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Hydrothermally Treated Coals for Pulverized Coal Injection

Final Technical Report

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EXECUTIVE SUMMARY

This project investigated the suitability of hydrothermally dried low-rank coals for pulverized fuel injection into blast furnaces in order to reduce coke consumption. Coal samples from the Beluga coalfield and the Usibelli Coal Mine, Alaska, were used for the study. Crushed coal samples were hydrothermally treated at three temperatures, 275, 300 and 325°C, for residence times of 10, 60 and 120 minutes. Products were characterized to determine their suitability for pulverized coal injection. Characterization included proximate and ultimate analyses, vitrinite reflectance and TGA reactivity. A literature survey was also conducted.

Characterization data confirm findings of earlier hydrothermal drying studies of Alaskan low-rank coals conducted by MIRL:

1. The degree of upgrading is affected by both process temperature and residence time, but process temperature has a greater influence than residence time on low-rank coal upgrading.
2. The upgrading takes place rapidly. Most occurs in the first 10-20 minutes of residence time and the rate of upgrading decreases thereafter. Significant upgrading can still take place given long enough residence time.
3. Beyond 40-60 minutes, the upgrading response versus residence time tends to level off.
4. Upgrading response increases linearly with increases in process temperature within the temperature range of 275-325°C.
5. Vitrinite reflectance increases significantly from the raw coal to the hydrothermally dried products, indicating significant structural changes, i.e. induced coalification has occurred during the hydrothermal drying process.
6. Small, laboratory scale, tubing reactor hydrothermal drying studies predict pilot plant performance well.

Isothermal reactivity of the char was then determined at 1000°C in a gas mixture consisting of 10.09% CO_2, 2.06% O_2 and 87.85% N_2, which simulates conditions in the raceway of a typical blast furnace during combustion of pulverized coal. Examination of the data reveals an increase in TGA reactivity for the coal after hydrothermal treatment, which is attributed to the increased porosity and permeability of the char, due to hydrothermal treatment. Reactivity of the char obtained from raw Usibelli coal was increased from 1.96 wt. % loss per minute to over 2.32 wt. % loss per minute, when the coal was hydrothermally treated at 325° C for 120 minutes. Beluga char reactivity was increased from 1.53 wt. % loss per minute for the raw coal to above 1.70 wt. % loss per minute, when the coal was hydrothermally treated at 325° C for 120 minutes. Reactivities of hydrothermally treated low-rank coal chars from this study were approximately twice those of Western British Columbian bituminous coals presently being used for PCI.

After only a 10 minute residence time, at all three treatment temperatures, a very significant improvement in coal quality was achieved. Ash levels remain essentially unchanged; the apparent increase in ash content above that of the raw coal being due to the dramatic decrease in equilibrium moisture (37% - 53%). Calorific values increased by 14% to 26% on an equilibrium moisture basis. After an initial increase above the raw coal level, volatile matter steadily decreased with increasing process temperature, while there was a steady increase in fixed carbon levels.

This project has been unable to obtain thermochemical modeling data, for the hydrothermally processed low-rank coal, from US Steel to date, despite assurances from US Steel's Mr. Tom Oshnock that the modeling data would be forthcoming. Should this data be received in the future, an addendum to this final report will be submitted.

Attempts to solicit Japanese and Korean interest in hydrothermally treated low-rank coal for PCI applications were also unsuccessful. None of the project's inquiries received replies, despite the assistance of Mr. John Sims, Vice-President of Marketing, Usibelli Coal Mine, Inc.

While there is significant upgrading of low-rank coal by hydrothermal treatment, these products have;

1) Considerably higher volatile matter content,
2) Considerably higher oxygen content,
3) Considerably higher moisture level,
4) Lower carbon content, and

5) Lower calorific value,

than coals presently being used for PCI. Their ash levels are comparable to coals used for PCI, while their sulfur levels are superior. Despite their lower carbon content and calorific value, the high volatile matter and high reactivity of the hydrothemally treated low-rank coals may have advantages for high rate PCI applications. These same properties may also prove attractive for blending opportunities with low volatile coals. The higher moisture levels of hydrothermally treated low-rank coal could be further reduced during pulverization and storage in the pulverized coal reservoir ahead of PCI into the blast furnace.

Based on these results, the following are recommended:

1) Thermochemical modeling of hydrothermally treated low-rank coal for PCI applications.
2) Bench scale testing of hydrothermally treated low-rank coal for PCI applications by U.S. and Pacific Rim steel companies.
3) Additional studies to define the effect of hydrothermally treatment of low-rank coal on grindability (HGI).
4) Bench scale testing of hydrothermally treated low-rank coal to define its oxidative and physical stability.
5) Use of a fast-flow reactor to evaluate the reactivity of hydrothermally treated low-rank coal char at higher temperatures (1200 - 2000° C) to simulate blast furnace raceway conditions.

I. INTRODUCTION

Injection of pulverized coal into blast furnaces to reduce coke requirements has been investigated in the past and serious trials took place in the 1960s. However, the cost of fitting blast furnaces with pulverized fuel systems and the ready availability of low cost oil at that time, encouraged Japanese steel plants to substitute oil for coke. The shortage of oil during the 1979 oil crisis forced steel industries to take a serious look at replacing oil with pulverized coals. The steel industry soon discovered the advantages of pulverized coal injection (PCI). Large scale conversions to PCI were made both in Japan and Europe.[1,2,3,4] By 1991, 26 of 33 operating blast furnaces in Japan were equipped with PCI systems, consuming 6 million tons of pulverized coal. Worldwide use of PCI was 13.2 million tons in 1990 and is expected to grow to 25 million tons by 1995. The current practice of pulverized coal injection at the Kakogawa No. 2 blast furnace, operated by Kobe Steel, uses coke (289 kg per ton of hot metal produced, kg/t) along with pulverized coal (129 kg/t) and oil (65 kg/t).[5]

Coal injection rates are limited by:[1,6]

1. Reactivity of injected coal
2. Chemical composition of injected coal
3. Particle size of injected coal
4. Gas distribution in the burden column
5. Furnace permeability

A proposed upper limit for pulverized coal injection, which would burn completely in the tuyere combustion zone, was estimated at 180 kg/t for a total fuel rate of 500 kg/t by Tamura, et al. in 1991[7], but that figure has been exceeded by US Steel (230 kg/t PCI rate for a total fuel rate of 555 kg/t).[1] Contemporary research activities by steel producers are targeting PCI rates up to 300 kg/t.[8] An upper limit for pulverized coal grain size of 0.7 mm has been proposed.[7] Calculations based on furnace trials have shown that pulverized coal can replace up to 30-50% of the coke previously used in blast furnaces employing oxygen enriched blast, without impairing productivity or hot metal quality.

The high price of coke has resulted in progressive substitution of coke by low cost pulverized coal. PCI combustion is differentiated from normal coal combustion by:[9]

1. High combustion zone temperatures; 1200 to 2000°C.
2. Short residence times in the presence of oxygen; less than 30 ms.
3. Gasification in the presence of CO_2.

Research conducted at the University of Alaska Fairbanks has shown that hydrothermal treatment of subbituminous 'C' coals can produce a high quality product suited for metallurgical use.[10] The hydrothermally dried product has low moisture, very low sulfur, 0.2%, and is highly reactive. This project addresses the suitability of hydrothermally processed low-rank coals (LRCs) for metallurgical use as pulverized coal for injection into blast furnaces.

The fact that Alaska has over 5 trillion tons of low sulfur coal reserves and that each trillion tons has an energy equivalency of approximately 5,500 years of Alyeska pipeline production (1.5 MM barrels/day) demonstrates the enormous economic potential for Alaska's coal resources (Figure 1). This includes over 4 trillion tons of bituminous and subbituminous coal located in the Northern Alaska basin, north of the Brooks Range. The Usibelli Coal Mine, the only current operating coal mine in Alaska, has an annual production of about 1.5 million tons. Half of this is exported to Korea while the remainder supplies heat and power for interior Alaska. The Beluga coal field, adjacent to Cook Inlet, is readily accessible to ice-free tide water. The field has been extensively drilled and evaluated by Placer Dome Inc. and Diamond Alaska Coal Co.

Coals from Usibelli Coal Mine and the Beluga coal field are of subbituminous 'C' rank and typically have calorific values of 8,000 Btu/lb. The moisture of these coals is about 28%. The combination of high moisture and low heating value has restricted most low-rank coal usage worldwide to mine-mouth power generation, which in turn has limited export sales of Alaskan coal to only 0.75 MM tons per year from Usibelli Coal Mine. This comes at a time when Australian coal exports have topped 100 MM tons per year and the steam coal market is

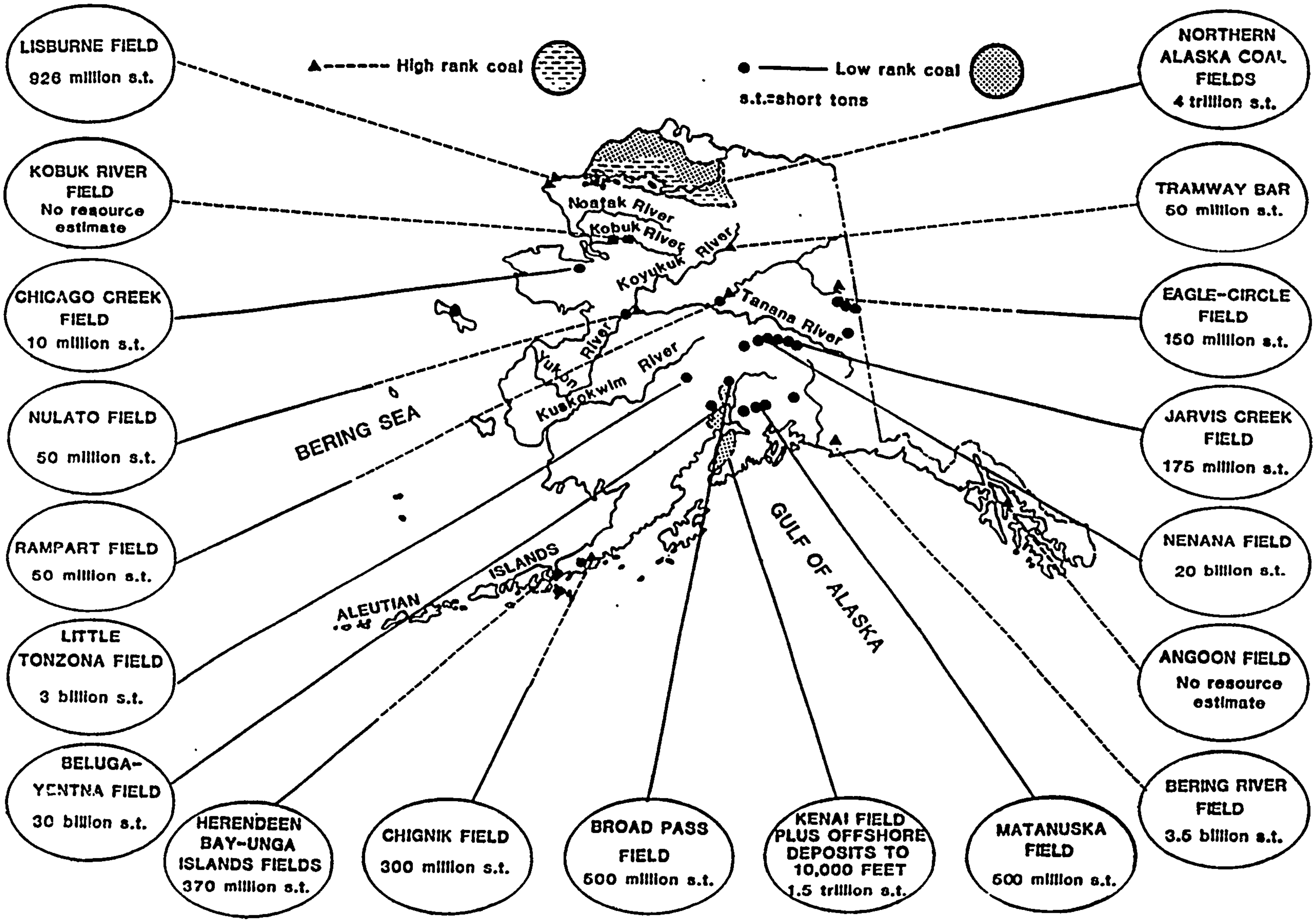

Figure 1. Coal resources of Alaska.

expected to more than double in this decade. Alaskan LRCs can participate in the Pacific Rim coal market by suitable upgrading to enhance quality.

While the objective of this project is applied research directed at Alaskan low-rank coal, the larger goal is to increase the available market for our state's and nation's coal resources. The relevance of such research is highlighted by the recent difficulty Usibelli Coal Mine, Inc. experienced in renegotiating its 1990-1995 coal export contracts with KEPCO, a Korean electric utility. Technology and sound engineering management must be applied to enhance the marketability of Alaska's low-rank coals. One potential market for hydrothermally dried, Alaskan, low-rank coals is pulverized coal injection.

Fischer and Schrader first studied hydrothermal treatment of coals in 1921.[11] The process involves heating coals to a temperature of 320° to 400°C in the presence of water in an autoclave. Koppleman's process, used in the K-fuels system, treats LRC's at temperatures of 500° to 600°C and high pressure. The process liberates moisture as well as light hydrocarbon gases and liquids. The process developed by University of North Dakota and Bechtel uses temperatures less than 400°C.[11] The gases liberated principally consist of CO_2. Some tars are mobilized and these tend to condense on the surfaces of coal particles and in pores.[10]

Beginning in 1989, the Mineral Industry Research Laboratory (MIRL), University of Alaska Fairbanks, undertook a preliminary investigation of hydrothermal treatment of Alaskan low-rank coals to 1) determine the quality of the product that can be prepared, and 2) study petrologic changes that accompany hydrothermal treatment that will eventually influence the mechanical properties, surface area and product reactivity. The products obtained during these tests were surprisingly different from conventional, evaporatively dried LRCs. Expected, as well as surprising characteristics are summarized below:[10-13]

1. As expected, the products had low equilibrium moistures and enhanced heating value.
2. Although rank enhancement was expected, vitrinite reflectance increased from 0.25% to over 0.7%, placing the product in a high volatile bituminous range.

3. Improvement in grindability was expected, but surprisingly the HGI increased from 34 for the raw coal up to 115 for the products.

4. Most surprising was the physical property of the product. The coals went through a plastic stage and the resulting products resembled coke produced from coking coal. Beluga coal showed the most thermoplastic behavior upon hydrothermal treatment.

Sectioning of the dried particles showed that the particles were devoid of cracks. The size distribution of cells in the product varied and could be related to petrology. Most significant of all, was that the surface of each particle so fused, that the presence of the inner cell structure is only revealed upon sectioning.[10] The density of the particles varied with the severity of hydrothermal treatment. Some particles have densities as low as 0.75 compared to 1.49 for the raw coal.[10] Of special significance is the fact that although the processed particles are very porous, the fused skin strength of the particle is high and may allow handling without significant degradation.

The high reactivity and porosity of the LRC products, coupled with low ash and sulfur, may make them unique raw materials for numerous metallurgical applications. It was therefore important to characterize the hydrothermally dried low-rank coal products for possible applications, which until now have not been considered for Alaskan coals. Among these alternative applications, pulverized coal injection appears to offer promise, and was the focus of this project.

II. PROJECT SUMMARY

This project investigated the suitability of hydrothermally processed low-rank coal for PCI. Coal samples from the Beluga coalfield and the Usibelli Coal Mine, Alaska, were collected and used in this study. A literature survey (bibliography included in appendix) emphasizing coal characteristics required for PCI into blast furnaces and a thorough review of previous hydrothermal drying research at MIRL were conducted.

4 x 8 mesh, crushed coal samples from the Beluga coalfield and the Usibelli Coal Mine, Alaska, were hydrothermally treated at three temperatures, 275, 300 and 325°C and three

residence times, 10, 60 and 120 minutes. Hydrothermal treatment was conducted using a reactor system similar to the one designed by Youtcheff.[14] The system provides excellent isothermality and accurate residence time measurements. The system consists of three components: 1) a stainless steel tubing reactor, 2) a fluidized sand bath for heating the reactor, and 3) an agitation device and holder for the reactor.

Two identical tubing reactors were used. Each reactor has a 75 ml capacity and is made of 28 mm outside diameter, stainless steel tubing. Two Swagelok threaded plugs cap the end fittings. A 6 mm hole is bored at the midpoint of the reactor and tubing is welded in a vertical position to support a fitting and 3000 psi Bourdon gauge. The reactors can be removed from their holder quickly, so that a rapid quench is attainable.

Hydrothermally dried products were filtered, dried and characterized using proximate and ultimate analyses, vitrinite reflectance and TGA reactivity. This characterization data was to be used to model hydrothermally treated low-rank coals' suitability for PCI using a thermochemical model developed by US Steel.

Letters were written and sent to PCI users in Japan and Korea, as well as to Japan's Center for Coal Utilization (International Cooperation Dept.) to solicit their interest in hydrothermally treated low-rank coal for PCI applications.

III. RESULTS AND DISCUSSION

Tables 1 and 2 show the proximate and ultimate analyses data for hydrothermally processed Usibelli and Beluga coals, respectively. Sulfur values of the hydrothermally dried products were, for all practical purposes, unchanged from those of the raw coals, remaining at approximately 0.2%. Tables 3 and 4 present TGA reactivity and vitrinite reflectance values for the treated coals. These data confirm earlier findings of hydrothermal drying studies of Alaskan low-rank coals conducted by MIRL[13]:

1. The degree of upgrading is affected by both process temperature and residence time, but process temperature has a greater influence than residence time on low-rank coal upgrading.

Table 1. Analyses of hydrothermally dried 4 x 8 mesh coal, No. 4 Seam Usibelli Coal Mine (all data expressed on a dry, ash-free basis except for equilibrium moisture, which is expressed on a raw coal basis).

PROCESS TEMPERATURE=275°C

Residence Time, minutes	Equilibrium Moisture %	Volatile Matter %	Fixed Carbon %	Calorific Value Btu/lb	C, %	H, %	N, %	O, %
Raw Coal	25.5	53.2	46.8	11980	67.9	4.9	1.2	25.8
10	15.8	50.1	49.9	12040	69.5	4.9	1.3	24.1
	16.2	52.2	47.8	12265	69.6	5.0	1.2	24.0
60	15.0	49.6	50.4	12485	71.1	5.0	1.3	22.4
	14.1	49.8	50.2	12605	71.5	5.0	1.3	22.0
120	12.4	48.9	51.1	12785	72.0	5.0	1.4	21.5
	14.0	49.1	50.9	12780	72.0	5.0	1.3	21.4

PROCESS TEMPERATURE=300°C

Residence Time, minutes	Equilibrium Moisture %	Volatile Matter %	Fixed Carbon %	Calorific Value Btu/lb	C, %	H, %	N, %	O, %
10	14.3	50.0	50.0	12535	70.7	5.0	1.2	22.9
	14.0	49.8	50.2	12580	70.2	5.1	1.2	23.2
60	11.3	48.0	52.0	12975	73.4	5.1	1.3	19.9
	9.9	47.4	52.6	13065	73.5	5.0	1.3	19.9
120	9.1	45.4	54.6	13200	74.2	5.1	1.3	19.2
	9.8	46.6	53.4	13130	74.1	5.1	1.3	19.3

PROCESS TEMPERATURE=325°C

Residence Time, minutes	Equilibrium Moisture %	Volatile Matter %	Fixed Carbon %	Calorific Value Btu/lb	C, %	H, %	N, %	O, %
10	11.6	48.5	51.5	12960	73.4	5.0	1.3	20.1
	13.4	48.2	51.8	12890	72.6	5.0	1.2	21.0
60	9.3	44.5	55.5	13285	75.3	5.0	1.4	18.2
	8.6	43.8	56.2	13290	75.0	5.0	1.4	18.4
120	7.6	42.3	57.7	13450	76.2	5.0	1.3	17.3
	9.0	43.9	56.1	13480	76.2	5.0	1.4	17.1

Table 2. Analyses of hydrothermally dried 4 x 8 mesh Beluga Coal (all data expressed on a dry, ash-free basis except for equilibrium moisture, which is expressed on a raw coal basis).

PROCESS TEMPERATURE=275°C

Residence Time, minutes	Equilibrium Moisture %	Volatile Matter %	Fixed Carbon %	Calorific Value Btu/lb	C, %	H, %	N, %	O, %
Raw Coal	23.8	52.8	47.2	12075	68.7	5.1	1.4	24.6
10	12.0	48.1	51.9	12645	71.5	5.2	1.4	21.8
	13.2	49.9	50.1	12405	70.5	5.1	1.3	22.9
60	12.6	47.4	52.6	12555	71.4	5.1	1.4	21.9
	14.2	48.6	51.4	12315	69.8	5.1	1.3	23.7
120	10.9	48.7	51.3	12630	72.0	5.1	1.4	21.2
	12.0	46.9	53.1	12695	71.7	5.0	1.4	21.7

PROCESS TEMPERATURE=300°C

Residence Time, minutes	Equilibrium Moisture %	Volatile Matter %	Fixed Carbon %	Calorific Value Btu/lb	C, %	H, %	N, %	O, %
10	11.0	49.2	50.8	12465	71.1	5.4	1.6	21.8
	13.3	48.1	51.9	12535	71.3	5.0	1.4	22.2
60	10.8	46.2	53.8	12810	72.9	5.0	1.4	20.5
	9.9	45.7	54.3	13045	73.8	5.2	1.4	19.5
120	9.8	46.1	53.9	13080	74.4	5.4	1.7	18.3
	10.5	44.7	55.3	13020	74.0	5.2	1.4	19.2

PROCESS TEMPERATURE=325°C

Residence Time, minutes	Equilibrium Moisture %	Volatile Matter %	Fixed Carbon %	Calorific Value Btu/lb	C, %	H, %	N, %	O, %
10	10.4	47.2	52.8	12805	73.2	5.0	1.4	20.2
	11.8	47.2	52.8	12680	72.2	5.2	1.4	21.0
60	10.5	44.0	56.0	13155	74.7	5.0	1.5	18.6
	9.9	43.2	56.8	13305	75.0	5.2	1.5	18.0
120	8.0	43.4	56.6	13385	76.3	5.1	1.5	17.0
	8.1	43.3	56.7	13410	76.5	5.1	1.4	16.7

Table 3. Ulminite reflectance and TGA reactivity of raw and hydrothermally dried Usibelli coal.

Process Temperature °C	Residence Time Minutes	Ulminite Reflectance $\bar{R}_{max}$, oil	Reactivity Wt.% loss/min
275	10	0.42	2.03
	60	0.54	2.09
	120	0.60	2.14
300	10	0.48	2.12
	60	0.60	2.21
	120	0.65	2.18
325	10	0.60	2.13
	60	0.77	2.25
	120	0.86	2.32
Raw Coal		0.30	1.96

Table 4. Ulminite reflectance and TGA reactivity of raw and hydrothermally dried Beluga coal.

Process Temperature °C	Residence Time Minutes	Ulminite Reflectance $\bar{R}_{max}$, oil	Reactivity Wt.% loss/min
275	10	0.48	1.65
	60	0.64	1.66
	120	0.72	1.69
300	10	0.60	1.62
	60	0.84	1.66
	120	0.88	1.70
325	10	0.72	1.68
	60	0.94	1.71
	120	0.98	1.70
Raw Coal		0.32	1.53

2. The upgrading takes place rapidly. Most occurs in the first 10-20 minutes of residence time and the rate of upgrading decreases thereafter. Significant upgrading can still take place given long enough residence time.
3. Beyond 40-60 minutes, the upgrading response versus residence time tends to level off.
4. Upgrading response increases linearly with increases in process temperature within the temperature range of 275-325°C.
5. Vitrinite reflectance increases significantly from the raw coal to the hydrothermally dried products, indicating significant structural changes, i.e. induced coalification has occurred during the hydrothermal drying process.
6. Small, laboratory scale, tubing reactor hydrothermal drying studies predict pilot plant performance well.

TGA reactivity studies were conducted using a Perkin-Elmer TGA, Series VII. Char from the raw coals and their hydrothermally dried products were obtained by heating 10 mg of coal in the TGA under a nitrogen atmosphere from room temperature to 1000°C at a scan rate of 20°C/min. Isothermal reactivity of the char was then determined at 1000°C in a gas mixture consisting of 10.09% CO_2, 2.06% O_2 and 87.85% N_2, which simulates conditions in the raceway of a typical blast furnace during combustion of pulverized coal. Tables 3 and 4 summarize the TGA reactivity data for the Usibelli and Beluga coals, respectively. Examination of the data reveals an increase in TGA reactivity for the coal after hydrothermal treatment, which is attributed to the increased porosity and permeability of the char, due to hydrothermal treatment. Reactivity of the char obtained from raw Usibelli coal was increased from 1.96 wt. % loss per minute to over 2.32 wt. % loss per minute, when the coal was hydrothermally treated at 325° C for 120 minutes. Beluga char reactivity was increased from 1.53 wt. % loss per minute for the raw coal to above 1.70 wt. % loss per minute, when the coal was hydrothermally treated at 325° C for 120 minutes. For comparison, two Western British Columbian bituminous coals, which are presently used for PCI, have reactivities of 0.88 - 0.90 wt. % loss per minute, and metallurgical coke, produced from a Western British Columbian bituminous coal has a reactivity of 0.73 wt. % loss per minute.[15] Reactivities of hydrothermally treated low-rank coal chars from this study were approximately twice those of Western British Columbian bituminous coals presently being used for PCI.

Vitrinite material (ulminite, telecollinite, desmocollinite) in low-rank coals is present in several forms as precursors to vitrinite in high-rank coals. Although vitrinite reflectance is not the best indicator of rank for low-rank coals, it is a very good indicator of structural changes accompanying thermal treatment of coals. In this study, ulminite and telecollinite were chosen as indicators. Mean, maximum reflectance was measured in oil. Ulminite reflectance is found to be very sensitive to process conditions. Ulminite reflectance for Usibelli coal increased from 0.30% in the raw coal to 0.86% for coal treated at 325°C for 120 minutes (Table 3). These changes in ulminite reflectance are indicative of induced coalification, i.e. rank progression, by the hydrothermal drying process. The data show that reflectances for Usibelli coal of 0.60% can be achieved by a treatment time of 120 minutes at 275°C or 10 minutes at 325°C. Treatment times can therefore be dramatically reduced by increasing the temperature of treatment. Beluga coal showed similar reflectance changes.

Table 5 compares raw Usibelli coal to its hot water dried products produced at the various process temperatures at a 10 minute residence time. After only a 10 minute residence time, a very significant improvement in coal quality was achieved. Ash levels remain essentially unchanged; the apparent increase in ash content above that of the raw coal being due to the dramatic decrease in equilibrium moisture (37% - 51%). Calorific values increased by 14% to 26% on an equilibrium moisture basis. After an initial increase above the raw coal level, volatile matter steadily decreased with increasing process temperature, while there was a steady increase in fixed carbon levels.

Table 6 compares raw Beluga coal to its hot water dried products produced at the various process temperatures at a 10 minute residence time. After only a 10 minute residence time, a very significant improvement in coal quality was achieved. Ash levels remain essentially unchanged; the apparent increase in ash content above that of the raw coal being due to the dramatic decrease in equilibrium moisture (47% - 53%). Calorific values increased by 15% to

Table 5. Improvement in Usibelli coal quality after hydrothermal drying (equilibrium moisture basis).

Property	Raw Coal	HWD PROCESS TEMPERATURE, °C (10 minute residence time) 275	300	325
Equilibrium moisture, %	25.5	16.0	14.1	12.5
Ash, %	4.6	5.5	5.8	6.2
Volatile matter, %	37.1	40.2	39.9	39.3
Fixed carbon, %	32.7	38.4	40.1	42.0
Fuel Ratio	0.9	1.0	1.0	1.1
Calorific Value, Btu/lb	8,365	9,545	10,050	10,505

Table 6. Improvement in Beluga coal quality after hydrothermal drying (equilibrium moisture basis).

Property	Raw Coal	HWD PROCESS TEMPERATURE, °C (10 minute residence time) 275	300	325
Equilibrium moisture, %	23.8	12.6	12.2	11.1
Ash, %	7.8	10.2	11.7	11.1
Volatile matter, %	36.1	37.8	37.0	36.7
Fixed carbon, %	32.3	39.4	39.1	41.1
Fuel Ratio	0.9	1.0	1.1	1.1
Calorific Value, Btu/lb	8260	9670	9520	9910

20% on an equilibrium moisture basis. After an initial increase above the raw coal level, volatile matter steadily decreased with increasing process temperature, while there was a steady increase in fixed carbon levels.

In contemplating the industrial scale production of hydrothermally treated lump coal from raw low-rank coal, a cost-benefit analysis of operating parameters versus product marketability would be helpful. While lower process temperatures would allow lower process pressures and enable lower capital costs to be realized, higher temperatures obviously produce a higher quality product. Process residence time and reactor volume would also need to be considered. Induced coalification is seen even at the lowest temperature (275°C) and a short residence time (10 minutes), yielding a hydrothermally dried Usibelli coal product with 37% lower equilibrium moisture and 14% higher calorific value and a Beluga coal product with 47% lower equilibrium moisture and 17% higher calorific value. Detailed studies would be required to demonstrate that hydrothermally treated lump coal has the physical and oxidative stability to withstand the rigors of transportation.

This project has been unable to obtain thermochemical modeling data, for the hydrothermally processed low-rank coal, from US Steel to date, despite assurances from US Steel's Mr. Tom Oshnock that the modeling data would be forthcoming. Should this data be received in the future, an addendum to this final report will be submitted.

Attempts to solicit Japanese and Korean interest in hydrothermally treated low-rank coal for PCI applications were also unsuccessful. None of the project's inquiries received replies, despite the assistance of Mr. John Sims, Vice-President of Marketing, Usibelli Coal Mine, Inc.

Review and analysis of the characterization data show hydrothermally dried, Alaskan low-rank coals have characteristics, which are comparable to those of coals presently being utilized for PCI. Prasad, Chatterjee and Mukherje of Tata Steel in India have published the data shown in Tables 7, for characteristics of two coals used for PCI at Tata Steel.[16] They note that while the ash content of coals used for PCI should be as low as possible, a wide range of coals from lignite to anthracite have been successfully injected, with volatile matter ranging from 5-

Table 7. Characteristics of Coals Used for Injection at Tata Steel, India.[16]

Parameter	South Black Water	West Bokaro
Proximate Analysis (db),		
Ash	9-10	17-18
VM	24-26	26-28
Ultimate Analysis (Typical), %		
Ash	10.1	18.30
C	77.72	71.47
H	4.22	4.24
N	1.75	1.63
O	5.73	3.75
S	0.48	0.61
Alkalis ($Na_2O + K_2O$), %	0.25-0.27	0.25-0.35
Initial Ash Fusion Temp., °C	1170	1250
HGI	55-65	60-70
Calorific Value, kcal/kg (Btu/lb)	7250 (13,050)	6820 (12,275)
Tar Yield, kg/t	31.5	39.85

Table 8. Specification of Coal for Blast Furnace Injection at Tata Steel.[16]

Moisture	: 4% max. at 40°C and 60% R.H.
Ash	: 10% max. for imported coal
	: 12% max. for Indian washed coal
	: 16% max. for Indian ROM coal
VM	: 25-30% on dry basis
Gieseler Fluidity	: 15-20 ddpm
CSN	: 2 max.
HGI	: 60 min.
Initial Ash Fusion Temp	: 1200-1250°C
Calorific Value	: 6000-7000 kcal/kg (10,800-12,600 Btu/lb)

50%. They have also published Table 8, listing the specifications of coals suitable for PCI base on Tata Steel's experience.[16]

Table 9 shows characteristics of 21 coals used for PCI as reported by Brouwer of Hoogovens.[3] Brouwer notes that mainly medium to high volatile bituminous coals, with ash levels of ≤ 7.5%, have been used. His data show that the highest coke replacement factors occur for high carbon and low ash, low moisture coals. Their injected coal characteristics showed the following ranges (dry basis): moisture, 4.6-12.6%; ash, 3.7-10.8%; sulfur, 0.5-1.1%; carbon, 73.6-84.9%; oxygen, 2.8-9.7%; and volatile matter, 17.5-39.3%. While calorific values are not noted, a range of 12,000-14,000 Btu/lb (dry basis) seems reasonable for medium to high volatile bituminous coals.

For comparison, characteristics of hydrothermally treated low-rank coals from Usibelli Coal Mine and Beluga coalfield showed the following ranges, respectively (dry basis): moisture, 8-16% and 8-12%; ash, 6-8% and 10-17%; sulfur, 0.17-0.21% and 0.13-0.24%; carbon, 63-71% and 61-66%; oxygen, 16-23% and 14-22%; volatile matter, 39-49% and 37-44% and calorific value, 11,260-12,460 Btu/lb and 11,000-11,400 Btu/lb. While there is significant upgrading of low-rank coal by hydrothermal treatment, these products have higher volatile matter, oxygen and moisture levels and lower carbon and calorific values than typical coals used for PCI. Hydrothemally treated low-rank coals ash levels were comparable to coals presently being used for PCI, while their sulfur values were superior. Table 10 presents ash analyses for Usibelli and Beluga coals from previous MIRL characterization research.

Despite the lower carbon content and calorific value of the hydrothermally treated low-rank coals, their higher volatile matter and reactivity may have advantages for PCI applications. In 1992, Yamagata, et al.,[6] of Japan's Sumitomo Metal Industries, Ltd., published work that investigated high PCI rates of up to 300 kg/t, using coals with volatile matter levels ranging from 19-45%. They found:

> "With the decrease of VM content, the combustibility in the raceway decreases and at PCI rate of 300 kg/THM the combustibility at the raceway boundary is reduced as low as 60%. .....the ignition of low VM coal is delayed and the combustion in the

Table 9. Main Characteristics of Injected Coals; Hoogovens.[3]

Coal number	Volatile matter (d.b.)	Ash (d.b.)	S (d.b.)	HGI	Ultimate analysis				Replacement factor (%) Coke/Coal	Moisture			
					C (d.b.)	H (d.b.)	N (d.b.)	O (d.b.)		Total	Inherent	Evaporated	Pulverized coal
1	36.6	3.7	0.97	51	81.1	5.3	1.3	7.7	91.20	7.7	2.3	6.0	1.5
2	32.6	5.8	0.88	62	81.2	5.1	1.4	5.6	93.41	5.7	1.4	4.7	0.9
3	31.8	6.3	0.88	62	81.6	5.1	1.5	4.6	94.98	6.5	1.0	5.8	0.7
4	34.7	6.3	0.67	61	78.3	5.1	1.2	8.5	87.22	8.3	2.6	6.0	1.8
5	33.0	4.7	0.73	56	83.0	5.0	1.4	5.3	95.82	4.8	1.3	3.9	0.9
6	32.5	7.5	0.62	46	77.8	4.8	1.6	7.7	86.62	6.6	2.6	4.9	1.7
7	32.5	5.7	0.75	59	81.3	5.2	1.4	5.7	93.93	5.0	1.2	4.2	0.8
8	35.5	6.3	0.74	45	77.7	5.0	1.5	8.8	85.73	5.6	2.7	3.8	1.8
9	33.8	5.7	0.66	48	80.4	4.7	1.4	7.1	89.48	8.7	2.1	6.0	2.2
10	31.3	8.5	1.07	75	78.7	5.2	1.5	5.0	88.35	10.2	1.2	6.0	3.7
11	30.5	5.4	0.79	60	82.6	5.1	1.4	4.7	95.99	5.3	1.1	4.6	0.7
12	17.5	5.6	0.79	100	84.9	4.4	1.4	2.9	97.84	8.5	0.7	6.0	2.0
13	33.9	5.7	0.88	58	82.1	5.1	1.3	4.9	95.43	6.5	1.0	5.8	0.7
14	31.6	6.1	0.94	61	82.3	4.8	1.3	4.6	95.29	4.6	1.2	3.8	0.8
15	33.3	5.5	0.78	62	82.2	5.1	1.3	5.1	95.34	6.0	1.2	5.2	0.8
16	39.3	5.9	0.64	54	78.0	5.0	1.3	9.3	82.79	11.4	7.1	6.0	4.9
17	33.8	9.2	0.67	57	76.8	5.4	1.3	6.6	87.78	7.3	2.4	5.7	1.6
18	32.7	10.8	0.54	55	73.6	4.4	1.5	9.2	79.52	8.8	3.6	6.0	2.4
19	24.4	8.3	0.87	80	81.8	4.7	1.5	2.8	93.62	9.7	1.2	6.0	3.2
20	30.7	8.0	0.62	58	77.7	4.4	0.7	8.6	84.40	8.9	4.8	5.7	3.2
21	30.7	8.6	0.54	50	76.4	4.1	0.7	9.7	78.69	12.6	7.1	6.0	6.1

Evaporated moisture = total moisture — 2/3 inherent moisture (maximum = 6 %)
Pulverized coal moisture = total moisture - evaporated moisture.
Replacement factor coke/coal = 2 % C + 2.5 % H + 0.9 % ash
Pulverized coal moisture : 86 %
(d.b. = dry basis, sur sec).

Table 10. Concentration of Major Elements in Coal Ash, percent.

Coalfield	SiO_2	$A1_2O_3$	Fe_2O_3	MgO	CaO	Na_2O	K_2O	TiO_2	SO_3	MnO	P_2O_5
Usibelli Coal Mine No. 3 Seam	34.5	14.7	8.1	3.7	30.5	0.6	1.0	0.6	3.2	0.1	0.1
Usibelli Coal Mine No. 4 Seam	38.4	17.7	6.7	3.5	23.8	0.2	1.1	0.7	5.1	0.1	0.2
Beluga, Chuitna (estimated, as-mined quality)[17]	41.0	20.2	8.9	4.1	13.4	1.6	1.1	1.3	2.5	0.1	1.0

> raceway is slow. As a result, the difference of the combustibility at the middle of the raceway increases as large as 50%. ... On the other hand, the combustibility of the injected PC measured at 700 mm above the tuyere level was at a high level of over 95% irrespective to the VM content."

Korthos,[8] in his dissertation from Aachen, Germany's Institute of Ferrous Metallurgy, postulated for coal particle sizes less than 0.1 mm, that is, within the size range of PCI applications, that only the volatile matter content is relevant for the degree of combustion. Tamura, et al., [7] of Nippon Steel have published an equation based on experimentation and modeling of PCI, that shows increased oxygen levels in the coal increase the maximum rate of PCI achievable.

Thus the high volatile matter content and reactivity of the hydrothermally treated low-rank coals may have PCI application advantages, especially at high PCI rates. They may also prove useful for blending with low volatile coals to produce blends mores suitable for PCI.

The higher moisture levels of hydrothermally treated low-rank coal could be further reduced during pulverization and storage in the pulverized coal reservoir ahead of PCI into the blast furnace. Moisture reduction for high rank coals during pulverization and storage ahead of PCI has been documented by Brouwer.[3] However, according to Brouwer, coals with an evaporable moisture content of more than 6% will decrease pulverizer capacity, in order to avoid surface moisture beyond the pulverized coal reservoir.

IV. CONCLUSIONS AND RECOMMENDATIONS

While there is significant upgrading of low-rank coal by hydrothermal treatment, these products have;

1) Considerably higher volatile matter content,
2) Considerably higher oxygen content,
3) Considerably higher moisture level,
4) Lower carbon content, and
5) Lower calorific value,

than coals presently being used for PCI. Their ash levels are comparable to coals used for PCI, while their sulfur levels are superior. Despite their lower carbon and calorific value, the high volatile matter and high reactivity of the hydrothemally treated low-rank coals may have advantages for high rate PCI applications. These same properties may also prove attractive for blending opportunities with low volatile matter content coals.

Based on these results, the following are recommended:

1) Thermochemical modeling of hydrothermally treated low-rank coal for PCI applications.
2) Bench scale testing of hydrothermally treated low-rank coal for PCI applications by U.S. and Pacific Rim steel companies.
3) Additional studies to define the effect of hydrothermally treatment of low-rank coal on grindability (HGI).
4) Bench scale testing of hydrothermally treated low-rank coal to define its oxidative and physical stability.
5) Use of a fast-flow reactor to evaluate the reactivity of hydrothermally treated low-rank coal char at higher temperatures (1200 - 2000° C) to simulate blast furnace raceway conditions.

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VI. APPENDIX

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