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Executive Summary

This report summarizes efforts for the third phase of a research project concerned with the development of a new environmentally friendly method to produce coke using a blend of Indiana and conventional metallurgical coals in a Multipurpose Coke Facility. In addition, the developed process provides for additional value streams that can further increase the benefit of the process and additionally reduce the influence of coke market price volatility. This effort has developed new technology that enhances the economics of the coke production process by enabling the use of blends of lower cost Indiana coal with conventional metallurgical coal as well as providing multiple value streams for the coke production process. The multiple process value streams are continuously optimized to maximize value. Coal samples from Indiana mines have been collected and tested for suitability in producing coke for use in blast furnaces in the steel industry. Initial test results indicate that it is possible to use coal blends containing up to 40% Indiana coal and maintain the desired chemical and physical properties for producing coke suitable for use in large blast furnaces. These samples will serve as a data base for the next phase of the testing and development efforts targeted at development of a larger scale test of the basic process.

Coke is a solid carbon fuel and carbon source produced from coal that is used to melt and reduce iron ore. Although coke is an absolutely essential part of iron making and foundry processes, currently there is a shortfall of 5.5 million tons of coke per year in the United States. The shortfall has resulted in increased imports and drastic increases in coke prices and market volatility. For example, coke delivered FOB to a Chinese port in January 2004 was priced at \$60/ton, but rose to \$420/ton in March 2004 and in September 2004 was \$220/ton. This makes clear the likelihood that prices will remain high.

The market price of coke has varied from \$130 to \$800/ton since 2009. Such fluctuations have caused considerable production planning issues. The current research, through the use of optimized multiple value streams, can reduce the effects of this market volatility by providing alternative revenue streams from multiple products including coke, fertilizer, electricity, Fischer-Tropsch transportation fuels, and hydrogen. It also helps to produce jobs and a new market for Indiana coal.

This effort has considered the suitability of and potential processes for using Indiana coal for the production of coke in a mine mouth or local coking/gasification-liquefaction process. Such processes involve multiple value streams that reduce technical and economic risk. Initial results indicate that it is possible to use blended coal with up to 40% Indiana coal in a non recovery coke oven to produce pyrolysis gas that can be selectively extracted and used for various purposes including the production of electricity and liquid transportation fuels and possibly fertilizer and hydrogen. At present essentially all of the coal used for coke production in Indiana's steel industry is imported from outside Indiana.

Indiana is home to roughly 22% of the domestic base steel production for the United States. One essential raw material needed by this industry is coke. Current 2005 forecasts indicate that the United States will produce 11,500,000 net tons of coke, but will require 17,000,000 net tons for blast furnace, foundry, and related uses.ⁱ At present, essentially no Indiana coal is being used for coke production. In 2002, Indiana's steel industry used an estimated 10.7 million tons of coal. Of this, approximately 8.1 million tons was used for coke production.ⁱⁱ Essentially all of this coking coal comes from Kentucky, West Virginia and Virginia.

The significant shortfall of needed coke has placed an enormous strain on Indiana's steel industries. This report describes initial results of the development of a process that can provide at least a partial resolution and/or mitigation of this formidable problem through the use of Indiana coal in a mine mouth or local, environmentally friendly, high efficiency coking/coal gasification facility which would increase coke supply and production, while, at the same time, reducing the cost for Indiana's steel and foundry industry.

In this phase of the research, a new furnace and test system was placed in operation. Pyrolysis gas is now transmitted directly to the gas chromatograph for analysis through tubing connected to a selector valve. This has improved the accuracy of the data. An expansion of the test system to have the capability of testing up to 5 samples of coal simultaneously is being developed.

The next steps in this effort entail additional laboratory testing of Indiana and other coals alone and in blends in conjunction with process design efforts. Washed coal samples were obtained from Indiana coal mines in southern Indiana. These samples were sealed under Argonne gas to minimize the influence of oxygen on the characteristics of the coal. Tests of physical and pyrolysis gas characteristics of the samples are currently under way. Blends of these coals with metallurgical coal are being tested for pyrolysis gas composition as a function of temperature as well as physical and chemical characteristics. Recently, a new design non recovery coke oven has been introduced in China. This is essentially a vertical non recovery coke oven. Initial indications are that it provides the emission reduction benefits of a vertical design and additional operational benefits. This design has the potential to further extend the economic and environmental efficiency and process benefits made possible by this research effort.

The general conclusion of this study is that it is possible to use a blend of Indiana and conventional metallurgical coal to produce coke for use in various industrial applications at reduced cost while maintaining quality requirements. In addition, there is also potential to also use gas produced in the coking process for a variety of purposes including production of electricity, liquid transportation fuel, fertilizer, and hydrogen.

A patent application has been filed for the developed process by the Purdue Research Foundation. Currently efforts are underway to conduct larger scale process testing that will lead to commercialization of the process and an associated increase in the use of Indiana coal.

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Introduction

This effort has considered initial design aspects of a new process for producing coke, which is a solid carbon fuel and carbon source produced from coal that is used to melt and reduce iron ore, in a Multi Purpose facility that uses up to 40% Indiana coal. Currently no Indiana coal is used for coke production. This effort was conducted by the Energy Efficiency and Reliability Center located at Purdue University Calumet in Hammond Indiana with funding from the Center for Coal Technology Research. The team members for this study were:

- Robert Kramer (Ph.D.) (PI) Professor and Director of the Purdue University Calumet Energy Efficiency and Reliability Center. Areas of expertise include energy research, electric system design and operation, energy system optimization, coal/coke gasification and liquid transportation fuel production, environmental engineering, and project management. He has over 30 years of industrial experience in the energy field, most recently as the Chief Scientist for NiSource.
- Libbie Pelter (Ph.D.), Assistant Professor, Department of Chemistry and Physics, Purdue University Calumet. Dr. Pelter has a background in surface chemistry and catalysis from the petroleum industry.
- Harvey Abramowitz (Ph.D.), Professor, Department of Mechanical Engineering, Purdue University Calumet. Dr. Abramowitz has had extensive experience in metallurgy and steel making processes in general.
- Hardarshan Valia (Ph.D.), President, Coal Science, Inc. Dr. Valia serves as a team member and consultant to the project. He has extensive experience in the steel industry and specifically in the utilization of coal and the coking process. He also has experience with various production and economic aspects of both the coal and steel industry.

A viable supply of iron is one mainstay of economies throughout the world. Issues associated with the supply and price of iron, which is used to produce steel, play either a direct or indirect role in all modern business operations. Indiana is home to approximately 22% of the base steel production for the United States and consequently there is enormous incentive to assure the supply, quality, and price of the raw materials that are used in its production. One of the major components used in the iron making process is coke.

Coke is a solid carbon fuel and carbon source used to melt and reduce iron ore. Coke production begins with pulverized, bituminous coal. In current operations, coal itself cannot be used in place of the central placement of coke in a blast furnace because it would not form a permeable bed of sufficient strength and porosity to support the weight of material in the blast furnace.

Coal is fed into a coke oven which is sealed and heated for 14 to 36 hours to about 1100 C (2000 F). Coke is produced by heating particulate coals of very specific properties in a refractory oven in the absence of oxygen (or with limited oxygen at the

top of the coal bed in the case of non recovery coke ovens). As temperature increases inside the coal mass, it melts or becomes plastic, fusing together as devolatilization occurs, and ultimately resolidifies and condenses into particles large enough for blast furnace use. During this process, much of the hydrogen, oxygen, nitrogen, and sulfur are released as volatile by-products, leaving behind a partially crystalline and porous carbon product. The quality and properties of the resulting coke is inherited from the selected coals, as well as how they are handled and carbonized in coke plant operations.

Heat is often transferred from one coke oven to another to reduce energy requirements. After the coke is finished, it is moved to a quenching tower where it is cooled with a water spray. Once cooled, the coke is moved directly to an iron melting furnace or into storage for future use. Currently essentially no Indiana coal is used to produce coke.

Coke production is traditionally one of the major pollution sources from steel production. At present there are two main methods of producing coke. First, a recovery process in which the coal is heated in a completely reducing atmosphere and the volatile products are recovered in an associated chemical processing plant. Major issues associated with this process include the complexity of the chemical processing and the production of potentially hazardous compounds. There is also a major concern with the tar that is left after processing. This material is also potentially hazardous and is generally stored on site and thus presents a significant future disposal concern. The complexity of the chemical process operational details that have restricted the use of this option in the past for coking and simultaneous power production.

Air emissions such as coke oven gas, naphthalene, ammonium compounds, crude light oil, sulfur, and coke dust are released from many coke ovens. Emissions control equipment can be used to capture some of the gases and heat can be captured for reuse in other heating processes. But, traditionally, some gases escape into the atmosphere as the coke oven ages. Air and water emissions from coke production can be reduced by using a non-recovery coke battery. In traditional plants, by-products are can be recovered. In non-recovery batteries, pollutants are combusted in the coke oven itself, which is often maintained at a negative pressure. This technique consumes the by-products, eliminating much of the air and water pollution.

In the non recovery process air is introduced above the top of the coke bed in the oven and the volatiles are combusted. The Environmental Protection Agency has stated that new ovens must meet non recovery standards. The hot gases from the oven can then be used in a heat recovery boiler to produce steam and subsequently generate electricity. Relatively small amounts of hydrogen are produced in this process and are recalculated to the bottom of the furnace to provide heat for the process.

Recently, a new design non recovery coke oven has been introduced in China. This is essentially a vertical non recovery coke oven. Initial indications are that it provides the emission reduction benefits of a vertical design and additional operational benefits. This

design has the potential to further extend the economic and environmental efficiency and process benefits made possible by this research effort.

In the iron making process, iron ore, coke, heated air and limestone or other fluxes are fed into a blast furnace. The heated air causes the coke to combust, which provides the heat and carbon sources for iron production. Limestone or other fluxes may be added to react with and remove the acidic impurities from the molten iron in the form of slag.

Process Description

One key issue in blast furnace iron making is the strength of the coke. The coke produced from Indiana coal has less strength than coke produced from current metallurgical coal sources and consequently is smaller in size. This means that it will be used in upper portions of the blast furnace. Typical characteristics of coke used in blast furnace operations are shown in Table 1.ⁱⁱⁱ

Physical: (measured at the blast furnace)	Mean	Range
Average Coke Size (mm)	52	45-60
Plus 4" (% by weight)	1	4 max
Minus 1"(% by weight)	8	11 max
Stability	60	58 min
CSR	65	61 min
Physical: (% by weight)		
Ash	8.0	9.0 max
Moisture	2.5	5.0 max
Sulfur	0.65	0.82 max
Volatile Matter	0.5	1.5 max
Alkali (K ₂ O+Na ₂ O)	0.25	0.40 max
Phosphorus	0.02	0.33 max

 Table 1: Typical Blast Furnace Coke Characteristics

This report details research that was conducted from March 2006 to the present to determine the viability of using Indiana coal for the production of coke. Specifically, the concept of locating a modified non recovery coking facility at a mine in Indiana or at an existing facility with energy recovery for the generation of electricity was considered. In addition, extension of the technology to include gasification and local power production were also considered. The results of this study indicate that there is a high potential to use Indiana coal for coking as well as other industrial purposes both within and outside Indiana. A flow diagram of the concept is depicted in Figure 1. A significant aspect of the research has been to develop multiple value streams from the coke production

process that are optimized in real time to produce the maximum value. There has been considerable complexity introduced in the planning process due to coke market price fluctuations ranging from Due to the market price fluctuations for coke experienced since 2009 which have ranged from \$130 to \$800/ton there has been considerable uncertainty in production planning. The use of Indiana coal in a Multipurpose Coke Facility can reduce the effects of this price volatility and at the same time open a new market and technique for the use of Indiana coal. The multiple value streams for the proposed process optimize value based upon market pricing for the various value streams. When there is a sudden decrease in one product value it is possible to at least partially shift production to optimize the production of a different and more valuable product. The interaction between these various value streams is depicted in Figure 2. Within the constraints of the physical production process the output from each of the value streams is optimized to produce the maximum net value in essentially real time. This provides the ability to quickly respond to market price changes. The general product value flow is depicted in Figure 3. The process flow is depicted in Figure 4.

To better understand the potential value of the proposed technology a risk assessment was conducted. The economic flows considered in the analysis are depicted in Figure 5. In this study, probability distributions for the value of products and operating costs were estimated and input to a production model through the use of a Monte Carlo simulation. Results from this study are depicted in Figure 6. In Figure 6 it can be observed that there is a very high probability (~92%) that there will be a net positive value for the process given the market uncertainties. Conservative assumptions were used for this estimate.

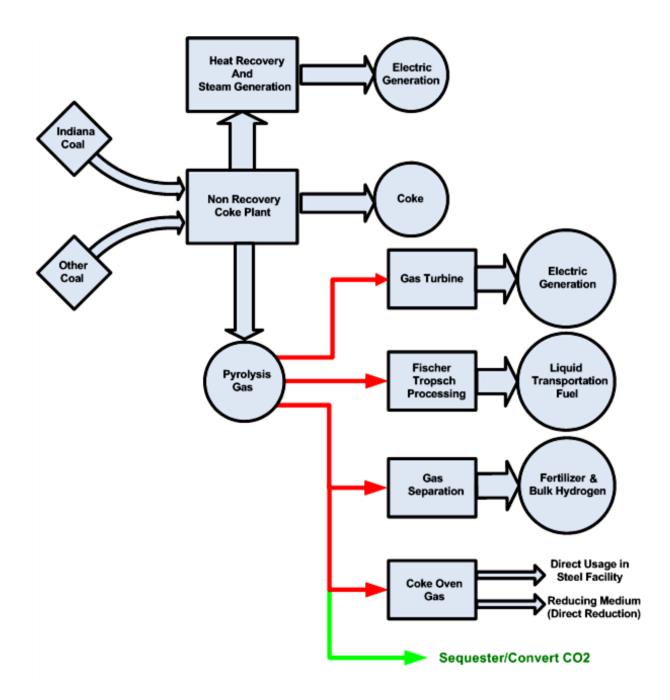


Figure 1: Concept Description

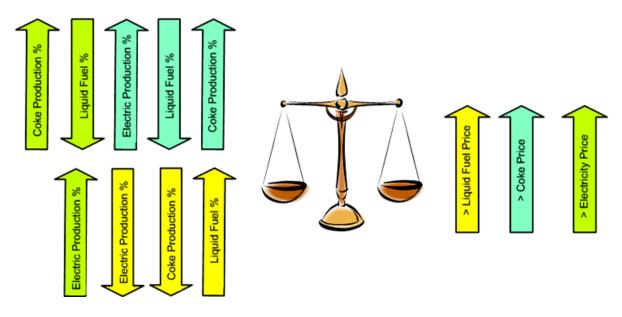


Figure 2: Economic Optimization

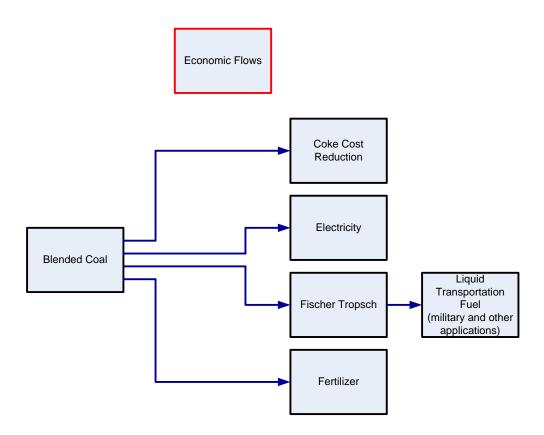


Figure 3: Value Streams

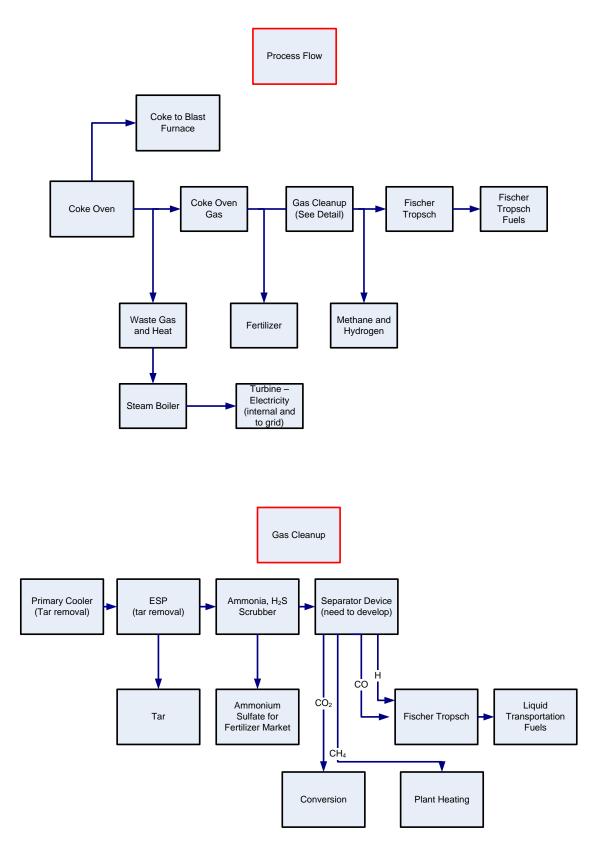


Figure 4: Process Flow

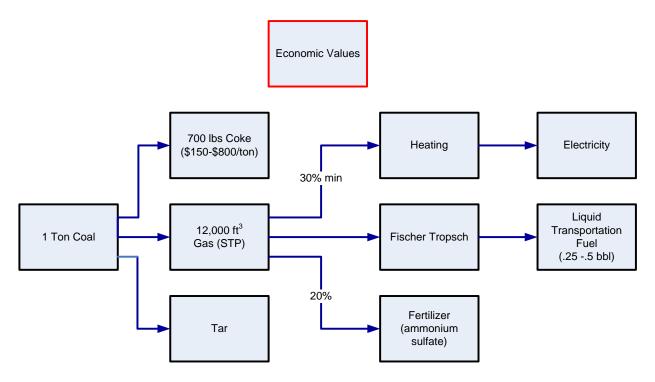


Figure 5: Economic Values

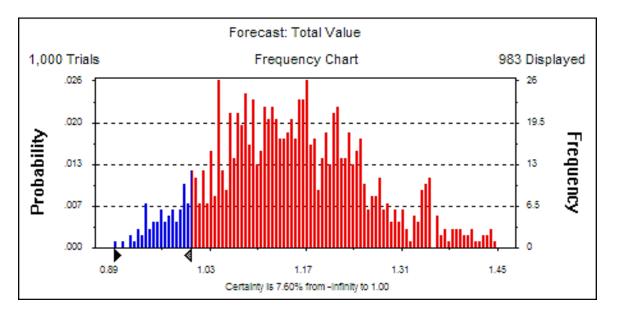


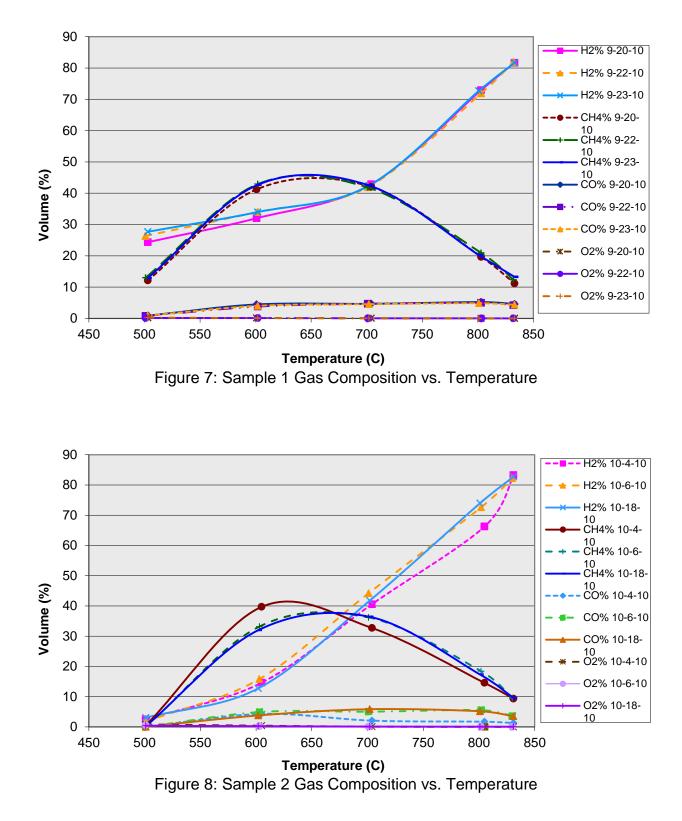
Figure 6: Risk Assessment Results

The coal used for the proposed coking process would be a mix of Indiana Brazil Seam and other Indiana coals, as previously identified by the Indiana Geological Survey, blended with other coals to meet metallurgical and emissions requirements. Currently this approach has been used successfully to dramatically increase coke quality. Several steel manufacturers have expressed interest in considering how Indiana coal might be used for various production processes. They also indicate that they have considered and/or are currently considering using Indiana Coal usually at low levels in blends. This research is investigating ways to increase the percentage of Indiana coal used for coke production. One approach is to blend different types of coals until a mixture is obtained that meets the coke quality requirements.

Samples of washed coal were obtained from mines in Southern Indiana. These coal samples are stored in an Argonne atmosphere to minimize the influence of oxygen. The samples were blended with metallurgical coal and the blends were evaluated for use in the coke production process.

Efforts to extend the blending to also consider optimizing the composition of the pyrolysis gas produced in the coking process are also underway. By optimizing both aspects simultaneously it will be possible to obtain coke of acceptable quality for use in blast furnaces and other applications and at the same time obtain a supply of pyrolysis gas that can be used for the production of liquid transportation fuels through the use of the Fischer-Tropsch process, and possibly fertilizer bulk hydrogen.

Laboratory tests of several Indiana coals were conducted to determine the suitability of Indiana coal for purposes of producing liquid transportation fuels, fertilizer, and hydrogen as part of the coke production process. As the temperature of the coal is increased in the coke production process pyrolysis gas of varying composition is released. In the proposed concept it is anticipated that portions of this gas will be gathered from the coke process at specific temperature ranges with the proper composition for the production of liquid transportation fuels, fertilizer, and hydrogen. Figures 6-8 depict typical test results for the pyrolysis gas composition from various Indiana coal samples at different temperatures. Figure 9 shows the apparatus used to test the pyrolysis gas produced by Indiana coal alone and in blends. This apparatus is connected to a gas chromatograph for analysis of the produced gas and has the capability of testing 10 samples simultaneously. The details of the pyrolysis apparatus developed as part of this research effort is shown in Figure 10.



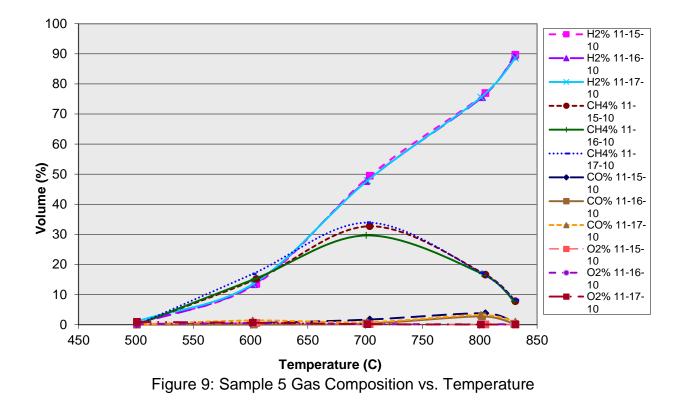




Figure 10: Pyrolysis Test Equipment

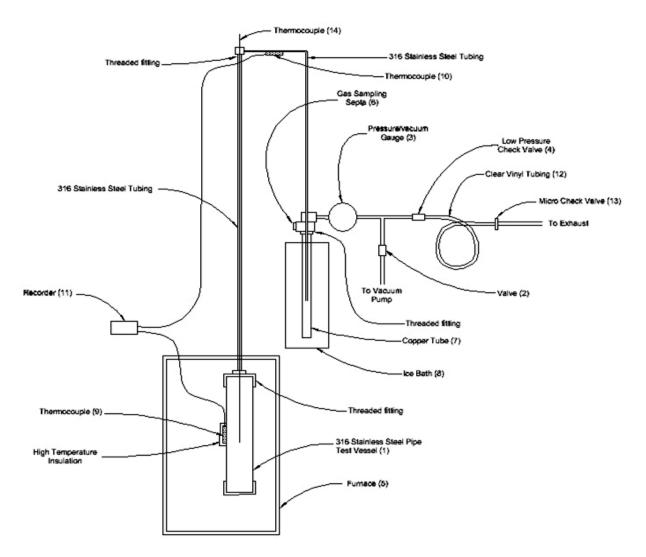


Figure 11: Coal Pyrolysis Test Apparatus

Further tests of the washed coal samples and blends are currently underway. Methods to evaluate optimal coal blends that maximize Indiana coal usage are being developed. Efforts to characterize coal blends and related chemical and physical characteristics are increasing since this directly influences the potential for Indiana coal markets for use in the Steel Industry. Initial tests results are shown in Table 2.

	Individual Coals					Blend Coals			
	Sample 1	Sample 2	Sample 3	Sample 4	Sample 5	Blend # 1	Blend # 2	Blend # 3	Blend # 4
Total Moisture	14.67	14.34	7.95	5.57	7.75	11.88	11.11	7.18	8.44
Ash	4.08	5.81	9.68	9.93	6.61	5.81	6.54	8.70	7.76
Volatile	39.27	38.09	20.26	29.28	15.87	33.67	34.44	21.89	24.67
Fixed Carbon	56.65	56.10	70.06	60.79	77.52	60.52	59.02	69.41	67.57
Sulfur	1.58	1.32	0.35	1.11	0.77	1.39	1.24	0.75	0.87
HV(BTU/lb)	14129	13873	14040	13980	14635	14154	14068	14195	14245
Carbon	78.32	77.70	80.57	78.86	86.05	79.18	78.91	81.43	81.44
Hydrogen	5.40	5.42	4.21	4.90	4.12	5.19	5.26	4.46	4.64
Nitrogen	1.59	1.67	1.16	1.61	1.31	1.62	1.65	1.41	1.45
Oxygen by difference	9.03	8.08	4.03	3.59	1.14	6.81	6.40	3.25	3.84
Initial Deformation Temperature (°F)	2176	2388	>2700	2259	>2700	2266	2104	2572	2633
Softening Temperature	2170	2000	22100	2200	22100	2200	2104	2012	2000
(°F)	2442	2419	>2700	2430	>2700	2302	2404	>2700	2689
Hemisphercial Temperature (°F)	2464	2466	>2700	2507	>2700	2514	2534	>2700	>2700
Fluid Temperture (°F)	2502	2624	>2700	2665	>2700	2660	>2700	>2700	>2700
Vineral Analysis of Ash									
Silicon dioxide	43.90	54.59	61.63	53.97	57.77	48.96	50.03	57.42	56.35
Aluminum oxide	27.80	23.92	26.82	25.52	31.18	28.50	27.36	27.85	28.04
Titanium dioxide	1.30	1.23	1.58	1.26	1.85	1.41	1.23	1.52	1.53
Iron oxide	24.24	16.73	4.17	10.92	4.75	15.70	14.21	6.13	7.91
Cacium oxide	0.99	0.73	2.22	1.57	1.30	1.24	1.70	2.05	1.66
Magnesium oxide	0.21	0.33	0.47	1.49	0.42	0.61	1.02	0.97	0.90
Potassium oxide	1.16	1.87	0.63	3.08	1.27	1.90	2.31	1.88	1.92
Sodium oxide	0.26	0.34	0.07	0.70	0.70	0.47	0.57	0.47	0.50
Sulfur trioxide	0.03	0.13	1.50	1.00	0.58	0.83	0.90	1.25	0.85
Phosphours pentoxide	<0.01	<0.01	0.66	0.15	<0.01	<0.01	0.27	0.17	0.08
Strontium oxide	0.04	0.06	0.06	0.13	0.07	0.21	0.19	0.10	0.10
Barium oxide	0.04	0.05	0.14	0.15	0.05	0.08	0.14	0.13	0.11
Manganese oxide	0.02	0.01	0.03	0.04	0.03	0.04	0.05	0.04	0.03
Undetermined	0.01	0.01	0.02	0.02	0.03	0.05	0.02	0.02	0.02
MAA Basis	Ignited	Ignited	Ignited	Ignited	Ignited	Ignited	Ignited	Ignited	Ignited
Alkalies as Na2O, Dry Coal Basis	0.04	0.09	0.05	0.27	0.10	0.10	0.14	0.15	0.14
Base Acid Ratio	0.35	0.03	0.08	0.18	0.07	0.23	0.22	0.10	0.14
Silica Value	63.32	75.42	89.96	79.43	89.93	73.61	74.72	86.24	84.32
MAA Sum	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00

MAA T250 (F)	2418	2658	2875	2692	2825	2582	2601	2790	2760
FSI	5.5	3	6.5	6.5	5.5	5.5	6.5	7.5	6.5
Vitrinite type(V-Type)									
V-4	20.00	3.00				14.00	6.00		
V-5	74.00	67.00				43.00	43.00		7.00
V-6	6.00	27.00				10.00	16.00		4.00
V-7		3.00					5.00	1.00	
V-8				3.00				8.00	3.00
V-9			3.00	27.00		10.00	14.00	21.00	20.00
V-10			14.00	58.00		7.00	12.00	21.00	8.00
V-11			14.00	12.00		1.00	4.00	2.00	10.00
V-12			4.00					3.00	4.00
V-13			23.00					6.00	1.00
V-14			33.00					3.00	5.00
V-15			9.00			2.00		8.00	5.00
V-16					61.00	6.00		13.00	20.00
V-17					39.00	7.00		14.00	13.00
Fluidity									
Softening Temperature									
(°C)			426	378	450	388	382	412	410
Solidification (°C)			499	489	510	464	481	505	500
Range (°C)			73	111	60	76	99	93	90
Temperature at Maximum			471	444	484	440	442	467	465
Maximum Fluidity ddpm			26	60,111	404 7	162	1056	407 507	405 259

Table 2: Coal Tests

In addition to the nature of the gas produced during the coke process it is also crucial that the produced coke meet standards for its use in blast furnace or other applications. One method to obtain the proper coke properties is through the blending of various types of coal.

An example of three types of coal blends used by the Japanese Steel Industry in 1975 for coke production is depicted in Figure 12.^{iv}

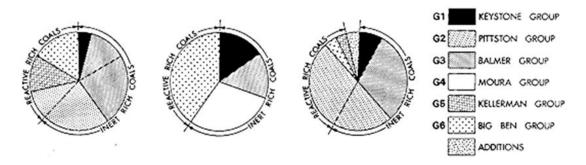


Figure 12: Coking Coal Blend Example

One way to rank coals is by the amount of volatile matter they contain. At the simplest level, mid-range prime coking coals will produce the best coke and the farther a particular coal is away from prime coking coal, the less suitable it is. Coke from high volatile coals tends to be too weak and reactive to be used in the blast furnace. Also, carbonizing low volatile coals can produce unacceptably high pressures on oven walls for slot ovens.

When coal is viewed under a microscope, it can be seen to be composed of three main components, or macerals, analogous to the minerals found in rocks.^v The first of these, vitrinite, softens on heating. It in association with the other components, liptinite and inertinite, forms the coke matrix. These components reflect light at different intensities. In general, the reflectance of the vitrinite is a measure of the rank of the coal and is inversely proportional to the volatile matter content. Usually a coal blend for blast furnace coke should have a reflectance between 1.25% and 1.35%. The reflectance of coals blends tends to vary linearly, but having the average reflectance of a blend in this range is not sufficient to assure that the produced coke will have the desired qualities. For this reason the reflectance distribution is considered.

If the reflectance values from a sample are plotted in a histogram, it is desirable to have a distribution that resembles a normal distribution with not too large a standard deviation. Unacceptable distributions have large standard deviations or have multiple peaks.^{vi} Attempts at using simple linear programming models to determine coal blends for coking have produced varying results due to the complexity of the coking process.^{vii} Modeling must also consider other characteristics such as dilatation and fluidity, which provide empirical measures of the extent of softening and fusion on heating, in the blending process.

Due to the physical characteristics of Indiana coal^{viii}, the coke produced will tend to be of a smaller size, but there are many opportunities to use this type of coke in blast furnace and other operations. Concerns with the relative strength of the coke produced from Indiana coal can be reduced by carefully blending various types of coal. Through blending many potential issues with coke characteristics can be reduced or eliminated. Classically, coal blending for coke production has been considered to contain a level of "art" to the process. The research team for the proposed project has had considerable experience in customizing coal blends used for coking processes in operating industrial coke production facilities. This experience will be a valuable attribute in customizing the process to maximize the use of Indiana coal. The research team will develop blending models and/or recommendations that will help to increase the use of Indiana coal for industrial purposes. The nature of the coal blend for the current proposal would be a function of the coking process detail and will require additional research to determine the optimal values.

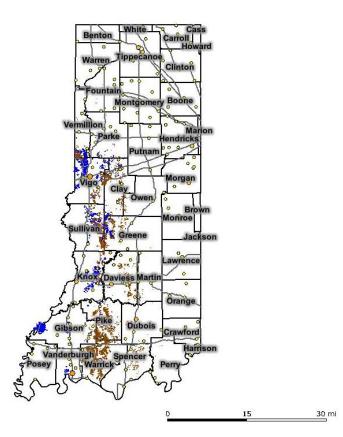
Another approach to increasing the percentage of Indiana coal for coke production involves locating that coke in upper regions of the blast furnace where higher reactivity is less of a concern. In this region there is also less mechanical pressure on the individual pieces of coke since there is less material above it. This would allow coke of reduced strength to be readily used in this region.

This research effort has also considered if it would be conceptually possible to modify the mass balance in the coking process in a way that would allow for a usable level of gas production that could be used to power a combustion turbine for electric production. In discussions with various operational, research, and engineering personnel it has been found that there is a possibility that a portion of the pyrolysis gas could be extracted from the gas stream as it is recirculated to the floor of a non recovery coke oven also referred to as the sole plate. The degree of gasification and influence on operations would need to be considered in a subsequent detailed study. Preliminary process modeling was done with the Metsim computer model^{ix} Metsim is a computer program that can model industrial processes, unit operations, and chemical and metallurgical processes.

The location of the proposed coke production process would take place either near or at an Indiana coal mine or at an existing production facility. The choice of location would be made based on business issues and also on the availability of transportation capabilities. Transportation of both coal and coke is necessary in this process since the coal used for coke production would be a blend of Indiana and other metallurgical coals. Production of coke at mine mouth would afford a transportation savings because a large portion of coal used by the coking facility would not have to be transported over a long distance. But, coal for blending as well as the finished coke would need to be transported. If sufficient transportation capability exists total transportation cost would be reduced since the mass of the product coke is less than the coal needed to produce it. Thus, a significant cost savings from the reduced weight per mile of material being transported would result if transportation capability was available. This effort is being correlated with research efforts regarding Indiana rail transportation capabilities being conducted by Purdue University North Central. Figure 13 depicts the location of Indiana coal resources.^x It will be important to consider issues of distance and availability of transportation as part of the commercialization planning process.



Index map of Indiana showing the coal-bearing rocks of the Pennsylvanian System in green, underground coal mines in blue, and surface coal mines in brown.



© Indiana University 2003 Figure 13: Indiana Coal Locations

Various industry contacts were established to obtain background for the project. Two coal mines were contacted and a coal sample was obtained. Two coal mine operators have indicated an interest in considering the concept for a mine mouth coking facility. Preliminary discussions have considered how such a facility might be developed. Two steel mills were visited and process applications of Indiana coal were discussed. One of the steel mills performed analysis of a sample of Brazil Seam Coal. A coke production facility was visited and issues regarding coking technology were considered.

The coking/coal gasification process would produce metallurgical grade coke using 20-40% Indiana coal and, at the same time, would produce a byproduct gas stream that would be usable in a cogeneration facility for the production of electricity to be sold in

the electric market. Initial power flow studies have been investigated to determine the potential value of the generated electricity. Preliminary studies indicate that 100 MW of electricity could be generated from a large coke production facility and that electric grid reliability in either the mine mouth or industrial locations considered would be increased as a result of the additional generation. Alternatives for electric production including heat recovery and potentially partial coal gasification were also evaluated. Results indicate that electric production in conjunction with coke production provides a significant economic benefit. Issues of the ability to produce electric ancillary services as part of the operation are also being considered.

In the proposed process, existing or planned coke production facilities would be used as part of the developmental process thereby reducing the process development risk as compared to construction of a dedicated test facility. The proposed process is based upon a design in which the risk and financing level required for development of an operating facility is reduced by developing the technology in conjunction with an operating or planned coking facility. The value of products, including liquid fuels, would be evaluated in comparison with conventional coke production operation. The amount of such products produced would be determined by optimizing the value of the various product streams. The process would adapt itself to changing market conditions. This would reduce the risk of developing new coal based liquid fuel production capability since the major capital expenditure, the gasifier, is already justified for conventional coke production. This technology can provides a knowledge base in the near term with significantly less cost than a dedicated gasification facility that could then be leveraged for the development of large scale dedicated gasification facilities. In a conventional Fischer-Tropsch liquid transportation fuel production facility a large portion of the cost is associated with the construction and operation of the gasifier. The cost breakdown for a conventional Fischer-Tropsch liquid transportation fuel plant is shown in Figure 14.^{xi} The Multi Purpose Coke Facility being developed as part of this research would avoid the majority of the cost associated with the gasifier since the coke oven itself is the gasifier and is cost justified for the production of coke. This drastically reduces the cost of the Fischer-Tropsch liquid transportation fuel produced by this process as compared to that for a conventional Fischer-Tropsch facility.

In the proposed process, pyrolysis gas is extracted over a temperature range in which there is a desired gas composition. This reduces the post process chemical treatment of the gas and further reduces the capital and operating costs in comparison to a conventional gasification plant.

Distribution of Total Overnight Capital Costs 15 wt% CBTL+CCS, 50k bpd Fuel Handling, Inventory, Pre-Prep & Feed Production & 11% **Owner's** Costs 6% **Contingencies** 20% Gasification Island 32% Engineering & Construction Mgmt. 6% CO₂ Removal. Comp., Transport, **Balance of Plant** Seq. & Monitoring **FT** Synthesis 9% 5% 11%

Figure 14: Conventional Fischer-Tropsch Costs

A concept for a process for the sequestration of the carbon dioxide produced by the process was also identified. Preliminary investigations indicate that it may be possible to produce a usable chemical product as part of the carbon dioxide sequestration process by the use of a nano catalysis. A concept for using a nano catalysist to enhance the coke oven gas based Fischer-Tropsch process for the production of liquid transportation fuels is also being considered.

Importance to Indiana Coal Use

The central theme of this effort has been to find ways to increase the use Indiana coal in coking and other related industrial operations in a way that increases overall value. By finding ways to increase the use of Indiana coal in such processes, imports of coal from sources outside Indiana will be decreased and there will be a potential to open new markets for Indiana coal.

A mine mouth coking/coal gasification facility will have many positive economic and employment effects for Indiana. This facility will be located in Indiana. Typically, a 1.3 million ton per year coke facility employs about 130 people. In addition, it is estimated that 13 new employees would be required in the Indiana mining industry. A new facility of the type considered would provide a significant employment opportunity for Indiana. Such a facility would allow the Indiana Coal Industry to open a new and expanding market. Metallurgical coal contracts increases by 20% to 40% in 2004. xii In 2002 Indiana imported 8.093 million tons of coking coal. The potential for use of Indiana coal for coke production for use in Indiana is between 2.0 and 3.6 million tons per year. Export potential is estimated to range from 6 to 11 million tons per year.xiii Current coke production at Indiana Harbor facilities is 1.2 million tons per year screened. The proposed facility would be of a comparable size and would result in an estimated cost savings of at least 5 % for delivered coke due to reduced transportation costs and would meet a portion of future demand growth. It would also reduce imports of metallurgical coal by several million tons per year and replace it with coal produced in Indiana. There would also be a potential to export coke to adjacent States including Ohio, Kentucky, and Illinois. The sale of electric power from the cogeneration function would also result in a significant revenue stream to further enhance the benefit of the project.

Indiana's steel industry is a major employer, as well as significant sources of revenue to the State in the form of taxes. This project will help to assure the health of this vital industry, generate new jobs and revenue streams, and advance the technical state of the art by using Indiana coal and simultaneously reducing environmental emissions.

Environmental emissions are often cited as a reason why Indiana coal is not used in the production of coke. The proposed process presents a different option that inverts the classic coke production paradigm. This project has done preliminary work to develop a process in which clean coal technology is used at the mine mouth or at an industrial location to produce coke, rather than transporting coal from sources outside Indiana to non attainment areas for coke production. Gas streams from the coking process will be collected and used for subsequent production of electricity at the site or possibly the production of liquid transportation fuel. This process will result in a net transportation savings, as well as a value stream from cogenerated electricity. Such a facility will provide base load electric generation, but will also have the capability to supply shoulder and peaking power, in addition to, potentially ancillary services.

The U.S. coke industry has two primary product markets (i.e., furnace and foundry coke) that are supplied by two producing sectors—integrated producers and merchant producers.^{xiv} Integrated producers are part of integrated iron and steel mills and only produce furnace coke for captive use in blast furnaces. Therefore, much of the furnace coke is produced and consumed by the same integrated producer and never passes through a market. However, some integrated steel producers have closed their coke batteries over the past decade and purchase their coke supply from merchant producers or foreign sources. A small number of integrated steelmakers produce more furnace coke than they need and sell their surplus to other integrated steelmakers. In 1997, integrated producers accounted for roughly 76 percent of U.S. coke capacity with

merchant producers accounting for the remaining 23 percent. These merchant producers sell furnace and foundry coke on the open market to integrated steel producers (i.e., furnace coke) and iron foundries (i.e., foundry coke). Some merchant producers sell both furnace and foundry cokes, while others specialize in only one.

Even though captive consumption currently dominates the U.S. furnace coke market, open market sales of furnace coke are increasing. As production costs increase, U.S. integrated steel producers increase their consumption of furnace coke from merchant coke producers, foreign imports, and other integrated steel producers with coke surpluses.

The research team for this proposal has extensive experience in the coking process, characterization of Indiana coal coking properties, electric generation, engineering, and system analysis. Major products from a facility using the proposed process will be coke, electricity, liquid transportation fuel, and potentially fertilizer and hydrogen. All are crucial to the economic future of Indiana. The locations of Indiana's coal mines provide many unique advantages for coke production.

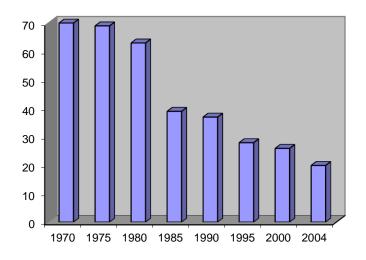
This proposal leverages experience from current coking facilities in Indiana. Additional research is required to extend these technologies for use in the proposed coking process, but the technical risk will be less than for a completely new experimental concept. Such an approach is made possible by the use of proven technology in the new coking paradigm of this research effort. This approach significantly increases the probability that an actual productive facility could operational within a 5 year time frame. The proposed coke production technology will provide many advantages over current production methods. These advantages will also be attractive both within and outside the United States. Due to current market shortages and the price volatility of coke internationally, there is an opportunity to market Indiana coal in a new way in the form of coke to a variety of new markets both within and outside Indiana.

Relevance to Previous Studies

Previously Indiana coal was used for coke production.^{xv} With the development of large blast furnaces Illinois Basin coal was not used for coke production. Based upon methods being developed in this research, it is now appropriate to again start using Indiana coal for coke production. To accomplish this it will be necessary to develop methods that alleviate issues with using Indiana coal for coke production and simultaneously add value to the process. This proposal presents an approach that is targeted at meeting these requirements.

Furnace coke also accounts for the majority of domestic coke usage.^{xvi} Figure 30 depicts the world distribution of coals suitable for coke production.^{xvii} Figure 31 depicts world coke production capacity minus consumption. The dotted line in this figure is a minimum level taking into account scheduled and forced outages.^{xviii} It can be observed that the supply of coke is anticipated to increase slightly in the future above the base level in 2004, but will level off at a relatively low value. This will result in a situation of elevated price and need for additional supply. Coke produced from Indiana coal could serve to meet a portion of this demand.

Due to a variety of circumstances including the tightening of emissions regulations, the number of coke ovens is decreasing as can be seen in Figure 15.^{xix} This indicates that there is clearly a need for new environmentally friendly coking production capability. The proposed research would support the development of such capability using Indiana coal.



US Operating By-Product Coke Plants

Figure 15: US Operating By-Product Coke Plants

In addition to decreasing numbers, a significant portion of the existing capacity is reaching end of life. Figure 16 depicts coke battery age at Mittal Steel.^{xx} This also supports the observation that there is need for new environmentally friendly coking production capability. As units reach the end of life, maintenance costs and outages increase dramatically.

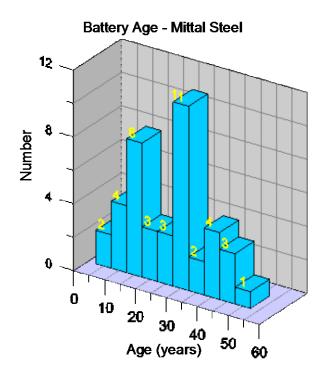
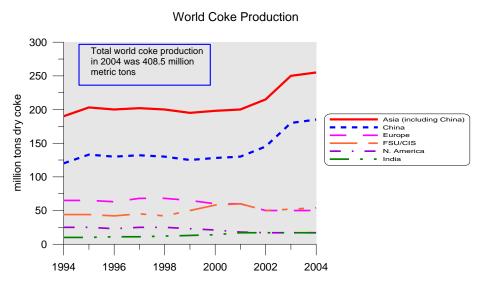
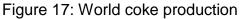


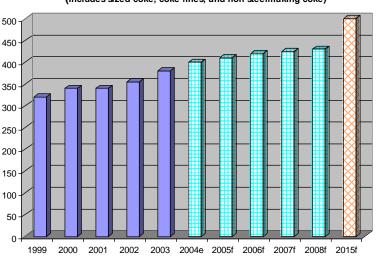
Figure 16: Battery Age – Mittal Steel

Figure 17 depicts the global production of coke^{xxi} and Figure 18 depicts the global consumption of coke products. From this figure it is clear there is a need for new coke production capacity.^{xxii} In general domestic supplies of coke are decreasing while international demand is increasing.^{xxiii} The estimated 2.2 billion tons of metallurgical reserves in the U.S. at an assumed consumption rate of 50 million tons per year would result in 40 years worth of recoverable reserves from currently operating mines. Using Indiana coal in the coking process described in this effort could improve economics and extend these reserves.

The price volatility experienced recently in China is a result of supply and export policies. In 2001 the cost of coke was \$80/ton FOB to a Chinese port. In 2004 it was \$410/ton. Currently it is \$200/ton.^{xxiv} In 2002 Chinese government decreased the number of coke export licenses to meet growing demand.^{xxv} It is anticipated that prices could stabilize at the \$200/ton level.^{xxvi} This would provide a clear incentive for the construction of additional coke production capacity.







Golbal Coke Product Consumption (Includes sized coke, coke fines, and non steelmaking coke)

Figure 18: Global Coke Consumption

Producing combustible gases from solid fuels has been done since ancient times. Pyrolysis is a process in which feed material is heated with little air present. It this context, blast furnaces can be considered to be large gasifiers of coke.^{xxvii}

In a recovery coke oven, typically the coke oven gas has a composition of 58% hydrogen, 26% methane, 5.5% nitrogen, 2.25% acetylene, 2% carbon dioxide, 6%

carbon monoxide, and .25% oxygen.^{xxviii} One metric ton of coal typically produces 600-800 kg of blast-furnace coke and 296-358 m³ of coke oven gas.^{xxix}

This hydrogen content is typically too high for use directly in Fischer-Tropsch processes for the production of liquid transportation fuels. Methods to reduce this to the range of 2-4 to 1 hydrogen to carbon monoxide ratio, possibly by combining with syngas streams, will be considered. Other processes for removing various constituents from the gas stream including oil, sulfur, and naphthalene will also be considered.^{xxx} It is also possible to adjust this ratio by blending various coals as was shown previously. Currently, an optimization scheme is under development that maximizes both coke properties and pyrolysis gas composition.

Conclusion

This study has shown that Indiana coal can become an important resource for the production of coke for the steel and other industries both inside and outside Indiana. As was noted in the study, currently there is a shortfall of 5.50 million tons of coke per year in the United States. This research effort has shown that Indiana coal can become one way to reduce current and future coke supply issues as well as reducing coke price by as much as 10%.

The significant shortfall of needed coke has placed an enormous strain on Indiana's steel and foundry industries. The need for additional coke production capacity is evident given plans for coke plant expansion being considered by Indiana's steel industry and others. The results of this study indicates that coke supply and high price volatility issues can be mitigated through the use of Indiana coal in a mine mouth or local, environmentally friendly, high efficiency multi purpose coke, liquid transportation fuel, fertilizer, electric, and hydrogen production facility. Such a facility would also increase coke supply and production, while, at the same time, reducing the cost for Indiana's steel industry. In addition, such a high efficiency coking facility would produce electricity for sale to the wholesale electric market, thereby reducing costs and environmental emissions and, at the same time, enhancing electric system reliability.

The following are major results from this study:

- 1. A mixture of Indiana Brazil Seam or potentially other Indiana coals, as previously identified by the Indiana Geological Survey, is being considered for development of blends with other coals to meet metallurgical coke quality and emissions requirements. Since there is a limited supply of Indiana Brazil seam coal, methods to use other Indiana coals are also being developed.
- 2. There is interest in the coal and steel industry to consider establishing a coke production process at an Indiana coal mine or steel facility. Moreover, there may be an opportunity to consider the value of some emissions credits, due to the "clean coal technology" as well as the different geographic location.
- 3. The total transportation cost could be reduced, since the mass of the product coke is less than the coal needed to produce it and also because coke is less dense than coal. Thus, a significant cost savings from the reduced weight per mile of material being transported would result. Issues regarding the availability of transportation need to be considered before a final recommendation on location can be made.
- 4. Results indicate that the developed Multi Purpose coking/coal gasification process can produce metallurgical grade coke using 30%+ Indiana coal and, at the same time, produce a byproduct gas stream that would be optimized for use in a cogeneration facility for the production of electricity to be sold in the electric market as well as other value stream products as described previously. By using a new blending approach that optimizes coke properties and pyrolysis gas composition it is possible to increase the percentage of coke produced from Indiana coal blended with coke from other coals in blast furnace operations.

- 5. Results indicate that it is possible to utilize the pyrolysis gas generated from a coke oven feed with a blend of Indiana and other coal to produce electricity, liquid transportation fuels by means of a Fischer-Tropsch process, fertilizer, and hydrogen. It may be possible to enhance this process with nano catalysis technology. There are also indications that it may be possible to isolate carbon dioxide from the process and use it to produce a marketable chemical product with nano catalysis technology.
- 6. Washed coal samples have been obtained from Indiana coal mines. These samples were sealed under Argonne gas to minimize the influence of oxygen on the characteristics of the coal. Tests of physical and pyrolysis gas characteristics of the samples are currently under way. Blends of these coals with metallurgical coal is being tested for pyrolysis gas composition as a function of temperature as well as physical characteristics related to coke production.
- 7. A new furnace and test system has been placed in operation. Pyrolysis gas is now transmitted directly to the gas chromatograph through tubing connected to a selector valve. This has improved the accuracy of the data. An expansion of the test system to have the capability of testing up to 10 samples of coal simultaneously is in process.

Indiana's steel and foundry industries are major employers, as well as significant sources of revenue to the State in the form of taxes. This technology will help to assure the health of these vital industries, generate new jobs and revenue streams through the use of Indiana coal at a facility to be located in Indiana, and advance the technical state of the art by using Indiana coal and simultaneously reducing environmental emissions.

Next steps in the technology development/commercialization process should include completion of the current coal blending tests with washed coal and verification of the suitability of blends of Indiana and other coals for use in coke production. After these tests are completed the proposed process model should be completed. Following completion of the initial process design, tests at an existing test facility capable of processing one ton of coal at a time (available through a consulting arrangement) should be conducted in conjunction with a proof of concept test of production of Fischer-Tropsch solids and subsequently liquids. Once the one ton tests are completed the next stage will be to implement the process at either an existing or new commercial coke oven. At this stage the process will be ready for full commercial implementation.

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