 10B; ROM and crushed sample.



Figure 6.3.1-9 The flow rates for LC 10BC ROM and LC 10BC ROM with added fines.



← LC 8U ROM added fines — LC 8U crushed



6.3.1.4 The effect of wettability of fines on the bulk coal handleability

The selected coal samples used in this study had different wettability characteristics. The transmittance and equilibrium moisture values indicated that some of the samples were very "wettable" (LC 8U), and other being somewhat wettable (LC 10B). LC 10BC and LC 3 samples are the most hydrophobic (Table 6.1-1).

As discussed in the previous chapter, the wettability of fines controls the behavior of particles in the presence of moisture. As a result, for hydrophobic coals, only mineral matter can have a significant effect on the handleability as determined from pelletization tests and confirmed by proposed wettability-aggregation model.

For hydrophobic coals, an increase in moisture may lead to deterioration of flow rate, but usually does not lead to "non-flowing" conditions. On the other hand, for hydrophilic coals it is not unusual that the deterioration of flow rate is more dramatic with the increase of moisture. Due to the fact that hydrophilic coals have higher equilibrium moisture, they can tolerate much higher moisture contents before the handleability of the bulk sample is impaired. Once the moisture content in the fines exceeds the equilibrium moisture, the sample becomes very cohesive. The effects of moisture on the flow rates for coals varying in wettability are shown in Figures 6.3.1-11 and 6.3.1-12.



Figure 6.3.1-11 The flow rates for three different colas; LC thermal, LC 10B and LC met.

Table 6.3-1. Bulk samples characteristics of the studied coals.						
Coal	Yield of (-0.5mm) (wt %)	Ash in fines (wt %)	Ash in bulk sample (wt %)	Equilibrium moisture (%)	Critical Moisture (%)	
•LC 3	36.0	12.61	30.12	1.30	5.0	
•LC 10BC	39.0	24.41	37.45	1.65	10.0*	
■LC 10B	45.0	12.94	24.30	2.96	9.0	
■LC 8U	37.0	15.37	23.63	7.34	10.0	
∎LS 20	22.0	33.7	35.00	8.02	11.0	
•LC met	44.0	12.66	9.77	1.56	10.0	
■LC thermal	45.0	15.86	15.15	2.09	6.0	
LC 3 OXY	36.0	12.61	30.12	4.38	12.0	

Table 6.3-1. Bulk samples characteristics of the studied coals.

* critical moisture calculated from the degree of fluctuation (Table G.1.1)

Figures 6.3.1-11 and 6.3.1-12 depict flow rate changes at increasing moisture levels for the studied coals. The LC met, LC 10B and LC thermal have a similar content of fines (about 45 % of the -0.5 mm material) and ash content (LC met =12.66 %; LC 10B =12.97 %) with the exception of LC thermal (15.86 %) as shown in Table 6.1-1. According to the pelletization tests, the LC thermal sample should be ranked as the most difficult to handle followed by LC 10B and LC met, and indeed this was found to be the case. These bulk coal samples have a very high amount of fines and this is reflected in their very low flow rates at a high moisture content.

For the other four coals, the trend is not so evident (Figure 6.3.1-12). LS 20 coal appeared to be the easiest to handle, followed by LC 8U, LC 10BC and LC 3. Coals LS 20 and LC 8U have similar wettability characteristics (Table 6.1-1 and Figure 6.1.1-1), however, LS 20 had only 22.0 % of fines (-0.5 mm fraction) as compared to 37.0 % in LC 8U sample. Since the amount of fines is a critical factor, it is not surprising that LS 20 exhibited better handleability than LC 8U. Both of these coals have equilibrium moisture in fines of about 8 %, implying that below that moisture content, there is no surface moisture present on these particles to lead to aggregation. Thus, only at total moisture levels exceeding 8 %, the particles will start to aggregate or stick together. To emphasize this effect, correlations shown in Figure 6.3.1-12 were re-plotted versus the surface moisture for these four coals (Figure 6.3.1-13). The surface moisture was calculated as the difference between actual moisture and equilibrium moisture.

The re-plotted data shows that LC 8U coal has the same amount of surface moisture as the LC 3 coal; the difference is that at 10 % surface moisture, flow from the Durham Cone practically ceased, while the LC 3 coal at this level of moisture is still handleable (Figure 6.3.1-13). The same can be concluded for LS20 and LC 10BC coals. The flow rate vs. surface moisture plot for the LC 10BC coal exhibits a very similar trend as that of the LC 3 coal. The shape of the curve for both of these coals suggests that they do not cease to flow at high moisture levels, which seems to be a feature of hydrophobic coals. This is related to the weak aggregative properties of hydrophobic fines, although they stick together but their aggregates' strength is not large enough to stop the flow of the bulk coal. In other words, the bridges that are built from these particles are more likely to be breakable, thus they do not stop the flow of the bulk sample completely, whilst hydrophilic

coal particles tend to form strong aggregates, which are difficult to break and this results in non-flow conditions. This is in agreement with the pelletization results. However, both of the hydrophobic coals (LC 3 and LC 10BC) exhibited overall poor handleability characteristics. From the mineralogical and the XRD analysis (Tables 6.1-8 and 6.2-1), it is evident that LC 10 BC coal have high amounts of clay minerals in the mineral matter, while in LC 3 coal clays were intermixed with quartz and perforating surface of coal particles.



Figure 6.3.1-12 The flow rates for LC 3, LC 10BC, LC 8U and LS 20.

Based on this information it can be concluded that handling behavior of LC 10BC coal is hindered by the presence of a large amount of clay material, while the high amount of clays in combination with highly contaminated with quartz coal surface led to the very poor handleability of LC 3 coal. The effect of clays on coal particles is especially pronounced at moisture content past the equilibrium moisture.

The surface moisture is the amount of moisture which is an excess moisture beyond the equilibrium moisture. The ability of fine particles to aggregate is controlled by the wettability characteristics and the mineral matter (ash) content of the fines. When fine particles aggregate, they stick together and reduce the handleability of the whole coal blend. The flowability of the coal blend should thus be influenced by the amount of fines, surface moisture and mineral matter (ash) content. This should be especially evident at the critical moisture level, where the deterioration starts. The correlation between amount of fines $(Y_{0.5})$, ash in fines and surface moisture (SM) was examined and is shown in Figure 6.3.1-14. The effect of amount of fines and moisture content in combination with the mineral matter (ash) content depends on the wettability. The effect is much stronger for hydrophilic particles and this is evident form the graph, an increase of fines with higher ash content along with the surface moisture reduces the flow rates of the bulk coal much faster than in hydrophobic coal samples, as discussed earlier.





To further investigate this effect, the correlation between the parameter derived from multiplying surface moisture and the ash fraction (AF_{0.5}) of the 0.5 mm fraction, (similarly as derived for 38μ m (AF_{0.038}) fraction by Brown and Atkin (2000)), at the critical moisture level (CM) and handleability index from the Durham Cone tests (DCI) was explored. The ash fraction was computed as follows:

$$AF_{0.5} = (\gamma_{0.5} A_{0.5}) / (A_{\text{tot}} \times 100)$$
(6-4)

where: $\gamma_{0.5}$ - yield of 0.5 mm fraction; $A_{0.5}$ - ash content in the 0.5mm fraction; A_{tot} - ash content in the bulk sample.



● hydrophobic coals ■ hydrophilic coals



The correlation is shown in the Figure 6.3.1-15. The higher the proportion of fines in the coal blend with a higher ash content, the lower the flow rate at the CM point of initial deterioration of handleability. The correlation factor is high ($R^2 = 0.79$) and indicates that a relationship exists between the surface properties related to wettability and ash content of fines. The correlations between amount of fines and flow rates (DCI index), as well as the correlation between surface moisture and DCI, handleability index were shown to be less significant and these plots are included in Appendix G. This confirms the theory that the amount of surface moisture and ash, has a prevailing effect on the bulk sample handleability and depends on wettability of coal samples.


● hydrophobic coals ■ hydrophilic coals

Figure 6.3.1-15 The effect of ash fraction $(AF_{0.5})$ and surface moisture on the flow rates for all studied coals.

6.3.1.5 Summary and discussion

From the bulk sample testing, it was confirmed that the amount of fines is a critical factor, but also the whole size distribution is an important parameter in determination of the bulk coal handleability. Widening the spectrum of sizes in the bulk sample was shown to improve handleability characteristics of the studied coals.

The critical moisture was derived from the flow rates at different moisture levels. The moisture content at which the highest fluctuations occurred was identified as the initial point of deterioration in handleability of the bulk coal. This parameter represents bulk sample characteristics, such as size composition of the sample as well as the quality of fines, that are responsible for deterioration of the bulk sample handleability. However, there was no meaningful correlation between the critical moisture and any other parameter (amount of fines, ash in fines or equilibrium moisture of fines) affecting handleability of the studied coal samples. The critical moisture is a result of all parameters acting together and reflects their shared effect on the bulk coal characteristic. The surface moisture of fines is an important parameter, which contributes to aggregation of fine particles. The surface moisture content depends on the equilibrium moisture, and equilibrium moisture is directly related to the surface properties of fine coal. This confirms that the wettability of fine coal particles is indeed a significant factor contributing to the handleability of the bulk coal. The handleability is hindered by the aggregation of fine particles and as confirmed by the bulk tests, the surface properties of coal particles have a strong effect on the handling characteristics of a coal blend. The pelletization tests confirm this relationship.

The bulk testing confirmed that coal samples with different wettabilities displayed different handling behavior. For hydrophobic coals, an increase in moisture was associated with decrease in the flow rate, but it did not lead to "no-flowing" conditions as confirmed by pelletization results. For hydrophilic coal particles, at low moisture contents, the flow rates were very high, but a drastic decrease in flow rate was observed past the equilibrium moisture content and this was also associated with reaching "no-flowing" conditions at high moisture content. This behavior was related to strongly aggregative properties of hydrophilic fines as predicted from pelletization tests. Summary of all parameters affecting bulk coal handleability is presented in Table G.4-1 in Appendix G.

6.3.2 Comparisons between the Durham Cone and the Handleability Monitor tests

The Handleability Monitor, as described in section 5.3.4 of the Chapter 5, has been used to measure the compressive strength of selected samples. In each test, approximately 50 kg of coal was used. The average value of the maximum pressure (P_{max}) obtained from " P_{max} -Time" trace was used as a measure of coal handleability as discussed in Chapter 5.

Since the value of the Handleability Monitor Index is expressed (in bars) as the Average Maximum Pressure, the AMP is used as the handleability assessment parameter. In the series of tests with the Handleability Monitor, two types of coal blends were used: two metallurgical and three thermal coals. The metallurgical coals were from Australia (coal #11 and #12) and thermal coals from the UK (coal #2, #4 and #5). These tests were carried out at several moisture levels and with different amounts of fines. The size distributions of the original samples and one with the added coarse fraction (coal #11) are presented in Table 4.1-2. The same samples were used for the flow tests using the Durham Cone. These

were performed immediately after the tests with the Handleability Monitor. The handleability indices from both methods were compared. The data for coal samples that were tested are given in Tables 6.1-1 and 6.3-2. The metallurgical samples were hydrophobic coals as indicated by low equilibrium moisture values and assessed by the penetration rate as shown in Figure 6.1.1-2. The thermal product coals had their equilibrium moisture increased and their penetration rate was similar to other hydrophilic coals.

Since these samples were coal blends, they were first examined at the moisture level at which they were received, then they were tested either at an elevated moisture content, increased fines or increased amount of coarse fraction. An increase in fines content was adjusted by adding the -9.5 mm fraction, while adding +9.5 mm fraction led to a relative reduction in yield of fines (Table 6.3-2).

Figures 6.3.2-1 and 6.3.2-2 are graphical illustrations of the results for metallurgical coals, and Figure 6.3.2-3 for thermal coals (summarized in Table 6.1-10). Figures 6.3.2-1 and 6.3.2-2 present flow patterns for Coal #11 and Coal #12, respectively, with varying amounts of fines and at different moisture levels as obtained from the tests with the Durham Cone. The AMP handleability index values are posted above the graph lines. The flow patterns for Coal #11 at 10 % moisture with 40 % fines coincides with the flow pattern for this sample at 6 % moisture with 50 % fines, with the greater variability found for the sample with higher fines content. At the same time, the AMP handleability index changed from 25 to 30 for these two samples. The difference between DCI (Durham Cone Index) for these samples was about 0.2 (0.74 and 0.92) (Table 6-10). There were similar trends in changes in handleability for these samples when tested by both methods. The sample with a higher amount of fines was slightly more difficult to handle, as assessed by both methods. Both indices were sensitive enough to detect these relatively small differences in the handleability. This particular fact allows us to match a change of 0.2 in DCI with the change of 5 in the AMP value. The marginal difference in these indices can lead to the conclusion that an increase in fines content from 40 to 50 % can have the same effect as changing moisture from 6 % to 10 % for this particular coal blend.



Figure 6.3.2-1 Flow rates for Coal #11, hydrophobic coal, at different moisture and fines content.

The flow rate (DCI) for coal #11 at a reduced level of fines and moisture appeared to be much improved, however the higher value of AMP indicated the opposite - i.e. deterioration in handleability for the same sample. This may be due to the size distribution in this sample being adjusted by adding an extra coarse coal and as a result the +9.5 mm fraction yield was increased by 10 % while the amount of fines was already significantly high (Table 4.1-2). This additional amount of coarse material probably affected more of the tests carried out using the Handleability Monitor than the Durham Cone. In the Handleability Monitor, the narrowing passage through which the sample is pushed is likely to be more restrictive for larger particles than the flow out of Durham Cone. Furthermore, for coal #12, an increase in the amount of fines from 23 % to 33 % had the same detrimental effect on the flow rate as determined from Durham Cone tests, as on the maximum pressure AMP determined using the Handleability Monitor.

The handling behavior of coals #2, 4 and 5 studied using the Durham Cone and the Handleability Monitor was relatively consistent. Coal #4 with the largest quantity of fines, high moisture and ash content, was characterized by the highest AMP and accordingly this sample did not flow when tested with the Durham Cone (Figure 6.3.2-3). For the two other samples similar indices were obtained from both methods. The correlation between DCI and AMP is shown in Figure 6.3.2-4. The correlation was obtained for all coal samples except the sample which displayed unusual behavior as discussed earlier.



Figure 6.3.2-2 Flow rates for Coal #12, hydrophobic coal at different fines content





6.3.3 Effect of wettability of coal on its handleability

In a separate series of experiments, seven samples were first tested using the Handleability Monitor and then with the Durham Cone. These were coal product samples of various sources and quality. The results are plotted in Figures 6.3.3-1, 6.3.3-2 and 6.3.3-3 (a, b). The tests were carried out at the "as received" moisture content, an increased moisture content and increased moisture content with added zeolite to absorb water from coal. The first point on each curve corresponds to the "as received" sample followed by the one with increased moisture and the last one, the sample with the absorbent.



Figure 6.3.2-4 Correlation between DCI and AMP handleability indices for studied coals.

For samples 3 and 7, the trends show that the change of the handleability with an increase in moisture from 8 to 10 % is consistently the same for both methods. From the shape of the curves, it is obvious that both samples are hydrophobic. A small decrease in flow rate with an increase in moisture content is consistent with the similar change in the AMP value (Figure 6.3.3-1a, b). Adding zeolite considerably improves the handleability of these coals and confirms their non-wettable character. This is consistent with the proposed wettability-aggregation model of the behavior of fines indicating that due to the hydrophobic nature of coal, water remains on the surface of coal particles and therefore is more available to be absorbed by zeolite powder.

Coal samples 1 and 5 (Figure 6.3.3-2 a,b) also appear to be hydrophobic and exhibit the same trends in behavior as samples 3 and 7 when tested using the Durham Cone. The AMP values obtained using the Handleability Monitor, however, indicate an improvement in the handling behavior with the addition of water. Both of these samples exhibit poorer handling when compared to the other samples. This may be due to the effect of the clay content or of a higher amount of fines. The improvement in handling with addition of water may be related to the fact that water added to the sample was not mixed properly and for hydrophobic coal stays trapped between particles in some pockets providing a lubrication effect when particles are pushed in the Handleability Monitor.



Figure 6.3.3-1 (a) Moisture versus DCI for samples 3 and 7. (b) Moisture versus AMP, (Pmax) for samples 3 and 7.



Figure 6.3.3-2 (a) Moisture versus DCI for samples 1 and 5. (b) Moisture versus AMP, (Pmax) for samples 1 and 5.

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Sample 2 - Sample 4 - Sample 6







In the Durham Cone tests, the sample was vibrated and this gave a chance for the water to spread around the particles and consolidate them. In addition, because water can penetrate through the sample, the effects of clays were also likely to be more pronounced. This is in agreement with other studies carried out using the Durham Cone that show that

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consolidating the coal sample prior to discharge decreases the flow rate (Hall and Cuttres, 1960).

Coal samples 2, 4 and 6 (Figure 6.3.3-3 a, b) displayed a different behavior as compared to the ones discussed above. Their patterns of change are typical for hydrophilic coals. For these three samples, dramatic change in flow rate for a relatively small raise in moisture content is indicative of being in close proximity to the critical moisture content. For sample #4, the addition of an absorbent improved flow rate only marginally, probably because only a small amount of water was available on the surface of coal. The 9 % moisture was possibly close to this coal equilibrium moisture.

This is possibly why in the Handleability Monitor test, where the penetration of water through the coal bed is less effective, there was no visible change in the AMP value. During the Handleability Monitor tests, it was likely that particles were acting as if they were "dry". For sample 2, the tested ranges of moisture content significantly exceeded the equilibrium moisture and as a result, the addition of the absorbent to the mixture radically improved the flow rate.

Whilst the behavior of sample 6 in the tests with the Handleability Monitor was similar to the hydrophilic sample, the behavior of this coal in the Durham Cone tests seemed unexpected. The fact that adding an absorbent did not improve handleability when tested with the Durham Cone can be explained by an insufficient quantity of absorbent that was added to the sample.

6.3.4 Summary and discussion

A comparison of the handleability indices derived from the two different experimental methods demonstrate that, when the samples are tested under the same conditions, the indices are comparable. However, when the samples are examined under changing conditions, increased moisture, or with the addition of an absorbent, the Durham Cone method is shown to be more sensitive to these changes. When considering both types of tests for assessing handleability, it needs to be recognized that the tests are carried out under different technical conditions; this results in some effects being more pronounced in one method than in the other. For handling conditions where the coal is subjected to vibrating or transfers that involve mixing or mechanical shaking, the Durham Cone test

would be more likely to be the appropriate choice. However, for coals which are handled without considerable vibration, or are pushed through the channels, the Handleability Monitor becomes the more suitable option.

Sample	Moisture %	Yield (wt %) fines	Ash (wt %) in fines	Ash (wt %) in bulk	DCI	DCI (std)	AMP	Equilibrium Moisture %
Coal 11 (added coarse)	8.0	35.52			1.51	0.11	45	1.51
Coal 11 (AR)	10.0	40.9	8.95	9.67	0.74	0.18	25	1.51
Coal 11 (added fines)	6.0	50.0			0.92	0.31	30	1.51
Coal 12 (AR)	15.7	27.3	15.13	10.46				1.91
Coal 12 (added fines)	14.0	33.0			0.66	0.21	35	1.91
Coal 12 (added coarse)	13.0	22.9			1.29	0.13	28	1.91
Coal 2 (AR)	9.0	8.5	14.87	11.70	1.02	0.16	18	4.43
Coal 4 (AR)	12.0	19.3	34.1	20.8	0.0		50	5.23
Coal 5 (AR)	10.0	12.5	23.6	11.5	1.03	0.12	12	1.98

Table 6.3-2. Sample characteristics of coal product samples.

Since the Handleability Monitor is much easier to use and offers a rapid way of obtaining the handling characteristics of the representative coal blend, it would therefore be more appropriate to prepare the samples according to their handling history prior to the testing with the Handleability Monitor.

Using the example of randomly chosen samples it was shown that understanding the effects of wettability on coal handling behavior can help to identify problems related to the increased moisture in the coal blend. The pattern of behavior for hydrophobic and hydrophilic coals can help in explaining some of the inconsistencies in the behavior of other coals.

6.4 Bulk density determination

The bulk density determinations were performed on three samples. The examined coal samples were: hydrophilic LC 8U, hydrophobic LC 3 and hydrophilic LC 3 OXY. The LC 3 and LC 8U samples were tested with different amounts of fines (-0.5 mm; 10 %, 20 % and 30 %) at increasing moisture levels. LC 3OXY was tested only at 30 % fines content. Samples were also tested at zero and 8 minutes conditioning-consolidation. The conditioning did not have a significant effect on changing the bulk density for either of the two tested samples (LC 3, LC 8U). For LC 8U (hydrophilic), the increase in bulk density (5 %) was only noticed for the sample with 30 % fines and at the moisture exceeding 8 % past the equilibrium moisture for this sample. For LC3, there was a 10 % increase in bulk density for the sample with 10 % fines over the entire range of moisture contents. The results from these tests are included in Appendix I. For all the other tests, 3 minutes conditioning was used as a standard procedure.

The change in bulk density for hydrophobic LC 3 samples with a varying amount of fines at different moisture levels is depicted in Figure 6.4-1. The size distribution of these samples was comparable to other tested coals, and only the amount of fines was varied. There was a consistent decrease in bulk density with the increase in moisture, and the minimum bulk density was reached at around 5 % moisture for samples with 10 % and 20 % fines. For LC 3 coal with 30 % fines, the minimum bulk density was found at a 5.9 % moisture content. The effect of moisture on the bulk density was found to be similar for all samples, the pattern of change was almost identical for samples with 10 and 20% fines and only slightly different for the sample with 30 % fines.



Figure 6.4-1 The change in bulk density (BD) with moisture for LC 3 coal, at different fines level.

For hydrophobic coals, the bulk density of coal decreases gradually with the increase of moisture, as the water stays on the surface of coal in between the solid particles, increasing the volume of the bulk solid (decreasing bulk density). The lowest bulk density (LBD) coincides with the funicular state, where most of the water already penetrated interparticle spaces, whereas the capillary state is reached at the highest bulk density level (HBD) and this is where particles fully aggregate and attain the lowest possible volume. This HBD is much higher than any bulk density achieved with dry particles due to the aggregation between all particles. This is common for both hydrophilic as well as hydrophobic coals, while the spread between the LBD and HBD is much greater for hydrophobic than for hydrophilic coals and it is also consistent with the findings by Hall and Cutress (1960).



Figure 6.4-2 The change in bulk density (BD) with moisture for LC 8U coal, at different fines level.

For the LC 8U hydrophilic coal, the patterns of change of bulk density with increasing moisture varied significantly for samples with different amounts of fines (Figure 6.4-2). The LC 8U with 10 % fines had a similar pattern of change of bulk density with moisture as hydrophobic coals, while for samples with 20 and 30 % fines, the change of bulk density was more dramatic over the same moisture range. All three plots intersected at 8 % moisture content and owing to the fact that equilibrium moisture for LC 8U fines is at 7.34 %, it is reasonable to assume that below 8 % moisture the coal particles in these samples should behave as dry granular material. As a result their packing characteristics are significantly different from the sample with surface moisture. The lowest bulk density for the LC 8U sample with 10 % fines coincided with 8 % moisture, while for samples with 20 % and 30 % fines content, the lowest bulk density was observed around 12 % moisture.

Comparison of the changes in bulk density with increasing moisture content for LC 3 and LC 3OXYcoals is shown in Figure 6.4-3. There is a different pattern of bulk density change with the moisture for these two coals. The pattern for LC 3 is typical for hydrophobic coal, while LC 3OXY behave as a hydrophilic coal. Since the LC 3OXY is the oxidized LC 3 coal, one can conclude that altering the wettability of coal can lead to changing packing characteristics of the coal particles in the presence of water. It is quite evident that the LBD moved from 5 % for LC3 to 12 % for LC 3OXY, while the HBD is almost in the same (20-22.5 %) moisture range. This further supports that surface wettability plays a dominant role in the behavior of particles in the presence of water.



Figure 6.4-3 The change in bulk density (BD) with increasing moisture for LC 3 coal (hydrophobic) and LC 3OXY coal (hydrophilic).

The effect of wettability on coal bulk density has only been addressed in a few studies (Hall and Cutress, 1960; Leonard et al., 1992 and Leonard et al., 1993). Generally, it has been accepted that the surface of coal is hydrophobic and the reason for bulk density to decrease when moisture increases could be explained by reaching consecutive stages from

the pendular stage to the capillary stage. This theory is based on the fact that water does not penetrate into the coal surface, but rather, stays as discrete droplets. Therefore, conflicting results have been obtained for coals of different rank or characteristics. For example, Hall and Cutress (1960) explained the effect of moisture on the Durham Cone handleability index and bulk density as going through all the above mentioned stages; however the fact that the lowest flow rate was observed at the highest bulk density (HBD) was left unexplained.

In at least two separate studies (Hall and Cutress, 1960 and Leonard et al., 1993) it has been shown that the addition of hydrocarbon oil to higher rank coals or modifying coal surface with chemicals to make it hydrophobic increased the bulk density over a wide range of moisture contents. The flattening of the bulk density curve resembles the pattern of bulk density change for hydrophobic coals and this also leads to the improvement of the handleability of these coals.

In this study, a bulk density assessment was performed to develop an understanding of bulk sample characteristics and their influence on the handleability of the tested coals. To accomplish this, the same samples were used for the bulk density measurements and the handleability tests using the Durham Cone. The results were analyzed in the view of the wettability-aggregation model proposed earlier with the emphasis on the wettability characteristics of tested coals. To facilitate this discussion, the results of the bulk density measurements and Durham Cone tests were summarized in Table 6.3-3.

In the case of hydrophilic LS 8U coal, it is apparent that as the amount of fines is increased, the LBD value moves to higher moisture contents. The values of critical moisture change differently for hydrophobic and hydrophilic coal. The increase in fines content has very little effect on the CM values for hydrophobic coals, and the LBD is exactly the same as CM. In the case of hydrophilic LC 8U coal, however, samples with more fines needed more moisture to reach the LBD. In both types of samples, the LBD coincided with CM or it was in a very close proximity to it. There is also a difference in the value of HBD for both of these coals. The HBD is the moisture content where the sample assumes the smallest volume and becomes the most compacted, or according to the proposed model, the particles fully aggregate. It can be noticed that HBD is much closer to the critical moisture for LC 8U hydrophilic coal, than it is for LC 3 hydrophobic coal. This

is also consistent with the handleability results obtained using the Durham Cone, where hydrophilic coals were reaching non-flowing conditions much sooner than hydrophobic coals.

Coals	Moisture % at (LBD) ¹	Critical Moisture % (CM)	Moisture % at (HBD) ²	Handleability index DCI at CM (kg/s)
LC3 (10% fines)	5.5	5.5	22.5	1.1
LC3 (20% fines)	5.0	5.0	20.0	0.7
LC3 (30% fines)	5.9	5.9	20.0	0.6
LC8U (10% fines)	8.0	8.5	17.0	1.7
LC8U (20% fines)	12.5	10.0	18.0	0.8
LC8U (30% fines)	12.0	11.0	20.0	0.6

Table 6.3-3. The results of the bulk density and Durham Cone tests for LC 3 and LC 8U coals.

¹ Lowest Bulk Density; ² Highest Bulk Density

6.5 Theory of flow based on the wettability-aggregation model

The particles are in a pendular state in the moisture content range from equilibrium moisture and the moisture level where coal sample assumes the lowest bulk density (LBD). The LBD moisture represents the point where the particles reach a funicular state. The funicular state extends from the LBD to the HBD moisture content, and at the HBD moisture level, capillary state is reached and particles fully aggregate. In order for hydrophobic coals to go through these three stages, they require the moisture content to increase somewhere from 1.3 % to over 20 %, while for hydrophilic coals this range is narrower and extends from 8 % to 20 % moisture. It is apparent that the funicular state is

more wide-ranging for hydrophobic coals than for hydrophilic ones. This implies that the capillary state is reached much quicker for hydrophilic coals than for hydrophobic coals leading to aggregation. The handleability deteriorates with increasing content of fines for both types of coals and so does the DCI index (Table 6.3-3). However, for a hydrophilic coal with very high fines content, and at the moisture content exceeding the critical moisture level, the sample eventually ceases to flow (Figure 6.3.1-1). It appears that for hydrophobic coals, more surface moisture is required to reach the capillary state and even when this stage is reached, the aggregation between the particles is not effective enough to stop the flow. Pelletization results confirmed this by the weaker pellets formed from hydrophobic coals.

According to the proposed wettability-aggregation model, for hydrophobic coals, in a pendular state, water stays in the form of discrete droplets, then with increasing moisture, the droplets join into a film in a funicular state, but the film is unstable due to the very small work-of-adhesion between the hydrophobic solid and water. Finally, as even more moisture is added, the capillary state is reached, but the surface forces between these particles are too weak to create a strong particle to particle bond (Figure 6.2-1b). For hydrophilic coal particles, once the film is formed around these particles, it is much stronger because porosity plays a role in creating a strong pulling force from within the coal structure. This film is very stable and as a result, less moisture is required to go from the funicular to the capillary state. The magnitude of the work of adhesion is also large and this results in a very strong particle-to-particle bonding, leading to strong and stable aggregates (Figure 6.2-2b). This is manifested by the strength of pellets formed from hydrophilic coals.

The bulk density results confirm the validity of the wettability-aggregation model, and provide evidence for the discussed flow theory of wet coal. The bulk density assessment also helps to link the effect of the bulk characteristics of coal particles in the presence of water with the handleability parameter such as critical moisture. This important conclusion can be used to set up a simplified procedure to predict handling properties of bulk samples. This is discussed in the following section.

6.5.1 Proposed quick method for evaluating coal handleability

The fact that the critical moisture (CM) always coincides with the moisture content at which the lowest bulk density (LBD) is reached, becomes an important point in setting up procedure for predicting the handling characteristics of the bulk sample from the bulk density determinations. From these tests, the lowest bulk density (LBD) can be determined and the critical moisture (CM) can be found (point of initial deterioration of handleability). The highest bulk density (HBD) is reached at the moisture content at which coal particles aggregate and handleability of the sample becomes critical. These two moisture levels will set the ranges for deterioration of handleability for a particular bulk sample.

Preparing final coal products at the coal preparation plant involves preparation of suitable blends from different cleaned coal fractions. The amount of fines (-0.5 mm) has a significant effect on coal handleability, therefore assessing the bulk density for the coal product with varying amounts of fines can help in setting up criteria for appropriate coal blending in order to avoid handling problems. Normally, to test handleability characteristics of the bulk sample, the series of Durham Cone tests are performed at a given moisture content and only behavior of the bulk sample at this particular moisture content is assessed. Testing handleability at various moisture levels is a lengthy and time consuming procedure. While with the use of new proposed method, the bulk density tests at various moisture limits within which coal handleability deteriorates, especially in terms of detecting the critical moisture level for a given bulk sample. This may be very important information for blending clean coal fractions as well as controlling the moisture content for the final coal blending problems, which are very costly.

The proposed method should include characterization of fines in terms of transmittance, equilibrium moisture, as well as the ash in the fines and in the whole blend. The transmittance and ash content determinations are routinely performed at coal preparation plants. The assessment of equilibrium moisture is also relatively easy to perform at the conventional coal preparation laboratory. The equilibrium moisture test will establish the moisture level in fines in order to prevent handleability problems.

To further investigate behavior of fines at high moisture content, pelletization of fines could be performed at the moisture content coinciding with the HBD. The pellets

strength should provide the information whether at this moisture content, the bridges built by such particles will be strong or weak.

Since changing wettability characteristics of coal particles leads to changing their flowability, the pelletization tests can also serve as a method to examine these changes in relatively simple way. The effect of adding surface altering chemicals can be quickly assessed with pelletization tests and this can provide valuable information in evaluating the effectiveness of such additives.

The prediction of DCI from the plot relating $AF_{0.5}$ and surface moisture versus DCI handleability index (Figure 6.3.1-15) may be possible; however more research is required to further verify this correlation. The method proposed here is based on the findings resulting from the research that included samples with varying wettability characteristics but of similar rank and origin. More tests are needed to confirm the validity of these findings for coals varying both in rank and origin.

CHAPTER 7. CONCLUSIONS AND RECOMMENDATIONS

The effect of surface properties of fine coal on the handleability of bulk coal was examined. The emphasis was placed on the wettability of coal fines, as the surface property which controls the behavior of fine particles in the presence of water. The following are the major conclusions.

7.1 Characterization of fines in terms of surface properties

- Three different methods were used to determine wettability of the studied coals: transmittance, penetration rate and equilibrium moisture. All three methods are shown to be in a very good agreement in assessing wettability of the studied coal fines.
- 2. The surface area and porosity measurements using N₂ as an adsorbate revealed that the porosity composition of the studied fines as well as the contribution of microporosity to the total surface area is an important wettability-related variable. The knowledge of the surface area and porosity configuration helped in understanding the effects of wettability on the aggregation ability of the tested coal particles.
- 3. Pelletization was used as a method of assessing the effect of coal particles interaction in the presence of water. It was shown that very strong pellets are formed from hydrophilic coal fines, while weak pellets are obtained from the hydrophobic coals.
- 4. Particle shape, roughness and mineral matter characteristics were studied in order to clarify the contribution from these physical properties of coal to the mechanical effects influencing the pellets formation and the resulting strength. It is concluded that properties such as aspect ratio, fractal dimension values or other surface characteristics such as rugousity or an external surface were not significantly different for tested fine coal and their influence on the pellets strength is negligible. Based on the mineral matter evaluations it was concluded

that clays and quartz are the dominant minerals present in the studied coals. Thus, the effects of mineral matter on the pellets' strength is determined by these two minerals. These effects were more pronounced for hydrophilic coals, indicating that the same mineral matter content has a much greater effect on pellets' strength for hydrophilic coals than for hydrophobic ones. The degree of liberation of mineral matter was shown to have a little effect on building the strength of aggregates formed from fine coal particles. It was concluded that the reason for overall bad handleability characteristics of two hydrophobic coals (LC3 and LC10BC) could be attributed to their high content of clays. Although, clays were present in high quantities in both of these samples, these coal samples were still handleable at high moisture contents due to their weak aggregative properties and hydrophobic nature.

- 5. Based on the assumption that pellet strength is the evidence for interparticle forces between the aggregating particles the wettability-aggregation model was proposed. The proposed model applies to hydrophobic and hydrophilic coals and uses wettability, surface area, porosity and mineral matter content (ash) to demonstrate the differences in pellets strength. The validity of the wettability-aggregation model has been confirmed by the pelletization results. Pelletization was found to be a valid method of testing ability of particles to aggregate and to predict the handling behavior of fine coal.
- 6. It was found from the angle of repose measurements that there are different patterns of flowing behavior with increasing moisture content for hydrophobic and for hydrophilic coal fines. For hydrophobic coal particles with increasing moisture up to the 10 %, the angle of repose increases, (flowability deteriorates) then levels off and then decreases again at a very high moisture content (20%). This indicates that at a high moisture content hydrophobic particles become less cohesive. For hydrophilic fines, the pattern is entirely different, there is a steady increase in the angle of repose indicating continuous deterioration in flowability for these fines without the improvement in flowability at high moisture content. This is especially obvious for the LC3 and LC3 OXY coal fines, where changed

wettability of fines led to the changes in the their flowability. These results demonstrate that indeed surface wettability affects flowability (handling behavior) of fines.

7.2 Bulk sample testing in assessing the handleability

- Bulk sample testing using the Durham Cone revealed that segregation of particles occurs during the testing which may affect the overall handleability results. It was also shown that not only the amount of fines is a critical factor, but the whole particle size distribution plays an important role. The wider the spectrum of sizes, the better the tolerance for an increased amount of fines (-0.5 mm). However, there is a limit for the top size in the bulk sample at which the large particles hinder the flow through a particular handling system.
- 2. "Critical moisture" is one of the important factors that affects bulk handleability, and is identified as the initial point of deterioration of flowing properties of bulk sample. In this study, the critical moisture was derived from the standard deviation of the flow rates at different moisture levels. It was statistically verified that, the moisture level at which the highest fluctuations occurred, was the primary point of deterioration of the handleability for the bulk coal sample.
- 3. It was observed that equilibrium moisture needs to be assessed in order to set the limits for good and bad handleability ranges for any given coal. For hydrophilic coals, equilibrium moisture is much higher and therefore these coals can tolerate higher moisture levels before they start to be difficult to handle. However, at the moisture levels exceeding the equilibrium moistures, a rapid deterioration of their handling behavior is observed, frequently leading to non-flowing conditions.
- 4. The effects of wettability of fine coal on the bulk sample handleability was shown to be significant as derived from the consecutive tests with Durham Cone. Different patterns of behavior were observed for hydrophobic and hydrophilic coals. In the case of hydrophobic coals only the mineral matter

affects significantly the handleability of such coals. For these coals, an increase in moisture content affects handleability to a certain level, but the flow does not cease completely. A high amount of clays in hydrophobic coals can have a very damaging effect on handleability. The only difference between the effect of clays on hydrophobic and hydrophilic coals is that for hydrophobic coals the aggregation of particles does not result in the formation of strong bridges (obstruction in the flow). Since the amount of fines is the most critical factor influencing handleability, in order to see the effect of coal surface properties, the tested samples must have the same size distributions, especially the fine fractions content.

- 5. It was concluded that the surface properties of the fines have a strong effect on the handling behavior of the bulk sample. The wettability, porosity and mineral matter content control the behavior of coal particles in the presence of water and thus particle-to-particle interactions. The surface moisture content, that is the amount of moisture in excess of equilibrium moisture, coupled with the mineral matter content, is the major cause for particles aggregation. For the bulk sample, the **critical moisture** is the point of declining of the handling characteristics. The correlation between the surface moisture and AF_{0.50}, ash fraction of fines (-0.5 mm) for tested coal samples was found to be valid at the critical moisture.
- 6. Comparison of the handleability results obtained using Durham Cone and Handleability Monitor revealed that the effect of testing conditions in both methods is quite significant. When the samples were tested under exactly the same conditions the indices were comparable, however, when the samples were examined under changing conditions (increase in moisture or addition of absorbent), the handleability indices were dissimilar. The Durham Cone method was shown to be especially sensitive to the effect of moisture changes. The Handleability Monitor method however, provides a quick way of obtaining handling characteristics of the representative coal blend.

- 7. Bulk density determinations for hydrophilic and hydrophobic coal samples revealed that there were visible differences in the way these samples behaved when the moisture was increased. For hydrophobic coal, the effect of increasing moisture is less pronounced than for hydrophilic coal. Altering wettability of coal surface as in LC3 and LC3 OXY samples, led to different effects of moisture on bulk density for these coals. It was shown that for hydrophilic coals, the range between critical moisture and moisture content at highest bulk density is much narrower, meaning that these coals will reach critical non-flowing or close to non-flowing conditions much sooner than hydrophobic ones. This was found to be in line with the findings from DC testing of other hydrophilic coals.
- 8. The observation that the lowest bulk density (LBD) was always reached close to the critical moisture was found to be useful in setting up simplified procedure for predicting the handling characteristics of the bulk samples. The surface properties parameters such as: (*i*) equilibrium moisture of fines (-0.5 mm); (*ii*) transmittance of fines; (*iii*) ash content of fines, and (*iv*) ash content of the whole coal blend could be used in the assessment of quality of fines as well as the wettability. From the bulk density tests the lowest bulk density value will determine the critical moisture (initial deterioration), and highest bulk density will be the critical point where a particular sample become unhandleable. The results of pelletization tests also indicate whether the fines are cohesive enough to form strong unbreakable bridges that affect handling.
- 9. The performed handleability tests have shown that understanding the effect of wettability on coal handling behavior helps identifying the problems caused by the increased moisture in the coal sample. This could also help in explaining some of the inconsistencies in the behavior of coals with different characteristics.

7.3 Recommendations

This is the first study in which the surface properties of coals have been used to explain the effect of moisture on handleability of fine coal. For the purpose of this research, the selected samples were chosen from one source and therefore, many of their physical characteristics were similar. As a result, in the final phase of the analysis, the proposed model for behavior of these coals was limited to certain type of handling behavior. To further confirm the wettability-aggregation model and to predict handleability from the proposed method requires more testing of coal samples with different handleability characteristics and perhaps of different origin.

More testing is also required with Handleability Monitor in respect to different preparation procedures to imitate handling conditions, and also in respect to the effects of moisture on hydrophobic and hydrophilic coals in order to derive relationship between surface moisture and ash fraction of fines (- 0.5 mm).

The future follow-up research on handleability should be directed towards testing bulk samples in relation to the wettability of the fines. The concept of critical moisture being in vicinity of the lowest bulk density (LBD) provides important information that can be used to detect handleability problems. Since the bulk density tests are simpler and quicker, they can be utilized as the alternative to the full-scale Durham Cone tests. As a result, more bulk density studies are required to test coals with different wettability characteristics and relate the bulk density values, at various moisture levels, to the handleability index (DCI) derived from the Durham Cone method.

The wettability of fines is a major factor that determines the way the particles behave in the presence of water. The addition of water absorbing chemicals (superabsorbent) should be tested on different types of fines. Pelletization can be used as a method for testing the aggregation behavior of these particles in order to assess deteriorating handling properties. The wettability-aggregation model should help in understanding the effect of surface properties of fines on the handleability and to select appropriate chemical additives to improve coal handling properties. An extensive research in the area of water adsorption on different types of fines, is necessary to evaluate the findings of the present study.

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APPENDIX

APPENDIX A

A.1 Additional size analysis of bulk samples

Table A.1-1.Size analysis of LS 20.

Size(mm)	LS 20 Cumulative wt% remaining
6.3-0.5	78.06
0.5-0.25	89.81
0.25-0.15	92.19
0.15-0.075	96.04
0.075045	97.43
0.045038	98.02
-0.038	100.00

A.2 Size analysis of fines (-0.5 mm fraction)

The size analyses were performed using Malvern Mastersizer 2000. The Mastersizer 2000, measures particle diameters from 0.02 to 2000 μ m. The Mastersizer examines the scattering pattern from a field of particles passed through a laser beam. With the knowledge of the properties of measured particles (i.e. refractive index, absorption), it is possible to calculate the size of particles that created a particular pattern.

The system is set up to measure particle size in wet or dry samples. All the size distributions of the studied fines (-0.5 mm) were performed in water. Each sample was placed in the tank with dispersant liquid, then sample was stirred from the controlled software. The pump forced sample to the cell via sample tubing located in the optical unit. With the use of a variable speed pump/stirrer, particles were suspended and circulated in the measuring field. A variable power ultrasonic system kept particles dispersed during the measurement. Figure A.2-1 presents photograph of the Malvern Mastersizer.



Figure A.2-1 Malvern – Mastersizer 2000 measuring setup.

Table A.2-1. Size analysis for LC met coal fines (-0.5 mm).







Table A.2-3. Size analysis for LC 3 coal fines (-0.5 mm).





Table A.2-4. Size analysis for LC 3OXY coal fines (-0.5 mm).



Table A.2-5. Size analysis for LC 10BC coal fines (-0.5 mm).

Table A.2-6. Size analysis for LC 10B coal fines (-0.5 mm).



Table A.2-7. Size analysis for LC 8U coal fines (-0.5 mm).





Table A.2-8. Size analysis for LS 20 coal fines (-0.5 mm).



Table A.2-9. Size analysis for Coal #2 coal fines (-0.5 mm).



Table A.2-10. Size analysis for Coal #4 coal fines (-0.5 mm).



Table A.2-11. Size analysis for Coal #5 coal fines (-0.5 mm).



Table A.2-12. Size analysis for Coal #11 coal fines (-0.5 mm).

		Result	Analysis Re	eport			. 1
lample Name: Coal #12 R3	sc)P Name:		Measured: Thursday, Janu	ary 29, 2004 1:27:4	0 PM	
ample Source & type: Iaria ample bulk lot ref:	Ma CO Re Ma	Measured by: contract Result Source: Measurement		Analysed: Thursday, Janua	PM		
article Name: arbon mixed with minera	Ac Hy	cessory Name: dro 2000S (A)		Analysis mode General purpose	lt	Sensitivity:	
article RI: .850 lispersant Name: Vater	Ab 0.2 Di 1.3	sorption: spersant RI: 330		Size range: 0.020 to 2000.000 Weighted Residual: 0.307 %		Obscuration: um 12.70 % Result Emulation Off	
oncentration: .0441 %Vol	Sp 3.7	кап ; 713		Uniformity:		Result units:	
pecific Surface Area: 163 m²/g	IC Surface Area: Surface Weighted Mean D[3,2]:		an D[3,2]:	Vol. Weighted Mean D[4,3]:			
d(0.1): 11.289	Jm	d(0.6)	: 109.637 uz		4(0.0	449 407	
4,5 4,5 4 (%) 3,6 3 3 3 3 3 2,5 2 5 1,5 1			10	100		000	
0.5 8 o	•	P	article Size (um)				
0.5 0.0 	Thursday, Janua	P iry 29, 2004 1:27 atrial [Sea Gun] Live	article Size (µm) :40 PM	time b %]	[Volume to 95]	The second second second	<u>ا</u> .

APPENDIX B.



B.1. Penetration rate measurements for studied coals. B.1.1. The effect of increasing compaction load.

Figure B.1.1-1. Penetration rates for LC 10BC. The effect of increasing compacting load (no-load, 5.5 kg, 11 kg).



Figure B.1.1-2. . Penetration rates for LC 10B. The effect of increasing compacting load (no-load, 5.5 kg, 11 kg).



Figure B.1.1-3. Penetration rates for LC 8U. The effect of increasing compacting load (no-load, 5.5 kg, 11 kg).



Figure B.1.1-4. Penetration rates for LC met. The effect of increasing compacting load (no-load, 5.5 kg, 11 kg).



Figure B.1.1-5. Penetration rates for LC thermal. The effect of increasing compacting load (no-load, 5.5 kg, 11 kg).



Figure B.1.1-6. Penetration rates for Coal #2. The effect of increasing compacting load (no-load, 5.5 kg, 11 kg).



Figure B.1.1-7. Penetration rates for Coal #4. The effect of increasing compacting load (no-load, 5.5 kg, 11 kg).



Figure B.1.1-8. Penetration rates for Coal #5. The effect of increasing compacting load (no-load, 5.5 kg, 11 kg).



Figure B.1.1-9. Penetration rates for Coal #11. The effect of increasing compacting load (no-load, 5.5 kg, 11 kg).



Figure B.1.1-10. Penetration rates for Coal #12. The effect of increasing compacting load (no-load, 5.5 kg, 11 kg).

B.2. Reproducibility of penetration rate results.



Figure B.2-1 Example of reproducibility of penetration rate results for LC met coal sample (at 5.5 kg compaction load).



Figure B.2-2. Example of reproducibility of penetration rate results for LC 3 coal sample (at 5.5 kg compaction load).

B.3. Transmittance measurements data.

Transmittance value / Coal sample	Transmittance readings (%)	Transmittance readings (%)- Average	Standard deviation (+/-)
LC 3	95.14	95.25	0.16
	95.36		
LC 10BC	80.93	80.90	0.99
	79.53		
LC 10B	56.19	56.24	0.03
	56.24		
LC 8U	26.06	26.06	0.14
	25.86		
LS 20	34.78	34.80	0.02
	34.81		
LC met	98.74	99.02	0.40
	99.31		
LC thermal	50.27	50.63	0.52
	51.00		
LC 3OXY	58.12	57.80	0.47
	57.47		

Table B.3-1. Transmittance (at 520 nm) data for studied coal fines.

APPENDIX C

C.1. BET Surface area measurements data







Figure C.1-2. Adsorption and desorption isotherms for LC 3OXY coal



Figure C.1-3.Adsorption and desorption isotherms for LC 3 coal.







Figure C.1-5.Adsorption and desorption isotherms for LC met coal.





C.2. Equilibrium moisture data

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Equilibrium moisture (%) / Coal sample	Equilibrium moisture readings (%)	Equilibrium moisture readings (%)-Average	Standard deviation (+/-)
LC 3	1.28	1.30	0.02
	1.31		
LC 10BC	1.67	1.65	0.02
	1.63		
LC 10B	2.95	2.96	0.01
	2.96	/	0.00
LC 8U	7.11	7.34	0.32
	7.56		
LS 20	8.02	8.02	0
	8.02	1.50	0.07
LC met	1.60	1.56	0.06
	1.51	0.00	0
LC thermal	2.09	2.09	0
	2.09	4.20	0.06
LC 30XY	4.34	4.38	0.00
G 1/2	4.42	4 42	0
Coal #2	4.43	4.43	0
0 1 14	4.43	5 77	0.22
Coal #4	5.04	5.25	0.55
C] #5	1.09	1 08	Ο
Coal #5	1.90	1.70	U
C1 #11	1.90	1 51	0.12
C041#11	1.37	1.31	0.12
Cool #17	1.42	1 01	0.13
Coal #12	1.01	1.71	0.15
	2.00		

Table C.2-1. Equilibrium moisture measurements data for studied coals.

APPENDIX D

D.1 Pellets' strength data

Coal	LC	LC	LC 3	LC	LC	LC	LC 8U	LS 20
/pellet	10BC	30XY		thermal	met	10B		
strength/	(kg/m^2)	(kg/m ²	(kg/m^2)	(kg/m^2)	(kg/m ²	(kg/m ²	(kg/m ²	(kg/m ²
reading	x10 ⁻⁶)	$x10^{-6}$)	x10 ⁻⁶)	x10 ⁻⁶)	x10 ⁻⁶)	$x10^{-6}$)	$x10^{-6}$)	x10 ⁻⁶)
1	14.25	14.30	8.53	20.27	7.55	11.45	22.06	258.68
2	14.25	14.30	9.15	19.50	6.99	11.92	19.41	289.86
3	12.83	15.73	8.56	20.27	8.13	11.80	20.17	300.50
4	15.67	12.87	8.50	21.29	7.35	12.61	20.83	317.20
5	12.80	15.39	7.53	20.24	7.16	11.13	19.05	266.20
6	14.11	13.11	8.80	18.20	7.20	11.04	19.95	299.97
7	11.55	14.50	8.13	21.59	7.61	12.44	20.43	271.21
8	14.30	14.10	9.06	18.50	7.79	10.44	20.13	320.75
9	13.90	15.99	9.10	21.30	8.20	10.82	18.11	260.43
10	14.10	12.55	8.06	18.90	7.09	12.98	20.10	280.24
Average	13.78	14.28	8.54	20.01	7.51	11.66	20.02	289.00
Standard deviation	1.07	1.13	0.52	1.20	0.43	0.83	1.05	20.59
95% confidence interval	(+/-) 0.66	(+/-) 0.70	(+/-) 0.32	(+/-) 0.74	(+/-) 0.27	(+/-) 0.51	(+/-) 0.65	(+/-) 12.76

Table D.11 Pellets' strens	2th of	f studied	i coal	fines
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D.2. Photographs of pellets.



Figure D.2-1. Pellets produced from LS 20, LC 8U and LC thermal coal.



Figure D.2-1. Pellets produced from LC met, LC 10BC, LC 3 and LC 30XY coal.

D.3 Fractal dimension calculation by NK method

Table D.3-1 Fractal dimension analysis for LC 10BC coal particles

Quantachrome Corporation Quantachrome Autosorb Automated Gas Sorption System Report							
	Aut	osorb for Win	dows® Vers:	ion 1.27			
Sample ID Description	MH-LC 10B/C MET Coal						
Jample Weight	3.6477 g NITROGEN	Outgas Temp	50.0 °C	Operator	MH/SF		
Cross-Sec Area	16.2 A ² /molecule	Outgas Time	70.0 hrs	Analysis Time	622.1 min		
NonIdeality	6.580E-05	P/Po Toler	0	End of Run	02/16/2004 22:15		
Station #	1	Bath Temp.	77.35	PC SW Version	1.27		
	Fina	l data temp	perature com	pensated.			
			•				
	NK Meth	od Fractal Din	nension (Ads	orption)			
	Radius of	V	apor-Liquid				
	Curvature [A] Inter	face Area (n	a²/g]			
	2.4962E+0	0	1.60798E+00)			
	2.1971E+0	0	1.77534E+00		방상법수는 것으로 문어 가지 않는다.		
	2.0311E+0	0	1.87857E+00	en in de la Breek Brein			
	1.9791E+0	0	1.92090E+00				
	1.8717E+0	0	2.01824E+00	5			
	1.8043E+0	0	2.08382E+00	2			
	1,7298E+0	0	2.16530E+00				
	1.5399E+0	0	2.39448E+00	5			
	1.38682+0	0	2.59944E+00)			
	1.3627E+0	0	2.63178E+00	2			
	1.3395E+0	0	2.709395+00				
	1,2864E+0	Ō	2.75399E+00	0			
	1.2596E+0	0	2.79826E+00	0			
	1.2247E+0	0	2.85838E+0				
	1.1241E+0	0	3.03148E+0	0			
	1.0347E+0	0	3.18064E+0	0			
	1.0237E+0	0	3.20565E+0	0			
	1.0076E+0 9.9037E-0	10	3.23/80E+0	0			
	9.7319E-0	1	3.30994E+0	0			
	9.5470E-0)1	3.34768E+0	0			
	9.3136E-0)1	3.39925E+0	0			
	9.0389E-0)1	3.49779E+0	0 0	n inghalalanga, salah		
a the second	8.4138E-C	1	3.53877E+0	0			
이 나는 문화	8.3508E-0)1	3.55078E+0	0			
la sulation 44 .	8.2739E-0		3.56323E+0	0	Weiter and the second		
	8.1075E-C)1	3.58887E+0	0			
	8.0027E-0)1	3.60213E+0	Ō			
	7.8225E-0)1	3.61598E+0	0			
	7.5694E-0)1)1	3.630/6E+0	0			
	7.0029E-0	ó1	3.66321E+0	Ŏ			
		Slope = -6.	825E-01				
	Thf	arcent = 4	998E-01				
	Correlation Coeff:	icient = 0	.990066				
NTV MA	thed Freetal Dimensi	(an n =) ?	6937400				

NK Method Fractal Dimension, D = 2.683E+00
Table D.3-2. Fractal dimension analysis for LC 3OXY coal particles.

Quantachroms Corporation Quantachroms Autosorb Automated Gas Sorption System Report Autosorb for Windows@ Version 1.27

Coal			۰.	
3.5289 σ				
3.5289 0				•
NITROGEN	Outgas 1	emp 50.0 °C	Operator	MH/SF
16.2 A*/molecule	Outgas 1	1me 27.0 hrs	Analysis Time	480.9 min
6.5808-05	P/Po Tol	er o	End of Run	02/24/2004 19:52
28.0134 g/mol	Equil T	tte 3	File Name	MHLC30XY . RAN
A set of the second second	Bath Ten	up. ≲ ?7.35	PC SW Version	1.27
Final	data	temperature com	pensated.	•
	•••••••••••••••••••••••••••••••••••••••	NAME AND A	· · · · ·	
NK Metho	d Fracts	1 Dimension /Ada	amption	
	a ssacca	T DIMONSTON (MUS.	·	
Radius of		Vapor-Liguid		
Curvature (A] I	nterface Area (m	*/a]	
2.5151E+00	-	6.38865E-01	· · · ·	
2.2119E+00	L.	7.22143E-01		
2.0338E+00		7.51217E-01		
1.97822+00		7.69273E-01		
1.9303E+00		7.85755E-01		
1.8749E+00	r	8.06009E-01		
1.8041E+00	ц.	8.33653E-01		•
1.73765+00		8.61641E-01		• •
1.64962+00		9.01646E-01		
1.53955+00		9.58344E-01		
1.38/15+00		1.04739E+00	*	
1.30805+00		1.05687E+00		
1.33905700		1.076048+00		
1 28728+00	· ·	1.00650E+00		·
1 26538-00	•.	1.1110/6+00		
1 22748400		1.12/235+00	1. A.	
1 19378-00		1.100428+00	i t	
1 13738+00		1.109435+00		
1 03418+00	··· 、	1.22//05400	•	
1 02018400		1.311905400		
1.0059E+00		1.339200400		•
9.9175E-01		1 352267+00	· · · · ·	
9:7628E-01		1 367505+00		
9.60248-01		1 383448+00		
9.4333E-01		1.400162+00	•	
9.3822E-01		1.40734E+00		
9.3266B-01		1.414838+00	1	
9.2591E-01		1.42284E+00		
9.1770E-01		1.43103E+00	, I	
9.0639E-01		1.43980E+00	F	
8.9074E-01		1.44907E+00	· · ·	
8.7206E-01		1.45920E+00		
8.4979E-01		1.47030E+00	ri i	
8.2501E-01		1.482622+00	I	
7.9328E-01		1.49613E+00		
7.55202-01		1.51208E+00		
6.05705-01		1.539892+00		
• •			•	•
S	lope =	-7.478E-01		· · · ·
Inter	cept =	1.1922-01	-	
Correlation Coeffic	ient =	0.002309		
		V. 372JUO		
od Fractal Dimensio	n, D =	2.748E+00		
	<pre></pre>	0.580 mol D/Po Toi 28.0134 g/mol Equil Ti 1 Bath Tam Final data NK Method Fracta Radius of Curvature (Å) 2.5151E+00 2.5151E+00 2.5151E+00 2.5151E+00 2.5151E+00 2.5151E+00 2.5151E+00 2.5151E+00 2.5151E+00 1.9702E+00 1.9702E+00 1.8749E+00 1.389E+00 1.389E+00 1.389E+00 1.389E+00 1.2872E+00 1.2872E+00 1.2872E+00 1.339DE+00 1.3274E+00 1.2872E+00 1.2872E+00 1.2872E+00 1.0059E+00 9.6024E=01 9.6024E=01 9.3622E=01	6.56(H=-03 P/PO Tole 0 28.0134 g/mol Equil: fime 3 Bath Temp: 77.35 Final data 3 NK Mathod Fractal Dimension (Ads Radius of Vepor-Liquit Curvature (A) Interface Area (m 2.5151E+00 6.36865E-01 2.2119E+00 7.22143E-01 2.0338E+00 7.5217E-01 1.9702E+00 7.69273E-01 1.8041E+00 8.3653E-01 1.6496E+00 9.01646E-01 1.649E+00 8.3653E-01 1.7376E+00 9.01646E-01 1.649E+00 1.05687E+00 1.3601E+00 1.06450E+00 1.3601E+00 1.07604E+00 1.3871E+00 1.0764E+00 1.3971E+00 1.2776E+00 1.2772E+00 1.1167E+00 1.2872E+00 1.1167E+00 1.393E+00 1.32466E+00 1.2274E+00 1.33196E+00 1.22776E+00 1.332466E+00 1.3344E+00 1.33196E+00 1.0201E+00 1.32466E+00 9.9175E-01 1.36246E+00	0:502-03 DYPO TOTET 0 End Of Run 28.0134 g/mol Equilifier 3 File Name 29.0134 g/mol Equilifier 3 File Name 29.0134 g/mol Equilifier 3 File Name Final data = temperature compensated. NK Method Fractal Dimension (Adsorption) Redination (Adsorption) 2.5151E+00 6.396852E-01 1.9702:2713E=00 1.87702:2713E=00 1.8792:2732E=00 1.8792:2732E=00 1.8602E+00 1.8602E+00 1.85352E=01 1.8532E=00

	Quantachrome A	Quantachron utosorb Automa	ted Gas Sor	on ption System Rep ion 1 27	ort
Sample ID	MH-LC3R2				
Description	Met Coal			Henrich (Martin 1. Standt Greenwald)	
Sample Weight	3.2713 g				n an Alland an Allanda an Allanda. An <u>Allanda an</u> Allanda an Allanda an Allanda.
Adsorbate Cross-Sec Area	NITROGEN 16.2 Å ² /molecule	Outgas Temp Outgas Time	50.0 °C 30.0 hrs	Operator Analysis Time	MH/SF 559.6 min
NonIdeality	6.580E-05	P/Po Toler	Q	End of Run	03/17/2004 00:46
Station #	1	Bath Temp.	5 77.35	PC SW Version	1.27
	Fine	l data temp	erature con	pensated.	
	NK Meth	od Fractal Din	nension (Ada	sorption)	
	Radius of		vor≁limid:		
	Curvature [A] Interi	ace Area [n	n²/g]	
	2.5070E+0	0	8.87130E-01		
	2.2021E+0 2.0388E+0	10 50	9.91451E-01 1.07152E+00		
	1.9872E+0	0	1.09879E+00	5	
	1.9323E+0	0	1.12784E+00)	
	1.8061E+C	0	1.20059E+00	5	
	1.7383E+(0	1.24359E+00	0	
신고 물건 이 영문	1.6496E+0)0)0	1.30577E+00	0 n	
	1.3859E+(0	1.52568E+0	Ő	mana ana ang ang ang ang ang ang ang ang
	1.3665E+(0	1.54121E+0	0	
	1.30438+0	0	1.564246+0		
	1.2923E+0	0	1.61947E+0	0	
	1.2600E+(0	1.65508E+0	0	
	1.2228E+0	0	1.096836+0	0	
	1.1246E+(50	1.81314E+0	Ŏ	
	1.0312E+(00	1.92259E+0	0	
	1.01956+0	JU 10	1.936806+0	0 0	
	9.9764E-	01	1.96427E+0	0	
an da an an an an an da da an	9.8714E-0	01	1.97867E+0	0	
	9.5462E-	J1 01	2.01625E+0	0	
그는 말을 하는 것이다.	9.2318E-	01	2.05440E+0	Ō	ede da local contra de la contra El contra de la contr
	9.1667B-	01	2.06339E+0	0	
	9.1089E-	01	2.07259E+0	0	鹿辺長 コンチャート しつ
	8.4634E-	01	2.14093E+0	0	
	8,3357E-	01	2.15446E+0	0	era zera de la de la c
	8.1846E-	D1 01	2.16865E+U	0	
	7.8098E-	01	2.19918E+0	0	
	7.5722E-	01	2.21578E+0	0	
	7.2734E-	01	2.23339E+0	0	병생님, 열양, 지난 글 신다
	0,70346-	V1	2.23230670		
		Slope = -7.	589E-01		
a da anti-constructional de la construcción de la construcción de la construcción de la construcción de la cons El construcción de la construcción d	Int	ercept = 2.	791E-01		
	Correlation Coeff	icient = 0	.990042		
NK Me	ethod Fractal Dimens	ion, D = 2.	759E+00		
·				***************************************	

Table D.3-3. Fractal dimension analysis for LC 3 coal particles

	Quantachrome A Aut	utosorb Automa osorb for Wind	ted Gas Sor lows® Versi	on ption System Rep lon 1.27	ort
ample ID escription	MH-LCThermal Bituminous coal				
omments ample Weight	3.2411 g				
lsorbate ross-Sec Area onIdeality olecular Wt	NITROGEN 16.2 Å*/molecule 6.580E-05 28.0134 g/mol	Outgas Temp Outgas Time P/Po Toler Equil Time	50.0 °C 20.0 hrs 0 3	Operator Analysis Time End of Run File Name	SF/MH 510.3 min 03/04/2004 16:52 MH-LCTHL.RAW
tation #	1 Fina	Bath Temp. 1 data temp	erature com	pensated.	1.21
		- 4			
	NK META	od Fractal Dim	ension (Ads	orption)	
	Curvature [Va A] Interi	ace Area [m	ı²∕g]	
	2.5006E+0	0	1.56533E+00		
	2.1946E+0 2.0361E+0	0	1.73700E+00 1.85486E+00		
	1.9841E+0 1.9298E+0	0 0	1.89372E+00 1.93649E+00		
	1.8688E+0 1.8115E+0	0	1.98754E+00 2.03884E+00		an cheville i suit. Benne setto contra
	1.7363E+0 1.6475E+0	0	2.11203E+00 2.20586E+00)	
	1.5413E+0 1.3843E+0	0	2.33058E+00 2.54289E+00)	
	1.3640E+0 1.3417E+0	0	2.56884E+00		
	1.3159E+0	0	2.64569E+00		
	1.2571E+0	0	2.74387E+00		
	1.1797E+0	0 0	2.88280E+00		
	1.1250E+0 1.0316E+0	10 10	2.98603E+00 3.16168E+00)	
	1.0256E+0 1.0093E+0	10 10	3.17695E+00 3.21336E+00)	shiriningi na art. Nigingi shirini shi
	9.9229E-0 9.7614E-0	1	3.25318E+00 3.29415E+00))	
	9.5388E-0 9.3376E-0)1)1	3.35261E+00 3.39985E+00	2	
	9.1148E-0 8.8161E-0	1	3.44827E+00 3.50002E+00	2	
	8.1105E-0 8.0150E-0)1	3.59486E+0) 	
	7.8979E-0	1	3.62585E+0	0	Handar († 1990) Hiller, skieler († 1990)
	7.6912E-0		3.65918E+0	0	
	7.5987E-(7.5254E-()1)1	3.67637E+0	0	
	7.4751E-(7.4545E-()1)1	3.71201E+0 3.73056E+0	0 0	
	6.8398E-()1	3.74958E+0	0	
		slope = -7.	026E-01		
	Inte	ercept = 4.	962E-01		
	Correlation Coeff:	lcient = 0	.992693		
NK Me	thod Fractal Dimens:	ion, D = 2.	703E+00		

Table D.3-5. Fractal dimension analysis for LC met coal particles.

	ort				
Sample ID Description	MH-STN2 MET Coal				
Sample Weight Adsorbate Cross-Sec Area NonIdeality Molecular Wt	2.8064 g NITROGEN 16.2 Å ² /molecule 6.580E-05 28.0134 g/mol	Outgas Temp Outgas Time P/Po Toler Equil Time	50.0 °C 20.0 hrs 0 3	Operator Analysis Time End of Run File Name	MH/SF 498.3 min 02/18/2004 16:42 MH-LCMET.RAW
Station #	1 Final	Bath Temp. data temp	77.35 Perature com	PC SW Version	1.27
	NK Metho	d Fractal Din	nension (Ads	orption)	
	Radius of Curvature [Å	Va [] Interi	apor-Liquid lace Area [n	ı²∕g]	
	2.5079E+00		1.02713E+00		
	2.0364E+00) 	1.23444E+00)	말했는 것 것 같은 것
	1.9838E+00)	1.26445E+00)	
	1.8687E+00	5	1.33636E+00		n de la companya de Novembra de la companya de la company
	1.8102E+00 1.7330E+00		1.37620E+00 1.43298E+00		
	1.6504E+00		1.49893E+00)	
	1.5366E+00 1.3848E+00		1.59869E+00 1.74831E+00) }	
a dha an shi ang baland. Ta ang ang ang ang ang ang ang ang ang an	1.3689E+00	2	1.76181E+00)	
	1.3442E+00 1.3194E+00	}	1.79020E+00 1.81951E+00		
	1.2932E+00	2	1.85098E+00	2	
	1.2616E+00	5	1.88977E+00 1.93725E+00)	
	1.1784E+0)	1.99549E+0)	
	1.1279E+00 1.0345E+00)	2.06235E+00 2.18402E+00)	
	1.0239E+0)	2.19958E+0)	en de la complete de En esta de la complete
	1.0154E+0)	2.21389E+00	2	
	9.8517E-0		2.26475E+0)	
	9.5958E-0. 9.4770E-0		2.30515E+0		
	9.0973E-0	.	2.37800E+0	0	
	9.0124E-0	1	2.38972E+0	0	
	8.8000E-0	I	2.41457E+0	0	
	8.6548E+0	1	2.42850E+0	0	
	9.3230E-0	1	2.45865E+0	0	
	8.1277E-0	1	2.47527E+0	0	
and the state	7.6351E-0	1	2.51238E+0	0	
ent in all a	7.3184E-0	1	2.53279E+0	0	
	0.920UE-U	1	2,555845+0	U	
		Slope = -7.	493E-01		Harland Angeler Her and a state of the state
	Inte	rcept = 3.	373E-01		
	Correlation Coeffi	cient = 0	.990130		
NK Me	thod Fractal Dimensi	on, D = 2.	749E+00		
					na straata oo shinta da ahaa ahaa ahaa Marka ahaa ahaa ahaa ahaa ahaa ahaa Marka ahaa ahaa ahaa ahaa ahaa ahaa ahaa

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Quantachrome Corporation Quantachrome Autosorb Automated Gas Sorption System Report Autosorb for Windows® Version 1.27							
Sample ID Description	MH-LC 10B - 0.5mm MET Coal						
Comments Sample Weight Adsorbate Cross-Sec Area NonIdeality Molecular Wt Station #	5.1467 g NITROGEN 16.2 Å*/molecule 6.580E-05 28.0134 g/mol 1 Final	Outgas Temp Outgas Time P/Po Toler Equil Time Bath Temp. data temp	50.0 °C 20.0 hrs 0 3 77.35 Perature com	Operator Analysis Time End of Run File Name PC SW Version pensated.	MH/SF 554.7 min 02/17/2004 19:31 MH-LC10B.RAW 1.27		
	NK Metho	d fractal Din	nension (Ade	orption)			
	Radius of Curvature (A	Vi] Interi	por-Liquid face Area [m	*/g]			
NK Mét	2.4939E+00 2.1937E+00 2.0341E+00 1.9823E+00 1.9274E+00 1.8677E+00 1.8677E+00 1.8101E+00 1.7345E+00 1.5396E+00 1.3630E+00 1.3630E+00 1.3630E+00 1.3630E+00 1.3630E+00 1.2291E+00 1.2249E+00 1.2249E+00 1.2249E+00 1.2249E+00 1.2249E+00 1.0334E+00 1.0268E+00 1.0334E+00 1.0268E+00 1.0091E+00 9.8000E=01 9.6284E=03 9.4066E=01 8.7437E=01 8.6527E=01 8.7437E=01 8.6525E=01 8.3696E=01 8.3696E=01 8.293E=01 7.9470E=01 7.9470E=01 7.7474E=01 2.293E=01 8.0855E=01 8.293E=01 8.293E=01 8.293E=01 8.293E=01 8.3696E=01 8.293E	Slope = -7. Scept = 2. Sient = 0 m, D = 2.	9.01014E-01 1.00654E+00 1.10397E+00 1.13276E+00 1.13276E+00 1.12476E+00 1.2467E+00 1.29647E+00 1.29647E+00 1.52214E+00 1.52214E+00 1.52214E+00 1.52214E+00 1.62164E+00 1.62164E+00 1.65721E+00 1.62164E+00 1.70713E+00 1.86457E+00 1.86457E+00 1.99639E+01 2.04219E+00 2.04219E+00 2.04219E+00 2.12319E+00 2.12319E+00 2.12319E+00 2.12319E+00 2.12319E+00 2.12319E+00 2.04219E+00 2.04219E+00 2.04219E+00 2.12319E+00 2.054E+00				

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Table D.3-6. Fractal dimension analysis for LC 10B coal particles.

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	Quantachrome Au	Quantachron tosorb Automa	ted Gas Sor	on ption System Rep	ort
	Auto	osorb for Win	dows® Vers:	ion 1.27	
Sample ID	MH-LC 8U 0.5mm				
Description Comments	Oxidized Coal				
Sample Weight	0.8836 g				
Adsorbate	NITROGEN	Outgas Temp	50.0 °C	Operator	MH/SF
NonIdeality	6.580E-05	P/Po Toler	0	End of Run	02/06/2004 19:03
Molecular Wt	28.0134 g/mol	Equil Time	3	File Name	MH-LC8U.RAW
Station #	I Final d	Bath Temp. lata NOT te	mperature c	PC SW Version	1.27
	NK Metho	d Fractal Dir	ension (Ads	orption)	
	Radius of	Ve	por-Liquid		
· · · · · · · · · · · · · · · · · · ·	Curvature [A] Interi	ace Area [m	1²/g]	
	2.5155E+00)	3.05383E+00)	
	2.1851E+00		3.40486E+00)	
	2.0369E+00 1.9862E+00)]	3.60368E+00)	
	1.9293E+00)	3.76976E+00	5	
	1.8693E+00)	3.88123E+00	2	
	1.7350E+00	, ,	4.17928E+00	5	•
	1.6505E+00)	4.37743E+00)	
	1.5383E+00)	4.67325E+00	2	
	1.3619E+00	j	5.17503E+00	5	an an an an 1919 an Alban an A
	1.3430E+00)	5.23293E+00	Qui talle a litta instantia.	n de la properte en la composition de l La composition de la c
	1.3146E+00 1.2937E+00))	5.32449E+00))	
	1.2621E+00)	5.50214E+00	5	
	1.2252E+0	2	5.63695E+00	Q	
	1.1309E+0	5	6.00586E+0	0	
	1.0381E+0)	6.38077E+00	0	
	1.0238E+00 1.0084E+00)	6.42891E+00	D 1	
	9.9177E-0	í i i i i i i i i i i i i i i i i i i i	6.54255E+00	0	
	9.7349E-0	1	6.60684E+0		eet and a second and a second s
a tanan ar	9.5454E-0		6.70428E+0	U	
	9.0670E-0	Ī	6.86000E+0	Ō	peti di de la combina de la
or the second	8.9673E-0		6,89532E+0	0	
	8.7633E-0	1	6.97527E+0	0	
	8.6616E-0	1	7.01970E+0	0	
	8.5469E+0	1	7.06607E+0	0	
	8,3135E-0	1	7.16357E+0	0	
	8.1520E-0	1	7.21642E+0	0	
	7.9514E-0 7.6804E-0		7.27098E+0	0	
an a	7.0929E-0	1	7.43805E+0	ŏ	
		Slope = -7	515R-01		
	Inte	rcept = 8.	026E-01		
	Correlation Coeffi	cient = 0	.992801		
NK Me	thod Fractal Dimensi	on, D = 2.	752E+00		

Table D.3-7. Fractal dimension analysis for LC 8U coal particles.

	Quantachrome Au Aut	Quantachron Itosorb Automa Osorb for Win	ne Corporati ited Gas Sor dows® Vers:	on ption System Rep ion 1.27	ort
Sample ID Description	MH-LS20 Subbituminous Coal				
Sample Weight Adsorbate Cross-Sec Area NonIdeality Molecular Wt Station #	3.0475 g NITROGEN 16.2 Å ³ /molecule 6.580E-05 28.0134 g/mol 1 Fina	Outgas Temp Outgas Time P/Po Toler Equil Time Bath Temp. 1 data temp	50.0 °C 48.0 hrs 0 3 77.35 Perature com	Operator Analysis Time End of Run File Name PC SW Version mensated.	SF/MH 1321.5 min 03/02/2004 12:05 MH-LS20.RAW 1.27
	NK Metho	od Fractal Din	nension (Ads	orption)	
	Radius of Curvature []	Va A] Interi	apor-Liquid face Area [m	i²/g]	
	2.7090E+0 2.4154E+0 2.1421E+0 2.0307E+0 1.9775E+0 1.9257E+0 1.8676E+0 1.8031E+0 1.7300E+0 1.302E+0 1.3629E+0 1.3822E+0 1.3838E+0 1.3142E+0 1.3142E+0 1.3142E+0 1.3142E+0 1.2558E+0 1.2193E+0 1.2558E+0 1.2193E+0 1.202E+0 1.0202E+0 1.0174E+0 1.0202E+0 1.0174E+0 0.3011E+0 1.0202E+0 9.5599E=0 9.3051E=0 9.3051E=0 0.3012E+0 0.3012E+0 0.3012E+0 0.3012E+0 0.3012E+0 0.3012E+0 0.3012E+0 0.3012E+0 0.3012E+0 0.3012E+0 0.3012E+0 0.3012E+0 0.392E+0 7.934E+0 7.3400E+0 0.3400E+0		7.53542E+00 8.09133E+00 9.19460E+00 9.75973E+00 9.75973E+00 1.01407E+01 1.2219E+01 1.2219E+01 1.32439E+01 1.32439E+01 1.55053E+01 1.55053E+01 1.55053E+01 1.55918E+01 1.63358E+01 1.63558E+01 1.9070E+03 1.9070E+03 1.9070E+03 1.9070E+03 1.9070E+03 1.9070E+03 1.9070E+03 1.9070E+03 1.9070E+03 1.9070E+03 1.9070E+03 1.9070E+03 1.9070E+03 2.03903E+03 2.03903E+03 2.03903E+03 2.03903E+03 2.03903E+03 2.15251E+03 2.15251E+03 2.15251E+03 2.25730E+03 2.3730E		
		Slope = -9.	340E-01		en marin (Communication), All and Aria 1993 - Communication (Communication) 1993 - Communication (Communication)
	Inte	rcept = 1.	279E+00		
NK Mei	Correlation Coeffi	cient = 0 on, D = 2.	.993203 934E+00		

Table D.3-8. Fractal dimension analysis for LS 20 coal particles.

D.4. Aspect ratio data

Coal /							
aspect	LC	LC 3	LC	LC	LC	LC 8U	LS 20
ratio	10BC		thermal	met	10B		
(d _{min} /d _{max}) readings							
1	0.82	0.85	1.00	0.68	0.86	0.67	1.0
$\frac{1}{2}$	0.84	0.85	0.69	0.48	0.88	1.0	0.43
3	0.89	0.90	0.87	0.96	0.52	0.98	1.0
4	0.97	0.67	0.91	0.88	0.49	0.89	0.50
5	0.72	0.66	0.55	0.57	0.72	0.76	0.93
6	0.84	0.78	0.98	0.80	0.98	0.71	0.70
7	0.66	0.93	0.96	0.61	0.98	0.77	0.87
8	0.89	0.76	0.88	0.41	0.79	0.98	0.61
9	0.64	0.75	0.78	0.81	0.80	0.71	0.48
10	0.93	0.86	0.62	0.92	0.38	0.90	0.92
11	0.93	0.73	0.62	0.54	0.89	0.50	0.68
12	0.82	0.59	0.71	0.65	0.82	0.90	0.88
13	0.68	0.97	0.85	0.56	0.56	0.63	0.67
14	0.75	0.72	0.82	0.84	0.95	0.56	0.78
15	0.69	0.76	0.79	0.84	0.83	0.60	0.85
16	0.98	0.80	0.79	0.87	0.90	0.67	0.74
17	0.67	0.88	0.57	0.95	0.68	0.73	0.80
18	0.67	0.59	0.89	0.51	0.90	0.83	0.80
19	0.82	0.93	0.62	0.87	0.72	0.81	0.86
20	0.36	0.40		0.62	0.69	0.58	0.90
Average	0.78	0.77	0.79	0.72	0.77	0.76	0.77
Standard							
deviation	0.15	0.15	0.14	0.17	0.17	0.14	0.17
95%	(+/-)	(+/-)	(+/-)	(+/-)	(+/-)	(+/-)	(+/-)
confidence interval	0.07	0.07	0.06	0.07	0.07	0.06	0.07

Table D.4-1. Aspect ratio measured for studied coal particles, using Olympus BX60 reflected light microscope with 500 magnification.

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APPENDIX E.

E. 1. Quantitative phase analyses using the Rietveld method and X-ray powder diffraction data. Table E.1-1. Results of quantitative phase analysis (wt. %) for LC thermal. LC met, LC 10BC, LC 8U, LC 10B, LC 3 and LS 20 coal samples.

	Ideal Formula	1	2	3	4	5	6	7
		LC thermal	LC met LTA	LC 10B/C LTA	LC 8U LTA	LC10B LTA	LC3 LTA	LS 20 LTA
Quartz	SiO ₂	29.5	26.8	35.8	27.3	31.1	33.7	42.2
Palygorskite	(Mg,Al) ₂ Si ₄ O ₁₀ (OH)·4H ₂ O	9.8	6.0	10.4	6.4	4.7	6.9	
Muscovite	KAl ₂ هاSi ₃ O ₁₀ (OH) ₂	13.9	9.9	10.1	9.6	9.4	6.9	32.3
Kaolinite	Al ₂ Si ₂ O ₅ (OH) ₄	36.9	41.7	36.7	47.1	46.2	48.4	16.9
Gypsum	CaSO ₄ ·2H ₂ O	6.7	4.3	2.6	6.4	3.7	2.4	3.0
Calcite	CaCO ₃							5.6
Ankerite	$Ca(Fe^{2+},Mg,Mn)(CO_3)_2$	4					0.1	
Hematite	α-Fe ₂ O ₃	1.2	4.7	1.8	0.6	3.2		
Magnetite	$Fe^{2+}Fe_{2}^{3+}O_{4}$		3.7		1.6			
Siderite	Fe ²⁺ CO ₃	0.6	0.7	2.6		1.1	0.9	
Rutile	TiO ₂		1.0		1.0	0.7	0.7	
Dawsonite	NaAl(CO ₃)(QH) ₂		1.3					
Zircon	ZrSiO ₄	1.3						
Total		100.0	100.0	100.0	100.0	100.0	100.0	100.0



Figure E.1-1. Rietveld refinement plot for sample 1 (LC thermal).



Figure E.1-2. Rietveld refinement plot for sample 2 (LC met LTA).



Figure E.1-3. Rietveld refinement plot for sample 3 (LC 10BC LTA).





Figure E.1-5. Rietveld refinement plot for sample 5 (LC 10B LTA).



Figure E.1- 6. Rietveld refinement plot for sample 6 (LC 3 LTA).



Figure E.1-7. Rietveld refinement plot for sample 7 (LS 20 LTA).

E. 1 The effect of mineral matter type on pellets' strength



□ hydrophilic coals ○ hydrophobic

Figure E.1-1 The effect of total clays on pellets' strength



o hydrophobic coals □ hydrophilic coals

Figure E.1-2 The effect of quartz content on pellets' strength

APPENDIX F.

F.1 Angle of repose data.

Coal	. The angle	of repose de	ita 101 Studi	eu coar mies	, at given inc	
/angle of repose	LC 10BC	LC 3OXY	LC 3	LC 8U	LC 10B	LS 20
(deg)/	at 1.42%	at 2.10%	at 0.2%	at 1.96%	at 1.00%	at 2.30%
reading	H ₂ 0					
Run #1					111, 11 - L	
1	50	45	48	48	45	43
2	48	40	43	40	45	40
3	48	45	4 <u>5</u>	. 48	40	41
4	44	43	45	42	40	39
Run #2						
1	48		50		45	40
2	45		47		45	40
3	42		45		43	35
4	48		45		40	38
Average	47	43	46	45	43	40
Standard deviation	3	2	2	4	3	2
95% confidence interval	(+/-) 2.08	(+/-) 1.39	(+/-) 1.39	(+/-) 3.92	(+/-) 2.08	(+/-) 1.39

Coal /angle of repose (deg)/ reading	LC 10BC at 1.79% H ₂ 0	LC 30XY at 2.80% H ₂ 0	LC 3 at 2.08% H ₂ 0	LC 8U at 3.40% H ₂ 0	LC 10B at 2.44% H ₂ 0	LS 20 at 2.90% H ₂ 0
Run #1					•	تو
1	55	50	45	50	55	48
2	50	45	45	50	50	50
3	55	45	40	50	55	50
4	52	45	48	44	52	45
Run #2						
1	48		48		48	46
2	52		48		52	45
3	50		42		50	48
4	54		45		54	50
Average	52	46	45	49	52	48
Standard deviation	3	3	3	3	3	2
95% confidence interval	(+/-) 2.08	(+/-) 2.94	(+/-) 2.08	(+/-) 2.94	(+/-) 2.08	(+/-) 1.39

Table F.1-2. The angle of repose data for studied coal fines, at increased moisture levels.

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Coal /angle of repose (deg)/ reading	LC 10BC at 6.60% H ₂ 0	LC 30XY at 4.20% H ₂ 0	LC 3 at 5.45% H ₂ 0	LC 8U at 8.05% H ₂ 0	LC 10B at 5.62% H ₂ 0	LS 20 at 9.47% H ₂ 0
Run #1						
1	65	57	45	65	65	52
2	55	52	50	65	60	58
3	60	55	50	60	60	58
4	65	50	55	55	65	60
Run #2						
1	55	55	48	65	55	55
2	58	52	50	60	60	50
3	55	48	45	60	63	50
4	62	52	52	58	58	60
Average	59	53	49	60	61	56
Standard deviation	4	3	4	4	3	4
95% confidence interval	(+/-) 2.77	(+/-) 2.08	(+/-) 2.77	(+/-) 2.77	(+/-) 2.08	(+/-) 2.77

Table F.1-3. The angle of repose data for studied coal fines, at increased moisture levels

Coal /angle of repose (deg)/ reading	LC 10BC at11.33 % H ₂ 0	LC 3OXY at 9.50 % H ₂ 0	LC 3 at 8.91 % H ₂ 0	LC 8U at 12.30% H ₂ 0	LC 10B at 8.79% H ₂ 0	LS 20 at 13.70% H ₂ 0
Run #1						
1	70	65	70	60	70	60
2	68	65	65	60	68	55
3	70	60	65	60	68	55
4	65	65	60	55	70	60
Run #2						
1	65	60	68	55	65	50
2	68	65	70	58	65	60
3	72	58	65	55	70	55
4	68	60	70	60	68	60
Average	68	62	67	58	68	57
Standard deviation	3	3	3	2	2	4
95% confidence interval	(+/-) 2.08	(+/-) 2.08	(+/-) 2.08	(+/-) 1.39	(+/-) 1.39	(+/-) 2.77

Table F.1-4. The angle of repose data for studied coal fines, at increased moisture levels.

Coal /angle of repose (deg)/ reading	LC 10BC at 21.8% H ₂ 0	LC 3OXY at 15.9 % H ₂ 0	LC 3 at 20.3 % H ₂ 0	LC 8U at 18.0 % H ₂ 0	LC 10B at 20.3 % H ₂ 0	LS 20 at 22.0 % H ₂ 0
Run #1						
1	70	70	60	70	75	60
2	70	70	60	70	70	65
3	70	62	60	68	70	70
4	68	70	60	. 65	68	- 65
Run #2						
1	75	65	65	70	70	65
2	72	70	65	75	68	65
3	70	68	60	70	70	70
4	75	68	60	68	70	68
Average	71	68	61	69	69	. 66
Standard deviation	2	3	1	3	1	3
95% confidence interval	(+/-) 1.4	(+/-) 2.1	(+/-) 0.7	(+/-) 2.1	(+/-) 0.7	(+/-) 2.1

Table F.1-5. The angle of repose data for studied coal fines, at increased moisture levels.

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APPENDIX G.

G.1 Examples of raw data from Durham Cone tests Table G.1-1 1

Cumulative	LC8U	LC 8U	LC 8U	LC 8U	LC10BC	LC10BC	LC10BC	LC10BC	LC 3				
time of runs, (s)	at4% H_20	at 10%	at 15%	at 19%	at 4%	at 6%	at 10%	at 15%	at 2%	at 5%	at 10%	at 15%	at 20%
		H ₂ 0											
								0.00	1 76	0.07	0.00	0.00	0.46
30	2.28	1.88	0.88	0.13	1.77	1.21	0.27	0.38	1.76	-0.07	0.29	0.00	0.46
60	2.65	0.37	0.75	0.14	1.67	0.71	0.29	0.65	1.66	0.09	0.55	0.50	0.37
90		0.85	0.50	0.00	1.84	0.94	0.41	0.60	1.49	0.08	0.00	0.19	0.36
120	2.42	1.14	0.30	0.00	1.89	1.28	0.22	0.17	1.88	0.85	0.51	0.18	0.24
150	2.29	0.44	0.00	0.00	1.57	1.12	0.80	0.61	2.17	1.04	0.07	0.34	0.30
180	2.29	0.91	0.00	0.00	1.48	1.16	0.79	0.25	2.06	0.34	0.84	0.35	0.26
210	2.41	1.63	0.00	0.00	1.80	1.09	0.71	0.61	1.96	0.11	0.00	0.59	0.31
240	2.43	0.63	0.00	0.00	1.85	1.15	0.54	0.44	1.56	0.36	0.69	0.00	0.42
270	2.27	0100			1.87	1.27	0.69	0.51	1.88	0.49	0.67	0.00	0.50
300	2.27				1.90	0.91	0.66	0.42	2.12	0.28	0.11	0.43	0.36
330					1.91	0.89	0.46	0.57	2.00	1.07	0.74	0.00	0.14
360					1.92	1 12	0.70	0.65	2.10	0.92	0.13	0.37	0.44
300					1.92	0.57	0.31	0.36	1.88	1.24	0.68	0.52	0.44
420					1.91	1 24	0.32	0.51	2 29	0.39	0.00	0.58	0.36
420					1.91	0.93	0.67	0.36	2 17	0.29	0.98	0.94	0.29
430	2 20	0.00	0.20	0.04	1.91	1.04	0.57	0.50	1 03	0.51	0.42	0 33	0.35
average	2.38	0.98	0.30	0.04	1.01	1.04	0.32	0.15	0.22	0.31	0.72	0.33	0.10
Standard	0.13	0.54	0.37	0.07	0.14	0.23	0.21	0.15	0.23	0.40	0.55	0.27	0.10
deviation													(
95%confidence	(+/-)	(+/-)	(+/-)	(+/-)	(+/-)	(+/-)	(+/-)	(+/-)	(+/-)	(+/-)	(+/-)	(+/-)	(+/-)
interval	0.09	0.37	0.26	0.05	0.07	0.12	0.11	0.08	0.12	0.20	0.18	0.14	0.05

Cumulative time of runs, (s)	LC 10B at 1%H ₂ 0	LC 10B at 5% H ₂ 0	LC 10B at 9% H ₂ 0	LC 10B at 17% H ₂ 0	LC 10B at 20% H ₂ 0	LC thermal at 1 %	LC thermal at 6%	LC thermal at 15%	LC met at 2% H ₂ 0	LC met at 5% H ₂ 0	LC met at 10% H ₂ 0	LCmet at 15% H ₂ 0	LC met at 19% H ₂ 0
						H ₂ 0	H ₂ 0	H ₂ 0					
20	1 2 2	0.46	0.63	0.20	0.26	1 70	1 1 5	0.32	1.60	1 20	0.74	0.18	0.45
30	1.52	0.40	0.05	0.50	0.30	1.70	0.22	0.52	1.00	0.61	0.14	0.10	0.45
60 00	1.02	0.47	0.40	0.21	0.23	1.40	0.25	0.15	1.02	1 27	0.19	0.10	0.10
90	1.08	1.29	0.47	0.31	0.17	1.52	1.23	0.10	1.42	1.57	0.19	0.00	0.00
120	1.23	1.03	0.79	0.33	0.14	1.24	0.33	0.10	1.21	0.00	0.04	0.48	0.00
150	1.29	0.78	0.82	0.19	0.18	1.08	0.23	0.10	0.90	0.83	0.40	0.45	0.50
180	1.13	0.68	0.98	0.28	0.16	1.13	0.68	0.10	0.92	0.82	1.04	0.57	0.00
210	1.12	0.83	0.89	0.19	0.17	0.85	0.77	0.10	0.82	1.13	1.04	0.10	0.42
240	1.21	0.78	0.00	0.00	0.16	0.76	0.90	0.10	1.23	0.70	0.65	0.56	0.26
270	1.38	0.42	0.06	0.11	0.16	1.46	0.39	0.10	1.32	1.32	0.96	0.45	0.80
300	1.19	0.86	0.90	0.41	0.12	1.21	0.00	0.04	1.39	1.09	0.80	0.33	0.01
330	1.25	0.46	0.25	0.23	0.13	1.34	0.67	0.10	1.50	0.93	0.72	0.70	0.49
360	1.32	0.75	0.00	0.37	0.08	1.50	0.22	0.10	1.56	1.32	0.22	0.76	0.56
390	1.28	0.47	0.90	0.34	0.11	1.38	0.75	0.04	1.40	1.10	0.80	0.70	0.47
420	1.25	0.98	0.00	0.29	0.12	1.35	0.27	0.04	1.30	1.13	1.13	0.38	0.76
450	1.19	1.08	0.11	0.33	0.12	1.36			1.36	1.51		0.69	0.36
average	1.22	0.76	0.48	0.26	0.16	1.29	0.56	0.10	1.30	1.05	0.68	0.43	0.35
Standard deviation	0.10	0.27	0.38	0.11	0.07	0.26	0.38	0.07	0.26	0.27	0.32	0.24	0.27
95%	(+/-)	(+/-)	(+/-)	(+/-)	(+/-)	(+/-)	(+/-)	(+/-)	(+/-)	(+/-)	(+/-)	(+/-)	(+/-)
confidence interval	0.08	0.11	0.30	0.06	0.04	0.13	0.19	0.04	0.14	0.14	0.17	0.12	0.14

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Table G.1-2

Cumulative	LC 10B	LC 10B	LC 10B	LC 10B	LC 10B	LS 20	LS 20	LS 20	LS 20	LC 3 `ROM	LC 3 ROM	LC 3 ROM	LC 3 ROM
(s)	ROM at 1% H ₂ 0	ROM at 6 % H ₂ 0	ROM at 11% H ₂ 0	ROM at 15% H ₂ 0	ROM at 20% H ₂ 0	at 4% H ₂ 0	at 11% H ₂ 0	at 14% H ₂ 0	at 20% H ₂ 0	at 3% H ₂ 0	at 7% H ₂ 0	at 12% H ₂ 0	at 15% H ₂ 0
2.0	0.05	0.07	0.48	0.56	0.22	2.57	1.06	1 20	0.14	1.85	1.21	0.70	0.42
30	2.25	0.96	0.48	0.56	0.32	2.57	1.90	0.69	0.14	1.65	0.07	0.70	0.12
60	1.89	0.73	0.83	0.66	0.35	2.15	1.00	0.00	0.17	1.54	0.97	0.02	0.97
90	1.74	1.00	0.28	0.52	0.33	2.98	1.00	0.02	0.00	1.02	0.24	0.97	0.72
120	1.90	0.62	0.80	0.57	0.31	2.08	1.54	0.97	0.00	1.57	1.00	0.04	0.72
150	1.76	1.27	0.97	0.52	0.30	2.30	1.24	0.62	0.00	1.65	0.49	0.01	0.02
180	1.61	1.01	0.55	0.67	0.24	2.36	1.10	0.90	0.00	1.30	0.40	0.70	0.75
210	1.89	0.56	0.68	0.34	0.24	2.52	0.90	1.00	0.00	1.79	0.01	0.91	0.52
240	1.60	1.38	0.98	0.64	0.29	2.47	0.70	0.85	0.00	1.92	0.49	0.72	0.85
270	1.71	0.93	0.97	0.52	0.29	2.30				1.79	0.97	0.75	0.70
300	2.10	0.85	0.81	0.52	0.35	2.02				1.92	0.97	0.53	0.08
330	1.79	0.91	0.30	0.46	0.36	2.40				1.97	0.96	1.06	0.78
360	1.77	1.05	0.95	0.50	0.35	2.43				1.80	1.30	0.72	0.90
390	1.73	1.10	0.82	0.64	0.37	2.32				1.91	1.28	0.80	0.87
420	1.59	1.13	0.43	0.44	0.38	2.28				1.95	0.69	0.77	0.70
450	1.82	0.65	0.69	0.60	0.35					2.13	1.04	0.77	0.71
average	1.81	0.94	0.70	0.54	0.30	2.41	1.34	0.91	0.04	1.81	0.89	0.76	0.72
Standard	0.18	0.23	0.24	0.09	0.04	0.24	0.65	0.16	0.07	0.18	0.31	0.16	0.14
05%	(+/_)	(+/-)	(+/-)	(+/-)	(+/-)	(+/-)	(+/-)	(+/-)	(+/-)	(+/-)	(+/-)	(+/-)	(+/-)
confidence	0.09	0.12	0.13	0.05	0.02	0.13	0.45	0.11	0.05	0.09	0 .16	0.08	Ò.09
interval	0.09	~~~~											

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Table G.1-4								·	
Cumulative time of	LC 8U added fines	LC 8U added fines	LC 8U added fines	LC 8U added fines	LC 10BC added fines	LC 10BC added fines	LC 10BC added fines	LC 10BC added fines	LC 10BC added fines
runs, (s <i>)</i>	at 4% H ₂ 0	at 10% H ₂ 0	at 15 %H ₂ 0	at 20% H ₂ 0	at 3 % H ₂ 0	at 5 % H ₂ 0	at 10 %H ₂ 0	at 15 % H ₂ 0	at 20 % H ₂ 0
30 60	2.88 2.45	1.38	1.00 0.95	0.58 0.54	1.06 1.64	0.76 0.48	0.00 0.00	0.29 0.22	0.13
90 120	2.28	1.42	0.85	0.42	0.90	0.86	0.00	0.22	0.09
150	2.20	1.43	1.03	0.33	1.53	0.72	0.25	0.28	0.12
210	2.74	1.33	0.96	0.30	1.62	0.74	0.64 0.56	0.29	0.10
240 270	2.50 2.78	1.34 1.44	0.84 0.85	0.23 0.23	1.09 1.65	0.55	0.00 1.33	0.24 0.29	0.15 0.09
300 330	2.35 2.84	1.45 1.36	0.76 0.81	0.21 0.20	1.19 1.68	0.95 0.94	0.00 0.00	0.20 0.29	0.14 0.11
360 390	2.91 2.41	1.36 1.50	0.71 1.09	0.23 0.20	0.99 1.21	0.94 0.44	0.30 0.53	0.22 0.25	0.09 0.10
420 450	2.78 2.97	1.22 1.35	0.91 1.07	0.23 0.22	1.38 1.43	0.93 1.00	0.39 0.67	0.29	0.09
average	2.61	1.38	0.92	0.32	1.31	0.80	0.32	0.26	0.12
Standard deviation	0.26	0.07	0.11	0.13	0.28	0.20	0.38	0.04	0.03
95% confidence interval	(+/-) 0.13	(+/-) 0.04	(+/-) 0.06	(+/-) 0.07	(+/-) 0.14	(+/-) 0.10	(+/-) 0.19	(+/-) 0.02	(+/-) 0.02

G.2 Reproducibility of Durham Cone results.

The reproducibility of selected results from Durham Cone tests is presented in Table G.2-1. Replicates of Durham Cone tests were performed for each coal on six to seven representative samples, at two conditioning times: 0 and 30 seconds. The examples of reproducibility for series of handleability tests for LC 8U and LC HR-8 (random coal product sample), at a given moisture level are shown in Figures G.2-1; G2-2 and G-2-3, respectively.

Table 0.2-1. DAu		reproduce								
Coal at a given	LC 3	LC 3	LC 3	LC 3	LC 3	LC 3	LC 3	LC 3	LC 3	LC 3
moisture level/	2 %	2 %	5 %	5 %	10 %	10 %	15 %	15 %	20 %	20 %
Sample #										
Conditioning	0	30	0	30	0	30	0	30	0	30
Time (s)										• •
1	1.88	1.76	2.31	0.07	0.99	0.29	0.00	0.00	0.12	0.46
2	2.20	1.27	1.77	0.93	1.00	0.55	0.80	0.47	0.61	0.44
3	2.77	2.31	1.49	0.76	0.98	0.61	0.71	0.58	0.57	0.26
4	2.31	1.66	1.08	0.66	1.11	0.64	0.80	0.38	0.73	0.41
5	2.12	1.62	0.53	0.61	0.99	0.00	0.63	0.54	0.26	0.31
6	1.98	1.56	2.61	1.29	1.10	0.13	0.94	0.54	0.29	0.31
7	2.01	1.79	1.98	0.24			0.58	0.36	0.21	0.13
average	2.18	1.71	1.68	0.65	1.03	0.37	0.64	0.41	0.40	0.33
Standard deviation	0.30	0.32	0.72	0.41	0.06	0.27	0.31	0.20	0.23	0.12
95% confidence	(+/-)	(+/-)	(+/-)	(+/-)	(+/-)	(+/-)	(+/-)	(+/-)	(+/-)	(+/-)*
interval	<u>0.22</u>	<u>0.24</u>	0.53	0.30	0.05	0.22	0.23	0.15	0.17	0.09

Table G.2-1. Example of reproducibility of Durham Cone tests for LC 3 coal.

G.2 Reproducibility of Durham Cone results continuation



Figure G.2-1. Duplicate runs for LC 8U coal sample (at 12 % moisture).



Figure G.2-2. Duplicate runs for LC HR-8 (randomly selected) coal product (at 8 % moisture).



Figure G.2-3. Duplicate runs for LC HR-8 (randomly selected) coal product (at 4 % moisture).





Figure G.3-1 The effect of amount of fines on DCI handleability index.



Figure G.3-2 The effect of surface moisture on DCI handleability index.

Table G.4-1 P	arameters infl	uencing bulk co	al handleabilty.		
Coals/	Amount of	Surface	Ash fraction of	Critical	DCI
parameters	fines (-0.5	moisture at	fines, AF	Moisture	kg/s
	mm)	CM- critical	$\gamma_{0.5}A_{0.5}/100A_{tot}$	CM	at CM
	content,	moisture			
	wt %	wt%			
•LC 3	10.0	6.20	0.03	7.5	1.1
•LC 3	20.0	3.70	0.07	5.0	0.7
•LC 3	30.0	3.70	0.12	5.0	0.6
•LC 3	36.0	3.70	0.15	5.0	0.5
■LC 3 OXY	36.0	7.62	0.15	12.0	0.6
•LC 10 BC	39.0	4.35	0.25	10.0	0.5
●LC met	44.0	8.44	0.57	10.0	0.7
■LC 10B	45.0	5.04	0.24	8.0	0.5
■LC thermal	45.0	3.91	0.47	6.0	0.6
■LC 8U	10.0	0.66	0.07	8.5	1.7
■LC 8U	20.0	2.66	0.12	10.0	0.8
■LC 8U	30.0	3.66	0.19	11.0	0.6
■LC 8U	37.0	2.66	0.24	10.0	0.98

G.4 Factors affecting DCI handleability index

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• hydrophobic coals; • hydrophilic coals

G.5 Segregation effects during Durham Cone testing



Figure G.5-1 Segregation effects during Durham Cone tests - initial seconds of conditioning.



Figure G.5-2 Segregation effects during Durham Cone testing – after a few more seconds of conditioning

G.5 Segregation effects during Durham Cone testing – continuation



Figure G.5-3 Segregation effects during Durham Cone tests – emptying.



Figure G.5-4 Segregation effects during Durham Cone tests – emptying sequence, last 30 seconds.

G. 6 Examples of raw Handleability Monitor tests results

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Reference	Source	Reference	Source	Reference	Source
Coal #11	SChEME	Coal #11	SChEME	Coal #11	SCHEME
8% moisture	Nottingham UK	10% moisture	Nottingham UK	6% moisture	Nottingham.UK
35% fines	Hottingham, ort	40% fines	, to may a second of the secon	50% fines	
	Pmax=45		Pmax=25		Pmax=30
Time.s	Pressure, bar	Time.s	Pressure.bar	Time, s	Pressure,bar
0.00	2.11	0.00	2.28	0.00	1.84
0.20	2.16	0.20	2.25	0.20	1.82
0.40	2.13	0.40	2.27	0.40	1.81
0.60	2.07	0.60	2.27	0.60	1.78
0.80	2.00	0.80	2.25	0.80	1.79
0.90	2.10	1.00	2.25	1.00	1.76
1.00	2.02	1.20	2.24	1.20	1.75
1.20	2.02	1.40	2.24	1.40	1.78
1.40	1.98	1.60	2.14	1.60	1.76
1.60	1.94	1.80	2.16	1.80	1.73
1.80	1.94	2.00	2.14	2.00	1.73
2.00	1.93	2.20	2.11	2.20	1.73
2.10	2.02	2.40	2.08	2.40	1.78
2.20	1.91	2.60	2.07	2.60	1.75
2.40	1.90	2.80	2.04	2.80	1.78
2.60	1.90	3.00	2.04	3.00	1.78
2.80	1.87	3.20	2.07	3.20	1.78
3.00	1.82	3.40	2.07	3.40	1.85
3.20	1.82	3.60	2.07	3.50	1.71
3.40	1.85	3.80	2.10	3.60	1.82
3.50	1.79	4.00	2.14	3.80	1.84
3.60	1.85	4.20	2.16	4.00	1.88
3.80	1.91	4.40	2.22	4.20	1.90
4.00	1.89	4.60	2.30	4.40	1.91
4.10	2.10	4.80	2.34	4.60	1.91
4.20	1.94	5.00	2.36	4.80	1.91
4.40	1.94	5.20	2.34	5.00	1.91
4.60	1.98	5.40	2.36	5.20	1.94
4.80	1.99	5.60	2.36	5.40	1.98
5.00	2.07	5.80	2.37	5.50	1.94
5.20	2.07	6.00	2.31	5.60	2.02
5.40	2.08	6.20	2.31	5.80	2.05
5.60	2.10	6.40	2.36	6.00	2.10
5.80	2.13	6.60	2.36	6.20	2.13
6.00	2.13	6.80	2.40	6.40	2.16
6.20	2.14	7.00	2.43	6.60	2.19
6.40	2.14	7.20	2.47	6.80	2.22
6.60	2.16	7.40	2.53	7.00	2.24
6.80	2.19	7.60	2.62	7.20	2.24
7.00	2.25	7.80	2.50	7.40	2.27
7.20	2.12	7.90	3.48	7.60	2.31

Table G.3-1. Handleability monitor data for Coal #11.

Reference	Source	Reference	Source	Reference	Source
Coal #11	SChEME	Coal #11,	SCHEME	Coal #11	SCHEME
8% moisture 35% fines	Nottingham,UK	10% moisture 40% fines	Nottingham,UK	5% moisture	Nottingnam, UK
	Pmax=45		Pmax=25		Pmax=30
Time,s	Pressure, bar	Time,s	Pressure,bar	Time, s	Pressure,bar
7.30	2.79	8.00	2.74	7.80	2.34
7.40	2.28	8.20	2.82	8.00	2.40
7.60	2.33	8.40	2.88	8.20	2.45
7.80	2.37	8.60	2.93	8.40	2.51
8.00	2.42	8.80	2.97	8.60	2.56
8.20	2.47	9.00	3.02	8.80	2.60
8.40	2.51	9.20	3.02	8.90	2.63
8.60	2.57	9.40	3.12	9.00	2.65
8.80	2.62	9.60	3.17	9.20	2.68
9.00	2.73	9.80	3.23	9.40	2.76
9.20	2.76	10.00	3.31	9.60	2.80
9.40	2.79	10.20	3.37	9.80	2.88
9.60	2.86	10.40	3.43	10.00	2.96
9.80	3.06	10.60	3.51	10.20	3.03
9.90	2.63	10.80	3.58	10.40	3.14
10.00	3.11	11.00	3.66	10.60	3.23
10.20	3.20	11.20	3.81	10.80	3.34
10.40	3.35	11.40	3.92	11.00	3.43
10.60	3.46	11.60	4.00	11.20	3.88
10.80	3.58	11.80	4.09	11.30	2.33
11.00	3.66	12.00	4.20	11.40	3.90
11.20	3.84	12.20	4.27	11.50	2.94
11.40	3.97	12.40	4.40	11.60	3.83
11.60	4.06	12.60	4.50	11.80	3.95
11.80	4.23	12.80	4.59	12.00	4.09
12.00	4.38	13.00	4.70	12.20	4.26
12.20	4.95	13.20	4.81	12.40	4.47
12.30	2.94	13.40	4.93	12.60	4.64
12.40	4.70	13.60	5.05	12.80	4.81
12.60	4.87	13.80	5.16	13.00	5.02
12.80	5.10	14.00	5.30	13.20	5.18
13.00	5.39	14.20	5.48	13.40	5.39
13.20	5.61	14.40	5.58	13.60	5.62
13.40	6.75	14.60	5.71	13.80	5.87
13.50	2.71	14.80	5.84	14.00	6.13
13 60	6 30	15.00	5.93	14.20	6.40
13.80	6 51	15.20	6.10	14.40	6.71
14 00	6.83	15 40	6.23	14.60	7.96
14.20	7 20	15.60	6 39	14.70	3.17
14 40	7.57	15.80	6 56	14.80	7.96
14 60	8.06	16.00	7.63	14 90	4 78
14.80	8.53	16 10	2.86	15 00	7 64
15.00	9.04	16.70	6.82	15 20	8 01
15 20	9 48	16 40	7.02	15.40	8.44

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Reference	Source	Reference	Source	Reference	Source
Coal #11	SChEME	Coal #11,	SChEME	Coal #11	SCHEME
8% moisture	Nottingham,UK	10% moisture	Nottingham,UK	6% moisture	Nottingham,UK
35% fines	Dmax=45	40% fines	D	50% fines	D
Time	Pmax=45	Time	Pmax=25	Time	Pmax-30
1 me,s	Pressure, bar	nme,s	Pressure, bar	Time, S	
15.40	10.08	10.00	7.23	15.60	8.87
15.60	10.65	16.80	7.44	15.80	9.34
15.80	11.31	17.00	7.69	16.00	9.82
16.00	11.96	17.20	7.97	16.20	10.34
10.20	12.04	17.40	0.21	16.40	10.00
16.40	13.31	17.60	9.34	10.00	10.91
16.60	14.02	17.70	5.47	16.70	13.28
16.80	17.48	17.80	8.78	16.80	11.92
16.90	4.01	18.00	9.04	17.00	11.14
17.00	15.63	18.20	9.36	17.10	17.80
17.20	16.53	18.40	8.15	17.20	13.08
17.40	17.48	18.50	15.96	17.40	13.71
17.60	18.46	18.60	11.69	17.60	14.37
17.80	19.38	18.70	3.63	17.80	15.03
18.00	20.31	18.80	8.84	18.00	15.70
18.20	21.25	18.90	17.11	18.20	16.41
18.40	22.24	19.00	10.91	18.40	17.07
18.60	22.30	19.20	11.35	18.60	17.74
18.70	27.53	19.40	11.76	18.80	18.40
18.80	24.50	19.60	12.22	19.00	19.10
19.00	25.63	19.80	13.40	19.20	19.84
19.20	26.84	19.90	9.45	19.40	20.60
19.40	28.02	20.00	12.96	19.60	21.29
19.60	29.08	20.20	13.27	19.80	22.03
19.80	30.15	20.40	13.68	20.00	22.64
20.00	36.47	20.60	14.03	20.20	23.19
20.10	11.29	20.80	14.19	20.40	23.70
20.20	32.60	21.00	14.49	20.60	24.24
20.40	32.53	21.20	17.28	20.80	24.66
20.50	38.25	21.30	5.54	21.00	25.03
20.60	34.90	21.40	15.20	21.20	25.43
20.80	36.14	21.60	15.52	21.40	25.78
21.00	37.17	21.80	15.84	21.60	26.13
21.20	38.34	22.00	16.22	21.80	26.46
21 40	39 75	22 20	16.41	22.00	26.78
21.50	37 72	22 40	16 71	22 20	27.07
21.60	40.41	22.60	17 10	22.40	27.31
21.80	41 38	22.80	16.92	22.60	27.57
22.00	47.30	22.00	10.52	22.00	27.07
22.00	43.08	22.00	17.82	22.00	27.88
22.20	40.00	23.00	10.02	20.00	27.00
22.40	40.00	23.20	10.21	23.20	21.33 28 08
22.00	44.20 11 70	23.40	10.00	20.40	20.00
22.00	44./9 45.20	23.0U	10.09	23.00	20.11
23.00	45.30	23.8U	19.21	23.00	20.10
Reference	Source	Reference	Source	Reference	Source
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Coal #11	SChEME	Coal #11,	SChEME	Coal #11	SCHEME
8% moisture 35% fines	Nottingham,UK	10% moisture 40% fines	Nottingham,UK	6% moisture 50% fines	Nottingham,UK
	Pmax=45		Pmax=25		Pmax=30
Time,s	Pressure, bar	Time,s	Pressure,bar	Time, s	Pressure,bar
23.20	45.78	24.00	19.53	24.00	28.11
23.40	46.05	24.20	19.85	24.20	28.10
23.60	46.27	24.40	20.22	24.40	28.08
23.80	46.54	24.60	20.64	24.60	27.97
24.00	45.30	24.80	21.06	24.80	27.80
24.10	52.73	25.00	21.58	25.00	27.67
24.20	46.88	25.20	22.01	25.20	27.57
24.40	44.80	25.40	22.46	25.40	27.56
24.50	54.80	25.60	23.07	25.60	27.62
24.60	46.54	25.80	23.61	25.80	27.57
24.80	46.40	26.00	24.07	26.00	27.53
25.00	46.33	26.20	24.73 ·	26.20	27.51
25.20	46.11	26.30	22.63	26.40	27.56
25.40	45.85	26.40	24.34	26.60	27.56
25.60	45.71	26.60	24.37	26.80	27.44
25.80	45.53	26.80	24.36	27.00	27.39
26.00	45.36	27.00	24.42	27.20	27.38
26.20	45.09	27.20	24.39	27.40	27.45
26.40	44.70	27.40	24.39	27.60	27.54
26.60	44.41	27.60	24.39	27.80	27.64
26.80	44.12	27.80	24.39	28.00	27.73
27.00	43.65	28.00	24.34	28.20	27.61
27.20	41.72	28.20	24.16	28.40	27.48
27.30	49.13	28.40	24.25	28.60	27.33
27.40	42.73	28.50	23.47	28.80	27.28
27.60	42.30	28.60	24.43	29.00	27.22
27.80	42.01	28.70	22.40	29.20	27.15
28.00	42.73	28.80	24.22	29.40	27.16
28.10	38.33	29.00	24.33	29.60	27.21
28.20	41.64	29.20	24.51	29.80	27.13
28.40	41.47	29.40	24.43	30.00	27.18
28.60	41.35	29.60	24.36	30.20	27.61
28.80	41.23	29.80	23.56	30.30	25.38
29.00	41.00	29.90	27.53	30.40	27.10
29.20	40.90	30.00	24.43	30.60	26.95
29.40	40.73	30.20	24.56	30.80	26.85
29.60	40.52	30.40	24.50	31.00	26.85
29.80	40.34	30.60	24.43	31.20	26.85
30.00	42.24	30.80	24.42	31.40	26.78
30.10	32.05	31.00	24.54	31.60	26.97
30.20	39.95	31.20	24.53	31.70	25.54
30 40	39.89	31.40	24.54	31.80	26.59
30.60	39 79	31 60	24.56	32.00	26.44
30.80	39.59	31.80	25.54	32.20	26.30

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Reference	Source	Reference	Source	Reference	Source
Coal #11	SChEME	Coal #11,	SChEME	Coal #11	SCHEME
8% moisture 35% fines	Nottingham,UK	10% moisture 40% fines	Nottingham,UK	6% moisture 50% fines	Nottingham,UK
	Pmax=45		Pmax=25	н.	Pmax=30
Time,s	Pressure, bar	Time,s	Pressure,bar	Time, s	Pressure,bar
31.00	39.42	31.90	23.32	32.40	26.20
31.20	39.48	32.00	24.56	32.60	26.26
31.30	38.18	32.20	23.64	32.80	26.29
31.40	39.10	32.30	26.69	33.00	26.32
31.60	38.91	32.40	16.81	33.20	25.75
31.80	38.80	32.60	10.08	33.40	18.66
32.00	38.93	32.80	9.56	33.50	26.99
32.20	38.62	33.00	1.99	33.60	10.00
32.40	38.61	33.20	0.52	33.80	5.15
32.60	38.51	33.40	0.55	34.00	5.09
32.80	38.41	33.60	0.55	34.20	0.23
33.00	36.78	33.80	0.54	34.40	0.34
33.20	13.63	34.00	0.52	34.60	0.34
33.40	0.55	34.20	0.52	34.80	0.32
33.60	0.47	34.40	0.54	35.00	0.35
33.80	0.52	34.60	0.54	35.20	0.35
34.00	0.54	34.80	0.54	35.40	0.36
34.20	0.52	35.00	0.50	35.50	0.34
34.40	0.54	35.20	0.51	35.60	0.37
34.60	0.54	35.30	0.49	35.80	0.37
34.80	0.57	35.40	0.54	36.00	0.35
35.00	0.57	35.60	0.52	36.20	0.34
35.10	0.49	35.80	0.52	36.40	0.35
35.20	0.54	36.00	0.49	36.60	0.37
35.40	0.57	36.10	0.53	36.80	0.36
35.60	0.57	36.20	0.52	36.90	0.41
35.80	0.60	36.40	0.57		
35.90	0.49	36.50	0.57		
36.00	0.57	36.60	0.54		
36.20	0.57	36.80	0.54		
36.40	0.55				
36.60	0.57				
36.80	0.55				
36.90	0.64				

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APPENDIX H.

H.1 Bulk density tests of LC 3 coal









LC 3 with 30% fines, effect of conditioning









LC 8U with 10% fines, effect of conditioning





LC 8U with 30% fines, effect of conditioning



