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UPGRADING LOW RANK COAL USING THE KOPPELMAN SERIES C PROCESS

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INTRODUCTION

Development of the K-Fuel® technology began after the energy shortage of the early 1970s in the United States led energy producers to develop the huge deposits of low-sulfur coal in the Powder River Basin (PRB) of Wyoming. PRB coal is a subbituminous C coal containing about 30 wt % moisture and having heating values of about 18.6 megajoules/kg (8150 Btu/lb). PRB coal contains from 0.3 to 0.5 wt % sulfur, which is nearly all combined with the organic matrix in the coal. It is in much demand for boiler fuel because of the low-sulfur content and the low price. However, the low-heating value limits the markets for PRB coal to boilers specially designed for the high-moisture coal. Thus, the advantages of the low-sulfur content are not available to many potential customers having boilers that were designed for bituminous coal.

This year about 250 million tons of coal is shipped from the Powder River Basin of Wyoming. The high-moisture content and, consequently, the low-heating value of this coal causes the transportation and combustion of the coal to be inefficient. When the moisture is removed and the heating value increased the same bundle of energy can be shipped using one-third less train loads. Also, the dried product can be burned much more efficiently in boiler systems. This increase in efficiency reduces the carbon dioxide emissions caused by use of the low-heating value coal. Also, the processing used to remove water and restructure the coal removes sulfur, nitrogen, mercury, and chlorides from the coal. This precombustion cleaning is much less costly than stack scrubbing.

PRB coal, and other low-rank coals, tend to be highly reactive when freshly mined. These reactive coals must be mixed regularly (every week or two) when fresh, but become somewhat more stable after they have aged for several weeks. PRB coal is relatively dusty and subject to self-ignition compared to bituminous coals. When dried using conventional technology, PRB coal is even more dusty and more susceptible to spontaneous combustion than the raw coal. Also, PRB coal, if dried at low temperature, typically readsorbs about two-thirds of the moisture removed by drying. This readsorption of moisture releases the heat of adsorption of the water which is a major cause of self-heating of low-rank coals at low temperature.

Ingram and Rimstidt¹ attribute increased moisture in naturally oxidized coals to the effects of weathering and an increase in organic acids, that are known to attract water to coal particles. Thus, an effective drying process must destroy or remove carboxylic acids and phenolic-like compounds to prevent readsorption of moisture.

The University of North Dakota Energy Research Center has explored the use of various techniques to upgrade low-rank coals². They found that coals processed at low temperature readsorb moisture and return to essentially the original equilibrium moisture level. In contrast, they found that the processes using high temperatures to alter the structure of the coal particles resulted in reduced readsorption of moisture. They also concluded that the lowered equilibrium moisture levels resulted from the rejection of carbon dioxide by the decarboxylation reactions which occur during high-temperature processing.

The readsorption of moisture by coals dried at temperatures much lower than about 260°C (500°F) is consistent with experience³ at Western Research Institute.

Mr. Edward Koppelman has developed technologies for over 50 years. U.S. aircraft used deicing and defogging equipment developed and produced by his organization during World War II. He has developed three-dimensional weaving techniques for fiber glass used to reinforce complex-shaped military components; antenna systems used on aircraft; the technology used to produce perlite insulation; and has patented processes used to dry wood under vacuum using radio frequency heating. Mr. Koppelman has also developed processes to recover furfural, carbon black, activated carbon, and sodium silicate from rice hulls. His activities have included exploration and production of oil and gas, and mining uranium.

Mr. Koppelman has worked to develop processes which produce clean-burning, high-value fuel from low-rank coals, wood waste materials, and biomass products for over 20 years. Early development work on the K-Fuel ® processes was conducted at the Stanford Research Institute (SRI) in Palo Alto, California. His work at SRI led to the development of numerous processes and many patents were awarded for these processes. The predecessor company of KFx Inc. was started in 1984 to further the development of the Series A Process developed by Mr. Koppelman at SRI.

In 1986 the Series A process pilot plant was relocated to the present site located near Gillette, Wyoming to develop the huge resource of low-rank PRB coal. Since that time KFx Inc. has developed and tested processes to upgrade PRB coal at the site. In the early 1990s Koppelman's Series B Process was tested at the same site. After successful testing, the technology for the Series B Process, in which coal is heated by direct contact with steam at high pressure, was licensed to Heartland Fuels Corporation. KFx Inc repurchased the rights to the Series B technology in early 1995.

OBJECTIVES FOR THIS PROJECT

The objectives of this project are to demonstrate the feasibility of the Koppelman Series C Process by constructing a 1,000 lb batch test unit and conducting tests to further develop the design basis for a commercial plant while producing samples of the product for testing and evaluation, and characterizing the emissions from the process. Further objectives are to support the shakedown of the commercial plant until it reaches commercially successful production; improve the efficiency of the process for future plants; expand the markets for the product by testing other feedstocks, and conduct development work to enhance the properties of the product.

APPROACH TO DEVELOPMENT OF THE SERIES C PROCESS

This development work, combined with Koppelman's long experience, has resulted in the Series C Process which is based upon the following principles:

- Nearly all of the moisture is removed in a efficient manner
- Decarboxylation is caused to remove much of the oxygen
- Particles are restructured and shrink during processing
- Sulfur content of sulfur-bearing coals is reduced

The above process concepts require that coal be heated to the range of 325 to 475°C (600 to 900°F) under pressure to confine condensable volatiles within the particles, and that gases and condensate be removed during the processing. The use of pressure, combined with careful control of temperature, causes nearly all of the energy content of the feed coal to be retained in the process while creating a high-calorific value product

Approximately \$50 million and ten years have been spent developing these patented processes. The technologies may be used to transform high-moisture, low-heat value coal and other carbonaceous feedstocks into a lower-moisture, higher-heat value solid fuel, which can be custom designed to meet the end user requirements.

Economic constraints make the process low-cost and efficient. The Series C Process avoids moving coal into or from high pressure. Condensing steam, formed from moisture in the coal, causes efficient heat transfer and uniform heating of particles.

Koppelman has conducted bench-scale tests using the Series C Process to show that subbituminous Powder River Basin (PRB) coal can be converted to a higher-value product having the characteristics of a low-sulfur bituminous coal.

In the tests, PRB coal having a heating value of 19.5 megajoules/kg (8367 Btu/lb) was upgraded to a higher-rank fuel having a heating value of about 28.8 megajoules/kg (12,400 Btu/lb) by removing all of the moisture and by reducing the oxygen content of the coal from about 29 wt % (dry basis) to about 7 wt %. The sulfur content of the low-sulfur PRB coal was further reduced in the process resulting in a product containing about 0.56 lb of SO₂/million Btu.

The successful conclusion of the bench-scale tests resulted in the need to construct and operate larger-scale test facilities.

Our early efforts focused upon determining the time-temperature-pressure cycle required to alter the structure of the product to produce a stable, high-heating value, clean fuel from the pilot plant. We later conducted tests to evaluate the effects of varying particle size and have recently conducted tests to gather in-depth information for use in designing effluent water and gas treatment systems for a commercial-scale plant.

DESCRIPTION OF THE SERIES C PILOT PLANT

The Series C pilot plant consists of coal feed and product discharge systems, a batch processor, a hot oil heating system, and effluent measurement and control systems (Figure 1). In the demonstration plant, crushed and sized coal from an enclosed bin is weighed and flows through a feed screw into a bucket elevator, where it is lifted into a feed bin located above the 13-meter (40-foot) high reactor system. The coal drains by gravity through an open valve into the top of the preheated reactor and fills the conical cavity below the reactor and the tubes within the reactor. The seats in the valve at the top of the reactor are cleaned by a blast of nitrogen gas, and the valve is then closed by a pneumatic operator. Air is swept from the processor by displacement using nitrogen gas and the pressure is then increased to about 8 atmospheres (atm) using nitrogen.

The processor is heated by a flow of 25 liters per second (400 gpm) of heat transfer fluid, which is continuously circulated through a 1050 megajoule (2 million Btu) per hour (net heat transferred) heater fired with propane. As the coal is heated by transfer of heat through the tubes, the water on the coal is evaporated to form steam which again condenses and heats more coal. The condensate from the moisture in the coal, after evaporating and condensing several times, is drained from the bottom of the processor. When the coal has been heated sufficiently to alter the structure and increase the heating value of the product, the reactor is depressured to less than 0.3 atm (5 psig), the valve at the bottom of the processor is opened, and the product falls into a nitrogen-blanketed product collection bin. The product is then transferred by a screw conveyor to a storage bin where it is held in a nitrogen atmosphere until it is cool enough to be exposed to air (about 120°C or 250°F). The product is then weighed, sampled, and held for evaluation.

Condensate drained from the processor is cooled by contact with cold water in a flash tank, filtered to remove coal dust and any coal tars released during processing, and transferred to a storage tank. The gas is vented from a gas-water separator, through a gas meter, and into the stack of the hot oil heater for incineration.

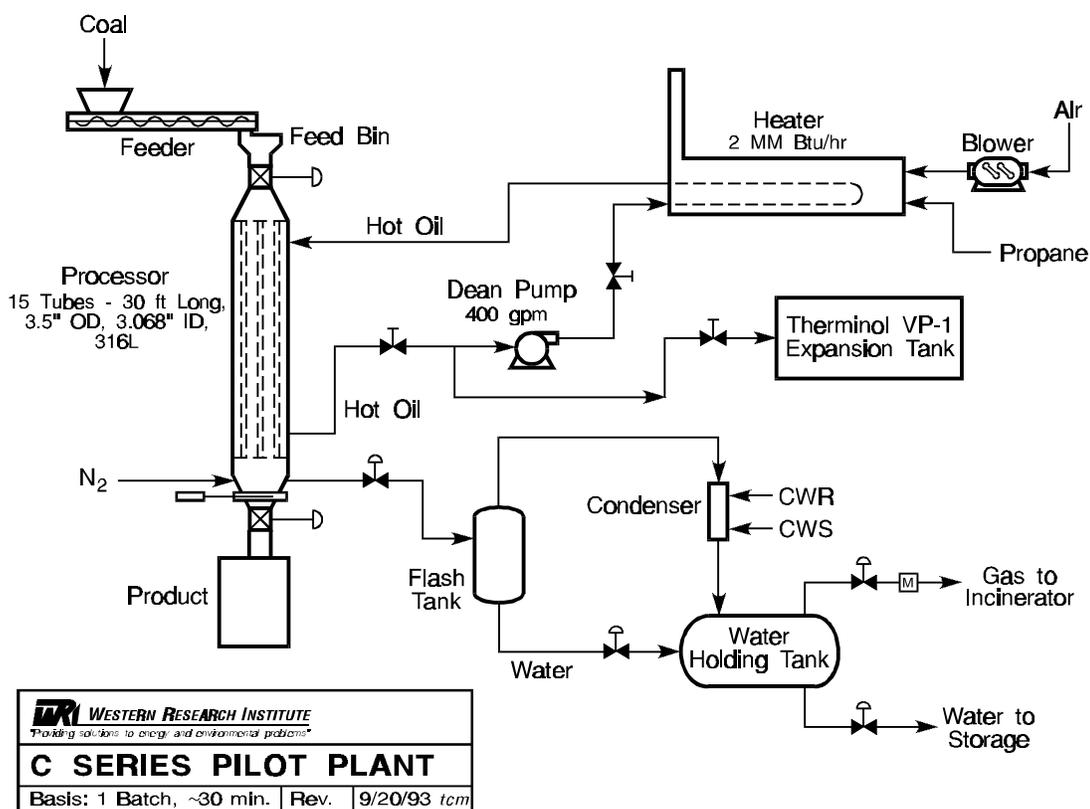


Figure 1. Simplified Process Flow Diagram of the Series C Pilot Plant

Temperatures within the processor are measured by thermocouples that are strategically placed within the coal bed. The temperatures of the feed coal, the product, the heat transfer fluid flowing into and out of the heater and into and out of the processor, as well as temperatures in the effluent gas and water lines are recorded. Pressures are recorded using pressure transducers located at the top and the bottom of the processor and in the flash tank. Pressures are also measured in the heat transfer fluid at the circulating pump suction and discharge, on both sides of the filter in the pump suction line, as well as in the cooling water system, the nitrogen system, and the propane supply system. Temperatures and pressures are recorded at preselected intervals using a computer-based data acquisition system. The timer, which controls the programmable logic controller, is used to establish a time-based series of data. The temperature and pressure readings are continuously visible upon a terminal located near the processor.

The rate of water and gas production are measured by instruments selected for those purposes. Samples of coal and product are analyzed following American Society for Testing and Materials procedures. Water samples are collected and analyzed following protocols established by the U.S. Environmental Protection Agency. Gas samples are analyzed using a gas chromatograph.

Characteristics of the Fort Union Coal Used for the Tests

The PRB feed coal used for the tests described in this report was taken from the Fort Union Mine which is located adjacent to the pilot plant northeast of Gillette, Wyoming. The coal was crushed and screened at the mine and was held in 2 ton capacity enclosed bins until used for testing. Typical composition of coal from the Fort Union mine is shown in Table 1.

Table 1. Composition of Fort Union Coal Used for Tests, wt%

Proximate Analyses				
Test	CSER-24	CSER-25	CSER-27	CSER-28
Moisture	30.4	30.7	32.2	32.0
Ash	5.9	5.8	5.3	5.5
Volatiles	33.8	35.2	32.6	32.8
Fixed C	29.8	28.3	29.9	29.7
Total	100.00	100.00	100.00	100.00
Heating value, MJ/kg	18.95	18.93	18.55	18.53
Sulfur	0.36	0.35	0.31	0.31
MAF calorific value, megajoules/kg	29.75	29.80	29.70	29.66
kg SO ₂ per gigajoule	0.39	0.38	0.30	0.34
lb SO ₂ per million Btu	0.90	0.88	0.78	0.79
Ultimate Analyses				
Moisture	30.4	30.7	32.2	32.0
Carbon	45.1	45.3	44.3	44.9
Hydrogen	2.4	2.5	2.5	2.5
Nitrogen	0.74	0.85	0.70	0.69
Sulfur	0.36	0.35	0.31	0.31
Ash	5.9	5.80	5.3	5.48
Oxygen	15.1	14.6	14.6	14.1
Total	100.00	100.00	100.00	100.00
Moisture is not included in the hydrogen and oxygen				

Design Conditions in the Processor

We selected the following product quality characteristics to produce a marketable product:

- Heating Value - 28.14-28.38 MJ/kg(12,100-12,200 Btu/lb)
- Yield - Approximately 64%
- Oxygen Content - 7 to 10 wt % (dry basis)

The bench-scale tests indicated that the product quality characteristics were most likely to be attained by using the following conditions in the processor:

- Initial Nitrogen Pressure - 8 atm (120 psig)
- Maximum Processing Pressure - less than 55 atm (800 psig)
- Heat Time (not including filling or discharge) - 25 minutes
- Initial Reactor Temperature (when loading) - less than 395°C (740°F)

Production Simulation Tests

At first, the coal would not flow into the top of the processor rapidly because gas being displaced from the processor was escaping through the same opening. A vent tube was installed into the bottom of the feed hopper, which permitted air to escape while rapidly loading the coal. No plugging of the processor occurred during this work. After test 11, a screen was added into the condensate vent opening which permitted faster draining of the condensate. The temperature of the heat transfer fluid entering the processor is the primary control variable. We also controlled the heating time carefully. Our initial effort with the demonstration plant was to make the processor function mechanically and to determine the inlet temperature of the heat transfer fluid needed to control the product heating value in the range of 28.14 to 28.61 MJ/kg (12,100 to 12,300 Btu/lb). Later tests emphasized variations in pressure, feed coal particle size and heating time. An inspection of the processor after test 29 showed that the center tube was free of any accumulation of particles and the tube walls were shiny clean.

RESULTS OF TESTS USING PRB COAL

Evaluation of Pressure and Temperatures in the Processor During a Typical Test (CSER-14)

Figure 2 shows the pressure in the processor, the temperature of saturated steam equivalent to the pressure (taken from steam tables), and the temperatures of coal in the tubes and in the chamber below the tubes. During the test the pressure rises quickly to about 8 atm (120 psig) as nitrogen is added. Pressure increases geometrically as the water and gas are driven from the coal and is then controlled at about 48 atm (700 psig) by venting gas and condensate from the processor.

The temperature of the coal corresponds to saturated steam temperature as the pressure rises to about 48 atm (700 psig). The presence of condensate during this period causes uniform heating throughout the processor. When the temperature exceeds about 260°C (500°F) the condensate becomes a heat sink and retards further temperature increase. Thus, control of the removal of condensate is important both for product quality and process thermal efficiency.

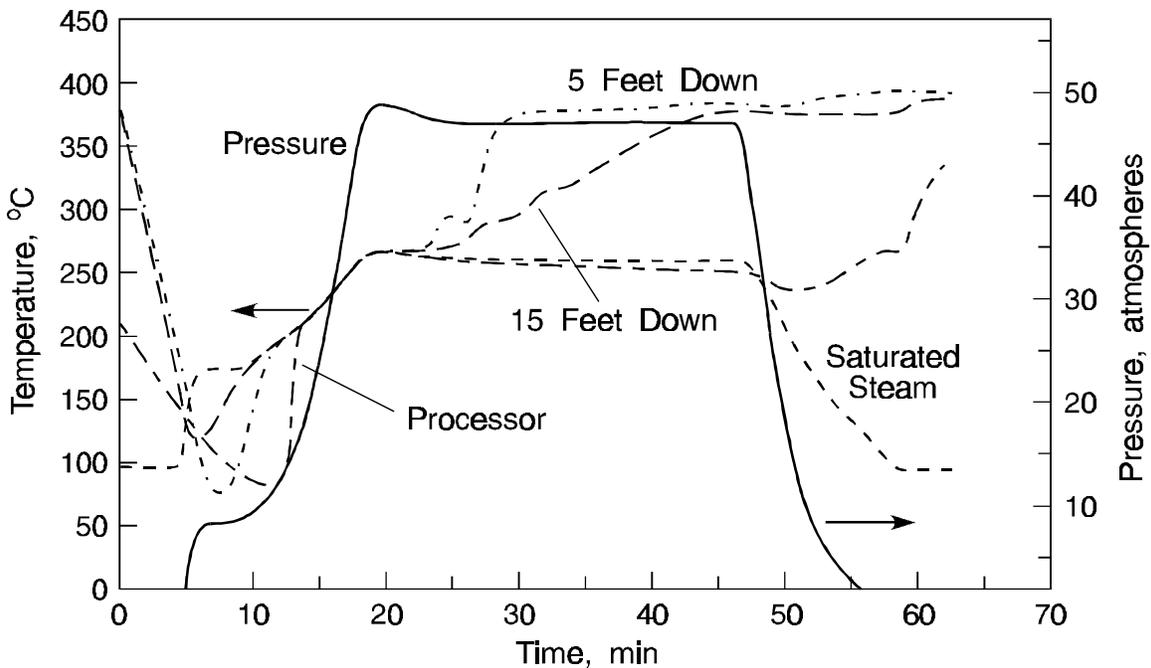


Figure 2. Pressure, Saturated Steam Temperature, and Temperature of Coal at Three Locations in the Processor During Test CSER-14

Figure 3 shows the large differential temperatures between the temperature of the hot oil and the coal early in the test. The differential temperature decreases as the coal is heated rapidly. Temperatures of the coal in the upper part of the processor increase first, followed by the temperatures in the middle, and finally the temperatures in the lower processor increase.

The presence of condensing steam causes uniform heating, but the condensate soaks up energy after the temperature reaches the saturation temperature of steam at the pressure in the processor. The high thermal efficiency for the processor occurs because condensate is removed from the processor as a liquid at low temperature. Nearly all of the energy in the steam driven from the coal is left in the coal.

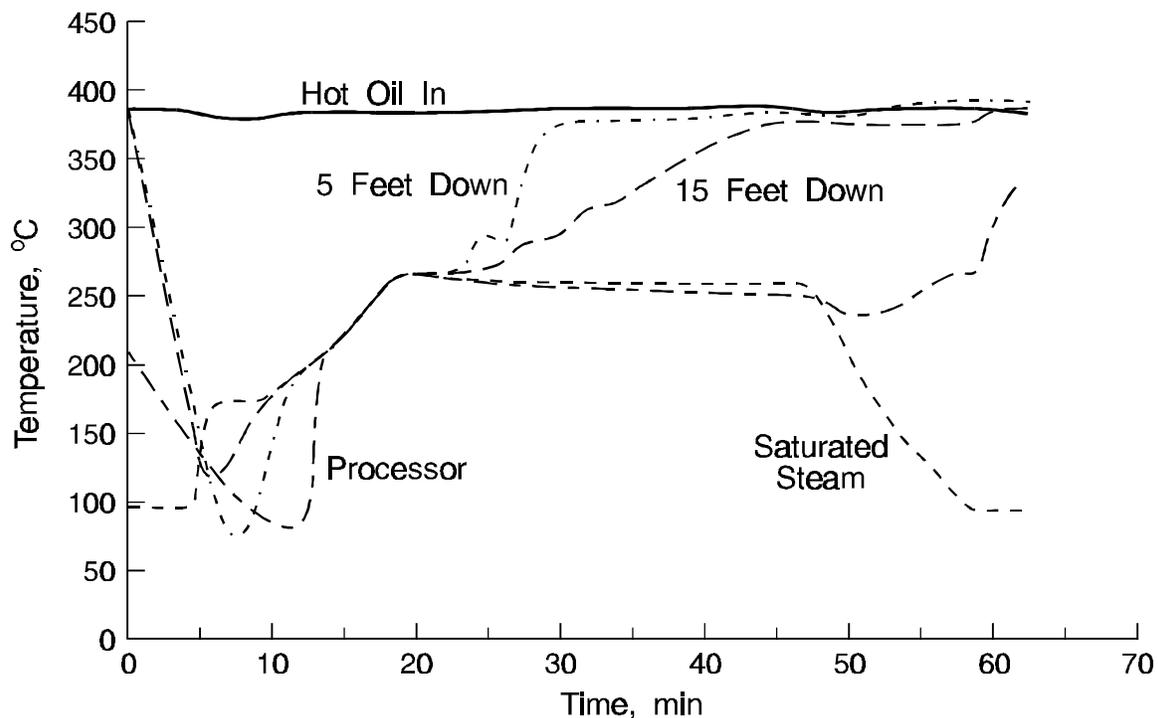


Figure 3. Temperatures of Hot Oil, Saturated Steam, and Coal at Three Locations in the Processor During Test CSER-14

The product characteristics (Table 2) are controlled by the temperature of the heat transfer fluid entering the processor and the heating time (Figure 4). Also, the oxygen content of the product decreases as the processor is operated at higher temperatures. The PRB coal used for the tests described in this paper is a low-sulfur coal in which most of the sulfur exists in the form of organic compounds. The PRB coal contains very little pyrite. The removal of sulfur is increased by using more severe processing conditions (Figure 5). The removal of sulfur in proportion to the weight of the gas and liquid byproducts is typical of organic sulfur compounds found in PRB coal⁴.

Table 2. Composition of K-Fuel® from Typical Pilot Plant Tests, wt %

Proximate Analyses				
Test	CSER-24	CSER-25	CSER-27	CSER-28
Moisture	1.4	0.310	0.40	1.5
Ash	7.8	8.9	8.9	8.0
Volatiles	44.5	42.3	43.6	44.6
Fixed C	46.4	48.6	47.2	45.9
Total	100.00	100.00	100.00	100.00
Heating value, MJ/kg	27.97	28.39	28.13	28.04
Sulfur	0.48	0.46	0.47	0.45
MAF calorific value, MJ/kg	30.78	31.25	31.00	30.99
kg SO ₂ per gigajoule	0.34	0.33	0.33	0.30
lb SO ₂ per million Btu	0.80	0.76	0.77	0.75

Ultimate Analyses				
Moisture	1.4	0.31	0.40	1.5
Carbon	70.1	72.0	70.6	69.4
Hydrogen	4.7	4.8	4.7	4.6
Nitrogen	1.1	1.1	1.1	1.0
Sulfur	0.48	0.46	0.47	0.45
Ash	7.8	8.9	8.9	8.0
Oxygen	14.5	12.5	14.0	15.1
Total	100.00	100.00	100.00	100.00

Moisture is not included in the hydrogen and oxygen

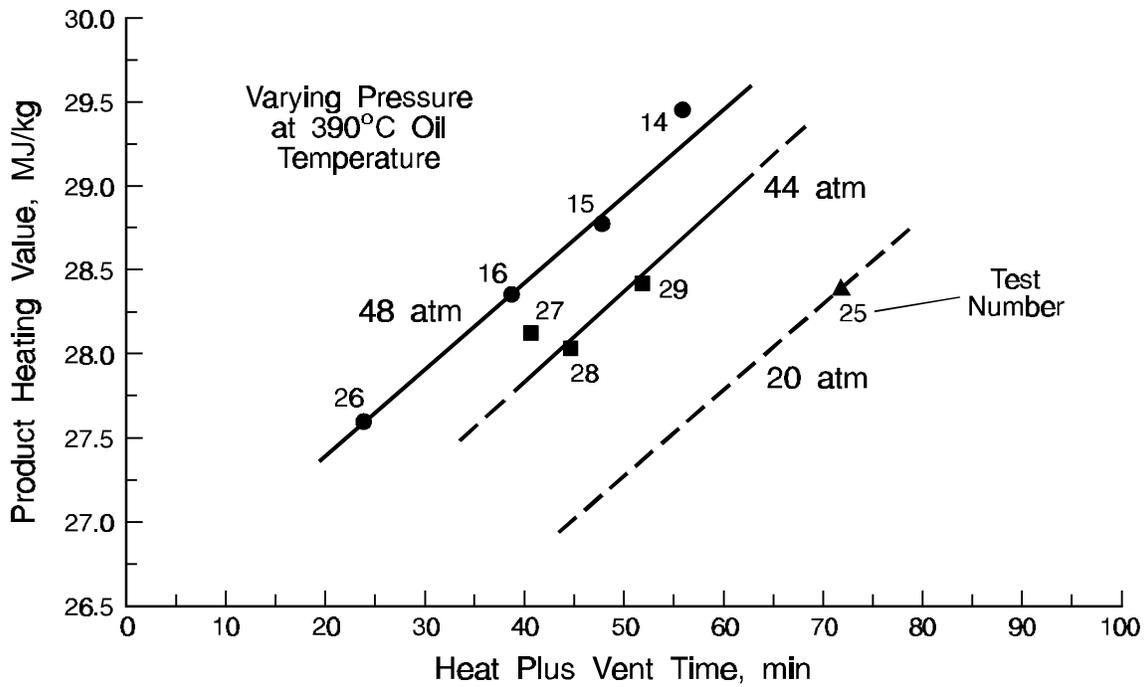


Figure 4. Use of Time and Temperature to Control Heating Value

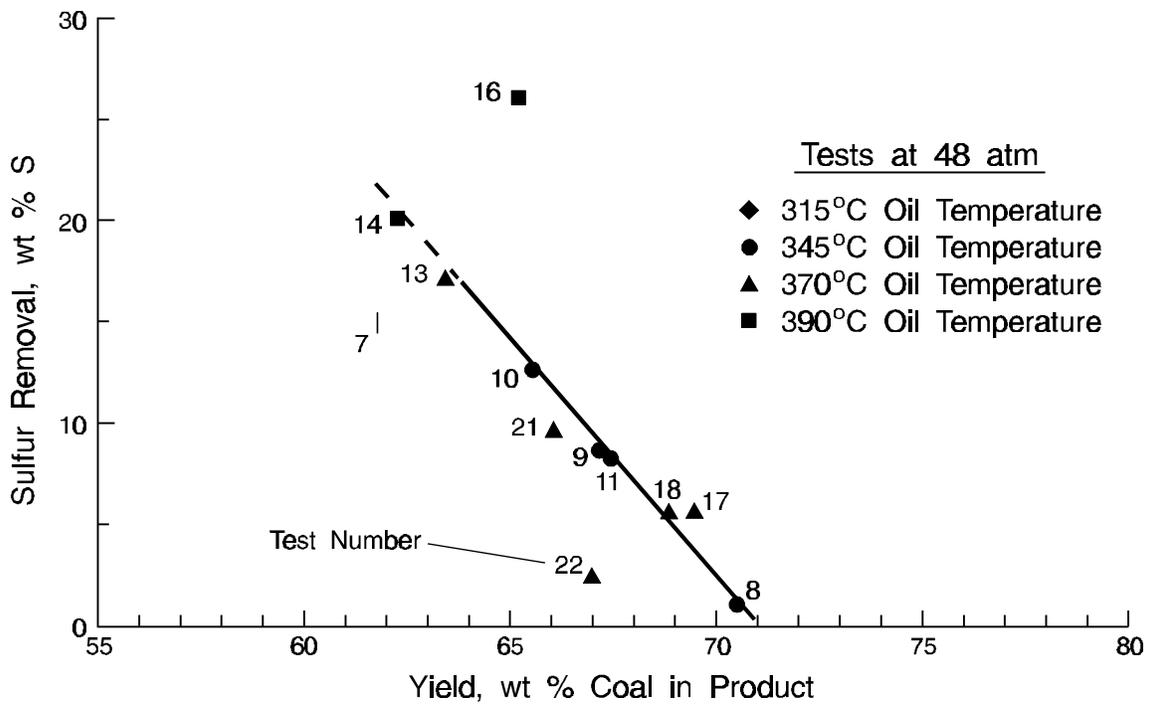


Figure 5. Sulfur Removal from Low-Sulfur Powder River Basin Coal

BENEFITS OF THE SERIES C DEVELOPMENT WORK

An extensive series of tests has been conducted using the Koppelman Series C process in a demonstration plant located near Gillette, Wyoming, USA. The demonstration plant has been operated successfully. We have used the temperature of the heat transfer fluid entering the processor and the heating time to control the heating value of the product within our target range of 28.14 to 28.61 megajoules/kg (12,100 to 12,300 Btu/lb). The oxygen content of the product decreases as the heating value increases, resulting in a product containing from 13 to 8 wt % oxygen. We have removed up to 25 wt % of the sulfur from the low-sulfur PRB coal using conditions which were not selected to maximize sulfur removal.

Typical product yields can be varied from about 63 to 70 wt % of the feed coal depending upon the severity of the conditions used. The energy content of the product varies from about 96% of the heating value of the feed coal with a yield of 64 wt % to nearly 100% energy recovery with a yield of 67 wt %.

In September 1994 KFx Inc. announced that a Memorandum of Agreement had been signed with Thermo Energy Systems Corporation, a subsidiary of Thermo Electron and SDS Petroleum, Inc. to finance, construct, own, and operate the initial commercial coal processing plant using the Koppelman Series C technology near Gillette, Wyoming. As a result of this agreement Thermo Ecotek put up \$42 million in exchange for 95% ownership of the plant that is designed to produce 500,000 tons / year of K-Fuel®. Construction of the plant was started in mid 1995 and completed in late 1996. In early summer of 1997 the plant is undergoing shakedown operations.

FUTURE ACTIVITIES

On April 4, 1996 KFx Inc. announced that it formed K-Fuel LLC, a company with Kennecott Energy and Coal Company (KECC). The new company will provide support for further technical advancement and the global commercialization of business opportunities arising from KFx's patented K-Fuel process. Under the terms of the agreement KFx will grant an exclusive worldwide license for the use of its technology to K-Fuel-LCC. KFx will initially own 51% of the joint venture ...while KECC will provide certain services and capital for all plants approved by KECC.

ACKNOWLEDGMENTS

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REFERENCES

1. Ingram, G.R., and J.D. Rimstidt, 1984, Natural Weathering of Coal. Fuel, 63(3): 292-296.
2. Willson, W.G., S.A. Farnum, G.G. Baker, and G.H. Quentin, 1987, Low-Rank Coal Slurries for Gasification. Fuel Processing Technology, 15: 157-172.
3. Boysen, J.E., C.Y. Cha, F.A. Barbour, T.F. Turner, T.W. Kang, M.H. Breggren, R.F. Hogsett, and M.C. Jha, 1990, Development of an Advanced Process for Drying Fine Coal in an Inclined Fluidized Bed, Final Report. Laramie, WY, WRI Report to DOE, DOE/PC/88886-T5.
4. Merriam, N.W., C.Y. Cha, T.W. Kang, and M.B. Vaillancourt, 1990, Development of an Advanced Continuous Mild Gasification Process for the Production of Co-Products-Task 4-Mild Gasification Tests. Laramie, WY, WRI Report to DOE, DOE/MC/24268-3075.