Coal Upgrading Program for Usti nad Labem, Czech Republic Task 8.3

**Topical Report
October 1994 - August 1995**

Brian C. Young
Mark A. Musich

October 1995

Work Performed Under Contract No.: DE-FC21-93MC30097

For
U.S. Department of Energy
Office of Fossil Energy
Morgantown Energy Technology Center
Morgantown, West Virginia

MASTER

By
University of North Dakota
Energy & Environmental Research Center
Grand Forks, North Dakota

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TASK 8.3

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EXECUTIVE SUMMARY

Coal has been a major energy source in the Czech Republic given its large coal reserves, especially brown coal and lignite (almost 4000 million metric tons) and smaller reserves of hard, mainly bituminous, coal (over 800 million tons). Political changes since 1989 have led to the reassessment of the role of coal in the future economy as increasing environmental regulations affect the use of the high-sulfur and high-ash brown coal and lignite as well as the high-ash hard coal. Already, the production of brown coal has declined from 87 million metric tons per year in 1989 to 67 million metric tons in 1993 and is projected to decrease further to 50 million metric tons per year of brown coal by the year 2000. As a means of effectively utilizing its indigenous coal resources, the Czech Republic is upgrading various technologies, and these are available at different stages of development, demonstration, and commercialization. The purpose of this review is to provide a database of information on applicable technologies that reduce the impact of gaseous (SO_2, NO_x, volatile organic compounds) and particulate emissions from the combustion of coal in district and residential heating systems.

This review reports on various coal-upgrading approaches, including beneficiation options of cleaning, drying, and agglomeration; pyrolysis and briquetting for smokeless fuels; and the use of additives for control of sulfur dioxide and smoke emissions from solid heating fuels. It also briefly discusses other upgraded products derived from Czech coals, in addition to smokeless fuels. Physical cleaning of lignites is generally not practiced because of the structural composition of the inorganics in these coals and the likely increase in moisture content and, hence, the increased cost of drying. However, exceptions have been noted, e.g., in Turkey and Austria. Advanced coal-cleaning methods, applicable to bituminous coals, are not an option, but new developments in magnetic cleaning offer some promise for both high- and low-rank coals. For some high-value-added products (e.g., activated carbons), chemical cleaning methods such as the Gravimelt process may be considered, but their high costs require careful justification.

New drying processes have been demonstrated for low-rank coals in recent years. SynCoal, hydrothermal treatment, SGI's "liquids from coal process," and the Koppelman Series C process are notable clean coal technologies for which various benefits have been claimed but for which commercial reality has yet to be realized. Nonetheless, some of these processes do offer flexibility in the product slate that may lead to increased financial return and justify the substantial investment. In some cases, the solid products need to be agglomerated to provide the necessary stability. Agglomeration through briquetting or pelleting/pelletizing often requires a binder which can appreciably add to the cost of the final product. However, a new, high-pressure, moderate-temperature briquetting process affords a lower-cost alternative. Key elements of briquetting with and without binders and the use of additives to improve briquette quality and environmental acceptability are briefly outlined.

Several of the many pyrolysis (or carbonization)/briquetting processes developed over the last 40 years for the production of smokeless fuels are summarized. Specific reference is made to those processes developed and/or commercialized for low- and high-rank coals in the United Kingdom

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(UK), Poland, Australia, Germany, and the United States. Similar standards for tarry volatile emissions currently exist in the UK and Germany. In these regions, the standard is based on combustion measurements specifically designed for solid fuels. In the Czech Republic, coals having at most 15 wt% volatile matter are considered smokeless. Kren-Consulting in the Czech Republic has conducted a feasibility study of the market for producing smokeless fuels from Ostrava hard coals. Assuming a market of 500,000 metric tons for the years 1998–2000 and a market price some 40% higher than that for existing anthracite coal briquettes, Kren-Consulting recommended that the Czech smokeless fuel project was a good alternative to scaling down the coal industry in the Czech Republic. The new smokeless fuel would exceed proposed future emission standards.

Control of sulfur dioxide emissions in the Czech Republic is of great concern since maximum daily concentrations in some cities of up to 20 times that of the permitted limits have been recorded (January 1992)—concentrations comparable to those exhibited in the 1952 London smogs. Apart from cleaning or pyrolyzing to reduce the sulfur content of solid fuels, additives such as hydrated lime can be incorporated into briquettes, as is being done with the lignite briquettes produced by MIBRAG of Germany. Indeed, the company plans to replace the hydrated lime with the waste product from a calcium carbide plant to produce some 600,000 tons of briquettes a year to conform with the government-prescribed Small-Scale Furnace Regulation for an emission-effective sulfur content of less than or equal to 1 wt%.

Smokeless fuels are not the only products being assessed or developed from low-rank coals in the Czech Republic and elsewhere. Value-added products such as humic acid, slow-release fertilizers, soil conditioners, drilling muds, activated cokes and carbons, and form coke for metallurgical applications are under different stages of development and commercialization worldwide. The Doly Bílina Company in the Czech Republic is now manufacturing about 3000 metric tons of sodium humate from lignite. The Rheinbraun company in Germany produces about 200,000 metric tons of active coke from lignite for wastewater purification and waste gas cleanup. Recently, the Coal Technology Corporation in the United States has demonstrated a newly patented continuous process for producing form coke for blast furnaces and iron foundry cupolas.

Clean energy from coal using some of the newly demonstrated technologies will require a large investment and substantial time on the part of the Czech Republic. Simpler and smaller-scale processes need to be examined, such as char briquetting with binders and additives, in order to reduce emissions and improve heating values (i.e., efficiency). In this way, small incremental steps can be made at a bearable cost to help reduce the current level of pollutant emissions to acceptable levels. Technology selection, risk assessment, and funding arrangements are major issues to be resolved by the Czech government and corporate decision makers over the next few years as they strive for a cleaner environment.

Three coals from the Czech Republic, including Bílina and Nástup lignites from Northern Bohemia and Ostrava bituminous coal from Moravia, were subjected to a limited technical investigation at the Energy & Environmental Research Center (EERC) to assess the potential for upgrading. All coals were analyzed for ash and sulfur contents, forms of sulfur, and ash composition. Bílina and Nástup lignites were treated by wet and dry physical cleaning methods to reduce ash and sulfur content. Physically cleaned Bílina lignite and raw Ostrava bituminous coal were carbonized to reduce volatile matter content. Select raw, physically cleaned, and carbonized coals were aggregated (tableted) with starch binder and calcium carbonate as a sulfur capture agent. The aggregated fuels were analyzed for ash and sulfur contents and ash composition, and then

subjected to bench-scale combustion tests to evaluate their potential application as district heating and home heating fuels.

The ash and sulfur contents of the Bílina lignite were reduced by 42 and 36 wt%, respectively, at Btu yields of 90% using float–sink testing. Further, dry physical cleaning via rare- earth magnetic separation (REMS) produced significant sulfur reduction (27 wt%) at a Btu recovery of 95%. Nástup lignite was not amenable to sulfur reduction at ¼ in. × 20 mesh via float–sink or REMS.

The tablet fuel analyses were compared to quality standards established in the Czech Republic for the allowable maximum sulfur content (g/MJ basis) of indigenous briquettes. Fuels prepared from raw Ostrava bituminous coal and Bílina lignite cleaned by float–sink methods would qualify as fuels for steam raising in business and energy installation applications. Further, tablet fuels prepared from carbonized Ostrava bituminous coal and carbonized Bílina REMS product would qualify as smokeless fuels for home heating applications by virtue of their sulfur and volatile matter contents. The Bílina lignite-derived fuels have potential emission effective sulfur contents comparable to the Ostrava fuels because of the sulfur capture agent.

COAL UPGRADING PROGRAM FOR ÚSTÍ NAD LABEM, CZECH REPUBLIC

1.0   INTRODUCTION

The major political changes occurring in Eastern Europe, and the Czech Republic in particular, since 1989 have engendered a dramatic reappraisal of the use of energy and subsequent impact on the environment. The Czech Republic is highly dependent on coal for its primary energy supply (64%), although a significant portion of the country's electrical generation is derived from nuclear power (27% in 1994). As of January 1993, the Czech Republic had over 4700 million metric tons of recoverable coal reserves, mainly consisting of brown coal and lignite (~3930 million metric tons) and hard or bituminous coal (~840 million metric tons) (Ministry of Industry and Trade, 1994).

These quoted reserves are apparently upper limits. An alternative source has quoted the following: 2340 million metric tons of lignite and 980 million metric tons of hard coal (IEA, 1994). This source also indicated that at current production rates, minable reserves of lignite and hard coal remain for more than 30 and 50 years, respectively. Given the contraction occurring in the Czech coal industry, the reserves could last substantially longer.

A substantial amount of the coal is low grade, characterized by both high ash and sulfur contents. For the brown coals of the Most and Sokolov regions, the respective ash content ranges are 18–36 wt% and 18–29 wt%, and the respective sulfur content ranges are 0.4–3.5 wt% and 0.5–5.6 wt%. In comparison, the bituminous coal in the Ostrava region has an ash content of 8–40 wt% and a sulfur content of 0.4–0.9 wt%. The reported analytical data for the Czech coal regions vary somewhat, depending on the information source and location or determination point. A representative data set of coal properties, compiled by the Energy & Environmental Research Center (EERC) and including its own analyses, is given in Table 1.

The Czech coal regions are shown in Figure 1. The Northern Bohemia coal basin (including Bílina and Nástup) covers 1400 square kilometers, of which the coal seam occupies over 60%. Existing surface mines contain 2.2 billion tons of brown coal, with additional reserves of 1.8 billion tons. At an annual output of almost 61 million tons of brown coal in 1990, this amount represented over 72% of the total brown coal output of the former Czechoslovakia and >57% of the total solid fuels output. By 2005, it is expected that the output will decrease to between 41 and 50 million metric tons.

Although the production of lignite and bituminous coal is declining, a strong economic incentive exists to use indigenous fuels. Upgrading or beneficiating the coal is a potential option for improving the efficiency of coal use as well as its environmental acceptability.

Ústí nad Labem, one of the cities in the Northern Bohemia region of the Czech Republic, is a typical example of a city that is very dependent on the use of coal. An industrial city, it contributes significantly to the high pollution levels of the "Black Triangle" region, which includes other cities of the Czech Republic, Poland, and former East Germany. The population of Ústí nad Labem, approximately 121,000 people, is mostly centrally located.

TABLE 1

Brown Coal and Lignite Properties Derived from Various Sources[1]
(analyses adjusted to a common basis)

Source:	EERC	SD a.s.	EERC	Martinek	Martinek	Martinek	Martinek[2]
Year:	1995	1994	1995	1988	1988	1988	1988
Mine/Region:	Bílina	Bílina	Nástup	N. Bohemia	N. Bohemia	Sokolov	Sokolov
Samples:	Avg. of 4	Range	1	Range	Average	Range	Average
Proximate Analysis							
Moisture, wt% as-mined	30.1	24–30	38.7	6–55	29.4	35–41	37.5
Ash, wt% dry basis	7.3	11–43	18.8	26–44	25.5	22–55	36.8
Volatile Matter, wt% daf[3]	51.5	52–54	55.3	52–56			
Fixed Carbon, wt% daf	48.5		44.7				
Ultimate Analysis, wt% daf							
Carbon	73.0		69.8				
Hydrogen	6.0		5.5				
Oxygen	18.7		20.9				
Sulfur	1.2		2.1	0.8–5	2.0	1.2–10	3.0
Nitrogen	1.1		1.6				
Sulfur Forms, wt% daf							
Organic	0.78	0.5–0.8	1.27				
Pyritic	0.38	0.6–1.0	0.73				
Sulfatic	0.05	0.08–0.16	0.15				
Total	1.21	1.18–1.96	2.14				
Heating Value							
HHV[4], as-mined							
Btu/lb	8442		6173		6505		4802
MJ/kg	19.6		14.4		15.1		11.2
HHV, daf							
Btu/lb	13032		12401		12367		12156
MJ/kg	30.3		28.8		28.8		28.3
LHV[5], as-mined							
Btu/lb	7769		5519		5934		4214
MJ/kg	18.1	10.8–17.4	12.8	9–18.6	13.8	9.7–11.7	9.8
LHV, daf							
Btu/lb	11994		11087		11791		11178
MJ/kg	27.9		25.8		27.4		26.0
Ash XRFA[6], wt%							
SiO_2	36.3	45–60	49.8				
Al_2O_3	24.2	23–30	10.9				
Fe_2O_3	10.2	6–14	14.6				
TiO_2	1.2	1.3–2.3	0.4				
P_2O_5	0.6	0.2–0.4	0.8				
CaO	8.0	3–9	7.4				
MgO	4.1	0.2–1.8	2.6				
Na_2O	1.4	0.4–1.1	0.6				
K_2O	1.1	0.6–1.8	0.4				
SO_3	12.8	0.1–2.1	12.5				

[1] Adapted from EERC, 1995.
[2] Taken from Couch, 1988.
[3] Dry, ash-free.
[4] Higher heating value.
[5] Lower heating value.
[6] X-ray fluorescence analysis.
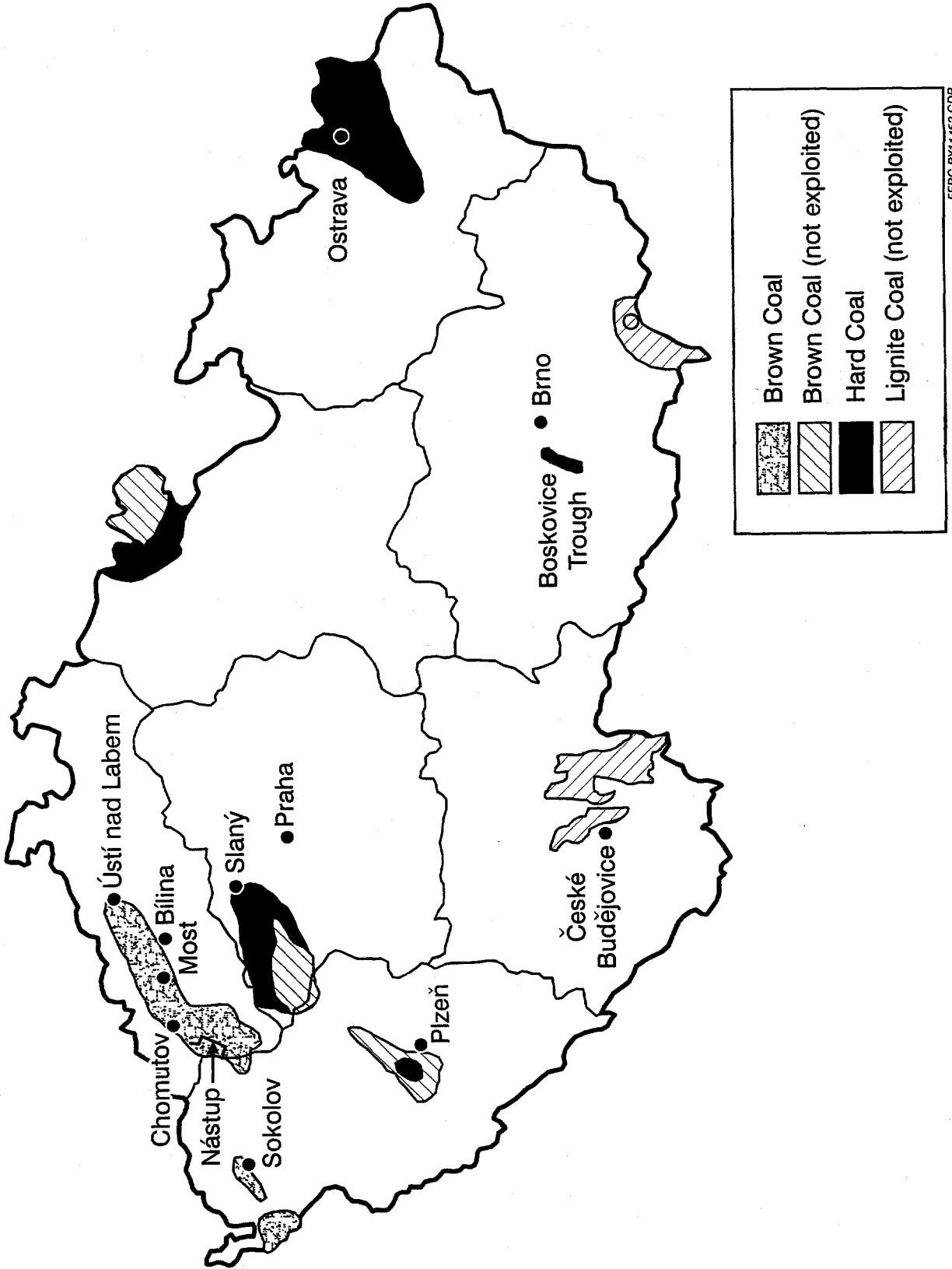
Figure 1. Coal regions in the Czech Republic (adapted from U.S. Environmental Protection Agency [EPA], 1992).

A critical concern of Ústí nad Labem is the substantial pollution attributable to emissions from sulfur dioxide (SO_2), nitrogen oxides (NO_x), and particulates arising from three large district heating systems that supply 80% of the heating energy demand, as well as from small residential coal-fired stoves. Average concentrations of SO_2 exceeding 100 mg m^{-3} and of particulates at typically 75 mg m^{-3} have been reported (Moldan and Schnoor, 1992). The city has eight heating plants (total output of 910 MW), about 100 local block boilers (125 MW total), and approximately 7400 small domestic boilers and stoves (Krivsky, 1994b).

Domestic heating in the Czech Republic uses some 7.4 million tons of raw lignite and 600,000 metric tons (Krivsky, 1995) of lignite briquettes (costing the equivalent of about US$42 [1100 Kc] per metric ton), as well as 640,000 metric tons of bituminous coal in a year. Combined, the power plants and large heating plants use 36 million tons per year of pulverized lignite, whereas the local boilers use 1.1 million tons per year (Krivsky, 1994b).

The lignite briquettes are produced at the one remaining briquette plant located at Sokolov, about 50 km west of the Tušimice mines. About 50,000 metric tons are exported to Slovakia and Germany (Krivsky, 1994b).

New environmental regulations for coal-burning units without desulfurizing devices came into effect on January 1, 1994, following the 1993 Act of the Ministry for Environmental Protection 206 on July 8, 1993 (Gavor, 1993). Two parameters will be used to control the amount of sulfur, based on the amount of sulfur per lower heating value (g/MJ): S^r_m aver., the average amount of sulfur in supplies of individual producers during a 3-month period, and S^r_m max., the maximum amount of sulfur in any fuel. The limits are initially divided between two regional groups:

- Regions requiring special air protection, such as Northern Bohemia

- Other regions for the period January 1 – December 31, 1994

The regulations define three categories of fuel use with respect to sulfur content. They are 1) fuel for energy installations (over 5 MW of installed heating capacity), 2) fuel for other business activities (over 50 kW of installed heating capacity), and 3) fuel for household use (up to 50 kW of installed heating capacity) (IEA, 1994).

Following January 1, 1995, the stricter limits of Group A will apply to all regions.
In particular, the national fuel quality standards for domestic lignite will be more stringent.
Table 2 lists the solid fuel standards from January 1, 1994, and includes the new ones for lignite from January 1, 1995. Special fuels such as K-Fuel or Gravimelt will be regulated by the limits for black coal.

The upgrading of low-grade coals may involve one or more techniques for the removal or reduction of the mineral species, moisture, or derived combustion products (e.g., SO_2). These techniques include dry beneficiation (e.g., magnetic separation of minerals), washing, oil agglomeration, ion exchange, hot-water drying, thermal pretreatment for moisture reduction, and briquetting. Depending on the type and grade of coal, coal-cleaning costs, including the cost of the

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TABLE 2

Fuel Quality Standards from January 1, 1994
(adapted from IEA, 1994)

	Maximum Sulfur Content, wt%	Mean Sulfur Content, g/MJ	Maximum Sulfur Content, g/MJ
Indigenous Lignite			
Households	–	0.65	1.20
Business Activities	–	0.95	1.70
Energy Installations	–	–	1.90
Indigenous Lignite – New Standards from Jan. 1, 1995			
Households	–	0.57	1.07
Business Activities	–	0.95	1.65
Energy Installations	–	–	1.90
Indigenous Lignite – Special Protection Zones from Jan. 1, 1994			
Households	–	0.57	1.07
Business Activities	–	0.80	1.20
Energy Installations	–	–	1.90
Indigenous Hard Coal			
Households	–	0.52	0.78
Business Activities	–	0.52	0.78
Energy Installations	–	–	1.90
Indigenous Briquettes			
All Uses	–	–	0.60
Imported Solid Fuels – All Uses			
Lignite	0.80	–	0.50
Hard Coal	1.00	–	0.50
Briquettes	1.00	–	0.50
Liquid Fuels			
Households	1.00	–	–
Business Activities	3.00	–	–

Source: Regulation 206 of 8 July 1993.

disposal of wastes, can be high. However, several advantages of beneficiation exist, namely, improved consistency of feedstock quality, enhanced heating value of the fuel, and reduced mineral content, resulting in less wear and corrosion of the boiler plant as well as lower emissions and reduced transport, handling, and storage costs for a specific power plant output (Couch, 1990).

The purpose of this review is to provide a database of information on applicable upgrading technologies for Czech coals, given the strategic perspectives and inherent coal reserves of the Czech Republic. The specific upgrading technologies to be addressed are those that reduce emissions of sulfur dioxide, nitrogen oxides, volatile organic compounds, and particulates released during coal combustion in district and residential heating systems.

2.0 TECHNICAL REVIEW

2.1 Beneficiation – Cleaning, Drying, and Agglomeration

2.1.1 Cleaning

Lignites, which, as already noted above, constitute a large fraction of Czech coal reserves, contain various amounts and species of inorganic matter, particularly alkali and alkaline-earth elements. As a consequence, they pose a difficult cleaning problem, owing to the nature of the mineral matter association and the cost of effective cleaning techniques. Because lignite inorganics are primarily associated or present as finely disseminated, difficult-to-liberate minerals, conventional cleaning procedures are often relatively ineffective. Other means are used to reduce or control the impacts of the inherent properties of lignite used as a fuel for power generation. For example, in the case of high alkali content leading to boiler fouling, boiler design changes or modified operation conditions are adopted. At power plants burning high-sulfur lignite, scrubbers will be installed rather than the coal being physically cleaned. However, where the cost of cleaning is a small fraction of the final coal product selling price, cleaning can be an option. Cleaning of coal for domestic heating use is not typically practiced. Nonetheless, if other beneficiation steps (e.g., to make a smokeless fuel) are to be used, the demonstrated enhanced value can make cleaning an attractive option in some circumstances.

Countries like the Czech Republic, which have a significant coal-based domestic heating and industrial fuel market, are likely to view the benefits of coal cleaning not only from an economic perspective but also from a strategic point of view. For example, Turkey, which has appreciable reserves of high-ash lignite, cleaned some 15% of this resource (Couch, 1990). A state-of-the-art coal washery with a capacity of 700 metric tons per hour was installed in the early 1980s, cleaning lignite with up to 40–50 wt% ash using a dense-media drum, dense-media cyclones, and jigs. Austria has tested a specially designed dense-media separator that has demonstrated a reduction in an ash level of almost 50% of that of the entering feedstock (Couch, 1990). The U.S. firm, Dow Chemical Company, has patented a specific arrangement of hydrocyclones, jigs, and classifying cyclones to minimize ash content and maximize coal recovery of Texas lignite (Jones and others, 1986; Lobbe Technologies Ltd., 1988). The process is particularly applicable at mine sites where substantial amount of overburden or seam partings are included in the excavated coal.

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Although conventional bituminous coal-cleaning technologies have been applied successfully to the cleaning of some poor-quality low-rank coals for power generation, the overriding concern is the handling, dewatering, and combustion of wet clean coals. Here, the increase in heating value arising from inorganic reduction has to outweigh any heating value reduction from increased surface moisture. A market for residential or stoker (industrial) fuels provides for the option of drying. Drying would further increase the cost, but the appropriate technology, e.g., high-pressure binderless briquetting (see below), would need to be selected to minimize this increase as well as to avoid increasing fuel dustiness, friability, moisture reabsorption, and the propensity to self-heat (leading to spontaneous combustion).

Advanced coal-cleaning technologies such as advanced froth flotation are not applicable to cleaning low-rank coals because of the hydrophilic nature of low-rank coal. Chemical cleaning using, say, carbonic acid, to remove mineral matter may be feasible and effective where value-added products, such as residential fuel briquettes, are in demand. The EERC has demonstrated this technology at the bench scale, achieving moderate ash reduction from North Dakota lignites (Anderson and others, 1993).

Dry cleaning using rare-earth magnetic separation (REMS) offers a convenient and beneficial alternative to conventional wet cleaning for high- and low-rank coals. Although not used commercially in the coal industry at present, it is used widely in the minerals industry. REMS technology involves feeding sized feedstock over a magnetic-head roller via a variable- speed belt. The roller consists of a neodymium–boron–iron disc that can separate mineral matter having paramagnetic and ferromagnetic susceptibility from coal which is diamagnetic (magnetic-repulsing). The technology is effective over the size range of 0.4–0.08 inches (10–0.2 mm) with good performance results, typically -1.2 to +0.3 inches (-3 to +0.8 mm) on high-rank coals (Donnelly and others, 1994). Recent results show the potential of REMS for a Czech Bílina lignite under testing at the EERC (Musich and Young, 1994). Donnelly and others (1994) report an engineering and economic evaluation of REMS as compared to wet cleaning, showing the cost-effectiveness of the technology when incorporated into a novel dry-cleaning plant design that they developed.

An alternative magnetic technology for cleaning dry coal cost-effectively and efficiently has also been very recently launched. It involves connecting the MAG-MILL upstream of an existing pulverizer system to remove hard and abrasive minerals (e.g., pyrite) by magnets before the coal is crushed to typically under 74 microns. The process is being developed by EXPORTech Co. Inc., New Kensington, PA (Coal & Synfuels Technology, 1995). Although initially aimed at high-sulfur bituminous coals, the MAG-MILL has the potential for application to some low-rank coals.

A very effective procedure to reduce substantially the level of sulfur and ash in high- and low-rank coals is the Gravimelt or molten-caustic-leaching process. This process, conceived and tested by TRW in the United States in the late 1970s (TRW Space & Electronics Group, 1993), involves blending coal with a melt of sodium hydroxide or mixtures of sodium and potassium hydroxides at 325°–415°C to eliminate almost all the sulfur and mineral matter in the coal. The process has been designed for small utilities or industrial boilers where the cost of flue gas desulfurization systems would be prohibitive. The Gravimelt product may have other applications also, such as an adsorbent (unactivated or activated) for pollutants, a precursor for metallurgical coke, or an ecological fuel for

home appliances. A schematic diagram of the Gravimelt process is shown in Figure 2. By using wash water as makeup and the effluent from the acid wash train without acid addition and by deleting the water treatment step, it is possible to gain large cost savings (almost 25%) and still generate a low-sulfur, moderate-ash coal product for commercial applications not requiring low-ash coal. The estimated processing cost for the moderate ash product is less than US$45 per ton. Such a product could be mixed with a binder and briquetted for use as an ecological fuel.

2.1.2 Drying

Technically and economically, drying is a key element in any upgrading process for low-rank coals. Drying low-rank coals increases heating value, improves combustion efficiency, and reduces transportation costs. However, drying low-rank coal can be a precarious process. Since high thermal shock and the removal of water result in decrepitation of these coals, the dried product is susceptible to dustiness, friability, moisture reabsorption, and self-heating as mentioned previously.

Several drying technologies—evaporative, nonevaporative, and pyrolytic—have been demonstrated, and a few have been commercialized or are near commercialization. The various technologies have been described elsewhere (Willson and others, 1992; Young and others, 1993a). Only a selected few will be briefly outlined here. Most conventional evaporative drying technologies operate at relatively low temperatures using hot flue gas as the drying medium. The design of the contacting equipment can involve entrained flow, a fluidized bed, or a rotary- louvered drum. The Parry entrained-flow drier and other driers in this class produce a product that lacks stability in terms of moisture reabsorption, dustiness, and self-heating. The dried product can be directly injected into a combustion boiler or aggregated to improve its stability. Following a recent patent by Davidson and others (1991), Tra-Det Inc., a U.S. company, has integrated the Parry dryer into a drying/briquetting process that avoids the use of binders but relies on high compacting pressure and the judicious selection of inlet moisture, particle size, and particle-size distribution (Davidson and Kalb, 1993; Young and Kalb, 1994). Although this technology does not reduce volatile matter, per se, it nonetheless produces a stable (towards moisture reabsorption and oxidation), strong, durable briquette having a moisture content around 8 wt% and providing a value-added product with enhanced heating value. This drying/binderless briquetting technology has been recently demonstrated at the pilot plant scale for subbituminous coals as well as for bituminous coal filter cake (Davidson and Kalb, 1993; Kalb, 1994).

Another evaporative drying technology is steam fluid-bed drying (SFBD), originally developed in Australia and now licensed to Lurgi Australia Pty. Ltd. (Hamilton, 1990). The SFBD process utilizes a heat exchanger connected to an external high-pressure steam source to dry finely ground brown coal. Heat is derived from a tube network immersed in the SFBD, and evaporated water is used to fluidize the coal. The moisture content of Victorian (Australia) brown coal can be reduced from about 60 to 15 wt%. The product will need to be stabilized by aggregation unless fed directly into a combustion boiler.

SynCoal has also developed a fluidized-bed, evaporative drying technology, now being demonstrated under the U.S. Department of Energy (DOE) Clean Coal Technology program. The drying plant is close to the Rosebud subbituminous coal mine near Colstrip, Montana. It entails a
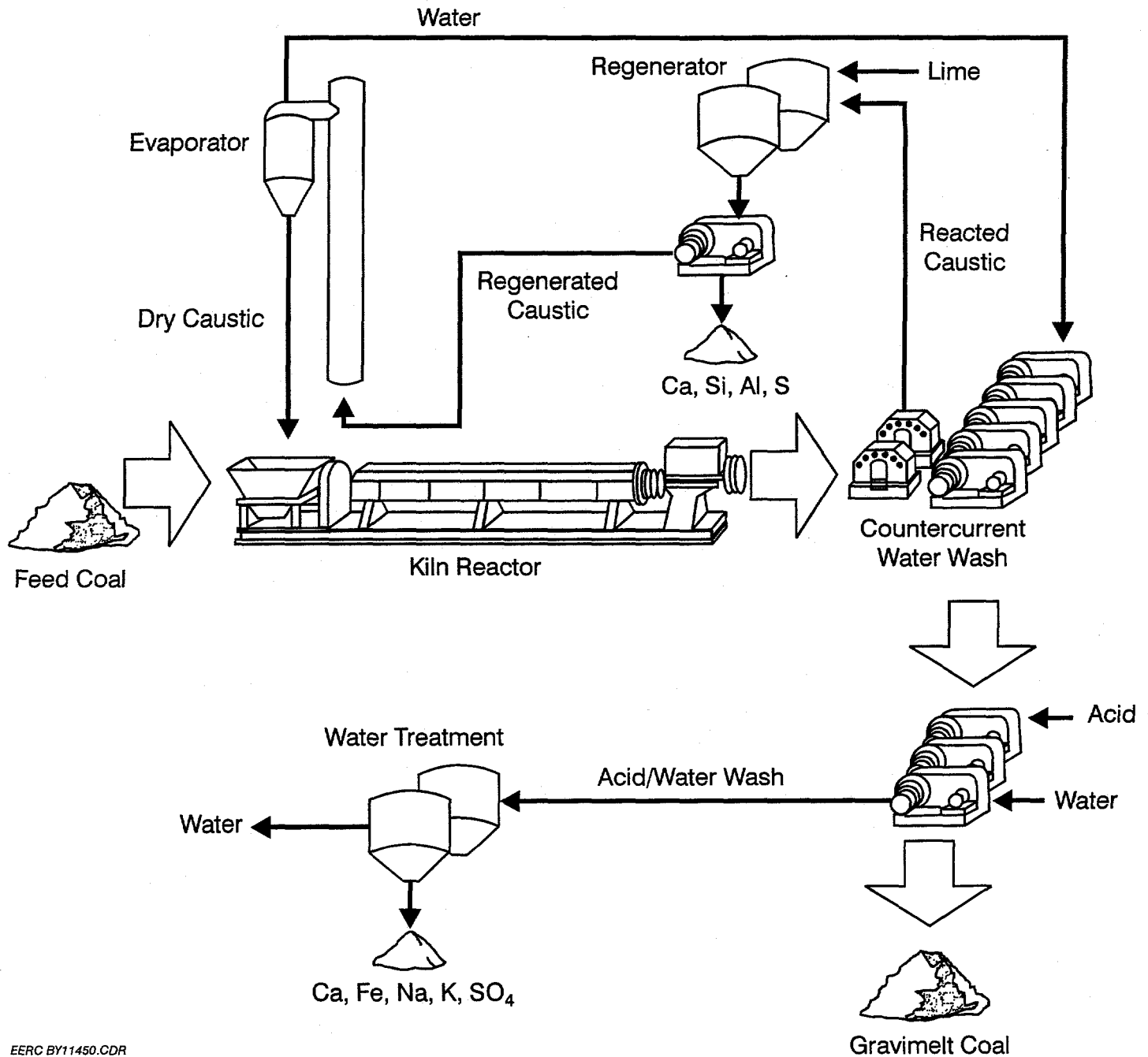
Figure 2. Schematic of the Gravimelt or molten-caustic-leaching process (from TRW Applied Technology Division).

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two-stage process using unique vibrating fluid-bed reactors designed to minimize fines generation. The first stage incorporates a relatively low-temperature evaporative drier, while in the second stage, additional heating is used to remove chemically bound water and some sulfur as well as to improve product stability. The process also involves inert gas cooling and pneumatic cleaning, the latter with air-table separators. Both fines and coarse products are obtained. Developments are in progress to enhance the stability of the fines product. Several coals, with moisture contents between 20 and 50 wt%, have been tested by the SynCoal process, resulting in coal products between 1 and 7 wt% moisture content. The 280,000-Mt/y plant has been constructed and demonstrated, and the process is now being optimized for commercialization (Greene, 1988; Niquette, 1994).

An alternative nonevaporative technology is hot-water drying (HWD), developed extensively for low-rank coals at the EERC. Producing a safe, easily handled, quasiliquid fuel or a briquette product, HWD is an advanced technology involving high temperature and pressure to remove irreversibly much of the inherent moisture in the original coal. During the process of liberating moisture and carbon dioxide, devolatilized tars and oils are pushed to the coal surface where they remain, forming a hydrophobic coating and sealing the micropores and preventing moisture reabsorption. The process allows the production of coal–water fuels (CWFs) with solid loadings rivaling those of commercial bituminous CWFs without requiring costly additives. Low-rank CWFs are a competitive replacement for heavy fuel oil in industrial and district heating boilers as well as utility boilers (Willson and others, 1994). Plans are currently in progress for establishing a commercial demonstration plant in Alaska. The EERC has also successfully made briquettes from HWD North Dakota lignite (Young and Musich, 1994).

Upgrading processes using pyrolysis are currently exemplified in the United States by the "liquids from coal (LFC) process" (Castro and others, 1994) and the Koppelman Series C (KC) Process (Gentile and Merriam, 1994). In the KC process, moisture reduction, decarboxylation, particle shrinkage, and sulfur reduction occur when the low-rank coal under pressure in a nitrogen atmosphere is heated indirectly by oil to 325°–475°C. In this automated batch process, gases and condensates are removed under high-pressure conditions without feeding or removing coal during processing. The technology has been demonstrated at the 1000-lb (450-kg)-batch level with Powder River Basin (Wyoming) subbituminous coal to yield an upgraded fuel having a heating content of about 28.4 MJ/kg (12,200 Btu/lb) and a 25% lower sulfur content than that of the feedstock. Typical product yields were 63–70 wt% of the feed coal, with nearly 100% energy recovery. Design data are still being collected for a commercial plant. The product is reported to exhibit good stability, but information on test conditions and product performance, particularly at the larger scale, is required to assess the product fully.

The LFC process, marketed by SGI International for upgrading subbituminous and lignite coals generates two value-added products: a liquid (coal-derived liquid [CDL]) and a solid (process-derived fuel [PDF]). The LFC process upgrades low-rank coal at reducing conditions and below fluidizing velocities by eliminating most of the moisture through convective drying and by significantly reducing the volatile matter (to about half of the original volatile content) through pyrolysis. The drying temperature is controlled to preclude the evolution of hydrocarbon gases, that are subsequently released during the pyrolysis step performed under controlled temperature conditions which depend on the selected coal (Nickell, 1993). The pyrolysis step also removes organic sulfur but not pyritic sulfur.

The SGI LFC technology is licensed to the ENCOAL Corporation by TEK-KOL (owners of the LFC technology), and ENCOAL is demonstrating the technology under the Clean Coal Technology program supported by DOE. Construction of the demonstration plant, located at Gillette, Wyoming, was completed in July 1992 (Castro and others, 1994). With operations under way, the plant has recently completed 68 consecutive days of operating at 90% capacity, producing in excess of 600,000 gallons of liquid product and 12,000 short tons of char. The latter, following stabilization, was stored for two months without significant self-heating. The stabilization step that follows the cooling of the pyrolysis products involves oxidative deactivation and rehydration to near-equilibrium moisture content. A sophisticated computerized control system maintains consistent end-product quality and minimizes cost. Within a limited range of sulfur control capability, the LFC process compares favorably in cost to that of flue gas desulfurization. Recently, the preliminary test results on Belchatów lignite from Poland showed the potential of this coal for treatment by LFC technology.

Operations at the demonstration plant are to continue for another two years following an extension of the agreement with DOE (Coal & Synfuels Technology, 1994). The cost-effectiveness of the ENCOAL technology requires that the cost of drying and pyrolysis be significantly less than the added value reflected in the price received for the char and coal liquids. The char, if not used directly as a boiler fuel, could be briquetted and sold as a relatively smokeless fuel.

2.1.3 Agglomeration

The extensive literature on coal agglomeration or aggregation can be only briefly summarized here based on the several recent references given below. Coal agglomeration entails the reconstitution of coal fines, prepared directly or from a process stream, into a lump product such as a pellet or briquette. The distinction between the two terms generally relates to size (the pellet being typically smaller than a briquette), the particle size of the material to be pelleted, and the type of compaction press used. The conditions used for making pellets will also differ from those used for making briquettes, creating cost differences.

The theory and practice of coal agglomeration has been comprehensively described in a recent book entitled *Size Enlargement by Agglomeration* by Wolfgang Pietsch (1991). Several other shorter publications on compacting machines, briquetting parameters and binders, and briquette testing include the following: "Briquetting" (Rhys Jones, 1963); "Briquetting" (Schinzel, 1981); *Elements II—Briquetting and Agglomeration* (Institute for Briquetting and Agglomeration, 1983); "Briquetting Bulk Solids" (Harris, 1992); "Physical Testing of Fuel Briquettes" (Richards, 1990); "Pressure Agglomeration of Fine and Finest Coal Granules: Kind and Composition of Binders" (Schafer, 1987); "A Survey of Briquetting Parameters and Binders" (Young and Kalb, 1994); and "Binders for Producing Smokeless Fuel Briquettes from Coal Chars" (Young and others, 1993b).

Key elements in briquetting of coal or coal chars include coal composition (e.g., volatile matter, carbon, sulfur, ash, and moisture contents), particle size and particle-size distribution, surface chemistry and morphology, porosity and hardness, total moisture, binder type and concentration, mixing conditions and time, briquetting temperature, roll pressure and speed, and curing conditions. Changes in one or more of these elements can have a significant impact on the quality of the briquette—its strength, durability, stability, and reactivity. Briquetting practices for brown coal and lignite generally differ from those for briquetting higher-rank coals. Typically, a binder is not used with brown coal or with some lignites; briquetting temperatures are substantially

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lower (e.g., 50°–70°C) than used in binderless bituminous coal briquetting; and ram extrusion presses are employed.

The required strength of coal briquettes depends on their application and the degree of handling and transportation. Briquettes for home heating or industrial stokers need sufficient strength to keep the generation of fines to a minimum, e.g., >75 psi (> 517 kPa) and preferably 150 psi (1032 kPa). Fines generation should not exceed 5 wt% of the weight of a briquette. Further, the briquettes need to be durable, withstanding wide changes in ambient conditions. Home heating or stoker briquettes should be water-resistant if outside storage is likely and to avoid growing mold in damp conditions. Here, a careful selection of the binder and possibly the use of an additive, e.g., a small quantity (say, 1 wt%) of a bitumen emulsion, is required. Stability of the briquettes towards absorption of water vapor, atmospheric oxidation, and self-heating leading towards spontaneous combustion is another important factor in solid fuel production. Unless judicious precautions are taken, the tendency is greater for self-heating to occur in lower-rank coals. For example, it has been known that brown coal briquettes, if stored incorrectly (such as when a critical stockpile volume in the open atmosphere is exceeded), will begin to self-heat under ambient conditions.

The literature on binders for coal briquettes is voluminous. A few references have already been noted above. For example, see Holley (1983), Schafer (1987), Young and Kalb (1994), Young and others (1993b). An additional reference is Waters (1969). As a guide to the variety of binders used for making smokeless fuels, Table 3 lists those that have been examined at the EERC. The binders are also characterized according to type of interaction (Holley, 1983).

Apart from binders, other additives are sometimes blended into briquettes to improve their quality, e.g., lubricants to assist densification (Holley, 1983), and biomass materials such as agricultural or wood wastes or paper to alleviate smokiness (Richards, 1985). Japanese and Australian researchers have shown that the insertion of biomass into coal briquettes reduces the smokiness of a variety of coals (Maruyama and others, 1985, 1988; Young and others, 1990; Young, 1992). Further work is continuing in Japan and Korea on Indonesian coal and the development of a "bio-type" coal briquette pilot plant to produce an environmentally clean briquette fuel (Choi, 1993; Honda, 1993).

Another form of agglomeration is pelleting/pelletizing. Schinzel (1981) provides a brief description of the fundamentals and conditions for pelletizing fuels along with a few of the processes. A distinguishing feature of pelletizing, apart from the type of press, is the particle size or particle-size distribution, with the top size generally <0.1 mm. As a recent example of a pelleting process for a low-rank coal, the Lignipel process using North Dakota lignite was developed in the early 1980s under DOE support. Here, ground lignite was wetted, and the product was pelleted with 4–8 wt% asphalt emulsion and then evaporatively dried in hot flue gas. Demonstration tests to produce 50 metric tons were undertaken (Baker and others, 1982; Blaustein and Garvin, 1983; Blaustein, 1991).

A significant, and often major, factor in the cost of briquetting or pelleting is the expense of the binder. However, the binder expense should be viewed relative to the added value of the briquette product and its potential market price. Smokeless fuel briquettes can command a premium price because of convenience in handling and storage, cleaner environmental emissions, and ease of burning. In the United States, the cost of the binder typically ranges from US$1 to US$20 per ton of briquettes, depending on the type and amount of binder used (which may be from 2–10 wt%). (It should be pointed out that coal briquettes in the United States are mainly used for the industrial stoker

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TABLE 3

Binders Used for EERC Tableting and Briquetting Tests[1]

Binder Type (classification)	Description	Form
Molasses (M[2], F[3])	Sugar beet processing by-product	Aqueous fluid
Molasses and Ca(OH)$_2$ (C[4])	Binders added independently (forms calcium sucrate)	Powder and aqueous fluid
CaO (C)	Lime	Powder
V1074 (M)	Coal liquefaction heavy distillate	Waxlike solid
Rose Resid (M)	Coal liquefaction residue	Brittle solid
A04 (M)	Pyrolysis-derived anthracene oil	Fluid
PVA 523-S (F)	Polyvinyl alcohol (PVA)	Powder
HFMS, CMS, CSS (M)	Petroleum asphalt emulsions	Fluid
Low-Sulfur Resid (M)	Petroleum distillation residue	Semisolid
High-Sulfur Resid (M)	Petroleum distillation residue	Semisolid
Form Coke Pitch (M)	Partially polymerized coal tar pitch	Brittle solid
MG Scrubber Tar (M)	Mild gasification coal tar	Solid
MG Cyclone Tar (M)	Mild gasification coal tar	Brittle solid
MG Heat Exchanger Tar (M)	Mild gasification coal tar	Solid
MG Distillation Resid (M)	Mild gasification coal tar	Solid
Coal Tar Pitch (M)	Coke-oven coal tar pitch	Brittle solid
E1242E Emulsion (F)	Cross-linked acrylic emulsion	Fluid
AIRFLEX RP-425 (F)	Vinyl acetate–ethylene copolymer	Powder
Flexbond 471 (F)	Acrylic copolymer emulsion	Fluid
Sta-Lok 600 (F)	Pregelatinized potato starch	Powder
Hamaco 196 (F)	Pregelatinized potato starch	Powder
Pearl Starch (F)	Unmodified corn starch	Powder
Potato Starch (F)	Unmodified potato starch	Powder

[1] Holley, 1983.

[2] Matrix-type binder.

[3] Film-type binder.

[4] Chemical-type binder.

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and barbecue markets, and very little is being burned as a residential fuel.) The addition of hydrated lime or limestone to capture sulfur costs about US$3 or US$0.75,
respectively, per ton of briquettes, assuming 3-wt% addition. Capital and operating costs for briquetting in the United States is estimated to be US$4–6/ton of briquettes, depending on the cost of labor, debt financing, power cost, and throughput of the briquetting machine. Thus, briquetting coal or char with a binder and including limestone for sulfur capture typically costs between US$10 and US$16 per ton of briquettes above the cost of the feedstock. To reduce this cost, alternative inexpensive binders can be used, e.g., low-grade wheat starch, which in the United States would cost between US$1 and US$2.50 per ton of briquettes.

2.2 Pyrolysis and Briquetting for Smokeless Fuels

The term smokeless implies a reduction in the tarry (mainly aromatic) volatiles released by fuel briquettes as compared to the parent coal during combustion. Different countries have different limits with respect to tarry volatiles. In the case of the United Kingdom, the approved rate is 5 grams per hour (based on electrostatic precipitator collection) for fuel briquettes classified as smokeless. Rheinbraun briquettes in Germany must meet a similar standard. The Irish standard is 10 grams per hour. While measured differently, the English and Irish test methods give similar results (Crowther and Cowburn, 1995). In the Czech Republic, coals with up to 15 wt% volatile matter content are classified as smokeless, e.g., anthracites having 0–10 wt% volatile matter content (Vúpek Ostrava, 1994). Crowther (1994) has suggested that emissions from smokeless fuel briquettes should be 10% or less of the emissions from burning coal in the same appliance.

Many pyrolysis (or carbonization)/briquetting processes have been developed and commercialized over the last 40 years for the production of smokeless fuels. Pyrolysis is a thermal treatment in either a nonoxidizing, or slightly oxidizing, atmosphere. Coals having a volatile content of up to 15 wt% volatile matter are classified as smokeless fuels, including anthracite and semianthracite. Other coals with higher volatile matter content can be made smokeless through pyrolysis. A comprehensive overview of the various carbonization processes is given by Rhys Jones (1963) and Schinzel (1981). The steps in the processes differ somewhat, depending on the nature of the coal (e.g., rank and hardness), the use or absence of a binder for briquetting prior to coking, and the carbonization temperature and atmosphere. Briquette properties can be greatly affected by oxidative heating (Schinzel, 1981).

The carbonization of brown coal to make char briquettes and low-temperature tar (Schinzel, 1981) has been carried out in the following ways:

- A two-stage process involving carbonization of coal to produce a low-temperature char, briquetting at 10 wt% moisture with a binder (e.g., pitch, coal tar, and bitumen), and then coking of the briquettes.

- A "one-and-one-half-stage" process involving carbonization of part of the coal in the first stage, briquetting the char together with dried coal, and then coking the briquettes.

- A one-stage process in which the coal is briquetted with a binder and then coked.

- A one-stage process whereby the coal is briquetted without binder and then coked.

Briquetting generally occurs between the first and second stages. A fluidized bed is generally used to heat the coal to relatively low temperatures (400°C) in the first stage. If a smokeless fuel is required, the second stage would involve devolatilization of higher temperatures at the lower end of the coking range (500°–1000°C). Germany, the former Yugoslavia, and Hungary in particular have utilized the two-stage process, typically incorporating roll presses (Schinzel, 1981).

Carbonization of bituminous coals at the commercial level is exemplified by the hot briquetting process of the former National Coal Board (NCB), later renamed British Coal. In this process, fine high-volatile coal is carbonized at 400°–440°C in a fluidized bed for 20–40 minutes, and then the hot char is immediately briquetted without a binder. At a mean residence time of about 20 minutes at 425°C, the volatile matter is reduced to 20 wt%. The briquettes are cooled in an inert gas to about 200°C and water-quenched. Two commercial plants for binderless briquettes were built in the UK, one using reciprocating presses (the Homefire Plant) producing 170-g hexagonal briquettes at about a million metric tons/year, and the other using double-roll presses (the Roomheat Plant) producing 75-g briquettes at about 250,000 tons/year (Schinzel, 1981). A third commercial plant, the Phurnacite Plant operated by Coal Products Limited in the UK, is based on a fluidized bed to carbonize blended bituminous coal–anthracite briquettes, incorporating a coal-tar-pitch binder. However, this process has been discontinued, and the briquettes are now made from anthracite dust and molasses–phosphoric acid binder, roll-pressed and heat-treated to about 300°C to produce ovoid briquettes weighing about 40 g with a sulfur content of <1.5 wt% (Vúpek Ostrava, 1994). The briquettes are used for open fires and combustion stoves. As of March 1995, the Homefire Plant, the Roomheat Plant, and the Phurnacite Plant produce about 30,000 metric tons/yr, 200,000 metric tons/yr, and 450,000 metric tons/yr, respectively (Crowther and Cowburn, 1995). Other coal briquetting plants exist in the UK, but these are generally based on anthracite and/or coke breeze and incorporate binders.

Poland, with a tradition of coal briquetting extending for over 100 years, has closed its conventional (noncarbonized) briquetting plants for economic and ecological reasons. At its Institute for Chemical Processing of Coal (ICHPW), development of environmentally friendly solid fuels–coal briquettes with low emissions, i.e., ecological and smokeless fuels—has been subsequently taking place. Ecological fuels can be produced from steam coal fines that have been carbonized at 500°–600°C to yield a char of 8–12 wt% volatile matter. This char is briquetted with ecological binders such as molasses and hydrolyzate of beech (along with phosphoric acid, presumably acting as a catalyst in the binding process) and sulfur-accepting agents. Anthracite or coke breeze can be substituted for the steam coal char, either partially or wholly. Poland produces two types of ecological fuels, one called ECOAL-T and the other ECO-FORMCO, with a total production goal of 180,000 metric tons/year in 1995 (Dreszer and Ściażko, 1994).

Poland has also developed a more effective ecological fuel, designated as smokeless and called ECOCOAL, with insignificant hydrocarbon emissions on combustion. The process is schematically shown in Figure 3. Here, steam coal is pyrolyzed and the char mixed with a preheated caking coal as a binder and hot briquetted at 400°–450°C. Subsequently, the briquettes are conditioned at 400°C for about 4 hours. The process has also been conceptualized for lignite. The steps would involve drying the lignite, carbonizing the lignite, and briquetting the lignite char with an ecological binder.

Table 4 gives a comparison of the properties of the Polish ecological and smokeless fuels. Organic vapor and gaseous inorganic emissions are lower for the smokeless fuel. Based on a typical-

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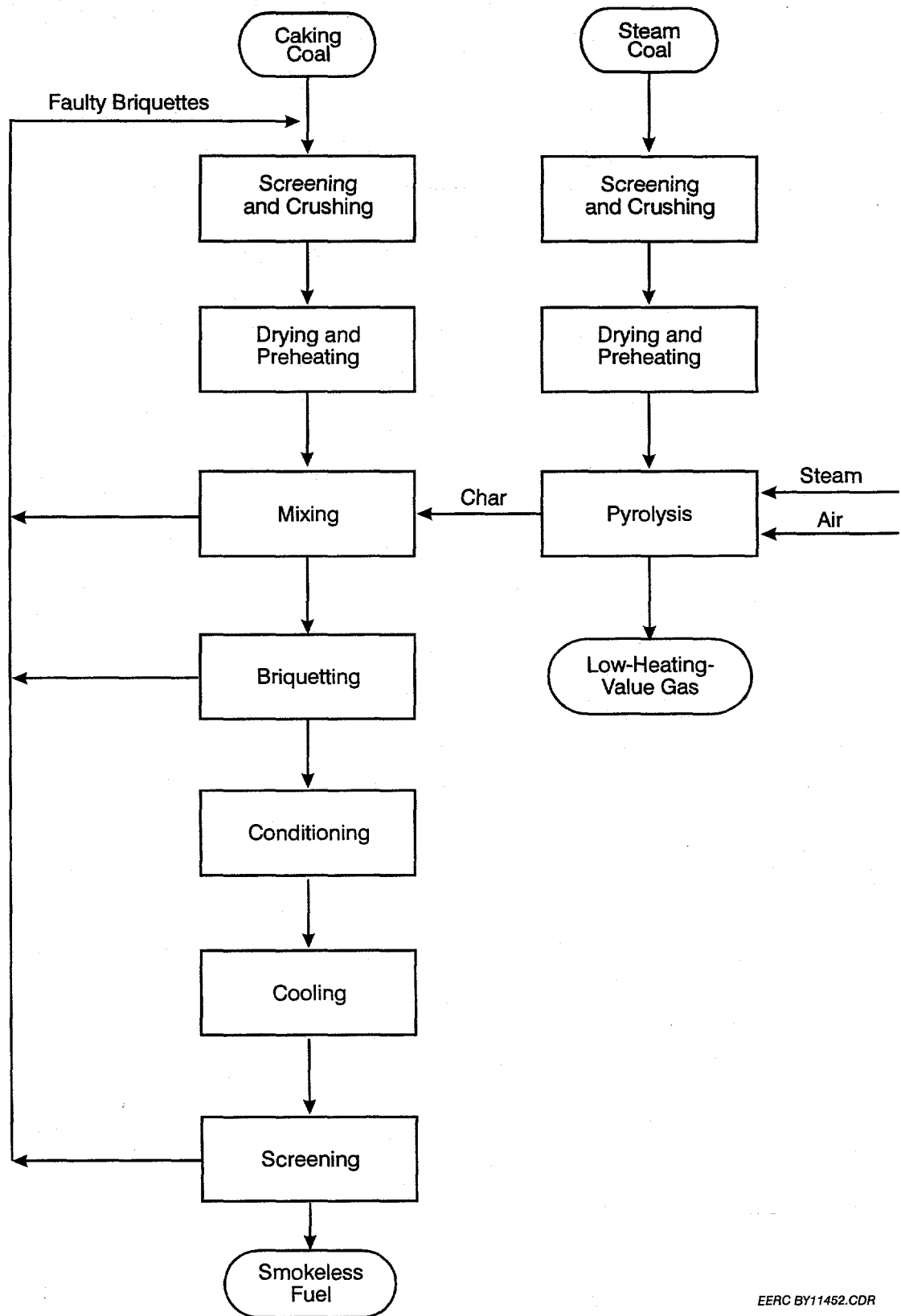
Figure 3. Block diagram of smokeless fuel production process (from Dreszer and Ściażko [1994]).

TABLE 4

Properties of Briquetted Fuels in Comparison to Raw Coal[1]

	Type of Fuel		
	Coal	Ecological Fuel	Smokeless Fuel
Content, %[2]			
Water	3.0–18.0	0.8–10.1	0.6–4.7
Ash	2.2–15.0	9.2–14.8	10.1–12.4
Volatile Matter	29.0–32.8	17.0–18.5	6.4–10.0
Volatile Matter, daf[3]	32.3–40.4	19.6–21.4	7.3–11.5
Sulfur, %[2]			
Total	0.7–1.30	0.60–0.70	0.48–0.56
Ash	0.09–0.49	0.10–0.15	0.35–0.47
Combined	0.18–1.21	0.50–0.55	0.08–0.18
Emission, mg/MJ			
C_nH_m	700–3000	470–700	135–330
CO	2000–5500	2000–4500	2000–4000
SO_2	350–700	130–270	125–185
NO_x	110–180	30–100	20–70
Org. Subst.	480–700	170–330	50–200
Benzo-α-pyrene[4]	400–600	100–275	20–80

[1] From Dreszer and Ściażko, 1994.
[2] As-received unless specified otherwise.
[3] Dry, ash-free.
[4] µg/MJ.

sized Polish apartment (60.6 m²) and 5-kW power demand, it has been estimated that for Poland, smokeless fuels would produce low pollution at a lower cost than using gas or electric heaters. The anticipated market in Poland for ecological and smokeless fuel briquettes is 1–2 million metric tons/year (Dreszer and Ściażko, 1994).

Pyrolysis of lignites and brown coals for fuel applications is not widely practiced. Carbonization plants for lignites and brown coals have been set up and are still operating in Victoria, Australia (brown coal), the former East Germany (lignite), and the former West Germany (brown coal). Pyrolysis temperatures vary, depending on the use of solid product and the desire to maximize the tar yield. At the high end, the pyrolysis temperature is typically around 800°–900°C, whereas at the low end and for maximization of tar yield, temperatures range from 450° to 650°C. A critical factor in carbonizing lignite and brown coal briquettes is to match the operational heating rate as closely as possible to the ideal heating rate to minimize shrinkage stresses and maximize strength (Higgins and Kennedy, 1966; Kennedy, 1971). Basically, two types of retorts have been used commercially: a moving-bed vertical retort in the Victoria plant (Kennedy, 1971, 1977) and the

former East German plants (Allardice and Newell, 1991), and a Salem-Lurgi rotary-hearth furnace in the former West German (Rheinbraun plant) (Scherrer, 1981; Scharf, 1985). At Swharze Pumpe, Germany, Thermobrite and Thermaglow briquettes are made in three sizes from milled lignite using drying, extrusion, and heat treatment above 900°C (Vúpek Ostrava, 1994). From the early 1920s until about 1991, North Dakota lignite was carbonized and briquetted, initially for use as a home heating fuel and later for use as barbecue briquettes. The lignite was carbonized in a Lurgi rotary-hearth furnace. The carbonized lignite was briquetted using a starch binder.

Kren-Consulting (1993) has carried out a prefeasibility study to assess the market for smokeless fuels based on Ostrava hard coal (0.8 wt% sulfur), which is in surplus. United Kingdom carbonizing technology was considered, and a business plan was drawn up with recommendations and privatization proposals for purchasing three coke plants. A market of 0.5 million metric tons processed into smokeless fuel briquettes was projected for 1998–2000. The flat, hexagonal-shaped briquettes (in retail packaging of 5–15 kg) could be burned in existing appliances. The fuel briquettes would have the following properties: 15 wt% ash, 0.8 wt% sulfur, 23–25 MJ/kg, and 1.9–2.2 kg/GJ total emissions. The market price for briquettes made from carbonized Ostrava coal would be 148 Deutche Marks (DM) (~US$114) per metric ton, which compares with anthracite coal briquettes (with high total emissions) costing 105 DM (~US$81) per metric ton. However, the new fuel from Ostrava coal would exceed proposed future emission standards, and anthracite supplies are distant from East Central Europe, resulting in high transportation costs. The smokeless fuel project was put forward as a good alternative to the government-proposed scaling-down program for the Czech coal industry (Kren-Consulting, 1993).

2.3 Sulfur Dioxide Emissions from Residential Solid Heating Fuels

Because of the relatively high sulfur content of Czech brown coal and lignite and their use as a residential fuel, the cities in the Northern Bohemia and Northern Moravia regions experience daily concentrations of air pollution substantially higher than those found in cities located outside of the Czech Republic. For example, per-capita emissions of SO_2 in the Czech Republic in 1992 were over five and one half times (totaling 182 kg) the average value for the European Organization for Economic Cooperation and Development (OECD) countries. At SO_2 levels of 3.0 and 2.4 mg/m^3 as maximum daily concentrations in Litvinov and Osek, respectively, and 3.2 mg/m^3 in Prague in January, 1992, these values are up to 20 times the permitted limits and are comparable to those during the London smog period of 1952. Pollutant levels were exacerbated in the winter of 1992–1993 (IEA, 1994).

Control of SO_2 pollution from residential solid fuels is largely by coal cleaning to remove pyritic sulfur; by pyrolysis (carbonization) to reduce organic sulfur; or by the inclusion of lime-based additives in coal briquette mixtures. Widely tested for various coal briquettes, calcium hydroxide or hydrated lime is now being added to lignite briquettes produced at MIBRAG's Mumsdorf factory in Germany. The treatment consists of adding hydrated lime slurry to the raw brown coal/lignite which contains 55% moisture, mixing intensively, and further adding anthracite dust to compensate for the reduced briquetting tendency of the treated brown coal/lignite, as well as to improve the heat content. The additive amounts are 3.5 wt% lime and 7.0 wt% anthracite. MIBRAG plans to replace the hydrated lime with the waste product from its calcium carbide plant to produce approximately 600,000 tons per year of treated briquettes. These briquettes will meet the government-prescribed Small-Scale Furnace Regulation for emission-effective sulfur content of ≤1 wt% at an additional cost

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of 15 DM (~US$12) per metric ton above that of untreated briquettes. Presently, 120,000 metric tons have been produced. An outline of the MIBRAG process is given in Figure 4. Table 5 provides a comparison of the properties of the treated and untreated briquettes. Except for a 30% gain in ash content (from 9–10 wt% to 13–14 wt%), the treated briquettes exhibit no detrimental effects (Mall, 1994).

2.4 Upgraded Products from Czech Low-Rank Coals Apart from Smokeless Fuels

Several upgraded products from low-rank coals have been investigated at the research and development stage, and a few have been marketed in other countries. These upgraded products include humic acid products, slow-release NPK (nitrogen–phosphorus–potassium) fertilizers, and soil conditioners based on brown coal, drilling muds from brown coal, and activated cokes and carbons from lignite and brown coal. Form coke is another value-added product derived from low-rank coals that has been investigated extensively worldwide and commercialized in the United States.

In the area of humic acid (organic acids with aromatic or aliphatic chains with –OH and –COOH groups with labile hydrogens) research and development, the Research Institute of Inorganic Chemistry in Ústí nad Labem has reported some promising results from work partially funded by Bílina Coal Mines (Kozler and others, 1993). Selected coals, with 13–15 wt% mineral matter containing 75 wt% humic acids, have ion-exchange potential for the separation of some heavy metals (Cd, Pb, Ra) from polluted industrial waters. Humic acids are also useful for improving fertilizers. The results are covered by several Czech patents. The Doly Bílina Coal Company is now manufacturing about 3000 metric tons per year of sodium humate from 5000 metric tons of raw coal.

Another upgraded product from brown coal and lignite is activated carbon, which can be utilized as a sorbent for environmental gaseous or liquid pollutants or for gas storage, e.g., methane. The Rheinbraun company in Germany produces about 200,000 metric tons/year of active coke from lignite for wastewater purification and waste-gas cleanup processing of SO_2, HCl, HF, heavy (trace) metals, and organic compounds such as furans and dioxins (Schieb, 1994). Currently, the coke for waste-gas cleanup is used mainly for waste incineration plants, but it is also being tested in power-generating utilities. The coke is produced in a rotary-hearth furnace and aged with water and air in a cooling system to reduce its reactivity toward oxygen. The product is marketed as a one-way adsorbent in three sizes, namely, < 0.4 mm as pulverized coke, 0–1.5 mm as extra-fine coke, and 1.25–5 mm as fine coke which, depending on size, can be used in a moving bed, filter bed, or circulating fluid bed.

Form coke, in spite of its wide investigation mainly as a substitute for coke-oven coke in blast furnaces and, to a lesser extent, in iron foundry cupolas, has been essentially commercialized in the United States at this stage only for use as a reductant for phosphorus ore. The FMC Corporation (Joseph, 1973; Schinzel, 1981; Eisenhut, 1981) produces the coke from a subbituminous coal at Kemmerer, Wyoming, in approximately 40–50-mm-square, pillow-shaped briquettes. The FMC form coke process involves several steps, including drying and catalyzing (to reduce caking), carbonizing, calcining of the coal, polymerizing the tarry liquids from the carbonizer, briquetting of hot char from calciner with polymerized liquids plus a supplement binder, curing, calcining, and finally cooling of the form coke briquettes (Schinzel, 1981). A rotary furnace, termed the pancake

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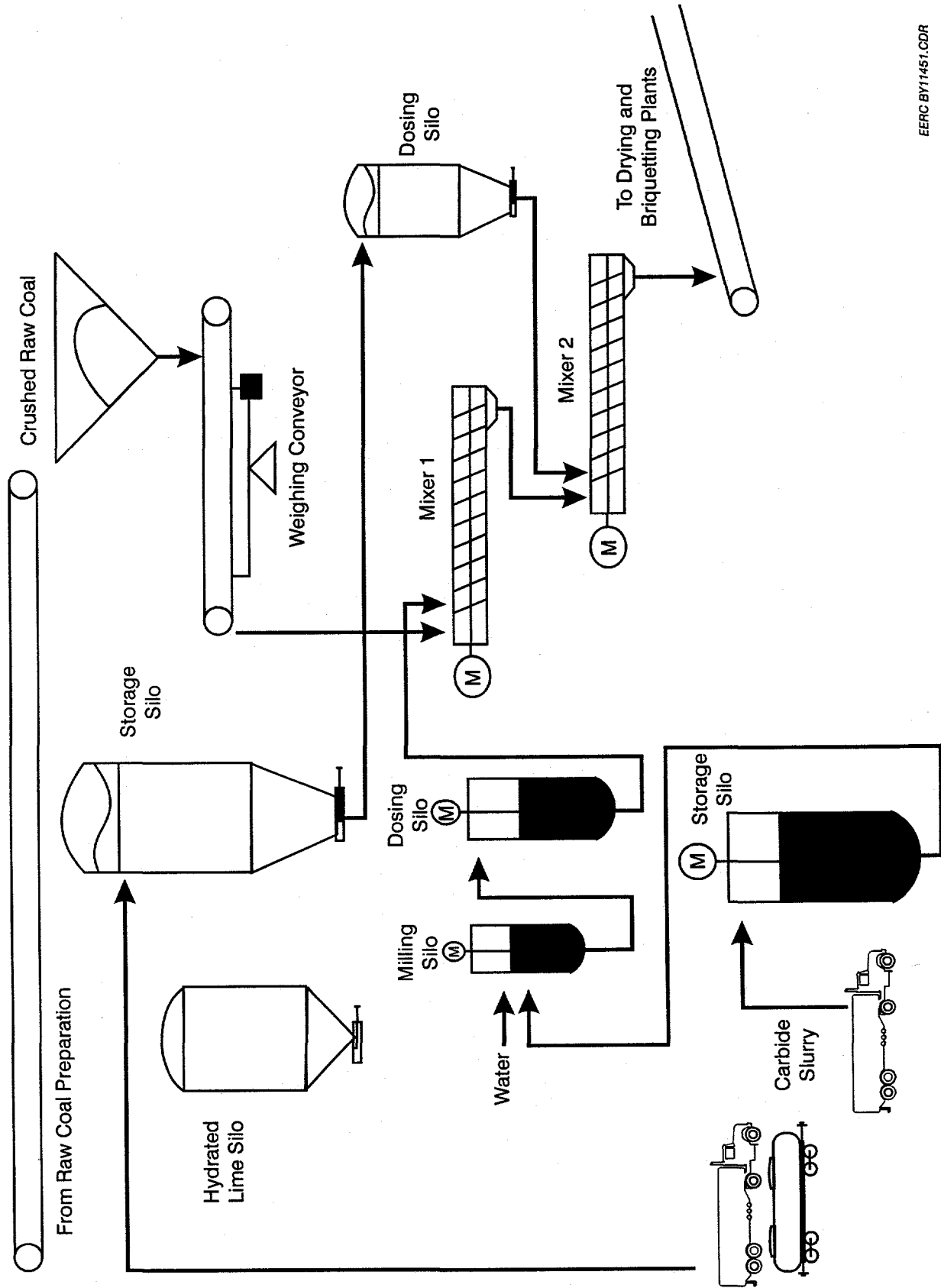
Figure 4.   Flowchart for treated-briquette mixing facility (adapted from Mall, 1994).

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TABLE 5

Quality Comparison Between MIBRAG's Untreated Briquettes and Treated Briquettes[1]

	Untreated Briquettes	Treated Briquettes
Calorific Value, MJ/kg	20.1–20.3	19.8–20.1
Moisture, %	19	17
Ash Content, %	9–10	13–14
Emission-Effective Sulfur Content, %	1.5–1.7	0.8
Compressive Strength, MPa	11–12	11–12

[1] Adapted from Mall (1994).

furnace, has been successfully used by the U.S. Salem Company and in Germany for the production of fuel semicoke. It is considered a promising process for the production of fuel semicoke from Czech coals (Buchtele and Straka, 1994).

The Coal Technology Corporation in the United States has taken a new approach to the production of form coke. It has recently developed, patented, and demonstrated a new continuous process for producing high-quality coke in under 2 hours without the use of coke ovens. The process uses a twin-screw mild gasification reactor at 650°–760°C under inert conditions and ambient pressure. It has been demonstrated at a pilot scale of 10 tons per day. At this stage, only bituminous caking coals have been tested (Wolfe and others, 1995).

3.0 TECHNICAL INVESTIGATION

3.1 Experimental Procedure

3.1.1 Coal Preparation and Analyses

Six coal samples, representing three producing mines in the Czech Republic, were supplied by Dr. Zdenek Krivsky of the North Bohemia Economic Association (Ústí nad Labem, Czech Republic). The coals included Bílina lignite (4 samples), Nástup lignite (1 sample), and Ostrava bituminous coal (1 sample) (see Figure 1). The Bílina lignite samples, designated as 1, 2, 3, and 4 for reference, were obtained from four active pit locations in the Bílina mine.

The six coal samples were each divided into four roughly equal fractions using a laboratory splitter, directing one-half to testing, submitting one-quarter for analyses (proximate and ultimate composition, heating value, forms of sulfur, and high-temperature ash composition), and storing one-quarter in reserve. The analytical procedures and methods are presented in Appendix A. Based on the analytical results, specifically sulfur forms, Bílina lignites 1, 2, and 4, and the Nástup lignite were selected for washability and dry beneficiation tests. The Ostrava bituminous coal was selected for carbonization tests only.

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3.1.2 Coal Beneficiation

The test coals were prepared to specified size ranges for the washability, dry cleaning, and carbonization evaluations. Briefly, the coals were sieved at ¼ in. and the oversize fraction stage crushed with a laboratory jaw-mill until 100% of the coal passed ¼ in. Jaw crushing was used as opposed to impact milling to minimize production of fine coal (−20 mesh) which is not amenable to washability via static float–sink testing. The crushed Bílina and Nástup lignites were bottom-sized at 20 mesh and the yield (recovery) of ¼ in. × 20-mesh and −20-mesh fractions determined; bottom sizing was not performed on the Ostrava bituminous coal. The −20-mesh fractions were subjected to short proximate (moisture, ash, sulfur, and heating value) determinations; the undersize fraction was not tested further. The ash, sulfur, and heating value numbers for the − ¼ in. × 20 mesh were calculated from the raw coal and −20-mesh coal analyses data.

Washability Testing

Washability testing was used to evaluate the cleanability (reduction of ash and sulfur) of select Czech Republic coals using an ideal gravity-based separation process that relies on differences in the specific gravity of lighter coal (carbonaceous matter) and heavier impurities (minerals and pyrite) to effect separation.

The head (¼-in. × 20-mesh) fractions of the Bílina 1, 2, and 3 and Nástup lignites were subjected to bench-scale static float–sink testing. Briefly, the ¼-in. × 20-mesh fraction was weighed and then charged to a separation vessel containing Certigrav true specific gravity solution. The impurities having a specific gravity higher than that of the Certigrav solution were allowed to settle under the influence of gravity (1 G). The suspended clean coal (float) and settled impurities (sink) fractions were recovered, with the resulting sink fraction subjected to further treatment at a higher specific gravity. Consecutive separations were performed at specific gravities of 1.3, 1.4 and 1.6, thus producing three float fractions and one sink fraction. All float and sink fractions were treated to remove trace Certigrav solution, weighed, and subjected to short proximate analysis.

Dry Cleaning

Dry cleaning (for reduction of sulfur and ash) via rare-earth magnetic separation (REMS) was evaluated for select Czech Republic coals. The principle of REMS is that pure coal, being diamagnetic, is repulsed by magnetic forces and pyrite and most coal minerals, being paramagnetic, are attracted by magnetic forces. Consequently, a magnetic field applied to a flowing coal stream can deflect the paramagnetic particles thus allowing them to be separated from the unattracted clean coal particles.

The head (¼-in. × 20-mesh) fractions of the Bílina 4 and Nástup lignites were subjected to REMS at Eriez Magnetics (Erie, Pennsylvania). Briefly, the head fraction was sieved at 6 mesh, producing ¼-in. × 6-mesh and 6-mesh × 20-mesh fractions. The narrower size distributions were deemed necessary to improve the efficacy of separation. The sieved fractions were weighed and then subjected to laboratory-scale REMS using a monolayer feed distribution and 100-ft/min linear belt speed. All magnetic and nonmagnetic fractions were weighed, and the nonmagnetic fraction was subjected to short proximate analysis.

Carbonization

Carbonization was used to produce low-volatile content feedstocks from select raw and beneficiated Czech Republic coals for home heating fuel preparation. Briefly, the ¼-in. × 6-mesh and 6-mesh × 20-mesh Bílina 4 REMS products were combined in proportion to their REMS yields prior to carbonization; for the Ostrava bituminous coal, ¼ in. × 0 in. was carbonized. The test coals were batch pyrolyzed at 650°C in an inert atmosphere using an electrically heated Lindbergh furnace. Replicate carbonizations were performed to provide enough char for tablet preparation. The recovery of char (carbonization product) for each coal was determined, and a representative sample was analyzed for total sulfur content.

3.1.3 Coal and Coal Char Tableting

The Bílina 4 washability product (at 1.3 specific gravity), the carbonized Bílina 4 REMS product, the raw Ostrava bituminous coal and the Ostrava carbonization product were tableted for testing as potential district and/or home heating fuels. Pregelatinized potato starch was used as binder at a concentration of 4 wt% (moisture-free feedstock basis). Pulverized limestone was blended with the Bílina 4 washability and REMS products as a sulfur capture agent.

Briefly, the coals and chars were stage-crushed to 1-mm (18-mesh) top size using a laboratory hammer mill. The crushed feedstocks were wetted with deionized water to assist dispersion of the pregelatinized starch binder and pulverized limestone. The heating fuel feedstocks were densified into 1¼-in.-diameter × ½-in.-thick cylindrical tablets using a hydraulic press. The "green" tablets were allowed to air dry to facilitate binder setting and strength development. Representative air-dried tablets were subjected to proximate, ultimate, heating value, and high-temperature ash analyses, compressive strength testing, and determination of apparent density.

3.1.4 Tablet Combustion

The tablets prepared from the Bílina 4 washability product, the carbonized Bílina 4 REMS product, the raw Ostrava bituminous coal, and the Ostrava carbonization product were subjected to laboratory-scale combustion tests to evaluate potential application as district and/or home heating fuels.

Combustion testing was performed using a DuPont model 951 thermogravimetric analyzer (TGA) with continuous combustion gas analysis performed using a Bomem model B100 Fourier transform infrared (FT-IR) spectrophotometer. Combustion was initiated by charging the fuels to the TGA furnace preheated to approximately 1000°C (1830°F). Air was supplied for combustion at a rate of approximately 200 mL/min. The mass loss was continually monitored until the sample was completely burned. Effluent gas from the TGA was analyzed for SO_2 and NO_x.

3.2 Results and Discussion

3.2.1 Coal Preparation and Analyses

The analysis data (proximate, ultimate, and ash composition, sulfur forms, and heating value) are presented in Table 6 for the Bílina and Nástup lignites, and the Ostrava bituminous coal. Among the six coal samples, the Ostrava bituminous coal had the lowest sulfur content (0.4 wt%) and the

TABLE 6

Analysis Results of Six Czech Republic Sample Coals

Analysis	Bílina 1	Bílina 2	Bílina 3	Bílina 4	Nástup	Ostrava
Coal Type	Lignite	Lignite	Lignite	Lignite	Lignite	Bituminous
Moisture, wt% ar[1]	31.3	29.8	29.7	29.7	38.7	13.6
Proximate, wt% mf[2]						
Volatile Matter	47.2	47.5	48.1	48.3	44.9	31.8
Fixed Carbon	46.4	44.5	45.0	43.9	36.3	62.3
Ash	6.4	8.1	7.0	7.8	18.8	5.9
Ultimate, wt% mf						
Hydrogen	5.3	5.6	5.7	5.8	4.5	4.7
Carbon	68.1	67.1	68.3	67.2	56.7	84.0
Nitrogen	1.1	1.1	1.0	1.0	1.3	1.2
Sulfur	1.02	1.23	0.95	1.14	1.74	0.44
Oxygen	18.1	17.0	17.1	17.1	17.0	3.8
Ash	6.4	8.1	7.0	7.8	18.8	5.9
Sulfur Forms, wt% mf						
Organic	0.68	0.76	0.80	0.67	1.03	0.29
Pyritic	0.30	0.53	0.15	0.44	0.59	0.10
Sulfate	0.02	0.05	0.02	0.08	0.12	0.01
Heating Value, Btu/lb mf	12,100	11,910	12,180	12,120	10,070	14,570
Ash XRFA[3], wt% as oxide						
Silicon	34.7	36.5	37.0	37.1	49.8	43.9
Aluminum	24.4	23.6	24.2	24.5	10.9	29.3
Iron	10.8	11.4	9.0	9.6	14.6	11.5
Titanium	1.1	1.1	1.3	1.3	0.4	1.0
Phosphorus	0.8	0.6	0.5	0.7	0.8	0.1
Calcium	8.4	7.7	8.0	7.8	7.4	3.2
Magnesium	4.2	3.9	4.0	4.2	2.6	3.2
Sodium	1.5	1.4	1.6	1.3	0.6	0.9
Potassium	1.0	1.1	1.1	1.1	0.4	1.6
Sulfur	13.1	12.6	13.3	12.4	12.5	5.3

[1] As received.
[2] Moisture-free.
[3] X-ray fluorescence analysis.

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lowest ash content (5.9 wt%). Not unexpectedly, the Ostrava exhibited characteristics superior to those of the other coals, namely lower moisture content (~14 wt%), higher heating value (~14,600 Btu/lb), and lower oxygen content (~4 wt%). The Bílina and Nástup lignites, by comparison, had sulfur contents ranging from 1.0 to 1.7 wt%, ash contents ranging from 6.4 to 18.8 wt%, moisture contents between 30 and 39 wt%, heating values ranging from 10,000 to 12,000 Btu/lb, and oxygen contents around 17 wt%. The Nástup lignite exhibited the lowest quality among the lignites.

The Bílina lignites were essentially identical in all analysis categories except for forms of sulfur where the pyritic sulfur (as a percentage of total sulfur) ranged from 15 wt% in the Bílina 3 up to 40 wt% in the Bílina 2.

Based on the analysis data, principally ash and total sulfur contents and sulfur forms, coals were selected or passed over as candidates for beneficiation and/or carbonization evaluations. The Ostrava bituminous coal was excluded from beneficiation tests because of its low ash and sulfur contents. Similarly, the Bílina 3 lignite was excluded from beneficiation evaluations because its low pyritic sulfur content (15 wt% of total sulfur) indicated low potential for significant reduction of total sulfur by physical cleaning methods. Conversely, sulfur forms analyses for the Bílina 1, 2, and 4 lignites and the Nástup lignite, indicated higher potential for sulfur reduction.

3.2.2 Coal Beneficiation

The analysis and recovery data for the −20-mesh and −¼-in. × 20-mesh fractions produced during jaw-mill crushing of the Bílina 1, 2, and 4 lignites and the Nástup lignite are presented in Table 7. Jaw-mill crushing and bottom sizing at 20 mesh appeared to be marginally effective for beneficiation, although the coarse coal Btu yields were high (above 80%), as ash was only slightly concentrated in the fines.

Washability Testing

Analysis data and mass and Btu recovery values for the individual float and sink fractions are presented in Table 8 for the Bílina 1, 2, and 4 lignites and the Nástup lignite. Cumulative recovery values and ash and sulfur data for the same coals are presented in Table 9.

Washability test results indicated that the Bílina 1, 2, and 4 lignites, sized to ¼ in. × 20 mesh, were amenable to substantial ash and sulfur reductions by wet gravity-based separation methods, and each sample exhibited similar properties at each separation specific gravity tested. At Btu yields between 90 and 93%, clean coal fractions were produced with ash and sulfur contents averaging 42 and 36 wt% lower, respectively, than values for the head (float−sink feed) fractions. The average ash reduction is more substantial when compared to the average raw coal ash content. Further, because of a modest improvement in the heating value, the average theoretical sulfur emission value, on a Btu basis, is 39% lower for the clean Bílina lignite relative to the raw coal.

Analysis of the ash from the 1.3 specific gravity float product indicated that silicon-bearing minerals were reduced while the typically organically associated species, such as calcium, magnesium, and sodium, were concentrated. The calcium concentration was doubled as a consequence of washability testing.

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TABLE 7

Analysis Results of Coals Prepared for Washability Evaluation
(moisture-free)

Analysis	Bílina 1	Bílina 2	Bílina 4	Nástup Lignite
−20-mesh coal				
Ash, wt%	11.9	11.5	12.2	22.9
Sulfur, wt%	1.14	1.18	1.18	2.48
Heating Value, Btu/lb	11,360	11,400	11,290	9,320
Mass Yield, wt%	15.1	19.7	16.9	22.7
Btu Yield, %	14.2	18.8	15.8	21.0
−¼ in. × 20-mesh coal				
Ash, wt%	5.4	7.2	6.9	17.6
Sulfur, wt%	1.00	1.24	1.13	1.52
Heating Value, Btu/lb	12,230	12,030	12,290	10,290
Mass Yield, wt%	84.9	80.3	83.1	77.3
Btu Yield, %	85.8	81.2	84.2	79.0

TABLE 8

Fractional Washability Results for Czech Republic Coals (moisture-free)

Coal Sample	Coal Yield, wt%	Btu Yield, %	Heating Value, Btu/lb	Ash, wt%	Sulfur, wt%
Bílina 1					
1.3 Float	89.6	92.7	12,740	4.0	0.72
1.4 Float	6.1	5.3	10,710	16.8	2.23
1.4 Sink	4.3	2.0	5,670	50.5	5.21
Bílina 2					
1.3 Float	86.8	90.2	12,630	4.1	0.71
1.4 Float	9.0	7.9	10,690	16.7	1.78
1.4 Sink	4.2	1.9	5,620	49.9	5.57
Bílina 4					
1.3 Float	89.2	92.3	12,580	3.9	0.69
1.4 Float	7.0	6.1	10,620	18.5	1.62
1.4 Sink	3.8	1.6	4,940	54.0	7.86
Nástup Lignite					
1.3 Float	73.6	88.8	11,280	9.2	1.79
1.4 Float	3.6	3.6	9,480	22.4	3.15
1.6 Float	2.8	2.1	7,020	40.3	2.48
1.6 Sink	20.0	5.5	2,560	74.4	1.49

TABLE 9

Cumulative Washability Results for Czech Republic Coals (moisture-free)

Coal Sample	Coal Yield, wt%	Btu Yield, %	Heating Value, Btu/lb	Ash, wt%	Sulfur, wt%
Bílina 1					
1.3 Float	89.6	92.7	12,740	4.0	0.72
1.4 Float	95.7	98.0	12,610	4.8	0.81
Total	100.0	100.0	12,310	6.8	1.00
Bílina 2					
1.3 Float	86.8	90.2	12,630	4.1	0.71
1.4 Float	95.9	98.1	12,450	5.3	0.81
Total	100.0	100.0	12,160	7.1	1.01
Bílina 4					
1.3 Float	89.2	92.3	12,580	3.9	0.69
1.4 Float	96.2	98.4	12,440	5.0	0.75
Total	100.0	100.0	12,150	6.8	1.03
Nástup Lignite					
1.3 Float	73.6	88.8	11,280	9.2	1.79
1.4 Float	77.2	92.4	11,200	9.8	1.85
1.6 Float	80.0	94.5	11,050	10.8	1.87
Total	100.0	100.0	9,350	23.6	1.80

Washability test results indicated that the Nástup lignite, sized to ¼ in. × 20 mesh, was amenable to substantial ash reduction by wet gravity-based separation methods. At a Btu recovery of almost 90%, an ash reduction of approximately 50 wt% was attained. Wet physical cleaning for sulfur reduction was not effective at ¼ in. × 20 mesh with the Nástup lignite. However, because of an improvement in heating value, the theoretical sulfur emission of the clean coal fraction at 90% Btu yield was approximately 6% lower than the value for the raw coal.

Dry Cleaning

Analysis data and mass recovery values for the REMS clean coal (diamagnetic) fractions for the Bílina 4 and Nástup lignites are presented in Table 10. Preliminary results indicated that the Bílina 4 was amenable to ash and sulfur reductions via REMS. At a coal recovery of almost 95 wt%, the sulfur content of the clean coal was ~27 wt% lower than the values for the head fraction and raw coal. Further, the heating value and ash and sulfur contents for the combined fraction closely paralleled washability data for the Bílina 4 lignite. Optimization with this coal may produce REMS yield and coal quality results equivalent to those from wet gravity-based methods.

TABLE 10

Magnetic Beneficiation Results for Czech Republic Coals

Coal Sample	Coal Yield, wt%	Heating Value, Btu/lb mf	Ash, wt% mf	Sulfur, wt% mf
Bílina 4				
−¼ in. × 6 mesh	97.8	12,300	5.9	0.86
−6 mesh × 20 mesh	90.3	12,410	5.2	0.73
Combined	94.5	12,350	5.6	0.80
Nástup				
−¼ in. × 6 mesh	93.1	9270	24.5	1.70
−6 mesh × 20 mesh	80.4	9860	19.5	1.76
Combined	87.7	9500	22.6	1.72

Preliminary results with the Nástup lignite indicate poor performance using REMS at a size of ¼ in. × 20 mesh, however, insufficient testing was done to completely assess the suitability of REMS for treating Nástup lignite.

Carbonization

The yields of char produced by carbonization of the Bílina 4 REMS product and the Ostrava bituminous coal were determined to be approximately 39 and 75 wt%, respectively. The sulfur contents of the char products were lower than the carbonization feedstocks, indicating that sulfur was liberated in gaseous form. The sulfur content of the Bílina 4 REMS product char was 0.62 wt% mf, which compared to 0.80 wt% mf for the Bílina 4 REMS product and 1.1 wt% mf for the Bílina 4 raw coal. The char product contained only 43% of the precarbonization feed coal sulfur. The sulfur contents of the Ostrava bituminous coal and coal char were 0.44 wt% and 0.40 wt%, respectively, indicating that approximately 20 wt% of the sulfur was liberated by carbonization.

3.2.3 Coal and Coal Char Tableting

Analysis data for the tablet products prepared from the Ostrava bituminous coal, the Bílina 4 washability product, the carbonized Ostrava bituminous coal, and the Bílina 4 REMS char product, are presented in Table 11. Preliminary results show that the sulfur contents of all four tablet fuels fall well under new quality standards in the Czech Republic (see Table 2). These standards regulate the maximum sulfur content for lignite and hard coals, briquettes, and liquid fuels utilized in household, business, or energy installations that are not equipped with sulfur emission abatement devices.

The sulfur contents for tablet fuels prepared from the Ostrava bituminous coal, the Bílina 4 washability product, the carbonized Ostrava bituminous coal, and the Bílina 4 REMS char product were determined to be 0.13, 0.25, 0.14, and 0.23 g/MJ (lower heating value), respectively. These

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TABLE 11

Analysis Results of Four Tableted Heating Fuels

Coal: Treatment:	Ostrava None	Bílina 4 Float–Sink	Ostrava Carboniz.	Bílina 4 REMS/Carboniz.
Moisture, wt %	1.0	6.2	1.5	2.2
Proximate, wt % mf[1]				
Volatile Matter	29.9	52.7	11.8	15.5
Fixed Carbon	64.3	40.3	80.8	72.9
Ash	5.8	7.0	7.4	11.6
Ultimate, wt % mf				
Hydrogen	4.8	4.8	1.7	1.5
Carbon	83.0	66.0	85.7	79.4
Nitrogen	1.2	0.9	1.3	1.2
Sulfur	0.43	0.61	0.38	0.56
Oxygen	4.8	20.7	3.5	5.8
Ash	5.8	7.0	7.4	11.6
Heating Value, Btu/lb mf	14,320	11,500	13,320	12,390
Ash XRFA, wt % as oxide				
Silicon	43.9	18.9	43.1	30.7
Aluminum	29.3	13.7	26.2	21.4
Iron	11.5	1.9	12.0	4.2
Titanium	1.0	0.6	1.0	1.1
Phosphorus	0.1	0.4	0.1	0.5
Calcium	3.2	41.5	5.6	26.2
Magnesium	3.2	4.4	3.4	3.2
Sodium	0.9	1.2	1.5	1.6
Potassium	1.6	0.6	1.4	0.8
Sulfur	5.3	16.8	5.7	10.4
Apparent Density, lb/ft³	69	53	65	64
Compressive Strength, lb/in²	290	80	220	90

[1]Moisture-free.

values compare to an allowable maximum of 0.60 g/MJ for indigenous briquettes prepared for residential heat or steam raising, thus qualifying all four fuels for application as district heating fuel. Further, adding calcium carbonate during tablet preparation produced molar calcium-to- sulfur ratios of 3.1 in the Bílina 4 washability product fuel and 2.7 in the carbonized Bílina 4 REMS product fuel, thus theoretically reducing the emission-effective sulfur content below 0.25 and 0.23 g/MJ, respectively.

The weight percentages of volatile matter in the tablet fuels prepared from the carbonized Ostrava bituminous coal and the carbonized Bílina 4 REMS product were determined to be 11.8 and 15.5, respectively. Volatile matter content of the carbonized Ostrava bituminous coal easily met the arbitrary, but long-established, limit of 15 wt% (see Section 2.2) to qualify as a smokeless fuel for home heating application. Although marginally above this value, the volatile matter content of the Bílina 4 REMS char product fuel can easily be brought into specification as a home heating fuel through slight modifications to the carbonization conditions (temperature, residence time).

Positive attributes of the tableted fuels relative to the parent coals include lower as-fired moisture contents and higher as-fired heating values, which would reduce fuel handling whether it be in the size of live or active storage bins or in the size and power requirements for conveyors, augers, and feeding devices. Similarly, for domestic or home heating applications, the time between fuel charges to the heating appliance would be reduced.

Preliminary strength analysis indicates that the tablets have good strength, especially for the Ostrava bituminous coal-based fuels. Further, the chemical homogeneity and size uniformity of tablet (or aggregated) fuels would lend to enhanced predictability in operation of heat or steam-raising stoves and boilers.

3.2.4 Tablet Combustion

The TGA combustion (mass loss) profile for the four fuels are presented in Figure 5, and the combustion gas FT-IR profiles for carbon dioxide, carbon monoxide, and hydrocarbons (HC) are presented in Figures 6, 7, and 8, respectively. The tablet fuels, designated 1, 2, 3 and 4, in the figures are respectively, raw Ostrava bituminous, float-sink cleaned Bilina No. 4 lignite, carbonized Ostrava bituminous coal, and REMS cleaned/carbonized Bilina No. 4 lignite. Preliminary TGA combustion tests indicated that there were no difficulties burning the four fuels. All four fuels exhibited an initial devolatilization stage during which the mass loss agreed quite well with the mass percentage of moisture plus volatile matter, as determined by proximate analysis. The rate of devolatilization was more rapid with the Bílina 4 lignite float–sink product and the raw Ostrava bituminous coal. The Bílina 4 lignite float–sink tablet fuel exhibited the fastest burnout rate after devolatilization, indicating a highly reactive char relative to the other fuels after the devolatilization stage. The carbonized Ostrava bituminous coal and carbonized Bílina 4 REMS product exhibited similar reactivity, with burnout rates slightly less than that of the raw Ostrava coal.

Preliminary results of gas analysis by FT-IR indicate that SO_2 and NO_x were below detectable limits of 10 and 50 ppm, respectively. Dilution caused by the high combustion air rate, inhibited quantitation of SO_2 and NO_x. FT-IR analysis also indicated that the carbonized fuel tablets had the lowest HC emissions relative to the noncarbonized fuel tablets. The initial HC emission levels were nearly proportional to the volatile matter content of each as-fired fuel.
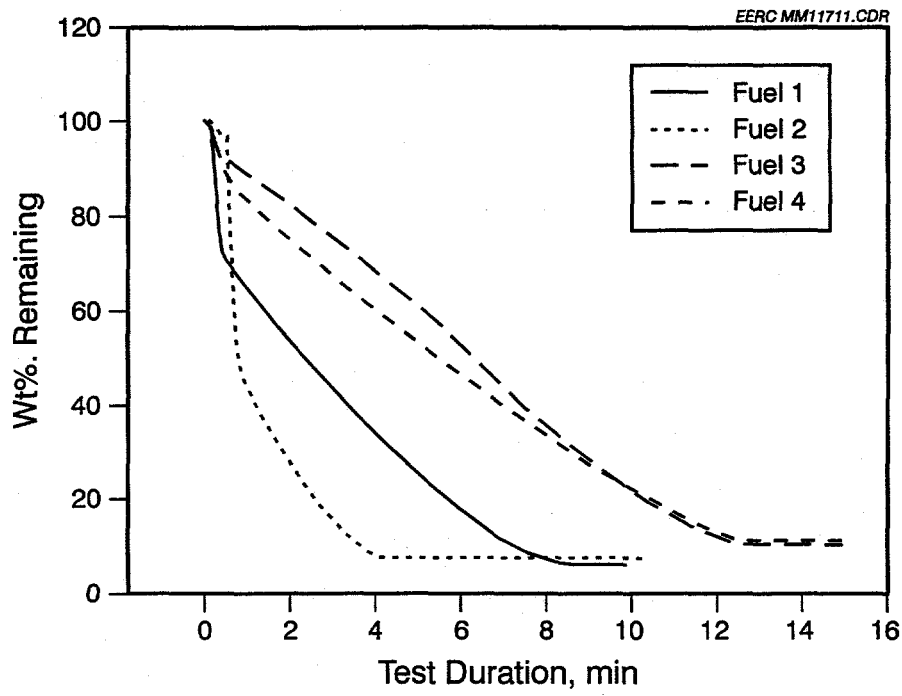
30

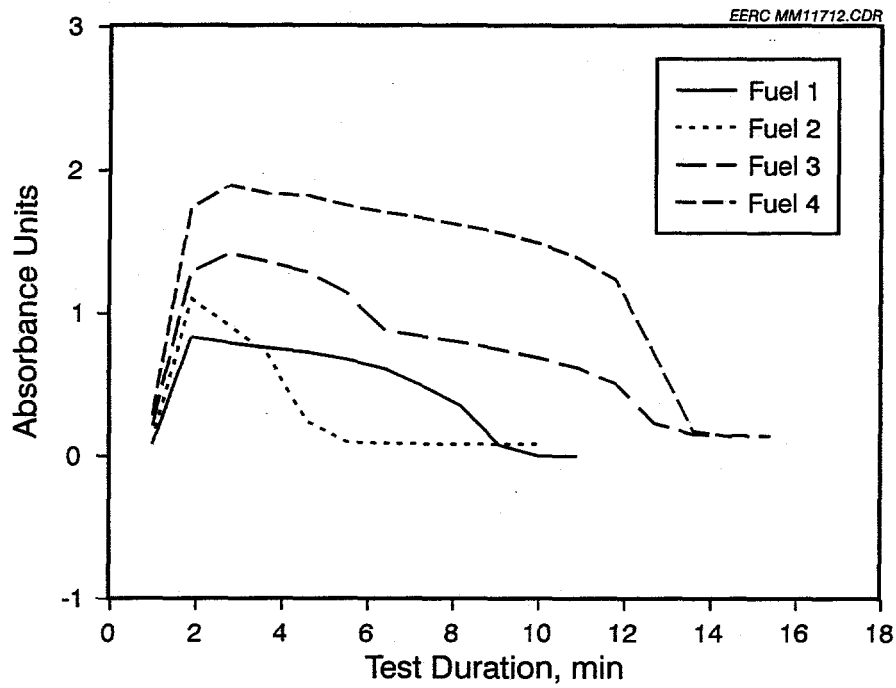Figure 5. TGA combustion profile for tableted Czech Republic coals.

Figure 6. FT-IR CO_2 profile for combustion testing of tableted Czech Republic coals.
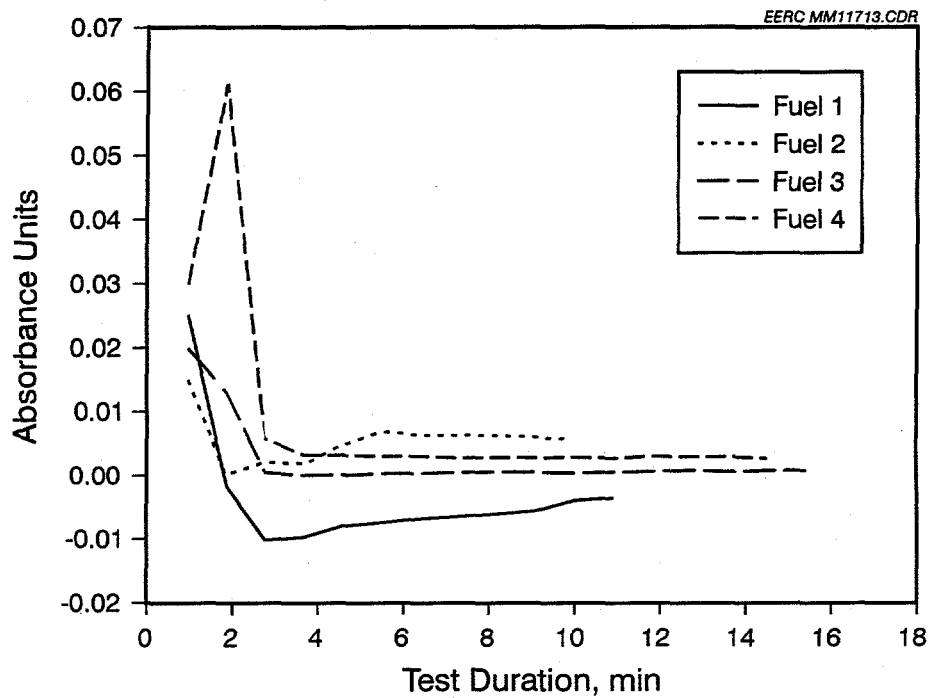
31

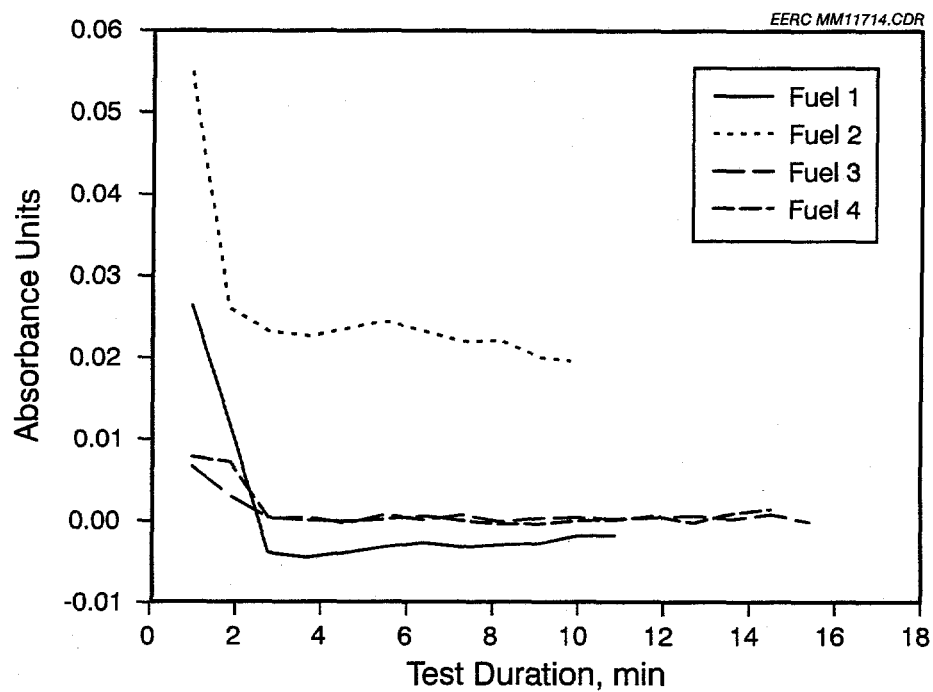Figure 7. FT-IR CO profile for combustion testing of tableted Czech Republic coals.

Figure 8. FT-IR HC profile for combustion testing of tableted Czech Republic coals.

4.0 CONCLUDING REMARKS AND RECOMMENDATIONS

4.1 Summary of Report

Coal mining output in the Czech Republic has significantly decreased over the last 6 years as the demand has declined owing to environmental constraints, government regulations, and available alternative energy sources such as gas and oil, as well as coal imports. By the year 2000, black coal production is estimated to be 14.5 million tons per year and brown coal to be 50 million tons per year. As of 1993, a little over 18 million tons of black coal and almost 67 million tons of brown coal were mined (Ministry of Industry and Trade, 1994). For residential use, the black coal, with its relatively low sulfur content (0.5–0.6 wt%) would need to be pyrolyzed, preferably under mild carbonization conditions, and briquetted to reduce the tarry vapors and soot formation during combustion of the briquettes in a home heating appliance. Alternatively, the coal could be mixed with biomass (e.g., agricultural waste products) and briquetted to help reduce the smokiness of the coal (Maruyama and others, 1985, 1988; Young, 1992; and Honda, 1993). In addition, the black coal would need to be cleaned because of its relatively high ash content (14–31 wt%). A recent report on North Moravian coal indicates that the cost of low-temperature carbonization is about 40% of the cost of coking (Krivsky, 1994a). This carbonization process would also provide chemical products for the chemical industry. As an example of a coal gasification process, the North Dakota Gasification plant at Beulah, ND, provides selected chemicals for the U.S. chemical industry. A new clean coal technology demonstrated by SGI, described above, has the potential of delivering chemical products.

The production of clean energy from brown coal/lignite is a major concern for the Czech Republic given this large fuel resource with not only high moisture but also relatively high sulfur content. An energy–chemical production complex based on brown coal is one option (Ministry of Trade and Industry, 1994). However, implementation of this concept will require a huge investment and substantial time. In the meantime, smaller-scale and simpler processes need to be examined, such as mild gasification, briquetting of the char, and briquetting of the coal with binders and other additives to reduce emissions and improve heating values (i.e., efficiency). An example is the new German briquetting process that blends lignite with hydrated lime and anthracite fines, dries the mixture to 17–19 wt% moisture, and briquettes with a stamp press (Mall, 1994).

Alternative approaches to upgrading brown coal include new pyrolysis methods, thermal treatment (K-Fuels C Process), and Gravimelt, each of which require a briquetting step to produce a residential heating fuel that is stable and convenient to handle. However, the cost of the processes will be critical, particularly in the case of the Gravimelt process and possibly the K-Fuels process, depending on plant module size. Costs of production can be spread if the adopted process produces other high-value products or can be integrated into a larger energy complex.

Regarding black coal utilization in the Czech Republic, various reports (e.g., Ministry of Trade and Industry, 1994; Kren-Consulting, 1993) have looked at the marketing options. A market is perceived for North Moravian black coal as a smokeless fuel. However, some 150,000 metric tons per year in 1995 would be the minimum projection that would be needed to make this approach viable (Kren-Consulting, 1993).

This report has dealt mainly with the fuel side of residential heating. The combustion appliance is also an important factor in determining the amount and type of emissions released to the

environment. The design and maintenance of the heating appliance will significantly affect the kind and level of emissions as well as the behavior of the type of fuel burned. Hence, the quality of the smokeless fuel, as well as the design and operation of the home appliance, should all be evaluated as a whole in determining overall improvements in environmental emissions in Ústí nad Labem or elsewhere.

Apart from the production of smokeless fuels, other value-added products can be generated from brown coals and lignite. Of the several products briefly discussed, activated carbons and form coke probably offer the most promise at this time because of the increasing thrust of environmental requirements and regulations. Different and competitive technologies exist, some presently better placed for commercialization than others, that enable serious investigation of nonfuel carbon product manufacturing. Matching the coal with the technology, determining the level of risk, and obtaining funding are the major challenges facing the Czech Republic government and corporate decision makers.

Home and district heating fuels can be prepared from lignite resources located within North Bohemia and close to the city of Ústí nad Labem using conventional beneficiation and carbonization technologies. Further, comparable quality fuels for similar applications can be produced from a high-quality bituminous coal.

Beneficiation technologies employing the float–sink principal can effectively reduce the sulfur and ash content of Bílina lignite, a coal of significant reserve in the North Bohemia region of the Czech Republic. Further, dry beneficiation via REMS can produce significant sulfur reductions; optimization of this process with the Bílina lignite may allow yields and sulfur reductions to approach values for wet gravity based separations. Nástup lignite, also mined in the North Bohemia region, is not amenable to sulfur reduction at ¼ in. × 20 mesh via conventional wet gravity-based methods or REMS.

Based on quality standards established in the Czech Republic for the allowable maximum sulfur content (g/MJ basis) of indigenous briquettes, tablet fuels prepared from a raw Ostrava bituminous coal and a float–sink-cleaned Bílina lignite would qualify as fuels for steam raising in commercial, industrial, and energy installation applications. Further, tablet fuels prepared from carbonized Ostrava bituminous coal and carbonized Bílina REMS product would qualify as smokeless fuels for home heating applications by virtue of their sulfur and volatile matter contents. Both Bílina lignite-derived fuels have potential emission-effective sulfur contents comparable to the Ostrava fuels because of a sulfur capture agent (calcium carbonate) added during tablet preparation and the inherent calcium in the case of Bilina lignite.

4.2 Recommendations

The opportunity exists to reduce the sulfur dioxide emissions in Northern Bohemia and, specifically, in and around the city of Ústí nad Labem by increasing the utilization of upgraded indigenous coals such as Bílina lignite and Ostrava bituminous coal. A program for the coals should include a detailed characterization of chemical and physical properties, pilot-scale preparation of briquetted fuel by conventional (with binders) and advanced (binderless) briquetting methods, pilot-scale combustion testing in stoker combustor and residential heating appliances, and an economic evaluation comparing upgraded fuels to other methods for emissions reduction. Further, the lignite

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should be subjected to pilot-scale assessment of cleanability by conventional wet physical cleaning and dry magnetic separation techniques.

5.0 ACKNOWLEDGMENTS

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APPENDIX A
Procedures and Methods for Coal and Coal Product Analyses

TABLE A-1

Procedures and Methods for Coal and Coal Product Analyses

Analysis	Method	Equipment
Moisture, Volatile Matter, Ash	ASTM D5192-91 (Proximate Analysis of the Analysis Sample of Coal and Coke by Instrumental Procedures)	LECO 501 thermogravimetric analyzer
Carbon–Hydrogen–Nitrogen	ASTM D5373-93 (Instrumental Analysis of Carbon, Hydrogen and Nitrogen)	LECO CHN-600 carbon–hydrogen–nitrogen determinator
Sulfur	ASTM D5016-89 (Sulfur in the Analysis of Coal and Coke Using High Temperature Tube Furnace with IR Cell Adsorption)	LECO SC-132 sulfur determinator
Heating Value	ASTM D2015-93 (Gross Calorific Value of Solid Fuel by the Adiabatic Bomb Calorimeter)	LECO adiabatic calorimeter
Ash Analysis	ASTM D4326-92 (Major and Minor Elements in Coal and Coke Ash by X-Ray Fluorescence)	FISONS 770 x-ray fluorescence analyzer
Ash Fusion	ASTM D1857-87 (Fusibility of Coal and Coke Ash)	