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# LOW-RANK COAL: Its Present and Future Role in the United States<sup>1</sup>

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# OCCURRENCE OF LIGNITE AND SUBBITUMINOUS COAL

Low-rank coals—lignite and subbituminous coal—are those that have been subjected to the least metamorphic change during the coal-forming process. As such, they retain greater fractions of moisture and volatile matter and contain less fixed carbon than the high-rank coals—bituminous and anthracite. The primary measure used to classify the lower ranks of coal is the heating value. Lignite is defined by the American Society for Testing and Materials (standard D-388) as coal with a heating value less than 8300 Btu/lb, on a moist, mineral-matter-free basis (m,mmf). Subbituminous coal ranges in heating value from 8300 to 11,500 Btu/lb (m,mmf). Bituminous coal and anthracite range from 10,500 to over 15,000 Btu/lb (m,mmf).

Low-rank coals represent a major but largely untapped energy resource in the United States. Very extensive deposits of lignite and subbituminous coal exist in the western states, the Gulf Coast, and Alaska (Figure 1). Major deposits of low-rank coal are also found in many other countries, most

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notably the Soviet Union, Australia, Canada, and the central and eastern European nations. The quantities of US lignite and subbituminous coal as shown in Table 1 (1, 2) are vast. Over one trillion tons of identified resources have been located or inferred by geologists. Over 130 billion tons occur close enough to the surface to be economically recovered by stripping. Another 108 billion tons of subbituminous coal are recoverable by underground mining.

When these reserve base quantities are multiplied by appropriate recovery factors of about 50% for underground mining and 90% for surface mining, and by average heating values, a comparison of recoverable reserves on a Btu basis can be made (Figure 2; 1). As indicated, the recoverable reserves of low-rank coals are comparable to the reserves of high-rank coal; these in turn are vastly larger than our remaining reserves of oil and gas.



Figure 1 Low-rank coal regions of the United States.

		=		
	Lig	Lignite		
Region	Identified resources <sup>b</sup>	Strippable reserve base <sup>c</sup>	-	
Fort Union Gulf Denver Others Total	465.3	-		
	463.3 68.3	26.3 35.0		
	10.0	2.9		
	0.2	na		
	543.8	64.2		
	Subbituminous coal			
Region	Identified resources <sup>b</sup>	Strippable reserve base <sup>c</sup>	Underground reserve base	
Powder River	238.1	57.5	97.2	
San Juan	50.6	5.5	0.9	
Alaska	110.2	0.5	4.8	
Others	147.2	4.4	4.8	
Total	546.1	67.9	107.7	

#### Table 1 US low-rank coal resources and reserve base (billion short tons)<sup>a</sup>

\* Strippable Gulf reserve from (2). All other values from (1).

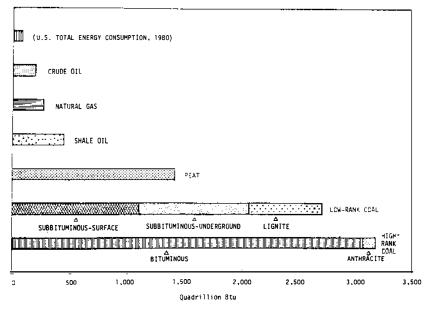
<sup>b</sup> Identified resources include demonstrated (measured and indicated) and inferred resources greater than 2.5 ft thick to an overburden depth of 3000 ft.

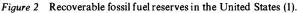
<sup>c</sup>The reserve base includes coal that is technically and economically mineable at this time. Criteria for minimum seam thickness and maximum overburden thickness vary from state to state.

#### MARKETS

During the past 60 years, the markets for coal in the United States have shifted substantially. Total coal consumption remained generally between 400 and 600 million tons/year until 1975, when it began a rapid climb to the current level of over 800 million tons/year. Railroad and residential/commercial direct use of coal—two very large markets at one time—had all but disappeared by the 1960s. Industrial use has been relatively constant for the last 30 years, with about half being coking coal for metallurgical production—a market reserved for high-quality bituminous coal.

Electric utility use of coal has grown steadily for the last 40 years. Lowrank coals played a minor role in that market until the late 1960s, primarily because the low-rank coal resources are remote from industrial centers, and cheap oil and gas were abundantly available. Beginning in the late 1960s, electric utility use of low-rank coals began to grow very rapidly, accounting for almost all of the nearly 240 million tons produced in 1982 (Figure 3; 1).





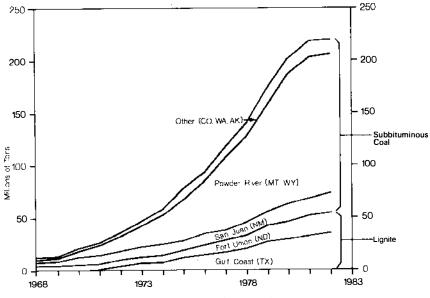


Figure 3 Low-rank coal production by region, 1968–1982 (1).

This rapid growth happened because several technological and regulatory developments changed the relative economics of low-rank coal use by the electric utility industry :

- 1. Very large pulverized coal-fired boilers, as well as extremely large stripmining machines, were adapted for low-rank coals. These allowed the industry to realize the economies of scale required to make low-rank coal use competitive.
- 2. Electric power transmission systems and regional reliability councils connected vast regions of the United States, which became interdependent in terms of power supply. Thus, mine-mouth power plants in the low-rank coal regions could penetrate large markets for the first time.
- 3. Limits were placed on emissions of  $SO_2$  from coal-fired power plants in the early 1970s. This created a huge demand in the midwest for low-sulfur western coal, despite the high transport costs per Btu of this low-rank coal.
- 4. The price and long-term supply uncertainty of OPEC oil and domestic gas rose rapidly in the early 1970s. This created a strong incentive to switch from these fuels to coal for power generation, particularly in Texas.

In this dramatic growth process (generally 15%-20% per year for lowrank coal overall), lignite has remained a regional fuel, used almost exclusively within the borders of the Fort Union and Gulflignite regions. In contrast, subbituminous coal has penetrated markets far from the coal mines. Only a small fraction of the subbituminous coal produced in the Powder River Region is used in power plants in Montana and Wyoming. The remainder is shipped by unit train to power plants in at least 20 other states. Nearly all of the San Juan Region subbituminous coal production is burned by utilities in New Mexico and Arizona.

Technological progress has been closely linked to penetration of the electric utility market by Fort Union Region lignite. In the 50-year evolution of the technology, phasing out of small stoker units, initial development of pulverized coal- and cyclone-fired boilers in the 100–250 MW size range, and most recently the development of very large 500-MW units have been primary events associated with the very rapid growth of this industry. The interconnection of major electricity transmission systems, mentioned above, is well illustrated: in 1976 more than half the power generated in North Dakota was transmitted out-of-state through regional power pool grids. Plants built or planned since 1976 send about three quarters of their power out of North Dakota. Lignite costs at mine-mouth plants as low as \$0.08 per million Btu (MMBtu) reported by Minnkota

Power Cooperative in 1967, and \$0.50 to \$0.75 per MMBtu in 1983, reflect the low mining costs achieved with high-capacity mining equipment.

In the Gulf lignite region, deposits extend through at least five states— Texas, Louisiana, Arkansas, Mississippi, and Alabama. However, significant utilization of this resource has occurred only in Texas to date. The Texas Utilities Company pioneered a 40-MW pulverized-lignite plant in the 1920s, and by 1972 operated three large units with a combined capacity of 1500 MW. During the 1974–1983 decade, an additional nine lignite-fired units were installed by several utilities, increasing the total capacity to 7005 MW. In the same decade, approximately 23 subbituminous coalfired power units were installed in Texas, Louisiana, and Arkansas, with a combined capacity of 12,900 MW.

Low-rank coals (roughly 45% Texas lignite and 55% western subbituminous coal) will generate approximately 40% of electricity in Texas in 1985, compared to 10% in 1975. The driving force for this major shift in the State's power-supply sources has been economics. In 1976 when the cost of intrastate gas had increased to about \$2.00 per MMBtu, Texas Utilities considered its lignite cost to be \$0.29 per MMBtu. The primary reason the Powder River Basin coals could also compete in Texas, despite the high costs of transportation, was that until 1979 these coals could be used in plants without scrubbers, whereas Texas lignite could not.

In comparison to the two lignite regions, the western region's subbituminous coal-fired generating capacity has grown only moderately. Some of the major constraining factors have been (a) relatively low demand growth for coal-fired capacity, due primarily to hydroelectric power growth in the northwest; (b) siting problems due to strict air-quality standards and low water availability in the major resource areas; and (c) the long distances from the coal reserves to the major electric load centers. The last two factors tend to increase the cost of power delivered to the consumers in a region where reasonably low cost alternatives have been available.

The states of Arizona and New Mexico have 15 large coal-fired units supplying both in-state and west-coast markets (primarily southern California). In Wyoming and Montana, six units supply power to the transmission loop, most of which moves to the northwestern states. In addition, the state of Colorado has ten small coal-fired units, primarily meeting local needs.

The export of Powder River Basin subbituminous coal was triggered by the mandatory limits imposed by the US Environmental Protection Agency (EPA) in 1970 and 1971 on ambient  $SO_2$  concentrations and plant emissions. Because of high transportation costs, the delivered price of western subbituminous coal exceeded the price of indigenous (but highsulfur) midwestern coal. However, when the cost of stack gas scrubbing was added to that of utilizing the local coal, the western low-sulfur coal had a cost advantage for many utilities in the Midwest. This incentive was reduced, but not removed, by the operating difficulties and costs involved in burning low-rank coal in boilers designed for bituminous coal. For example, the higher moisture content (lower heating value), lower grindability, and high ash resistivity of the low-rank coal generally increase the boiler, pulverizer, and electrostatic precipitator costs compared to bituminous coal. Also, the boiler capacity may be reduced somewhat by fouling and slagging.

As a result of this regulation-created demand for low-sulfur coal, western subbituminous coal penetrated the midwestern electric utility market very rapidly during the 1970s. From a level of essentially zero before 1970, the low-rank coal-fired capacity grew to approximately 13,600 MW by 1980 in the states of Minnesota, Wisconsin, Illinois, Iowa, Arkansas, Missouri, Kansas, Oklahoma, and Nebraska. Another 7400 MW of subbituminous coal-fired capacity was planned in these states during 1980–1985.

The 1977 Clean Air Act Amendments and the 1979 New Source Performance Standards significantly altered the situation by requiring that all new coal-fired plants reduce  $SO_2$  emissions by at least 70%. While western low-sulfur coals require substantially less sulfur removal capability than high-sulfur bituminous coals, the previous large incentive to use western coals in midwestern locations has been dramatically reduced. Coals of different rank from different regions once again compete primarily on the basis of delivered cost (extraction plus transportation) to the consumer.

Industrial markets for low-rank coal are currently very small, amounting to only about 1% of total production, or 1–2 million tons/year, and occurring near the coal deposits. The basic problem for low-rank coal in penetrating industrial markets is that an industrial-size coal-fired boiler is about three times higher in capital cost than an oil- or gas-fired boiler. Even in cases where the total cost of producing a pound of steam from coal is lower than from oil or gas, many firms reject the coal option. Reasons cited are the uncertainties and difficulties in meeting environmental regulations and the inconvenience of handling solid fuel.

A major impediment to the use of coal by small consumers is their inability to achieve economies of scale in delivery, utilization, and cleanup systems. For example, single-car rail rates are double (or more) the unit train rates negotiated by utilities. Solving this problem would require the development of a "wholesaling" infrastructure for low-rank coals providing large, efficient delivery systems to industrial distribution centers. Either coal slurry pipelines or unit trains could be used for long-distance transportation. Other options to consider include (a) central combustion facilities feeding steam and power distribution networks, and (b) central gasification facilities feeding fuel gas to individual boilers. One advantage of these options compared to individual direct use would be the convenience and economy of centralizing all important environmental control and disposal operations. Ensuring steady, long-term demand for such systems may require the development of new cooperative investment and financing approaches.

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By the mid-1980s, the industrial use of low-rank coal in the Gulf Coast is projected to be fully competitive with the use of oil and gas in conventional boilers. As fluidized-bed combustion technology gains acceptance, the cost incentive to use Gulf lignite could be substantial. The huge petrochemical industry in the Gulf region also represents a large potential market for lowrank coal via the manufacture of synthesis gas.

Coal-based synthetic fuels will begin contributing to our energy supply in significant amounts when they pass the cost "crossover" point with petroleum and natural gas, possibly starting in the 1990s. The electric utility industry will use coal-derived gas in highly efficient "combined cycle" (gas turbine/steam turbine) plants, and will use coal-derived liquids in existing oil-fired boilers and new peak-load units. Transportation and distribution infrastructures for oil and gas products from coal are in place, and the market can expand rapidly.

Several characteristics of low-rank coals (e.g. high reactivity, low sulfur content, noncaking properties) make them generally favorable feedstocks for many of the synthetic fuel processes. These advantages, coupled with the low extraction cost per Btu, indicate that low-rank coal could account for a very large portion of the eventual synfuels market.

The first commercial synthetic natural gas-from-coal plant in the United States is the Great Plains Gasification Associates project, which will use North Dakota lignite. Almost all of the synthetic natural gas (SNG) projects proposed during recent years (most of which have been cancelled or deferred owing to high costs and the regulatory climate) intended to use western low-rank coals.

Commercial production of synthetic liquids from coal appears to be farther in the future than that of SNG. In addition, the advantages of lowrank coals as feedstocks to direct liquefaction processes are not as great as their advantages in first-generation gasification processes.

The October 1983 National Energy Policy Plan (NEPP) (3, 4) mid-range projection for the future use of coal in the United States is plotted in Figure 4. Under this scenario, total US coal production would rise from the 1982 level of about 820 million tons/year to about 2 billion tons/year in the year 2010. In the predominant electric utility segment of the market, low-rank coals are expected to increase their present share from 37% to about 50%. The use of electricity will also continue its growth as a percentage of total US energy consumption, from 31% in 1980 to a NEPP projection of 42% by the year 2000. Current overcapacity in the electric utility industry (reserve margins of 30%-35% as compared to a desired 20%) is predicted by a recent US Department of Energy (DOE) study (5) to disappear within a few years, so that 438 gigawatts (GW) of new generating capability would be needed by 2000. Coal's share of electricity generation is projected to increase from about 50% in 1980 to 59% by 2000.

The coal export market may include some western subbituminous coals shipped to the Pacific importers such as Japan. The NEPP calls the United States the world's "coal source of last resort" and says that because the country is projected to be the world's marginal supplier of coal, small changes in oil prices and economic growth could radically alter projected US exports.

At present it is clear from examining the US coal markets that low-rank coal growth is constrained mainly by demand. Competing fuels have the edge in many markets. Low-rank coal capacity currently exceeds produc-

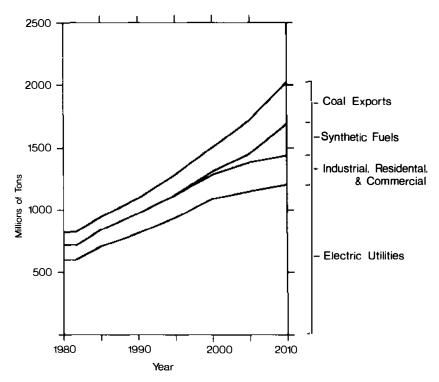


Figure 4 DOE projections for US coal production to the year 2010 (3, 4).

tion substantially, and is projected to do so at least through the mid-1980s. As coal use expands, supply-side constraints will also emerge. Impacts of greatly expanded surface mining and coal use in the arid and semi-arid west, water availability, and inadequate coal transportation capabilities are the major factors that could hold back continued growth.

The current technology for mining, transporting, and firing low-rank coal to generate electricity for major regional power grids will continue to predominate for some time. However, this technology is being challenged by emerging market opportunities and more stringent environmental control requirements. Cost-effective solutions to these problems require technology development.

### PROPERTIES OF LOW-RANK COALS

Low-rank coals have a porous structure containing between 20 and 40 wt% moisture. (Foreign brown coals contain up to 65% moisture.) Lignites from the Northern Great Plains retain a woody character, whereas Gulf Coast lignites appear to have undergone more decay during coalification. All low-rank coals crumble upon air drying owing to shrinkage and loss of elasticity. Texture and friability vary widely.

Differences in the properties of coal vitrinites, the predominant form of pure organic matter in coal, are shown as a function of rank in Table 2 (6). The effects of metamorphic change on heating value and moisture content, as well as on the carbon/oxygen ratio, are evident. The volatile matter contents of low-rank coals are higher owing to carbon dioxide rather than

	Lignite	Subbituminous	Bituminous	Anthracite
Analysis (wt% daf) <sup>b</sup>	_			
Moisture capacity	40	25	10	<5
Carbon	69	74.6	83	94
Hydrogen	5	5.1	5.5	3
Oxygen	24	18.5	10	2.5
Volatile matter	53	48	38	6
Heating value				
(MJ/kg)	27.0	29.5	34.2	35.3
Aromatic				
C/total C	0.7	0.78	0.84	I
Density				
$(He, g/cm^3)$	1.43	1.39	1.3	1.5

Table 2	Properties of vitrinites in US coals <sup>a</sup>
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<sup>a</sup> Source : (6).

<sup>b</sup> daf = dry, ash = free basis.

combustibles; only 17% of the calorific yield occurs in volatile products from North Dakota lignite, compared with 30% for some bituminous coals (7).

Average analyses of low-rank coals and their ashes are given by region in Table 3 (1, 2, 8–10). The ash analyses are for standard American Society for Testing and Materials (ASTM) ash reported as oxides on a  $SO_3$ -free basis. A notable point is the higher proportion of the alkaline components (CaO, MgO, and Na<sub>2</sub>O) in low-rank coals compared to higher-rank coals.

The Fort Union Region lignite deposits currently mined in western

	Region and Coal Rank				
	Ligi	Lignite		Subbituminous	
	Fort Union (8)	Gulf (2)	Powder River (9)	San Juan (10)	
Proximate analysis, wt% (as-received)					
Moisture	37.2	30.8	25.4	12.8	
Volatile matter	27.3	30.8	29.6	33.1	
Fixed carbon	30.3	24.9	38.7	40.6	
Ash	6.2	13.5	6.3	13.5	
Heating value, Btu/lb	6820	5803	8820	10020	
Ultimate analysis, wt% (dry, mineral matte	er-				
free)					
Hydrogen	4.9	6.1	5.1	5.5	
Carbon	71.9	62.1	75.6	77.6	
Nitrogen	1.1	1.1	1.2	1.4	
Oxygen	21.0	28.8	17.3	14.3	
Sulfur	1.1	1.9	0.8	1.2	
Number of samples for proximate and					
ultimate analyses	212	495	79	82	
Ash analysis, wt% (SO <sub>3</sub> -free basis)					
SiO <sub>2</sub>	24.5	47.5	40.2	64.7	
$Al_2O_3$	13.8	18.0	21.8	23.0	
Fe <sub>2</sub> O <sub>3</sub>	11.3	7.8	9.7	5.0	
TiO2	0.5	1.3	0.8	1.2	
$P_2O_5$	0.4	0.1	0.4	_	
CaO	30.6	15.3	19.4	2.9	
MgO	8.6	3.4	5.4	0.7	
Na <sub>2</sub> O	8.1	1.0	2.0	2.5	
K <sub>2</sub> O	0.5	0.7	0.3	0.7	
Ash fustion temp., °F (softening)	2290	2240	2180	2250	
Number of samples for ash analyses	212	97	104	16	

Table 3 Average analyses of US low-rank coals<sup>a</sup>

\* Sources : (1, 2, 8-10).

North Dakota and eastern Montana are typically 10–20 ft thick, and stripping ratios are very favorable. Within local deposits, the elemental analysis for H, C, N, and O is relatively uniform on a dry and mineral-free basis. However, the coal moisture, sulfur, and ash contents, and the ash analyses all vary widely. Moisture content ranges from about 32% to 44%; sulfur content from 0.1% to 1.9%; ash content from 3% to 12%; and heating value from 5800 to 7600 Btu/lb.

Lignite deposits in the Gulf Region occur primarily in Texas, Louisiana, Arkansas, Mississippi, and Alabama. The largest commercial deposits are found in sediments of the Wilcox Group with thin overburden. Seams are quite irregular and generally less than 10 ft thick. Since the quality of Gulf lignite improves from east to west, the average regional properties shown in Table 3 represent somewhat poorer quality lignite than is currently mined in Texas.

The subbituminous coal deposits mined in the Powder River Region are typically 10-100 ft thick and are low in sulfur and ash contents. Moisture content ranges from 20% to 31%, ash content from 3% to 16%, heating value from 7300 to 9600 Btu/lb, and sulfur content from 0.1% to 3.5%.

The San Juan Basin in New Mexico and Colorado contains both subbituminous and bituminous coal deposits, with heating values from 8900 to 11,900 Btu/lb and moisture contents from 10% to 20%. Ash contents range from 10% to 25%, and the ash is relatively low in alkaline elements compared to those of other low-rank coals.

The forms of inorganic species in low-rank coal before combustion include ion-exchangeable cations, minerals such as carbonates soluble in dilute acid, and minerals resistant to dilute acid, such as quartz and pyrite (11, 12). In addition to quartz and pyrite, some of the more familiar minerals that have been observed in lignites include calcite, kaolinite, and gypsum as commonly occuring species and dolomite, hematite, and a wide range of fedspar and clays as minor constituents (13, 14). The identification of specific minerals in coal may lead to better prediction of the behavior of the inorganic constituents in combustion or other processes than predictions based on the elemental composition alone.

The organic structure of lignite is interpreted in current literature (14-16) to consist of one-, two-, or three-ring aromatics and hydroaromatics connected by alkyl links. The ratio of aromatic carbon to total carbon is typically 0.7 or lower for lignite, compared to 0.84 for bituminous and 1.0 for anthracite (Table 2; 6). The prominent aliphatic content of lignite is further indicated by the 3%-5% yields of straight-chain alkanes up to C-32 that are obtained in liquefaction reactions. Pyrolysis also yields a homologous series of straight-chain alkanes and 1-alkenes. The length of aliphatic bridges is an issue of debate, with evidence suggesting both long

	Sulfur content, % (dry basis)				
	1.0 or less	1.1-1.8	1.8-3.0	Over 3.0	Total
Subbituminous	89.5	8.1	2.0	0.4	100.0
Lignite	40.7	33.4	23.4	2.5	100.0

Table 4 Distribution of US low-rank coal reserve base by sulfur content<sup>a</sup>

<sup>a</sup> Source : (17).

chains of ten or more carbon atoms and short methylene and ethylene bridges.

A significant difference between low-rank and bituminous coals is the 10 to 30 wt% of oxygen incorporated into the organic structure of low-rank coals (16). This oxygen occurs in hydroxyl, ether, and carboxylate functional groups, each of which contributes to the chemical behavior. The cleavage of ether groups plays a role in the initial stage of coal liquefaction. The hydroxyl groups give rise to the rich diversity of phenol derivatives found in liquefaction products and gasification effluents. The carboxylate groups, which account for an estimated two thirds of the coal oxygen, bind the alkali and alkaline earth cations (principally Na, Ca and Mg), which affect all aspects of low-rank coal utilization.

The other heteroatoms in low-rank coals are nitrogen and sulfur. Subbituminous coals and Fort Union lignites typically contain 1% or less of sulfur (Table 4; 17), whereas many bituminous coals contain 2% to 5% sulfur. Gulf Coast lignites are also higher in sulfur. In low-sulfur low-rank coals, most of the sulfur is present as organic sulfur rather than pyrite. Coal nitrogen content increases slightly with rank (Table 3) but varies only slightly within a local deposit. The chemical forms of sulfur and nitrogen in low-rank coal are not well established.

#### PREPARATION METHODS

The only preparation steps commonly applied to the low-rank coals currently burned by utilities are primary crushing at the mine, storage, and pulverization at the power plant. Cost-effective preparation methods would help to lower transportation costs and reduce ash deposition and sulfur dioxide emissions. Existing technology used for bituminous coals will not meet these requirements, and methods suited to the properties of low-rank coals are only now beginning to be developed.

Processes for removing some of the 10%-40% moisture in low-rank coals involve drying in hot gas, superheated or saturated steam, or pressurized

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hot water. Low-rank coals dried in hot gas reabsorb moisture and are dusty and subject to spontaneous combustion. Although large-tonnage rail transport and storage of such dried coal has been successfully demonstrated (18), it has not been commercially adopted.

Drying in high-pressure saturated steam or hot water produces a dense product that reabsorbs only a small amount of moisture. By heating a water slurry of lignite to 310°C at 1440 psig (pounds per sq inch gage), 67% of the moisture is expelled from the coal particles, about 50% of the sodium is removed, and some carboxylate is eliminated as carbon dioxide (19). The process can be optimized by separating excess water to produce highheating-value slurries tailored to pipeline transport, retrofit combustors, or entrainment gasifiers. Incorporation of other preparation techniques such as fine-coal cleaning may extend these applications. The costs of these lowrank coal-drying processes have not yet been analyzed owing to lack of engineering data.

Size reduction methods are affected by a loss in the strength of low-rank coal particles as they are heated to remove moisture. Although low-rank coals are typically more difficult to grind than bituminous coals, performance is improved by in-mill drying with hot air.

Gulf lignites and San Juan Basin subbituminous coals contain sufficient extraneous mineral matter and pyritic sulfur to warrant cleaning by conventional wet or dry methods. Separation from cleaning media would probably require special attention owing to coal particle attrition. However, adoption of conventional cleaning methods for even these suitable coals is doubtful, since partial cleaning would still not meet strategic goals applying to transportation cost reduction, ash deposition and erosion, or sulfur control.

If methods effecting a more complete separation of inorganic constituents and sulfur from low-rank coals can be developed, their range of applicability will depend on data correlating the separability of minerals with fineness of coal grind, which are now being compiled for low-rank coals (20). Fine-coal cleaning techniques such as oil agglomeration, froth flotation, and magnetic separation, as well as chemical cleaning methods for removing organic sulfur, will need to be tested for their applicability to low-rank coals. Removal of ion-exchangeable cations by acid washing has already been demonstrated at laboratory and process development scale. Its cost has been estimated at \$3 to \$6 per ton in preliminary engineering studies (21, 22).

#### COMBUSTION OF LOW-RANK COALS

The fundamental burning characteristics of US low-rank coals are only beginning to be investigated (20, 23), but available indications provide

important background for understanding more applied work. Unlike caking bituminous coals, the low-rank coal particles retain their size and shape during ignition, devolatilization, and initial burning rather than coalescing into a plastic mass. Low-rank coals have a high internal porosity that contributes to their reactivity and appears to allow uniform release of volatiles over the external surface rather than the release by individual pores noted with bituminous coals. During devolatilization, low-rank coals emit low yields of tar and light oil, some combustible gas in the form of  $CH_4$ , CO, and  $H_2$ , and an abundance of inert gas that is principally  $CO_2$ . The high moisture content of low-rank coals greatly affects the initial heating of coal particles leading to ignition. Also, ignition characteristics of low-rank coals are believed to be more dependent on char reactivity than on volatiles, owing to their lower yield of the combustible volatiles that stabilize the ignition of high-volatile bituminous coals. Ignition properties of low-rank coals are also importantly affected by their ion-exchangeable cations.

Many low-rank coals contain quantities of alkaline constituents (Ca, Mg, and Na) that are more than stoichiometrically equivalent to their sulfur content. In combustion, however, only a portion of the sulfur is retained in the alkaline ash. In an oxidizing atmosphere, higher retention is favored by lower combustion temperature and longer contact between the ash and the  $SO_2$ -bearing combustion gases. In pulverized-fuel-fired boilers the fraction captured varies from near zero to 40% depending on the amount of sodium and calcium in the coal (24).

For the lower temperature (e.g.  $1500^{\circ}$ F) and extended gas/solid contact of fluidized bed combustion (FBC), sulfur retention on ash is higher, approaching 100% for some low-rank coals (Figure 5; 25, 26). Utilization efficiency for either ash alkalinity or added sorbent varies widely from 25% to nearly 90% (26) depending on the alkaline constituent (sodium being more effective than calcium) and the mass action effect of the coal's sulfur level. At a coal sulfur content of 1%, a 100% or greater excess of alkaline constituents (alkali/sulfur = 2) is typically required to achieve 90% sulfur control.

Slagging combustors are now being developed by TRW and Rockwell International that operate under reducing conditions (0.7–0.9 stoichiometric oxygen) in the slagging section and reject 90%–95% of the coal ash as slag. Rockwell has reported some preliminary data indicating very high sulfur retention in the slag (up to 95% with a subbituminous coal having a 2/1 Ca/S ratio), and both companies report very low NO<sub>x</sub> emissions from the overall furnace (27, 28).

At the relatively low flame temperatures produced by low-rank coals, thermal  $NO_x$  formation from combustion air is also relatively low. Fuelbound nitrogen is of greater concern (23). It is initially released primarily in either HCN gas or char. The fraction initially appearing as HCN is rapidly reconverted to N<sub>2</sub> in a reducing atmosphere, which allows for some control of NO formation by properly distributing (staging) the combustion air. NO formation from the fuel nitrogen in the char is more difficult to control because an oxidizing atmosphere is required to burn the carbon. In practice the overall NO<sub>x</sub> emission from the pulverized-coal (pc) firing of highmoisture lignite can be controlled to 20%-40% of the fuel-bound nitrogen, depending on burner staging (29).

The release of inorganic constituents during combustion (30) depends on their modes of occurrence in the coal, the fineness to which the coal is ground, and the combustion temperature. Discrete mineral particles may retain their identity or coalesce into larger ash particles. Ion-exchangeable cations are more uniformly distributed throughout the coal and produce particulates whose fineness is proportional to the fineness of the coal grind. Ash volatiles also affect the variation of composition with size of particulates, as evidenced by the enrichment of sodium and sulfur in the finer fraction of the ash. The degrees of both alkali volatility and ash vitrification depend importantly on combustion temperature.

Ash deposition has caused severe fouling and slagging in conventional boilers burning low-rank coals, and a related problem of bed agglomeration has been encountered in fluidized-bed combustors. The mechanism of ash deposition has been extensively studied and in general terms, results from the occurrence of a partial melt phase that binds individual fly ash

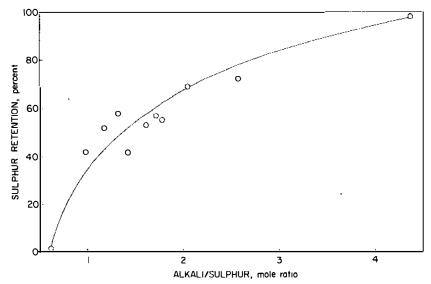


Figure 5 Fluidized-bed combustion sulfur retention as a function of total alkali to sulfur ratio (25).

particles (Figure 6; 31, 32). Diffusion and sulfation within the melt phase increase deposit hardness with time.

Sodium content is strongly correlated with fouling (33) and agglomeration (25, 26) and is known to play a very important role in the mechanism of ash deposition (31, 32). The organically bound sodium in low-rank coals is volatilized in combustion and subsequently condenses on fly ash particles and deposits, where it acts as a flux for a susceptible fraction of the ash. The resulting melt phase has been variously suggested to consist of complex sulfates, an eutectic mixture of sodium and calcium sulfates, or a crystalline mineral melilite. All of these materials are known to melt at lower temperatures if sodium predominates.

The impact of sodium-induced boiler fouling at seven power plants operating 23 boilers on western low-rank coals was estimated in a 1980 study (34) to involve losses in power output due to outages and curtailments

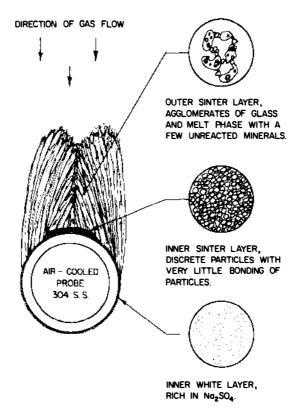


Figure 6 Ash-fouling deposits from high-sodium, low-rank coals divided into three primary layers (31).

ranging up to 17% of annual production. Losses were greatest for North Dakota lignite (up to 17%), moderate for subbituminous coals (1% to 6%), and very low for Texas lignite. Remedies for fouling include load reduction, boiler modification, added soot blowers, additive injection, coal switching and blending, and (in the future) coal washing and ion exchange.

### ENVIRONMENTAL CONTROL TECHNOLOGY

Wet scrubbers installed on 4200 MW of capacity at western utilities use the alkalinity in the coal fly ash in place of part or all of the lime or limestone that would normally be used (35). These scrubbers have suffered many of the same mechanical operating problems as conventional lime/limestone scrubbers, and their application to new units has been phased out.

The present state of the art in sulfur control for low-sulfur low-rank coals is based on spray dryers that introduce a finely atomized soda ash solution or lime slurry into the hot stack gas ahead of a fabric filter or electrostatic precipitator (36, 37). Both sodium-based (1981) and lime-based (1983) units are operating on 450-MW boilers burning North Dakota lignite. This technology offers significant cost and operating advantages for low-sulfur coals, but extension of the technology to high-sulfur coals requires higher use of lime reagent.

An extension of the spray dryer concept to accomplish simultaneous control of both sulfur and nitrogen oxides is being developed at the University of North Dakota Energy Research Center. As presently conceived, this technique will involve dry injection of finely divided pressure-hydrated lime and throwaway  $NO_x$ -reduction catalyst ahead of a ceramic filter baghouse operated at 800°F to 1000°F (20).

Low-rank low-sulfur coals will continue to find applications in retrofit sulfur control technology. Low-rank coals are particularly suitable for direct furnace injection of sorbents, as in the limestone injection modified burner (LIMB) program sponsored by EPA or the pressure-hydrated lime study under DOE, owing to their lower flame temperatures and the smaller impact of a relative excess of reagent on cost and boiler operability when using low-sulfur coal.

Particulate control for low-rank coals has shifted gradually from electrostatic precipitators to fabric filter baghouses. The performance of the first precipitators installed for low-rank coals in the late 1960s was poor owing to the high electrical resistivity of fly ash from low-sulfur and lowsodium coals. Newer units were designed with very large specific collecting areas or for elevated (hot side) operating temperatures. Some baghouse installations have also experienced problems, including bag failures, poor cleaning, and high back pressure. Hot-gas cleanup methods for protecting gas turbines from particulates and alkalies in the combustion gases of pressurized fluidized-bed combustors burning low-rank coals have received little attention. However, fluidbed particulates are being studied to provide the data needed for future assessment.

# GAS CONVERSION PROCESSES FOR LOW-RANK COALS

The low-rank coal synfuel industry arrived on July 25, 1980, with the ground breaking for the Great Plains Gasification Associaties (GPGA) plant at Beulah, North Dakota. When brought on line late in 1984, this plant will convert 14,000 tons of lignite per day (4.7 million tons per year) to 137 million standard cubic feet per day of pipeline-quality synthetic natural gas (SNG) at an energy efficiency of 66%. This first-of-a-kind project had to survive a highly complex permitting process and protracted negotiations on financing options between 1972, when it was proposed, and January 1982, when the federal loan guarantee was signed by DOE. The project, which is 90% constructed and is on schedule and below budget, now faces the challenge of surviving a significant downturn in projected oil prices (to which its product gas price is linked by the loan guarantee agreement) and another potentially protracted negotiation involving DOE, the US Synthetic Fuels Corporation, and the five major energy companies that have invested in the plant.

The technology used by the GPGA plant is based largely on the proven experience at the SASOL plants in South Africa (Figure 7; 38). Lurgi Mark IV gasifiers react lignite screened to a size of  $2 \times 1/4$  inch with steam and oxygen at a pressure of 430 psig to produce a raw synthesis gas. Coal fines representing approximately one third of the crushed coal are sold to the adjacent power station operated by the Basin Electric Cooperative. Condensible tars, oils, wastewater, and acid gases are removed, and gas is shifted to a 3:1 ratio of hydrogen to carbon monoxide before methanation.

The environmental control systems for the Great Plains plant are designed to achieve "zero discharge" of surface water and over 90% control of sulfuremissions. The plant will have less apparent environmental impact than a fully controlled electric generating plant using a similar amount of lignite (and will produce about twice the final energy product). However, some areas such as the recovery and reuse of tar, oil and contaminated wastewater, and the disposal of solid process wastes represent new problems that warrant special study.

The water balance for the GPGA plant is maintained by treating process wastewater by extraction (Phenosolvan process) and steam stripping

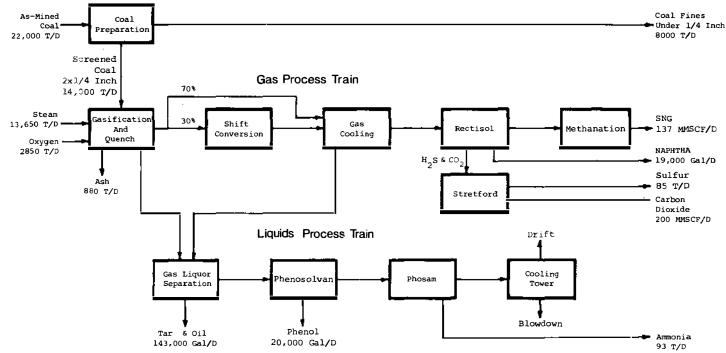


Figure 7 Great Plains Gasification Associates process flow diagram (38).

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(Phosam process) and using the partially purified water containing some residual organics as makeup in the process cooling towers. Blowdown from the towers is concentrated in evaporators and then incinerated to produce a solid waste.

A study of this method of wastewater treatment and disposal is being conducted by DOE, using the slagging fixed-bed gasifier (SFBG) at the University of North Dakota Energy Research Center (39). Analyses of this raw waste-water and stripped gas liquor indicate markedly different organic contaminants than those expected from GPGA design data, with SFBG wastewater containing high concentrations of hydantoins rather than the fatty acids reported by GPGA. These differences are most likely caused by the differences in gasification temperatures between slagging and dry-ash gasifiers. It is becoming apparent from this work that gasification waste characteristics will vary significantly with different coal types and process conditions.

Several alternative gasifier designs may also find application to lowrank coals. The slagging fixed-bed design investigated by British Gas and Lurgi at Westfield, Scotland, and by DOE at Grand Forks represents improvements in throughput, thermal efficiency, and steam consumption. High-throughput entrained-flow gasifiers such as the atmospheric Koppers-Totzek and the pressurized Texaco gasifier operate in a hightemperature slagging mode that consumes organic condensate.

Lignitic and subbituminous coals, with their noncaking property, are probably the only feedstocks that will be used in first-generation fixed-bed gasifiers, although the more friable of these coals may produce quantities of fines unsuitable for fixed-bed units. Low-rank coals, being more reactive than higher-rank coals, provide higher throughput and carbon conversion than bituminous coals in gasifiers like the dry-ash (low-temperature) Lurgi.

The high inherent moisture content of low-rank coals acts as a diluent and increases wastewater volume, and predrying in hot gas causes particle degradation. Moisture also affects operability, as in the case of the DOE slagging fixed-bed gasifier that could not sutain reactions at coal moisture contents greater than 40%. Also, the slurry-fed Texaco gasifier cannot be operated with acceptable levels of oxygen consumption and efficiency unless high-moisture coals are first dried in a manner that limits their reabsorption of moisture (such as hot-water coal drying, discussed earlier).

The ash-forming mineral constituents in lignite contain alkaline components that catalytically promote gasification reactions and also influence the melting properties and temperature/viscosity relationships of the slags produced. The corrosive behavior of these ash-forming components toward refractory materials is also different from that of the ashes of higherrank coals. These factors mean that gasifier design must be tailored to the coal ash analysis.

#### LIQUEFACTION OF LOW-RANK COAL

Indirect coal liquefaction involving the catalytic synthesis of hydrocarbons or methanol from coal-derived synthesis gas is coal specific only in the initial steps of coal preparation and gasification. The advantages of the technology are that it is commercially proven and produces easily refinable hydrocarbons similar to those found in petroleum. Commercial plants are operating at SASOL in South Africa, and others have been proposed for sponsorship by the US Synthetic Fuels Corporation.

Direct liquefaction processes react coal with a process-derived solvent and a reducing gas. Three processes aimed at distillate production have been developed to the pilot stage: (a) solvent-refined coal (SRC-II, 25 ton/day), (b) Exxon Donor Solvent (EDS, 250 ton/day), and (c) H-Coal (600 ton/day). These processes were developed for the liquefaction of bituminous coal and use hydrogen as the reductant. Common features of the three processes include reaction temperature ( $440^{\circ}C$ - $460^{\circ}C$ ), residence times (30--60 min), and pressures [14-21 MPa (2000-3000 psi)]. All three processes have operated on subbituminous coals; only the EDS process has operated on lignite.

Optimum distillate yields from US lignites (40) have been shown to be as high on a dry, ash-free basis as those obtained from bituminous coals, but only if the vacuum bottoms are recycled. Lignites are more readily liquefield if the reducing gas contains some carbon monoxide and water, which together with recycling the bottoms yields the highest conversions and the best bench-unit operability.

Under liquefaction conditions, a portion of the carbon skeleton in lignite breaks down easily to form highly reactive free radicals that are either immediately capped with hydrogen or reacted to form char. In preheater studies at Project Lignite at the University of North Dakota (41), soluble oil yield reached about 30% in only 30 sec. However, optimum yields are typically achieved only after residence times greater than 30 min preferably with recycling the bottoms—indicating that other portions of the organic structure break down far more slowly. Elevated temperatures in the range of  $440^{\circ}C-460^{\circ}C$  are also required to obtain high distillate yields in the absence of added catalyst.

In early research, the combination of lignite's high initial reactivity and the apparent need for an elevated reaction temperature seemed to provide only a very narrow operating range between low conversions and rapid charring. In single-stage reaction systems using hydrogen, the necessary conditions were best met in the modified EDS process, which operated successfully on Texas lignite using both a hydrotreated distillate (H-donor) recycle and bottoms recycle. Other means of process optimization are now becoming available, including use of carbon monoxide reductant,  $H_2S$ /iron sulfide catalysts, and staging of reaction temperatures.

Carbon monoxide or synthesis gas has been shown to be a preferred reductant for lignite at all temperatures from  $350^{\circ}$ C to  $480^{\circ}$ C (42). However, the reaction mechanism and the effectiveness of this step differ depending on the coal, its moisture content, and the reaction temperature. Appell & Wender (43) attributed the effectiveness of carbon monoxide to the formate ion intermediate, which in the presence of alkaline earth oxides reduces carbonyl groups to alcohol. Since formate is unstable at temperatures greater than  $400^{\circ}$ C, they proposed the concept of staged heating with sufficient hold time below  $400^{\circ}$ C to take advantage of the formate mechanism. This concept is currently being studied at the University of North Dakota Energy Research Center as the first step in a two-stage process that involves a higher temperature in the second stage. At higher temperatures CO reacts directly with lignite and its liquefaction products to form volatile compounds.

Partial removal of moisture from lignite before liquefaction results in lower process pressure, increased coal throughput, and less process-derived wastcwater. However, drying in air with accompanying oxidation, or in nitrogen, also reduces product yield. Alternatively, lignite can be slurried in recycle oil and heated to drive off water, which has been shown to be beneficial if about half the moisture is removed.

Recent research by Stenberg and co-workers (44, 45) has demonstrated the importance of sulfur chemistry in lignite liquefaction, and particularly the effectiveness of  $H_2S$  in promoting lower temperature reactions that favor high distillate yield, lower gas yield and reductant consumption, and improved operability. In model compound studies,  $H_2S$  is an important promoter of hydrogen donor activity, hydrocracking, cleavage of aromatic rings, and water gas shift catalysis. The beneficial effect of  $H_2S$  compensates for a low hydrogen-donor solvent quality. The superiority of  $H_2S$  as a liquefaction promoter for lignite over iron compounds and sulfur is illustrated in Figure 8 (40), which graphs total conversion at 460°C. Even more significantly, the total oil yield from one Texas lignite in recycle liquefaction using syngas was shown to be higher at 400°C with  $H_2S$  added than at 460°C without  $H_2S$ , and both gas production and reductant consumption were reduced by two thirds. Other tests confirm these trends and show that  $H_2S$  stabilizes recycle slurry viscosity.

Continuing liquefaction research on low-rank coals funded by DOE at the University of North Dakota Energy Research Center (20) is investigating the advantages of combining  $H_2S$  with alumina-supported iron sulfide catalysts and combining  $H_2S$  with CO in temperature-staged reactions. Other activities include using isotopic tracers (<sup>13</sup>C and <sup>2</sup>H) to follow

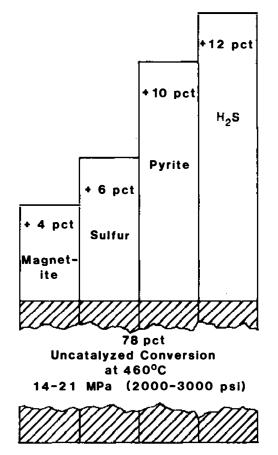


Figure 8 Averaged effects of added promoter on conversion of Beulah, North Dakota, lignite. Reactions were carried out in the 54 kg/day (120 lb/day) GFETC CPU (tubular reactor) on asreceived and slurry-dried lignite using either  $H_2$  or syngas. Benefits were more pronounced with syngas and water than with hydrogen (40).

hydrogen transfer and carbon monoxide reactions with coal and solvents. The results of these ongoing studies suggest that direct liquefaction processes can be designed for low-rank coals to outperform current technologies such as SRC-II and EDS in terms of cost and operability.

#### CONCLUSIONS

Resources of low-rank coals in the United States are comparable in quantity to available resources of bituminous coals but are found primarily in the western part of the country. With the exception of the Gulf region lignites, these coals are generally remote from major energy-consuming areas. In most cases, the relatively thick seams of low-rank coal found close to the surface can be strip-mined at a fraction of the cost required to produce US bituminous coals.

The importance of low-rank coal in the US energy system increased dramatically in the 1970s. This increase resulted from a combination of factors, including the escalating prices of oil and natural gas, the possibility of using low-rank coal to comply with clean-air regulations in some regions, the development of improved combustion technology, and the increased wheeling of power from region to region.

Low-rank coals are noted for causing ash-related problems during combustion. However, the high alkali and low sulfur contents of many lowrank coals make them attractive for reducing sulfur emissions without much addition of limestone or lime.

Some low-rank coals are especially attractive for gasification and liquefaction and are likely to dominate the early development of synthetic fuels plants and coal gasification/combined-cycle utility power plants. With all these factors considered, the role of low-rank coals in the United States is likely to continue growing as process improvements are made.

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