EVALUATION OF BIOMATERIAL SUBSTITUTION IN METALLURGICAL COKE

THE EVALUATION OF BIOMATERIAL IMPACT ON METALLURGICAL COKE STRUCTURE FOR SUBSTITUTION OF COAL IN OPERATIONAL COAL BLENDS

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Lay Abstract

Metallurgical coke remains the main fuel and reductant source for ironmaking by blast furnace operation. Quality metallurgical coal, a fossil fuel, is required to produce coke. This work continues ongoing steel industry research investigating biomaterial substitution of coal as a more sustainable option. Coal is considered a new release of greenhouse gas (GHG) emissions when used in the steelmaking process compared to a biomaterial which is regarded as a GHG neutral replacement. Three raw biomaterials, available to industrial users, were evaluated for substitution at low amounts and compared to an operational coal blend. The substitution could allow for GHG emissions of the cokemaking processes to be reduced if quality coke can be produced. The interaction of the biomaterial substitutions with coal during the coking process is evaluated in this work to better understand the resultant coke textures related to reduction in coke strength from the substitution.

Abstract

Carbon is a necessary reductant in steel production to convert iron ore to metallic iron. The use of coal and coke causes CO_2 emissions to be released into the environment. Using bio-based carbon sources has the potential to offset these emissions and reduce cokemaking overall carbon footprint. The use of biomaterial in coal blends reduces the fossil fuel requirements but to what capacity and type of biocarbon can replace coal is unknown. The full effects of coal and coke characterization from the addition of biomaterial are unknown. In this work, raw biomaterials available to industrial users were evaluated for substitution at low amounts in operational coal blends. Physically, the optical properties of carbon coke forms can provide insight into the strength, reactivity, and performance in the blast furnace, resulting from coal rank and type. The interaction of the biomaterial substitutions with coal during the coking process is evaluated to better understand the reduction in coke strength after reaction (CSR). For this purpose, a series of the pilot oven and sole heated oven tests were performed. When coal was substituted with low amounts of raw biomaterials, the most notable changes in coke texture analysis were to incipient and circular textures. In this work, data from a series of pilot oven and sole heated oven tests showed that fine coke textures and overall inerts increased. The changes in coke textures can be linked to decreases in coke strength after reaction (CSR).

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Table of Contents

List of Figures

List of Tables

List of Equations

List of Abbreviations and Symbols

CBI	Coke Basicity Index
CRI	Coke Reactivity Index
CSPA	Canadian Steel Producers Association
CSR	Coke Strength after Reaction
DDPM	Dial Divisions per Minute
FC	Fixed Carbon
FSI	Free Swelling Index
GHG	Greenhouse Gas
GVC	Gross Calorific Value
ISO	International Organization for Standardization
MBI	Modified Basicity Index
MFL	Metallurgical Fuels Laboratory
MOECC	Ontario Ministry of the Environment and Climate Change
MWO	Movable Wall Oven
PAH	Polycyclic Aromatic Hydrocarbons
PO	Pilot Oven
SHO	Sole Heated Oven
SSS	Site-Specific Standards
SS-SOP	Steel Sector Strategic Options Process
TNT	Trinitrotoluene
TSG	Total Specific Gravity
TSMP	Toxics Substance Management Policy
VM	Volatile Matter

Declaration of Academic Achievement

The author is the first author and main contributor of the written work included in this thesis with consultation from adviser Dr. Neslihan Dogan. No part of this work has been published or submitted for publication or for a higher degree at another institution.

Research test work was completed by employees at the Metallurgical Fuels Laboratory (MFL) of CanmetENERGY-Ottawa (Canmet), a national research laboratory within the Department of Natural Resources Canada under the guidance of Ka Wing Ng, Marc Saul, and Rino Canaj. Private consultation was held with industry expert Kevin DeVanney.

1. Introduction

The modern global steel industry has evolved from the centuries old process of ironmaking to producing one of the most highly used materials worldwide. The blast furnace remains the most commonly used process for iron production (Geerdes, Chaigneau, & Kurunov, 2015). Quality metallurgical coke is essential for the operation of the blast furnace process. Coke is required chemically for reduction reactions of iron ore and physically for burden support and permeability for gas movement of the blast furnace process.

Metallurgical coke production requires the use of metallurgical coal which is a fossil fuel. A blend of coals is needed to meet the required properties for cokemaking operations and consumption in the blast furnace. The chemical and physical properties are very important for operational choices at the coke batteries and blast furnace fuel needs. The largest challenge cokemaking operations face is the environmental impact and resource availability of quality coal. The steel industry is a large contributor to greenhouse gas (GHG) emissions, including carbon dioxide (CO_2). The use of coal and coke causes CO_2 emissions to be released into the environment. Coal is considered a new release of GHG emissions when used in the steelmaking process compared to a biomaterial which is considered GHG neutral. Using bio-based carbon sources has the potential to offset these new release emissions and reduce cokemaking overall carbon footprint.

Reduction of GHG emissions in cokemaking not only has a positive environmental impact but there are potential cost reduction and coke consumption for the blast furnace. As a result, there has been an increase in research for more sustainable options of coal and coke replacement for the ironmaking process (Ng, MacPhee, & Giroux, 2009). Injection of coal or natural gas has become standard in North American blast furnace operations for coke reduction, but a further reduction is desired (Ng, Huang, & Giroux, 2019). Ongoing steel industry research is investigating biomaterial substitution of coal as a more sustainable option. Substitution could allow for GHG emissions of the cokemaking process to be reduced if quality coke can be produced.

This study aims to assess biomaterial substitution of coal as a more sustainable option for blast furnace operations. Three raw biomaterials, available to industrial users were substituted at low amounts. The test work was divided into two sets of trials. The first set consisted of four pilot oven trials and the second set eight sole heated oven trials. Data from both series of trials were collected on the physical, chemical, and texture properties of the resultant coke using biomaterial substitution and were compared to an operational coal blend. The specific objectives of this study are as follows:

- Study the interaction of the biomaterial substitutions with coal during the coking process using pilot oven tests
- Analyze the physical and chemical properties of coal blends using biomaterial substitution
- Investigate the connection between coke strength after reaction and texture properties of coke after biomaterial additions to the coal blend

This thesis consists of six chapters. The first chapter introduces the key concepts and works the thesis will cover. The second chapter is the literature review, where the fundamentals of the blast furnace operation, metallurgical coke production requirements, and quality testing of coal and coke are covered. This section also gives details of coke texture analysis, the environmental considerations of cokemaking in Canada, and the ongoing industrial research of biomaterial use in iron & steelmaking for coal substitution. The gap of knowledge is presented in chapter three. The current limited knowledge of the impact of biomaterial on coke quality from a microscopic level is addressed. The research methodology of the thesis is reviewed in chapter four. The procedures, test types, and standards followed are reviewed. The fifth chapter presents the results and discussions of the test work conducted. This chapter is broken into two main sections. The first section presents the outcome of pilot oven trials and the second section includes results of SHO trials. The final chapter is chapter six, where a summary of the thesis conclusions is listed as well as the recommendations for further work are presented.

2. Literature Review

2.1. Blast Furnace Process and Fuels

Ironmaking is a centuries old process that has developed into a global steel industry producing one of the most highly used materials worldwide. There are two main options for steel production, specifically, the blast furnace and the electric arc furnace. The blast furnace remains the primary source for steel production as of 2015 with 70% of the steel produced globally (Geerdes, Chaigneau, & Kurunov, 2015). In 2019 there was 6.3 million tons of hot metal produced in Canada from five operating blast furnaces (Technology, 2020).

The blast furnace is a continuous process that relies on coke as its primary fuel source and reducing agent. Coke for blast furnaces is generated from the transformation of metallurgical coal to coke in coke ovens. The blast furnace process can be compared to a packed bed countercurrent reactor with inputs and outputs identified in Figure 2-1. Alternating layers of metallurgical coke and fluxed iron material are inputs to the top of the blast furnace, known as the burden layers. Heated wind with controlled oxygen and moisture levels along with injected fuels (natural gas, coal, or oil) are input through the tuyere level, travel upward through the burden bed, and blast furnace gas leaves the top of the furnace. The intended product of reduced liquid iron is output through tapholes along with the byproduct of liquid slag.

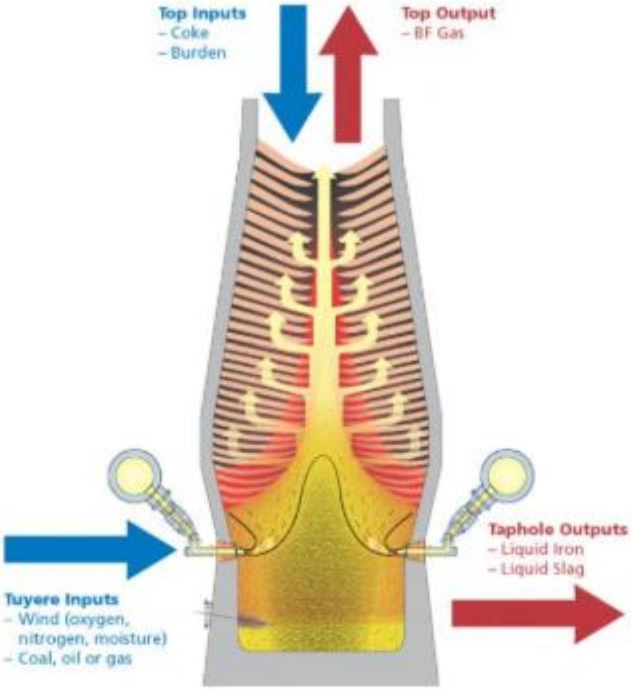


Figure 2-1- Input and outputs of a blast furnace (Geerdes, Chaigneau, & Kurunov, 2015)

Coke is essential for the operation of the blast furnace process as the thermal fuel source and reductant. Coke's chemical role is to participate in reduction reactions of iron ore. Coke's physical role is for burden support and permeability for gas movement in the blast furnace process. Understanding how coke passes through the blast furnace zones and the role it plays in each zone is important for coke selection. The blast furnace can be broken into five zones starting from the top moving down as temperatures increases. The top of the blast furnace is referred to as the Lumpy Zone, where raw materials are charged into the process, beings to move down the shaft to start the reduction process and are heated. The second zone is the Cohesive Zone; this zone is critical for the reduction of the iron ore material, and gas permeability of the coke burden layers must be maintained. The Raceway Zone is the third zone where tuyere level inputs are injected and react with the burden coke to combust producing carbon monoxide (CO) and hydrogen (H_2) gases. The fourth zone is known as the Active Coke Zone, where coke is still permeable to allow for gas flow up and liquid iron to pass down. The final zone is the Deadman or Hearth Zone. This zone is where the coke bed is found and is being pushed down on the liquid iron and slag is extracted from the blast furnace. (Lu, 2018)

2.2. Coke Requirements

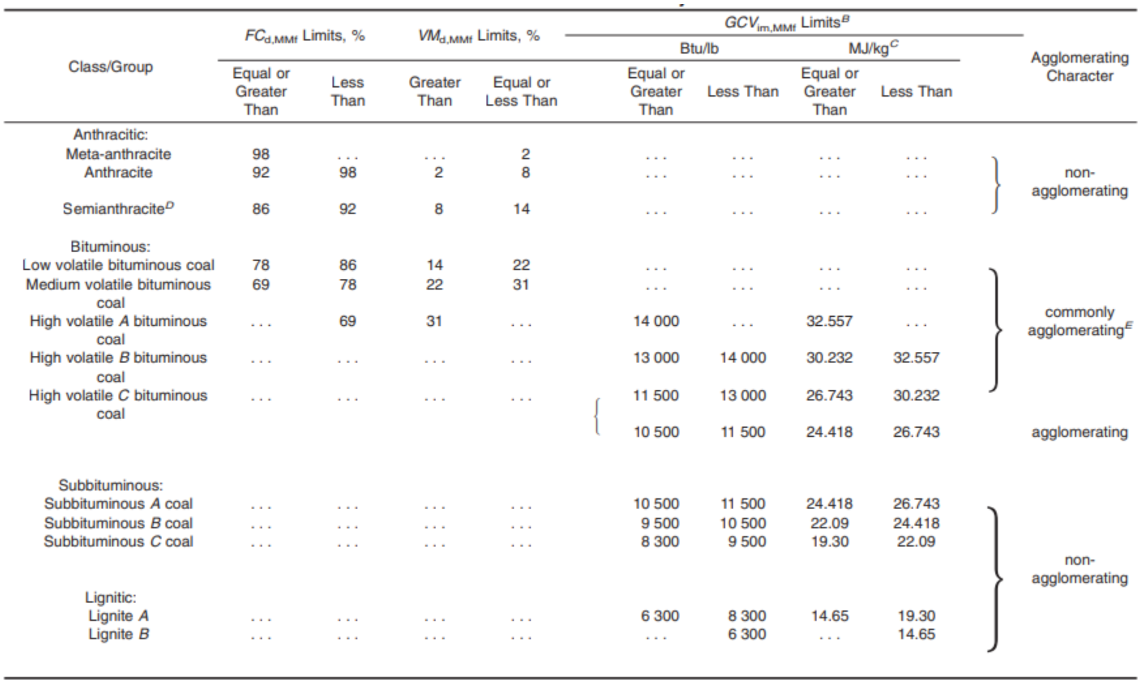
As the primary fuel source for the blast furnace, there are several attributes of coke that affect the blast furnace operating conditions. The most impactful attributes are strength, stability, size, moisture content, and chemical composition (Geerdes, Chaigneau, & Kurunov, 2015). In North America stability is measured by the cold strength index which measures the degree of breakdown expected when the coke is handled. The size of coke fed to a blast furnace is impactful to the operation because of the impact on gas permeability. Smaller coke, typically less than ½ inch, will increase the pressure drop of the furnace and reduce the permeability of gases. Coke moisture is continuously monitored for coke fed to the blast furnace to ensure a consistent dry weight of coke is charged. If coke with too high of moisture enters the blast furnace, this reduces the carbon which impacts the thermal balance. The chemical composition of coke is essential not only to the blast furnace but also to the downstream steelmaking operation because the goal is to have a high source of carbon with little impurities to fuel the iron reduction process and ensure a consistent iron chemistry. Supplying consistent coke to the blast furnace is extremely important. Coke is not only a fuel for the reduction of iron ore in the blast furnace process, but also the physical strength maintains the permeability for gas flow and supports of the burden as it descends the shaft. Quality coke that meets the need of the blast furnace process is determined by the coal quality, coal preparation, and coke oven operating parameters. (Poveromo, 2019) The applicable standards and their test methods will be reviewed in the following sections.

2.3. Metallurgical Coal and Coke

2.4. Coal Sources

Coal is a material found in underground deposits around the world that contains greater than 50% by weight carbon derived from plant material that is readably combustible. Coal is formed in underground deposits naturally from peat over time in swampy environmental conditions. Peat formation is the required start to coal transformation and is affected by climate, geology, chemistry, type of plants and conditions of the swamp to form different types of coals. Peat becomes coal once the peat has been buried, compressed and the moisture content is less than 75%. Peat accumulation and the process of coalification where the peat matures varies depending on the geographical location, but all require time, temperature and pressure. (Osborne, 2019)

Determining the maturity and classification of coal has been refined and in North America is most commonly defined by the American Society for Testing and Materials (ASTM) standards. As the coal change in the underground conditions, the properties determine where on the ASTM classification of coals they fall. The classification is based on rank, type and grade as seen in Figure 2-2 from ASTM Standard D388 – Standard Classification of Coals by Rank. The coals are broken into class based on the degree of metamorphism and then broken into group which are subdivisions of the class. The table displays identifying limits for attributes of Fixed Carbon (FC), Volatile Matter (VM), Gross Calorific Value (GCV) and the associated agglomerating character. The agglomerating character is used to describe how the coal will react, and the state of the resultant material when heated and the cooled per the standard's definition. These are the first properties of coal evaluated to determine if and how the coal is to be mined, processed and the end use.



Class/Group	$FC_{d,MMf}$ Limits, %		$VM_{d,MMf}$ Limits, %		$GCV_{im,MMf}$ Limits^B				Agglomerating Character
					Btu/lb		MJ/kg^C		
	Equal or Greater Than	Less Than	Greater Than	Equal or Less Than	Equal or Greater Than	Less Than	Equal or Greater Than	Less Than	
Anthracitic:									
Meta-anthracite	98	. . .	. . .	2	. . .	. . .	. . .	. . .	non-agglomerating
Anthracite	92	98	2	8	. . .	. . .	. . .	. . .	
Semianthracite^D	86	92	8	14	. . .	. . .	. . .	. . .	
Bituminous:									
Low volatile bituminous coal	78	86	14	22	. . .	. . .	. . .	. . .	commonly agglomerating^E
Medium volatile bituminous coal	69	78	22	31	. . .	. . .	. . .	. . .	
High volatile *A* bituminous coal	. . .	69	31	. . .	14 000	. . .	32.557	. . .	
High volatile *B* bituminous coal	. . .	. . .	. . .	. . .	13 000	14 000	30.232	32.557	
High volatile *C* bituminous coal	. . .	. . .	. . .	. . .	11 500	13 000	26.743	30.232	
					10 500	11 500	24.418	26.743	agglomerating
Subbituminous:									
Subbituminous *A* coal	. . .	. . .	. . .	. . .	10 500	11 500	24.418	26.743	non-agglomerating
Subbituminous *B* coal	. . .	. . .	. . .	. . .	9 500	10 500	22.09	24.418	
Subbituminous *C* coal	. . .	. . .	. . .	. . .	8 300	9 500	19.30	22.09	
Lignitic:									
Lignite *A*	. . .	. . .	. . .	. . .	6 300	8 300	14.65	19.30	
Lignite *B*	. . .	. . .	. . .	. . .	. . .	6 300	. . .	14.65	

Figure 2-2 - ASTM Standard D388 (ASTM, 2020)

Coal classification is broken into three categories of grade, rank, and type. The plant materials that accumulated in the swamps influence the type of coal that is formed and its properties. All three classifications of grade, rank and type affect the potential coke quality of a coal and coal blends.

2.4.1. Coal Grade

The grade of a coal is dependent on the purity of the materials with the plants that go through coalification. This is environmentally dependant on which minerals were deposited with the plant material during the first stage of coalification. (Price, 2017) The fewer inorganics such as ash, sulfur and phosphorus, the higher the coals grade. A high-grade coal would be less than 10% ash, and a low grade would be 30-50% ash content. (Osborne, 2019)

2.4.2. Coal Rank

The rank of a coal is the degree of coalification the coal has reached. Plant structures of roots, stems, leaves, flowers and their tissues all have varying physical and chemical components that will decompose differently. The decomposition and fossilization of these materials is the organic material found in coal. The rank of a coal is determined chemically or petrographically and identifies the potential use of the coal. ASTM standard D388 Standard Classification of Coals by Rank identifies shown in Figure 2-2 - ASTM Standard D388 (ASTM, 2020)breaks coal rank into four categories: lignites, subbituminous, bituminous and anthracite. Lignites are the lowest ranking and cannot be used in metallurgical coal processes as they have a low calorific value and do not agglutinate. Similarly, high ranking anthracite coals cannot be used due to their lack of adhesion properties needed in metallurgical coal. Bituminous coal is the focus for coking coals because of their adhesion properties (Price, 2017) Ranking of a coal from low to high designate the typical use. A low ranked coal (lignite and sub-bituminous) is used for electricity generation, high ranked (bituminous and anthracite) are used for thermal purposes, while premium (bituminous and anthracite) are used for metallurgical coking coals. Figure 2-3 relates the rank of coal from low to premium. (Osborne, 2019)

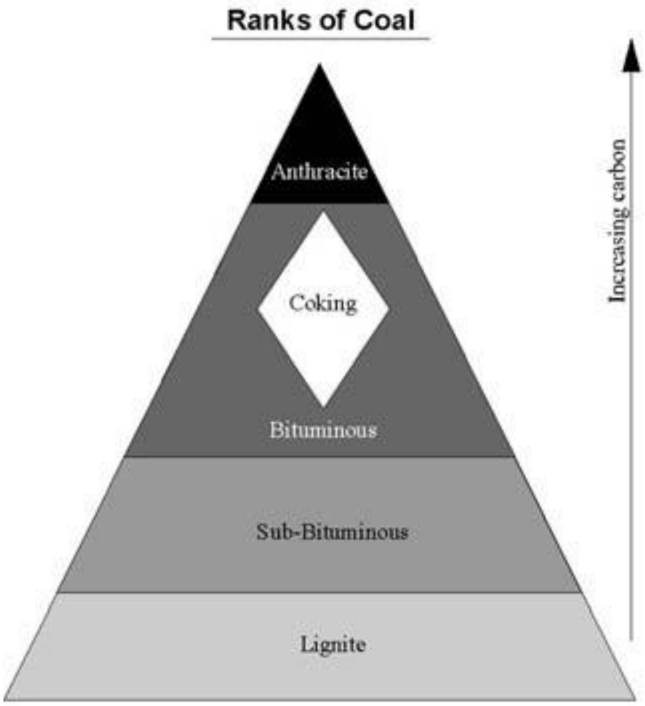


Figure 2-3- Coal Rank (IEA, 2020)

2.4.3. Coal Type

Coal type is the third property used to define coal properties. The type of coal is identified by the plant material residues when reviewed through petrographic microscopes. (Osborne, 2019) Type allows coal particles to be categorized to understand the origin, appearance, chemical and physical properties. The different macerals that are found in coal are how coal type is assigned. The three main groups of macerals are: liptinite, vitrinite, and inertinites. (Price, 2017) The properties of the different maceral types will be discussed further in the petrographic analysis section to follow.

2.5. Coal to Coke Process

Cokemaking is an ancient process that can be traced back to 371 BC. Like any process of this age, there have been many changes to the art of cokemaking with the industrial revolution and technological advancements. In North America, slot style byproduct recovery batteries, as seen in Figure 2-4, and heat-recovery batteries, as seen in Figure 2-5, are the two main processes used for cokemaking (Kobus, 2017). Currently, byproduct recovery batteries are more common as many were commissioned in the world war eras, and the byproduct of toluene for TNT production was highly sought after. On the other hand, heat-recovery batteries are a newer development and have the ability to capture excess heat for steam or electrical power generation.

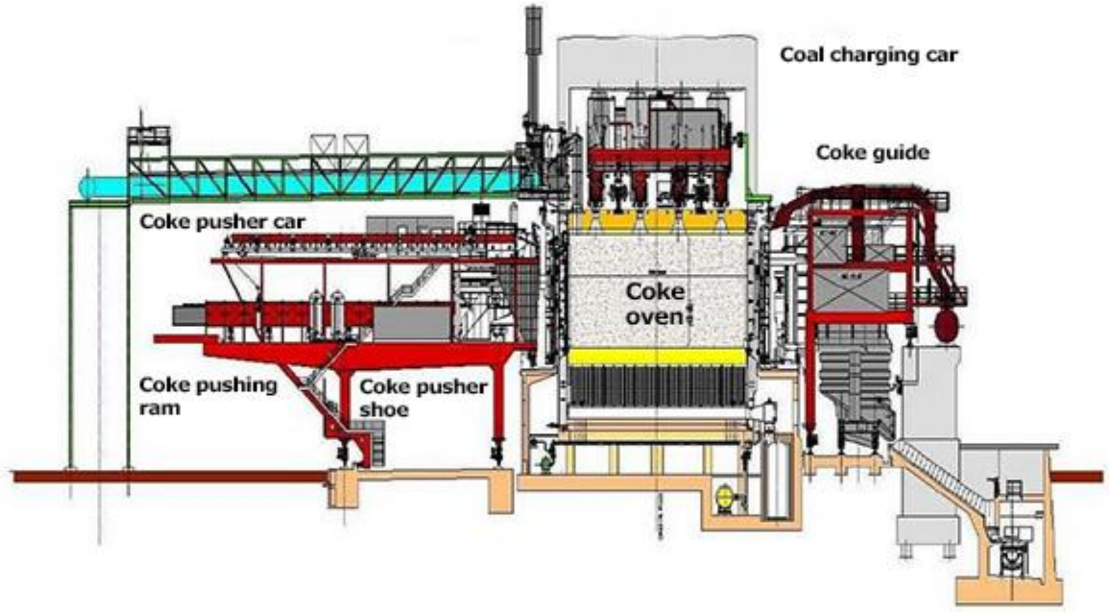


Figure 2-4 - Coke Oven Design for By Product Design (Anyang , 2021)

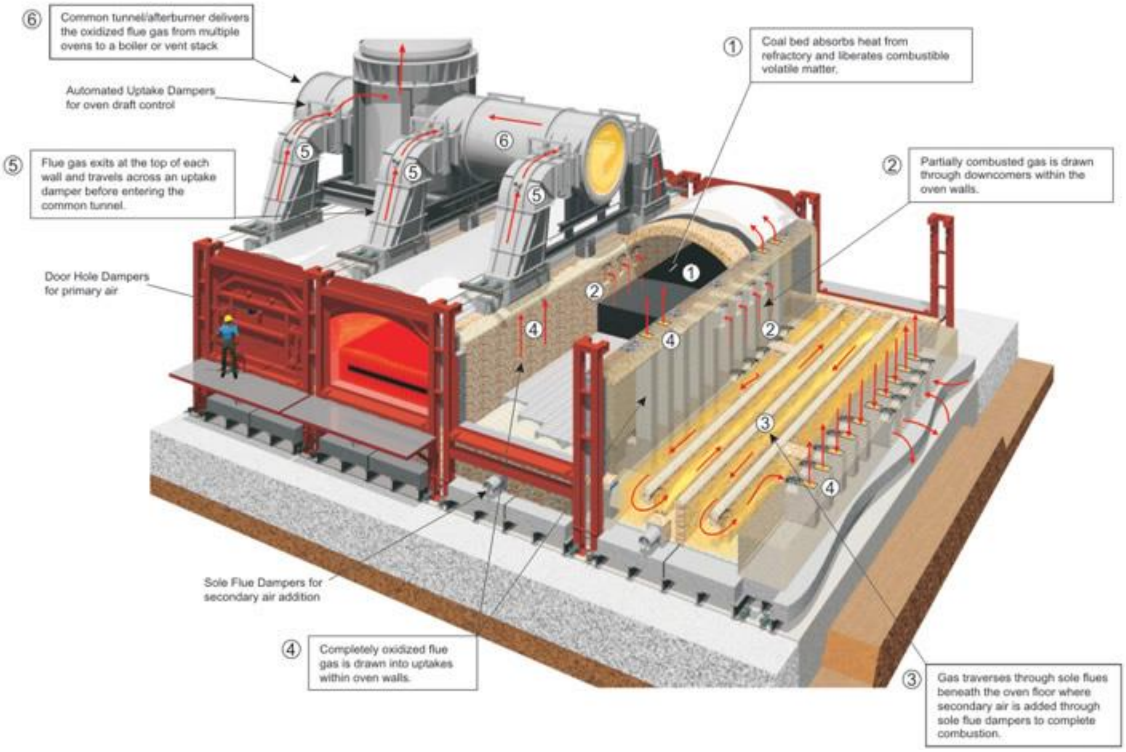


Figure 2-5- Heat Recovery Battery Design (SunCoke , 2021)

In either battery type, the cokemaking process begins once the coal blend is charged into the oxygen deficient hot battery. Batteries are lined with refractory and continuously heated to provide the desired coking environment. In slot style batteries the cokemaking process usually takes 16-24 hours at 1000 - 1100°C depending on the height, width and heating profile of the battery. The cokemaking process does not have a consistent profile throughout the oven which is often referred to as a zipper effect. The oven bottom coke transforms first and then from the walls into the middle as the transformation progresses with completion of the oven top middle completing coking last. The variation and nonuniformity of the phase transformation from coal to coke are primarily due to the coal bulk density and charging nature. Coal charged at the top of the oven is less dense than the bottom of the oven due to the concentration and size of material when charged. (Todoschuk T. W., 2017)

As seen in Figure 2-6, coal must progress through several key transformation stages to reach the coke stage. There are three stages to the swelling portion of the coking process. First, the coal is charged into the oven and begins to heat, the coal's moisture is first driven off. As the coal continues to heat, it progresses into the plastic phase, where the coal softens allowing tars and oils to be driven off from the bubbling action that occurs. The plastic phase is the most impactful to coke quality for pore structure formation, optical characteristics, surface area, and strength development possible from the coking conditions. The third stage is the progression to semi-coke where gases are driven off as temperature and pressure increase. The process begins to move into the shrinking portion of the coke transformation, and the bubbles collapse. The final stage in the transformation is for the hydrogen to be driven off, causing shrinkage, leaving a high carbon material referred to as coke. During the shrinking phase, coal particles are fusing together, and fissures are formed, which affects the tensile strength of the final coke product. The tapering off where no further heating is observed is the soak time. This time is needed to ensure the coke has been adequately heated all the way through. (Todoschuk T. W., 2017)

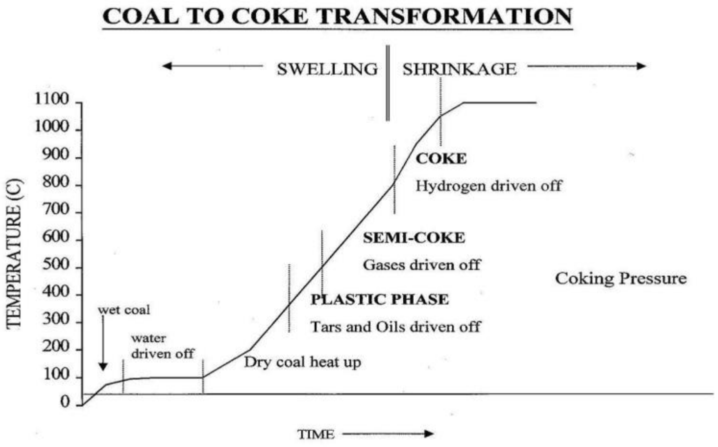


Figure 2-6 - Coal to Coke Transformation Diagram (Todoschuk T. W., 2017)

2.6. Coal Quality

In early cokemaking processes, a single bituminous coal could be used, but this is no longer possible with the properties of limited coal resources available today. Thus, a blend needs to be used. To prepare a coal blend for the metallurgical cokemaking process, the properties and characteristics predicted for cokemaking need to be evaluated. The objective of creating a coal blend is to reduce cost, meet production performance targets, meet chemical and physical requirements, meet blast furnace coke requirements, and to analyze the economics of blend design. Each producer relies on internal blend models, coal specifications from suppliers, pilot oven test results, and coal blend selection analysis.

2.6.1. Coal Petrography

A petrographic analysis is the most commonly relied upon tool for analyzing coal properties and predicting coke strength. There are several standards worldwide, with the ASTM standards being used in North America. All examination relies on three steps for petrographic analysis of maceral identification, grouping of macerals into categories, and determining the rank of the coal. These three steps allow for a predicted coke strength to be calculated and is a crucial component to determining a blend that should proceed to testing. Petrographic analysis is used to monitor coal quality and variation in blends when in operational use. (Valia, 2019)

To complete a petrographic analysis the fundamentals of maceral identification and components of a coal must be understood. From the organic plant remains that are fossilized to form coal the final remains are known as macerals. Petrography is the act of identifying, examining and classifying the units of macerals under a microscope to determine the type of coal that has formed (Osborne, 2019). This is a visual observation of bright, dull and banded layers and mineral matters of the coal. There are three categories of macerals, each with subcategories, used for classification which are the vitrinite macerals, the inertinite macerals and the liptinite macerals. (Osborne, 2019)

Vitrinite maceral group is the most abundant component in coal. Vitrinite contributes the most to coke quality and is used to determine critical properties in coal. This group is used to find the reflectance of the coal which is the measure of the reflected light from the vitrinite maceral viewed under a microscope. The reflectance of the coal is used to categorize the coal into v-types which are then used to determine the coal rank. (Price, 2017) Under a microscope vitrinite is grey in color, as seen in Figure 2-7 below. (Osborne, 2019)

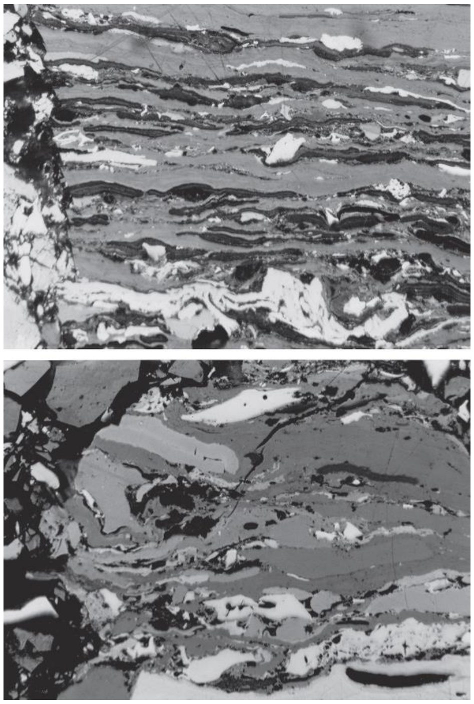


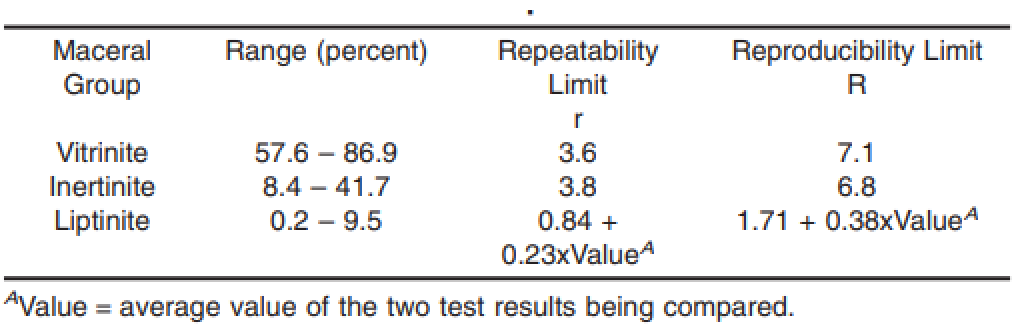
Figure 2-7- Vitrinite (grey layers) and fragments of inertinite (white) are emended. Scale 200 µm left to right. (Osborne, 2019)

Inertinite macerals are white in color when viewed under the microscope, as seen in Figure 2-7 above. Inertinite macerals formed from fossil wood that had been burnt in ancient fires or exposed to aerobic bacterial degradation during decomposition. (Osborne, 2019) The four types of inert macerals are micrinite, macrinite, fusinite and semifusinite. Micrinite and macrinite are very similar but defined by size, with micrinite being <5 μm in size and macrinite >10 μm in size. Fusinite is similar to vitrinite but has been oxidized during coalification. Semifusinite macerals are between vitrinites and fusinites in properties as there are both reactive and inert semifusinite. All inertinite macerals are inert materials but act as fillers and binders for the reactive macerals during coking transitions. Inert macerals are needed in coal blend properties as they increase coke contraction, reduce fissurization, and reduce coking pressure, which are all critical for operational tolerances of a coal blend in a battery. (Price, 2017)

The final group is liptinite macerals, which were often referred to as exinite in the past. This group of macerals is derived from specialized hydrogen, waxy, and oil-rich plant materials and has a low oxygen content (Osborne, 2019). When viewed under the microscope, they are seen as dark material and have the lowest reflectance values of the maceral groups. These macerals are generally low in density and are the most fluid maceral group during carbonization (Loison, Foch, & Boyer, 1989). Additionally, they have a high volatile content, which contributes to more byproducts than coke formation. (Osborne, 2019)

Mineral matter is also reviewed in coal petrographic analysis. Coking coals are washed to remove mineral matter before supplied to the customer, but there is residual mineral matter expected. Minerals include those deposited with the plant matter as well as those that have formed during the peat transformation process. However, the mineral matter may be classified as included or excluded. Included mineral matter is bound into the organic cellular structure of coal whereas, excluded are mineral matter excluded from the coal structure or coal seam. The most common types of mineral matter found in coal are: clay, quartz, Pyrite, and carbonate minerals. Identifying and understanding the different mineral matter found in coal is important as they can affect equipment wear, gases emitted when heated, ash fusion temperature and can reduce coking capacity. (Osborne, 2019)

In North America the procedure for petrographic analysis follows ASTM standard D2799 - Microscopical Determination of the Maceral Composition of Coal, which is like the ISO standard 7404-3 used in other labs. This procedure uses any microscope with a mechanical stage and a vertical illuminator capable of 1 to 2 µm resolution of the object. The eyepiece should have a crosshair for references, and the stage should be able to move in measured increments. A pellet of coal is prepared according to ASTM D2797 to be analysed under the microscope for reflectance, other optical properties, and morphology measurements. The microscope operator places the prepared sample under the microscope and uses the crosshair points to quantify the maceral groups, minerals and other observations optically noted on a counter tool. The pellet is moved in increments of 0.5 mm until the desired length of the specimen in that direction has been covered before stepping down and repeating in the opposite direction. This continues until 1000 counts have been recorded. As a measurement tool, there are limitations to petrographic analysis as it is qualitative, and operator based with the repeatability and reproducibility of manual counts of macerals seen in Figure 2-8. (ASTM, 2020)



Maceral Group	Range (percent)	Repeatability Limit r	Reproducibility Limit R
Vitrinite	57.6 – 86.9	3.6	7.1
Inertinite	8.4 – 41.7	3.8	6.8
Liptinite	0.2 – 9.5	0.84 + 0.23xValue[A]	1.71 + 0.38xValue[A]

[A]Value = average value of the two test results being compared.

Figure 2-8-Limits of Repeatability and Reproducibility for Maceral Groups (ASTM, 2020)

2.6.2. Coal Reflectance

Metallurgical coals are composed mainly of vitrinite and ranked accordingly. ASTM Standard D388 Standard Classification of Coals by Rank uses the rank of coal to correlate the maximum reflectance of coals by vitrinite groups. Following ASTM Standard D2798 Standard Test Method for Microscopical Determination of the Vitrinite Reflectance of Coal over 800 U.S coals were plotted to show the relationship between mean max reflectance and all varieties of vitrinite in volatile matter seen in Figure 2-9. This allowed for the understanding that low volatile coals are typically greater than 1.4 reflectance, medium volatile coals are found between 1.1 to 1.4 mean max reflectance and high volatile coals are less than 1.1. A blend of coals would fall within these limits depending on the composition. (ASTM, 2020)

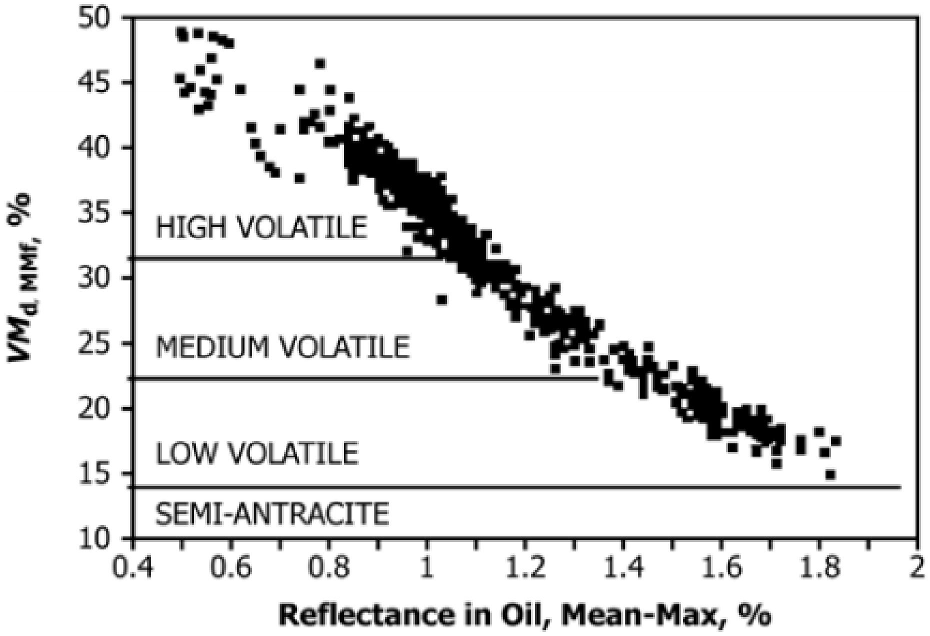


Figure 2-9 - Relation Between Rank of U.S. Coals and Vitrinite Reflectance (ASTM, 2020)

2.6.3. Coal Proximate

ASTM standard D7582 Standard Test Methods for Proximate Analysis of Coal and Coke by Macro Thermogravimetric Analysis. This standard is used for determining the moisture, ash, volatile matter, and fixed carbon of a coal. Each property has its own test method but are collectively used for characterization, which is important for knowing the use of the coal. Moisture, ash and volatile matter are determined by tests while the fixed carbon is a calculation. (Speight, 2015) Moisture is determined by measuring the loss in mass of the coal sample after heating under controlled conditions. Volatile Matter is also measured through heating a sample of coal in controlled conditions and measures the loss of material after correcting for the moisture loss. Ash measures the amount of residue remaining after burning the coal sample. Fixed carbon is the last component determined in the proximate analysis. It is calculated by taking the difference of 100 and the sum of moisture, volatile matter and ash. (ASTM, 2020)

2.6.4. Coal Ash Analysis

From the proximate analysis following ASTM standard D7582, the ash composition and percent present in the coal are determined. The ash determination is the most fundamental of quality parameters from the proximate analysis. The ash is a critical component to monitor for coal quality as it replaces carbon in coke formation. The amount of ash present in a coal is crucial for determining the coal's end-use for cokemaking. Ash remains in the coke and becomes incorporated in blast furnace slag, which has limitations. (Poveromo, 2019)

2.6.5. Coal Ultimate

ASTM standard D3176 is the Standard Practice for Ultimate Analysis of Coal and Coke. This analysis is for determining the elemental composition of coal and coke, including weight percent carbon, hydrogen, sulfur, nitrogen, and oxygen. Carbon and hydrogen are important for coal compositional review as they make up 70-90% of the weight of coal that combine to make up complex organic compounds of coal. (Speight, 2015)

The sulfur content is critical to determine as it contributes to coal grade. The average percent of sulfur in coal is 1%, and after losing some sulfur in the coking process, coke has approximately 0.8% sulfur (Price, 2017). Coal ash and sulfur are closely related to coke ash and sulfur. During the coking process, almost all coal ash becomes coke ash, and most of the coal sulfur (≈80%) becomes coke sulfur. Therefore, monitoring and controlling the ash and sulfur in coal is crucial. (Osborne, 2019) Decreasing sulfur by

0.1% in coke increases the blast furnace productivity by 0.8% and reduces coke consumption by 1% (Price, 2017)

While there is very little information on nitrogen compounds in coals, it does seem to only come from organic components. The nitrogen content is used for comparison of coals and for determining the oxygen content. The nitrogen content can also be used for the potential prediction of nitrogen oxides that could be emitted from the coal being evaluated. (Speight, 2015)

Oxygen in coal comes from both organic and inorganic components. The portion from organic to inorganic changes with the rank of coal with the higher rank coals having less organic origin. There is no direct test method for determining oxygen content in coal. The common practice is to subtract the percent of carbon, hydrogen, nitrogen and organic sulfur from 100 as seen in Equation 1 below. (Speight, 2015) The oxygen is measured from the volatile components and excludes ash.

Equation 1 - Oxygen Content of Coal Calculation

$$\%\,Oxygen = 100 - (\%C + \%H + \%N + \%S_{organic})$$

Oxygen levels, oxidization and age of coal are all monitored at a regular interval at operating facilities to monitor for impacts to coal quality. When coal is mined, it is exposed to oxygen and begins to react. The reaction of oxygen and carbon of coal is not desired as it lowers the potential for the coals to swell and soften and impacts the coking capabilities. Exposure to oxygen is unavoidable once mined, but the extent of oxidization can be limited by reducing the time between mining and charging into a coke oven (Todoschuk & Giroux, 2019). Time is reduced by coordinating the time of mining, transportation, delivery, storage, and use for each operational facility.

2.6.6. Coal Sieve Analysis

Coal size can impact the interactions during coking and is monitored for impact on coke quality. Coal is transported and stored at different points before being processed through a coal handling system for charging into a coke oven. The coals can be crushed to the final desired size before or after blending. This can depend on the individual properties as well as transportation logistics. Coal is charged into batteries at a target of 80% minus 3.2 mm (1/8 inch) in size. ASTM standard D4749 Standard Test Method for Performing the Sieve Analysis of Coal and Designating Coal Size is used for raw and prepared coal analysis and does not give specification to set coal size for operations. To conduct the test, a series of square mesh sieves are stacked by standard size number as noted in Table 2-1 below. The coal to be measured is weighed before being passed through the standard set of sieves. The passing of the coal can be done manually or mechanically, depending on the equipment available. There are specifics to the procedure depending on the method being used. The resulting coal in each sieve is weighed and a percent assigned to determine size distribution.

Table 2-1 - Wire Cloth (U.S.A. Standard) Sieves with Square Openings for Coal used in Coke Ovens

50.0 mm [2 in.]
37.5 mm [11⁄2 in.]
25.0 mm [1 in.]
19.0 mm [3⁄4 in.]
12.5 mm [1⁄2 in.]
9.5 mm [3⁄8 in.]
6.3 mm [1⁄4 in.]

2.6.7. Gieseler Fluidity

The plastic properties of a coal blend are known to be one of the most impactful factors on coke strength control. The Gieseler Plastometer is used for fluidity determination as a measure of the coal's ability to form a plastic phase and make quality coke. (Lin & Hong, 1986) Fluidity affects the Coke Strength after Reaction (CSR) which is an important parameter of coke quality. A lower CSR was linked to lower fluidity as a result of fewer inerts from high fluid coals and not enough binders from low fluid coals as seen in Figure 2-10- Effect of Fluidity on CSR (Kumar, Barman, Singh, & Ranjan, 2008) Figure 2-10 below. The maximum fluidity of a coal or coal blend is an indication of the bonding process during coke making and the Gieseler Plastometer is a practical test for comparing this critical rheological property. (Kumar, Barman, Singh, & Ranjan, 2008)

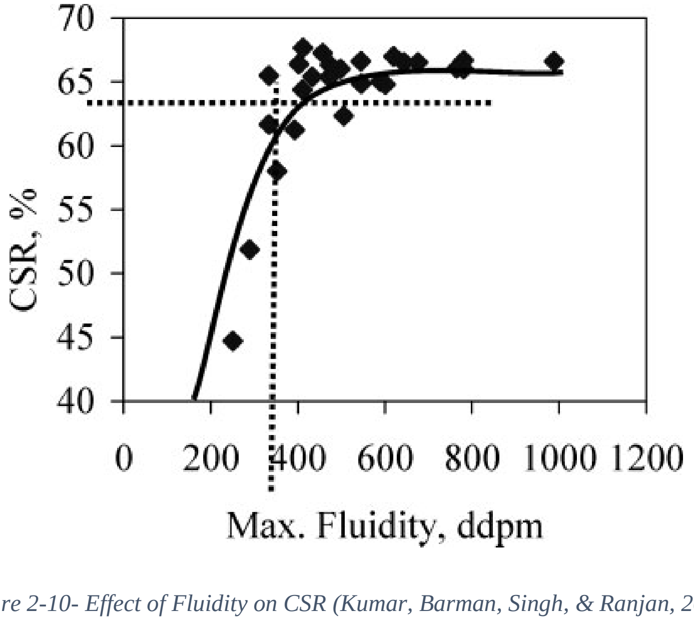


Figure 2-10- Effect of Fluidity on CSR (Kumar, Barman, Singh, & Ranjan, 2008)

The fluidity reading follows ASTM Standard D2639 Standard Test Method for Plastic Properties of Coal by the Constant-Torque Gieseler Plastometer where the coal sample is heated at a constant rate and monitors the softening and melting temperatures of the coal.

When the sample reaches the maximum fluidity and starts to soften then solidify is the final measurements of the test. (ASTM, 2020)

Fluidity readings should not be the only factor considered when evaluating the age of the coal and the potential coking properties. The fluidity reading can be impacted by many factors during testing including sample size, petrographic composition of the sample and errors with the test equipment. The temperature range of the difference between the coal's initial softening and resolidification is measured during the test. In 2019 a study on US Appalachian coals showed that there is variability in the fluidity results and that the temperature range results of fluidity and dilatation are a more consistent indicator on coal's aging and should be evaluated in conjunction with the fluidity and dilatation testing (Todoschuk & Giroux, 2019). This is important for Canadian coke makers as aging of coal is monitored due to the restriction of coal delivery to warmer seasons through vessel delivery on the Great Lakes.

2.6.8. Dilatation

The second measurement tool for coal rheological properties is dilatation. Dilatometry of a coal and coal blend provides information on the expansion and contraction (Price, 2017). Like the fluidity reading, the dilatation is an indicator of the coal's plastic properties and its potential coking ability, which is essential information when creating coal blends for evaluation. It should be noted that both parameters are functions of the coal size, petrographic properties, and the bulk density of the samples. (Todoschuk & Giroux, 2019) This measurement procedure follows the ASTM Standard D5515 Standard Test Method for Determination of the Swelling Properties of Bituminous Coal Using a Dilatometer. A small sample of the coal that has been ground to pass a 250μm is loaded into the dilatometer apparatus, Figure 2-11, and heated in a controlled manner. The piston height changes for the percent contraction and dilatation values along with the softening temperature, maximum contrition temperature, and maximum dilatation temperature, are recorded before the test concludes. From the values, collected the percent of dilatation is determined and reported.

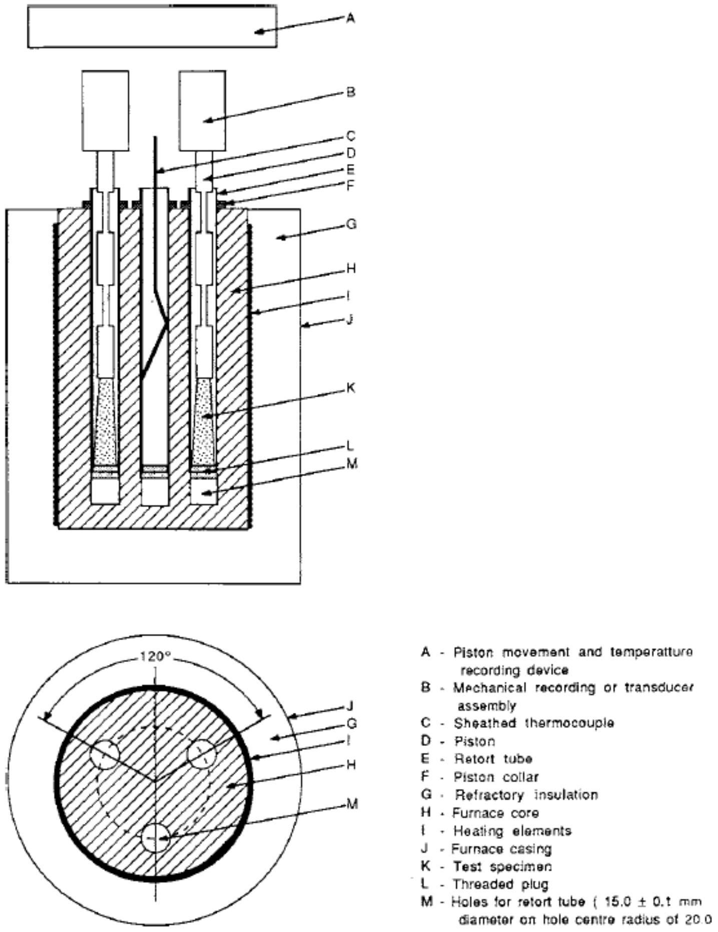
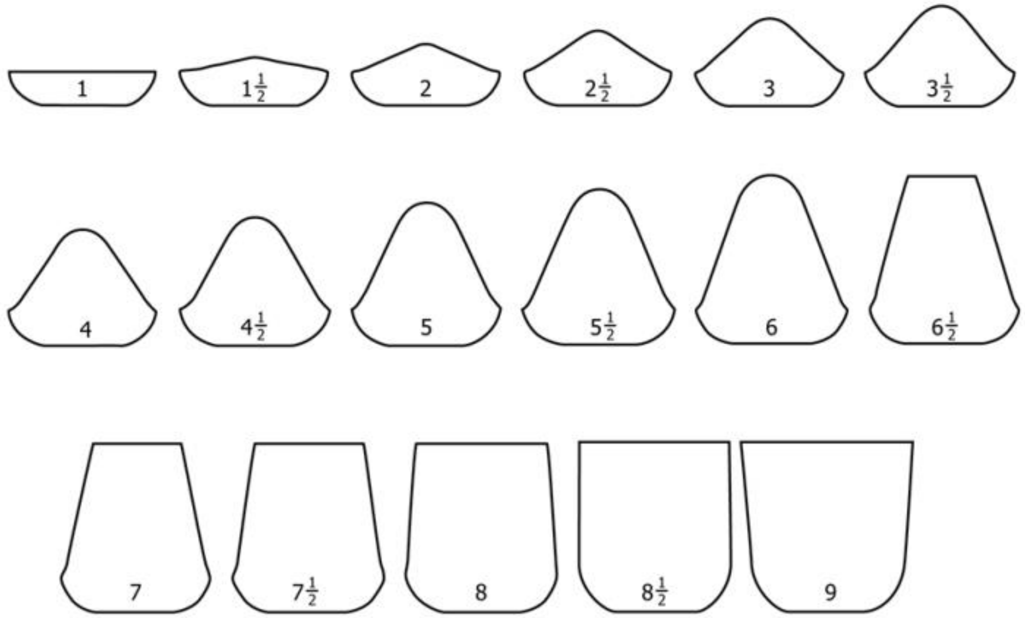


Figure 2-11 - Typical Dilatometer Apparatus (ASTM, 2020)

2.6.9. Free Swelling Index

The Free Swelling Index (FSI) of coal is determined by following ASTM D 720 Standard Test Method for Free-Swelling Index of Coal. This is a lab scale test used to determine the caking properties of a coal, not the expansion properties. (ASTM, 2020) This test takes a small crucible coal sample of 1 gram and places it into a controlled heated furnace for 2 ½ minutes. The profile of the coke button is formed from the heating process where the coal swells from the volatile matter, and when it solidifies, the coke mass is lighter and larger. (Loison, Foch, & Boyer, 1989) The resultant coke button is compared to the standard profiles index, as seen in Figure 2-12. The profile that best matches the coke button is the assigned FSI value. An FSI value of over 4 is considered a good caking coal. An FSI of 6.5 to 9 is considered a high-quality coking coal (Price, 2017).



Courtesy of British Standards Institution

Figure 2-12 - Standard Profiles for FSI Index (ASTM, 2020)

2.6.10. Sole Heated Oven

The Sole Heated Oven (SHO) is a lab scale oven that uses small amounts of coal or coal blends for expansion and contraction evaluation. These tests are performed in duplication, and the amount of contraction/expansion is reported. The coke produced from SHO testing can be used for Coke Strength after Reaction (CSR) and Coke Reactivity Index (CRI) and coke texture analysis. Industrially the aim expansion/contraction range targeted for a blend is from -8 to -12. This value is not only impacted by the coal properties. The expansion/contraction can be impacted by operational set points, which result in individual producers requiring different target ranges for the SHO limits. ASTM standard D2014 for Standard Test Method for Expansion or Contraction of Coal by the Sole-Heated Oven is used for both single, as seen in Figure 2-13, and double chamber ovens. The advantages of a SHO are that only a small 30 kg (66 lbs.) sample of the coal blend being reviewed of is needed, and coking times are much shorter than those of the pilot oven test discussed later in this document. The coking time is determined by the time to reach 500°C at the thermocouple on the top surface of the SHO. The coal properties of bulk density, moisture and mass along with the final and initial thicknesses of the coal charged are all measured in reference to the standard oven operating parameters to determine the expansion/contraction value. If a double chamber oven is being used, the average of the two is used for reporting purposes. (ASTM, 2020)

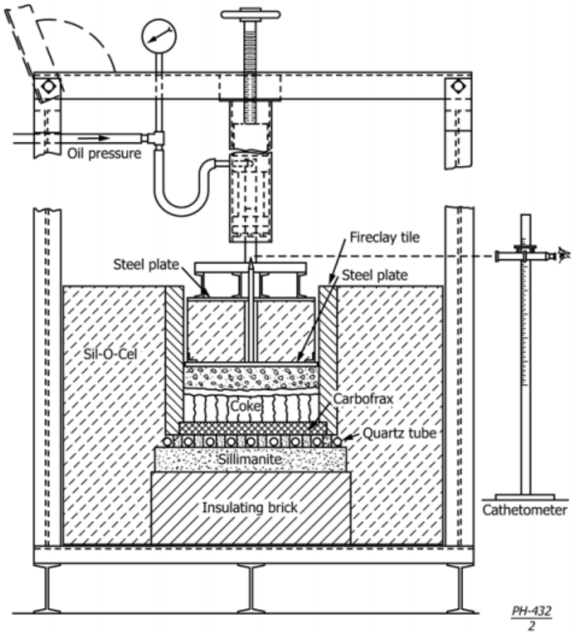


Figure 2-13- Single Chamber Sole Heated Oven Design (ASTM, 2020)

2.7. Coke Quality

The ultimate goal of coal evaluation, blending, and selection is for the coal to be used in coke making operations to produce metallurgical coke for use in the blast furnace process. Coal blends that are selected for cokemaking should be tested before implementation as their properties are not additive and testing of the coke quality at a smaller scale is used to avoid unacceptable blends. There are standards and procedures followed for coke evaluation of pilot oven testing as well as operational coke production. In North America, ASTM standards are the most commonly followed compared to ISO standards.

2.7.1. Pilot Oven Testing

General industry practice is to conduct pilot oven testing of coal blends prior to implementation in an operating battery. This allows for suppliers to evaluate their products as well as producers to assess coal blends before costly full-scale implementations. Pilot oven dimensions vary depending on the laboratory and type of oven. The width of the ovens varies from 310 to 600 mm (12.2 to 23.6 inches) and in height 1 to 1.5 meters (3.2 to 4.9 feet). Most laboratories use Movable Wall Ovens (MWO) for pilot testing as they allow for the expansion and contraction of the coal blends being tested without damage to the oven. MWO are used in North America for their ability to simulate coke battery conditions (Price, 2017). The coke produced from the pilot ovens is used for a chemical and physical evaluation. These tests include coke proximate analysis, coke sizing, coke stability, coke hardness, Apparent Specific Gravity (ASG), Total Specific Gravity (TSG), Coke Strength after Reaction (CSR), and Coke Reactivity Index (CRI). The operational set points of the pilot oven tests are set to best predict the cokemaking conditions of a full-scale battery. The coal charged, bulk density of the coal, heating conditions and the mechanical and thermal stresses the coke is subjected to are all controlled for this reason. (Loison, Foch, & Boyer, 1989)

2.7.2. Coke Strength after Reaction

Coke Strength after Reaction (CSR) is the measurement of hot coke strength, which is an important attribute of how the coke will react in the high temperatures of the blast furnace process. Coal rank is one of the most important factors contributing to coke strength. When a coal blend is being selected, the rank is optimized for CSR impact in reactive and inert components. Inert macerals aid in binding of vitrinite macerals for increased coke strength and CSR. (Price, 2017) Other factors of coal properties that impact the CSR are bulk density, reflectance, ash chemistry, and cokemaking process conditions. The thermal coke strength of coke increases with reflectance. Ash chemistry significantly affects the coke's CSR. The modified basicity index (MBI) measures the amount of basic oxide elements in the coke (Price, 2017). The equation for MBI calculation is from the ash chemistry, volatile matter, and ash present in the coke:

Equation 2 - MBI Calculation

$$MBI = 100 * \%ash * \frac{Na_2O + K_2O + CaO + MgO + Fe_2O_3}{(100 - VM) * (SiO_2 + Al_2O_3)}$$

MBI is inversely proportional to CSR.   Lower MBI values are favorable for CSR, which is shown in Figure 2-14 below:

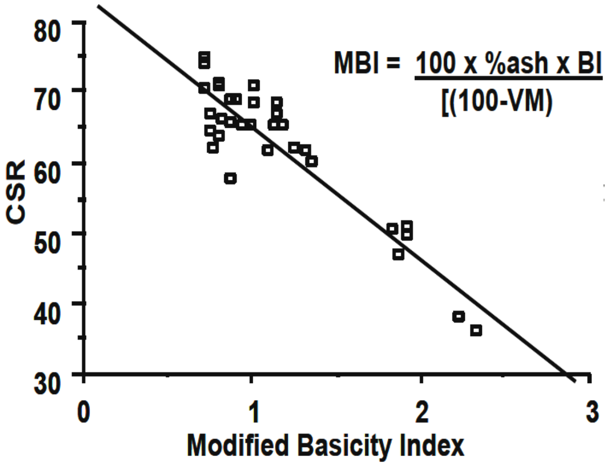


Figure 2-14- MBI to CSR Relationship (Price, 2017)

ASTM standard D5341 Standard Test Method for Measuring Coke Reactivity Index (CRI) and Coke Strength After Reaction (CSR) is followed to evaluate the physical coke; however, there are many predictive CSR calculations used within the industry from coal properties. For the physical test, prepared and weighed dried coke sample is reacted with carbon dioxide gas in a high temperature-controlled retort chamber. The cooled residual coke residue is weighed, sieved for size, and then tumbled at a consistent turning rate and weighed to determine the CSR. Coke for this test can be from a SHO, pilot oven, or operational product. (ASTM, 2020)

2.7.3. Coke Reactivity Index

Coke Reactivity Index (CRI) is inversely related to CSR and used for the control of CSR. CRI is also impacted by many different aspects of coal properties, including ash chemistry, inert content and cokemaking process conditions. The results from the CSR and CRI allow for an understanding of the physical stamina and reactivity properties in the blast furnace operations. (Loison, Foch, & Boyer, 1989)

ASTM standard D5341 Standard Test Method for Measuring Coke Reactivity Index (CRI) and Coke Strength After Reaction (CSR) is followed to evaluate the CRI. The same sample that was prepared and reacted with carbon dioxide gas in a high temperature-controlled retort chamber for CSR is used. The cooled residual coke residue is measured for weight loss to determine the CRI. Both CSR and CRI testing are done in duplicate, and the average value is reported. The design of the apparatus used for CSR and CRI testing can be seen in Figure 2-15 below. The prepared sample is placed inside the retort furnace with a controlled inlet of nitrogen and carbon dioxide gas fed into the bottom of the sample chamber. The design allows for a uniform high temperature of 1100°C surrounding the sample chamber. A thermocouple is suspended from the top of the chamber to allow it to be positioned in the coke bed. (ASTM, 2020)

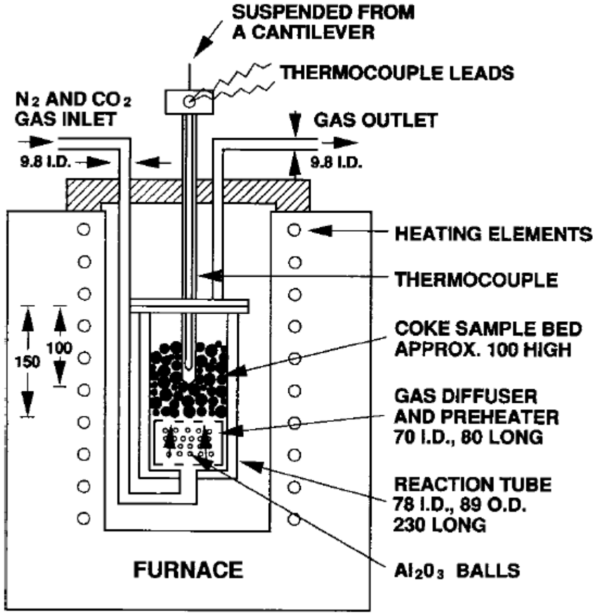


Figure 2-15 - Apparatus design for CSR and CRI testing (ASTM, 2020)

2.7.4. Stability

Stability is the cold strength index used in North America for the degree of breakdown expected when the coke is handled. Stability is affected by the coking process and coal quality. (Price, 2017) The coking process conditions are unique to each battery, with the temperature control and timing of when coal contraction impacting the stability. If the coal transitions and contraction occur too fast, the rapid contraction causes weakness, and the coke will break. Control of the heating allows time for the coal to soak and contract and reduce the possibility of the finished coke being broken. The coal density and moisture charged into the oven also affect stability because these properties influence the heating and contraction rate and temperatures in the oven. (Todoschuk T. W., 2019) ASTM Standard D3402 Standard Test Method for Tumbler Test for Coke is followed to determine the stability and hardness factor of coke. Stability is often measured daily in manufacturing facilities as poor stability will impact the amount of coke required in the blast furnace, influencing furnace productivity and process stability.

2.7.5. Fissure Formation

When coke is formed in a battery, fissures naturally form from the transformation stages when material shrinks from volatile matter leaving the coal. Fissures that are perpendicular to the oven walls are most common. The fissures can fully traverse through a coke piece but can also stop inside of a coke piece. The fissures formed impact coke size and strength. (Loison, Foch, & Boyer, 1989) A visible fissure in a fully coked piece can be seen in Figure 2-16 after 20 hours of coking time. Research work in pilot oven testing has shown that fissure formation begins early in the transformation stages and spreads from the oven wall to the oven centerline as the plastic phase grows. Large coke pieces are required to feed the blast furnace to produce the bed permeability and strength needed, which is why fissure free coke is desired. (Jenkins, Mahoney, & Keating, 2010b)

Figure 2-16 - Visible fissure in the fully coked piece (Jenkins, Mahoney, & Keating, 2010b)

Cokemaking operating parameters are one of the largest influencers to coke fissure development. The heating profile of an oven with a high heating rate will promote fissures and smaller coke pieces. The size of the coke is not only impacted by the heating rate and this is just one factor that must be kept in mind when evaluating coke produced. (Jenkins, Mahoney, & Keating, 2010a)

2.7.6. Apparent Specific Gravity

Apparent Specific Gravity (ASG) is a measurement influenced by the rank of coal, the ash content and the operating conditions of a battery. The charge bulk density, heating rate, and pulverization level will impact the performance of the blast furnace. ASTM D167 Standard Test Method for Apparent and True Specific Gravity and Porosity of Lump Coke is followed for determining the ASG of coke. Coke can be from the SHO, Pilot Oven or coke battery for testing. The sample coke is first dried, cooled to room temperature and weighed. The sample is placed in a sample cage and lowered into a known volume vessel of water. The caged coke is left in the water for 15 minutes before the coke is removed. The weight of the displaced water and mass of the wet coke are taken to determine the apparent specific gravity from Equation 3. (ASTM, 2020) High amounts of inerts in a coal or coke blend will increase the ASG. An increased ASG will see a decrease in the coke porosity only if the inerts are incorporated well into the binder phases of the coke. (Price, 2017)

Equation 3 - Apparent Specific Gravity

$$SG_{app} = \frac{W}{(W_1 + (W_2 - W)}$$

Where W is the mass of dry coke, W_1 is the mass of water displaced by wet coke and W_2 is the mass of wet coke.

2.8. Coke Carbon Forms

Like coal, many tests were designed for physical and chemical analysis of the metallurgical coke produced. The chemical and physical properties essential for operational choices at the coke batteries and blast furnace fuel needs. Physically, the optical properties of carbon coke forms can provide insight into the strength, reactivity, and performance in the blast furnace, resulting from the coal rank and type. In the 1950s to 1980s, significant advancements were made to understand the coke forms and textures of industrial metallurgical coke (Gray & DeVanney, 1986).

During this time, studies on coke wall, pores, and crystallinity of coke were used to develop coke petrography techniques for microscopically characterizing the properties of coke structure and texture. (Gray & DeVanney, 1986) The coke structure relates to the amount and size of the coke pores and wall in review, whereas the texture is the optical properties of the coke carbon. Coke petrographic analysis is performed under polarized light from a microscope. A pellet for coke analysis is prepared by placing coke in a plastic powder that is heated to solidify. The pellet is polished before being placed under the microscope that is calibrated before every sample reading. To read the reflectance, the sample is spun on a microscope table where the density of reflection changes. Smaller pieces of coke are harder to read, and larger sized pieces of coke are used to look for cracks and textures. (ASTM, 2020)

There are two major categories for carbon forms of coke when completing an analysis: Binder Phase and Filler Phase. The binder phase carbons are from the coal macerals of vitrinite, exinite, resinite, and reactive semifusinite. There are five binder phases of carbon: isotropic, incipient, circular anisotropic, lenticular anisotropic, and ribbon anisotropic. Each phase has a vitrinoid type (V-type) associated with them that marks a 0.1% change in reflection that allows for the coke carbon forms to be distinguished from one another. The summary of carbon form with the associated V-type, coal rank, texture and size are found in Table 2-2 below (Price, 2017). When performing reflectance observations of the textures, using a polarized light and tint plate, rotating the stage for reflectance readings will see different colors and transformations of light typical to each carbon type. The isotropic carbon is seen as pink to purple in color. The incipient carbon forms remain pink to purple in color but appear to have a light texture when rotated. Circular anisotropic, often referred to as granular structures, are fairly circular in form and increase in size as the V-type increases. Lenticular anisotropic, also known as leaflet structure, has a ratio of 2 to 4 with respect to length and width that increase from fine to medium to coarse with V-type increase. Whereas ribbon anisotropic carbon forms, also known as flow structure, have a 1 to 4 ratio of length to width with a color range of pink or red to blue when observed. Counts of the coke pellet sample are taken and used for analysis and property determination. By counting the proportions of carbon forms and assigning them to the appropriate V-types, the blend proportions of coal volatilities can be estimated. (Gray, 1995)

The technique and ability for coke texture analysis and counting takes training and time. The different structures, forms, and reflectance characteristics must be learned and are operator dependent to a certain degree. There are anticipated actions of the different structures when rotated on the microscope table. Mosaic structures will be seen due to the light effect by appearing to change from on to off in light when rotating table. Lenticular structures see light fade and have sweeping color effects in one direction when rotating a sample on the table with size measured as width to length ratio. Domain structures are seen as bright or not when revolving on the table and are tubular-shaped in 3D form. Examples of what binder forms and their coke texture would look like are shown below in Figure 2-17. An isotropic binder is associated with isotropic texture, an incipient binder form is associated with very fine mosaic texture, and a circular anisotropic binder is associated with fine, medium, and coarse mosaic textures.

Table 2-2- Binder Phase Carbon Forms for Coke Petrography (Price, 2017)

Binder Phase Carbon Form	**Coal V-type**	**Coal Rank**	**Texture**	**Size**
Isotropic	V 6-7	From High Volatile Coal (Poor Coking)	Isotropic	
Incipient	V 8	From High Volatile Coal (Fair Coking)	Very Fine Mosaic	<0.5 μ
Circular Anisotropic	V 9 - Fine V 10 - Medium V 11 – Coarse	From High Volatile Coal (Good to Excellent)	Mosaic • Fine • Medium • Coarse	• 0.5-1.5μ • 1.5-5μ • 5-10μ
"Flow" Lenticular Anisotropic	V 12 – Fine V13 – Medium V14 – Coarse	From Medium Volatile Coal	Elongated Flow • Fine • Medium • Coarse	
Ribbon Anisotropic	V15 – Fine V16 – Medium V17&18 – Coarse	From Low Volatile Coal	Domains • Flat • Undulating • Ribbon	

Filler phase carbon forms are from the coal macerals of inert semifusinite, fusinite, and potentially from micrinite, macrinite and inertodetrinite that come from organic, inorganic and oxidized coals. A summary of the filler phase forms, their potential sources and textures can be seen in Table 2-3 below (Price, 2017). Pyrite is listed as a mineral matter for counting but is often counted separately.

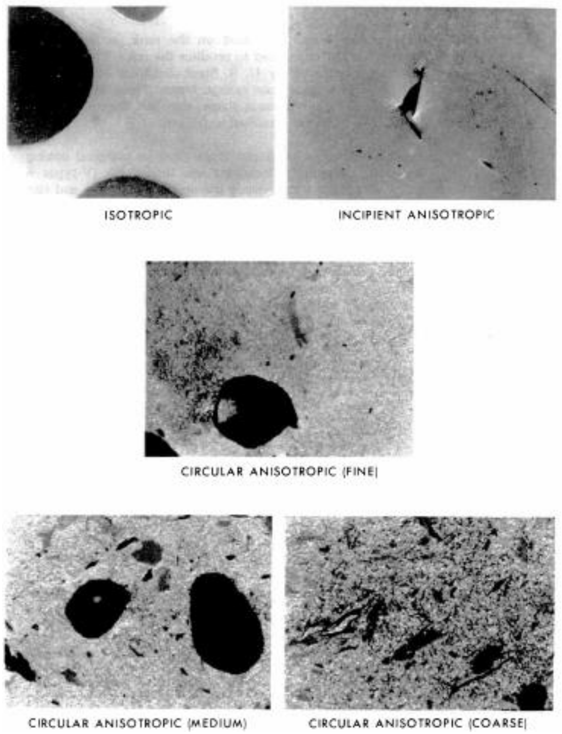

ISOTROPIC
INCIPIENT ANISOTROPIC
CIRCULAR ANISOTROPIC (FINE)
CIRCULAR ANISOTROPIC (MEDIUM)
CIRCULAR ANISOTROPIC (COARSE)

Figure 2-17- Binder Phase Forms Polarized reflected light, 400x (Gray & DeVanney, 1986)

Table 2-3 - Filler Phase Carbon Forms for Coke Petrography (Price, 2017)

Filler Phase Carbon Form	**Source**	**Texture**
Organic Inert	**Fine** <50 μ – Micrinite, macronite, inertodetrnite **Coarse** >50 μ – fusinite, semi fusinite, macronite **Coal Derived Mineral Matter** – clay, shale, pyrite	Inerts: • Fusinite • Semi-fusinite • Unidentified Inert
Inorganic Inert		
Oxidized Coal		Altered Vitrinite

Similar to binder phase carbon identification, there are typical interactions of filler phase forms for counting. Inert Fusinite forms will not reflect light. They stay dark on rotation of the stage but can be identified by their bone structure and crisp lines. Semifusinite can look similar to fusinite pieces and can have some light change on pieces; but most stay dark. Mineral Matter can show up circular in pieces of coke, quartz shows as a transparent mineral and looks different at different magnifications.

Coke for microscopic analysis can be produced from the pilot oven or SHO when conducting lab tests. It has been shown that the texture results and distribution are similar as presented in the 2011 work for determining CSR values of coke produced from the pilot oven compared to SHO coke (McPhee, et al., 2011). This work showed that there was no bias on CSR results from the pilot or SHO coking method. A 95% confidence interval of 1.6% with a standard deviation of 3.6% was found in the test work. Seven samples of coke were produced in both pilot oven and SHO tests for texture with the result that the distribution of binder and filler phases were found to be similar (McPhee, et al., 2011). Often there is an increased bulk density in the SHO due to the pressure plate, but the textures from the SHO are comparable to MWO. Texture analysis performed at the Canmet lab is a combination of the US Steel method (Gray & DeVanney, 1986) and Canmet method following the ASTM D5061 Standard Test Method for Microscopical Determination of the Textural Components of Metallurgical coke standard discussed.

2.9. Coke for the Blast Furnace

The ultimate end-use for metallurgical coal is coke in the blast furnace. The coal selection and coke quality monitoring are all critical for process optimization and stable blast furnace operation. (Poveromo, 2019) The physical and chemical characteristics of the coke charged into a blast furnace impact the operation from raw material requirements, thermal interaction, chemical interactions, and structural support for the porosity to allow gasses to pass as the material descends the blast furnace shaft. The coke requires physical strength to support the ore material, allow for gas passage and the abrasion of the process. Thermally the coke needs to react to produce heat to melt the burden once it descends. Chemically the coke needs to generate gasses for the removal of oxygen from the ore material and the carbon is needed for carbon dissolution. As discussed in the coal and coke properties, due to the impact to blast furnace operations, the coke quality needs to meet specific standards. The characteristics of the coke produced for the blast furnace have general rules of thumb for their impact, as seen in Table 2-4 below but are not limited to these impacts. The ash content of a coke fed to the blast furnace would require the addition of approximately 1 kg of flux per 1% increase in ash and will increase coke required per ton hot metal production (Price, 2017). For every 0.1% decrease in sulfur in coke fed to the blast furnace, there will be an increase in blast furnace productivity and a coke consumption decrease of 1% (Price, 2017). Phosphorus content in coke goes through the blast furnace process right into the hot metal and must be controlled by the input to the coke (Poveromo, 2019). The moisture of coke entering the furnace needs to be kept to a minimum not only because it will replace carbon content but also due to the decrease in blast furnace top temperature by 7°C for every 1% increase in coke moisture (Poveromo, 2019). Coke stability is important for the amount of coke required in the blast furnace process. When stability is above 62, every 1% increase reduced the coke rate needed by 1 kg. When stability is lower than 62, every 1% increase reduced the required coke rate by 2.25 kg. (AIST, 2000)

Table 2-4 - Rules of Thumb for Blast Furnace Coke Quality Impact (Poveromo, 2019) (Price, 2017)

Characteristic	Change	Coke Quality Impact	BF Impact
Ash	+1%	Replaces carbon	Increases required flux 0.7 – 1.1 kg +6 kg/thm coke rate -0.35% blast furnace production
Sulfur	-0.1%	60-70% coal Sulfur converts to coke Sulfur	Increases the blast furnace productivity by 0.8% Decreases coke consumption by 1%
Phosphorus	+0.1	No effect on coke quality	Travels through BF process and needs to be removed by hot metal pre-treatment or secondary refining
Moisture	+1%	Replaces carbon	Decreases top temperature by 7°C
Stability	+1%	Increased stability	-1 kg/nthm coke rate for stability >62 -2.25 kg/nthm coke rate for stability <62

2.10. Environmental Consideration

The largest challenge cokemaking and ironmaking operations now face is their environmental impact and resource availability. The steel industry is a large contributor to greenhouse gas (GHG) emissions, including carbon dioxide (CO_2). For North American cokemaking facilities, primarily of the byproduct design, there are many environmental regulations in place for operating and monitoring of the operations that are continually being updated and pushing the environmental stewardship.

There are many components of cokemaking operations that will need to be considered from an environmental responsibility standpoint moving into the future. Since the 1990's cokemaking operations in Canada have followed the Toxics Substance Management Policy (TSMP) under the Canadian Environmental Protection Act (CEPA). With respect to the CEPA, the management of the cokemaking environmental impact is under the Steel Sector Strategic Options Process (SS-SOP). Representatives from the Provincial and Federal governments, industry, and non-governmental organizations joined to create sixteen recommendations to address programs and their regulatory and socio-economic impacts to improve environmental performance (Sebestyen, 2019). In 2015, the SS-SOP set the environmental direction for Canadian steel manufacturing with two of the sixteen recommendations specific to cokemaking control of polycyclic aromatic hydrocarbons (PAHs) and benzene emissions of cokemaking byproduct plants. All coke ovens in Canada are in the province of Ontario, where each location has Site-Specific Standards (SSS) overseen by the Ontario Ministry of the Environment and Climate Change (MOECC). The regulations and standards for air quality determined by these regulatory bodies with respect to acidification management, SO_2 emissions, particulate matter, byproduct production, and water standards are diligently monitored and managed as stricter limits are pushed. (Sebestyen, 2019)

In 2017, the three operating blast furnace companies in Canada reported a total of 10.9 million tonnes CO_2 released and 11 million tonnes Greenhouse Gas equivalent CO_2. (Canada, 2018). An estimated 1.5 tonnes of CO_2 is released per tonne of hot metal produced from the blast furnace ironmaking operation (Ng, Giroux, MacPhee, & Todoschuk, 2012). By 2050 the Canadian Steel Producers Association has set an aspirational goal to be net-zero CO_2 emissions through a call to action released in 2020. There are several pillars identified in the call to action document that will be required to see this goal be executed (CSPA, 2020).

2.11. Biocarbon for Cokemaking

In order to reduce the environmental impact of the coke and ironmaking operations as well as for, cost reduction and coke requirements for the blast furnace, there has been an increase in research for to investigate more sustainable options for coal and coke replacement. Injection of coal or natural gas has become standard in North American blast furnace operations for coke reduction, but the latest approach has been the investigation and implementation of biocarbon based materials. Biomass or biocarbon can be any plant or animal-based material. The focus in the steel industry has been on biomaterials that can be used as a fuel source. The addition of biomass material has the potential to lower GHG for the coke and ironmaking processes. While biocarbon materials are still carbon based and will release CO_2 gas when used as a fuel, there is a difference in the timing of CO_2 release. For coal, a fossil fuel, it can take millions of years to transform the biomaterial into coal and this is considered a new release of GHG emissions when used in the steelmaking process. For biomaterials, this combustion is considered GHG neutral. The CO_2 released from the combustion of biomaterials is balanced by the CO_2 that was captured during the recent growth of the material. With bio-based sources being considered carbon neutral, it can reduce the reportable GHG emissions of the process even at low usage. Publication of results of the addition of biomaterials with successful results have been at low replacement amounts. As the impact of the additives to a coal blend cannot have a large effect on physical or chemical properties to be used in the blast furnace process low amounts have been found to be the recommended option. Additions of biomaterial to coal blends result in a product known as bio-coke versus coke produced from only coal. (Ng K. W., MacPhee, Giroux, & Todoschuk, 2011)

In countries with steel production facilities and large biomaterial availability like Canada and Brazil, there are numerous programs in progress for the evaluating of biomass materials for fuel sources. For Canada, biomass is the second most abundant renewable energy source. This includes wood pellets, wood chips, crop residues, waste-derived fuel, and biocarbon. The challenges faced in Canada to increase biomaterial consumption for replacement of fossil fuels is the logistics, infrastructure, and capital costs needed to bring the technologies and material for to use in Canadian industrial settings. Current investment limitations to incorporate more biomaterials as fuels in Canada include the current low cost of natural gas, capital risk assumptions, and policy changes. (Group, 2019)

2.11.1. Coal Additives

In the past decade, there has been an increase in the quantity of research and published works evaluating biomaterial addition to coal blends. The interest in biomaterials for coal substitution has grown due to their availability, options, and the potential GHG reduction they could provide. Substituting coal with a renewable biomaterial can be done with little modification to the existing process. Operational batteries have well established coal handling practices. Process modifications to coal handling systems to allow biomaterials additions at operational batteries would need to be minimal to not incur major process interruptions. Biomaterial additives can be raw, treated, waste or pyrolyzed material. The selection of the material is impacted by the availability, cost, carbon structure of the material, and operating constraints. In literature, the consensus is that the production of coke with biomaterial additives is a complex initiative to achieve due to the physical and chemical requirements of the blast furnace process (Ng, MacPhee, & Giroux, 2009). In the following sections, reviews of published works that have analyzed the effects of different biomaterials and their forms after addition to coal blends are reviewed.

2.11.2. Raw Material

Raw biomass material is a simple additive not requiring processing before addition. Canada is considered a biomass rich country with 45% covered in forests, making raw biomaterials a reasonable fuel substitution. (Ng, MacPhee, & Giroux, 2009) One of the biggest concerns with using raw biomaterial for coal substitution is transportation costs and logistics. Raw materials usually have a low bulk density and low carbon content. Transporting unaltered raw biomaterial would not be very cost effective due to the location of most Canadian cokemaking facilities relative to the forested areas where raw materials would be readily available. Pelletizing is an option to densify the raw biomaterial and ease the cost of transportation. As of 2010, there were 33 pellet plants with 2 Mt annual production capacity operating at about 65% capacity in Canada, which would allow for potential feedstock supply to Canadian cokemaking facilities. In 2012, a joint life cycle analysis of pelletized raw biomaterial was conducted by the Canadian Carbonization Research Association (CCRA) and Metallurgical Fuels Laboratory (MFL) of CanmetENERGY-Ottawa (Canmet). The analysis found that the use of pelletized and pyrolysis/carbonization raw biomaterial could reduce GHG CO_2 equivalent per tonne hot metal by 83%. (Ng, Giroux, MacPhee, & Todoschuk, 2012)

2.11.3. Waste Wood Material

The availability or transportation of raw biomass to be used in coal blends is not always possible, and in some areas, wood waste material is more readily available. One of the largest waste wood sources in North America is construction and demolition (C&D) wood waste. There is no standard classification of C&D waste wood, but there are grades commonly used in the wood industry and waste wood management. Based on discussions with ArcelorMittal Dofasco, the following is typically used in waste management found in Table 2-5.

Table 2-5 - Waste Wood Classification (Todoschuk T, 2021)

Type	Composition	Contamination	Typical Markets
Grade A	Clean-Dimensional Lumber, Pallets	None	Animal bedding, Panel Board, Mulch
Grade B	Grade A, OSB, Particleboard, Chipboard, MDF, some Painted wood	Plastics, Ash, Drywall, Metals, <1% Debris	Industrial Grade Feedstock, Biomass Fuel
Grade C	Grade B, Fencing, Transmission Poles, Railroad Tie, Cooling Towers	>1% Debris, Pain, Pressure-Treatment Chemicals	Hazardous Waste Material/Disposal

There are challenges associated with the energy density of each of these types of wood waste as well as for their storage requirements as wood absorbs water unlike coal. While Grade C wood is considered hazardous waste and is regulated due to creosote and requires special handling, it is considered to have a high energy content for coal substitution. Torrified wood waste has similar heat and carbon content to coal; however, there are very few companies within Canada that can supply it. The limited availability makes the cost of torrified wood high. (Liew, 2018)

A Spanish study by Montiano, Barriocanal, & Alvarez conducted in 2013 at the Instituto Nacional del Carbón evaluated three different waste wood materials from furniture and flooring factories for their impact on the thermoplastic properties of industrial coal blends. The study focused on the thermoplastic changes of coke during the plastic transformation phase to produce quality coke. From previously published work referenced in this study, it is suggested that biomaterials can only be substitute for coal at a low percentage of the blend. This study substituted 2% by weight into the trial blends using a sawdust size, less than 0.212mm, material that had been heat-treated to reduce oxygen content. The oxygen reduction was required as there are two suggested disadvantages of higher oxygen containing materials. The first is for low rank coals the higher oxygen content increases the reactivity of carbonization and increases isotropic structures. The second is that higher oxygen containing additives of a coal blend will impact the fluidity interactions which limits the thermal depolymerization inhibition of the coal. This study found that the addition of the three waste wood materials resulted in a significant reduction in the thermal properties of the coking process. The untreated waste materials saw less impact than the heat-treated samples on the coal fluidity properties. The heat-treated samples saw a greater decrease in fluidity, however, the heat-treated biomass saw a lower volatile matter content. The biomass could interact more with the volatiles from the coal, which is a positive impact. (Montiano, Barriocanal, & Alvarez, 2013)

2.11.4. Pyrolyzed Material

Pyrolysis is the process of changing the chemical composition of a material through heating at a high temperature. Charcoal is a pyrolyzed biomaterial that has been considered for coal substitution in published work. (MacPhee, Gransden, Giroux, & Price, 2009) Canadian resources could provide charcoal to be used in the steel industry. In 2009 a series of lab tests were conducted to evaluate the use of charcoal to replace coal at up to 10% in an industrial coal blend at the Canmet MetFuels Research Facility. This work found that larger charcoal, − 3/8 inch to +1/4 inch, produces significantly better coke than finely divided charcoal. The CSR and CRI were impacted differently by the size of coke but showed at low percentage of coal replacement, there was no significant impact. (MacPhee, Gransden, Giroux, & Price, 2009)

Additional work at Canmet in 2009, trialed the substitution of charcoal for coal, at 5% and less, at different sized charcoal pieces resulted in different physical properties. The trials conducted with finer additions (<0.07mm) did not see an impact on stability and hardness compared to the coarse materials (>2.4mm) which saw a significant reduction in these parameters. Microscopically the charcoal additions were identified by their cellular structure. As seen in Figure 2 17, the larger charcoal was not embedded in the coke, which explains the decrease in stability and hardness. In comparison, the finer charcoal was embedded in the coke, which saw comparable stability and hardness to coke produced with no additives in the study. This study also saw negative changes in CRI and CSR that were related to the increased mineral content with charcoal added. The coarser charcoal additions saw less impact on CSR because the reactivity of calcium with CO2 was more localized than with the finer charcoal. (Ng, MacPhee, & Giroux, 2009)

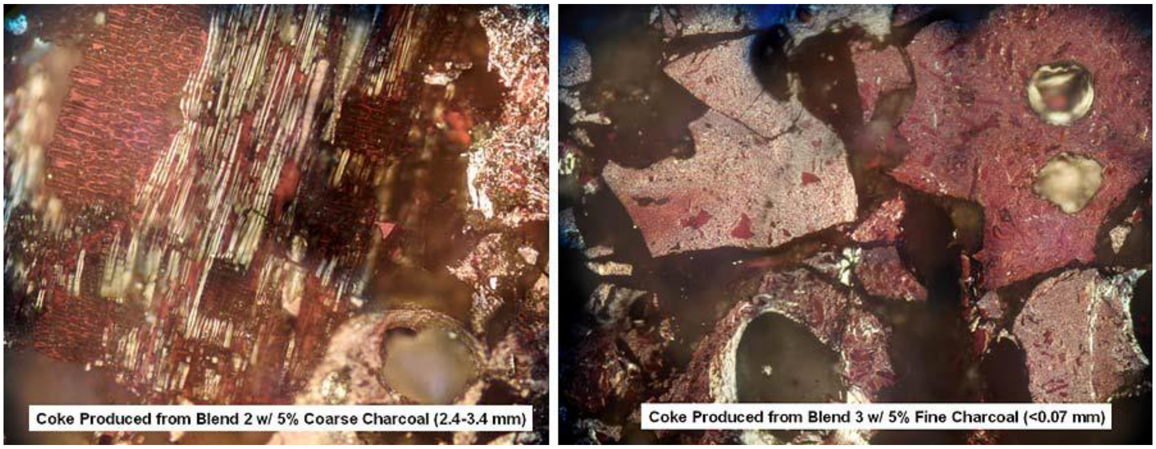


Figure 2-18- Microscopic Image of Charcoal in Coke (Ng, MacPhee, & Giroux, 2009)

In 2011 United States Steel Corporation completed a study to evaluate the substitution of coal at low percentages with torrified (lightly pyrolyzed) biomass. The work parameters were determined by literature review and completed at United States Steel Corporation's USSE Research facility in Kosice, Slovakia. They indicated that a low weight percent substitution could be accepted in an operational coal blend but may also help decrease wall pressure, ash content and sulfur content of the resultant coke. Pyrolyzed biomaterial was selected in this study because it is easier to grind, has less moisture and a higher calorific value than a raw or untreated biomass alternative. It was also noted that the benefits of a torrified biomaterial having lower sulfur and nitrogen contents than a typical high volatile coal it could replace in the study could potentially reduce SOx and NOx emissions in the operating processes. A series of pilot oven tests were conducted in duplicate, comparing the biomass additions to an industrial base blend of coal. The size of the torrified biomaterial was used as received and at a smaller grind to evaluate the size impact. Results of these tests found that the higher ash composition of the torrified biomaterial lowered CSR and increase CRI. The larger sized material saw a slightly higher reactivity than the smaller sized material, which was associated with the higher porosity of the larger material. There were no coke texture results included in the study, only comments on the visual appearance of the coke and that the coke petrographic results reported higher filler phase carbons than the baseline. The report stated that the biomaterial present in the coke texture analysis appeared to be isotropic but did not appear to bond well to the coke matrix and likely acted more as an inert material. (Thomas, McKnight, & Serrano, 2011)

In 2019, work at CanmetENERGY reported on the use of pyrolyzed biomaterials added to coal blends in a briquette form. From ongoing industrial research, it is known that loose biomaterials added to coal blends reduced coke quality because of the highly reactive nature of the additions in the coal to the resultant coke matrix formed. The briquette increases the density of the pyrolyzed biomaterial being added, which could limit the reactivity interactions for the coke that will be produced. The experiment used lab scale coking equipment, the Sole Heated Oven (SHO), to evaluate the impact on CSR and CRI of three biomaterials. Three different materials from different pyrolysis technologies were evaluated after addition to the same coal blend, all at 5 weight percent replacement of coal. A similar reduction in CSR and CRI was seen in all three tests, even with the additive materials all having different ash compositions. This finding leads to the theory that the chemical composition of the biomaterials being added has a limited impact in the reduction. The biomaterials were briquetted and added to two different industrial coal blends and tested in the sole heated oven. The resultant coke had notable visual indication where the biomaterial briquette had been in the coal blend. In these locations, notable fissures through the coke also went through the residual briquette location, signifying the briquette bonded well with the coke matrix. The CSR of this coke was not as significantly reduced as the loose material coke, but CRI did increase compared to the base blends. This finding suggests that coke with high reactivity and high CSR can be produced when a briquetted biomaterial is added to the coal blend. The biomaterial briquettes allowed for a 10 percent by weight substitution of coal in comparison to 5 percent by weight loose biomaterial with less impact on CSR. This resultant coke could potentially increase blast furnace efficiency by lowering the thermal reserve zone temperature required for the coke, which could reduce the GHG impact of the blast furnace process. (Ng, Huang, & Giroux, 2019)

3. Gap of Knowledge

There has been an increased amount of research into assessing biomaterials substitution in metallurgical coal blends to reduce fossil fuel consumption. Biomaterials have been selected for coal substitution due to their availability and range in material options. Substitution of coal with a renewable biomaterial can be done with little modification to the existing process, however, the type of biomaterial best suited to this purpose has not been established. This study investigates three different biomaterials that would be available to potential industrial users in Canada.

The effects of including biomaterial in a coal blend have been shown in published literature and previous industrial test work. This includes the reduction of coke stability and hot strength. However, the reason for these reductions and the connection to coke texture changes has not been investigated in depth. The work presented in this study aims to fill this gap by focusing on how the added biomaterials interact with a known coal blend at a microscopic level. The potential correlation of the added biomaterial interactions and impact on coke quality would allow operational facilities to implement the substitution with known quality impacts.

4. Research Methodology

The overall aim of this study is to compare a base coal blend used in operating coke ovens to blends containing low level substitution of the different biomaterials and evaluate the subsequent impacts on the resultant coke. To investigate the relationship between coke strength and texture changes from the substitution of coal with different biocarbons, a series of pilot and lab scale Sole Heated Oven (SHO) experiments was organized. The resulting coke was prepared for coke microscopy analysis for each trial.

The experiments were divided into two sets of trials. The first was a set of four pilot oven tests. The second, a set of eight lab scale SHO tests. Each set of trials used a fresh base blend of coal and compared to the base blend coal properties. The biomaterials that were selected for addition to the base blend are available waste materials to the operational user. Material A is a non-treated material whereas Material B and Material C have additives. Material D is a pyrolyzed biomaterial and Material E is a cokemaking operational waste. All biocarbon samples under evaluation were prepared in the same manner and substituted to the same base coal blend of each set of trials.

The experimental procedure followed for the test work can be visualized in Figure 4-1. Coal and biomaterials that were selected for testing were prepared for use following the laboratory and ASTM standards. Once prepared, the coal was evaluated for physical, chemical, and petrographic properties before biomaterials were added. For the four pilot oven trials, the coal blend was added into the charge car for loading into the pilot oven and a portion saved for SHO testing. In the second set of trials, the coal blend is only charged in the SHO and not the pilot oven. The resultant coke from the pilot oven and SHO tests were collected for chemical, physical and microscopic analysis. The data produced from all the test work was analyzed and statistical analysis was completed on the texture results from coke microscopy. All pilot oven and SHO tests along with coke analysis and textures conducted for this work were completed at the Metallurgical Fuels Laboratory (MFL) of CanmetENERGY-Ottawa (Canmet), a national research laboratory within the Department of Natural Resources Canada. In this section, the details of the trials conducted, and the procedure followed for evaluation are discussed.

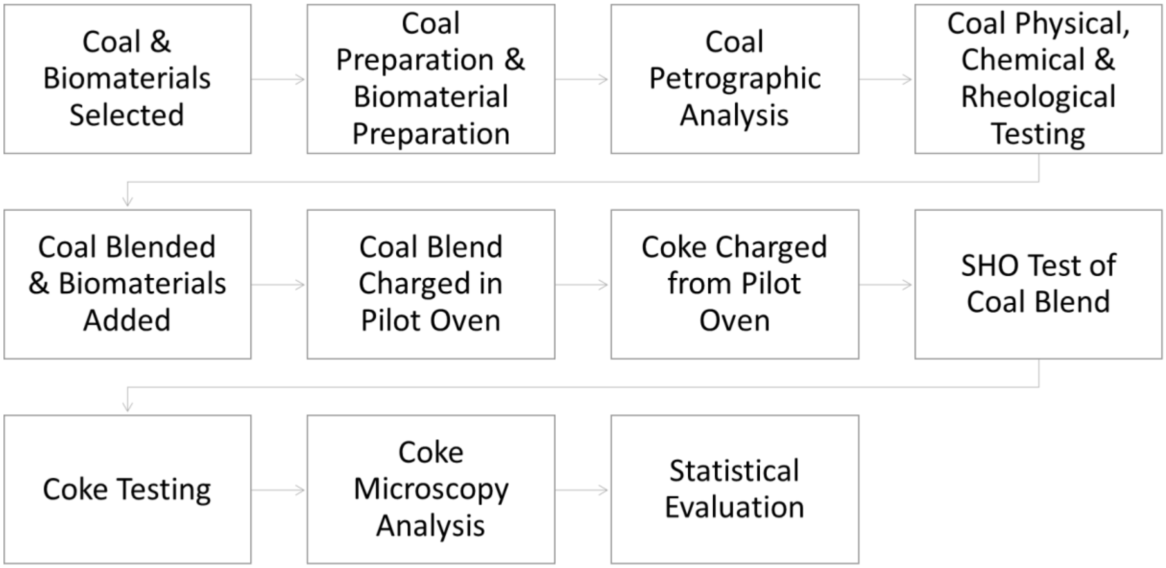


Figure 4-1- Process Flow Diagram of Experimental Procedure

4.1. Petrographic Sample Preparation

Petrographic analysis on the coal being used in all four pilot oven tests was only conducted on the base blend, as the added biomaterials do not affect the results of the analysis. The coal sample was prepared for analysis following ASTM standards as summarized in the literature review. A 1-inch mold was used to set a 50/50 ratio of -20 mesh coal with epoxy and left to set for 24 hours. Once set, the pellet underwent a four-step series of grinding in an automatic grinder and polisher for preparation for microscopic analysis. The light microscope used at the Canmet lab is seen in Figure 4-2 which is an oil immersion microscope with a simple polarized reflected light.

Figure 4-2 - Optical Light Microscope with Polarized Light Source, Rotating Stage and Photomultiplier (manual or automatic) for Coal Petrography at Canmet

The procedure for petrographic analysis follows ASTM standards under an oil immersion lens. Counts are taken manually of the different maceral groups which are identified by their reflectance, size, and shape as discussed in the literature review. The three main groups of macerals have variations in properties, with vitrinite seen as grey in color, inertinite macerals seen as white in color and liptinites seen as dark material. Liptinites have the lowest reflectance values of the maceral groups. The inertinite group can be broken down into varying degrees of grey scale depending on the rank of coal and what type of inertinite. The lower the rank, the higher volatile, the darker the grey scale. The higher the rank, the lower volatile, the lighter the grey scale. These color variations will correlate to each rank of coal with respect to their corresponding macerals. A full maceral analysis is time consuming and operator dependant. The petrographic counting collects 1000 manual counts on all maceral types. An additional 150 manual counts are taken on the sample to measure the reflectance on only the vitrinite following ASTM standard procedure. The average of each type is calculated and reported along with the mean max reflectance. The distribution of the vitrinite types can give insight into the individual coals used in a blend. The petrographic results of the vitrinite distribution and mean max reflectance of the base coal blends used in all four pilot oven tests and eight SHO tests are presented in Table 4-1. The blends have some variation in vitrinite type with the pilot oven test containing 15.3% low volatile coal (v-type 15-18), 15.3% medium volatile coal (v-type 12-14) and 46.1% high volatile coal (v-type 8-11). The SHO base blend saw a distribution of 10.3% low volatile coal, 10.3% medium volatile coal and 51.8% high volatile coal. It is typical of an operation blend of coals to have the majority in high volatile as this is where the fair to excellent coking properties are derived from. The mean maximum reflectance of the two blends is within the similar range expected for a blend of metallurgical coal.

Table 4-1 - Base Blend Petrographic Results for Pilot Oven and SHO Trials

Coal Petrographic Analysis	V-type		Pilot Oven	SHO
	V-7	%	2.60	0.9
	V-8	%	2.60	4.1
	V-9	%	13.80	19.8
	V-10	%	11.80	22.6
	V-11	%	15.30	4.4
	V-12	%	7.20	0.0
	V-13	%	6.10	1.8
	V-14	%	2.00	8.3
	V-15	%	4.10	6.4
	V-16	%	3.60	2.3
	V-17	%	5.10	0.9
	V-18	%	2.00	0.5
	V-19	%	0.50	0.0
	Mean Max. Reflectance	%	1.17	1.14

4.2. Coal Physical and Chemical Testing

A coal blend of Appalachian coals was selected as the base blend used in industry in both sets of experiments. The base blends vary between the two sets due to coal freshness and availability at the time of testing. Each set is compared to the base blend of the experiment, but the comparison of base blends was not made or needed. When coal is being evaluated for pilot oven or SHO tests, a fresh new base blend is always run for comparison. A fresh sample of coal allows for the most consistent and representative sample of the blends to avoid any contamination, aging or oxidization that could occur if an old sample is compared to.

Coal Proximate, Coal Ultimate, Coal Sieve Analysis, and Sole Heated Oven (SHO) tests were performed on the base blend of the pilot oven tests with the results presented in Table 4-2. The rheological properties of the blend were also evaluated to understand the blend and to give insight into the potential coke product. The base coal blend for the four pilot oven tests was crushed and blended for the trials before arrival at the Canmet testing lab together. The coal proximate and ultimate analysis are reported on a dry basis (db) as per ASTM standards. Moisture is variable between the samples and the dry basis allows for a standard evaluation to be made. The SHO results determined the amount of expansion or contraction experienced from the coal blend in the lab scale set up. A typical value expected for an operational coal blend is from -8 to -12. The result of -9.2 for the base coal is within an acceptable range. The coal sieve analysis ensures that the coal has been prepared and crushed to operational size with 80% or greater passing 3.35 mm. The ash composition of the base coal is tested for comparison to the coke ash composition which will include residuals from the biomaterial in the results discussion.

Table 4-2 – Pilot Oven Base Blend Coal Proximate, Ultimate, Sieve Analysis & Ash Composition Characteristics

Moisture		**%**	**1.06**
Coal Proximate Analysis (db)	Ash	%	8.09
	Volatile Matter	%	28.54
	Fixed Carbon	%	63.37
Coal Ultimate Analysis (db)	S	%	1.12
Sole-Heated Oven Test	Expansion/Contraction	%	-9.2
Coal Sieve Analysis, cumulative	6.30 mm	%	10.1
	3.35 mm	%	18.1
	1.70 mm	%	31.1
	0.85 mm	%	47.4
	0.50 mm	%	59.4
	passing 3.35 mm	%	81.9
Ash Composition	SiO_2	%	50.93
	Al_2O_3	%	29.23
	Fe_2O_3	%	8.71
	TiO_2	%	1.49
	CaO	%	2.43
	MgO	%	0.88
	Na_2O	%	0.32
	K_2O	%	2.26
	P_2O_5	%	0.44
	SO_3	%	2.48

The base coal blend for the four pilot oven tests was crushed and blended for the trials before arrival at the Canmet testing lab. However, due to coal availability, the base blend used for the eight SHO lab scale trials was prepared at Canmet for testing. The blends were different in composition, but both blends were used in operational facilities. The SHO expansion/contraction of -14 is outside the normal range of -8 to -12 but acceptable with the operational conditions of the battery it is used for, making it acceptable. The proximate, ultimate and ash analysis will be used to evaluate the impact of the biomaterial additives in the resultant coke characteristics.

Table 4-3 - SHO Base Blend Coal Characteristics

Moisture		%	0.95
Coal Proximate Analysis (db)	Ash	%	7.79
	Volatile Matter	%	29.65
	Fixed Carbon	%	62.55
Coal Ultimate Analysis (db)	S	%	0.9
Sole-Heated Oven Test	Expansion/Contraction	%	-14.0
Ash Composition	SiO_2	%	52.32
	Al_2O_3	%	30.52
	Fe_2O_3	%	7.35
	TiO_2	%	1.54
	CaO	%	1.79
	MgO	%	0.91
	Na_2O	%	0.37
	K_2O	%	2.18
	P_2O_5	%	0.31
	SO_3	%	1.51

As seen in the initial base blend analysis in Table 4-2 and Table 4-3, the blends fall within reasonable limits of an operational coal blend, as discussed in the respective sections of the literature review. The initial review of the coal allows for validation the blend has been prepared to the specifications and critical properties are as expected before charging into a test oven.

4.3. Rheological Testing of Base Blend Coals

The rheological properties of coals are important for evaluating the coking potential of a blend, and the Geisler Plasticity, Dilatation and Free Swelling Index (FSI) were all tested on the pilot oven base blend. The Geisler Plasticity and Dilation were tested for the SHO base blend for comparison, however, the FSI was not available. All rheological properties of the base blend results, seen in Table 4-4, fall within acceptable ranges of an operational coal blend. The max fluidity is greater than 500 ddpm, which is expected to allow for a desirable plastic phase transformation to occur, resulting in quality coke. As expected with different coal blends there are some differences between the pilot oven and SHO base blends results due to the individual coal properties of each blend. As discussed in the literature review, the maximum fluidity should be evaluated with the temperature range and dilatation results due to variability. The maximum temperatures and plastic ranges have some variance, but due to different coal composition are acceptable.

Table 4-4- Rheological Base Blend Coal Properties

Gieseler Plasticity	**Pilot**	**SHO**
Max. Fluidity, DDPM	1046	4664
Max. Fluidity, Temp C	443	449
Softening Temp. C	402	404
Solidification Temp. C	481	498
Plastic Range	79	94
ASTM Dilatation %	99	170
FSI	8.2	

4.4. ASTM Standards

The coal and coke analysis testing in this work followed the ASTM standards, as identified in Table 4-5. ASTM is an internationally recognized organization for technical standards primarily used in the North American coal and coke industry. Some tests adhere to the ISO standards system or are equivalent to an ASTM standard.

Table 4-5 - ASTM Standards for Coal Analysis

Coal Proximate Analysis	ASTM D7582
Coal Ultimate Analysis	ASTM D3176
Coal Moisture	ASTM D7582
Coal Ash Analysis	ASTM D4326
Coal Sieve Analysis	ASTM D 4749
Petrography Analysis	ASTM D 2799
Coal Reflectance	ASTM D 2798
Gieseler Fluidity	ASTM D 2639
ASTM Dilatation	ASTM D 5515
FSI	ASTM D 720
SHO	ASTM D 2014
CSR & CRI	ASTM D 5341
Coke Stability	ASTM D 3402
ASG	ASTM D 167
Coke Texture	ASTM D 5061

4.5. Raw Biocarbon Sample Preparation

In the first set of trials in the pilot oven, three different forms of raw wood waste biocarbon materials were evaluated for their effect on the coke characteristics of the same base coal blend. The proximate, ultimate and heating value were tested for the three materials to understand potential changes seen in the coke produced, as seen in Table 4-6. There is a significant variation in the moisture of the materials. The coal blend with material additions is adjusted for moisture content during coal handling to meet the targeted bulk density set for the test work. The ash, volatile matter and fixed carbon variations in the material are due to the level of processing or treatment from use the wood waste has undergone before being available for addition to the coal blend. The materials selected for test comparison were chosen based on the availability of supply to industrial users. The advantage of potentially substituting coal with biomaterials is the undesirable elements identified in the ultimate analysis can be directed to cokemaking byproducts over waste disposal costs associated with these levels in wood waste. Material A is a raw wood waste product that has not been treated, Material B is a wood waste product that is from cokemaking operations, and Material C is a wood waste product that would require hazardous disposal once it is finished being used in the initial application of use from other industries.

Table 4-6 - Biomaterials Coal Substitute Characteristics

	Material A	**Material B**	**Material C**
Proximate analysis– wt. %			
Moisture	12.89%	26.21%	47.95%
Ash	1.31%	19.45%	8.57%
Volatile Matter	72.13	50.70%	41.83%
Fixed Carbon	13.67	3.64%	1.65%
Ultimate Analysis – wt. %			
Carbon	44.04%	37.32%	30.69%
Hydrogen	6.41%	6.80%	8.01%
Nitrogen	0.63%	1.25%	0.43%
Sulfur	0.12%	5.68%	0.45%
Ash	1.31%	19.45%	8.57%
Oxygen	47.49%	29.50%	51.85%
Heat Value - BTU/lb	7215	7043	6050

All three materials were to be ground to the same size determined but the industrial users' specifications after any potential contaminants were removed. Due to an error, Material A was not ground to the same size as the other two materials and was charged into the pilot oven before the error was caught. This is taken into consideration when considering differences in the results to be discussed. In the second series of tests all three materials were ground to the same size for use in the SHO tests.

In the SHO series of tests, two additional materials were added for evaluation. The added materials are not raw biomaterials and are considered processed materials. Material D undergoes a transformation of pyrolysis before being added to the coal base blend and Material E was a waste cokemaking product. These materials were selected to compare the interactions and effects of the raw biomaterials to further processed materials. The same testing of proximate, and ultimate analysis for the two additional materials used in the SHO testing is found in Table 4-7. Both materials were smaller than Materials A, B and C as 100% passed the standard size when screened. These materials would require processing to be the same size as other materials. The advantage to these materials over the raw biomaterials is the higher carbon content and lower volatile matter. The ultimate analysis will be used for the evaluation of the coke ash composition.

Table 4-7- Additional Processed Materials for SHO Testing Characteristics

	Material D	**Material E**
Proximate Analysis - wt%		
Ash	1.43%	21.04%
Volatile Matter	26.56%	2.64%
Fixed Carbon	72.01%	76.32%
Ultimate Analysis – wt. %		
Carbon	79.9%	75.15%
Hydrogen	3.8%	0.01%
Nitrogen	0.29%	1.03%
Sulfur	0%	0.79%
Oxygen	14.63%	1.98%

A picture of Material D before addition to the coal blend and a SEM image of this material can be seen in Figure 4-3 from Canmet. It is essential to note the cellular structure of the biomaterial for identification in coke texture analysis.

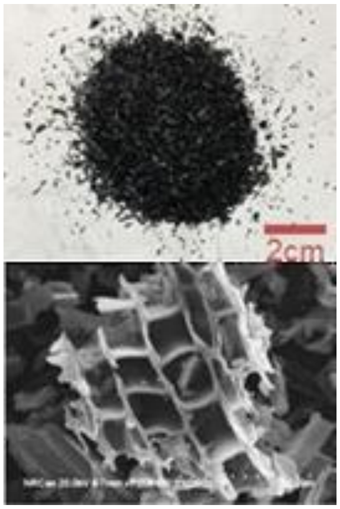


Figure 4-3 – Pyrolyzed Biomaterial Material D Before Addition to Coal Blend at Canmet

For the SHO trials, it was also decided to add two tests with increased biomaterial substitution. The amount was tripled for a trial with Material A and Material C to collect data on the impact of increased substitution. This brought the total number of SHO tests to eight for the second set of trials: SHO Base Blend, SHO Material A, SHO Material A3X, SHO Material B, SHO Material C, SHO Material C3X, SHO Material D and SHO Material E.

4.6. Addition of Biocarbon to Base Coal Blend

The petrographic analysis was not conducted on the coal blends with biomaterial, as petrographic analysis only notes non-coal materials as contaminants. Previous work and work reviewed in the literature survey were referenced for the size of biomaterial to be used (Thomas, McKnight, & Serrano, 2011). A series of SHO tests were conducted to determine the effects of the size of biomaterial to be added to the coke making process and coke quality in previous work which was applied in these experiments. Once the materials were ground to the desired size, the materials were manually mixed into the prepared coal blend based on the weight percent of the charge. The final weight of the coal blend and biomaterial was validated for the addition amount. The biomaterials were manually mixed per Canmet’s standard procedure and not mechanically mixed to avoid potential contamination of the mixing drums. The coal blend with the biomaterial added was then charged into the hopper for charging to the pilot oven or into a clean bin for charging into a SHO test.

4.7. Pilot Oven Testing

The initial set of tests conducted were four pilot oven tests in the Movable Wall Oven (MWO) at the Metallurgical Fuels Laboratory (MFL) of CanmetENERGY-Ottawa (Canmet) seen in Figure 4-4Figure 4-4- 18" MWO Canmet. The MWO is an 18” (460 mm) wide oven lined with SiO_2 refractory walls that are maintained at 1200°C constant wall temperature. The four pilot oven tests were conducted under the same procedure to reduce possible errors. The oven parameters were set for all four tests to simulate the industrial users’ settings and verified before and during charging of the coal blends into the coke oven. Tests were charged with an average of 314 kg net dry weight, 780 kg/m^3 ASTM bulk density, 5% moisture and charged for 18 hours of coking. The resultant coke was pushed and collected for testing.

Figure 4-4- 18" MWO Canmet

4.8. Sole Heated Oven (SHO) Testing

SHO tests were conducted on the initial set of four pilot oven tests as well as the eight tests in the second set of the experiments at Canmet. The Canmet facility has three ASTM sole-heated ovens in use for testing. The dual chamber SHO equipment was used for all twelve tests conducted in this study, as seen in Figure 4-5. The dual chamber is used to run duplicate tests and the average is reported. Each chamber has a 12kg capacity where the prepared coal blend is charged. The chambers were preheated before a charge and underwent the coking process until the top surface of the coal reached a temperature of 500°C on the equipment's top thermocouple. The resultant coke from the pilot oven coal blends was disposed of after the expansion/contraction values were determined. The resultant coke was collected and prepared for further testing in the eight trials only using SHO coke. The duplicate sample results are required to fall within a 3% tolerance to be accepted.

Figure 4-5 - SHO duplicate chamber design at Canmet

4.9. Coke Testing

The physical properties of the resultant coke in the pilot oven tests are important for understanding the potential use of a coal blend in the blast furnace operations. The Coke Sieve Analysis for size analysis, Coke Tumbler test, CSR, CRI and ASG tests were conducted on all coke produced from the pilot ovens. The coke was sized according to ASTM sieve standards and the percent value of each size was reported. The coke tumbler test results in the ambient coke strength reported in stability and hardness values. For the tumbler test, 10 kg of dry 3 x 2-inch coke was tumbled at 24 rpm for 1400 revolutions in the ASTM tumbler shown in Figure 4-6. The coke removed was screened and divided into 1 and ¼ inch screens. The stability is the cumulative coke plus the amount screened greater than 1-inch coke and the hardness is the cumulative plus the screened ¼ inch coke. The results will be presented and discussed in the following section.

Figure 4-6 - ASTM Tumbler at Canmet

The coke from the SHO eight trials was collected and prepared for hot coke strength testing of CSR and CRI, ASG and coke ash analysis in the same method as the pilot oven coke for the first four trials. The equipment used at Canmet, see Figure 4-7, charges 200 grams of 19.0 x 22.4 mm dry coke into the test chamber where it is heated to 1100°C for two hours and reacted with CO_2 at a flow rate of 5 L/min. Once cooled, the CRI results are determined from the loss of coke weight after this reaction time. After the coke is measured for CRI it is tumbled in an I-drum for 600 revolutions at 20 rpm and screened for coke greater than 9.5 mm using a standard sieve. The CSR is determined from this for the cumulative percent of coke after reaction greater than 9.5 mm. The results will be presented and discussed in the following section along with the ASG and coke ash composition results. The resultant coke duplicate samples must be within 10g range of each other to be acceptable results at Canmet for CSR and CRI reporting.

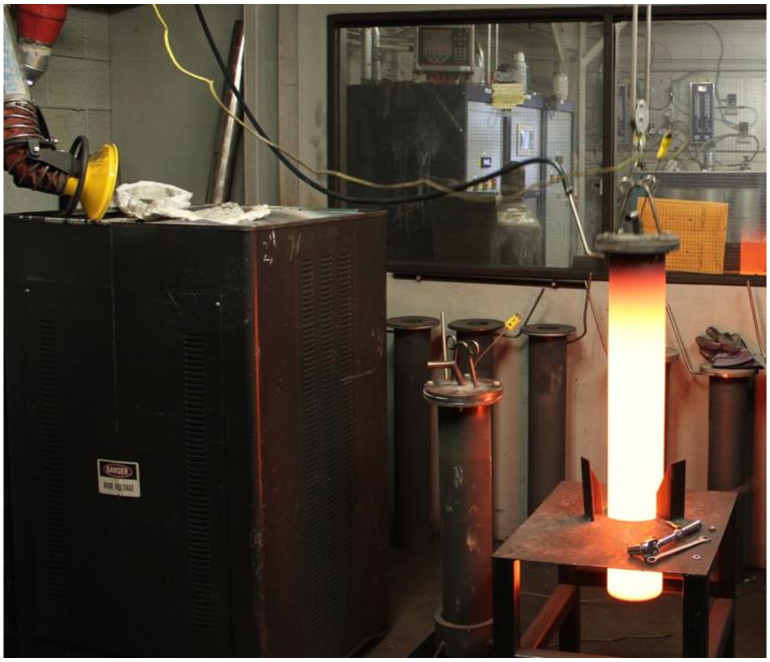


Figure 4-7 - Hot Coke Strength Test Equipment at Canmet

4.10. Coke Microscopy

Coke produced from the pilot oven was used for coke texture analysis in the first set of coke trials. Coke from the SHO was used for texture analysis in the second set of coke trials. Published work indicates the origin of the coke does not change the sample preparation for texture analysis (McPhee, et al., 2011). Following ASTM Standard D 5061, the coke is ground to -16 mesh and mixed at a 50/50 ratio with Loosite powder to make the pellet for analysis. Once mixed, it is put in a heated press for 25 minutes, cooled then polished.

Each pellet was reviewed under an oil immersion microscope, Figure 4-8, with cross-polarized reflected light and tinted plate to collect 1000 points per sample. The counts were manually taken in a step-wise fashion to identify the distribution of the coke textures. The main groups of identification are: (1) mosaics which see a light on/off effect when rotated on the microscope stage, (2) lenticular which sees the light fades with a sweeping color effect in one direction when rotating sample on stage, (3) domain which are seen as bright or not when the stage is rotated and (4) inert material that does not reflect light when rotated. The vitrinite type and sub categories of coke texture classifications used for this set of experiments are highlighted in the literature review Table 2-2.

Figure 4-8 - Petrographic Microscope at Canmet

The predicted coal reflectance can be calculated from the coke texture results and used to compare to the actual coal reflectance reading taken in the petrographic analysis. The reflectance calculated should be close to the reflectance reading of the coal, which is a validation of the tests being conducted. The predicted reflectance value is predicted using Equation 4:

Equation 4- Predicted Coal Reflectance (Price, 2017)

$$Ro = 100 * \frac{(.0165 * TD + .0145 * TF + .0095 * FM + .0113 * MM + .01CM + .0074 * (VFM + IS))}{(TD + TF + FM + MM + CM + VFM + IS)}$$

where TD represents total domains, TF represents total flows, FM represents fine mosaic, MM represents medium mosaic, CM represents coarse mosaic, VFM represents very fine mosaic, and IS represents isotropic mosaic

4.11. Statistical Evaluation

From the data collected in the pilot oven, SHO test trials and the texture analysis Minitab version 18 was used for analytical evaluation.

5. Results and Discussion

In this section the results of the pilot oven trials and SHO trials along with the characteristic analysis of the coke produced, are presented and discussed. All pilot oven and SHO tests along with coke analysis and textures conducted for this work, were completed at Metallurgical Fuels Laboratory (MFL) of CanmetENERGY-Ottawa (Canmet), a national research laboratory within the Department of Natural Resources Canada. The test work was divided into two sets of trials. The first set of four pilot oven MWO trials were for the base blend and one of the three biomaterials being evaluated. The second set of tests resulted from the discoveries of the first set and the need for further data collection. The three initial biomaterials were used again, along with a pyrolyzed biomaterial and a cokemaking waste product. A different base blend of coal was used between the first and second sets of trials to ensure fresh coal was being evaluated.

5.1. Pilot Oven (PO) Test Results

From previous work of the industrial partner's research, a series of four pilot oven (PO) tests were selected. The goal was to compare a base coal blend used in operating coke ovens to three different raw biomaterials being evaluated for substitution of coal at low amounts in the base blend. Coal evaluation tests were only conducted on the chemical properties of the base blend as the added biomaterials will not impact the coal properties measured in petrographic analysis.

5.1.1. Change in Physical Properties of Coke

The three materials selected for addition to the base blend are available waste materials to the operational user. Material A is a non-treated material, whereas Material B and

Material C have additives. The advantage of using the materials with additives in the coal blend is the volatile matter that is expelled from them during the coking process is captured and moved into the byproducts process over a landfill waste. The SHO and coal sieve analysis were conducted on each, and the variation from the base blend is presented in Table 5-1 before the blends were charged into the pilot oven. The values are the difference between the coke additive product from the base blend. The base blend of coal was in the desired range of SHO expansion and contraction limits for operating. The blend had a +/- 2 % tolerance range for the SHO to still be acceptable. The SHO results saw Material A with a slight increase in expansion and Materials B and C with an increase in contraction. Coal size and sieve analysis were expected to have no impact as the base coal blend was prepared for all tests at the same time, with the only difference being the biomaterials added. However, Material A was not ground down to the same size as the other two, and its size was doubled. This size difference could be the reason for the increased variance from the base blend for Materials B & C.

Table 5-1 - Coal Properties Biomaterial Blends for Pilot Oven Tests Variance from Base Blend

Properties			**PO Material A**	**PO Material B**	**PO Material C**
Sole-Heated Oven Test	**Expansion/ Contraction**	**%**	0.14	-0.34	-1.36
Coal Sieve Analysis, cumulative	**6.30 mm**	**%**	3.16	-2.84	-2.87
	3.35 mm	**%**	4.73	-3.45	-2.54
	1.70 mm	**%**	4.53	-3.23	-1.79
	0.85 mm	**%**	3.61	-2.41	-1.27
	0.50 mm	**%**	3.00	-0.25	-0.77
	passing 3.35 mm	**%**	-4.70	3.45	2.50

The four pilot oven tests were conducted, and coke was collected from each trial. Testing of the resultant coke from the pilot oven tests included: coke proximate analysis, coke sizing, stability, hardness, and Apparent Specific Gravity (ASG). Coke produced from the pilot oven tests for each of the four blends was used for testing Coke Strength after Reaction (CSR) and Coke Reactivity Index (CRI). The physical properties variance from the base blend is presented in Table 5-2 for the size, stability, hardness, CSR, and CRI results. These results are the delta of the trial from the base blend. Pilot oven coke is intended to be representative of coke produced in operational batteries. The coke size, stability, and hot strength results are often less than operational results, with each producer knowing the relationship and limits of the pilot oven results to their batteries. For this reason, the values are presented as the delta change of the coke with biomaterial additions from the base blend and not the raw values. A negative value is a decrease from the base blend.

From previous work reviewed in the literature review, it is known that the size and strength of coke are reduced with the addition of biomaterials, and this was observed in the pilot oven tests. Material A saw the most significant drop in mean coke size, but this can be explained as the size of Material A was larger than that of Materials B and C. Material C saw the smallest change in mean coke size. Material A saw the largest decrease in stability but the lowest reduction in CSR compared to Materials B & C. A drop in CSR by 2 points could be expected depending on the base blend in use. However, a 5-point drop for CSR as seen for Material C would be hard to justify not only for the reduction in quality but also the increased coke requirements in the blast furnace to make up for the quality drop. The relationship between stability and CSR is relatively poor and should not be the only factor considered.

Table 5-2 – Variance in Coke Physical Properties from Base Blend after Pilot Oven Tests

Properties			**PO Material A**	**PO Material B**	**PO Material C**
Sieve Analysis of Coke, cumulative	**100 mm sieve**	**%**	-0.17	-0.43	-0.09
	75 mm sieve	**%**	0.14	-1.09	0.36
	50 mm sieve	**%**	-6.72	-2.35	-0.13
	37.5 mm sieve	**%**	-3.17	-2.84	-1.75
	25.0 mm sieve	**%**	-1.25	-1.71	-0.62
	19.0 mm sieve	**%**	-0.63	-0.84	-0.16
	12.5 mm sieve	**%**	-0.40	-0.64	-0.16
	Passing 12.5 mm sieve	**%**	0.40	0.64	0.16
	Mean coke size	**mm**	-1.89	-1.47	-0.26
ASTM Coke Tumbler Test	**Stability**		-1.50	-1.32	-0.77
	Hardness		-0.14	-0.09	0.36
Hot Strength	**CSR**		-0.29	-2.26	-5.09
	CRI		0.08	0.08	3.13

5.1.2. Change in Coke Textures

The effects of biomaterial on reducing the coke stability and strength have been shown in published literature reviews and previous industrial test work (Ng, Huang, & Giroux, 2019). However, the reason for these reductions and the connection to coke texture changes has not been investigated. For this purpose, a pellet was prepared from each of the two coke samples collected from each of the four pilot oven tests, and coke microscopy analysis was performed by counting the different coke textures as reviewed in Table 2-2. No photographs were taken of the coke textures from the pilot oven test samples but would be similar to those shown in the literature review Figure 2-17.

The results from the microscopic analysis of the textures were broken into nineteen different sections for binder and inert coke textures. The percent of each coke texture for the sample was reported. A review of the texture results of each sample compared to the base blend found that there were only notable changes in the isotropic, incipient, and circular mosaic textures of the pilot oven coke samples. The full microscopic results can be found in Appendix A – Pilot Test Coke Texture Results. These coke texture groups were the focus of the statistical analysis to determine contributing effects. The average of the two samples from the base blend and each biomaterial addition can be seen in Table 5-3 for these results.

Table 5-3- Change in Coke Texture of Pilot Oven Tests

Binder Phase	Coke Texture		PO Base Blend	PO Material A	PO Material B	PO Material C
Isotropic	**Isotropic**	%	2.5	2.95	3.05	3.55
Incipient	**Very Fine Mosaic**	%	3.8	2.75	5.3	5.5
Circular Anisotropic	**Fine Mosaic**	%	8.95	7.9	11.2	11.6

To better understand the effects of the coke texture changes from the biomaterials, the main results of each group of isotropic, incipient, and circular were plotted using Minitab. On each plot, the average value of the two pellet samples analyzed for the base blend and three different materials are presented. The materials are ordered in the least to most processed material. As seen in Figure 5-1 for isotropic binder and isotropic texture, Figure 5-2 for incipient binder and very fine mosaic texture, and Figure 5-3 for circular binder and very fine mosaic texture, as the amount of processing increases, there is an increase in each coke texture type observed. Material A is the exception that sees a drop in incipient and circular from the base blend. The individual textures are plotted before they are combined in Figure 5-4- Pilot Oven Isotropic, Very Fine Mosaic and Fine Mosaic Coke Texture vs. Material TypeFigure 5-4 to show how each texture varies with both samples from each trial being included. The dotted horizontal line on each plot is the overall average of all of the samples. The plots are not horizontal in any of the individual plots that validate the effect of the different additives on affect the texture variation from the base blend. For example, Material C is above the data set average, which signifies a notable difference from the original base blend. Material A and Material B are close to the overall average and do not see as significant an effect as Material C. The base blend being below the average signifies it is clearly different in properties than the blends with the biomaterials added.

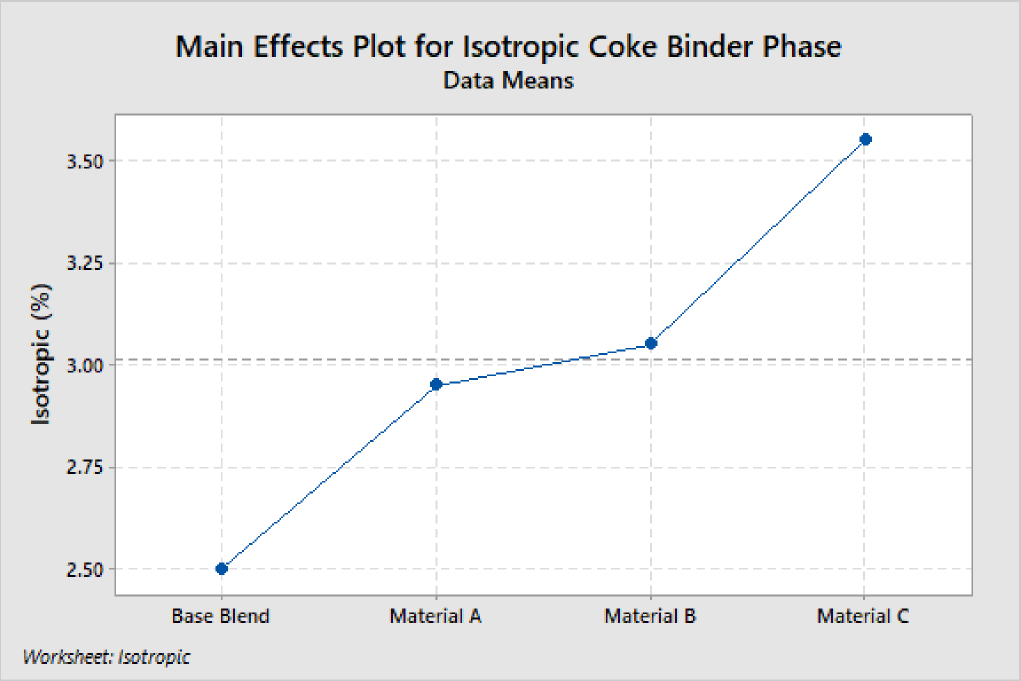


Figure 5-1- Main Effects Plot for Isotropic Coke Binder – Isotropic Coke Texture for Base Blend and Materials A, B and C

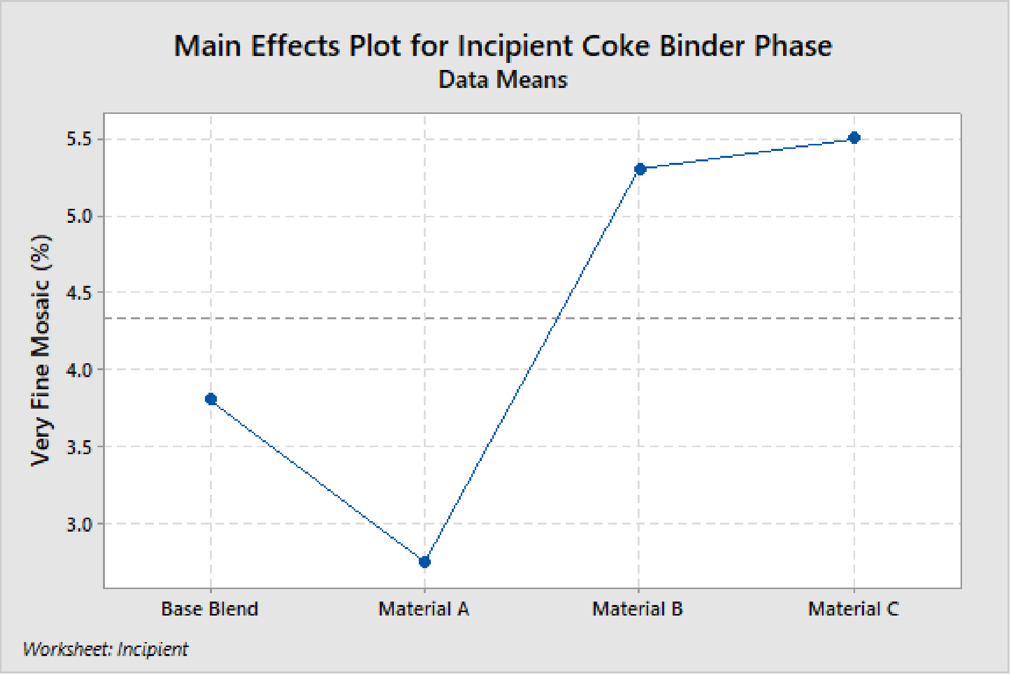


Figure 5-2 – Main Effects Plot for Incipient Coke Binder – Very Fine Mosaic Coke Texture for Base Blend and Materials A, B and C

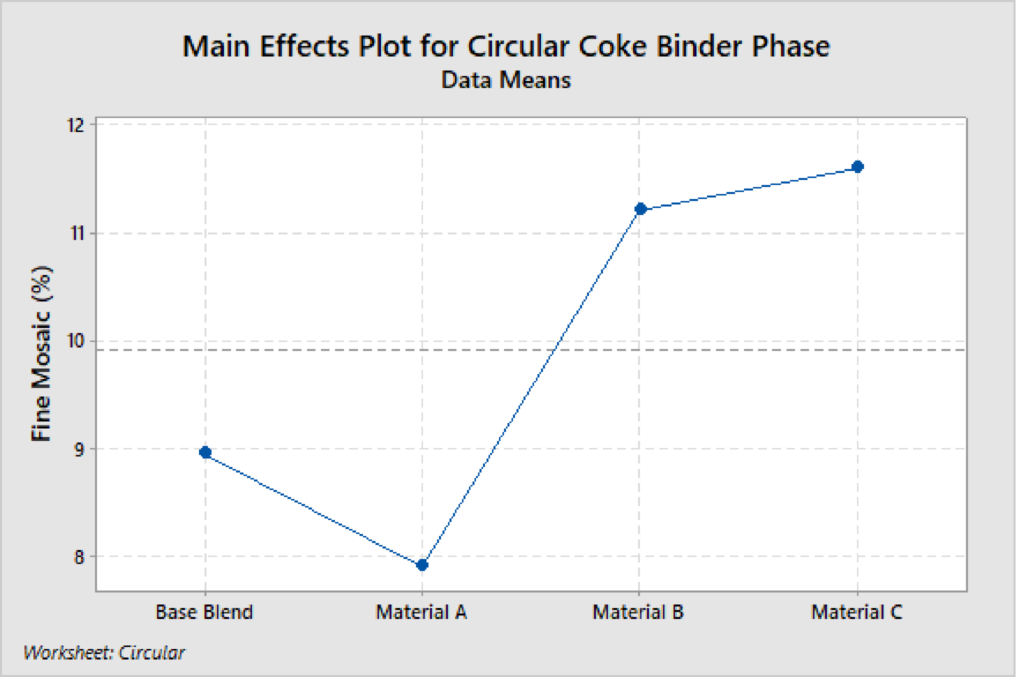


Figure 5-3 – Pilot Oven Main Effects Plot for Circular Coke Binder – Fine Mosaic Coke Texture for Base Blend and Materials A, B and C

These three individual characteristic plots of texture are replotted onto a multi-vari graph in Figure 5-4. The x-axis has the results of the base blend and three biomaterial blends with the isotropic, very fine mosaic, and fine mosaic average textures of each plotted on the y axis. As the materials transition from the least processed in Material A to the most processed of the three Material C, the total sum of all properties sees an increase between each texture. Within each blend, the characteristic of higher fine mosaic, then very fine, then incipient increase as the material processing level increases. The difference between the isotropic and incipient and the incipient to circular increases with material processing as well. As the trend goes from isotropic to fine mosaic the coke texture is coarser and stronger binder type property. This trend verifies that adding biomaterial to a coal blend would increase the amount of finer textures observed which is a negative impact. The red dotted line is the overall average of all of the samples. It was increasing as the materials processing increases with the exception of Material A. As discussed, this could be due to the error in the size and was recommended to be retested in the SHO trials.

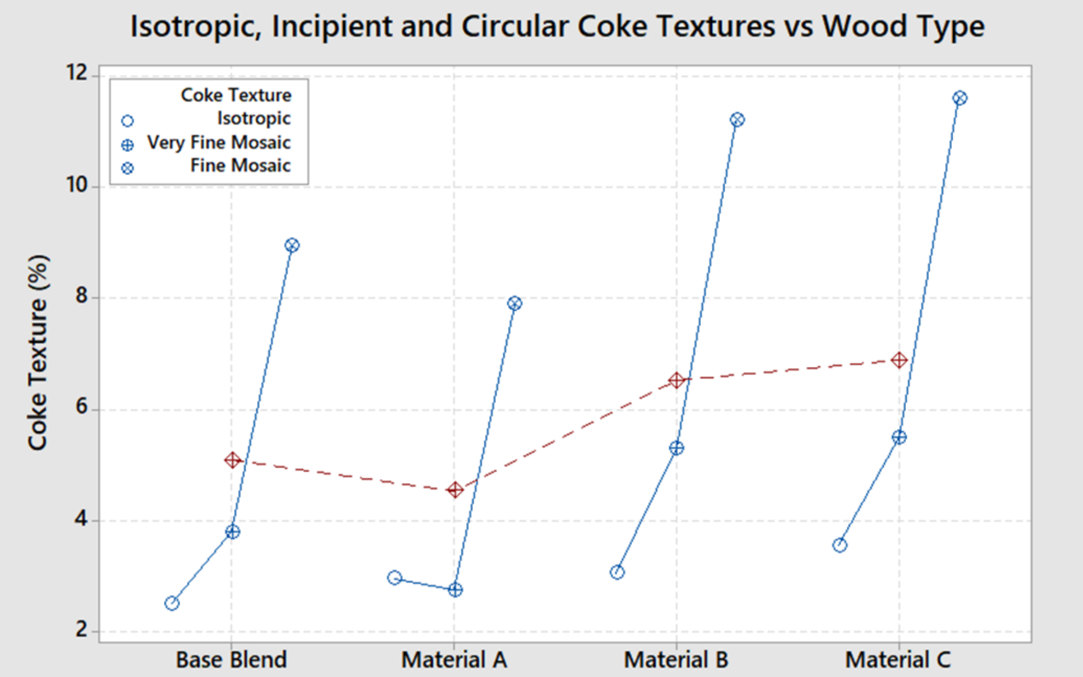


Figure 5-4- Pilot Oven Isotropic, Very Fine Mosaic and Fine Mosaic Coke Texture vs. Material Type

Looking at the texture changes, the next step was to investigate the relationship in changes to strength and reactivity. Plotting the same sequence of main effect plots for mean CSR, there was a decrease in the CSR with the addition of biomaterials in Figure 5-5. The values presented are percent change from the base blend to better visualize the impact of the change from the delta change presented in Table 5-2. There was a reduction in CSR as the processing of the biomaterial addition increased from Material A to Material C. There decrease for CSR value of Material A was relatively small, and this could be related to Material A having a larger size than Material B and Material C. As the CSR decreased, there was an increase in isotropic and very fine textures which are more reactive carbon forms.

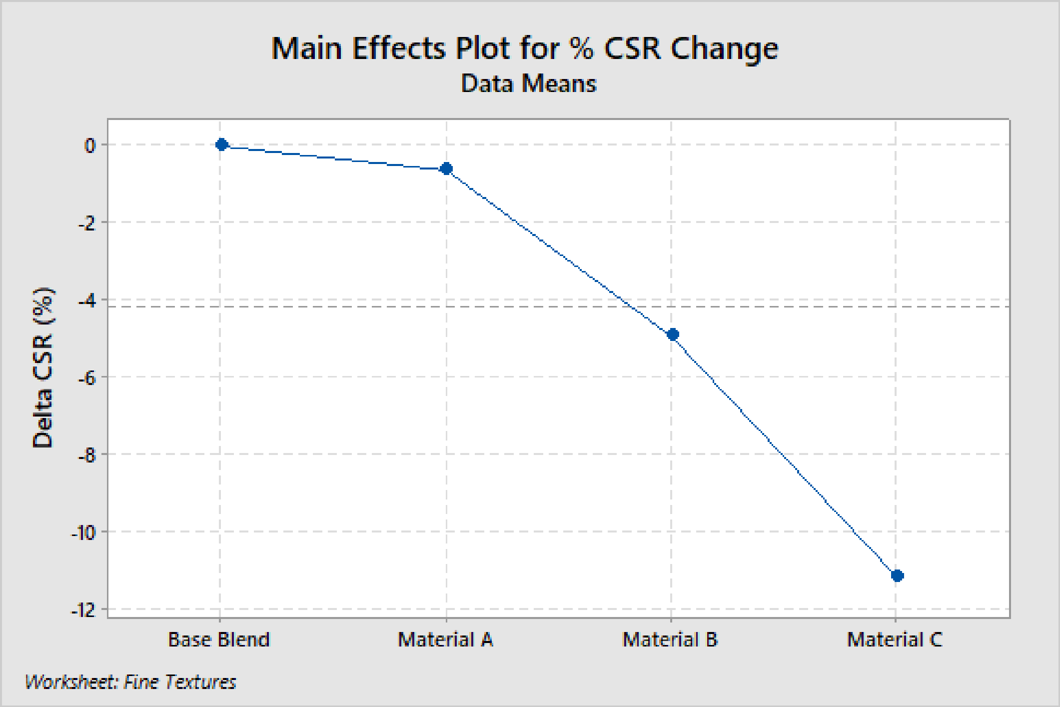


Figure 5-5 - Main Effects Plot for Pilot Oven CSR vs Material Additions

The main effect plot of the mean CRI, as seen in Figure 5-6, shows as expected, the inverse relationship of CSR. Like the CSR plot, the values presented are percent change from the base blend to better visualize the impact of the change from the delta change presented in Table 5-2. As the amount of processing incurred by the biomaterials before addition increases, the CRI also increases. The change in reactivity in Material C shows a sharp increase which could be related to the more significant shift seen in Material C’s very fine isotropic increase seen in Figure 5-1 and Figure 5-4.

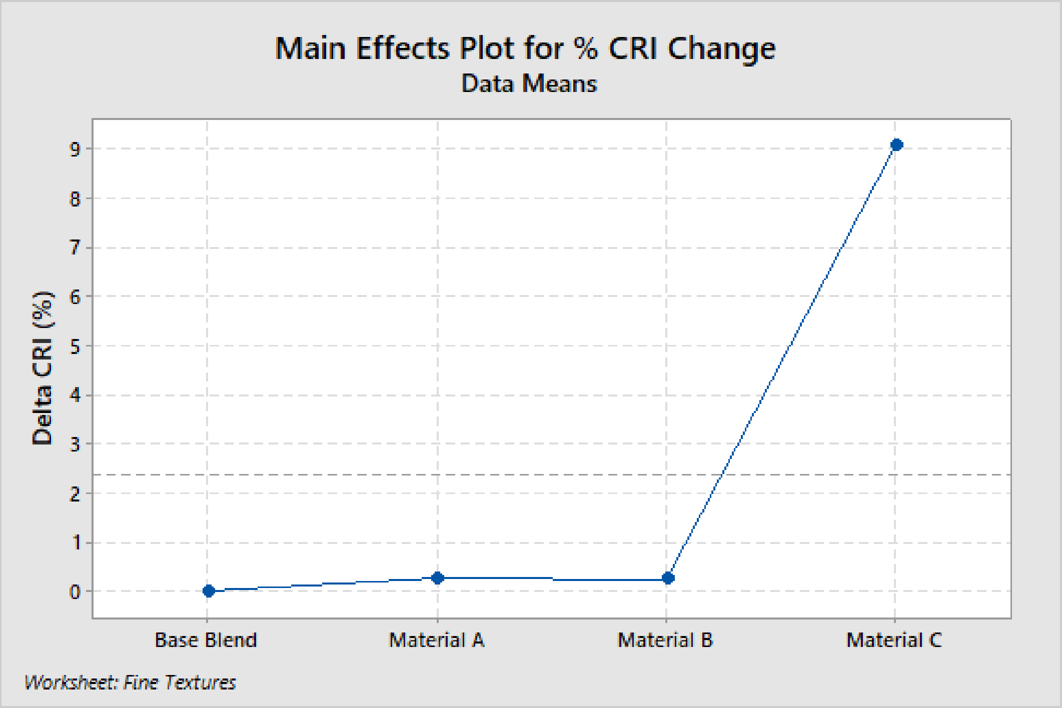


Figure 5-6 - Main Effects Plot for Pilot Oven CRI vs Material Addition

5.2. Sole Heated Oven (SHO) Results

From the initial four pilot tests it was determined additional tests would be required to validate the findings for coke texture interactions. Pilot oven tests are time-consuming, costly and require a large amount of coal. The coke textures analysis was conducted from the SHO coke in the initial tests which are less time-consuming, less costly, and uses smaller samples of coal. The second set of tests were conducted only using SHO coke. Biomaterials A, B, and C were tested again at the same blend substitution amount (1X) as well as materials A and C were tested with the substitution tripled (3X). The changes observed in coke textures between the initial three materials showed as the amount of processing the biomaterials underwent before addition increased the negative quality impacts on results.

One additional pyrolyzed biomaterial and waste coke material were included in the second set of SHO tests. The analysis of coke characterizations of biocarbon additives in different forms will allow for a better understanding of the texture interaction. The pyrolyzed Material D has less volatile matter and higher carbon than the raw biomaterials which made it closer to traditional coal properties. The cokemaking process waste of Material E is to compare the strength impact of a biomaterial compared to a coke material.

5.2.1. Sole Heated Oven Physical Tests Result

The SHO coke allows the data to relate changes in expansion/contraction, CSR, and ASG to the different additives. In Table 5-4, the variation of the expansion/contraction, CSR, CRI, and ASG are presented in comparison to the base blend. The values are presented as the delta change of the coke with biomaterial additions from the base blend and not the raw values. The SHO coke allows for measurement of expansion/contraction with the additives to be evaluated to predict the impact on operational uses of the blends. All variations from the base blend presented in Table 5-4 saw a greater contraction in the coking processes with the biomaterials added. As in the pilot oven tests, the CSR was dropped as the degree of processing increased. Material A a lower drop compared to Material C. As expected, increasing the amount of coal substituted by material saw a decrease in the CSR for trials Material A3X and Material C3X. CRI increased as the more processed materials were added, as in the pilot oven trials, but Material C did see a slightly lower increase compared to Material B. Again, as expected the two tests with increased amounts of Materials A and C saw a greater increase in CRI compared to the original substitution. The ASG results compared to the base blend of coal did not see any significant changes as the processed material increased.

Table 5-4 - Variation from Base Blend of Material A, A3X, B, C & C3X for SHO Coke Trials

Test		SHO Material A	SHO Material A3X	SHO Material B	SHO Material C	SHO Material C3X
SHO Test	**Expansion/ Contraction**	-0.81	-1.96	-2.23	-2.10	-3.05
CSR	**CSR**	-1.61	-5.93	-4.65	-4.93	-7.50
	CRI	1.64	5.58	4.01	3.59	5.44
Density	**ASG**	0.01	0.02	0.02	0.01	-0.02
Coal Ro Calculated		0.00	-0.02	-0.06	-0.01	0.00

The two additional additives, Material D and Material E were compared to the base blend for expansion/contraction, CSR, and ASG as seen in Table 5-5. Material D is a pyrolyzed loose biomaterial. Material D saw an increase in expansion, the greatest reduction in CSR, and a greater increase in CRI compared to all other additive variations from the base coal blend at the same substitution amount. Material E was a fully coked waste material from operations, not a biomaterial, and was added at the same substitution amount for comparing the effects of processed materials, this is not a biomaterial. The impact of this addition fell between Material A and B. Material D saw a significant drop of seven points in CSR which would not be acceptable for addition to an operational coal blend. Material E would need justification of cost savings to be considered in an operational blend as the general range is a variance of 2 points. The ASG is within an acceptable variance from the base blend for both materials.

Table 5-5- Variation from Base Blend of Materials D & E

	Test	SHO Material D	SHO Material E
SHO Test	**Expansion/Contraction**	0.27	-1.68
CSR	**CSR**	-7.01	-3.00
	CRI	4.68	1.65
Density	**ASG**	-0.02	0.02

5.2.2. Coke Microscopy Sole Heated Oven Results

After the strength density results were reviewed, the microscopic analysis of the SHO coke was studied. A single pellet for each of the eight trials was prepared according to ASTM standards as explained in section 4.10 Coke Microscopy . The manual counts of each microstructure observed in a sample will be reviewed below. A report summarizing the percent of each texture observed by the operator is reported. The Canmet microscope is not equipped with a built-in camera. The pictures presented in this section were taken manually through the view lenses while the microscope was set at 625 magnification. There is visible scale from the crosshair marking that are 1.7 μm between each line on the images.

The base coal blend used in the SHO is the reference point for all variations in the analysis as it is a known operational blend in use. Figure 5-7 shows two well-established coke structured pieces in the base blend. Both images show desired coke pore walls that are smooth and well-bonded binder phases of the coal. Inerts are well incorporated into the coke pieces. The microscopic analysis did not note any significant coke structure reduction from inert materials or large cellular structures that would identify biomaterials in the coke. The coke structure on the top is an example of a well-established circular carbon form of fine mosaic. The coke structure on the bottom shows more circular carbon forms in fine mosaic on the bottom half with ribbon carbon binder of flat domain textures notable on the top twelve o’clock position of the coke piece.

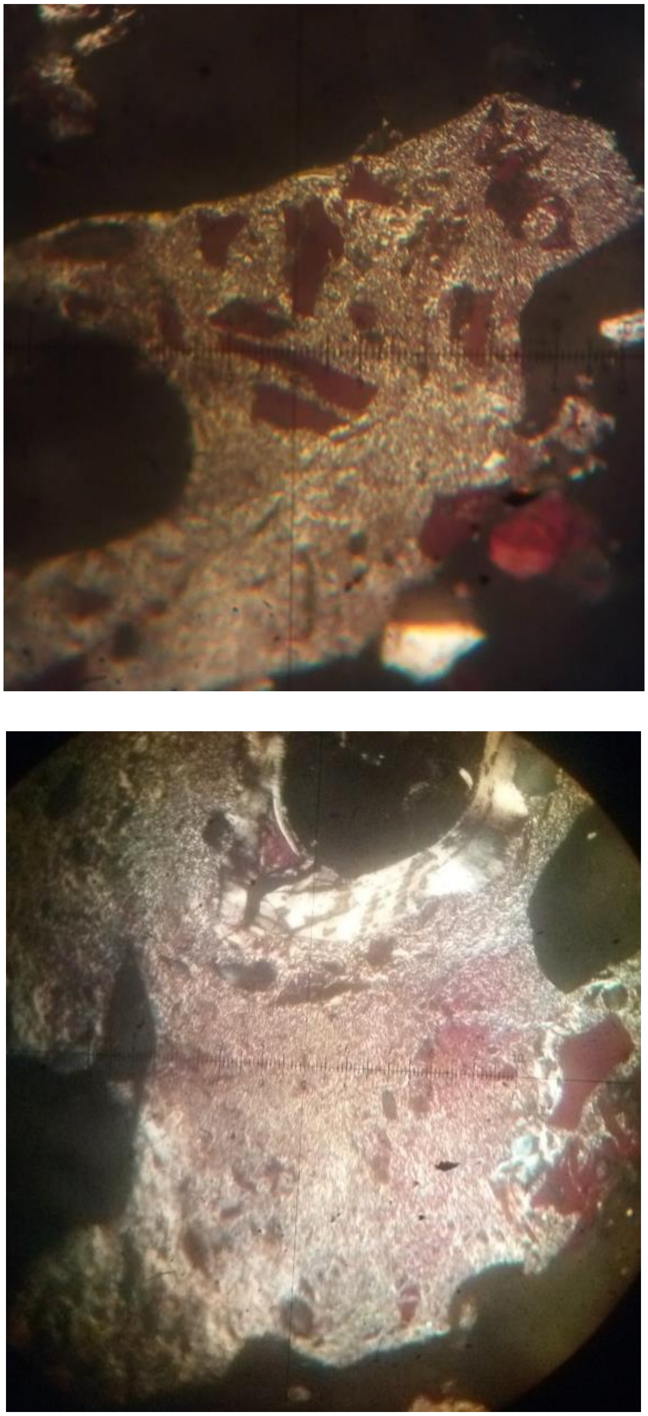


Figure 5-7 – Images of SHO Base Blend Coke Structure taken manually using microscope at Canmet with crosshairs visible

When evaluating the coke from the SHO tests that incorporated additive, it is important to understand what a biomaterial can look like. The biomaterials evaluated are noted as inert phases in the microscopic analysis with the most common counts in the coke products analysis showing properties of fusinite. This inert structure is cellular looking which is tubular in structure with degraded features. The coked biomaterials typically have a thinner-walled structure than a fusinite structure. The thinner cell walls of the biomaterials will allow for increased reaction zones for CO_2 which will also lower CSR (DeVanney , 2021). Figure 5-8 and Figure 5-9 give two different views of the cellular structures left behind after coking with a biomaterial. In the first figure, Figure 5-8, the long tubular openings can be seen in the lower half of the coke piece.

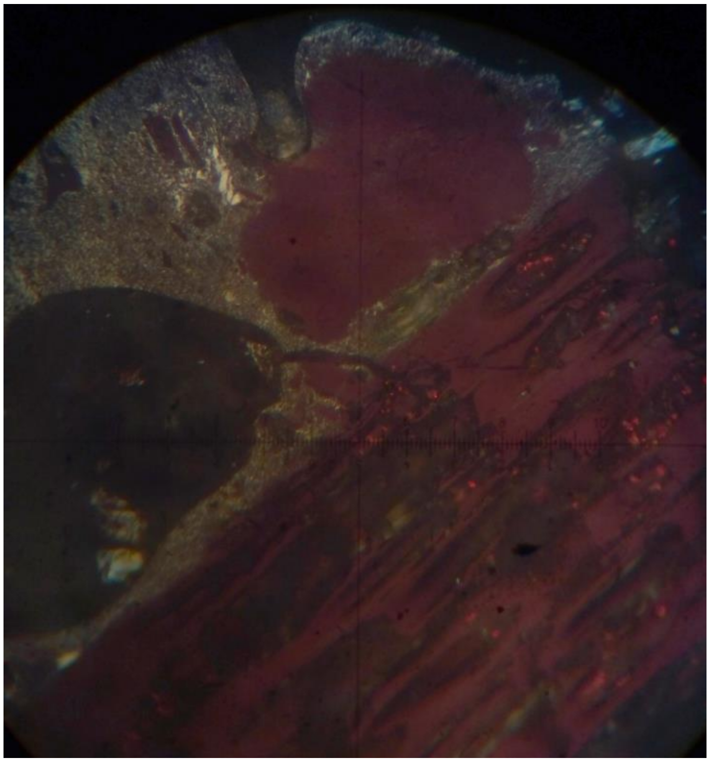


Figure 5-8- Manually Captured Image of SHO Biomaterial Additions in Coke Structure Showcasing the Tubular Structure of a Biomaterial Visible After Coking

In the second figure, Figure 5-9, a different plane of the material being cut shows the ends of the tubular structures. These structures were not seen in the base blend analysis.

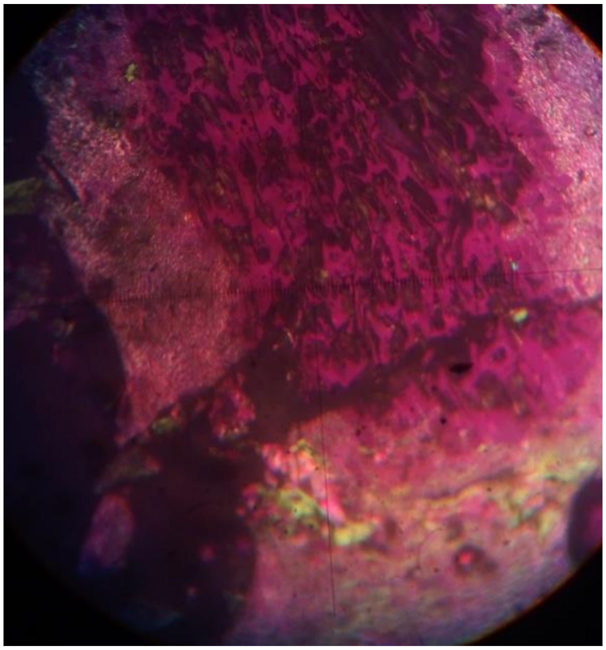


Figure 5-9 - Manually Captured Image SHO Showcasing the Tubular End Structure of a Biomaterial Additions in Coke Structure (Tinted)

The interaction of the inerts and biomaterial residues is different than the incorporated inerts of the base blend. There was notable interaction of the cellular structures of the biomaterials with mosaic coke textures. Some of the larger biomaterial deposits that were found looked as if they spread through the coke structures into the coke pore walls. Figure 5-10, shows the interaction of the fusinite inert of Material C in the centre of the coke structure with the mosaic textures on the left and domain textures on the bottom. There are notable pores and openings in these areas of texture changes that are not well bonded and could be sources of poor strength or potential fissure formation.

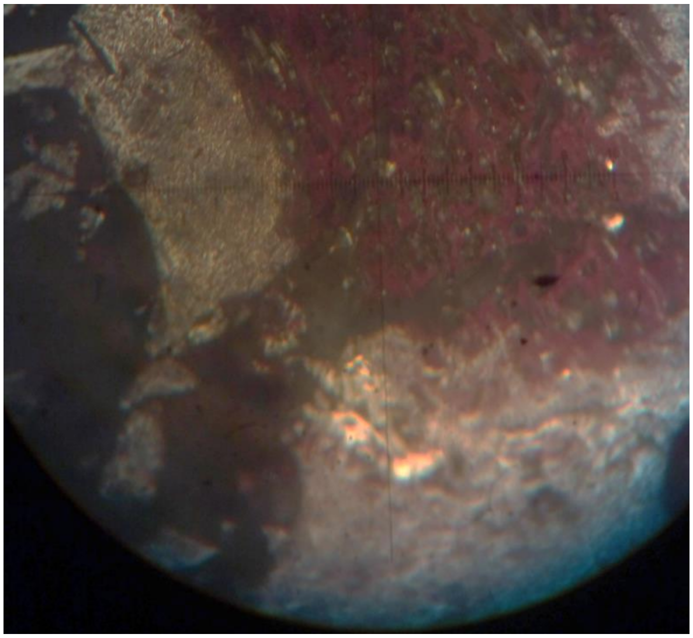


Figure 5-10 – Manually Capture Image of Coke Structure Bonding Interaction of Material C

There were interactions of the inert biomaterial with coke textures incorporated into it. As seen in Figure 5-11 with trial Material C3X the cellular structure of the inert material has notable mosaic coke texture incorporated into the porous structure in the lower quadrant.

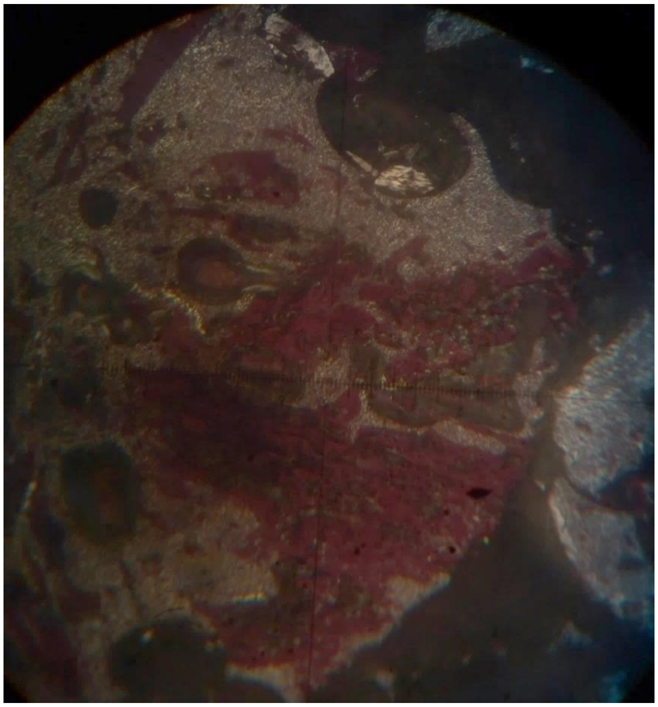


Figure 5-11 – Manually Capture Image of Mosaic Coke Texture Incorporated in Material C3X Inert

In some interactions of the biomaterials, it was found that the minerals from the biomaterial are affecting the coke structure rather than the biomaterial structure itself. As seen in Figure 5-12, the bright spots within the biomaterial structure are mineral matter. To the left of the cellular structure mosaic coke textures can be seen and domain coke texture on the bottom of the cellular structure. In the bottom left quadrant, the joints between the coke textures and the inert can be seen to be disrupted and cracked pore walls are observed compared to the smooth clean pore walls seen in the base blend textures. Minerals will affect the coal and lower the v-type assigned which can lower the classification of coke texture. If the minerals are not fused well into the reactive coke structures this will lower the CSR and increase the CRI.

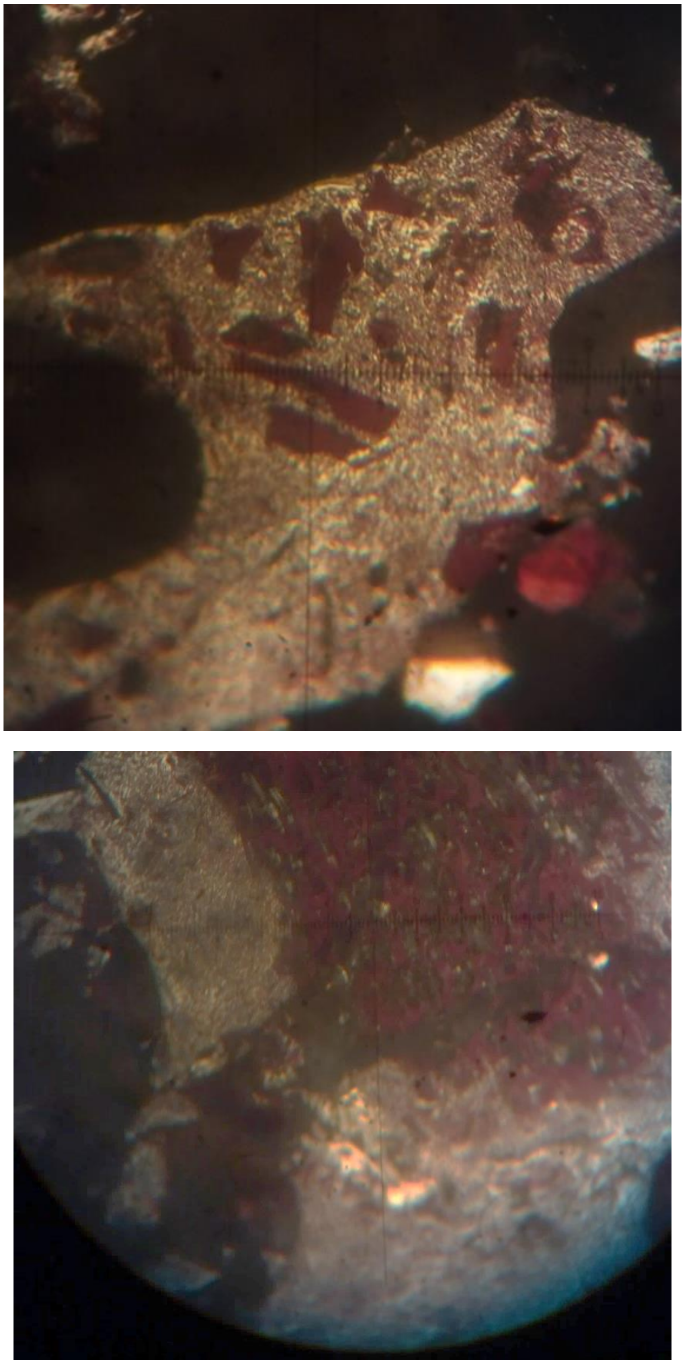


Figure 5-12 – Manually Capture Images of Base Blend Coke Structure (Top) Biomaterial Inert Mineral Matter in Coke Structure (Bottom)

Many of the observations of the biomaterial inerts present in the coke structures were found not to be well bound with the carbon forms. There were clear distinctions between the inerts of coal that were incorporated well and defined in the base blend compared to the biomaterial inert deposits that were degraded and not well fused to the carbon binders. There were increased levels of inert material found in the coke texture analysis of pellets with the biomaterial additions. Coked biomaterials can be misidentified as filler phase coke forms of inerts (DeVanney , 2021).

Figure 5-13 is an example of well incorporated inert materials in the base blend on the top and the defined cellular structure of the biomaterial in the middle of the coke structure on the bottom. The porous properties saw a fissure connecting a pore in the biomaterial that spreads into the coke textures above and below on the right side of the figure. Defining if the fissure started from this pore during coking or during crushing is not clear in this figure. The fissure could have occurred from the sample preparation for microscopic analysis. To identify whether a fissure started from the pore wall or the weaker structure of the biomaterial required identification where the thicker part of the fissure occurs as the starting point. An increased count of fissures and the size of them will correlate to a drop in CSR.

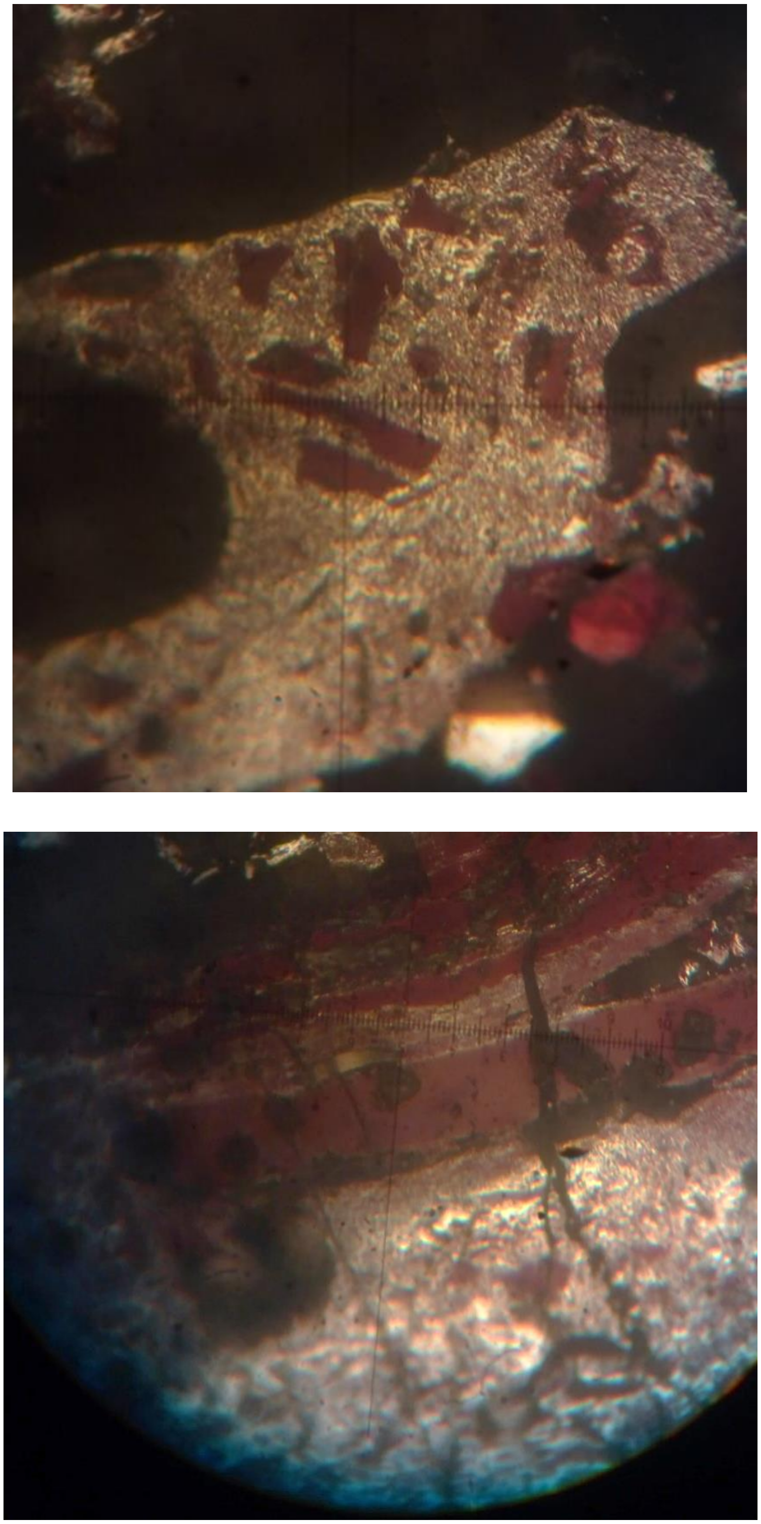


Figure 5-13 – Solid Base Blend Coke Structure (Top) Fissure Through Biomaterial and Coke Structure (Bottom)

In the texture analysis increased levels of lower rank binder phase, carbon forms were noted from the base blend. The material properties of the biomaterials in Table 4-6 and Table 4-7 show higher levels of oxygen compared to the base blend of coal. When coked, the increased levels of oxygen create an oxidization environment which would change the carbon forms in the area surrounding the biomaterial additions. The change in carbon form will result in a lower rank of binder phase to be found which will negatively impact the texture and lower the CSR of the coke produced. (DeVanney , 2021)

5.2.3. Sole Heated Oven (SHO) Texture Distribution Results

The distribution of the counts of the eight samples for texture analysis were conducted on the microscopic results. Similar to the results of the pilot oven tests, the three coke binder phases that saw the most significant change in distribution were isotropic, incipient, and circular. However, the medium mosaic coke texture of circular binder form is also included as it showed notable variation from the base blend as well. The full microscopic results can be found in Appendix C - SHO Test Coke Texture Results. The variation in coke texture change from the base blend is displayed in Table 5-6. A positive value is an increase in percentage from the base blend. In all additives, the largest variation of coke textures appeared in the circular binder phase and the least impacted were all in the isotropic binder phase.

Table 5-6 – Percent Variation in Coke Texture from Base Blend for SHO Tests

Binder Phase	**Isotropic**	**Incipient**	**Circular Anisotropic**	**Circular Anisotropic**
Coke Texture	**Isotropic**	**Very Fine Mosaic**	**Fine Mosaic**	**Medium Mosaic**
SHO Material A	-0.7	1.7	5.2	-9.8
SHO Material B	-0.70	2.40	10.3	-15.2
SHO Material C	-0.90	3.50	9.5	-14.2
SHO Material A3X	-0.70	4.70	16.2	-17.6
SHO Material C3X	-0.70	3.20	6.6	-11
SHO Material D	0.30	1.10	-0.6	-0.7
SHO Material E	-0.30	1.00	3.8	-4.4

Materials A, B, and C used in the initial pilot oven tests were tested again in the SHO. To compare to the multi-vari plot of the pilot oven results these three materials were replotted from the SHO results in Figure 5-14. All plots are comparing the percentage change observed in the coke analysis from the base blend. As seen from the plot there is little variation on the isotropic, but all saw a slight decrease. There is an increase in very fine mosaic textures in all three materials from the base blend with Material C having the largest increase. There is a notable increase in all materials in the fine mosaic with Material B having the largest change. x. There is a difference in results from the pilot oven texture multi-vari plot in Figure 5-4 as all three materials see a shift up in the average counts of these fine textures from the base blend. The difference is likely that all three materials were prepared to the correct size in the SHO trials unlike in the pilot oven trials where one material was larger due to an error. Very Fine Mosaic textures are associated with fair coking coal properties, so an increase is not as desirable as a decrease in isotropic coke textures which are from poor coking coals. Fine mosaic coke textures are from good coking coal properties, so an increase is desired over a decrease.

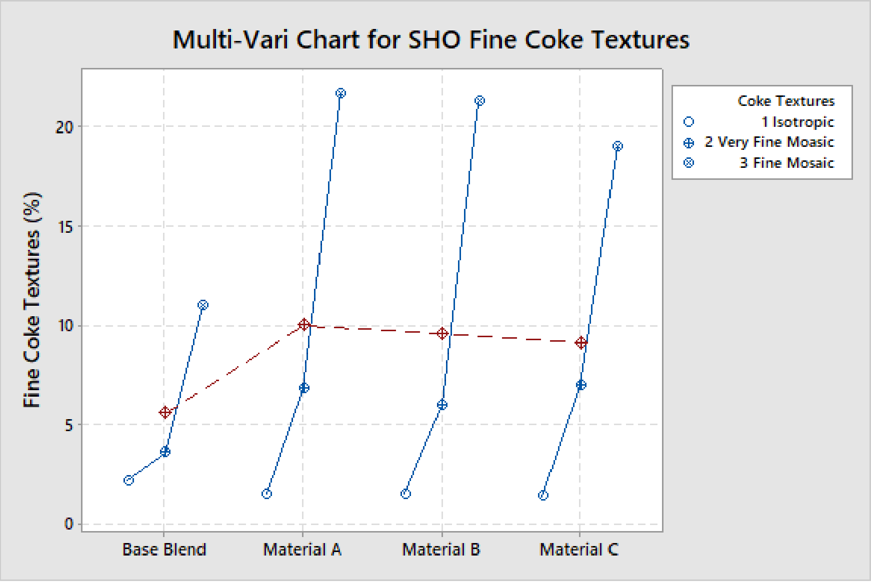


Figure 5-14 - SHO Multi-Vari Chart of Textures for Materials A, B and C

In the SHO texture distribution results, it was found that the medium mosaic results also saw significantly different amounts than the base blend. The multi-vari plot was created with the medium mosaic coke textures included and a notable decrease from the base blend can be seen in Figure 5-15. The variation in the circular anisotropic binder phase textures of the fine and medium mosaic are critical to coke production as these are where the good to excellent coking properties come from. A drop in these good coking properties could explain the observed drop in hot strength observed and the increase in CRI. The average of all samples noted by the red dotted line becomes less variable with the impact of the decrease in medium mosaic textures which are the opposite of increases in the finer textures.

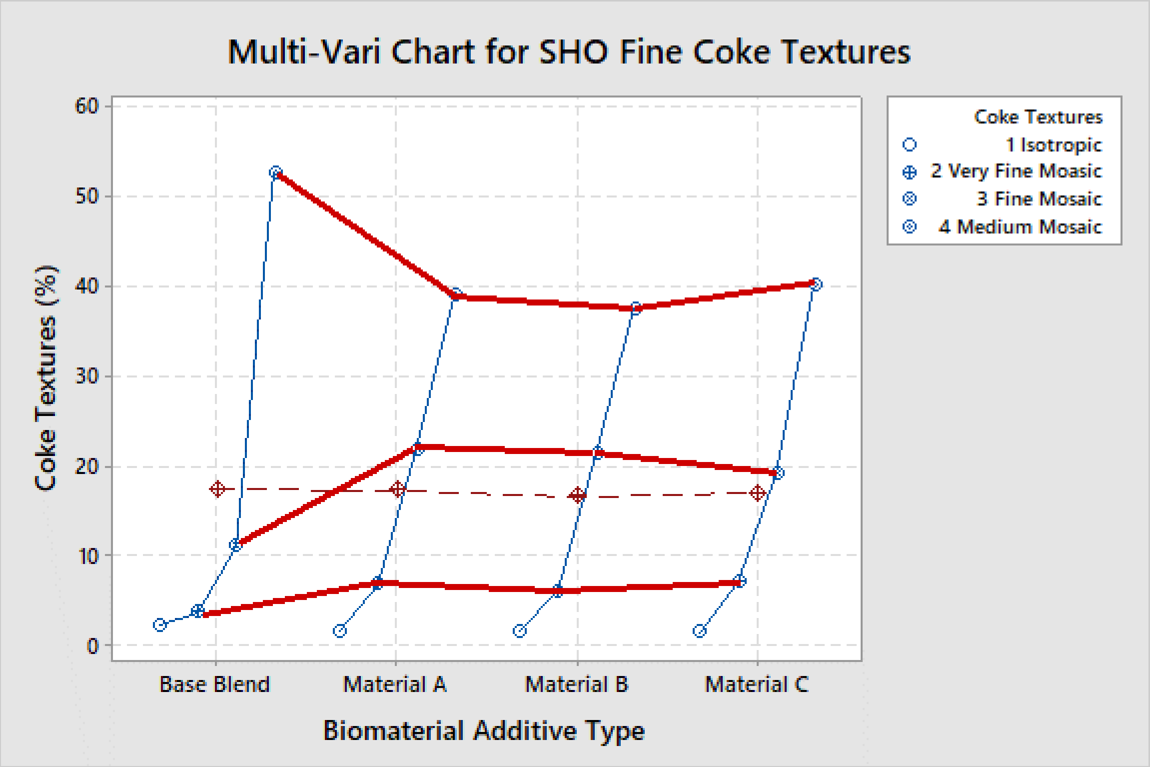


Figure 5-15- SHO Multi-Vari Chart of Fine Textures & Medium Mosaic for Materials A, B and C

In Table 5-4, the CSR variation from the base blend was presented to look at the changes of Materials A, B, and C from the main effects plot is presented in Figure 5-16. Like the pilot oven results, as the CSR decreases there is an increase in very fine textures which are more reactive carbon forms. The average decrease is a 4.5-point drop in CSR with Material A being above this average and Material C being below with the most negative impact on CSR. This is a similar trend found in the pilot oven test results plotted in Figure 5-5 where Material A was above the average and Materials B and C were below. This shows the increased size of Material A in the pilot oven test was not the only factor why Material A did not have as large of a CSR drop as Material B and C. However, it does confirm that the reduction in CSR increases as the processing of the biomaterial addition increases.

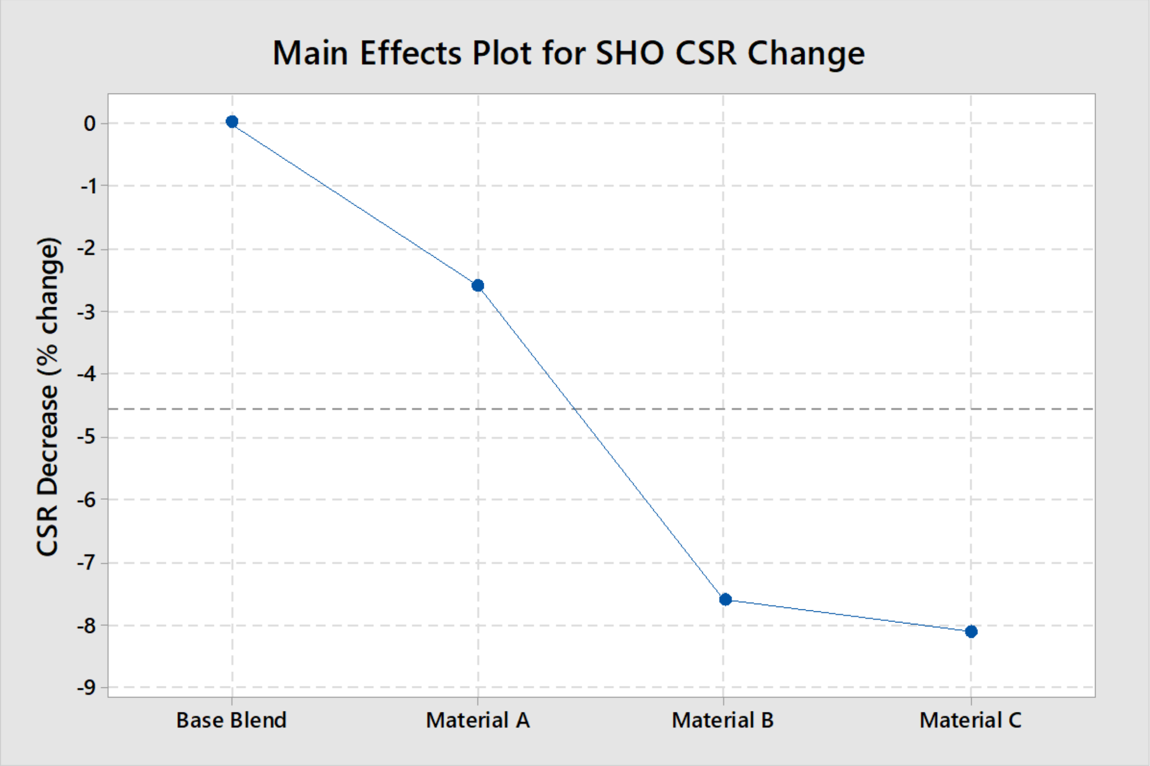


Figure 5-16- SHO CSR Variation from Base Blend

The main effect plot of CRI for the SHO tests with Materials A, B, and C is seen in Figure 5-17. The SHO has a similar trend from the pilot oven test plot in Figure 5-6. Both tests show that as the biomaterials processing incurred before addition increases the

CRI also increases. The SHO has a more step change increase in CRI increase than the pilot oven results did. This increase could be due to the change in reactivity in Material C having a bigger shift of very fine isotropic in the pilot oven results that was not seen in the SHO results. In the SHO results, Material B had the largest change from the base blend on CRI which could be related to the largest decrease in fine mosaic and coarse mosaic textures.

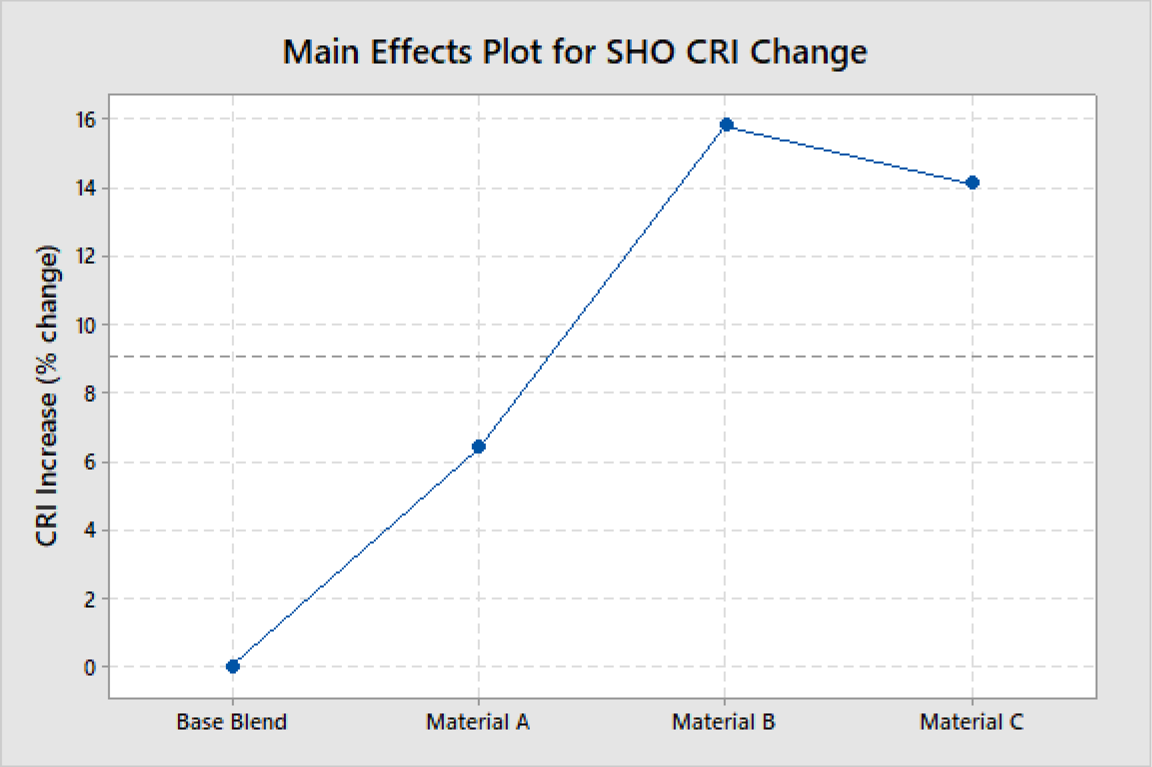


Figure 5-17 - SHO CRI Variation from Base Blend

The amount of biomaterials substituted in the base blend was selected by the industrial user. To confirm the understanding of how much material can be substituted in a coal blend without exceeding acceptable quality parameters Material A and C were tested with triple the amount (3X). Material A and C were selected as they have shown the least and most variation from the base blend in the work presented so far. The standard lower substitution (1X) amount used in all the pilot and SHO tests discussed is plotted in comparison to triple (3X) the amount of substitution in Figure 5-18. There is a noteworthy drop in CSR from the base blend in both Material A and Material C which justifies the amount selected for the test work. The drops in CSR aligns with the increase in fine and very fine mosaic textures observed in the analysis presented in Table 5-4. Material A saw a larger drop in quality when the substitution was tripled which could be due to the reactivity and higher volatile matter of this material. Material C shows that the decrease in CSR will occur regardless of the material added.

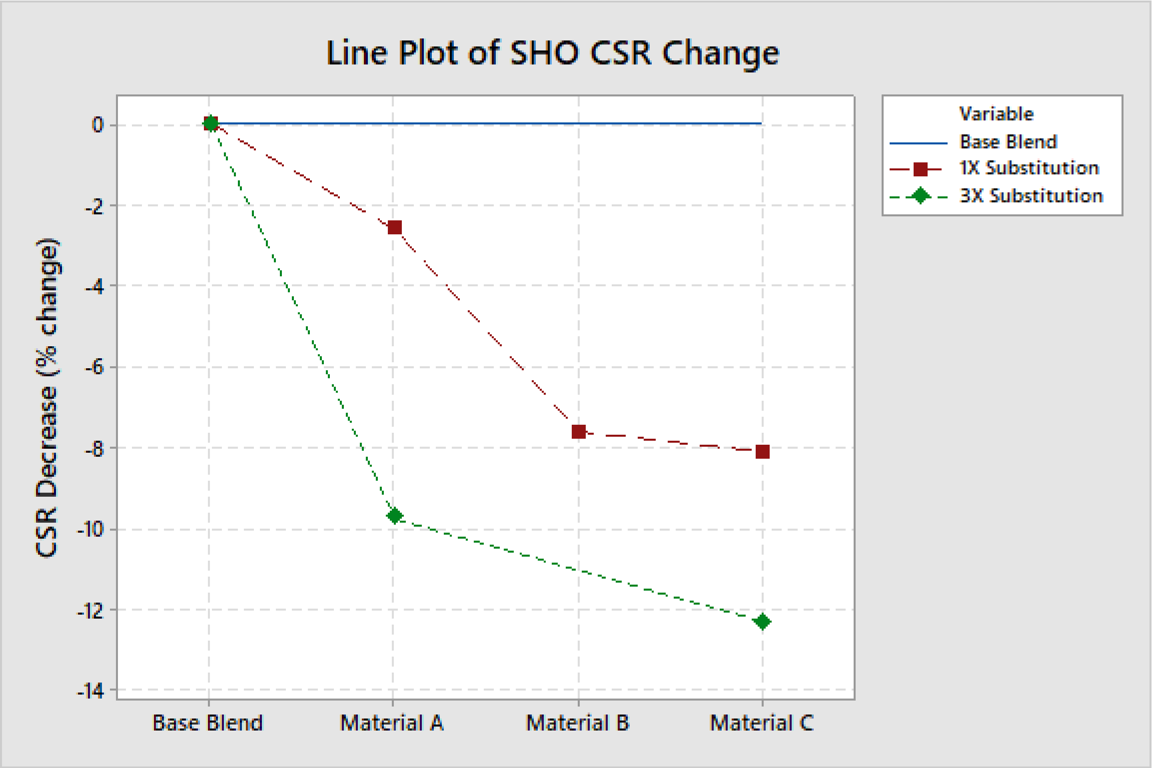


Figure 5-18- Comparison on Standard Substitution and Triple Substitution of Material A and C CSR Results

The main effect plot of CRI, as seen in Figure 5-19, verifies the inverse relationship of CSR and CRI. As the biomaterials increase in amount of processing they incurred before addition the CRI increases are amplified with the increase (3X) of substitution. The increase of finer textures aligns with the increase of CRI and CSR. The biomaterial substitution was tripled but would still be considered as low amounts, 2-10% substitution referenced in literature, as the tripled amount is less than 10% coal substitution (Thomas, McKnight, & Serrano, 2011).

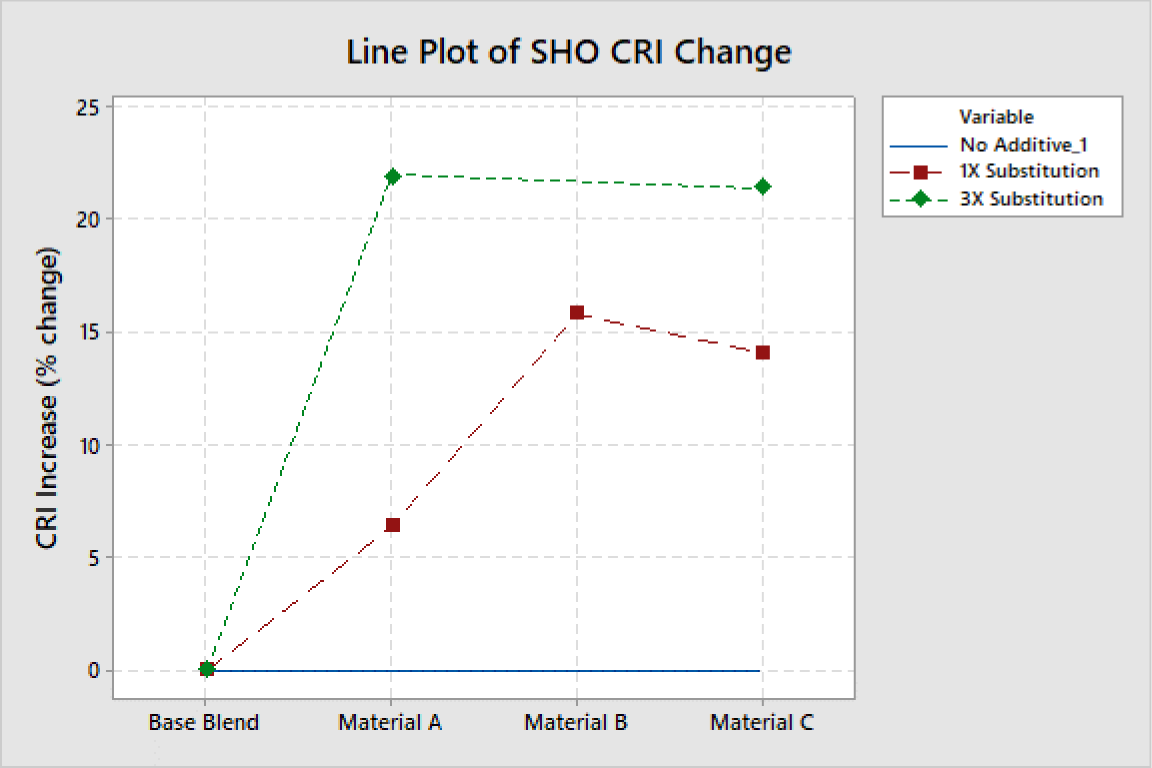


Figure 5-19 - Comparison on Standard Substitution and Triple Substitution of Material A and C CRI Results

Material D and Material E are not raw biomaterials which is beyond the initial scope of this work. They were tested for comparison in the changes in coke textures with a pyrolyzed biomaterial and cokemaking waste. The texture changes as shown in Table 5-6 did not see similar changes as the raw biomaterials for these two materials. The textures were not altered on the same magnitude, but the CSR and CRI were.

Material D saw the most significant drop of CSR of all materials tested in the SHO compared to the base blend at the same standard substitution amount (1X) as presented in Table 5-5. Material E did not see as large decrease in CSR as Material D but the variation in the fine coke textures was larger. A larger decrease in CSR of Material D was expected because of the size and reactivity of the pyrolyzed biomaterial. In the literature review, the work investigating loose pyrolyzed biomaterial in loose compared to a briquetted pyrolyzed biomaterial was reviewed. The study found the briquetted material did not impact the coke quality as significantly as loose and could be used at higher amounts. The advantages of the pyrolyzed biomaterials chemistry, volatile matter, carbon content, and higher reactivity can be limited to localized areas by briquetting without significant reduction in CSR (Ng, Huang, & Giroux, 2019). Running a SHO trial with a pyrolyzed biomaterial validated this when compared to the raw biomaterial trials.

Unlike raw biomaterials, there is ongoing research into the addition of pyrolyzed biomaterials for coal substitution in the industry. The results of the raw biomaterial substitutions at low amounts show that there are opportunities to have less impact on CSR reduction than the addition of low amount of loose pyrolyzed addition. A further advantage reviewed in the literature search is raw biomaterial additions require less processing, altering of the material and energy for production in order to be added to a coal blend. A review of the energy required for addition of the raw biomaterial compared to the higher energy needed to produce the substitutions of pyrolyzed material should be investigated.

5.2.4. Mineral Content Impact

In the texture analysis, the predicted coal reflectance can be calculated from the coke texture results and used to compare to the actual coal reflectance reading taken in the petrographic analysis. The reflectance calculated should be close to the reflectance reading of the coal which is a validation of the tests being conducted. The reflectance only accounts for the coal components of the sample so comparing to the coke predicted reflectance gives an insight into how the mineral content of the different additives and the hindrance on bonding good coke forms. Table 5-7 shows the difference of the SHO calculated reflectance from the base blend with a negative value being a decrease. With reflectance (Ro) there is a low tolerance of variation, all tests apart from Material B would fall within acceptable variation as seen in Table 5-7. This variation aligns with the notable difference in Material B having significantly higher ash content, mineral matter, than the other biomaterials substituted from Table 4-6. As noted in the coke texture optical analysis, increased mineral matter will negatively impact the bonding and strength of the produced coke.

Table 5-7- Calculated Ro Variation from SHO Base Blend

Test	**Coal Ro Calculated**
SHO Material A	0.00
SHO Material A3X	-0.02
SHO Material B	-0.06
SHO Material C	-0.01
SHO Material C3X	-0.01
SHO Material D	-0.01
SHO Material E	0.00

To further review the impact of the composition of the biomaterial on the coke quality, the ash composition analysis was conducted on the coke produced from each of the SHO tests. The variation in properties used to calculate the coke basicity index (CBI) using Equation 5 for each trial are presented in Table 5-8. A negative value is a decrease in percent amount from the base blend percent of the composition.

Equation 5 - Coke Basicity Index

$$Coke\ Basicity\ Index = \frac{\mathrm{N}a_2\mathrm{O}+K_2\mathrm{O} + \mathrm{CaO} + \mathrm{MgO} + \ \mathrm{Fe}_2O_3}{\mathrm{Si}O_2 + \mathrm{A}l_2O_3}$$

Table 5-8 - Variation from Base Blend of Coke Ash Composition from SHO tests

Ash Component	Na2O	K2O	CaO	MgO	Fe2O3	SiO2	Al2O3
SHO Material A	0.01	-0.05	0.5	0.18	0.5	-1.06	-0.62
SHO Material B	0.02	-0.03	0.35	0.06	0.11	-0.51	-0.28
SHO Material C	0	-0.06	0.88	0.38	0.08	-0.95	-1.04
SHO Material A3X	0.05	-0.07	0.96	0.46	0.59	-1.47	-1.41
SHO Material C3X	0.02	-0.15	1.9	0.9	0.22	-2.71	-2.15
SHO Material E	0	0.03	0.27	0.06	0.57	-0.65	-0.88
SHO Material D	-0.01	-0.2	0.13	-0.02	-0.24	3.89	-3.47

The coke basicity index was calculated for each test , as found in Table 5-9, using the coke basicity index (CBI) calculation of Equation 5. All tests used the same base blend of coals, the variation in coke ash basicity is a result of the material added. Table 4 6 and Table 4 7 capture the material properties of the added materials all with higher ash and the texture analysis all verified higher inerts than the base blend. The highest increase in CBI was found in the triple (3X) substitution trials of Materials A and C which further validates the standard lower substitution (1X) rate selected for the tests. All tests with the exception of Material E saw an increase in the CBI. This is expected from the drop in

CSR found in the tests from the base blend. Higher basicity and higher inerts will result in a lower CSR (Todoschuk, Price, & Gransden, 2004).

Table 5-9 - Coke Basicity Index of SHO Coke Tests

Test	**Coke Basicity Index**
Base Blend	0.156
Material A	0.174
Material B	0.164
Material C	0.176
Material A 3X	0.187
Material C 3X	0.203
Material D	0.171
Material E	0.152

All tests used the same base blend of coals, the variation in coke ash basicity is a result of the material added. Table 4-6 and Table 4-7 capture the material properties of the added materials all with higher ash and the texture analysis all verified higher inerts than the base blend. The highest increase in CBI was found in the triple (3X) substitution trials of Materials A and C which further validates the standard lower substitution (1X) rate selected for the tests.

6. Summary and Conclusions

Quality metallurgical coke will remain the primary reactant in steel production to convert iron ore to metallic iron in blast furnace operations for the foreseeable future. Using biomaterial carbon sources to substitute low levels of coal has the potential to offset GHG emissions and reduce cokemaking's overall carbon footprint. In this work, three raw biomaterials available to industrial users were evaluated for substitution of operational coal blends. The focus was to investigate the relationship between changes in coke texture and the decrease in coke strength after reaction (CSR). Data from a series of pilot oven and sole heated oven (SHO) tests found the following conclusions:

- When coal is substituted with low amounts of raw biomaterials, the most notable changes in coke texture were found in incipient and circular binder phases which are linked to lower quality coals.

- Analytical analysis of the coke texture changes saw that as the CSR decreased there was an increase in isotropic and very fine textures which are more reactive carbon forms.

- As the amount of processing of the substituted materials increased there was an increase in fine coke textures which is a negative impact and a reduction in medium coke textures which is also a negative change. These changes to the coke structure properties are different than those from the base coal blend.

- Mineral matter and chemical composition of the substituted biomaterial saw an increased negative impact on coke ash and a decrease in CSR results as the amount of processing on the raw biomaterial was increased.

- Increased oxygen in biomaterial additions negatively impacted the carbon form by lowering the rank of the binder phase due to localized oxidization which will lower the CSR of the coke produced.

- The selection of raw biomaterial for coal substitution needs to keep the inert and oxygen content as close to coal as possible. Of the three raw biomaterials evaluated, Material A had the least impact on coke quality.

- The negative changes in coke texture distribution, CSR, CRI, and coke ash composition found in the triple (3X) substitution trials of Materials A and C validated the standard lower substitution (1X) rate selected for the work.

Recommendations for further work include:

- Additional raw biomaterials that are available to industrial users and are similar in characteristics to Material A should be evaluated.
- Lab scale testing, both SHO and pilot oven, to evaluate the biomaterial microstructure interactions with different coal blends and individual coals should be considered.
- A review of the GHG generation from use of the different biomaterials should be investigated for Life Cycle Analysis (LCA).
- Yield impact of the biomaterial additions should be reviewed to justify the cost impact of coke quality reduction.
- Further research is warranted to study the impact of binder additives to biomaterial to improve the interaction and bonding to coke structures.

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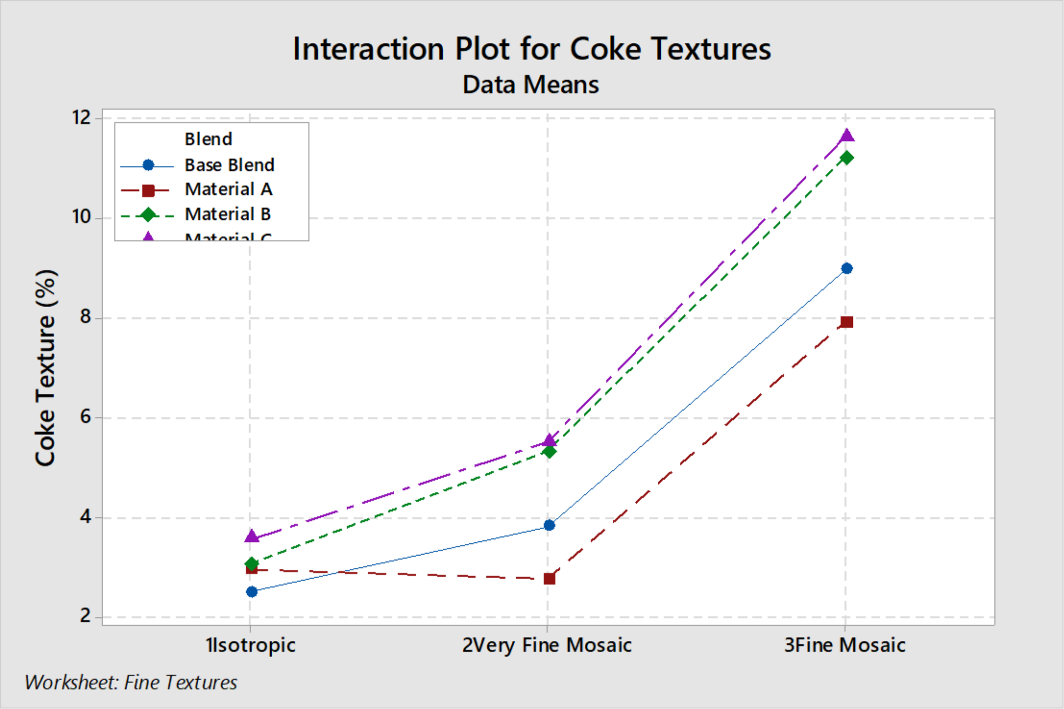
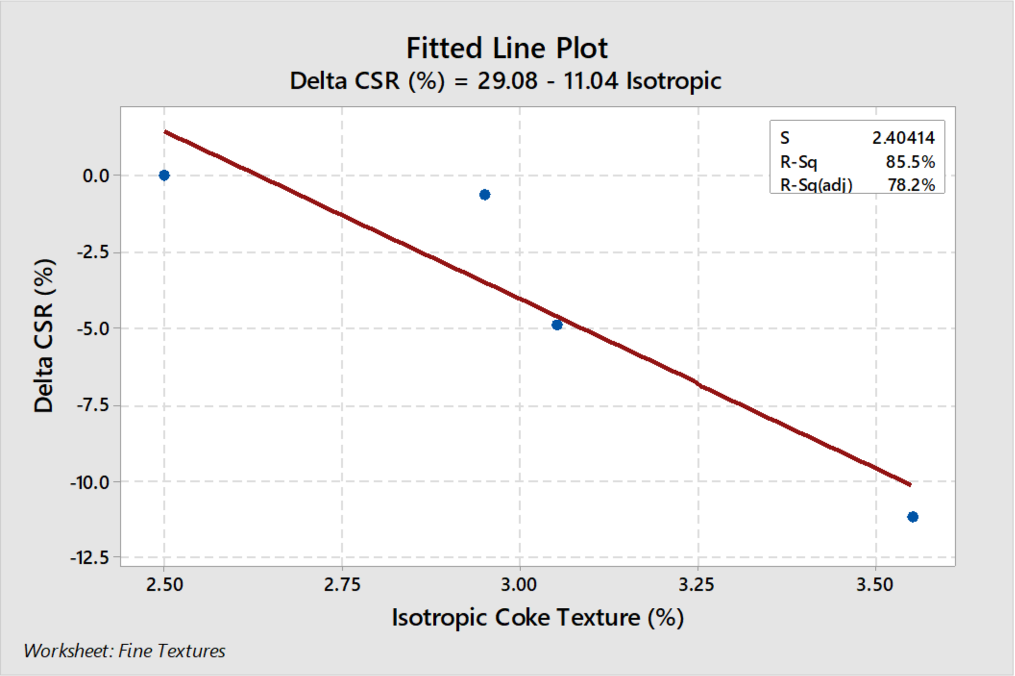
Appendix A – Pilot Test Coke Texture Results

Coke Texture Group	**Texture Identification**	**Base Blend 1**	**Base Blend 2**	**Material A1**	**Material A2**	**Material B1**	**Material B2**	**Material C1**	**Material C2**
Isotropic	Isotropic	2.60	2.40	3.00	2.90	3.10	3.00	3.10	4.00
Incipient	Very fine mosaic	3.30	4.30	2.40	3.10	7.60	3.00	5.30	5.70
Circular Anisotrpoic	fine mosaic	7.10	10.80	8.40	7.40	13.20	9.20	8.60	14.60
Circular Anisotrpoic	medium mosaic	47.50	46.00	41.80	43.10	38.00	42.30	43.00	35.50
Circular Anisotrpoic	coarse mosaic	11.50	11.30	13.90	16.20	11.70	16.20	11.70	11.00
Circular Anisotrpoic	Total mosaic	69.40	72.40	66.50	69.80	70.50	70.70	68.60	66.80
Lenticular Anisotropic	elongated fine flow	9.30	7.90	8.40	9.30	8.20	8.10	7.00	9.00
Lenticular Anisotropic	elongated medium flow	2.50	1.50	3.00	3.20	2.20	3.40	3.10	3.20
Lenticular Anisotropic	elongated coarse flow	1.10	0.70	2.20	1.50	0.90	1.50	1.10	1.40
Lenticular Anisotropic	Total flow	12.90	10.10	13.60	14.00	11.30	13.00	11.20	13.60
Ribbon Anisotropic	domain flat flow	5.60	4.10	5.20	3.90	5.90	4.20	6.50	5.70
Ribbon Anisotropic	domain undulating	2.30	1.40	2.30	1.70	1.90	1.40	2.10	1.80
Ribbon Anisotropic	domain ribbon	3.50	4.10	5.70	3.20	3.30	3.20	4.90	4.60
Ribbon Anisotropic	Total Domain	11.40	9.60	13.20	8.80	11.10	8.80	13.50	12.10
Inerts	fusinite	1.60	2.00	1.20	1.20	1.40	1.10	1.40	0.90
Inerts	semifusinite	1.30	2.00	1.70	2.30	1.60	2.40	1.10	1.10
Inerts	unidentified inerts	0.80	1.50	0.80	1.00	1.00	1.00	1.10	1.50
Inerts	altered vitrinite	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00

Appendix B – Additional Plots of Pilot Oven Texture Analysis

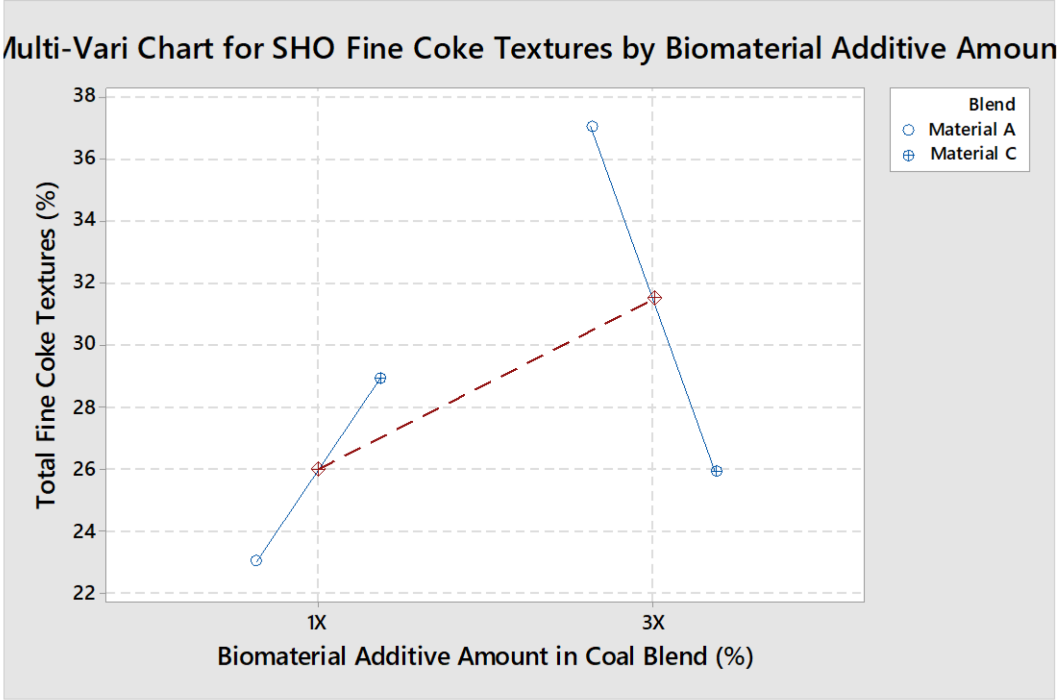


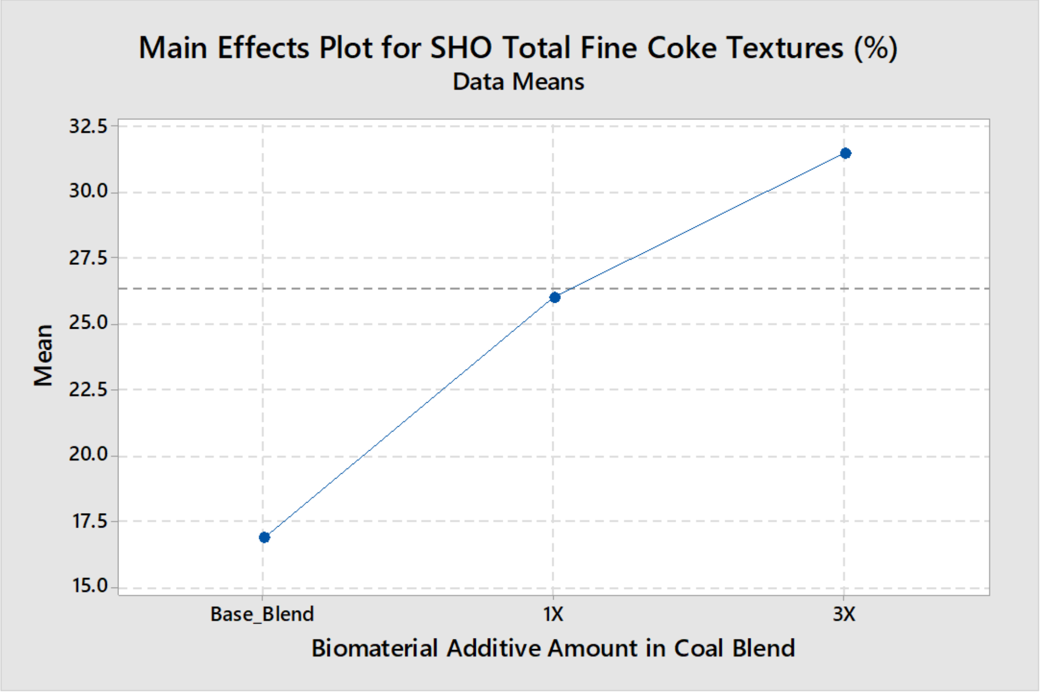


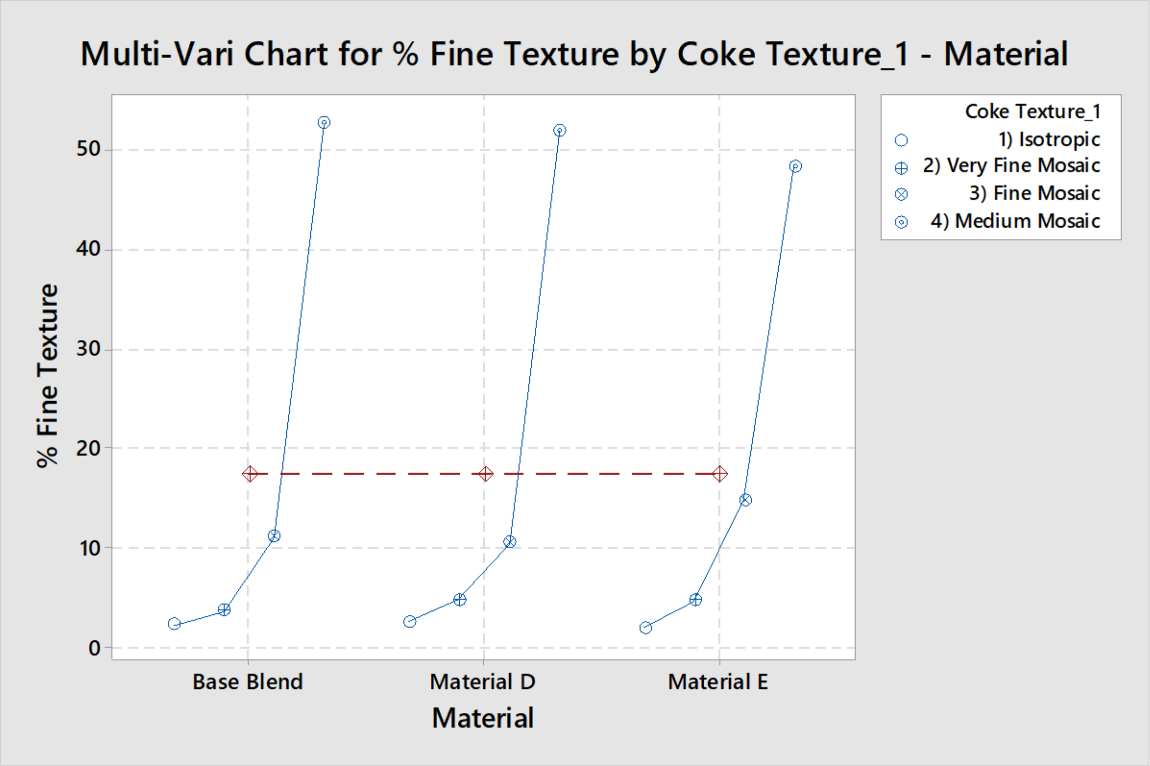
Appendix C - SHO Test Coke Texture Results

Coke Texture Group	**Texture Identification**	**Base Blend**	**Material A**	**Material A3X**	**Material B**	**Material C**	**Material C3X**	**Material D**	**Material E**
Isotropic	Isotropic	2.2	1.5	1.5	1.5	1.3	1.5	2.5	1.9
Incipient	Very fine mosaic	3.6	5.3	8.3	6	7.1	6.8	4.7	4.6
Circular Anisotrpoic	fine mosaic	11	16.2	27.2	21.3	20.5	17.6	10.4	14.8
Circular Anisotrpoic	medium mosaic	52.6	42.8	35	37.4	38.4	41.6	51.9	48.2
Circular Anisotrpoic	coarse mosaic	7	5	3.2	3.6	5.2	5	7.1	3.7
Circular Anisotrpoic	Total mosaic	74.2	69.3	73.7	68.3	71.2	71	74.1	71.3
Lenticular Anisotropic	elongated fine flow	6.6	4.9	3.1	3.6	4.3	4.7	3.8	4.6
Lenticular Anisotropic	elongated medium flow	2	2.3	1.3	2	1.8	2.8	1.9	2.3
Lenticular Anisotropic	elongated coarse flow	1.6	1.9	0.5	1.3	1	2.3	1.2	1
Lenticular Anisotropic	Total flow	10.2	9.1	4.9	6.9	7.1	9.8	6.9	7.9
Ribbon Anisotropic	domain flat flow	1.4	3.5	2.8	2.8	2.8	2	3.8	2.8
Ribbon Anisotropic	domain undulating	2	2.6	1.5	5	2.8	1.9	0.8	3.3
Ribbon Anisotropic	domain ribbon	5.2	6.6	6.6	7	7.1	7.3	5.3	6.6
Ribbon Anisotropic	Total Domain	8.6	12.7	10.9	14.8	12.7	11.2	9.9	12.7
Inerts	fusinite	2.4	4.1	3.8	4.6	4.7	5.4	3.5	2.6
Inerts	semifusinite	2	2.3	4.5	3.1	2.1	0.7	2.1	3
Inerts	unidentified inerts	0.6	1	0.7	0.8	0.9	0.4	1	0.6
Inerts	altered vitrinite	0	0	0	0	0	0	0	0

Appendix D – Additional SHO Texture Analysis






Multi-Vari Chart for % Fine Texture by Coke Texture_1 - Material
% Fine Texture
0
10
20
30
40
50
Base Blend
Material D
Material E
Material
Coke Texture_1
1) Isotropic
2) Very Fine Mosaic
3) Fine Mosaic
4) Medium Mosaic