Development of a Multipurpose Coke Plant for Synthetic Fuel Production

Robert A. Kramer, Ph.D.
Director, Energy Efficiency and Reliability Center
Professor, Electric and Computer Engineering
Purdue University Calumet
Hammond, Indiana
219-989-2147

Liberty Pelter, Ph.D.
Assistant Professor, Chemistry and Physics Department
Purdue University Calumet
Hammond, Indiana

Hardarshan S. Valia, Ph.D.
Coal Science Inc.
Highland, IN

Keywords: Multipurpose Coke Facility, Indiana Coal, Coal Blending, High Volatile Coals

Introduction

World coal and coke market prices have recently been in a state of flux. Chinese Coke before the year 2000 was priced at about $50/t FOB Port. Since that time there has been extreme volatility in coke prices. In 2004 prices increased to $320/t FOB Port with subsequent prices of $180/t FOB Port in 2005, $150/t FOB Port in 2007, and estimates in early 2008 of up to $450/t FOB Port [1]. This makes long term production planning difficult since coke is the costliest raw material in steel making.

A prudent approach to this situation is not to have a standalone coke plant but a multipurpose coke facility in which multiple product value streams are optimized to reduce technical and economic risks. In such a coke plant, it would be possible to selectively extract pyrolysis gas to produce electricity, natural gas, liquid transportation fuels, fertilizer, and hydrogen in addition to coke. With this scenario in mind, a research project was initiated to consider the use of Indiana/ Illinois Basin Coal for the production of coke at mine mouth or locally at a steel production facility in addition to other ancillary products. Initial results indicate that it is possible to use blended coal with up to 50% 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, liquid transportation fuels, fertilizer and hydrogen. Since Indiana coal is less expensive than conventional metallurgical coals, coking coal costs would be significantly reduced.

In 2005 the United States produced 11,500,000 net tons of coke, but required 17,000,000 net tons for blast furnace, foundry, and related uses.[2] 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.[3] Essentially all of this comes from Kentucky, West Virginia and Virginia. The significant shortfall of needed coke has placed an enormous strain on the steel industry. A multipurpose coke plant using Indiana coal would help reduce the coke shortfall and lower the cost of coke produced.

This paper 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/Illinois Basin coal in either a mine mouth or local, environmentally friendly, high efficiency coking/coal gasification facility. Such a process would increase coke supply and production, while, at the same time reduce coal costs. Current results indicate that it is possible to use an optimized blend of Indiana/Illinois Basin and conventional metallurgical coals to produce coke that has acceptable hot and cold strength and other physical and chemical characteristics as well as producing a gas stream that can be used for the production of additional products such as Fischer-Tropsch liquid transportation fuels, fertilizer, hydrogen, and electricity. Methods to use nanoscale catalysis to reduce the carbon foot print of coking operations by producing salable chemical products from the produced carbon dioxide are also being considered.

Background

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. The inherent strategic importance of steel production throughout the world provides an 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 approximately 18 for a slot oven and approximately 48 hours for a non recovery oven to about 1100 C (2000 F). 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 depends on the nature of the coal as well as how it is processed.

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. A typical blast furnace operation indicating the location of the coke is depicted in Figure 1.[4]

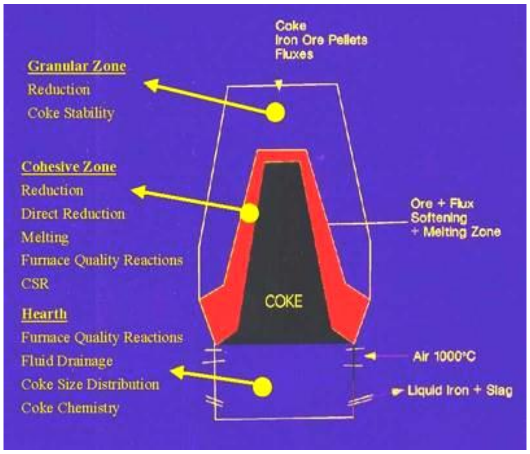

Figure 1: Typical Blast Furnace Zones

Due to the physical characteristics of Indiana coal[5], 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. The size of this coke is in two general classes. One class, often referred to as Buckwheat or Nut coke, is on the order of 1 inch x ¼ inch as compared to conventional blast furnace coke which is on the order of 1 inch x 4 inches. The other class is called coke breeze and is much finer.

Indiana/Illinois Basin coals are highly suitable for gasification and can produce good quality coke if blended with other coals or lower quality coke if carbonized alone. Some of these coals when analyzed petrographically under white light and fluorescence light show higher amounts of exudatinite, sporinite, and cutinite macerals, all grouped as Liptinite in Table I. Liptinite is reported to give higher liquefaction yield than associated vitrinite maceral.[6] Liptinite contributes to higher hydrogen and higher liquid fuel yield. Additional hydrogen contributes to higher fluidity resulting in enhancement of coking properties. Hence, some Indiana/Illinois Basin coals are good candidates for coke making and production of pyrolysis gas.

Table I. Liptinite Content in White Light and Fluorescence Light

	% Liptinite (White Light)	% Liptinite (Fluorescence Light)
Indiana Coal (c)	7.40	14.40
Indiana Coal (R)	7.20	10.80
Indiana Coal (a)	8.0	9.40
Indiana Coal (LFC)	23.6	No Data

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. Methods are under development to customize the blending process maximizing the use of Indiana/Illinois Basin coal.

Two examples of coke quality produced via pilot oven carbonization using Indiana and Illinois coals (as a single component and as blended components) are given in Table II and III[7]:

Table II. Carbonization Pilot Oven Test Results of 100% Indiana and Illinois Coals

	5-RL	7-MC	6-AQ
Coking Parameters:			
Moisture (%)	7.42	8.0	11.2
Grind(%, -3.55mm)	89.2	80.8	85.0
D.O.B.D.(kg/m3)	776	737	739
M.O.W.Pressure(kpa)	5.44	2.20	2.96
C. Time(h)	18.62	17.9	20.15
Coke Properties & Charge Contraction:			
Stability	31	25	33
CSR	27	46	30
Hardness	71	65	69
C.Size(mm)	48.5	57.8	55.1
C.Yield(%)	67.9	60.1	67.0
SHO Contraction(@52.2%)	-2.1	-10.10	-3.67

Table III: Carbonization Pilot Oven Test Results of Indiana and Illinois Coals in Blends

	TC1931	TC1933	TC1935	TC1940	TC1941	TC1951	TC1952	TC1953	TC1954	TC1995
	30% Ill 30%EHV 40% EMV	30% Ind 30% EHV 40% EMV	80% Ind 20% PC	45% Ind 15% EHV 40% EM	45% Ind 15% EHV 40%WCM	30% Ind 30% EHV 40%WCM	30% Ill 30% EHV 40%WCM	20% Ind 10% PC 30% EHV 40%WCM	20% III 10% PC 30% EHV 40%WCM	50% Ind 50%LVM
Moisture (%)	2.94		2.5	4.98	5.15	4.48	4.03	3.29	3.24	3.4
Grind (%, -3.35mm)	97.1	93.3	87.6	90.7	91.1	91.9	92.7	94.6	96.9	91.0
Dry oven bulk density (kg/m^3)	792	816	754	801	788	801	804	804	805	794
Max oven wall pressure (kPa)	5.65	6.27	2.55	4.62		3.45	4.07	4.07	3.58	7.23
Coking time (h)	16.87	16.37	16.05	17.13	17.03	17.05	17	16.6	16.1	17.02
Stability	61	60	42	58	63	57	61.1	60.5	60.7	62
CSR	61	68	24	57	65	65	70	72	71	66
CRI	30	22	44	32	24		21	20		28
Hardness	70	70	51.3	70	68	70	70	69	68	72
Coke size (mm)	61.73	65.53	70.9	70.74	69.3	62.8	59	61.3	64.2	62.6
Coke yield (%)	73.58	70.15	69.6	73.39	74.6	74.9	76.3	78	76.9	74.9
SHO contraction %	-7.99	-9.57	-11.94	-11.13	-10.14	-12.82	-7.93	-10.59	-12.93	
Coke sulfur (%)						0.66		0.93		
Coke ash (%)						11.1		8.9		

Note: Ill=Illinois Coal; Ind=Indiana Brazil Formation coal; WCM=Western Canadian medium volatile coal, EHV=Eastern high volatile; EMV=Eastern medium volatile coal; LVM=Alabama low volatile coal; PC=petroleum coke

In Table III it can be observed that a blend of 45% of Indiana Coal can be used in a slot oven to produce acceptable coke quality (see TC1941 containing 45%Indy-15%EHV-40%WCM). TC 1995 (50% of Indiana Brazil Block coal when blended with 50% of LVM Alabama coal) also produces acceptable coke quality in a slot oven. If such a blend is carbonized in a Heat Recovery/Non Recovery Coke Oven, it is anticipated that there will be similar or better performance in coke quality. One example is depicted in Table IV where a blend containing 12% of Indiana coal was carbonized in six ovens of a Non Recovery Battery producing excellent quality coke[8].

TableIV. Non Recovery Six Oven Carbonization Test Results

	65%HVM, 35%MVM/LVM (with 12% Indiana coal)	64%HVM, 35%MVM/LVM
CSR	70	71
CRI	21	20
Stability	64	64
Hardness	70	72

Another approach to increasing the percentage of Indiana/Illinois Basin coal for coke production involves locating the coke produced from this coal in upper regions of the blast furnace where higher reactivity is less of a concern. In such regions there is also less mechanical pressure on the individual pieces of coke since there is less material above it. This allows coke of reduced strength to be used.

The location of a multipurpose coke production process could be either near or at a coal mine or at an existing steel production facility. The choice of which 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 developing a new approach to coke production in which there are multiple value streams from the conventional coke production process. These value streams include coke, electricity, liquid transportation fuels, fertilizer, and hydrogen. Product value is maximized by selectively extracting a portion of the coke oven gas at various stages of the coking process with a composition suitable for production of the particular ancillary product. Figure 2 depicts the general concept for extraction of gas from a non recovery coke oven.

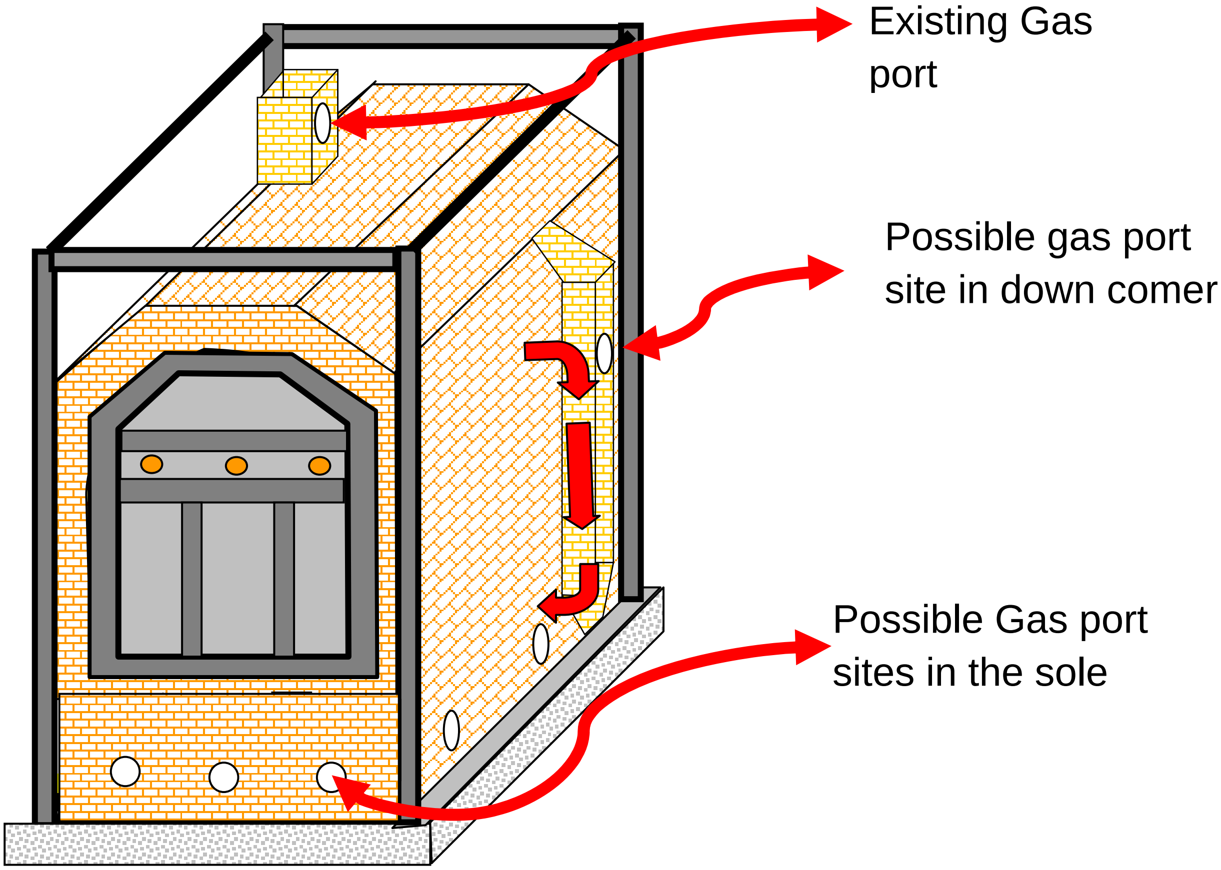

Figure 2. Possible Gas Extraction Locations

It will provide a new source of coke, produced at mine mouth or locally, using Indiana/Illinois Basin coal which is up to 30% lower in cost. This coal has a higher volatile content than what is normally considered for current coking operations. But, the process under development uses the gas from the volatile material as feed material for production of other product value streams. This allows for a considerable increase in the acceptable level of volatile material in the coal. The process will also selectively extract portions of the gas and assist in increasing operational flexibility. For example, extraction of pyrolysis gas could reduce the occurrence of hot spots in the oven sole plate.

Process Description

During the coke production process pyrolysis gas evolves as the coal is heated. Producing combustible gases from solid fuels has been done since ancient times. Pyrolysis is a process in which feed material is heated with a restricted air supply. Towards the end of the eighteenth century gas was produced from coal by pyrolysis on a larger scale. In 1812 the London Gas, Light, and Coke Company commercialized gas production. The most important gas produced at this time was Town Gas. Town Gas can be produced by pyrolysis (producing gas with a heating value of 20,000-23,000 kJ/m^3) or by the water gas process (coke is converted into a mixture of equal parts of hydrogen and carbon monoxide with a heating value of approximately 12,000 kJ/m^3).[9] The coke oven was developed for the metals industry in order to provide a substitute for charcoal during the second half of the eighteenth century. One metric ton of coal typically produces 600-800 kg of blast-furnace coke and 296-358 m^3 of coke oven gas.[10] Preliminary it is estimated that from .1 - .25 barrels of liquid transportation fuel could be produced from each ton of coal used in the coking process.

Much of the pyrolysis gas from coal arises from the contained alaphatic material. This gas consists of various amounts of hydrogen, methane, carbon monoxide, nitrogen, acetylene, and carbon dioxide. The composition of the pyrolysis gas changes as the coal is heated. For example, hydrogen concentration increases with increasing temperature. Depending on its composition, Pyrolysis gas can be used in Fischer-Tropsch processes for the synthesis of liquid transportation fuels. Producing gas from coal or coke is common in the steel industry and hence new methods to maximize the value of this important material can be most beneficial. There is much discussion today about methods to produce gas from coal. It should be noted that the steel industry has been actively involved in gasifying coal and coke for over 100 years. In this context, blast furnaces can be considered large gasifiers of coke.[11] The current effort is attempting to leverage the extensive pool of steel industry coke production knowledge to reduce the technical and financial risk associated with new approaches to coal gasification.

A device has been developed to facilitate the testing of pyrolysis gas from various coal samples. This device is constructed of 316 stainless steel with the exception of the vapor traps which are copper. A schematic of the device is depicted in Figure 3.

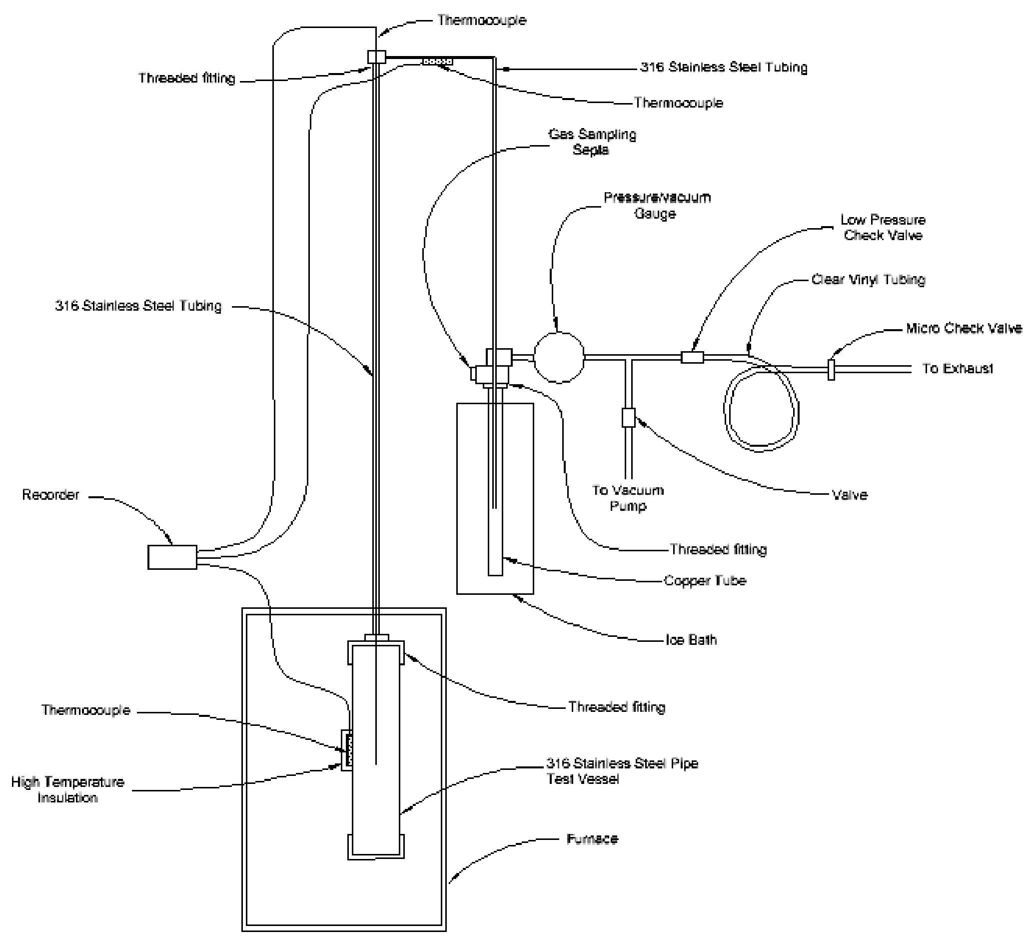

Figure 3. Pyrolysis Gas Apparatus

Coal samples are placed in the test vessel. A vacuum is then drawn to test the seals of the system and the system is backfilled with dry nitrogen. This process is repeated twice to assure removal of air from the system. The vessel is then heated in the furnace. At various temperatures a partial vacuum is momentarily drawn on the system and pyrolysis gas from the coal at that temperature refills the apparatus. Gas is then extracted from the apparatus at the particular temperature with a gas tight syringe through the sampling septa and is analyzed in a gas chromatograph.

Representative results for three different coal samples from Indiana are depicted in Figures 4 - 6. The samples were split 3 ways and tested on different days.

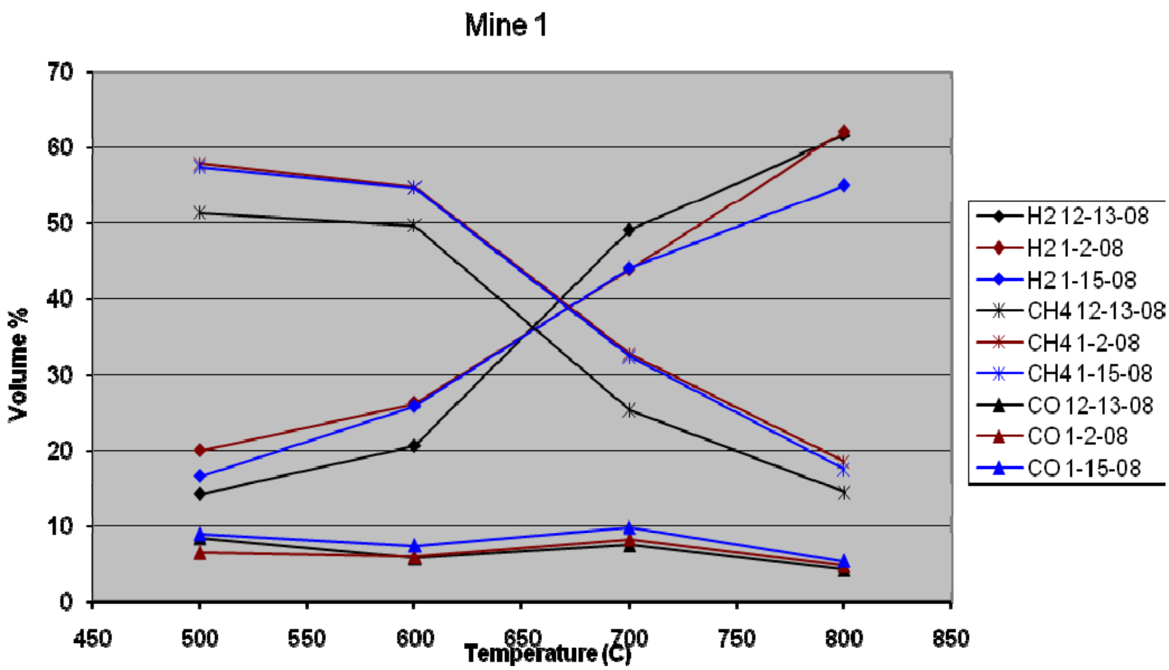

Figure 4. Pyrolysis Gas Analysis, Mine 1

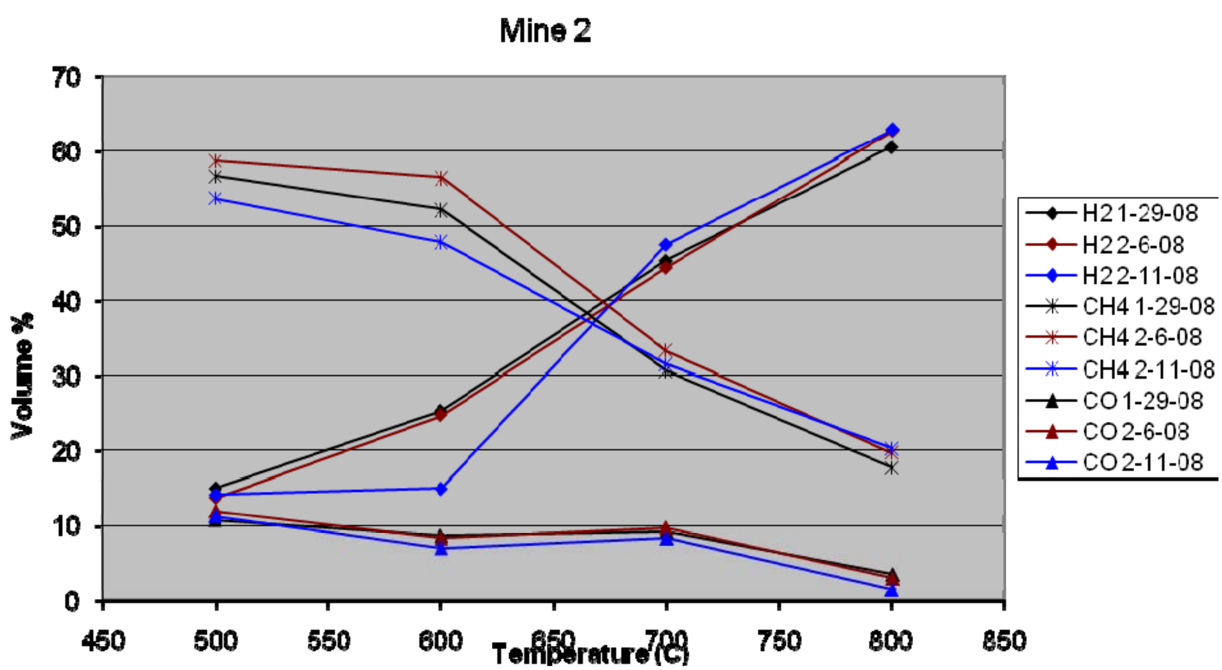

Figure 5. Pyrolysis Gas Analysis, Mine 2

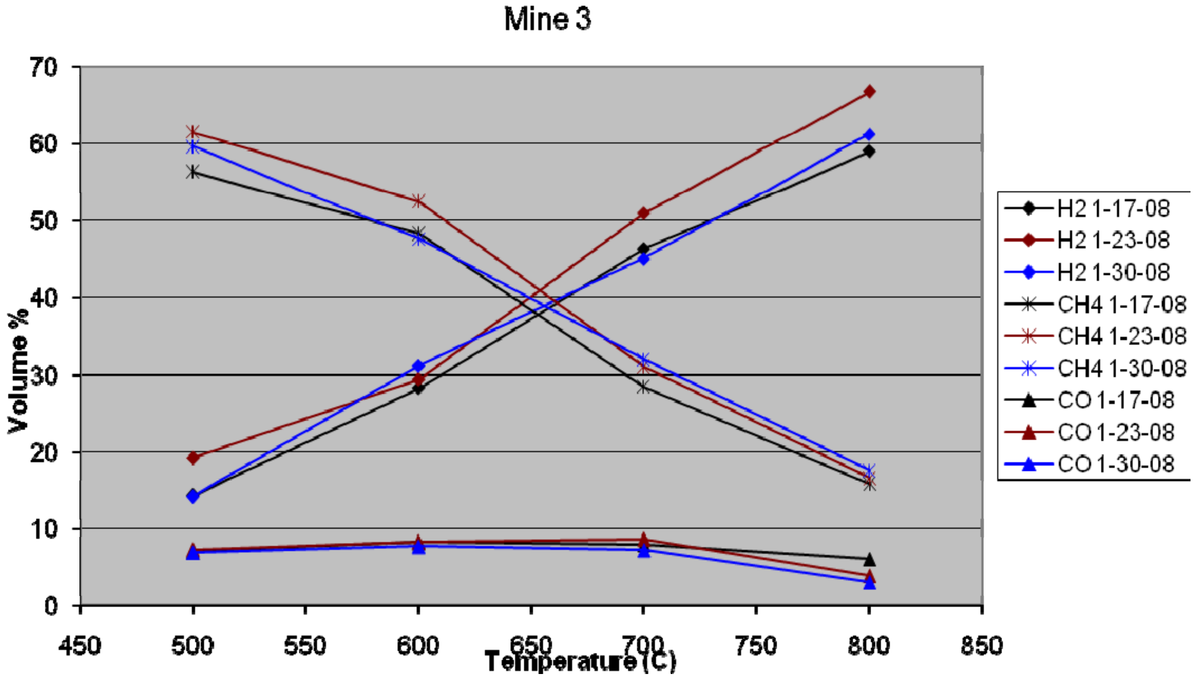

Figure 6. Pyrolysis Gas Analysis, Mine 3

As can be observed in Figures 4 - 6, the composition of the pyrolysis gas varies with temperature for each type of coal. For a Fischer-Tropsch process producing a product that can be converted to liquid transportation fuel, a ratio of hydrogen to carbon monoxide of up to 4 to 1 is acceptable with an iron catalyst. Pyrolysis gas from the tested coals in the temperature range of 500-700 C could be readily used for such a process. The temperature range could be extended further by post processing if needed. A coal blending methodology is currently under development that will simultaneously optimize coke physical and chemical properties, such as CSR, as well as pyrolysis gas composition. By blending various coals it will be possible to adjust the temperature ranges over which the pyrolysis gas can be used for different purposes including production of liquid transportation fuels, fertilizer, hydrogen, and electricity. The coal blend will be optimized to maximize the value for the various products depending on market conditions at the time. The blending process is being optimized to maximize the value of both the coke and gas streams while minimizing coal costs. A graphic representation of the process is depicted in Figure 7.

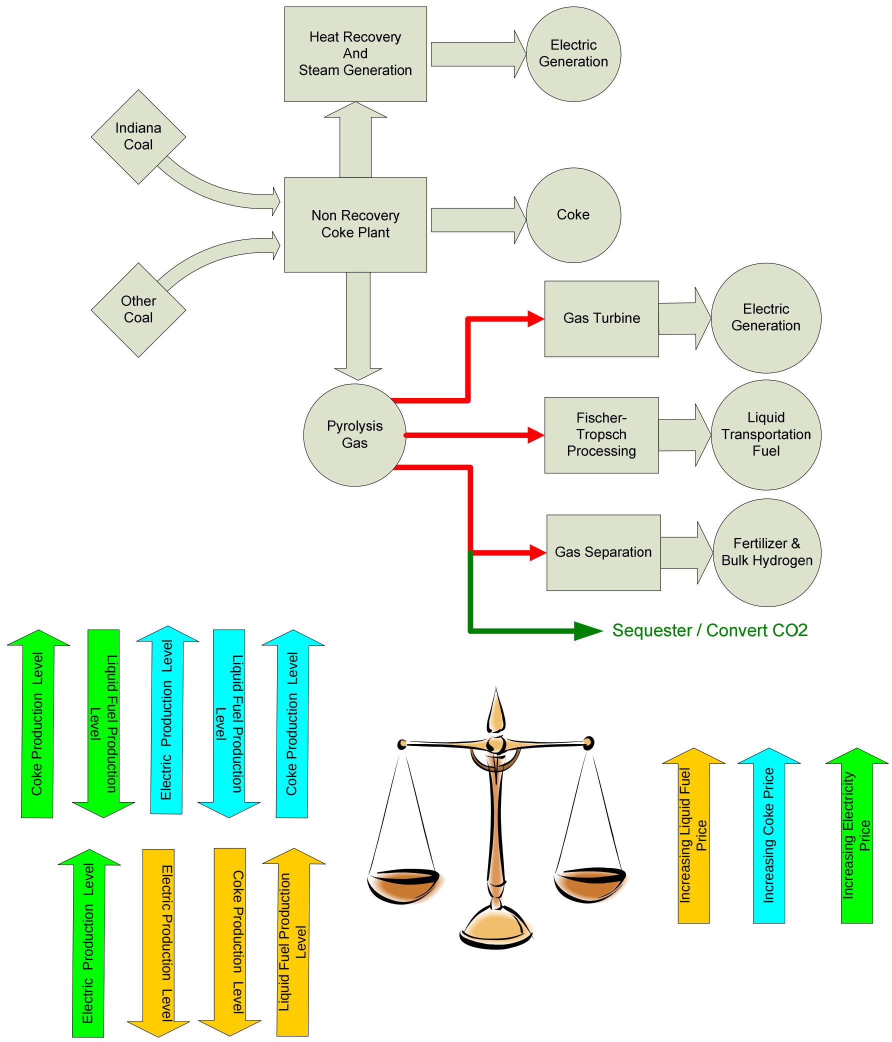

Figure 7. Process Flow Concept

The technology and financial risk of this process will be reduced by leveraging existing process technology. This process can provide a new direction and approach for the production of coke in the future that optimizes value over multiple product streams while reducing both business and technological risk.

Summary

The research described in this paper is developing a new approach to the production of coke. In this process multiple optimized value streams are produced from a coke facility located at mine mouth or locally at an existing plant. As part of the process, lower cost Indiana/Illinois Basin coals will be blended with conventional metallurgical coals. The blending process will be optimized to meet coke quality requirements and simultaneously to obtain a pyrolysis gas composition suitable for production of ancillary products including liquid transportation fuels, fertilizer, hydrogen, and electricity. By using lower cost Indiana/IllinoisBasin coal it will be possible to reduce net coal costs. This process can provide a new direction and approach for the production of coke in the future that optimizes value over multiple product streams while reducing both business and technological risk by leveraging existing coke production technology. Methods to reduce the carbon foot print of the plant are also being considered.

Acknowledgement

The author would like to thank the Center for Coal Technology Research and the Illinois Clean Coal Institute for their support of this effort. Without their assistance this research would not have been possible.

References

[1] Private communication, Industry Sources; CRU Presentation at Intertech 2006
[2] Valia, H., Mastalerz, M., “Indiana Coals and the Steel Industry, Indiana Geological Survey Special Report 64,” 2004.
[3] Bowen, B., Holland, F., Sparrow, F., Rardin, R., Gotham, D., Zuwei, Y., Black, A**.,** “*Expanding the Utilization of Indiana Coals,*” Center for Coal Technology Research, Purdue University, August 2004.
[4] Valia, H., “Coke Production for Blast Furnace Ironmaking”, American Iron and Steel Institute.
[5] Mastalerz, M., Drobniak, A., Rupp, J., Shaffer, N., “Characterization of Indiana’s Coal Resource: Availability of the Reserves, Physical and Chemical Properties of the coal, and the Present and Potential Uses,” Indiana Geological Survey Open-File Study 04-02.
[6] Elliott, M.A., *Chemistry of Coal Utilization*, 2nd Supplementary Volume, Wiley, NY, pg. 155, 1981.
[7] Valia, H. S., “Coal Cost Reduction Using Low Rank Coals”, AIST, pg. 170-175, March 2006.
[8] Valia, H. S., “Coal Cost Reduction Using Low Rank Coals”, AIST, pg. 170-175, March 2006.
[9] Gumz, W., *Gas Producers and Blast Furnaces*, New York, John Wiley & Sons, 1950.
[10] *The Making Shaping and Treating of Steel*, Association of Iron and Steel Engineers, Herbick & Held, Pittsburgh, 1985.
[11] Gumz, W., *Gas Producers and Blast Furnaces*, New York, John Wiley & Sons, 1950.