Clean Production of Coke from Carbonaceous Fines

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Executive Summary

In order to produce steel (a necessary commodity in developed nations) using conventional technologies, you must have metallurgical coke. Current coke-making technology pyrolyzes high-quality coking coals in a slot oven, but prime coking coals are becoming more expensive and slot ovens are being shut-down because of age and environmental problems. The United States typically imports about 4 million tons of coke per year, but because of a world-wide coke scarcity, metallurgical coke costs have risen from about $77 per tonne to more than $225. This coke shortage is a long-term challenge driving up the price of steel and is forcing steel makers to search for alternatives.

Combustion Resources (CR) has developed a technology to produce metallurgical coke from alternative feedstocks in an environmentally clean manner. The purpose of the current project was to refine material and process requirements in order to achieve improved economic benefits and to expand upon prior work on the proposed technology through successful prototype testing of coke products. The ultimate objective of this project is commercialization of the proposed technology.

During this project period, CR developed coke from over thirty different formulations that meet the strength and reactivity requirements for use as metallurgical coke. The technology has been termed CR Clean Coke because it utilizes waste materials as feedstocks and is produced in a continuous process where pollutant emissions can be significantly reduced compared to current practice. The proposed feed material and operating costs for a CR Clean Coke plant are significantly less than conventional coke plants. Even the capital costs for the proposed coke plant are about half that of current plants. The remaining barrier for CR Clean Coke to overcome prior to commercialization is full-scale testing in a blast furnace. These tests will require a significant quantity of product (tens of thousands of tons) necessitating the construction of a demonstration facility. Talks are currently underway with potential partners and investors to build a demonstration facility that will generate enough coke for meaningful blast furnace evaluation tests. If the testing is successful, CR Clean Coke could potentially eliminate the need for the United States to import any coke, effectively decreasing US Steel industry dependence on foreign nations and reducing the price of domestic steel.

Project Description

Original Project Goals and Objectives

Sixty percent of the world's crude steel production is in blast furnaces[1], and metallurgical coke is required to operate a blast furnace. Coke is the most important feedstock in a blast furnace; it reduces the iron ore to metallic iron, supports the burden, provides thermal energy, and maintains permeability for the flow of gases and liquids inside the furnace. Due to coke shortages and foreign control of markets, typical metallurgical coke costs have risen from about $77 per tonne in 1999 to $121 in 2002 to $165 in 2003[2]. Current 2004 long-term contract costs are reportedly even higher; this coke shortage is a long-term, world-wide challenge[3]. Current coke-making technology uses high-quality coking coals and pyrolyzes them in a slot oven. Prime coking coals are becoming scarce, hard to mine, and expensive. In 2003, the cost of steam coal for electricity was $25/ton while the cost of met-coal for coke production was over $50/ton[4]. The US currently imports about 4 million tons of coke per year, mainly from China. This places nationally strategic steel production under the influence of a foreign country. Additionally, the traditional slot ovens used to produce coke are an environmental hazard due to the release of particulate and sulfur gases, as well as emissions of carcinogenic and mutagenic polycyclic aromatic hydrocarbons and benzene-toluene-xylene's. These factors all increase the cost of steel production and force steel makers to search for alternatives.

Combustion Resources has developed a technology to produce metallurgical coke from alternative feedstocks in an environmentally clean manner. The original project objective was to refine material and process requirements in order to achieve improved economic benefits and to expand upon prior work on the proposed technology through successful prototype testing of both metallurgical and foundry coke products. The final goal is to commercialize the coke technology (CR Clean Coke).

The tasks set forth to accomplish the project objectives and goals were:

Task 1. Obtain Raw Materials and Equipment
Task 2. Conduct Studies to Reduce Feed Material Costs
Task 3. Conduct Parametric Studies to Reduce Cost-Intensive Processes
Task 4. Identify a Production Calcining Furnace
Task 5. Conduct Prototype Testing of Foundry Coke
Task 6. Conduct Prototype Testing of Metallurgical Coke
Task 7. Solidify Industrial Partners
Task 8 . Revise Business Plan
Task 9. Project Management and Reporting

Variance from Original Goals and Objectives

The project was very successful and each task was completed with the exception of Task 5, which stated that CR would conduct prototype testing of foundry coke. As the project progressed, a high degree of interest was received from the steel industry in metallurgical coke and it was decided to direct project funds and time solely towards met-coke. Producing foundry coke is still of future interest, but the greatest potential for commercialization of this technology comes from met-coke production.

Combustion Resources performed a significant amount of additional work that was not included in the DOE sponsored tasks. Multiple trips were made to potential partner companies to present the technology and conferences were attended where the technology was presented and displayed. This additional work was not included in the invoices to the DOE and will greatly aid in the solidification of future strategic partners.

Discussion of Work Performed

Over the past two years, much work has been done to complete the outlined tasks. Each task is listed below with a short discussion of the work done in each area.

Task 1. Obtain Raw Materials and Equipment

In order to further the research and development of CR Clean Coke, adequate raw materials and other supplies were identified and acquired. At first this task proved quite challenging as many coal suppliers were not willing to send a couple barrels of coal to a research facility, but eventually willing suppliers were found and multiple and varied feedstocks were obtained. As the project continued to progress and the promising results were displayed at various conferences, finding eager suppliers became much easier. A large variety of feedstocks were obtained including the following:

Steam coals	5	Recovered waste coals	2
Petroleum coke	4	Anthracite	2
Low-vol coking coals	3	Mid-volatile coals	2
Biomass waste	5	Coal fines	3
Coal tars	3	Coke breeze	1

These feedstocks were acquired from the across the US, Canada, and India, and vary in volatiles percentage (3-80%), ash (0.2-24%), sulfur (0.4-4.6%), and free-swelling index (0-9).

The cost, quality, and quantity of each feedstock was noted and incorporated into the business plan for future production evaluation. As the project evolved, feedstock acquisition efforts were focused on obtaining high quantity, low-cost feedstocks. Oftentimes, the feedstocks were acquired from potential partners interested in commercializing the technology.

Task 2. Conduct Studies to Reduce Feed Material Costs

A major portion of all the work performed for this project was centered on reducing the feed materials costs. Not just any feed material will work to produce high-quality coke; metallurgical coke must meet specific blast furnace requirements. Table 1 contains a summary of many of the requirements for metallurgical coke.

Table 1: Metallurgical Coke Properties

Property	Typical Specified Value
Composition	
Ash %	10.5-12.5
Sulfur %	0.6-1.0
Alkali %	0.2-0.3
Moisture %	<1.0
Volatile Matter %	0.4-1.1
Phosphorus %	<0.055
Size, cm	
Mean	>5
Strength, Reactivity	
CSR	56-65
CRI	25-30
Stability	>60
Hardness	68-70

Possibly the most important requirement for metallurgical coke is the CSR score. The CSR test attempts to mimic conditions in a blast furnace and provides information about two different parameters; (1) the strength of the briquettes after reacting with CO_2, and (2) the amount of dust produced by CO_2 attack and bed agitation. Fine dust can be detrimental to blast furnace operation since it can decrease the permeability of the bed, requiring increased blast pressure to force the air up through the bed. Each formulation of CR Clean Coke was subjected to the CSR test, along with other tests, to determine if it would be suitable for blast furnace use.

The best coal/coke/tar ratios were determined. Multiple formulations using high percentages of petroleum coke met the industry specification for CSR, but met-coke specifications require a low sulfur percentage and low-sulfur petroleum coke is not readily available and is expensive. A significant effort was directed to finding lower cost feed materials that could imitate the properties of pet-coke in the CR Clean Coke briquettes. Through multiple tests, blends of coal and pet-coke were found that produced high quality briquettes. The cost of different feed materials was always part of the design of formulations. Table 2 provides an overview of the different feedstocks that yielded high-quality coke briquettes, both in the small test size and in the larger prototype coke size.

In addition to blending feedstocks, it was found that by changing other process parameters, raw material costs could be lowered. Varying briquetting pressure and calcining temperature ramp rates allowed production of acceptable coke that contained smaller quantities of expensive feedstocks. There were tradeoffs between reducing the cost of feedstocks and reducing the operating costs and energy requirements of the plant.

Table 2: Summary of Feedstocks in Acceptable Coke Briquettes

Feedstock	Number of Feedstocks	No. Products
A. Small-scale briquettes		
Steam coals (hi-vol)	2	2
Mid vol. coals	2	5
Low-vol coking coals	4	19
Pet cokes	3	28
Tars	2	29
Coke breeze	1	1
Coal fines	1	4
Anthracite	2	7
Total	**17**	**30**
B. Large-scale briquettes		
Low-vol coking coals	2	6
Pet cokes	1	7
Tars	1	7
Coal fines	1	1
Anthracite	1	2
Total	**6**	**7**

Early in CR Clean Coke development it was established that relative reaction rates of various feedstocks influenced coked briquette strength (CSR). To aid in the selection of feedstocks, reaction rates in CO_2 were measured with a thermogravimetric analyzer. Later, a more extensive research effort was initiated. This effort was intended to develop a method to predict successful feedstock formulations from low-cost raw materials without the need for time intensive testing. The extensive research was performed to:

(a) Make use of the extensive CR data base to identify the importance of various parameters.
(b) Make use of applicable correlative methods and insights into CRI and CSR for slot oven coke production[5].
(c) Develop a physically-based model of the coke–CO_2 reaction process in a briquette in order to evaluate the importance of selected variables on product reactivity and strength (CSR).
(d) Measure fundamental properties of feedstocks including proximate and elemental analysis, particle size distribution and reactivity.
(e) Measure fundamental properties of green, coked and reacted briquettes including true and apparent density, void fraction, ultimate and proximate analysis and internal surface area.

(f) Measure extent of reaction (CRI) and strength (CSR) for briquettes of various compositions as a function of reaction time from 30 minutes to two hours.

Through this research effort, it was discovered that correlations developed for slot oven coke production do not completely describe the results seen from CR Clean Coke technology. This innovative technology has the ability, through careful formulation and briquette preparation, to take feedstocks that can not be accepted in a traditional slot oven and make them into met-quality coke. The model of the coke-CO_2 reaction can predict the reactivity and strength of coke briquettes to a particular accuracy. Work continues on this research effort to fully understand how the possible variables in CR Clean Coke production affect final product properties.

Task 3. Conduct Parametric Studies to Reduce Cost-Intensive Processes

There are three major cost-intensive processes in the production of CR Clean Coke; (1) crushing and briquetting the feed material, (2) calcining the briquettes, and (3) cleaning the coke oven gas (COG). Specific equipment that can perform each task was identified and compared on the basis of capital cost, operating costs, and functionality.

To aid in the selection of machinery for the coke plant, a preliminary plant design was completed. The plant design included four main sections: feedstock preparation, briquetting, calcining, and COG cleanup

Some of the raw materials used to produce the coke product must be crushed and ground. Studies were performed to optimize the cost of grinding the feed material with product performance. A mill that dries the materials as it grinds was chosen, eliminating the need for an additional drying oven.

For calcining, multiple furnaces were identified that could accomplish the task. Some furnaces heat the oven by direct burning of the pyrolysis gas released from the green briquettes and others were indirectly heated by either electricity or gas. With these furnaces, there seemed to be a tradeoff between operating costs and capital costs.

The COG cleaning system was designed from currently available technology. Pyrolysis gas cleaning is associated with high capital costs. However, COG consists mainly of hydrogen and methane and its value after it is cleaned offsets the costs of cleaning. There are also options for using uncleaned COG such as lime and cement kiln firing if commercialization partners wish to lower the risks of a high capital investment.

Task 4. Identify a Production Calcining Furnace

The calcining furnace is the single most important and expensive piece of equipment used to produce CR Clean Coke. There are many furnace designs that will adequately pyrolyze green briquettes into coke. The desired aspects of a furnace include:

- Appropriate temperature range
- Simple operation
- Low investment cost

- High efficiency
- Zone control
- High throughput
- Low operating and maintenance cost
- No vagrant emissions

There are many directly fired furnaces in use today that could be applied to production of CR Clean Coke. The furnace designs include rotary hearths, rotary kilns, roller hearths, tunnel kilns, and others. However, directly fired furnaces combust the tars released from the green briquettes, causing a need to buy tar as a feedstock. Therefore, much effort was expended to find suitable ways to modify direct fired furnaces to be indirectly fired. Indirect firing is generally more complex, but the byproducts recovered from indirect firing contain substantial amounts of energy that are useful within the process.

Many basic drawings and price quotes of acceptable furnaces were obtained from various furnace designers. The best candidate was a roller kiln. It is simple to operate, can be indirectly heated with either gas or electricity, has easy temperature zone control, and has a high-throughput per capital investment dollar. The furnace is also very clean; there are no issues of gas leakage through doors like traditional slot ovens. The coke is cooled before exiting the furnace, so no vagrant emissions result from the coking process.

Task 5. Conduct Prototype Testing of Foundry Coke

With the shortage and resulting high price of metallurgical coke causing great concern in the steel industry, foundry coke testing was delayed. Foundry coke is still of future interest, and it is assumed that the technological gains accomplished in producing met-coke will apply to future foundry coke research.

Task 6. Conduct Prototype Testing of Metallurgical Coke

Met-coke, using many different formulations, was made in the laboratory at Combustion Resources. The small briquettes (1”x 2”) produced in the laboratory were adequate for analysis, but too small for blast furnace use. For this reason, full-size prototype briquettes (3” x 2.5”) were made at the K.R. Komarek test facility on a commercial-scale briquetter. The prototype briquettes were then tested at CR. It was discovered that prototype briquette properties mirrored the small, lab-scale coke briquette properties. Most test results for quality were the same for both briquette sizes, except CSR numbers which are slightly lower for the full-size coke briquettes. However, the CSR numbers are still significantly higher than the specified 60 for traditional metallurgical coke. Table 3 gives some CSR testing numbers for both the lab-scale and the full-size coke briquettes.

Table 3: Lab-Scale and Full-Size Coke CSR Numbers

	Formula 1		Formula 2		Formula 3		Formula 4	
	Large	Small	Large	Small	Large	Small	Large	Small
CSR	74	79	73	76	70	77	72	77

Many different process parameters were able to be studied on the prototype briquettes. The pressure applied to the briquettes was varied on the commercial briquetter, as well as the amount of binder added to the carbon mix. The heating ramp rate for the prototype briquettes was also studied to optimize product quality and production time. Overall, the prototype coke performed exceptionally well.

Task 7. Solidify Industrial Partners

Solidifying industrial partners is one of the key tasks to get the product to market. Industrial partners were sought who could provide feedstocks, contacts to coke consumers, production experience, and possible start-up capital. CR found multiple suppliers of feedstock and is currently working with potential partners to commercialize the coke product.

Task 8 . Revise Business Plan

Based on the results of testing and potential partner desires, CR has revised its business plan. The market potential of CR Clean Coke is very good; the US Steel industry is eager for additional suppliers of domestic coke. The major market barrier is expected to be the unfamiliarity of the coke briquettes to current lump coke users. These users include industries that tend to be very conservative and resistant to change. This resistance can be attributed to the enormous capital cost generally associated with blast furnaces. However, with the sharp increase in coke costs, these users are more amenable to alternative cokes.

CR has developed and advanced the coke as far as can be expected in the laboratory and testing in a blast furnace is the next step needed. In order to run a thorough testing program, tens of thousands of tons of coke are required. To produce this much coke, a demonstration facility needs to be constructed and strategic partners are needed for coke plant construction and management. Significant attention is currently being directed toward aligning strategic partners and finalizing plant design.

The economics of CR Clean Coke are highly favorable. Depending on the feedstocks utilized, the coke is expected to cost significantly less to produce than conventional coke.

Task 9. Project Management and Reporting

The project was overseen by Dr. Craig N. Eatough. Semi-annual reports were regularly submitted. Dr. L. Douglas Smoot also managed portions of the project.

Conclusions and Recommendations for Future Work

The project was successful in moving the technology towards commercialization. With the exception of prototype testing of foundry coke, the outlined tasks were completed. CR was able to obtain a variety of raw materials from coal mines, coal fines recovery operations, petroleum coke producers, and others. Equipment to crush the feed was identified, and an appropriate calcining furnace was identified. Prototype coke was produced from a wide variety of feedstocks and it meets the requirements for metallurgical quality coke. Very positive discussions are underway with potential commercialization partners to produce CR Clean Coke in large enough quantities to be tested in a blast furnace. For the future, in order to attract the attention of serious investors, the coke product must be proven in a blast furnace. In order to perform meaningful blast furnace tests, a large supply of coke is needed (30-40 thousand tons). This will require the construction of a demonstrations facility requiring about $20M in capital and raw material investment. Ultimately, the technology has the potential to eliminate the need for the US Steel industry to import coke from foreign countries.

References

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6. Amendola, Gary A., "Preliminary Assessment of Energy Requirements and Environmental Releases for Four Alternative Steel Making Technologies," Amendola Engineering, Inc., July 1999.

Appendix A

Final Task Schedule

Task Number	Task Description	Task Completion Date				Progress Notes
		Original Planned	Revised Planned	Actual	Percent Complete	
1	Obtain Raw Materials and Equipment	11/30/02	5/30/03	2/25/04	100%	completed
2	Conduct Studies to Reduce Feed Material Costs	2/28/04	7/30/04	8/15/04	100%	completed
3	Parametric Studies of Cost-Intensive Processes	2/28/04	5/30/04	3/31/04	100%	completed
4	Identify a Production Calcining Furnace	2/28/04	6/30/04	7/15/04	100%	completed
5	Conduct Prototype Testing of Foundry Coke	7/31/04			0%	of future interest
6	Conduct Prototype Testing of Metallurgical Coke	7/31/04	8/31/04	8/31/04	100%	completed
7	Solidify Industrial Partners	8/31/04			90%	very positive interest
8	Revise Business Plan	8/31/04		8/31/04	100%	completed
9	Project Management and Reporting	8/31/04		8/31/04	100%	completed

Appendix B

Final Spending Schedule

Final Spending Schedule **Project Period: 09/01/02 to 08/31/04**

Task	Approved Budget	Final Project Expenditures
Task 1 Obtain Raw Materials and Equipment	28,000	28,416
Task 2 Conduct Studies to Reduce Feed Material Costs	44,000	79,667
Task 3 Parametric Studies of Cost-Intensive Processes	44,000	42,477
Task 4 Identify a Production Calcining Furnace	44,000	44,582
Task 5 Conduct Prototype Testing of Foundry Coke	34,000	
Task 6 Conduct Prototype Testing of Metallurgical Coke	34,000	36,535
Task 7 Solidify Industrial Partners	2,000	3,500
Task 8 Revise Business Plan	6,000	8,045
Task 9 Project Management and Reporting	21,035	21,314
Total	257,035	264,506
DOE Share	200,000	200,000
Cost Share	57,035	64,506

Appendix C

Final Cost Share Contributions

Final Cost Share Contributions

Funding Source	Approved Cost Share		Final Contributions	
	Cash	In-Kind	Cash	In-Kind
Combustion Resources		57,035	9,021	55,485
Total		57,035	9,021	55,485
Cumulative Cost Share Contributions				64,506

Appendix D

Energy Savings Metrics

The following is a comparison of the energy requirements for two coke production facilities; the CR Clean Coke plant and a Heat Recovery plant.

A One Million Ton per Year Capacity CR Clean Coke Plant:

CR Clean Coke production has four main process steps: feedstock preparation, briquetting, calcining, and gas cleanup. The feedstock preparation and briquetting steps use small amounts of energy compared to the calcining step, and the proposed calcining furnace is assumed to be very efficient. Therefore, the energy needed to prepare, briquette, and calcine one ton of coal in the CR Clean Coke plant is assumed to be the same as the energy needed to calcine one ton of coal in a Heat Recovery plant. The CR Clean Coke plant will clean the coke-oven-gas (COG) and recover byproducts in a similar manner to existing byproduct plants with the clean COG being used to fire the calcining furnace. One main difference between the CR Clean Coke plant and existing coke plants is the amount of product loss during calcining. Traditional coke plants lose 7.5% of their feedstock to coke breeze and 28% of their feedstock to volatile components. CR Clean Coke recycles all fines generated in the process and only loses 20% of the feedstock to volatile components.

A One Million Ton per Year Capacity Heat Recovery Plant:

Although most current coke is produced in byproduct plants, all new coking capacity proposed to be built in the United States is of the Heat Recovery plant type because of environmental concerns over vagrant emissions. In a Heat Recovery plant coal is charged into a slot-type oven and heated for several hours to become coke. The most efficient plants partially burn the volatiles coming off the coal bed as heat for the oven and then recover the rest of the volatiles for use in an electricity cogeneration facility. Since the volatile components are combusted before they are cleaned, no byproducts are recovered. It can be assumed that the coke breeze generated in a Heat Recovery plant is wasted and can not be used by the blast furnace.

Discussion of Energy Savings:

The biggest energy savings between the two plants is in the increased utilization of the feedstocks. More of the feedstock in the CR Clean Coke plant goes directly to product; therefore, less feedstock is needed to produce one ton of coke than in an existing coke plant. A decrease in the amount of feedstock needed means less feedstock to transport, and less feedstock to mine. The inherent energy of the feedstock (assumed to be all coal for energy calculation purposes) and the energy necessary for transporting the feedstock are all quantified. Additional energy savings would be seen in the extraction or mining of the coal and in disposal of coke breeze.

Energy Savings Metrics

Type of Energy Used	Current Technology (BTU/yr/unit)	Proposed Technology (BTU/yr/unit)	Energy Savings (BTU/yr/unit)	Estimated Number of Units in U.S. by 2010 (units)	Energy Savings by 2010 (BTU/yr)
Diesel	9.793×10^{10}	7.984×10^{10}	1.81×10^{10}	2	3.62×10^{10}
Coal	3.878×10^{13}	3.162×10^{13}	7.16×10^{12}	2	1.43×10^{13}
Electricity	-2.172×10^{12}		-2.172×10^{12}	2	-4.34×10^{12}
Steam	-1.802×10^{12}		-1.802×10^{12}	2	-3.60×10^{12}
COG		-1.972×10^{12}	1.972×10^{12}	2	3.94×10^{12}
Byproducts		-1.119×10^{12}	1.119×10^{12}	2	2.24×10^{12}
Total	3.490×10^{13}	2.860×10^{13}	6.30×10^{12}	2	1.26×10^{13}

The Heat Recovery plant produces energy in excess of its oven needs in the form of electricity and steam. The CR Clean Coke plant produces energy in excess of its furnace needs in the form of COG and byproducts. Energy calculations are based on values taken from a presentation given by Gary Amendola at the EPA sponsored Cokemaking Stakeholder Meeting in July of 1999[6]. COG and byproduct values were taken from traditional byproduct coke plant numbers and adjusted for volatile release percentages.