FLUIDITY OF WESTERN CANADIAN COALS AND ITS RELATIONSHIP TO
OTHER COAL AND COKE PROPERTIES

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textures.

INTRODUCTION

This paper is intended to provide the reader with an understanding of Gieseler Plastometer measurements of coal fluidity and how they are influenced by other coal properties. It uses data from western Canadian coals and consequently is also a review of the fluidity of western Canadian coals (WCC). The paper introduces some new data but it also makes extensive use of data that are available in scattered and less accessible articles. One of the most important sources for data on Canadian coking coals is the Canadian Carbonization Research Association, which has advanced the understanding of the quality characteristics of WCC to the great benefit of the companies marketing metallurgical coal overseas.

In simple terms there are three types of coal differentiated by rank (maturity): thermal (low-rank steaming coal), bituminous (middle-rank metallurgical coal) and smokeless coal (anthracite rank). The bituminous coals, which cover the rank from high-volatile to low-volatile bituminous, are so called because bitumen can be obtained from them. In fact, the coke making process involves the destructive distillation of bitumen derived from coal as it is heated in a coke oven. The coal goes through a plastic phase in the coke oven as bitumen is distilled out of the coal. As the temperature continues to rise the bitumen is destroyed and the coal is finally annealed into coke at temperatures of over 1000°C. The formation of a plastic or sticky phase is critical to the formation of quality coke. One measure of coal's ability to make quality coke is fluidity, a test which measures the rheological properties. test, which measures rheological properties.

The fluidity of bituminous coal samples is important in predicting coke quality but often its importance as one technical parameter for predicting coke quality is overshadowed by its importance as a bargaining chip in the high-stakes game of coal sales. A recent paper (Coin and Broome, 1997) de-emphasizes the importance of fluidity and suggest that its usefulness for predicting coke quality is extremely limited. They point out many problems with the various models that use coal rheology or petrography to predict coke properties. They suggest that the two most important coal properties are rank and ash chemistry because they correlate strongly with coke strength after reaction (CSR), which they consider to be the most important coke quality parameter.

Intense efforts are underway in countries like Japan to reduce the costs of raw materials. For example, the use of pulverized coal injection (PCI) allows use of weak or soft coking coals in the coke blend to reduce the overall cost of the blend. This means that there is increased emphasis on quality control as it pertains to the coal leaving the mines and the coke made in the coke ovens. Contracts include reference to an increasing number of quality parameters, which are increasingly more rigorously defined.

One can measure the quality of coke by its performance in the blast furnace. This is too late for the coke oven operator to respond to changes in coke quality. It is better to test coke before it gets to the blast furnace. This requires making test coke and establishing tests that duplicate conditions in the blast furnace. The data are reasonably expensive to obtain but provide a good prediction of the performance of coke in the blast furnace. The least costly method is to predict the quality of coke by using data from tests on coal. Fluidity is one of the tests used to predict the quality of the coke and the blending potential of coals used to charge coke ovens.

COAL RHEOLOGY

Many of the performance characteristics of coke, that are measured by tests on cold rather than hot coke, are heavily influenced by the coal rank and rheology (i.e. dealing with flow and deformation) of the parent coals. Consequently the three tests, described in the next paragraph, that measure the rheological properties of coal, are frequently used to predict coke properties. The coke property most often determined is the resistance of cold coke to abrasion and impact. This property is measured by tumbling sized coke in a drum under fixed conditions and then measuring the amount of material remaining on a screen. There are two frequently used variations of the test. In America the
Stability Factor (ASTM test) is measured while in Japan the JIS DIN 30/15 drum test is performed. Results for the two tests are compared in Figure 1; it is useful to be able to convert from the Japanese index to the ASTM index. Many diagrams exist relating cold coke tests to various coal properties. Because the two tests involve slightly different procedures there is no exact relationship between the tests as indicated by the data scatter in Figure 1.

The three tests that measure rheological properties are FSI, dilatation and fluidity. The FSI test is the oldest and most universal, though its usefulness is limited because there are only 20 possible increments to the range of FSI values. In some countries FSI is referred to as CSN (crucible swelling number). In Europe the dilatometer test, in which a small pencil of coal powder is heated through a contraction and expansion phase, is preferred as a measure of coal rheology. In North America the Gieseler Plastometer is more widely used. In this test a sample is slowly heated and measurements made of its fluidity (viscosity) at elevated temperatures.

The most extensive investigation of the rheological properties of western Canadian coals is by Price and Gransden (1987). It indicates that dilatation is the least accurate way of predicting cold coke strengths followed by fluidity and then FSI. This means that as long as a sample has good FSI there is a good chance it will make acceptable coke. If the dilatation is low, fluidity and FSI measurements should be made to see if they contradict the initial prediction of poor coke quality.

**SIGNIFICANCE OF MAXIMUM FLUIDITY AND FLUID TEMPERATURE RANGE MEASUREMENTS USING A GIESELER PLASTOMETER**

Fluidity is measured using the Gieseler Plastometer (Figure 2), which documents the changing fluidity of a coal sample in air as it is heated at a constant rate of 2° to 3°C/minute through its softening and melting temperature. The sample is 5 grams of coal crushed and screened to pass 35 mesh (0.5 mm) and loosely compacted. The heating rate is similar to that employed in commercial coke ovens. The temperature range extends from about 350°C to 550°C. The maximum fluidity is recorded as are the temperatures at which the coal reaches maximum fluidity, first starts to soften, and finally solidifies. The difference between the last two temperatures is the fluid temperature range (FTR). The fluidity is measured using a paddle, under constant torque. The paddle is inserted in the sample and as it softens the paddle starts to rotate, rotating faster as the sample becomes more fluid. Eventually at high temperatures the coal sample hardens and the paddle stops rotating. The speed of rotation is measured in dial divisions per minute (ddpm). There are 100 divisions per revolution, consequently a fluidity of 100 ddpm corresponds to a speed of 1 revolution per minute of the paddle. Most instruments can not record a rotation of more than 280 revolutions per minute, therefore, fluidities above 28 000 can not be measured and measurements over 10 000 probably are not reproducible.

Measurements of maximum fluidity (MF) and FTR are used to predict the behaviour of the plastic phase during coking. The property of “fluidity” measured by a spinning paddle is not completely analogous to the properties of softening, sticking, smearing and binding, that in loose terms describe what is happening in a coke oven. Based on the heating rate of about 3°C/minute coal is plastic for about 10 to 30 minutes (bottom scale in Figure 2) in the plastometer or in the coke oven. If the maximum fluidity is 300 ddpm, which is above average for most British Columbian coals, then the paddle is turning once in 20 seconds. Considering these conditions one can envisage a transitory phase in which the reactive macerals become soft, sticky and swell as
vesicles form. This phase forms a front, which moves slowly inwards towards the centre of the coke oven, leaving behind it semicoke heated to temperatures greater than the solidification temperature of the coal. The process is not one in which large volumes of coal become fluid for long durations during coke making.

The MF is an empirical and approximate measure of the minimum viscosity of the sample achieved at a particular temperature. Unfortunately the instrument sets up shearing forces in viscous coals and in fluid coals forms froth so that the paddle stirring a mixture of coal and gas. Consequently fluidity measurements do not correlate exactly with the true viscosity of the sample and the results for highly fluid coals probably do not correlate well with coke-making processes.

There are many variations of the plastometer but they have not gained wide acceptance and do not provide a basis for comparing coals. For example, there have been attempts in France to use a constant speed variable torque version of the Gieseler Plastometer to overcome some of the problems described above.

The fluidity measurement is an attempt to provide a practical test for comparing the rheology of coals. It is important that coal samples are prepared consistently. Samples are crushed to pass 35 mesh (0.5 mm). This is much finer than the 80% minus 3.35 mm size consist used in commercial coke ovens. The finer size inhibits fluidity, samples would exhibit greater fluidity if crushed to a coarser size consist. The softer nature (higher HGI) and susceptibility to crush to fine size consists are two possible reasons why some WCC exhibit fluidities lower than expected based on rank and petrography. Values of FTR and MF are improved if the heating rate is faster. In fact the FTR can increase by as much as 20°C if the heating rate is increased from 2°C/min to 5°C/min (Loison et al, 1988).

In general fluidity is controlled by the relative proportions of plastic components (vitrinite+lpiptinite) and inert components (inertinite + mineral grains). The plasticity does not survive at higher temperatures because the plastic bitumen looses hydrogen and solidifies into carbon grains that cement the sample. The instrument is therefore providing information on a number of processes.

- Generation of a plastic/fluid component that is mobile enough to move through the mixture of plastic and solid fragments and coat them so that the combination is plastic.
- The temperature range over which the sample remains plastic.
- The maximum fluidity or minimum viscosity obtained.

In the coke-making process the coal must provide a phase that also eventually acts as a cement. A hydrogen-rich material might provide very high fluidity, but after the hydrogen evolves at high temperatures there is not enough carbon cement to stick the inert grains together. For example a mixture of the hydrogen-rich hydrocarbons tar and anthracite, will not make coke. There is therefore an optimum amount of fluidity required by a coal blend to make good coke.

Coin and Broome (1997) suggest a slightly different model for coke formation, in which vitrinites swell and become sticky as the charge is heated. The swelling is caused by the development of pores. The vitrinite grains eventually partially enclose the inert fragments and stick the heterogeneous solid together. This model obviously emphasizes dilatation behaviour.
Fluidity of coal is controlled by:
- petrography and rank.
- grain size.
- chemistry.
- the amount, grain size and chemistry of ash.
- aging and oxidation.

**Petrography and rank**

At constant rank, fluidity increases as the amount of total reactivities increases in a sample (Figure 3c). The rate of increase varies depending on rank, probably influenced by the hydrogen content of the vitrinite. High-volatile coals are characterized by high values of fluidity that are insensitive to changes in petrography. This may in part be due to limited amounts of in medium-volatile coals. Medium to low-volatile coals are characterized by low fluidity values, that are zero at intermediate concentrations of total reactivities and increase rapidly at higher concentrations. The approximate linear relationships of total reactivities to log MF and FTR for Canadian coals is apparent in a number of plots in Figure 3b and 3c. It should be remembered that there is no natural law, which states log MF versus total reactivities relationship should be linear and many exponent relationships may give a better empirical relationship. The apparent linear relationship has some very important implications for WCC, which are characterized by major variations in total reactivities within and between seams. An increase in total reactivities from 50% to 60% changes fluidity from 5 to 20 dpdp whereas the same 10% increase from 70% to 80% reactivities changes the MF from 100 to 700 dpdm. High-volatile coals will exhibit some fluidity even if they are rich in inertinite where as medium-volatile coals have zero fluidity if total reactivities are below about 45%. This threshold probably rises for low-volatile coals. It is very important to monitor changes in petrography of low fluidity medium-volatile coals, especially when they have high contents of inertinite.

Using the linear relationships in Figure 3c, it is possible to produce a plot of MF versus rank for samples with constant total reactivities content (Figure 3a). Apart from the very fluid, low rank coal data, log MF is roughly linearly related to rank of coals with similar total reactivities content. Fluidity is therefore a measure of the combined effect of petrography and rank. If a version of Figure 3a was constructed containing more tracts of coals with similar reactivities contents but varying rank, then it would be possible to estimate the fluidity of any coal. It is not clear if Figure 3a will work for a blend of samples of different rank.

Three samples of a single high-volatile seam Rmax = 1.03% representing average seam (banded lithotypes), bright lithotype bands and dull lithotype bands were collected. The bright and dull lithotype samples were also combined in equal proportions to make a mixed sample. For these samples the macerals were grouped petrographically into structured vitrinite (tellinite and telocollinite), unstructured vitrinite (desmocollinite and detrovittinite), structured inertinite (semifusinite and fusinite) and unstructured inertinite (macrinite, inertodetrinite and micrinite). The most striking difference between the bright and dull lithotypes is the increased amount of structured vitrinite in the bright lithotype (Figure 4). The amount of inertinite increases in the dull lithotype. Fluidity is very sensitive to changes in total reactivities, more so than for other high-volatile data suites (Figure 3c). This suggests that it is the larger changes in percent structured vitrinite that are controlling the fluidity and this maceral has the highest fluidity. This may be because the cell structure of tellinite is often filled by other maceral material, such as resinite and liptinite, which have high fluidities. It appears that maximum fluidity is very sensitive to small additions of bright lithotypes, which contain high concentrations of structured vitrinite.

Cameron and Babu (1968) measured fluidity of whole coal and bright lithotype samples from a
medium-volatile seam. Generally the bright lithotypes had fluidities 2 orders of magnitude higher than the whole coal samples. Coke breeze is added to some coal blends to increase the inert content of the blend and because it is available as a waste product from the coke ovens. Their data illustrate that fluidity is decreased and the FTR shrinks symmetrically about the maximum fluid temperature as the amount of inerts increases.

Fluidity is dependent on rank, which is measured by vitrinite reflectance. But for any coal, vitrinite reflectance measurements produce a range of values. Some grains have higher than average reflectance and others lower. This range of values is composed of the V types also used to calculate the stability index (Schapin and Gray, 1964). Probably each V type has its own characteristic fluidity with low V types having high MF, large FTR and low T_max temperatures and high V types having low MF, small FTR and high T_max temperatures. It is the mix of V types and the amount of inert material in a sample that control its fluidity. The fluidity characteristics of a particular V type vitrinite may be the same irrespective of whether it is from the upper part of a histogram from a low-rank coal or from the lower part of a histogram from a high-rank coal.

The fluid temperature range (FTR) decreases as rank increases (Figure 3b) and in general has similar relationships to rank and petrography as maximum fluidity. At a rank of medium-volatile, FTR is sensitive to changes in total reactive content whereas at lower ranks it is relatively insensitive. If coke quality is related to FTR, then the coking properties of WCC coals of medium-volatile rank may be susceptible to small changes in petrography or ash contents.

Petrographic composition can vary within a seam and there are a number of papers that document increased inertinite in the upper parts of seams (Lamberson and Bustin, 1997; Ryan, 1997). It can also be influenced by washing as illustrated by Bustin (1987) who analyzed the petrography of a number of seams from the Fording River coal mine. The samples were subjected to float-sink analysis and individual specific gravity (SG) increments analyzed (Figure 5). The inertinite/reactives ratio on an ash-free basis reaches a maximum at intermediate SG values. Petrography is also influenced by crushing and sizing, which concentrates the inert macerals in the intermediate sizes (Table 1). The fluidity of a sample may be improved if either the intermediate sized material, or the material with SG values in the 1.4 to 1.6 range, is removed.

Fluidity is very sensitive to the amount of exinite in high-volatile coals (Figure 6). Unfortunately exinite is usually destroyed by the time the rank reaches medium to low-volatile bituminous. However for coals with Rmax values less than 1.3%, small changes in the amount of exinite may explain unexpected variations in fluidity.

### Grain size

The grain sizes of both the maceral components within the solid coal and the fragments of coal can potentially influence fluidity. No data were located on the relative significance of maceral sizes. Macerals such as tellinite and telocollinite usually occur as large masses whereas as desmocollinite and dextra-vitrinite usually occur in small masses. Similarly within the inert macerals, microinite and inertodetrinite form smaller fragments within the solid coal than macrinite, fusinite and semifusinite. A high proportion of inertodetrinite and low proportion of fusinite plus semifusinite in the inertinite component of a coal significantly increases the surface area of inert material that must be cemented. Changing proportions of macerals can effectively change the relative homogeneity of the coal blend without even considering particle size. The size of the maceral fragments compared to the size of coal fragments

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**Table 1**

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<th>Size mm</th>
<th>S g 1-1.2</th>
<th>S g 1.2-1.4</th>
<th>S g 1.4-1.5</th>
<th>S g 1.5-1.6</th>
<th>S g 1.6-1.7</th>
<th>S g 1.7-1.8</th>
<th>S g 1.8-1.9</th>
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<td>17.9</td>
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<td>18.1</td>
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<td>17.9</td>
<td>17.5</td>
<td>17.9</td>
<td>18.1</td>
<td>18.1</td>
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**Figure 5:** Variation of inert/reactives ratio for different specific fractions. Data from Bustin (1982).
Figure 6: Maximum fluidity and fluid temperature range of blends of a medium-volatile coal and an exinite-rich cannel coal.

Figure 7: Relationship of hydrogen content to maximum fluidity for Canadian coals of different ranks. Probably plays an important part in aiding or inhibiting the ability of the reactive macerals to coat and cement inert fragments. Possibly the most important factor is the number of coal particles that are composed of a combination of reactive and inert maceral components. These coal particles will have much less difficulty sticking together than coal fragments composed of a single maceral. Crushing to a finer size increases the number of monomaceral coal fragments.

It is also possible that the fine fragment size speeds the distillation process, shrinks the FTR and gives the reactive components less time to coat the inert fragments. Decrease in grain size lowers fluidity and the effect is particularly pronounced in the case of dull coals (Lowry, 1963). There have been a number of studies by the CCRA in which vitrinite-rich coal from the fine circuits of wash plants has been added to the clean single coal product in an attempt to improve coke quality. The results were generally not as good as predicted by petrography, indicating that for a single coal, fine size may have a negative effect on coal rheology, in particular fluidity and coke strength. However finer size does not lower FSI values, possibly because of the higher heating rate (Price and Gramden, 1987).

There are data that indicate that crushing blend coals to a finer size-consist improves coke quality (Leeder et al., 1997). It is not clear why this differs from the single coal data. Possible factors influencing coke quality are that these blends were crushed to an overall finer size where as additional fine coal was added to an existing size consist for the single coals. Obviously the average coal fragment separation is influenced by fragment size and size distribution.

Hydrogen chemistry

Hydrogen is concentrated in the reactive macerals, especially the liptinites, and is less abundant in higher rank coals. Since these are coals with poorer rheology for making coke, it could theoretically provide a good indication of the fluidity of a coal. Fluidity is very sensitive to small changes in hydrogen content in medium-volatile coals but much less sensitive to changes in high-volatile coals (Figure 7). In general the hydrogen content of coals can not be used to predict fluidity or coke properties. Hydrogen can effect fluidity, if it is available to be donated to solvent species (Clemens and Matheson, 1991). The process is similar to what happens when hydrogen is introduced to help liquefy coals.

The relationship of fluidity to the general chemistry and molecular structure of coal is complex. In simple terms as rank increases coal molecules become larger and take on the form of interlocking hexagonal plates (aromatic structures), which inhibit the formation of fluidity in the coal.

Ash

Ash contents are reported as weight percents but fluidity is probably more dependent on the relative volume of inerts in the sample. An ash content of 10% by weight is equivalent to about 6% by volume. Price and Gransden (1987) studied the effect of ash on a number of rheological and coking properties of WCC. They washed three coals (Rmax 0.91%, 1.32%, and
Most of these changes in MF seem large but are weight percent ash; emphasizing the important petrography but different ash concentrations were volatile data set for the three coals has a similar trend to this case the size of the grains is very important. The way they increase the total surface area of inerts, there also seems to be a chemical influence on fluidity where as additions of inert compounds, such as sulphur and Fe$_2$O$_3$, caused larger decreases in much as 36%. Quartz and kaolinite caused minor minerals can cause maximum fluidity to decrease as much as 36%. Quartz and kaolinite caused minor changes in MF where as additions of inert compounds, such as sulphur and Fe$_2$O$_3$, caused larger decreases in MF. Most of these changes in MF seem large but are similar to expected decreases based on an equivalent 1% increase in inherent ash content (Figure 9).

The mineral additions can effect fluidity in three ways. They increase the total surface area of inerts that must be incorporated into the plastic phase and in this case the size of the grains is very important. The introduced grains may have different surface tension characteristics with respect to the plastic phase. The mineral additions may react chemically with plastic components. It appears that in most cases the mineral additions mimic changes in ash content in a normal sample. The sulphur and Fe$_2$O$_3$ (magnetite) additions change fluidity much more, probably indicating a chemical reaction with the plastic components in the coal.

Minerals in the experiments were added as a fine powder and therefore mimicked natural ash occurring as finely dispersed extraneous fragments. Natural ash in washed samples can occur finely disseminated in desmocollinite, filling cells in semi fusinite, or as ions forming part of the coal molecules. The mineral matter that is locked in inert coal fragments can have negligible effect on fluidity where as that dispersed in desmocollinite or external to maceral fragments can have a pronounced effect on fluidity. In this regard a sample with calcite filling cells in semi fusinite may have a reasonable fluidity, but the calcium oxide will have a devastating effect on CSR at higher temperatures. The way mineral matter is dispersed in the macerals will effect the way it influences MF and other coke properties.

The main effect of mineral addition is to decrease MF and FTR in amounts comparable to normal coal ash with the exception of additions containing sulphur and iron. These substances obviously get involved in chemical reactions, which inhibit MF but do not have an equivalent effect on stability factor. In contrast the 1% additions of some minerals caused major changes in CSR illustrating the ash chemistry control on this property. Obviously fluidity will be a poor indicator of stability factor if there is a chemical reaction between the mineral matter and the plastic components in the coal. This is the case for coals with Fe-rich ash, which lowers fluidity causing an under estimation of stability factor. CSR is even more sensitive to ash chemistry than fluidity, therefore in some cases it will be over estimated if based on fluidity data.

**Aging and oxidation**

Fluidity is more sensitive to oxidation than FSI, though when MF values are plotted on a log scale the effects are of the same magnitude as those experienced by FSI values. Pyrite has a moderate effect on fluidity but Fe$_2$O$_3$ has a major effect (Figure 9). Obviously if pyrite oxidizes, or if magnetite is introduced by heavy medium cleaning, fluidity will be effected. Price et al. (1992) measured the deterioration of fluidity over 200 days (Figure 10). Log MF decreased slightly where as stability factor and CSR values were unchanged and FSI values varied from 7.5 to 8.5 with no downward trend. When plotted on an MOF diagram (Figure 10) the changes in fluidity appear insignificant. There has been a lot of emphasis on the rapid deterioration of fluidity as samples oxidize. For many coals, the changes due to oxidation are irrelevant in terms of coke quality. The initial decrease in fluidity may be associated with adsorption of oxygen, which occurs rapidly to all coals when they are brought to surface as a result of mining. This occurs long before they are used for coke making. Fluidity is probably a very sensitive, but expensive, test for oxidation. The alkali extraction test is becoming widely accepted by the coal industry because it is faster and, cheaper.
COAL BLENDS AND THE ADDITIVE PROPERTIES OF FLUIDITY

A number of experiments aimed at calculating the fluidity of coal blends indicate that fluid properties are not additive. It is not possible to derive an average fluidity by calculating the weighted average of log fluidity values of a number of samples. In fact this process nearly always predicts average fluidities that are too low. This can be explained by the fact that fluidity is proportional to VM (daf), which has a non linear relationship to reflectance. Averaging ranks produces an intermediate rank that is higher than would be calculated from the average VM (daf) value for the mixture.

The 50/50 mix of lithotypes (open square, Figure 4) produced a noticeably greater fluidity than predicted (solid square), based on averaging the log fluidity values of samples with identical rank.

One blending experiment looked at the effect of mixing a sample containing 84% exinite (cannel or needle coal from the Elk Formation in south-eastern British Columbia) with a medium-volatile coal containing 0.2% exinite. Addition of small amounts of cannel coal produced major changes in MF that could not be predicted by adding the log MF values of the two coals (Figure 6 and 11c). In fact this process consistently predicted MF values that were too low. Additions of cannel coal in amounts of less than 1% can increase fluidity significantly. Consequently varying amounts of exinite in high-volatile coals may destroy the linear log MF versus total-reactives relationship seen in medium-volatile coals. It should be noted that exinite-rich coals often have high contents of inherent ash and organic sulphur. Additions of exinite-rich cannel coal have been found to decrease coke oven pressure and coke strength (Stell, 1986).

A similar blending experiment was conducted by Fawcett and Dawson (1990: Figures 11a & b). They blended high and low-volatile coals and were unable to accurately predict MF by weighted log averages. However the errors when comparing log MF values are less significant. Plotting the results on an MOF diagram illustrates that 2 component blends do not plot on chords joining the plotted positions of the component coals.

Generally very little attention is given to the MF temperature relative to the start and final temperatures. If it is not mid way between these two temperatures then the fluidity curve is asymmetric. There is some indication that blending produces asymmetric log MF versus temperature plots. It is apparent in Figure 6 that

Figure 9: Effect on fluidity of 1% mineral additions to coal samples. Data from Price et al. (1992). The vertical line is the decrease in MF expected for a 1% increase in weight percent ash based on the data from Figure 8.

Figure 10: (a) Effect of oxidation/aging on fluidity, stability factor and coke strength after reaction for up to 200 days. (b) fluidity data plotted into an MOF diagram. Data from Price et al. (1992).
as the amount of exinite in the blend increases the start temperature is largely unaffected but the MF temperature decreases. Figure 11 b, illustrates the effect on MF temperature (T_{max}) and FTR of blending the coals. The FTR of the blend is increased but not as much as the full overlap.

It is not surprising that fluidities can not be predicted by weighted averages of log values because fluidity is a measurement of the amount of plastic material in the coal sample at a particular temperature. For a single coal the amount of plastic material starts at zero at a low temperature, reaches a maximum at T_{max} and then decreases to zero as the temperature continues to increase. A second coal goes through the same process but over a different temperature range and with a different relationship of plastic components to measured fluidity. Any attempt at predicting blend fluidity should start by summing the plastic and inert components of the individual coals in the blend over a range of temperatures. The resulting plot of proportion of plastic phase versus temperature can then be converted into a fluidity versus temperature plot and the maximum fluidity determined. Generally fluidity data are not reported on a continuous basis as the temperature increases, so that there are no data on the shape of fluidity versus temperature plots for blends of coal to use to help the modeling.

Sakurovs et al. (1994) modeled blend fluidity and in cases where there was no chemical interaction between components that enhanced or suppressed fluidity, the predicted results were close to experimental results.

Simple plots of Log MF versus total reactives are approximately linear for coals of similar rank (Figure 3). This means that if the total reactives of the component coals are known then it is possible to calculate the total reactives of the blend. The data on a total reactives versus log MF diagram can be used to predict the MF value for the blend. This method for predicting blend fluidity will probably only work for coals of similar rank.

The MOF diagram, which plots log MF versus mean maximum reflectance (R_{max}) is used to predict blend fluidities for coals of different rank. It appears that blend coal values plot near the line joining the two end members based on calculating the average R_{max}. But as discussed earlier, the blends will generally plot above the line. It is therefore not possible to accurately predict MF values of blends of coals with different R_{max} values using the MOF diagram. It is also questionable if the term average rank has any significance. It does not necessarily indicate what the average petrography is in terms of the distribution of V types.

**COKE FORMATION**

Coke is made by crushing coal to 80% minus 6 mesh, 1/8 inch or 3.5 mm and then charged into a slot oven. During the coking process, the coal charge goes through a number of changes over a period of 15 to 25 hours as the temperature rises to a maximum of about 1100°C. Initially at temperatures up to about 100°C, water and CO_{2} evolve with some increase in pressure. As the temperature rises to 350°C the coal expands. At temperatures above 350°C the outer layer of the coal in the coke oven starts to form a plastic layer characterized by the distillation of bitumen, tars and oils. As the temperature increases the distillation products evaporate and the plastic layer moves inwards leaving behind, on the outer side, semi-coke, which continues to evolve gases such as CH_{4}. The plastic layer meets in the middle of the oven, either trapping gases to form a final pressure surge (test oven) or
allowing the last gas to escape upwards (commercial oven). As the temperature increases towards 1100°C hydrogen gas is evolved, the charge usually shrinks and true coke forms.

The various rheology tests attempt to provide information about different stages in the coke forming process as indicated in Table 2, which is adapted from Lowry (1963). As the coal is transformed into coke it initially expands and then contracts. The amount of expansion is very important in terms of coke oven pressure and the amount of contraction is important in terms of ease of pushing (removing coke from oven). The balance of expansion and contraction depends on:

- porosity of the charge.
- porosity of the coke.
- weight of volatile matter lost.

Also based on the chemistry of the ash there may be a tendency of the coke to stick to the brick lining of the ovens.

The expansion or contraction of a coke charge varies with rank. When a high-volatile coal is coked there is an increase in porosity as the plastic layer forms vesicles, but this is more than offset by the substantial loss of volatile matter. The charge will generally not generate a lot of pressure. The plastic components in higher rank coals lose less volatile matter so that there is a tendency to generate more oven pressure and to contract less in the final stages of coking.

Reactive macerals in the coal swell, soften and melt during coking and the less viscous components flow around the inert fragments eventually solidifying into a mosaic of anisotropic carbon grains that cement the inert particles together. These grains can have a number of shapes that generally indicate the amount of plastic flow that took place. A classification for coke textures is suggested by Gransen et al. (1991; Table 3). Vitrinite from high-rank coal (Rmmax>2%) does not melt and forms large anisotropic grains descripted as domains; usually with length to width factors greater than 2 (Photo 1). Vitrinites from medium-volatile coals form a lot of medium-sized, anisotropic mosaic grains (Photo 2). Vitrinite from high-volatile coals forms fine anisotropic mosaic grains and isotropic areas indicating high fluidity (Photo 3). Gransen et al. (1991) studied the relative proportions of textures in coals made from coals of different ranks (Figure 12).

At any rank there is a distribution of reflectance values of the vitrinite. These are the V types that are fundamental to predicting stability index using petrographic data. Generally there appears to be a reasonable correlation between vitrinite V type and coke texture it generates. For example V types of 7 to 9 will produce a lot of isotropic, very fine or fine mosaic texture whereas V types of 10 to 15 produce medium mosaic and domain textures. As the rank of a coal increases the V type distribution shifts to the higher numbers and the proportions of coke texture types changes accordingly.

The volatile content of the reactive macerals can decrease by 30% from high-volatile rank to low-volatile rank reatives which results in 30% less carbon cement left after loss of the volatile component. Strong cokes require a balance between medium and fine mosaics, domain structure and inert material. The inert macerals, fusinite and inert semifusinite, remain isotropic and undergo little change in shape. Mineral matter contained in cells in the semifusinite will have minimal effect on fluidity or coke textures, but may cause fracturing of the inert carbon grains at higher temperatures. Mineral matter that is disseminated in desmocollinite may inhibit fluidity and formation of medium mosaic structures. In fact Cameron and Botham (1964) found that the accuracy of predicted stability indexes is improved by considering desmocollinite to be a semi inert maceral. Mineral matter or inertinite fragments that are large can severely weaken the coke.
Photo 1: Coke from low-volatile coal with large highly anisotropic grains described as domains; usually with length to width factors greater than 2.

Photo 2: Medium-sized anisotropic mosaic grains formed from a medium-volatile coal.

PREDICTING COKE QUALITY USING PETROGRAPHY OR RHEOLOGY

Most models that predict cold coke strength (ASTM coke stability factor or JIS DI 30/15 index) use petrographic or rheological criteria. The petrographic methods can be made to predict accurate results for coal with high concentrations of semifusinite by empirically choosing the proportion of semifusinite that is considered reactive. This proportion is more than 1/3 but varies from one coal to another. CANMET assumes that 50% of the semifusinite is reactive when making the calculations and Pearson (1984) calculates the amount of reactive semifusinite based on the rank of the coal. Figure 13 illustrates the agreement between predicted stability index and measured stability factor for a number of western Canadian coals using the CANMET 50/50 method of assigning inerts.

Coin and Broome (1997) have recently called into question the usefulness of plots which use petrography to predict coke stability index, such as those of Schapiro and Gray (1964). The data set used by Schapiro and Gray is quite restricted and the vitrinite contents of the samples are strongly correlated with rank. Vitrinite contents increase from about 60% at Rmax of 0.8% to 75% at an Rmax of 1.8%. This trend is counter to that found in coal from the Mist Mountain Formation in south-east British, in which vitrinite contents decreases as the rank increases (Ryan and Khan, 1998). Coin and Broome suggest that, based on data from Australian coals, stability factor is almost independent of vitrinite content at constant rank (Figure 14). Experience with WCC suggests that stability factor is influenced by variation in petrography at constant rank but that the influence may be less than previously thought.

Predictive methods based on measurements of coal rheology include the MOF diagram and the FSI versus volatile matter diagram. Noticeably absent from all models for predicting cold coke strength is any consideration of ash chemistry, particle size effects and operational parameters. Ash chemistry has a major effect on CSR but has not been shown to have much effect on cold coke strength indexes. Particle size can have a major effect on coke quality as can changes in coke battery operating conditions. Price et al. (1987) have attempted to put iso-stability factor lines on the MOF diagram (Figure 10b).
Table 3
CANMET CLASSIFICATION OF COKE TEXTURES

<table>
<thead>
<tr>
<th>TEXTURE</th>
<th>DESCRIPTION</th>
<th>MACERALS</th>
</tr>
</thead>
<tbody>
<tr>
<td>ISO TROPIC</td>
<td>no change in form</td>
<td></td>
</tr>
<tr>
<td>WEAK ANISOTROPY</td>
<td>little change in form</td>
<td>INERT SEMIFUSINITE</td>
</tr>
<tr>
<td>MOSAIC</td>
<td></td>
<td></td>
</tr>
<tr>
<td>very fine mosaic</td>
<td>&lt; 0.5 UM</td>
<td></td>
</tr>
<tr>
<td>fine mosaic</td>
<td>&lt;1.5&gt;0.5 UM</td>
<td></td>
</tr>
<tr>
<td>medium mosaic</td>
<td>&gt;1.5&gt;1.5 UM</td>
<td></td>
</tr>
<tr>
<td>coarse mosaic</td>
<td>&gt;10&gt;5 UM</td>
<td></td>
</tr>
<tr>
<td>ELONGATED FLOW TEXTURES</td>
<td></td>
<td></td>
</tr>
<tr>
<td>fine</td>
<td>L = 2xW</td>
<td></td>
</tr>
<tr>
<td>medium</td>
<td>W =&lt;1.5&gt;0.5 um</td>
<td></td>
</tr>
<tr>
<td>coarse</td>
<td>W =&lt;5&gt;1.5 um</td>
<td></td>
</tr>
<tr>
<td></td>
<td>W =&lt;10&gt;5.0 um</td>
<td></td>
</tr>
<tr>
<td>DOMAIN</td>
<td>large</td>
<td></td>
</tr>
<tr>
<td>ribbon</td>
<td>anisotropic</td>
<td></td>
</tr>
<tr>
<td>creased</td>
<td>areas</td>
<td></td>
</tr>
<tr>
<td>undulating</td>
<td></td>
<td></td>
</tr>
<tr>
<td>flat flow</td>
<td></td>
<td></td>
</tr>
<tr>
<td>BASIC ANISOTROPIC</td>
<td>anisotropic</td>
<td>high rank</td>
</tr>
<tr>
<td></td>
<td>form</td>
<td></td>
</tr>
</tbody>
</table>

1 so that the diagram can be used to estimate coke quality. It is unlikely that coke stability factor can be accurately and consistently predicted using only rank and a single rheology term. Coin and Broome (1997) consider that rank is the most important indication of the ability of a coal to make good coke and in this regard it should be noted that the iso-stability factor lines tend to be vertical for fluidities greater than 10 in the rank window 1% to 1.4%. This is very important, because it implies that in the rank window of Rmax =1% to 1.4%, stability factor is not sensitive to changes in MF.

Price et al.,(1987) and Pearson (1980) have both produced FSI versus VM (daf) diagrams with iso-stability factor contours (Figure 15). Pearson examined 109 British Columbia experimental cokes and Price et al. used the CANMET database, which includes oxidized and high ash samples. The contour patterns differ some what probably because of the inherent difficult in fitting contours through the data. It is interesting to note that where as, at a medium-volatile rank, stability factor is not sensitive to changes in fluidity, based on the contour lines in Figure 15, it is sensitive to changes in FSI. There appears to be some decoupling of the relationships FSI versus VM and fluidity versus rank.

The combination of FSI versus VM (daf) may be a more sensitive indicator of coke quality than the MOF diagram. This may be in part because VM (daf) is sensitive to changes in petrography as well as rank. In fact for a sample of constant rank both FSI and VM (daf) will increase as the amount of vitrinite in the sample increases and this explains in part the positive slope of the iso-stability factor contour lines.

One advantage of the FSI versus VM (daf) plot is that it requires less sophisticated analytical data than the fluidity versus Rmax plot. Price et al. found that, for diagrams utilizing rank and a single rheology term, the FSI versus VM (daf) diagram was the most accurate for predicting stability factor.

The iso-stability factor contours on the MOF and FSI versus VM (daf) diagrams are curved and two coals of different rank with low stability factors can be mixed produce a blend with a stability factor value higher than that of either component coal. A check of a number of Canadian coals (Figure 15a, 17a) indicates that based on the iso-stability factor contours in the diagrams they both often predict low values of SF.

There is a weak tendency for the data in the plots to increase in the direction of high iso-stability factor contour lines but the general consistency between posted values and contour lines values is poor.

Coin and Bloome (1997) suggest that there is no correlation between stability factor and petrography at constant rank (Figure 14) and they also question the
usefulness of Gieseler Plastometer results in predicting coke quality. This is not completely born out for WCC based on the limited data in Figure 16b in which there is some tendency for stability factors to increase as maximum fluidity increases at constant rank. There is an approximate linear relationship between total reactivities and maximum fluidity at constant rank (Figure 3), implying an increase in stability factor with increasing total reactivities content. Certainly the general steepness of the iso stability factor contour lines in the MOF diagram indicates a large measure of insensitivity of stability factor values to changes in fluidity at constant rank.

Without attempting to quantify the usefulness of the MOF and VM (daf) versus FSI plots, it appears that the data are more consistent with the contour pattern in the FSI versus VM (daf) plot in agreement with the conclusion of Price et al. (1987). Data in Figures 16b and 17c are for blends of coals of differing ranks and in both cases the diagrams do not appear to be useful in predicting blend stability factor values.

COMPARISONS BETWEEN EASTERN USA, AUSTRALIAN AND WESTERN CANADIAN COALS

The fluidities of Western Canadian coals (WCC) are generally less than that of eastern USA coals of similar rank. Part of the reason may be because WCC generally have less vitrinite. It has also been suggested that vitrinite from the USA coals has more hydrogen than vitrinite from WCC of similar rank (Price and Gransden, 1987). This would explain the lower fluidity of WCC because there is a moderate correlation of fluidity to hydrogen content for medium-volatile coals. The average hydrogen content of vitrinite from WCC at R_{max} = 1.22\% is approximately 5.05\% and that of Appalachian coals is 5.25\%. This is somewhat surprising because usually vitrinite reflectance corresponds with the hydrogen content of vitrinite. One possibility is that there are variations in hydrogen content of the vitrinite sub macerals. Possibly desmocollinite contains less hydrogen that telocollinite. Mastalerz and Bustin (1993) measured the hydrogen content of individual maceral grains and found no statistically significant difference (telocollinite 2.64\% H and desmocollinite 2.63\% H). If there is a variation in the fluidity of the sub macerals, then probably telocollinite and tellinite are more fluid than desmocollinite. Reflectance is measured on telocollinite, which may have similar hydrogen contents in both coals. If WCCs have more desmocollinite, then the overall hydrogen content of WCCs may be lower than that of Appalachian coals. Unfortunately most of the literature that provides petrography and rheology data does not differentiate the vitrinite sub macerals.
The $R_{\text{max}}$ is rank dependent, but also tends to increase as the amount of inertinite in a sample increases (Ryan, 1997).

A partial explanation for the different hydrogen contents of the vitrinites from the two areas may be found in the biaxiality of the WCC compared to the possible uniaxial negative optical properties of vitrinite from the eastern USA coals. It is possible that the rank of the Carboniferous eastern USA high-vitrinite coals was imposed during a simple burial history and they are uniaxial negative, whereas the more complicated maturation history of WCC ensured that they are predominantly biaxial negative. Bustin et al. (1986) heated and deformed samples of anthracite and was able to increase the reflectance, as well as the biaxial negative component of the reflectance indicating surface. Clearly deformation does change a uniaxial negative reflectance indicating surfaces into a biaxial negative one, possibly with higher $R_{\text{max}}$% values.

The value $R_{\text{max}}$% (true maximum reflectance) is greater than $R_{\text{mmax}}$% (mean maximum reflectance) for biaxial coals and the relationship for Mist Mountain coals (Grieve, 1991) is given by:

$$R_{\text{max}}\% = R_{\text{mmax}}\% \times 1.044$$

Grieve suggests that the biaxiality is caused by a differential stress factor imposed upon the hydrostatic stress, which is responsible for the uniaxial component of the reflectance indicating surface. If this is the case, then coals that experienced a simple burial history will have uniaxial reflectance indicating surfaces, $R_{\text{mmax}}$% will equal $R_{\text{max}}$%, and $R_{\text{mmax}}$% will be a true measure of rank. However for coals whose rank was established during folding, $R_{\text{mmax}}$% will be an underestimate of rank and the true rank will be indicated by $R_{\text{max}}$%. Vitrinites from WCC have approximately 2% less volatile matter and 0.1% less hydrogen than their Appalachian counterparts. Many WCCs are biaxial negative consequently their $R_{\text{mmax}}$% values could have been under estimated by about 0.05%. This, based on the hydrogen versus $R_{\text{mmax}}$% relationship in Stach et al. (1982) could explain about 0.06% of the missing hydrogen. If the true rank of the western Canadian vitrinites is used, then they may appear to be less anomalous in terms of rheology and hydrogen content.

There is some data that indicate that Australian coals of the same rank as WCC are more fluid (Leeder et al., 1997). Part of this effect may be due to variations in petrography. A limited amount of fluidity and petrographic data grouped by average rank are plotted in Figure 18. It appears that at a rank of high-volatile bituminous there is a distinct difference between WCC and Australian coals, however at higher ranks the apparent difference may be due to higher concentrations of inertinite in WCC.

Gransden et al. (1991) studied the relative proportions of reactive coke textures in cokes made from WCC and eastern USA coals of different ranks (Figure 12). There is a strong correlation between vitrinite V type and the type of coke texture generated. They found very little difference in the proportions of textures for cokes derived from the two types of coals at similar rank. If the amount of reactive coke and the relative proportions of the textural types are important in determining the quality of the coke, then the proportions of vitrinite V types must be more important than any subtle changes in fluidity or chemistry of vitrinite. This point is very important for WCC because it means that fluidity at a particular rank is not as important in determining coke quality as petrography especially V type distribution. This conclusion is similar to that reached by Coin and Broome (1997) with regards to fluidity but there is still the implication that petrography in terms of the amount of reactivcis and their V type distribution are important.

**CONCLUSIONS**

Fluidity is one of three primary measures of coal rheology, the other two being FSI and dilatation. The fluidity of a coal is dependent on rank and the content of reactive macerals. For medium-volatile coals there is a threshold reactive maceral content below which the coal has no fluidity. Above the threshold fluidity can increase rapidly as the content of reactive macerals increases.

Fluidity data provide information about the formation of a plastic phase during coke making. This is only one of the processes that takes place as coal is converted into coke and its usefulness in predicting coke quality has been questioned by Coin and Broome (1997). The steepness of the iso stability factor contours in the MOF diagram also indicates that coke quality is insensitive to changes in coal fluidity over a wide range of rank values.

There is no simple way of predicting the fluidity of a blend of coals. It is not possible to weight average the log values of MF, though it may be possible to derive a blend fluidity using petrographic data for the component coals. More complicated methods of adding reactivcis exist but they do not take into account
Figure 16: (a) MOF diagram with posted stability factor values for a number of WCCs and one eastern USA coal (solid symbol) with stability factor values are posted next to the data. (b) Window of (a) showing data for a number of single seam samples of similar rank. (c) Data for blend samples composed of seams of different ranks. The data are for single coals but not necessarily product coals. The A following a stability factor values indicates an average value for overlapping data points.

Figure 17: (a) VM (daf) versus FSI diagram with posted stability factor values for a number of Canadian coals. (b) Window of (a) showing extra data indicating some agreement between posted stability factor values and iso-stability factor contour lines. (c) Data for blend samples composed of seams of different ranks.
interaction between the reactive particles from the different component coals.

As with petrography and other measures of rheology, fluidity is used to predict coke quality, in particular cold coke strength as measured by ASTM stability factor or the JIS DI 30/15 index. Coke quality is strongly dependent on coal rank and somewhat dependent on petrographic composition. It also depends on coal size consist and coke battery operational parameters. No single measure of coal rheology will consistently and accurately predict coke quality.

If a single measure of rheology suggests that a coal will make poor coke, this should not be taken at face value. Other measures of rheology should be checked for consistency. Low FSI is probably the best current coal-derived indicator of poor coking potential for a single coal. In fact the FSI versus VM (daf) diagram, can be much more useful than the MOF diagram in providing a rough estimate of coke quality. It has the added advantage that it is much cheaper.

Western Canadian coals contain high proportions of semifusinite and as yet there is no accepted way of predicting what proportion of this material will become plastic. This introduces a level of uncertainty in petrographic techniques for predicting coke quality. Measures of coal rheology effectively take into account the semifusinite problem but the results are often inconsistent because they utilize small samples. No test approaches the quality of the data obtained from a movable wall coke oven test which links coal rheology data to industrial coke oven experience.

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