

# **DETERIORATION OF COKING COAL QUALITY IN SAMPLES AND STOCKPILES**

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## **Abstract**

Coking coal is a perishable commodity as its quality properties can deteriorate from exposure to air, increased moisture content and handling. These factors, which affect the quality of the coke product, can occur during the sampling, mining, transportation, storage and utilization of these coals.

This paper discusses several studies to evaluate the impact of these variables on the quality of the coking coals, including Canadian work undertaken by the Canadian Carbonization Research Association and its industrial members using laboratory, barrel and small stockpile sized samples.

This paper reviews studies that show that there is a significant reduction in coal thermal softening/melting properties and some coke quality characteristics after approximately one year from mining. The rate of deterioration for coal and coke properties depends on the coal and can be reduced if it is stored under vacuum, at lower temperatures (~freezing point) or in larger volumes such as stockpiles.

Increased coal moisture can cause handling problems and also reduce the coke oven charge

weight (i.e. bulk density of the charged coal), which reduces both the coke yield (productivity) and coke quality. This can be exacerbated by additional handling which creates more fines (higher surface area) that increase the exposure to air and the amount of water that can be retained.

Canadian coking plants must deal with these problems for several months during the winter when new coal is not being delivered. Coke quality is maintained by using winter planning practices that adjust coke oven blends.

## **Key Words**

Coal oxidation, coal moisture, coal fines, coal thermal softening, bulk density, coke quality

## **Introduction**

Coking coals are a perishable product with a shelf-life similar to food products, whose quality decreases over time - for samples or coal at mines, ports or coke plants.

At least three factors can contribute to the change/deterioration of coking properties over time –

- exposure to air (oxygen),
- moisture (rain/snow) content and
- re-handling (size reduction) of the coal.

The time between mining and carbonization of seaborne coking coals can often take a number of weeks or months. Canadian coke-makers stockpile coal for winter use and each year must deal with this issue.

This paper will present and discuss the impacts of these variables on coking coal quality and address, where possible, practical ways to manage the impact of these factors on the coal/coke quality.

## Weathering (oxidation) of Coking Coals

Coking coals have one unique quality property that differentiates them from all other coals – they soften/melt when heated to high temperatures (i.e. >300 °C) and solidify into a solid mass “coke” at around 500 °C. In-situ, coking coal seams contain methane. When these seams are exposed to water or air or broken, i.e. during mining or taking of samples, the methane desorbs from the coal structure which can then re-absorb oxygen from air or water (i.e. rain or ground water). Oxygen reacts with the coking coal (called oxidation or weathering) and reduces its chemical ability to thermally melt to ultimately produce coke. Very little oxygen is required to bring about some change and once the protective methane blanket starts to dissipate, weathering begins. This section will discuss the quantitative impact of oxidation on coal and coking properties.

### Impact of Weathering on Coal Gieseler Maximum Fluidity

Many studies have been undertaken to determine the changes due to oxidation in the coking coal using lab-scale measurements of Gieseler Plastometer and/or Dilatometer. Some literature data as well as internal studies on the impact of ambient temperature weathering on coal Gieseler maximum fluidity (MF), are summarized in Table 1 (1,2,3,4,5). It includes a number of coals of differing sample weights/volumes.

Inspection of the table shows that the Gieseler MF of all samples, no matter the sample size (weight), significantly decreases over time with exposure to air (weathering). However, sample weight (size) appears to play a role, as the Gieseler MF of the larger sized (barrels, 100 t stockpile) samples decreased more slowly than a 10 kg (lab) sample. Once exposed to air, even storing lab sized sample under vacuum does not stop the weathering process,

although it slows it down. Studies have shown that storing 10 kg sized samples at near freezing temperatures also slows the weathering process – and can have almost the same impact as storing under vacuum (4). However, once coal is exposed to air and some oxygen enters the coal structure, the thermal softening/melting properties start to degrade, no matter if the coal was stored under vacuum, at low temperatures or in large stockpiles. It has also been observed at the Teck coking coal mines that the Gieseler fluidity of freshly mined coal drops very quickly and then the rate drops to a slower long term rate, Table 1 (5).

To attempt to quantify this impact, the results for samples with at least 300 days of data were plotted in Figure 1. The data included large (100 t and barreled) samples and a 10 kg sample exposed to air at room temperature or stored under vacuum where no further exposure to air occurred. The larger samples (barrels, 100 t stockpile) degraded at close to the same rate and were averaged together in Figure 1.

The thermal softening/melting results degrade in all cases, and as might be expected - more slowly in the vacuumed stored sample. After a year, these samples had lost 50% of their MF, the bulk samples 75% and the lab samples 95% of their MF.

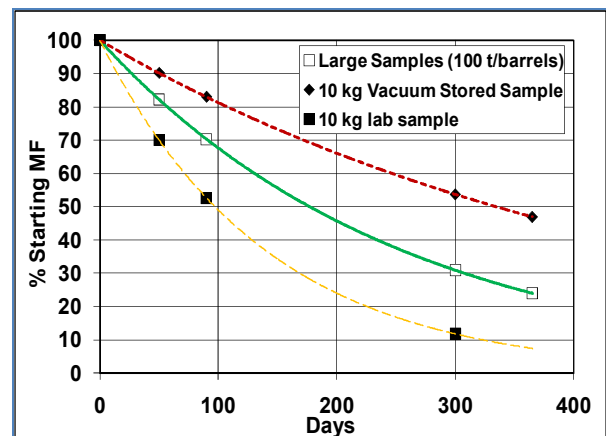


Figure 1: Impact of weathering on MF

Degradation studies conducted on coking coals found that MF decreases significantly on/near the surface of a 20 m high stockpile whereas MF degradation at the centre was far less, even after several years (6). As an older stockpile is reclaimed, the surface coal would be expected to have more serious MF deterioration than the coal underneath.

### Impact of Weathering on Coke Strength

The impact of oxidation on the strength properties of the resulting coke of individual coals and blends has also been studied by a number of organizations using technical scale and semi-industrial coke oven tests (1,2,3,7). In all cases the coal or blend thermal softening/melting properties had degraded over the testing period which ranged for up to a year. It was found that coke strength trends varied, depending on the coal or blend being studied and the type of coke strength test being used.

Selected ambient temperature coke tumble test results from coke made from coal or blends weathered for up to a year are shown in Figure 2.

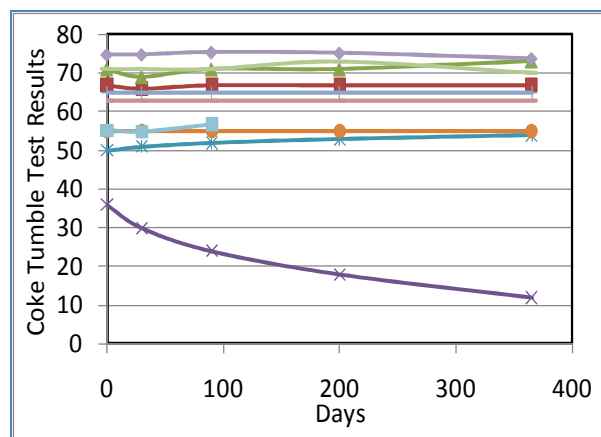


Figure 2: Impact of weathering on coke strength

There was little if any change in coke tumble test results for coke ASTM Stability/Hardness, JIS 30/15 and 150/15 and IRSID and Micum coke tumble strength indexes from any of the coals or blends,

except for one low rank, low MF coal where some deterioration was noted (7). This isn't unexpected from studies of predicting coke tumble strength, since the thermal softening/melting properties of the coals/blends during the testing period were still sufficient to ensure proper coke formation – i.e. 20-100 ddpn (8), with the exception of the one low MF coal. A coal with very high MF may improve its coking strength with weathering, if the coal is inert poor (7).

The high temperature evaluation of coke strength by CSR is a popular method of assessing coke quality. The impact of weathering time on the CSR of the coals and blends in Table 1 was also studied. The results are summarized in Figure 3 (see Table 2).

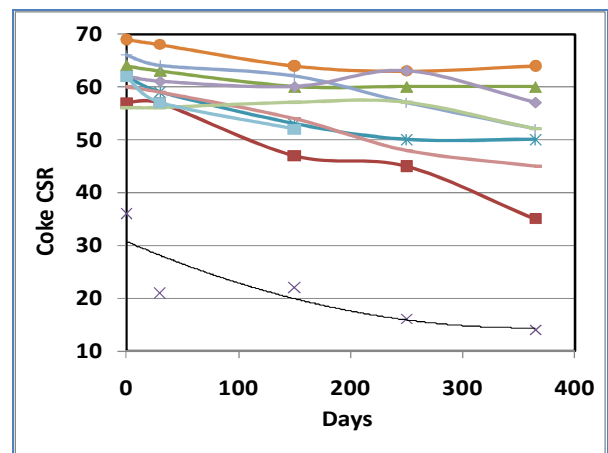


Figure 3: Impact of weathering on coke CSR

With the exception of one sample, the results indicate there is a general drop in CSR for the coals/blends studied over time. Some samples exhibited a significant decrease in CSR of 5 to 10 units after only 90 days. On average, the trends showed that the CSR degraded by almost 1 CSR unit for each month the coals were stockpiled, or about 3 units after 90 days. One year old coals had CSR's that on average were 11 units lower than at the start.

It was concluded after reviewing this information that

weathering can cause a significant decline in coal MF and coke CSR properties, over a time interval of about 1 year. In contrast, the resulting coke tumble strength properties were not as impacted for good coking coals, although an increase or decrease is possible because of differences in the characteristics of the coal (i.e. rank, inert level, MF). Therefore, coking coal being assessed in labs should be tested as soon as possible after the coal sample is taken and where possible stored in a cold environment. Commercial cargoes should also be used as quickly as is practical to maintain the optimal coal/coke properties.

### Impact of Moisture on Coking Coal

The water content of a coal is controlled by the production/shipped moisture, amount of rain-fall during transportation and stockpiling, amount of water sprayed on coal at various stages for dust control and stocking practices. Typically, seaborne commercial coking coals are delivered at around 7 to 10% moisture. Coal moisture content influences the dustiness, handling and coking aspects of utilizing coking coals, which will be discussed in this section.

### Impact of Moisture on Coking

Coal moisture directly impacts coking – by changing the bulk density (weight) of the oven charge and by impacting the quality of the coke produced.

The impact of moisture on coal bulk density is seen in Figure 4 for a Canadian and a US coal. The bulk density for both was lowest at 8 to 10%, the level most often found in seaborne coking coals.

The bulk density of the charge in either a technical and conventional industrial scale coke oven, changes the weight of coal in the oven. Actual results from an industrial oven appear in Figure 5. This directly influences industrial coke oven productivity.

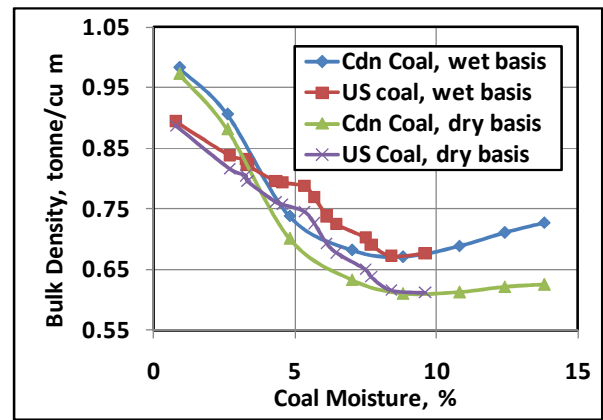


Figure 4: ASTM Bulk Density of Two Coals

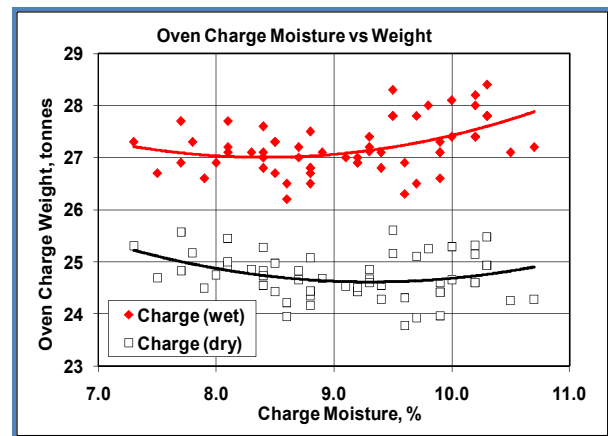


Figure 5: Industrial charge moisture impact on charge weight

Increasing the charge oven bulk density also increases coke density and coke tumble strength (9), Figure 6, and coke CSR (10).

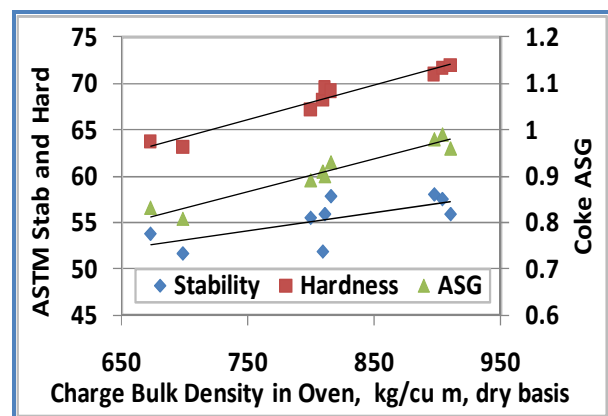


Figure 6: Technical oven coal charge bulk density impact on coke quality

The cokes in Figure 6 were produced in a technical scale coke oven, whose charges were prepared to three target moisture levels from 3 to 4% (higher bulk density in oven) to 8 to 10% (lower bulk densities).

If industrial coke oven charge moisture can be reduced below the 8-10% moisture range, the bulk density and weight of coal charge in the oven would increase, improving oven productivity and coke strength and CSR. This could reduce the requirements for higher priced prime coking coals and has led to the development of coke oven add-on processes such as DAPS and CMC that dry coal before coke oven charging (11).

### Coal Handling

The moisture content of the coal has a big impact on the handling of the coal. For example higher (i.e. 10 to 12%) moisture levels can lead to handling issues as the coal become sticky and at very high levels (i.e. 12-14%) this can lead to instability in stockpiles or in coal in vessel holds (12).

Seaborne coking coal mine products typically have 7 to 10% moisture, which is good for handling. In addition, this level is required to ensure the coals are not unduly dusty during transportation and storage (stockpiling). In addition to rainfall, further water is often used on stockpiles during transportation and storage to control dust at those locations.

### Moisture Migration in Stockpiles

Most coking coal shipments from Canada are stockpiled and shipped from the western ports of Vancouver or Prince Rupert in British Columbia, both which have “rain forest” climates which increases the moisture content of the stocked coal. Consequently clean coal products are both mechanically and thermally dried at mines to levels around 6-8%

before railing. After drying, moisture levels increase from rail car capping sprays, from rain en-route to the coast, during stacking, vessel loading and stockpile storage, and from water used for dust control during stacking, storage and loading vessels. This typically increases the mine product moisture by about 1 to 2%, more in rainy winter months and less in dry months. Fortunately, it has also been found that coal shipments to Asia and Europe typically arrive with about 1% lower in moistures as a result of water drainage from the coal in the vessel holds (13).

These sources of water addition cannot be controlled but an understanding of moisture behavior in coal stockpiles may allow some management of cargo moisture consistency. Consequently the CCRA and CanmetENERGY undertook investigations of port stockpile moisture distributions, as well as studied water migration in coal using vertical pipes (14).

A port stockpile study was conducted in February 2002, at Westshore Terminals, Vancouver. One stockpile was filled with new coal 4 to 6 weeks before re-claiming and sampling was conducted the morning after loading the vessel. Sampling was possible from the re-claimer benches that cut through the entire height/width of the pile. A cross-section of the coal stockpile showing the moisture levels in the areas accessible for surface sampling are seen in Figure 7.

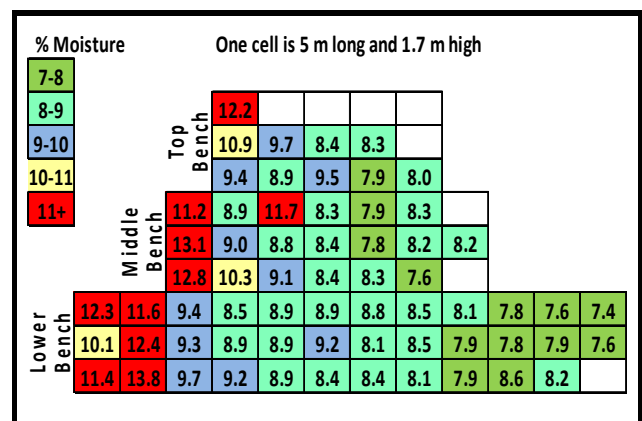


Figure 7: Vancouver Port Stockpile Moisture Levels

The Figure clearly shows that the interior of this stockpile (originally 25 m high) was lower in moisture and similar to the railed mine product (i.e. 7-9%) compared to higher moistures near/at the surface (10– 13%). Moisture contents at the very bottom of this pile were not particularly elevated compared to the coal above it. Certainly, within the 4 to 6 weeks that the coal was at Westshore, there was little/no migration of moisture in the pile.

To better understand moisture migration in coal, detailed moisture drainage tests were undertaken at CanmetENERGY using 0.1 to 0.2 m diameter plastic pipes packed with coal. Short lengths of plastic tube were packed with coal to a constant bulk density then mounted together to form 2 to 7.3 meters high columns. Drainage from the bottom was monitored over periods of one to four months. Moisture migration was followed by cutting the columns in 0.3 m sections and analyzing each section for moisture. A number of variables were studied, including the effect of initial coal moistures (i.e. 8, 10, 12, 14, 16 %), the impact of rain by adding water daily, the use of a surfactant in the water to increase drainage and the impact of vibration (14).

The effect of initial coal moisture on the rate of drainage and water migration within the columns was studied in several tests. Initially two meter high columns containing coal at different initial moisture contents varying from 7.8 to 15.6% were studied. Immediate drainage was observed for the 14 and 16% moisture coals but a month later there was still no drainage from coal moisture lower than 14%. Profiles of the moisture content in the columns showed that the moisture had migrated down for all coals and the bottom 0.6 m of the columns was significantly higher in moisture than the coal above.

The results from a 7.3 m high column with a 10.5% moisture coal, dismantled after 4 months are seen in Figure 8.

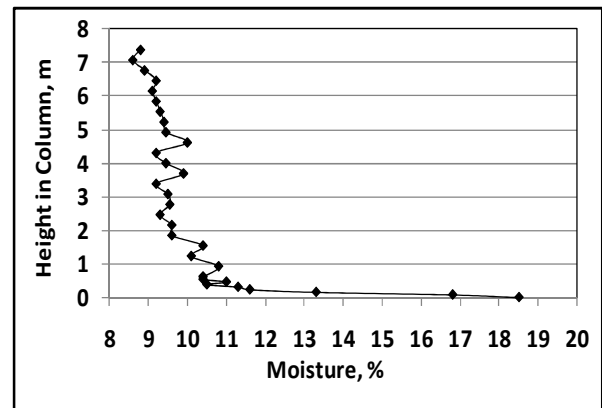


Figure 8: Coal moisture gradient (120 days, 7.3 m column, initial moisture 10.5%)

Some drainage had started after 2 months, however the total that drained by the end of the 4 month test was very small (i.e. initial average coal moisture was 10.5% compared to the final average of 10.4%). In a parallel test, the addition of surfactant didn't appear to make any difference to the results.

These test show that western Canadian coals with moistures from 8 to 10% don't drain even after 4 months, but the moisture does migrate towards the bottom of the column (stockpile) where the moisture levels are high (i.e. over 12%).

Drainage and migration results from a vibrated column, to simulate vessel vibrations during shipment, increased moisture migration rates and drainage was six times higher than a non-vibrated column. The effect of the vibration helps explain why water readily drains from the coal during vessel shipment.

At Westshore, rain and water dust sprays add moisture to the coal in the stockpiles. To simulate the impact of these additions, which occur mostly in

the four rainy winter months, water was added to the top of the column each day, equivalent to the average rainfall for the winter months. When coal starting at 6.5% moisture was studied in 7.3 m columns over a period of 4.5 months, drainage started after approximately 2 to 2.5 months. The moisture of the coal in the column had increased to around 14% and then leveled off as the drainage rate balanced the water addition. This level is similar to saturated coal piles that have issues with stability (12). When the water additions were stopped, the columns continued to drain and the average coal moisture gradually decreased to about 11 to 12%.

In tests with columns of different heights to estimate how long drainage would take for different height piles with “rain” additions, the time to initial drainage was found to be directly related to the column height, Figure 9.

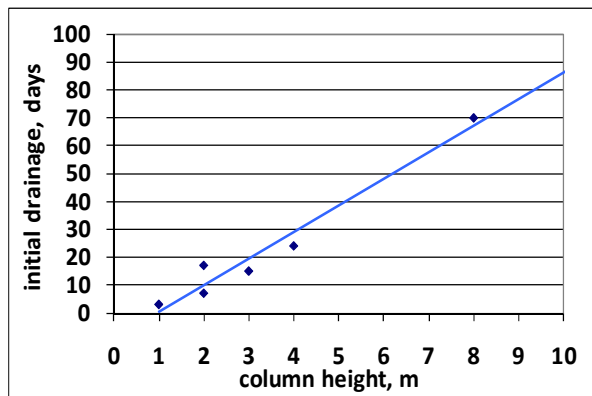


Figure 9: Days to drainage with Vancouver equivalent rain-fall moisture added, (coal moisture 6.5 – 8%)

Even when Vancouver rainfall is considered, it is concluded that normally any rain or dust suppressant moisture falling on a stockpile will be absorbed and not have time to drain before normal reclamation to vessels (i.e. < 4 weeks for the last 2 years), except when extremely heavy rainfall causes stockpile surface moisture saturation and the rain runs down the outside of the piles (13).

These observations point out several ways to control the moisture of coal being loaded to vessels. These include – (i) minimizing stockpile footprints, (ii) filling in reclaim benches with new coal and (iii) reclaiming coal through the top, middle and bottom of stockpiles, (iv) compacting stockpiles and (vi) using coatings (i.e. latex) to help shed rain/sprays. Application of some or all of these strategies could help to manage the consistency and level of moisture in coal cargoes and all these methods are used in various coal storage sites around the world (13).

### Stockpiling Impact on Size

The level of fines (i.e. % - 0.15 mm or - 0.5 mm) in a coal has a direct relationship to the ease of handling a coal and the ability of the coal to hold moisture (15). Generally a finer coal with a higher surface area holds more moisture and becomes stickier and more difficult to handle than coarser sized coal. Continually re-handling coal in stockpiles (dozing, reclaiming, etc) leads to increased coal fines content.

It is well documented that mining and handling of coal leads to the generation of fines (<0.5 mm) (15,16,17). Test work conducted on bituminous coals from BHP’s Illawarra Collieries, Kemira and Cordeaux Australia, in the late 1980s and early 1990s, found that dropping 20 kg of these coals (100% < 63 mm) from a 45 m height resulted in the formation of 20% fines, < 0.5 mm. These investigations derived a relationship between extent of breakage of a material to  $(\text{height})^{0.5}$ , for a given particle size and found that fines generation can be reduced by increasing the number of drops made to achieve a given drop height.

Increased fines in coal can create a number of problems. This makes the coal dustier, increases the coal moisture holding capacity and stickiness, makes it pack less densely in coke ovens (i.e. reduces bulk density with drop in coke quality and oven

productivity) and can increase the rate of degradation of the thermal softening/melting properties of the coals by increasing its surface area which enhances its exposure to air (4,6).

For all these reasons, minimizing handling/re-handling to a minimum is a prudent operating strategy.

## **Coking Plant Winter Strategy**

Stockpiling of coal for longer times (i.e. 3 – 6 months) is practiced in Canada and several European countries as the coal cannot be delivered over the winter months. This can lead to increased moistures in the coal which can be as high as 12 to 14% in the spring with all its associated ticking/handling and oven control problems. In addition the coal has continued to weather. This section will discuss how this issue is dealt with at ArcelorMittal Dofasco, Canada.

All coal deliveries to ArcelorMittal Dofasco in Hamilton are by “Laker” vessel as there is no rail delivery available. Coal is received from the end of March until the locks close at the end of December and the coal has to be stockpiled for several months during the winter. The coals used in the oven blends are low, medium and high volatile bituminous components, along with a percentage of lower rank high volatile coal.

Due to historical coke quality winter problems, ArcelorMittal Dofasco has developed a good understanding of the shelf life of their coals. It has been found that certain coals are not capable of being stored over the winter months without causing coke quality issues. This work was completed over several shipping seasons and took into account coal fluidity, coal dilatation, carbon form analysis (coke

microscopy) as well as moveable wall technical scale coke oven testing.

ArcelorMittal Dofasco's coal stockpiles are packed to about 60-65 lb/cu ft. The issues of coal storage that must be dealt with are coal aging (weathering, oxidation) and moisture gains.

Aging of the coal in the stockpiles leads to decreased coal caking properties. Traditional oxidation tests such as the ASTM alkali extraction test (D5263-98 (2008)), does not really show values that would indicate actual oxidation. Aging is a much milder process, but its impact on lower than desirable thermal softening/melting coal can result in significant drops in both cold coke strength and CSR over storage time. Aging leads to a shift in the coke carbon forms, especially the lenticular and more significantly the circular and isotropic forms. This mostly occurs with the high volatile coals. As a result, lower coke stability and CSR is observed.

In order to solve this issue, the strategy is to purchase high volatile coals having adequate fluidity and dilatation to begin with, and avoiding coals that are high in inherent oxygen. High volatile coals that have fluidity values <20,000 ddpm and dilatation values <150% are avoided for winter usage. However, these coals can be used during the shipping season since they can be utilized in the blend when they arrive. Dilatation has been found to be the most critical property measured, for maintaining an adequate caking capacity over the winter months. If a coal has a fluidity value of 30,000 ddpm and dilatation value of >150%, storage during the winter months does not lead to a major influence on the subsequent coke quality.

Stockpiles have increased moistures during the winter months especially if there are high levels of rain and snow. Oven operating rates are adjusted to compensate for the wetter coal since less coal is



charged per oven. The coal blend ASTM cone bulk density (D291-07) aim is monitored every two hours using a control chart. Oiling of the charge is used to decrease the moisture effect on bulk density, but there is an upper limit that is effective.

The packed coal piles are worked to expose the drier coal in the interior. As the pile is depleted towards the end of the winter, coal is moved from the bottom to the top to try to "dry" this coal. In the spring, when a new shipment arrives this can be combined with the very wet stockpiled coals to lower then net moisture content. Stockpiles are also designed with adequate drainage paths along the sides of the piles to allow for run-off from rain and melting snow.

## Conclusions

Since coking coal is a perishable commodity as the quality of coke produced from the coal deteriorates over time, coals samples should be kept as cool as possible and analyzed immediately. Commercial coal should be utilized as soon as is practical as longer term delays leads to a reduction in the coking properties of the coal. In several studies, the MF and CSR decreased noticeably after one year of storage.

Moisture levels in coking coals impact the resulting coke strength and CSR. High moisture levels (i.e. 12%) in oven charges can lead to lower quality coke and oven productivity.

For Canadian coals, chances are that most moisture added to coal during transportation and storage will be loaded with the coal to the vessel. Improvement in the consistency of the moisture of coal shipments is possible with stockpile management.

Minimizing the handling of coals will help to control the level of fines in the product. Increasing fines content can lead to additional moisture, handling and weathering problems with a coal.

Where stockpiling for longer periods is required, strategies for coal selection are imperative. Laboratory studies including carbon form analysis have been instrumental in helping determine why coke quality issues have occurred due to storage time. Coal will age naturally and adsorb oxygen in the process. Adequate caking and coking properties at the outset are most important. Stockpile management is also critical during the winter months.

Time is of the essence in the evaluation and use of coking coals.

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