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USE OF PELLETIZATION TO ASSESS THE EFFECT OF PARTICLE-PARTICLE INTERACTIONS ON COAL HANDLEABILITY

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Although there is no widely accepted rigorous definition of handleability, the handling coal characteristics often referred to as handleability define whether a coal has the ability to flow unhindered through the processing and transportation systems. The handleability may be severely affected if fine coal particles tend to aggregate. In the pelletization process, the rolling action of the drum is applied to bring the individual particles into proximity with each other so that they can aggregate and form pellets. Because of apparent similarities between these two processes, the pelletization tests are carried out in parallel to the handleability tests in this project, and the pelletization results are used to explain coal handleability properties.

Key words: coal, pelletization, handleability

INTRODUCTION

British Columbia produces nearly 30 million tonnes of coal annually. Almost all of the produced clean coal is exported outside of the province, and transported by trains across the continent for the use in the eastern Canada or United States, and to other overseas destinations by cargo ships. Coals produced from western Canada have large amounts of fines and high surface moisture at the point of load, which affect these coals handleability characteristics. The western Canadian coals are also very friable and the content of very fine particles continues to increase during transportation.

Many handleability studies carried out over the years mostly related physical parameters of coal samples such as amount of fines, mineral matter type and moisture content on handling characteristics. The quality of fines in terms of surface properties has never been studied before to any significant extend. Neither the porosity of coal has been considered as a factor influencing behavior of fine particles in the presence of water.

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The objective of this research project is to study the effect of surface properties of fines with the special emphasis on wettability, surface properties such as surface area, pore volume and relating these factors to the handleability of bulk coal. Pelletization experiments have been used as a method of testing fine coal particle interactions. The pellet strength is used as an evidence of forces acting on these particles owed to their surface properties. The surface properties of fines are then related to the bulk sample behavior in handling of the selected western Canadian coals.

BACKGROUND

HANDLEABILITY OF COAL

A number of techniques have been developed over the years to assess handleability; for example using specially designed cone to measure the flow of coal and referred to as Durham Cone (Brown, 1997; Cutress et al., 1960; Vickers, 1982; Brown et al., 1997). Other methods focused on either measuring the shear strength of fines (Jenike, 1961; Arnold, 1992; Barois-Cazenave, 1999) or tensile strength of fines, as in the method developed in Poland (Polish norms, 1982; Wawrzynkiewicz, 2003). Extrusion through Index method that was developed by Brown at the University of Nottingham aimed at measuring compressive strength of the bulk sample (Brown, 1997; Brown, Atkin, 2000). The advantage of this method being that, it actually measures the strength of the representative sample of the handled material not only the strength of fines.

The study by (Blondin et al., 2000) showed that handleability is determined by the content of clays and fine particles and developed their own handleability classification system based on these two parameters. (Mikka, Smitham, 1985) showed that bentonite effect on fine coal handleability is much larger than the effect of kaolin.

FACTORS INFLUENCING HANDLEABILITY OF COAL

Factors that influence handleability of coal are: moisture, ash, size distribution and fines content. In general, as the moisture content of coal increases the handleability deteriorates until it reaches a point where coal is so moist that behaves as a fluid. The same trend is observed with increase in fines content, the high fines content leads to deterioration of handleability. It was found that increase in fines content has greater effect on coal handleability than increase in moisture content (Arnold , 1992).

An increasing amount of fines (<0.5mm) can have similar effect as raising moisture content. Widening the spectrum of sizes in bulk coal improves handleability; larger particles counteract moisture effects (Mikka, Smitham, 1985; Holuszko, Laskowski, 2003). It has been shown that the effect of moisture is greater in the case of fines (<0.5mm) than in case of coarse coal (Wawrzynkiewicz, 2003). Mineral matter content has the greatest effect on handleability when clays are present. Clays tend to swell in presence of water and act as glue between particles leading to buildup of cohesiveness within the fines as well as coarse particles.

PELLETIZATION

Pelletization is the particle-enlargement process, in which tumbling of moist fines in drums, discs or in conical devices leads to formation of pellets. Physical forces, such as interfacial attraction, surface tension, van der Waals interactions, combined with the applied mechanical energy of tumbling, that bring particles together are responsible for pelletization (Sastry et al., 1982).

In an extensive study on pelletization of coals of various ranks by (Sastry and Fuerstenau, 1982), the fundamental principles of coal pelletization have been formulated. It was concluded that moisture addition is critical for successful pelletization and lies in a narrow range for each coal and it varies inversely with the content of ash.

There are apparent similarities between behavior of fine particles in pelletization and in a 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. Using pelletization as the model for particle aggregation, 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. 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 up cohesive strength affecting handleability of a bulk sample. Therefore, pelletization can be used as the method to elucidate fine coal particle behavior that determine coal handleability.

EXPERIMENTAL

SAMPLE CHARACTERISTICS

Five raw and two coal products were used for this study. Three coals were of metallurgical grade (LC3, LC 10BC, LC 10B) and two of thermal grade; one lower rank coal (LS20) and one higher rank (LC 8U). One of the raw coals was oxidized under laboratory conditions and used for testing (LC 3OXY). One metallurgical and one thermal sample represented product coals.

Two types of samples were used for testing. For bulk testing 10 kg samples were prepared in such a way that they were closely representing the coal in terms of the size distribution. All raw coal samples were crushed down to 100% passing 25.4 mm. For pelletization and surface properties characterization minus -0.5mm size fraction was sieved out from the bulk sample and used for evaluations. The product coals were used as received, and fine fraction of 0.5mm was also isolated from the bulk sample for surface properties characterization.

The coal analyses are given in Table I. The equilibrium moisture and transmittance are included to demonstrate different hydrophobicity (wettability) characteristics of selected coals. Equilibrium moisture indicates water-holding capacity and was determined following ASTM D 1412-93. Transmittance values were determined following the ASTM D5263-93 method.

Table 1. Samples Characteristics

Coal Sample	Yield of 0.5mm fraction	Ash in Fines %	Ash in bulk Sample %	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

Penetration rate tests were used to characterize wettability of fines obtained from the studied coals. The gravimetric version of the method based on Washburn equation (15) was used as described by (Laskowski et al., 2003). In this method, 3 grams of coal was packed in a tube and compressed by a column-packing device under load of 5 kg. The tube with 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. The patterns of penetration for the studied coals are shown in Figure 1.

SIZE DISTRIBUTION OF THE FINES

Size analyses were performed using Malvern Mastersizer 2000. In this method laser beam is used to obtain scattering patterns of particles in order to determine the size distribution. Figure 2 presents cumulative size distribution of 0.5mm fractions from all seven coals.

N_2 SURFACE AREA AND PORE VOLUME DETERMINATION

Surface area was determined by Quantachrome Autosorb Automated Gas Sorption System using BET method with N_2 as an adsorbate at 77.35 K. The samples were dried at 120^{o}C overnight in a vacuum oven and then outgassed at 50 ^{o}C . The BET surface area was calculated according to the Brunauer-Emmett-Teller equation. The total pore volume was derived using calculations from Density Functional Theory (DFT) and micropore volume was computed from Brauner MP method (Lowell et al. 1998).

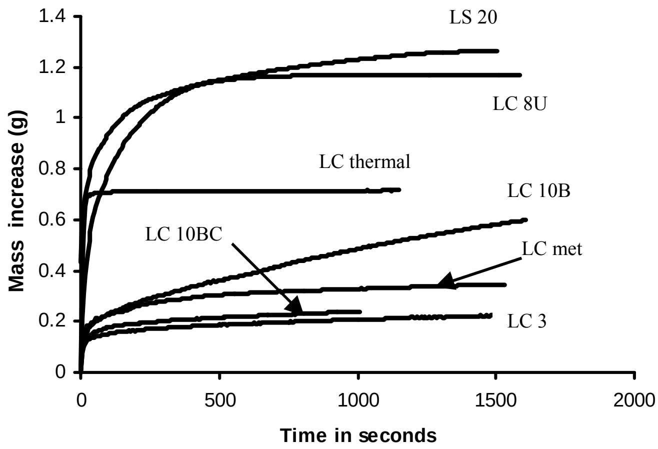

Fig. 1. Penetration rates for studied coals

PELLETIZATION

Pelletization was used as a method to characterize surface properties of fines (-0.5mm). The tests were set up for all the samples under the same operating conditions. Approximately 1kg of dry coal was placed onto the pelletizing disc (Syndron) rotating at 25 rpm and sprayed with water at a rate of 10 ml/min. The amount of water added to each sample was calculated from equation derived by (Sastry and Fuerstenau, 1982). Since the addition of water for each coal depends on the ash content, duration of pelletization run had to be adjusted accordingly. The pelletization tests were carried out until suitable pellets were formed and for most of the coals this was completed within 30 minutes. The pellets were collected and strength was measured using a Tritester tensiometer. The compressive strength of single pellet was measured by crushing the pellet between flat parallel surfaces and the loads at which failure occurred was recorded. The rate of loading was strictly controlled and was digitally set at 0.75mm per minute.

HANDLEABILITY TESTS

The handleability of bulk samples was tested with the use of Durham Cone (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 it was allowed to flow out through the opening. The time to discharge the coal from the vibrating cone was measured and flow rate in kg/s was calculated and referred to as Durham Cone Index. (DCI) The test was repeated for at least 10 times and average value of DCI was calculated.

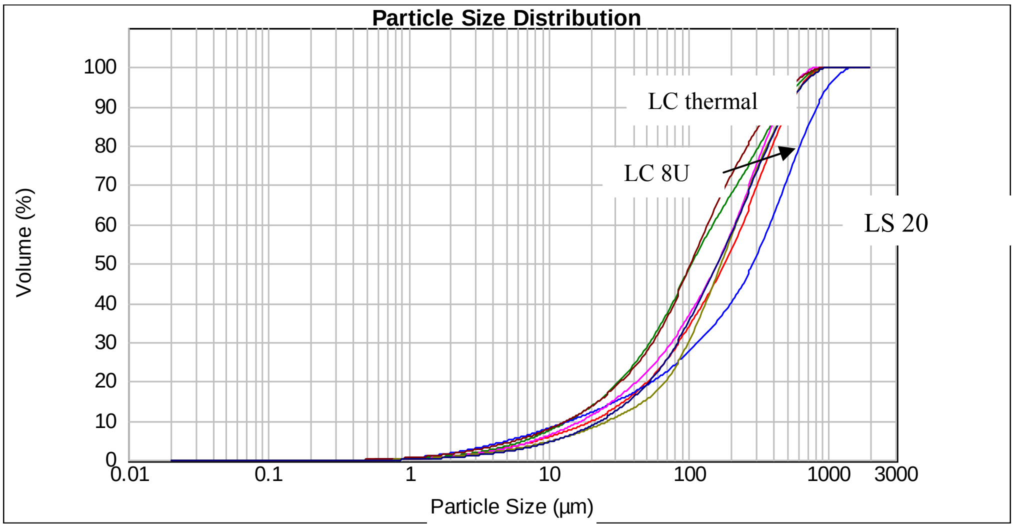

Fig. 2. Particle size distribution of 0.5mm size fractions for studied coals. The coarsest distribution is for LS 20 coal and the finest two are; LC thermal and LC 8U. Distributions for all the other coals are in between

RESULTS AND DISCUSSION

COAL SURFACE PROPERTIES CHARACTERIZATION

The surface properties of all studied coals are summarized in Table II. The selected coals were typical metallurgical coals from British Columbia. These coals were of medium volatile bituminous rank. LC thermal and LC met (metallurgical) were the coal products received from one of the local mines. The exception was LS 20 coal, which was the lower rank coal (high volatile A) from Alberta. These samples varied significantly in wettability characteristic, from very fresh to heavily oxidized, as observed from the transmittance values. The LC 3OXY sample was oxidized by heating at 180°C in the oven for eight weeks.

As Figure 1 shows, the penetration rate patterns for studied coals are in relatively good agreement with both equilibrium moisture and transmittance data. The surface area and pore volume data for studied coals are included in Table II.

As already indicated, in this study pelletization was used as a method of elucidating the effect of surface properties of coals which are responsible for aggregation of particles in presence of moisture. The strength of the resulting pellets was used to evaluate the effect of interparticle interactions on aggregation. However, from the strength and surface area data alone it will be difficult to judge what is causing formation of strong pellets for some coals.

PELLETIZATION

Pelletization of coal particulates is controlled by interfacial and capillary forces due to presence of a liquid phase (Kapur et al., 1966; Sastry et. al. 1977, 1981;Capes, 1980). For pelletization to take place, first 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.

Wetting of hydrophobic coal 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 high interfacial tensions between liquid and solid. Although capillary state may be reached, the layer of water around aggregating particles remains relatively thick and unstable. As a result, strength of the formed pellet is much smaller. (Kaji et al., 1986) made an important observation that for some coals, adsorbed water occupies only ~30-75% of the total pore volume, while for other coals the adsorbed water exceeds by 2 to 3 times their pore volume.

Table 2. Surface Properties Characterization

Coal Sample	Ash in fines %	N_2 BET Surface area m^2/g	Total pore Volume cc/g E-0.5	Micropore Volume cc/g E-0.5	Pellets strength g/cm
LC 3	12.61	0.390	32.4	2.6	8.5
LC 3 OXY	12.61	0.276	18.9	9.8	14.3
LC 10BC	24.41	0.652	45.2	4.2	13.8
LC 10B	12.94	0.376	24.1	0.8	11.6
LC 8U	15.37	1.440	97.3	2.9	20.0
LS 20	33.70	4.140	280.0	27.8	289.0
LC met	12.66	0.439	32.0	7.7	7.5
LC thermal	15.86	0.661	26.5	3.1	20.0

In 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, only at the moisture content exceeding the equilibrium moisture, an aggregation can proceed almost instantly to the capillary state. The stable layer of water on the surface of particles is much thinner; therefore, strength of pellets is much greater due to the strong capillary pressures from within the coal structure.To facilitate discussion the data from Table 2 will be analyzed as separate case studies.

Case 1. Comparison of the behavior of the LC 3 and LC 3OXY coal samples. Since the LC 3OXY sample was prepared by oxidizing the LC 3 sample, their ash contents remain the same. Although the pore volume changes a little and the micropore volume is also smaller after the oxidation, the strength of the pellets almost doubled for the oxidized sample (LC 3OXY). Following our assumptions that for oxidized – hydrophilic coal, the strength of the pellets results from strong capillary forces due to surface hydrophilicity, porosity as well as the ash content and surface area, it is evident that decrease in hydrophobicity has a larger overall effect.

Case 2. Comparing LC met and LC thermal coals shows similar trend; much stronger pellets are produced from the hydrophilic LC thermal coal. Pore volume along with microporosity is much greater in LC met, however due to hydrophobicity of these coal particles, this does not have any significant effect and does on increase the pellet strength. The wetting water on the LC met coal is not able to penetrate the pores to create significant force for aggregation.

Case 3. Two coals LC met and LC 10BC of similar hydrophobicity (Figure 1), one gives pellets 1.83 times stronger than the other. In both cases, contribution of porosity to the force pulling particles together is apparently negligible; the only difference is in the ash content. In this case it appears that only the ash in the LC 10BC sample contributes to the strength of the pellets, and is almost exactly the same magnitude as the difference between the strength of the pellets formed for these two coals.

Case 4. The LC 8U and LS 20 are the two hydrophilic coals. Pellets formed LS 20 are 14 times stronger than pellets formed from LC 8U. If we consider that LS 20 having twice as much ash content (2.2) and approximately three times as much greater surface area (2.8) and pore volume (2.9) as compared to LC 8U, this should give (2.2 x2.8x2.9 = 17.8) 17.8 times stronger pellets from LS 20 coal. According to penetration rate both coals have similar level of hydrophilicity with similar equilibrium moisture value. However, moisture requirement for pelletization predicted from (14) was based on the ash content of the fines and did not take into account surface area; as a result, it underestimated the amount of water required for pelletization of LS 20. The ratio of the underestimated amount of water is equal to 0.73, hence 17.8 x 0.73 = 13. Thus according to these calculations 13 times stronger pellets are expected to be formed from LS 20, which is very close (14 times) to the actual difference between strength of pellets from these coals.

Likewise, all the other cases seem to confirm our conclusion that the strength of the formed pellets is related to the hydrophilic/hydrophobic character of coal particles, and their porosity, microporosity and ash content. In summary, the effectiveness of pelletization is dependant on wettability. The wettability is the most important factor because it decides whether the porosity can create a strong capillary force that affects particle-to-particle adhesion. This capillary force and mineral matter on coal surface determine behavior of the water films between particles. In the case of hydrophobic coals, the strength of the pellets results only from the ash (mineral matter), since water will not penetrate into the pores, at least not to the great extent, to create strong pulling force from within coal pores.

HANDLEABILITY

During coal transportation, fine and coarse particles are tumbled together; however, fines are responsible for the cohesiveness of the whole mixture that affects coal handleability. Depending on the hydrophobic/hydrophilic character and ash content of coal, fine particles can strongly affect coal handleability.

If the same surface properties are responsible for fine particle behavior in handleability as in pelletization, then hydrophobic coals should be much easier to handle than hydrophilic ones. Further on, it can be concluded from the conducted pelletization tests that for hydrophobic coals only ash (mineral matter) content can influence their handling characteristics.

The handleability tests were performed using Durham Cone on the bulk samples of all seven coals. The tests were carried out varying moisture levels; flow rates at different moistures are plotted to examine the trends (Figure 3 and 4). The LC met, LC 10B and LC thermal coals have similar amount of fines (about 45% of 0.5mm material) and ash content with the exception of LC thermal (Table I). According to pelletization tests LC thermal sample should be ranked the most difficult to handle followed by LC 10B and LC met and indeed this is the case.

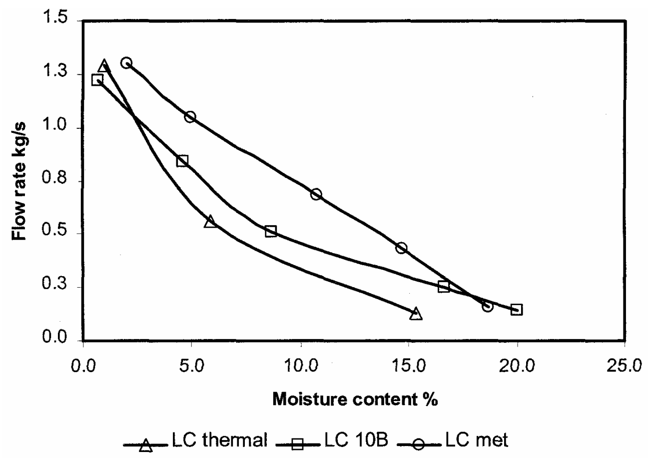

Fig. 3. Flow rates at different moistures content for LC thermal, LC 10B and LC met coals

For other four coals the trend is not so evident (Figure 4). Coals LS 20 and LC 8U have similar wettability characteristics (Figure1) however, LS 20 has only 22.0% of fines (0.5mm fraction) as compared to 37.0% in LC 8U. Since the amount of fines is the most critical factor, it is not surprising that LS 20 has much better handleability than LC 8U. Both of these coals have their equilibrium moisture at about 8%; this implies that below that moisture content, there is no surface moisture present on the surface of particles which would lead to the aggregation. Thus, only at the moisture levels exceeding 8% the particles will start to aggregate or stick together. To emphasize this effect, correlations shown in Figure 4 were replotted versus the surface moisture for these four coals (Figure 5). Surface moisture was calculated as the difference between actual moisture and equilibrium moisture.

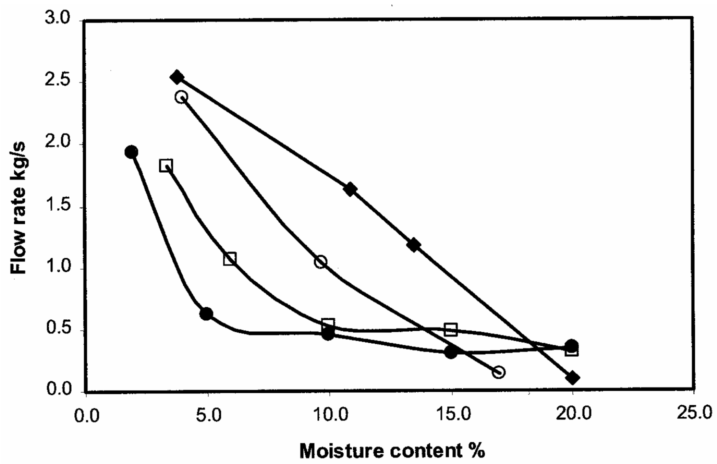

Fig. 4. Flow rates at different moistures content for LC 3, LC 10BC, LC 8U and LS 20 coal

Lines showing the trends of the flow rate change vs. surface moisture for LS 20 and LC 8U moved to the range of much reduced handleability (Figure 5). The LC 8U coal has the same amount of surface moisture as the LC 3 coal; the difference is that at 10% surface moisture this coal almost entirely stops to flow, while LC 3 is still handleable.

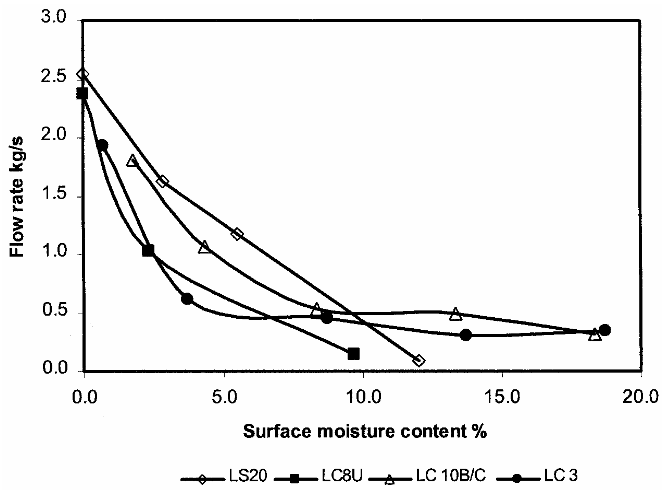

Fig. 5. Flow rate change vs. surface moisture for LC 3, LC 8U, LC 10BC and LS 20

The LC 10BC coal exhibits a very similar trend in flow rate vs. moisture to that of the LC 3 coal, suggesting that being hydrophobic it does not cease to flow at high moisture levels. However, both of these coals exhibit overall poor handleability characteristics. To clarify why these two coals have such bad handling properties, X-ray diffraction (XRD) of low-temperature ash (LTA) was carried out to learn more about mineral matter type in these samples. Table 3 presents XRD results for these coals.

As Table 3 indicates, both of these coals have very high amounts of clay minerals. The LC 3 coal has 5% more clays in total than LC 10BC. Based on this information it can be concluded that handling behavior of these two coals is hindered by the presence of high amount of clay material.

Table 3. Mineralogical Composition of LTA from LC 3 and LC 10BC

Mineral	LC 3%	LC 10BC %
Quartz	19.6	24.4
Kaolinite	36.4	26.1
Illite	24.1	27.8
Muscovite	18.9	20.7
Total clays	79.4	74.5
Pyrite	0.3	0.4
Dolomite	0.6	0.6

CONCLUSIONS

The following conclusions can be drawn from this project:

- Pelletization can be used as a method of characterizing surface properties of fines that affect behavior of these particles in handling. Testing coal for pelletization under controlled conditions can help to reveal the effect of surface properties (such as wettability, surface area and porosity) on ability of fine coal particles to aggregate. In general, easy to pelletize coals are difficult to handle.
- Equilibrium moisture is an important parameter that needs to be assessed in order to set the limits for good and bad handleability ranges for any given coal. For lower rank and oxidized coals, equilibrium moisture is usually much higher, therefore these coals can tolerate higher moisture levels before they start to be difficult to handle. However, at the moisture levels exceeding their equilibrium moistures, a rapid deterioration of their handling behavior is observed, leading practically to non-flowing conditions.

- In case of hydrophobic coals only the mineral matter and type of association with coal organic matter affects significantly these coals handleability. For such coals an increase in moisture content tends to affect handleability to certain level, but the coal does not cease to flow completely. Apparently high amount of clay material even in very hydrophobic coals can have very damaging effect on handleability.
- 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 levels of fines.

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„Coal hadleability'', termin, którego polskim odpowiednikiem jest "podatność transportowa węgla", charakteryzuje przepływ węgla przez różne węzły technologiczne i opisuje zachowanie się drobnego węgla podczas transportu, składowania i dozowania. Podatność transportowa węgla zależy w bardzo dużym stopniu od agregacji ziarn węgla. W procesie peletyzacji ruch obrotowy bębna/dysku powoduje stykanie się drobnych ziarn, które w wyniku tego mogą tworzyć kuliste pelety. Ta sama podatność ziarn do tworzenia agregatów w trakcie ich transportu powoduje agregację ziarn, co zasadniczo wpływa na podatność transportową takiego materiału. W tej pracy badano równolegle podatność transportową różnych próbek węgla i peletyzację tych próbek. Wyniki peletyzcji zostały użyte do scharakteryzowania właściwości drobnego węgla, które określają jego podatność transportową.