

6.0 ASSESSMENT OF LIGNITE-UPGRADING TECHNOLOGIES

6.1 Lignite Properties Affecting Upgrading

Various upgrading technologies are applicable to different ranks of coal and ranges of coal analyses. Representative ranges in analysis for U.S. low-rank coal regions are compared in Table 14 with ranges for East Central European brown coals and lignites. The ranges for East Central Europe include higher levels of moisture, sulfur, and ash and lower heating values. The ranges of variation in ash analysis are generally similar for Czech and U.S. coals, although some U.S. lignite ashes contain higher concentrations of sodium, calcium, and magnesium oxides.

The molecular structure of U.S. low-rank coals has been interpreted based on a broad range of analytical data, including elemental analysis, pyrolysis, extraction, controlled oxidation, instrumental Fourier transform infrared analysis (FT-IR), nuclear magnetic resonance spectroscopy (NMR), and differential scanning calorimetry (DSC) (Schobert, 1990; Knudson, 1986; Kube et al., 1984; Benson and Schobert, 1982). Idealized molecular models for low-rank coal, such as the one in Figure 13 indicate one-to three-ring aromatic clusters, a greater abundance of aliphatic and hydroaromatic carbon chains than in bituminous

TABLE 14

Variability in Properties of Low-Rank Coals by Country

	Czech Republic	Poland	Hungary	Bulgaria	Spain	Germany	U.S.
Location	Bilina Melnik Nastup Most Sokolov	Belchatow Konin Turow Adamow Rybnik	Oroszlany Matraalja Borsod	Maritsa East Sofia Bobov Dol Pernik	Teruel	Cologne Leipzig	Wyoming Montana New Mexico Texas North Dakota
Moisture, % as mined	6-55	9-55	19-48	14-62	13-24	48-63	10-42
Ash, % db	7-44	8-40	18-40	28-58	14-70	4-40	3-30
Sulfur, % daf	0.7-9	0.5-7	0.8-5	3-11	3-12	0.4-3	0.3-4
Lower Heating Value, MJ/kg	9-19	7-22	6-15	5-14	12-17	7-12	8-25
Ash Analysis, % Oxides							
SiO ₂	35-50			15-65			18-63
Al ₂ O ₃	11-25			4-32			13-25
Fe ₂ O ₃	9-15			5-20			4-20
CaO	7-8			1-60			5-39
MgO	3-4			1-5			2-13
Na ₂ O	0.6-1.6			0.2-0.4			0.1-12
K ₂ O	0.4-1.1			0.2-0.6			0.1-2

Sources of data include papers by Brix, Couch, and Zakrzewski from the April 1992 Energy and Environment Conference in Prague; case study reports submitted by East Central European participants at the Least Cost Power Course at the EERC in 1992; coal analyses obtained from Bulgaria, Poland, and the Czech Republic for EERC studies performed in 1994; papers by Franke (1977), Stefanski (1981), and Hein (1986) from EERC Lignite Symposia; and U.S. analyses compiled by Selle in his 1986 Review of Slagging and Fouling from Low-Rank Coal.

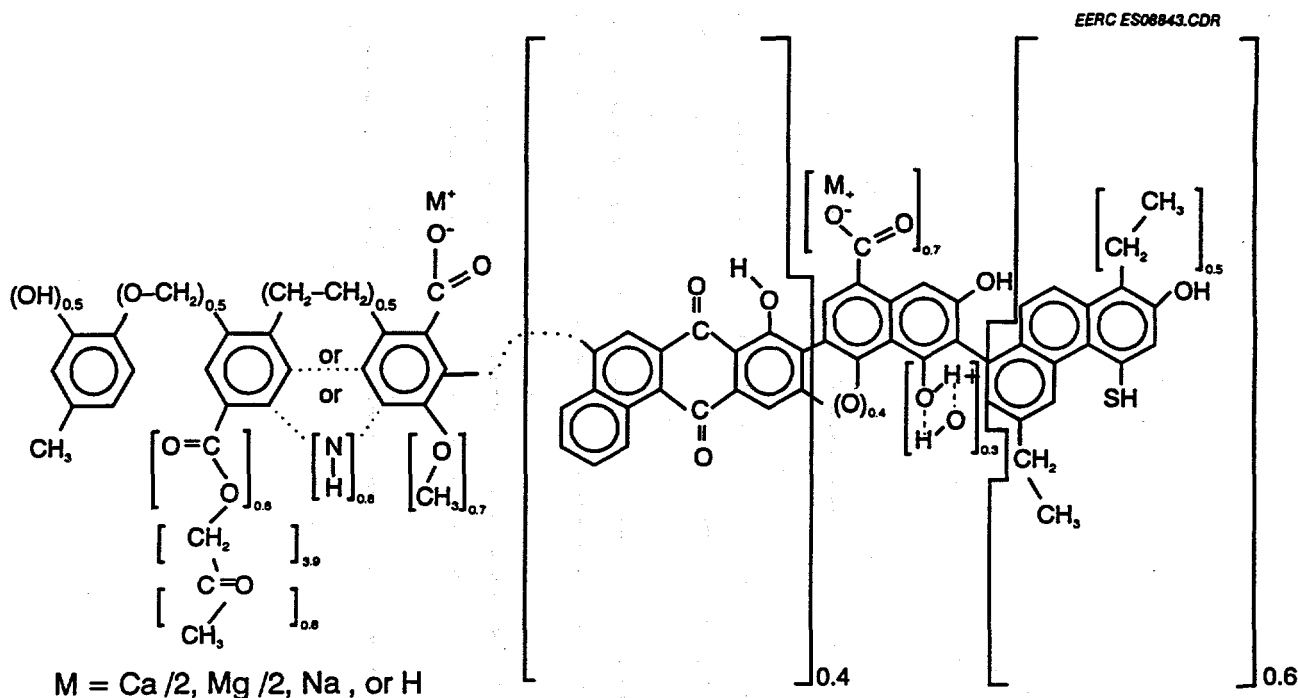


Figure 13. A molecular model for Wyodak subbituminous coal (Knudson, 1986).

coal, and a much higher oxygen content. The most important bridging groups between aromatic ring clusters are polyethylene chains ($-\text{CH}_2-$) and ethers ($\text{R}-\text{O}-\text{R}$). The atomic H/C ratio for moisture and ash-free (maf) low-rank coals is in the range of 0.75 to 0.9, which is comparable to hvA bituminous coal. Between 50% and 75% of the carbon in low-rank coal occurs in aromatic rings, compared to about 85% for bituminous coal. In laboratory calorimetry, the aliphatic and hydroaromatic carbon burns off at a lower temperature than the aromatic clusters, leaving the graphitized char.

The physical structure of low-rank coal is believed to be determined by the effect of oxygen functional groups on hydrogen bonding and the role of moisture as a structural component (Schobert, 1990). Phenolic groups provide a framework for hydrogen bonding, whereas carboxyl groups may hinder structuring. The roughly 20% of total moisture held tightly by hydrogen bonding is believed to contribute to structural rigidity in low-rank coal in the same manner as in wood, and the loss of this structure when coal is dried to low moisture levels accounts for its friability and dustiness.

The high oxygen content in low-rank coal, up to about 22% maf, includes carboxyl ($-\text{COOH}$)-, phenolic ($\text{Ar}-\text{OH}$)-, ether and methoxyl ($\text{R}-\text{O}-\text{R}$ and $\text{R}-\text{O}-\text{CH}_3$)-, and carbonyl ($-\{\text{C}=\text{O}\}$)-type functional groups. Carboxyl groups, which account for one-quarter to one-half of the total maf oxygen, are important ion-exchange sites for alkalis, alkaline earths, and trace elements. The carboxyl oxygen content thermally decomposes with the release of carbon dioxide starting above about 300°C , tending to increase the mobility of the absorbed cations in thermal processes.

The molecular form of sulfur and nitrogen in low-rank coal is not as well characterized. Their forms in pyrolysis products indicate that they occur in both aliphatic and aromatic structures. The ASTM-defined sulfur forms (pyritic, sulfatic, and organic) used in the United States have only limited value in understanding sulfur chemistry. For U.S. lignites and subbituminous coals, the ASTM determination typically indicates 50%–60% organic, 40% pyritic, and 5% sulfatic. However, research at the EERC using supercritical fluid extraction (SFE) to separate and analyze the true sulfur forms has shown that up to 36% of the ASTM "organic" sulfur is elemental sulfur (Louie et al., 1993).

High reactivities are observed for low-rank coals both in thermal processes and at near ambient temperatures where spontaneous heating begins. In high-temperature processes, reactivity is increased by the noncaking property of the low-rank coal and by the release of free radicals formed by decarboxylation. High-temperature reactivity correlates with minerals and cations that catalyze thermal reactions, but it is not directly linked to surface area measurements (Smith et al., 1993; Timpe et al., 1989). At lower temperatures, spontaneous heating correlates with reduced particle size and drying (Sondreal and Ellman, 1974).

The distribution of pore size and surface area in low-rank coals characteristically involves a relatively large microporosity scaled below 2 nanometers (nm) and a smaller macroporosity larger than 20 nm (Smith et al., 1993; Sharkey and McCartney, 1981). In higher-rank coals, this distribution tends to be reversed. The accessible pore volume increases during carbonization to form a high surface area char. Questions concerning the interpretation of surface area measurements on coal are related to the different chemical and physical gas absorption interactions that are possible.

Inorganic forms in low-rank coals are primarily categorized as either ions associated with coal carboxylate or discrete mineral grains. Most of the alkali and alkaline earth elements in U.S. low-rank coals are ionically associated, typically including 80%–90% Na, 70%–80% Mg, 60%–70% Ca, and 30%–40% K. The percentage for potassium is lower because of its occurrence in clay minerals. The major classes of minerals in U.S. low-rank coals, in approximate order of decreasing abundance, are silicate and aluminosilicate, pyrite, oxides of iron and titanium, sulfates as gypsum and barite, calcite, and phosphate as apatite (Benson et al., 1993).

6.2 Beneficiation

6.2.1 Background on Cleaning U.S. Coals

Coal cleaning is widely practiced in the U.S. to reduce the ash and pyritic sulfur contents of both metallurgical and steam coals, with most cleaning plants currently processing bituminous coal. However, cleaning has long been viewed as one key to expanding markets for low-rank subbituminous coal and lignite. Conventional gravity cleaning has been correlated with improved capacity and performance in pc-fired boilers owing to reduced erosion, mill wear, slagging, and fouling and to increased pulverizer capacity (Durant et al., 1989; Busch and Vaninetti, 1980; Everitt and Jones, 1980). More advanced fine coal cleaning methods involving oil agglomeration, froth floatation, and leaching in acid or caustic can be used to achieve low levels of ash if they are tailored to the feed coal. Testing is required to determine the effectiveness of different methods for a particular low-rank coal.

Washability data on U.S. low-rank coals indicate wide variability in cleanability and limited value in coarse gravity cleaning alone (Energy Resources Co., Inc., 1980; Brown et al., 1985; Cavallaro et al., 1976; Malterer et al., 1988). This is principally due to the significant fraction of the ash-forming constituents that are chemically bound as absorbed cations, which cannot be removed by physical cleaning. Ion-exchangeable cations account for over half of the ash-forming material in some low-ash U.S. low-rank coals, including most of the sodium content which adversely affects ash deposition in boilers. Also, the discrete mineral grains in U.S. low-rank coals are typically finer than those in bituminous coals and, therefore, more difficult to physically separate (Zygarlicke et al., 1990).

In the past, coal-cleaning methods have not played a significant role in sulfur control strategy for U.S. utilities because they do not have the capability of meeting the 70% to 90% removal requirement under the U.S. NSPS. The more flexible provisions in the 1990 U.S. acid rain legislation mandating retrofit of older boilers, including the provision for emission trading, should serve to increase the role of coal cleaning. In general, the effectiveness of physical cleaning to remove sulfur depends on the fraction of total sulfur occurring as pyritic sulfur, which is potentially separable. In the United States, this fraction is lower for low-rank coals than for bituminous coal, averaging 34% for North Dakota lignite (Sprouls, 1989) compared to 58% for Pennsylvania bituminous coal (Cavallaro et al., 1976).

6.2.2 Applicable Methods for Cleaning Low-Rank Coals

Gravity Cleaning Processes: Coarse gravity cleaning methods are applicable to low-rank coals within the limitations imposed by the forms of ash and sulfur present. Clean coal has a specific gravity of 1.12-1.35, compared to 2-3 for sand, shale and clay, and 4.8-5.2 for pyrites. Various equipment for gravity cleaning includes 1) jigs operating by stratification in pulsating water; 2) dense-media vessels achieving gravity separation in finely dispersed magnetite in water; 3) concentrating tables using differential movement of coal in water over an inclined, ribbed surface; 4) hydroclones achieving centrifugal separation in water; 5) dry pneumatic concentrators that substitute air for water on a dry table or jig; and 6) launders which use the differential velocity of coal and minerals down an inclined water-washed surface as influenced by specific gravity, friction, and hydrodynamic boundary layer effects.

Fine-Coal-Cleaning Methods: Finer mineral particles and pyrites are liberated as the coal is progressively ground to a smaller particle size. The principal methods used for physically cleaning fine coal include 1) dense-media cyclones which combine the effects of specific gravity and centrifugal force up to 200 g, 2) dry magnetic separation in a high-intensity magnetic field based on the paramagnetic properties of minerals that are attracted by magnetic force and the diamagnetic property of clean coal which is repulsed, 3) oil agglomeration based on the adhesive property of oil to agglomerate and separate clean coal from water, and 4) froth flotation based on the differential adhesion of water and fine air bubbles on coal and minerals. The applicability of dense-media cyclones and dry magnetic separation methods depends primarily on the properties of the minerals rather than the properties of the clean coal, and they may or may not be applicable to a particular low rank coal depending on the mineral forms present. The use of oil agglomeration for cleaning low-rank coals requires that a phenolic oil or another polar additive be included in the oil phase to allow it to adhere to the otherwise hydrophilic coal surface. Froth flotation has not been effective for low-rank coals because of the hydrophilic property of the coal surface which causes it to remain in the bulk water phase.

Chemical Cleaning Methods: Chemical cleaning methods that are potentially suitable for low-rank coals include 1) acid leaching to remove ionically absorbed cations, 2) an alternative ion-exchange treatment that substitutes calcium for sodium to alleviate boiler fouling, 3) various combinations of aqueous oxidation and chemically enhanced (e.g., caustic) leaching to remove principally pyritic sulfur, and 4) molten caustic leaching to reduce both ash and total sulfur to low levels. The cost of chemical cleaning is typically much higher than physical cleaning, with chemical methods estimated at US\$12 to US\$41/ton in the 1980 Low-Rank Coal Study (Energy Resources Co., 1980), which corresponds to approximately US\$0.60 to US\$2/GJ for a 23-GJ/kg product. The EERC has demonstrated that U.S. low-rank coals can be cleaned by a combination of wet gravity separation and acid leaching to achieve 60%–80% ash removal and 30%–80% sulfur removal, yielding a product with typically 2%–3% ash and 0.2%–0.5% sulfur (Potas et al., 1989). The combined cleaning cost was estimated at about US\$0.75/GJ. Cleaning of low-rank coals to lower ash levels of 1%–2% has been demonstrated by the EERC using a combination of polar oil agglomeration and acid leaching (Potas et al., 1990).

Other principles that have been tested for coal cleaning include microwave treatment, separation in liquid CO₂, biodesulfurization, and hydrocarbonization with dolomite. These and other experimental methods may gain commercial status in the future, but for now assessment should focus on more proven physical or chemical cleaning methods.

6.2.3 Physical Cleaning of Czech Lignites

The EERC has recently completed an evaluation of cleanability for lignites from Bílina and Nástup for Ústí nad Labem in the Czech Republic (Young and Musich, 1995). Test samples were crushed and screened to separate a 0.84- to 6.35-mm size fraction for cleanability testing. The discarded finer fraction, which contained the higher concentration of ash and was not cleaned, represented 14% to 22% of the coal heating value. Wet gravity separations were performed at specific gravities of 1.3, 1.4, and 1.6. Dry separation using a rare-earth magnetic separation method (REMS) was performed on two subordinate size fractions.

Significant reductions in ash were achieved by wet gravity separation at 1.3 specific gravity for the Bílina and Nástup lignites at reasonably high energy recoveries of 89%–93%. For three samples of Bílina lignite, ash contents in the range of 5.4% to 7.2% after screening (moisture-free basis) were reduced to between 3.9% and 4.1%, representing a 23% to 43% percentage reduction. The combination of screening and cleaning reduced the ash contents of Bílina lignite samples by 38% to 51%. For the one sample of Nástup lignite tested, the ash content was reduced from 17.6% to 9.2%, representing a 48% percentage reduction for wet gravity cleaning alone and a 51% percentage reduction for screening and cleaning combined.

Substantial percentage reductions in sulfur content of 29% to 43% achieved by wet gravity cleaning on three Bílina lignite samples were in direct proportion to the percentage of the total sulfur occurring as pyrite, which also ranged from 29% to 43%. The range of total sulfur content in the Bílina samples before screening and cleaning was 1.02% to 1.23% on a moisture-free basis. Screening did not significantly lower the sulfur content. For Nástup lignite, with 34% of its 1.74% total sulfur content occurring as pyrite, essentially no sulfur reduction was achieved by wet gravity cleaning, possibly indicating that the pyrite in this sample was very finely divided and not amenable to separation.

Dry magnetic cleaning by REMS was not as effective as wet gravity separation at 1.3 specific gravity, but it did achieve significant removals of ash and sulfur for the one Bílina lignite sample tested. Combining results for two sizes tested, the REMS method achieved a 19% reduction in ash and a 29% reduction in sulfur at 95% energy recovery, compared to 43% and 39% reductions, respectively, for wet gravity cleaning at 92% recovery. For Nástup lignite, the REMS method achieved essentially no reduction in either ash or sulfur content.

The varied results of cleaning lignites from two Czech mines illustrate the difficulty of providing a general assessment on the overall cleanability of lignites and the need for testing a representative sampling of any coal being evaluated.

6.3 Evaporative Drying

A wide range of evaporative drying processes are available where coal temperatures remain below 100°C. They differ according to the design of the dryer, the size consist of the feed coal, the heat source, the quenching method, the product stabilization procedure, if any, and the utilization characteristics of the dried product. Commercially available dryers include entrained-flow (e.g., the Parry dryer), rotating drum, fluidized bed, flash mill, and steam tube designs. The application of these drying methods to upgrade coals has been constrained by problems of product stability and cost-effectiveness. Low-temperature drying unfortunately does not significantly reduce the equilibrium moisture content, causing the product to be subject to moisture reabsorption. Evaporative drying to low moisture levels also reduces particle strength, increases dust and friability, and increases the danger of spontaneous heating in handling and storage (Willson et al., 1992). However, even with these problems, the rail shipment and long-term storage of evaporatively dried U.S. lignite and subbituminous coals have been successfully demonstrated on an experimental scale (Paulson et al., 1973). The most successful example of commercial drying in the United States is the Parry dryer at the Texas Utilities generating plant in Texas, which dries lignite for direct use in slagging boilers. A fluidized-bed dryer built by the Amax Coal Company to dry U.S. Powder River Basin subbituminous coal for shipment to utilities in the U.S. midwestern and eastern regions has thus far proven unsuccessful because of fines generation and extreme dust problems.

Flash mill and steam tube dryers were developed for European and Australian brown coals for applications that include 1) the direct integration of flash mill drying with pc-firing, 2) stand-alone drying for production of briquettes, and 3) the production of pneumatically conveyed dry coal powder for use as an industrial fuel. The integrated flash mill (Kramer) design uses recirculating boiler flue gas for in-mill drying, with centrifugal separation of the dried product from the moist gas. The stability and efficiency of this process depends on careful management of the in-mill drying conditions. Steam tube drying, which was developed in Australia and licensed to Lurgi Australia, dries coal in a fluidized bed heated by immersed steam tubes, with the fluidizing agent being the moisture released from the coal. The reported capabilities of steam tube drying on brown coal include a large reduction in moisture content (from 60% to 15%) and reduced carbon dioxide emissions from the product upon heating (decreased by 17%) (Hamilton, 1990).

Experimental drying methods operating at low coal temperatures include multiple-effect fluidized beds, solar ponds, and a pulse-jet design that eliminates the need for blowers. These methods offer potential advantages where relatively large amounts of water need to be removed

efficiently, but various limitations have prevented their commercial application—including the capital cost of multiple fluidized beds, the climatic dependence of solar drying, and the noise of a pulse-jet design.

The capital costs of low-temperature evaporative dryers are in the range of US\$63 to US\$109 per product ton year, based on a 1984 study update to current dollars (Davy McKee, 1984). The processing cost including capital charges for upgrading low-rank coal by evaporative drying is on the order of US\$1/GJ of dried product, which can only be justified by offsetting savings in freight cost, boiler performance, or other utilization benefits. Freight savings alone do not come close to covering this level of processing cost, and credits must be identified in areas of coal handling, pulverizers, boiler capacity and availability, and emissions control performance. In the United States, these offsetting savings are being investigated where there are opportunities for substituting low-sulfur subbituminous coal for high-sulfur bituminous coal without incurring extensive charges for boiler modifications

6.4 Intermediate Temperature Drying

Drying processes that raise the temperature of the coal above 240°C permanently change the physical and chemical properties of low-rank coals and yield a product with a lower equilibrium moisture content. At these temperatures, tar migrates to the coal surface where it seals surface pores and significantly reduces the ability of the coal to reabsorb moisture. Evolution of CO₂ also reduces the capacity of the coal to chemically bind water by removing hydrophilic carboxyl groups.

Different intermediate temperature drying processes have been developed using hot gas, steam, hot water, or oil. The quality of the dried product depends more on temperature than on the drying medium. Updated capital costs for intermediate temperature dryers are in the range of US\$101 to US\$146 per product ton year, or about 30% to 50% higher than for low-temperature evaporative dryers (Davy McKee, 1984). Processing costs including capital charges, based on the same source, are in the range of US\$1.36 to US\$1.55/GJ, or about 10% to 30% higher than for low-temperature drying. Process improvements in solids separation and handling or heat recovery have been reported to reduce both capital and operating costs.

6.4.1 The Syncoal Drying Process

The Syncoal process dries low-rank coal in hot combustion gas to produce an upgraded boiler fuel. The technology has been demonstrated under the U.S. Clean Coal Technology program, operating on Montana subbituminous coal and North Dakota lignite (Sheldon and Heintz, 1995; Niquette, 1994). The process incorporates staged drying in two vibratory fluidized beds followed by a water quench, cooling in another vibratory fluidized bed, and pneumatic separation of ash, including up to 90% of the pyritic sulfur. When applied to lignite feed, the process has achieved a 31% reduction in ash (db), a 53% reduction in specific sulfur emission (g of SO₂/GJ), and a 53% increase in fuel heating value (16.4 to 25.1 GJ/kg). Boiler tests on the dried coal product have shown improvements in boiler cleanliness, efficiency, and generating capacity, with no adverse effect on NO_x emissions. A commercial plant has been proposed to produce 500,000 tons per year of product from North Dakota lignite at a plant construction cost of US\$43 million. The economic justification for this project in boiler fuel

markets depends on the U.S. alternative fuel tax credit and the offsetting cost savings expected in transportation, boiler performance, and SO₂ emission reduction.

6.4.2 Hydrothermal Drying Processes

A number of intermediate temperature drying processes for upgrading lignite as a solid fuel have been designed to operate by heating coal under saturated steam/water to temperatures between 240°C and 380°C, including development efforts performed by the EERC, K-Fuel, Bechtel, IGT, Shell and others—all somewhat related to the older batch Fleissner process. The semicontinuous K-Fuel Series C process has been demonstrated on a 450-kg-per-batch scale in Gillette, Wyoming, in the United States. When operating on subbituminous coal, the process has achieved up to 25% sulfur removal and an increase in heating value from 18–19 GJ/kg to above 28 GJ/kg (Merriam and Gentile, 1995). The advantage of using moderately higher temperatures, as discussed above, is that the structure of the product is altered by decarboxylation and migration of tars to the coal surface such that moisture reabsorption is reduced and product strength and stability are improved.

6.4.3 Hot Oil Drying

The Carbotech hot-oil drying process was initially selected in Round IV of the U.S. Clean Coal Technology Demonstration Program, but the demonstration was not implemented because of technical and financial questions. The concept of drying in hot oil was developed as early as 1926 and was later used by Exxon in their donor-solvent direct liquefaction process (Willson et al., 1992). The Carbotech technology uses two stages: first to dry the coal in hot oil and then to recover oil by flue gas stripping. A small amount of oil remains absorbed on the coal, which reportedly serves to stabilize the product. Some difficulty has been experienced in achieving the high level of oil recovery needed to make hot-oil drying economically feasible.

6.5 Upgrading Processes Based on Pyrolysis

6.5.1 The LFC/Encoal Process

The “liquids from coal (LFC) pyrolysis process” is being demonstrated under the U.S. Clean Coal Technology Demonstration Program (Castro et al., 1994; McCord et al., 1993) to optimize techniques for producing both premium solid fuel and liquid fuels from low-rank coals. Coal is processed in three stages, including controlled drying, pyrolysis, and cooling in inert gas with rehydration and dust suppression. Liquids are collected in quench columns and an electrostatic precipitator, and the low-calorific value gas produced is burned to supply process heat. The economics of the process have been evaluated for Polish lignite containing 58.7% moisture (Castro et al., 1994) based on utility-type financing at a 10% cost for capital, with the following findings being reported: 1) a capital cost of US\$560 million for a char plant producing 4 million tonnes/yr (heating value 28 GJ/kg), sufficient to supply fuel for four 360-MW_e generating units; 2) a levelized processing cost including capital recovery of US\$0.95 per GJ of product (char and oil), excluding the cost of the coal feedstock; and 3) energy recovery of 77% in char and 8% in oil in relation of lignite feed. In special circumstances where a one-third reduction in sulfur emission (kg SO₂/GJ), as offered by the LFC process, meets the required emission standard, the LFC process is estimated to offer a

significant cost advantage over FGD (cost ratio 1/1.5) after credits are taken for the sale of oil at US\$15/bbl and for a 9% improvement in electrical generating efficiency (Castro et al., 1994).

6.5.2 The FMC Process and Other Form Coke Technologies

The FMC Corporation in the United States operates a commercial pyrolysis plant at Kemmerer, Wyoming, producing briquetted formcoke® from low-sulfur subbituminous coal for use in reducing phosphorus ore. The FMC process can be configured in different ways by appropriately selecting a series of fluidized-bed reactors to accomplish drying, preoxidation to reduce caking, carbonization, and/or calcination as required. The process can be applied to coals of different rank, including caking bituminous coals. At Kemmerer, the hot char is combined with polymerized tar and supplemental binder to form briquettes which are cured, calcined, and finally cooled to give a high-strength low-volatile formcoke®. A 20,000-ton test at Inlant Steel demonstrated that the FMC formcoke® can be successfully used in a blast furnace.

Many other form coke processes have been investigated over the last 40 years using both high- and low-rank coals. The principal goal has been to develop a technically, environmentally, and economically satisfactory substitute for slot-oven coke for use in blast furnaces and, to a lesser extent, in iron foundry cupolas. Driving forces behind this development effort have been 1) decreasing supplies of good quality coking coal, 2) increasing prices for good quality coke, and 3) the need for a fully contained process that can control the pollution arising from traditional slot ovens. Despite extensive interest, only two commercial form coke plants are known to be operating: the Sastech plant in South Africa and the FMC plant in the United States. In addition, the Samchully plant in South Korea produces cylindrical briquettes from a mixture of petroleum coke, coke breeze, anthracite, and coking coal.

Recent developments in form coke technology have been undertaken in the United States, Canada, and Germany, most notably within the U.S. DOE mild gasification program discussed below. The U.S. Salem Company has successfully tested a rotary pancake furnace in the United States and Germany for the production of semicoke and for smokeless fuel production in Canada. This process has been considered for the production of semicoke from Czech coals (Buchtele and Straka, 1994).

6.5.3 The U.S. DOE Mild Gasification Program

Starting in the mid-1980s, the U.S. DOE sponsored research on four carbonization (mild gasification) processes at the scale of 100 lb (45.5 kg/hr) of coal feed per hour for the purpose of optimizing char properties and liquid yields and qualities from a variety of U.S. coals under low-severity conditions that could potentially be economic in existing markets. The most significant differences among the processes were in the reactors and the gasifying atmospheres, with all operating at near-atmospheric pressure. The EERC investigated a two-stage spouting fluidized-bed reactor using hot combustion gas and steam as the gasifying atmosphere; the process was operated on a mildly caking high-sulfur Indiana (U.S.) bituminous coal under conditions that optimized sulfur removal from the char and separation of liquid condensates (Sondreal et al., 1989; Aulich et al., 1991). The Coal Technology Corporation developed a

twin-screw reactor for using highly caking eastern U.S. bituminous coals; a continuous process for producing form coke was demonstrated at a pilot scale of 10 tons per day and patented (Wolfe et al., 1989, 1995). The Western Research Institute performed studies in an inclined fluidized-bed reactor operating on indirectly heated recycle gases; products from a western U.S. subbituminous coal were a char that was converted to carbon black and a relatively high heating value gas. Finally, IGT used an externally heated, spouted fluidized bed optimized to operate on caking coals using preheated inert gas (Babu et al., 1989). The IGT process is being scaled up under U.S. DOE sponsorship for production of form coke.

The market assessments performed in conjunction with the EERC mild gasification project (Sinor, 1992, 1988) indicate that there are many technically feasible opportunities for producing upgraded coal products, including metallurgical coke substitutes, activated carbons, sorbent chars for control of toxic emissions, and liquid fuels and chemicals. Some of these markets are reviewed in section 7.0. The general conclusion is that the high cost of producing marketable products and the challenge of meeting traditional market specifications will constrain commercial development. For example, the financial analysis for a form coke plant sized to produce 129,000 short tons per year from a bituminous coal priced at US\$20 per ton indicated a discounted cash flow (DCF) rate of return of only 8.1% at a form coke selling price of US\$150 per ton, which is higher than prevailing prices for conventional slot-oven coke (Sinor, 1982). The best prospects for profitability would appear to be in making low-volume products having a very high added value, such as activated carbons, where market success depends primarily on the performance of the product.

6.6 Low-Rank Coal-Water Fuel (LRCWF)

6.6.1 Process Description

The technology for producing LRCWF from brown, lignitic, and subbituminous coals was pioneered by the EERC in a 6-tpd pilot plant by hydrothermally treating normal pc-grind coal in saturated hot water at about 285°C and a pressure of 1100 psi (75 bar). A commercial-scale demonstration project based on the EERC's LRCWF technology is being planned for Alaska in the United States. Also, a consortium in Japan and the State Electricity Commission of Victoria in Australia have recently constructed LRCWF pilot plants modeled after the EERC design, and interest has been expressed for a dredge-mined Kovin lignite in northern Serbia (Ljubičić et al., 1994; Yui, 1995). Under the conditions of this process, water is irreversibly expelled from the coal; carbon dioxide is released; and devolatilized tar is deposited on the surface of coal particles to prevent water reabsorption. After hydrothermal processing, excess water is removed to produce a stable pumpable slurry with a dry solids content of about 60%. Sufficient oxygen content is retained in the coal to maintain its hydrophilic character, so that the LRCWF has little tendency to settle and exhibits good viscosity characteristics without additives.

6.6.2 Economics of LRCWF

The projected selling prices for LRCWF cover a range from about US\$1.60 to US\$3.70/GJ depending on a number of variables: 1) coal cost, 2) product specifications (e.g., level of coal cleaning), 3) plant size, and 4) varying capital costs. This overall range is competitive with refined petroleum in the Rotterdam market, based on April 1995 prices for

1% sulfur residual oil at US\$17/bbl (US\$2.54/GJ) and No. 2 fuel oil at US\$21.60/bbl (US\$3.43/GJ) (Oil and Gas Journal, 1994). LRCWF is generally not competitive with steam coal in international trade, for which the International Steam Coal Market ranged from US\$34 to US\$42/ton (US\$1.24/GJ to US\$1.51/GJ) during 1994 (International Coal Report, 1994). The capital cost of a plant for producing 4 million short tons per year (tpy) of LRCWF is estimated to be on the order of US\$300 million, generating a capital cost of about US\$0.70/GJ. A breakdown of the nominal US\$2/GJ production cost for a 4-million-tpy plant includes US\$0.66/GJ for coal, US\$0.13/GJ operating labor, US\$0.27/GJ utilities, US\$0.24/GJ general maintenance, and US\$0.70 in debt payment for 12% financing over a term of 20 years (Anderson, 1995). Adjusting for plant size based on a 0.6 exponential factor, the corresponding costs at 1.5- and 10-million-tpy capacities would be approximately US\$1.60 and US\$2.70, respectively. At 4-million-tpy capacity, the incorporation of physical coal cleaning would add nominally US\$0.30/GJ, and chemical cleaning by acid leaching would add an additional US\$0.70/GJ. The added cost of chemical cleaning would be considered only for direct firing of LRCWF in diesel or gas turbine engines, which is being studied under the U.S. DOE Research & Development program. Lower processing cost estimates down to US\$0.73/GJ, exclusive of coal feed cost, have been advanced for a small modular plant design for producing 500,000 tpy (International Coal Preparation Consultants Limited, 1995). Lower costs could also be achieved by integrating the LRCWF process into an advanced IGCC power system that uses a coal slurry feed, such as the Texaco and Destec gasifiers. Where slurry feed equipment is already included in an IGCC power system, the incremental cost of adding hydrothermal treatment and slurry concentration is estimated to be on the order of US\$0.30/MJ (Anderson, 1995). Current uncertainty in process cost estimates will be narrowed by the demonstration project being planned for Alaska in the 1995-1999 time period.

6.6.3 Pipeline Transportation

Pipeline transport is economically justified at product volumes above 5 million short tons per year. The low settling rate of LRCWF allows pipelines to be designed for laminar flow and provides flexibility for interrupted flow, which would not be possible for a coarse coal-water slurry pipeline. For long-distance shipment, costs are estimated to be US\$0.025/ton mile plus US\$2.50/ton for terminal transfer charges (International Coal Preparation Consultants Limited, 1995). For a LRCWF heating value of 15.8 MJ/Kg, the resulting 500-mile shipping cost of US\$15/ton is equivalent to US\$1.05/GJ. This estimate is in general agreement with U.S. pipeline transportation costs for petroleum on a tonnage basis, which averaged US\$0.017/short ton-mile in 1994 (Oil and Gas Journal, 1994). However the unit cost of a short pipeline could be considerably higher, based on an estimate of US\$0.06/ton mile for a 21-mile (34-km) pipeline of nominal 20-inch (51-cm) diameter studied for transporting 5 million tpy of LRCWF in Alaska (Willson et al., 1991). The design and cost studies were based on calculations of pressure drop for a pseudoplastic fluid in laminar flow, which predicted a pressure prop of 1.75 bar/km at a slurry velocity of 0.72 m/sec; capital cost was estimated at US\$25 million, annual operating cost at US\$2.8 million, and annualized capital cost at US\$4.3 million.

7.0 ALTERNATIVE MARKETS FOR LIGNITE

Traditional markets for lignite in the Czech Republic are expected to decline in the coming decade with the decommissioning of older coal-fired power plants after completion of new nuclear units and the conversion of district heating plants and other use sectors to natural gas. Alternative uses for extensive reserves of lignite, which make up the largest economically viable energy resource in the Czech Republic, represent an important goal for the Czech national economy and for the welfare of displaced miners.

The search for new uses for lignite under current market realities in the Czech Republic and elsewhere should take into account previous successes and failures in the field. The availability of natural gas and crude oil in world markets at prices in the range of US\$1.50 to US\$3.00/GJ at the wellhead places a strict upper limit on the value of coal-derived fuels. Any successful upgrading process must provide a substantial added value to pay for the cost of processing and a profit. This tends to focus attention on limited-tonnage high-value fuel products such as smokeless briquettes for space heating and on non-fuel products such as activated carbon rather than on upgraded boiler fuels.

7.1 The Experience of North Dakota Lignite in the United States

A challenge similar to that experienced in the Czech Republic is faced by the North Dakota lignite industry in the United States, where production reached a plateau in 1985 and is currently poised between future expansion into new markets or possible decline due to environmental and price competition from other fuels. The past history of the North Dakota lignite industry also parallels current trends in the Czech Republic, with the closing of 320 small underground mines in North Dakota during the 1940s and 1950s and progressive consolidation of production into five large surface mines that efficiently produce 30 million tons annually at a productivity of 129 tons per miner per 8-hour shift. Major expansion in lignite production occurred between 1965 and 1985 with the building of eight large regional electric generating plants and the Great Plains Gasification Plant producing synthetic natural gas (SNG). However, growth has peaked, and future expansion is constrained by electric transmission line capacity and the availability of premium low-sulfur subbituminous coals in the bordering states of Montana and Wyoming at mine-mouth prices as low as US\$0.25/GJ.

To meet this competitive challenge, the state of North Dakota in 1987 formed a strategic partnership with the lignite industry to perform research and development for the purpose of expanding lignite production and employment through market diversification. One of the activities under that program has been a marketing feasibility study performed to identify ways for improving the competitive position of lignite through other resources as a fuel or raw material (Sinor, 1992). Selected findings of this report and other upgrading studies which have potential application to Czech lignite are summarized in the sections below.

7.2 Upgraded Utility Boiler Fuels

Thermal processes for drying or carbonizing lignite to produce upgraded solid boiler fuels typical involve capital costs in the range of US\$68 to US\$157 per product ton of annual capacity and processing costs including capital recovery on the order of US\$1 to US\$1.50 /GJ (Davy McKee, 1984; Sinor, 1992 [costs updated to 1995]). These levels would effectively

double the unit energy cost of a Czech lignite feedstock, making the upgrading methods at best only marginally competitive for large-scale electric power generation. In the United States, a niche utility market for coal upgrading is being explored under the Clean Coal Technology demonstration program for applications involving fuel switching from high-sulfur bituminous coal to low-sulfur compliance fuels, which are being prepared by cleaning and drying or carbonizing subbituminous coals or lignites (U.S. DOE, 1994).

At current oil price levels, the cost of coal carbonization processes cannot be supported by selling coal liquids, and financial success depends on receiving a premium price for the char or dry-coal product. Economic benefits that can be used to justify a price premium for char fuels in utility boilers include reduced costs for transportation, handling, and storage; pulverizers; boilers; and emissions controls. Also, boiler tests on upgraded coal products have shown operating improvements in boiler cleanliness, efficiency, and generating capacity, with no adverse effect on NO_x emissions. Currently in the United States, the economic feasibility of upgrading coal depends on the availability of an alternative fuels tax credit which may offset up to about US\$1/GJ in processing cost.

In the past, a number of commercial coal pyrolysis plants for producing char and liquid fuels were built in the United States starting in the 1920s, but none remains in operation. Texas Utilities built a power plant in the 1950s to burn char from a pyrolysis plant, but the production of liquid fuel was not economically successful, and the power plant was modified to burn dried lignite (Sinor, 1988). The Amax Coal Company constructed a commercial fluidized-bed dryer for U.S. subbituminous coal in 1988 which failed because of its extremely fine and dusty product (Woessner, 1993). The Syncoal and Encoal upgrading projects currently under way as part of the U.S. Clean Coal Technology program are addressing problems of product quality.

In summary, the primary barrier issue to be addressed in upgrading lignite to a premium solid boiler fuel is cost, followed by the need to be sure that the product has sufficient strength and stability to prevent unacceptable moisture reabsorption, decrepitation, dust, and spontaneous heating during transportation, handling, and storage (Willson et al., 1992).

7.3 Smokeless Briquettes

The production of smokeless fuels from brown coals and lignites was the topic of a workshop sponsored by the U.S. Department of Energy in Prague in November 1994 (Energy & Environmental Research Center, 1995). The term smokeless is defined by a reduction in the tarry volatiles released during coal combustion. Both briquetted fuels and lump coal are used extensively throughout East Central Europe for residential space heating, as well as commercial and small industrial heat generation. Most of these applications do not involve high-quality low-sulfur fuels, and conversion to a smokeless solid fuel at significant added cost will likely be accomplished only through emissions legislation. Standards are set either by the emission rate (e.g., 5 grams per hour in the United Kingdom) or by the volatile content of the fuel (a maximum of 15 wt% defines smokeless fuel in the Czech Republic). Technically feasible improvements in production methods that would substantially resolve dispersed air pollution problems from the household use of coal or briquettes are available based on a variety of advanced methods for coal cleaning, drying, carbonizing and briquetting with additives.

The potential size of the market for smokeless fuels is illustrated by the estimated 50 million tons of residential solid fuel used throughout East Central Europe, including the Ukraine (Crowther, 1994). Briquettes of varying quality account for only about one-fourth of this total, and briquette production has been declining for economic and ecological reasons. Poland has closed its conventional (noncarbonized) briquetting plants and is now producing 180,000 tons of smokeless fuel per annum. Production of conventional briquettes in the former East Germany has declined by 90% to under 6 million tons per year for economic and ecological reasons. In western Germany, the current production of 10 million tons of brown coal briquettes represents only 15% of capacity. In the Czech Republic, only one plant, producing 600,000 tons per year of partially devolatilized briquettes, remains in operation at Vřesová, after the plant at Tisová was closed in 1993.

The consensus of the 1994 workshop in Prague was that a significant market for smokeless fuels in East Central Europe can be served by small plants using indigenous coals and drawing on some of the developmental methods described in Section 3.0 of this report. Imported natural gas and anthracite are the competing alternatives. Several smokeless fuel projects are being implemented or planned, including the ECOCOAL process in Poland (Dreszer and Ściażko, 1994), MIBRAG in central Germany (Mall, 1994), and an Ostrava hard coal project in the Czech Republic (Kren Consulting, 1993). Specifications for future smokeless fuels will typically include a volatile content below 10%; sulfur content below 1%; high heat content (e.g., 25 MJ/kg); good strength—both wet and dry; good ignition characteristics; low ash, size, and shape appropriate for heating appliances; and reasonable cost. Representative costs given for smokeless briquettes are in the range of US\$80 to US\$120/metric ton, or US\$3.20 to US\$4.80/GJ at a heating value of 25 MJ/kg.

7.4 Dry Lignite Powder

Dried lignite or brown coal powder is generally used on-site in combustion boilers following drying to reduce excessively high moisture (e.g., up to 70 wt% moisture in brown coals). The coal can be dried directly as in the Kramer mills used for Victorian (Australia) brown coal (Garner, 1984) or in an entrained-flow reactor (e.g., Parry dryer) as with Texas lignite (Texas Utilities, 1977). An indirect drying method being used in Australia for Loy Yang (Victoria) brown coal incorporates a steam heat exchanger immersed in a fluid-bed system (Hamilton, 1990).

Drying lignite for off-site use was pioneered in Germany, where it has been practiced for over 15 years. Currently, some 2 million metric tons/year of lignite powder are produced by grinding the dried material from a tubular steam drier and mixing it with the fines collected from ESPs. The dried powder is transferred pneumatically by air and stored in silos, carefully designed and with the necessary safety controls. The powdered product is delivered by road or rail tanker and handled pneumatically for use in the cement industry (Couch, 1990).

Germany is also producing pulverized and granular coke from Rhenish lignite for a variety of uses, including waste gas and wastewater cleanup, steel manufacture, electric furnace metal smelting, graphitization, and activated carbon manufacture. The noncaking Rhenish lignite containing alkaline constituents is produced in the following three sizes: <0.4, 0-1.5, and 1.25-5 mm. Grain sizes are selected to match the application (Schieb, 1994).

In the Czech Republic, fine dried lignite products having a calorific value between 21 and 25 MJ/kg and a low sulfur content of 0.5%–0.8% are obtained from the gasification and briquetting plants operated by the Sokolov Coal Company at Vřesová (Keller et al., 1994). Lignite dust is separated from the dried feed stream going to Lurgi fixed-bed gasifiers, taken from ESPs downstream of a steam tube dryer. Lignite powder is also produced by grinding dried coal from the briquetting plant. The dried products are pneumatically conveyed in nitrogen to special storage tanks and trucks to be supplied to consumers.

7.5 Marketing Opportunities for Low-Rank Coal–Water Fuel

CWF is a mixture of finely ground coal, water, and a chemical stabilizing agent that is designed to replace heavy fuel oil in industrial and utility boilers. The product is handled like oil, using similar tank storage, pumps, and atomizing equipment. With deep cleaning to reduce ash and sulfur, future markets for CWF may include marine and railroad diesel engines and gas turbines. CWF can be produced from either bituminous coal or low-rank coals (subbituminous, lignitic, or brown coal), but the processes involved are substantially different.

Until recently, development of CWF has been primarily based on bituminous coal, which, because of its low moisture-holding ability, can be used directly to produce CWF without prior thermal treatment to remove inherent moisture. However, since bituminous coal is hydrophobic (the surface repels water), additives are needed to reduce the viscosity of the CWF and to prevent settling. The added cost of additives has been a major deterrent to commercial development of CWF in the United States. However, in some regions of the world where coal pipelines are more economic than new rail transport facilities or fuel diversity is desired to avoid dependence on imported oil, commercial development of CWF is already well advanced. Production of 4.3 million tpy of bituminous CWF commenced in Russia in 1989 to supply fuel for six 220-MW utility boilers through a 163-mile pipeline from Belevo to the Novosibirsk power plant. An Italian firm, Snamprogetti, is involved in the Russian project and also recently opened a 500,000-tpy CWF plant in Sardinia this year. In Japan, where fuel diversification is a strategic policy issue, over 1 million tpy of CWF capacity has been constructed to utilize imported bituminous coal, and an additional 1-million-tpy capacity has been built in Yanzhou, China, to supply bituminous CWF. Japan is expected to utilize up to 8 million tpy of CWF by the year 2000.

A potentially large market for CWF exists around the world because of the many oil-fired utility boilers that are not fully utilized. For example, only about 40% of the oil-fired capacity in western Europe is currently used (Sinor, 1992). Bituminous CWF, at an estimated cost of about US\$3/GJ, can be economically competitive with fuel oil in some oil-fired utility applications, but it cannot compete with coal in boilers designed to burn coal directly. Industrial boilers represent a future market with different retrofit requirements.

The combustion characteristics of CWF are linked to coal characteristics and hardware design, including the atomizer, burner, and configuration, as first summarized by Beér (1985) and more recently by Anderson et al. (1994). Combustion programs at ABB Combustion Engineering in the United States have developed retrofit burners for a range of CWF applications involving different bituminous coals and ash levels (Sinor, 1992). Initially, bituminous CWF experienced poor combustion performance because of particle agglomeration during heating through the plastic state, but this has been partly remedied by improving

atomizer design to achieve acceptable carbon burnout in boilers having a residence time of several seconds. In compactly designed oil-fired boilers and coal-fired diesels or gas turbines, where burnout must occur in a fraction of a second, carbon burnout can only be achieved for bituminous CWF by fine grinding, at significant added cost. Since ultrafine grinding can also result in undesirable (dilatant, shear thickening) flow behavior and poor atomization, a compromise must be reached between the fineness of grinding and the rheological characteristics of bituminous CWF.

Low-rank coal-water fuels (LRCWF) offer important advantages in achieving rapid carbon burnout without fine grinding because of the high reactivity of the feed coal and the retention of volatile tar on particle surfaces during processing. Combustion tests on normal pc-grind subbituminous LRCWF produced by the EERC have demonstrated superior carbon burnout in both conventional residence time firing (e.g., 99.8% burnout) and short residence time firing in a gas turbine (>99% compared to 97% for micronized bituminous CWF) (Anderson et al., 1994). Sulfur emissions can be reduced by precleaning the feed coal. Also, the addition of small amounts of limestone to LRCWF has been shown to be effective in reducing sulfur emissions in preliminary tests at the EERC. Processing in hot water also serves to remove soluble alkalis which are a principal cause of severe boiler fouling when burning high-sodium coals. LRCWFs from various test coals have typically produced a fine, powdery fly ash which is less likely to cause erosion and deposition problems in compact boilers than bituminous coal ash.

The cost of processing LRCWF is estimated to be in the range of US\$1 to US\$3/GJ without the cost of the feed coal, depending on the product specifications (e.g., ash and sulfur contents); the moisture, ash, and sulfur contents of the feed coal; the size of the plant and economy of scale; and variations in the cost of capital. LRCWF is competitive with both fuel oil and bituminous CWF and may offer economic advantages in retrofit cost and emissions compliance depending on the feed coal properties and precleaning. The high reactivity and favorable ash characteristics of LRCWF are expected to allow for less boiler derating and auxiliary fuel use compared to bituminous CWF.

In the Czech Republic, interest in the production of LRCWF would likely be restricted to industrial boilers currently burning fuel oil and not involve utility boilers. Lignite feedstocks for producing LRCWF for oil-fired industrial boilers should contain minimum amounts of sulfur and ash. Cleaning by wet gravity or dry magnetic methods should also be considered to improve the quality of the product. Based on the data for Czech lignites given previously in Table 1, the selected samples of Bílina lignite analyzed by the EERC show an average ash content of 7.3% (db) and sulfur content of 1.2% (daf). Wet gravity cleaning of these samples at 1.3 specific gravity yielded 4.0% ash and 0.7% sulfur contents at 90%-93% energy recovery, and dry magnetic cleaning resulted in 5.6% ash and 0.8% sulfur at 95% energy yield. These analyses indicate that selected sources of Bílina lignite would be suitable feedstocks for producing LRCWF and that precleaning would offer significant benefits in improving product quality.

7.6 Form Coke

Trends in the metallurgical industry are generating a growing interest in alternative sources of coke because of heightened environmental regulation, scheduled shutdowns of aging slot ovens, increased prices for high-quality coke, and specialized needs of foundry iron and steel producers. Manufacturers of coke are demanding new processes that are environmentally acceptable and that offer increased efficiency and high quality at market-competitive prices. The growing impact of environmental legislation around the world is forcing coke producers to expand their interest to include low-sulfur low-ash noncaking coals. The form coke process is adaptable to the nature of the coal since the processing conditions can incorporate appropriate steps for either caking or noncaking coals. Further, the form coke process is continuous, in contrast to the slot-oven coking process.

Formed coke can be produced in different sizes, shapes, and qualities to meet the needs of selected markets. Metallurgical processes depending on high-quality carbon, such as that derived from wood, are increasingly being forced to use higher-cost products as supplies of timber decrease, labor costs increase, and environmental regulations become more stringent and widespread. Char made from low-ash low-sulfur coal offers an acceptable high-quality alternative. The Japanese steel industry, in particular, sees the need for a range of form coke products for iron, steel, and metal smelting operations. For example, Nippon Steel has developed dumbbell-shaped briquettes that improve gas permeability in its blast furnaces. By modifying the furnace heating cycle, Nippon Steel has also been able to control the problem of fissure generation in the utilization of form coke (Kato and Komaki, 1994).

7.7 Synthetic Liquid Fuels

Synthetic liquid fuels produced by the direct hydrogenation of coal are estimated to cost approximately US\$30/bbl using the most recently developed liquefaction technologies. These fuels cannot compete with refined petroleum products at current world crude oil prices of about US\$20/bbl. However, starting in the 1930s and 1940s, substantial quantities of liquid fuels from coal, totaling more than 1 million tons annually, were produced in a number of European countries, including a plant in the Most region of the Czech Republic built in 1942 for the hydrogenation of lignite tar. The Most plant and other coal liquid plants that continued to operate into the 1960s, including plants at Leuna, Bohlen, and Zeitz in central Germany using brown coal, have all since been decommissioned for economic reasons. The only plants currently producing substantial amounts of motor fuels from coal are the three Sasol synthesis gas plants in South Africa with an estimated total production of 150,000 bbl/day (about 6 million tons per year) of gasoline, diesel fuel, waxes, and specialty products. The special conditions that justified European plants to be built starting in the 1930s and in South Africa in the 1950s and 1980s do not exist in Europe or the United States today. Changing circumstances that would favor a renewal of interest in synthetic coal liquids would be either a substantial rise in world oil prices, which is likely in the long run but unpredictable in the short term, or else a breakthrough in the technology for producing liquid fuel from coal. There is some scientific basis for expecting a significant reduction in the cost of liquid fuels produced from lignite based on highly favorable yields from staged low-severity laboratory studies (Hetland et al., 1995).

7.8 Methanol

Methanol is being produced from coal in the United States at the Tennessee Eastman plant in Kingsport, Tennessee. In general, methanol made from coal is not currently competitive with production from natural gas because of the much higher capital cost of a coal-based plant. However, demand for methanol is rising because of its use along with isobutylene for producing methyl *tertiary*-butyl ether (MTBE). MTBE and other oxygenates are being added to gasoline in the United States to promote combustion efficiency to meet the requirements of the 1990 U.S. Clean Air Act Amendments. Market analysts predict that a substantial amount of new methanol production capacity will be needed for several years (Sinor, 1992), and in special circumstances, some of that capacity may be coal based. In particular, coproduction with electric power generation has been evaluated as a promising approach, where the methanol would be produced in a once-through process and the unconverted synthesis gas would be used as fuel for an IGCC plant. The one-step liquid-phase process developed by Chem Systems and Air Products in the United States offers the additional benefit of using carbon monoxide-rich synthesis gas directly without water/gas shift conversion from CO to hydrogen and subsequent removal of carbon dioxide.

7.9 Synthetic Natural Gas

Synthetic natural gas (SNG) is being produced in the United States from North Dakota lignite at the Great Plains plant, which was commissioned in 1984. This plant uses fourteen Lurgi Mark IV gasifiers to convert 6 million tons of screened lignite (6- to 50-mm size) to synthesis gas that is cleaned and converted to 50 billion standard cubic feet of methane or SNG annually (1.6 billion standard cubic meters), equivalent to about one-fourth of natural gas demand in the Czech Republic. The project was conceived in the 1970s when it was believed that the United States would run short of natural gas supplies and that prices would rise above the US\$6/GJ level necessary to support the cost of producing coal-derived methane. That expectation was not realized, with the wellhead price of natural gas instead dropping to current low levels of under US\$1.50/GJ in the United States. As a consequence, the US\$2 billion capital investment in the plant could not be repaid, and the private consortium of five regulated gas pipeline companies that built the plant defaulted on the federal loan guarantee in 1985, causing ownership of the plant to pass first to the U.S. DOE and then to the Dakota Gasification Company at a substantially reduced investment cost. However, even the operating cost for producing SNG is approximately US\$2.50/GJ, and the plant is operating under sales agreements with four of the sponsoring gas pipeline companies that continue to provide demand payments compensating for the low market price of the gas until the year 2001. The Dakota Gasification Company has devoted considerable effort to increasing revenues through by-product sales, which accounted for about 12% of plant income in 1993. By-products include phenol, cresylic acids, naphtha, creosote, krypton and xenon gases, nitrogen, ammonia, and sulfur. Plans are under way to change the plant's sulfur control technology to a first-of-a-kind ammonia based scrubbing process that will produce ammonium sulfate fertilizer instead of elemental sulfur. Also, future plans call for converting one-fourth of the plant's capacity to ammonia production, which recently reached historic high price levels in U.S. fertilizer markets. In summary, the Great Plains plant is expected to continue to operate profitably by shifting its product distribution away from low-priced SNG to higher-value fertilizer and fuel products. Commercial success rests on the favorably low purchase price for the plant when it

was acquired by Dakota Gasification Company in 1988, and new ventures for producing a similar mix of products from coal would be doubtful in the near future.

7.10 Activated Carbon

Activated carbons of various qualities are widely used both in liquid-phase applications for separation of organic or inorganic compounds and in gas-phase applications for solvent recovery and air toxics control. At present, the purification of drinking water and treatment of wastewater are the largest uses of activated carbon in the United States. However, as air pollution standards become stricter and more widely applied, the use of activated carbons to control gaseous effluents from numerous sources such as metallurgical and chemical plants, wood product facilities, waste incinerators, and even paint shops and dry cleaning establishments will increase substantially. Activated carbons can be tailored to achieve some degree of selectivity in separating acid gases, halogen compounds, heavy metals, mercury vapor, volatile organic carbons, and other pollutants from liquid and gaseous effluent streams. The average value of activated carbons in the United States was approximately US\$1000/ton for powdered activated carbon and US\$2000/ton for granular activated carbon in 1990 (Sinor, 1992), providing a potential opportunity for producing a high value-added product for a growing market.

Lignite from the U.S. Gulf Coast region is presently used to produce activated carbons, but overall, a wide range of raw materials are used, including higher-rank coals and other materials such as coconut shell, wood, petroleum coke, peat, and bone. The starting material importantly affects the surface area and pore size distribution of the product, and it is these properties that determine the suitable applications for various activated carbons produced from different raw materials. Currently, brown coal- and bituminous coal-based carbons are being used for adsorbing SO₂, HCl, mercury vapor, and volatile organic carbons from waste incinerator stack gases in Germany and Japan. With the addition of ammonia to the flue gas stream, NO_x can also be controlled. As environmental regulations become more stringent in the United States, these capabilities may be applied to power plants, cement kilns, and other combustion sources, as well as waste incinerators. The EERC is performing research to modify the surface of U.S. lignite char to provide optimum capture of mercury vapor in both metallic and chloride forms.

7.11 Humate Soil Conditioners and Ion-Exchange Products

Several upgraded products in research and production stages are based on the exchange properties of humic acids in agricultural uses, wastewater treatment, and other applications. The cation exchange, pH buffering, and water retention properties of coal humates are similar to those of organic matter in soil and peat. Various formulations prepared from coal have been used to improve the physical, chemical, and biological fertility of deficient soils. The use of brown coal directly to increase the tilth and friability of clayey soils has been demonstrated, but it is not practiced commercially. Research worldwide to produce long-lasting fertilizers based on coal humates have met with varied success (Heng, 1991). High-temperature treatment of coal humate in air and ammonia at 300° to 350°C have had limited success because of tar toxicity and poor nitrogen availability. However, near-ambient sequential processing in dilute nitric acid, ammonia, concentrated nitric acid (for oxidation), and ammonia for final neutralization has been used to produce ammoniated polycarboxylic acid (AMP) fertilizers that

are as effective as urea. A related process in Japan is used to produce Azumin, a nitrogen fertilizer for gardening. Other researched applications for coal humates include the treatment of contaminated soils, reclamation of mined land, control of odor, and the coating of seeds to improve germination.

Purified humic acids are also used in various applications such as oil well drilling mud formulations, binders, and wastewater treatment. Extraction of humic acids from oxidized coal at yields as high as 80%–90% can be accomplished by treating with alkali (NaOH, KOH, or NH₄OH) and reacidifying to precipitate the solubilized humic material. The useful properties of the purified humic acids include high cation exchange capacity, anion complex formation, pH buffering, transfer of nutrients in plant physiology, and the formation of viscosity-controlling gels. The action of humic acid as a biostimulant is not firmly established.

The Research Institute of Inorganic Chemistry in Ústí nad Labem has reported promising results from work on humic acids supported by Břilina Coal Mines (Kozler et al., 1993). Selected coals containing 13%–15% mineral matter and 75 wt% humic acids have been shown to have good ion-exchange potential for separating heavy metal (Cd, Pb, Ra) from industrial wastewater. Humic acids have also been used in fertilizer formulations. Results are covered by several Czech patents. The Doly Břilina Coal Company is currently manufacturing about 3000 tons per year of sodium humate.

7.12 Coal Combustion By-Products

Increased cost and regulation are making the disposal of coal ash and calcium sulfate scrubber wastes an increasingly undesirable option in the United States and many other places. Only 31% of the coal ash and slag produced in the United States in 1991 was beneficially used, and only 1.9% of the scrubber waste was used (American Coal Ash Association, 1991). Several countries in the European Union have a strong emphasis on coal ash utilization and achieve remarkably high levels of beneficial use, including the Netherlands (100% utilization) and Denmark (90% utilization) (Clarke, 1993). The quality characteristics of coal combustion by-products vary widely at present and will become more diverse in the future with the increased application of FGD, FBC, and gasification combined-cycle systems. There are important environmental and economic benefits to be gained from the increased utilization of coal by-products and other waste materials as resources. Environmental benefits are obtained from reduced solid waste, conservation of natural resources, and reduced energy consumption and CO₂ generation where by-products can be substituted for energy-intensive primary products such as portland cement. Economic benefits include reduced construction costs, savings in ash handling and disposal costs, and the creation of marketing opportunities for by-product sales.

The value of many coal combustion by-products has been well established by research and commercial practice (Manz, 1984, 1985, 1993). As an engineering construction material (e.g., cement replacement), they enhance strength and durability while reducing cost. In agriculture and mine land reclamation, ash and gypsum by-products can improve the tilth of deficient soils and provide some plant nutrients. In waste stabilization, their pozzolanic and cementitious properties can be used to immobilize hazardous nuclear, organic, and toxic metal wastes for safe environmental disposal. Numerous products can be manufactured from coal combustion by-products, including blended cements, gypsum, aggregate, concrete block, and other cast concrete products, mineral wool, brick and other ceramic products, fillers for metal

and plastic products, and even paint. Specialized construction applications include road base, controlled-strength fill, and marine applications in break waters and piers. In all of these applications, it is vitally important to match the properties of the coal by-product with the requirements of the application.

Product specifications for coal by-product application should address environmental, engineering, and economic performance criteria together rather than separately. The environmental safety of a range of coal combustion by-products has already been determined, with the overwhelming weight of evidence indicating that these products are generally not hazardous under regulatory definitions and that they pose no greater concern than a large number of common solid materials, including cement, rock, and soil (Schwartz et al., 1993; Smith, 1991; Hassett et al., 1991, 1993). However, for particular coal combustion by-products or sources that have not been environmentally tested for beneficial use, additional validation may be required before unrestricted use for a class of application can be approved. Engineering and economic performance criteria should be pursued through standards organizations (e.g., ASTM in the United States), government agencies that are potential by-product users (e.g., the Highway Administration), and universities performing technology-based research and development.

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