Kirk-Othmer Encyclopedia of Chemical Technology.

LIGNITE AND BROWN COAL

Lignite and brown coal are common names for coals having properties intermediate between peat and bituminous coal as a result of limited coalification (see Coal). In general, brown coal designates a geologically younger, ie, less coalified, material than the firmer, fibrous lignite. In the ASTM classification (1), both kinds of coal are classified as lignite. In many English-speaking countries, the consolidated coals are termed lignite, and unconsolidated coals are termed brown coal. In Australia, and in Germany and a number of other European countries, the generic term brown coal is used for the whole class, including some coals that are included in the ASTM classification as subbituminous. Lignite signifies the firmer, fibrous, woody variety. Herein lignite is used as the comprehensive term.

Selection of coal for a particular use requires a knowledge of composition greater than that supplied from the ASTM classification. Progress is being made toward classifying all kinds of coal, including lignite, by correlating properties with composition and other qualities (2).

The primary use of lignite is combustion in steam (qv) generation of electric power (see Power generation). Lesser amounts generally in the form of briquettes are used for industrial and domestic heating outside of the United States. Briquettes are pressed and often carbonized at low temperatures to provide a smokeless fuel. The by-product tars obtained from briquette production have been used for liquid fuels and chemical manufacture (see Tar and pitch). Lignite is also converted by gasification to synthesis gas for motor fuels, chemicals, and ammonia-based fertilizers (qv) in large integrated plants (see Coal conversion processes; Feedstocks, coal chemicals; Fuels, synthetic).

Worldwide production of lignite was over 1.13×10^9 tons in 1990 (U.S. production was 8×10^7 t). U.S. production has risen at ca 2% yr in the early 1990s. The world's proved lignite reserves can be recovered under local economic conditions with existing available technology, and were over 3.8×10^{11} t in 1990, representing almost 300 years of production in 1990 terms (3, 4).

Lignite is less valuable than coals of higher rank, primarily because its much higher (30–70% as mined) water content (5) and high chemically combined oxygen content result in a relatively low heating value (LHV). In the past, the expense of shipping limited the market largely to the vicinity of the mine. However, in the United States the low sulfur content of lignite has made long distance shipments economically feasible, in order to limit sulfur oxide emissions at electric power generation plants. The increasing worldwide demand for energy together with desire for national self-sufficiency has increased the importance of low heating value coals.

1. Geology

Lignite was deposited relatively recently (ca $2.5 - 60 \times 10^6$ yr ago), mainly during the Tertiary era. U.S. deposits include those in the Dakotas, Alaska, Montana, and Wyoming. Other deposits exist in Saskatchewan and northwestern Canada, Germany, Asiatic Russia, Pakistan, northern India, Borneo, Sumatra, and Manchuria. The Miocene period provided the brown coal deposits that are up to 300 m thick in the Latrobe Valley of Victory

Table 1. International Classification of Coals having a Gross Calorific Value below 23.8 MJ / kg[abc]

Group number	Group parameter tar yield, [d]	Code number					
40m	25	1040	1140	1240	1340	1440	1540
30	20–25	1030	1130	1230	1330	1430	1530
20	15–20	1020	1120	1220	1320	1420	1520
10	10–15	1010	1110	1210	1310	1410	1510
00	≤10	1000	1100	1200	1300	1400	1500
class number		10	11	12	13	14	15
class parameter, [e]		≤20	>20 – 30	>30 – 40	>40 – 50	>50 – 60	>60 – 70

[a] Moist, ash-free basis (30°C and 96% RH).
[b] Ref. 7.
[c] To convert MJ/kg to Btu/lb, multiply by 430.2.
[d] Dry, ash-free basis.
[e] Numbers represent % of total moisture of ash-free freshly mined coal.

in Australia. In addition, deposits in Venezuela, Mexico, southern Germany, the Volga region, and northern China were laid down during this period (6). The oldest deposits, which occur in the Moscow basin, were formed in the lower Carboniferous period, ca 200×10^6 years ago.

The Pliocene lignites in Alaska, southeastern Europe, and southern Nigeria are the youngest coals. A number of peat accumulations in different parts of the world representing a range of climates indicate that the process of coal formation continues to take place.

2. Classification

Several classification schemes have been used in different places and for different purposes. In the United States the ASTM method is used for all kinds of coal, from lignite through anthracite (1). The criterion for classification of lignite through high volatile B bituminous is moist, mineral–matter-free energy content. The term moist refers to bed moisture only, and the bed samples must be collected as described in ASTM Standard D388. In this method of classification, lignite and brown coal have moist energy <19.3 MJ/kg (8300 Btu/lb). Before 1960 consolidated coals were called lignite and unconsolidated coals, brown coal. The 1994 ASTM Standard D388 distinguishes lignite A from lignite B. The heating values of dry lignite A range from 14.6 to 19.3 MJ/kg (6300 – 8300 Btu/lb); the values of dry lignite B range below 14.6 MJ/kg (6300 Btu/lb). In the United States, the terms soft coal and hard coal refer to bituminous and anthracite, respectively. However, in Europe soft coal refers to lignite and brown coal, whereas hard coal refers to bituminous. In 1958 the Coal Committee of the Economic Commission for Europe (ECE) officially adopted the classification of lignite and brown coal given in Table 1 (7). Total moisture, which can be correlated with heat value, provides a guide to the use of coal as a fuel. Tar yield provides a measure of value as a raw material for the chemical industry. This nonscientific classification provides a guide for use. Another system based on a two-figure classification index derived from moisture content, tar yield, and petrographic values was adopted in 1961 as the International Classification for Brown Coals for the Peoples' Democracies (8).

In 1963 a classification of coals by rank (differing from the ECE scheme) was published by the International Committee for Coal Petrology (Table 2) (9). This includes a classification of brown coal that correlates a number of important properties including the percent reflectance of vitrinite in the coal. This is a simpler version of that used in German practice, which further subdivides soft brown coals into foliaceous and earthy. Most brown coals belong to the latter group.

Table 2. Classification of Coals by Rank, International Committee for Coal Petrology

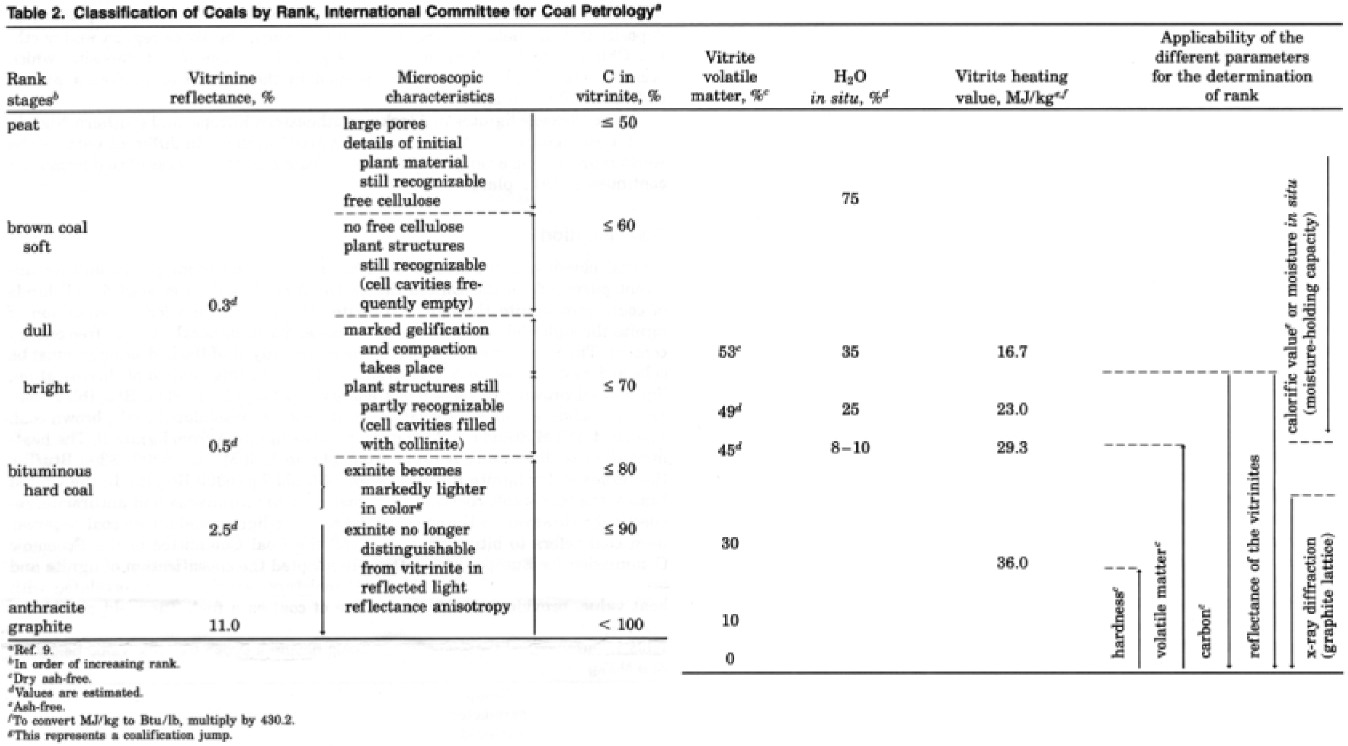


Table 2. Classification of Coals by Rank, International Committee for Coal Petrology[a]

Rank stages[b]	Vitrinine reflectance, %	Microscopic characteristics	C in vitrinite, %	Vitrite volatile matter, %[c]	H_2O *in situ*, %[d]	Vitrits heating value, MJ/kg[e,f]	Applicability of the different parameters for the determination of rank
peat		large pores details of initial plant material still recognizable free cellulose	≤ 50		75		calorific value[f] or moisture *in situ* (moisture-holding capacity)
brown coal soft	0.3[d]	no free cellulose plant structures still recognizable (cell cavities frequently empty)	≤ 60				
dull		marked gelification and compaction takes place		53[c]	35	16.7	
bright		plant structures still partly recognizable (cell cavities filled with collinite)	≤ 70	49[d]	25	23.0	carbon[c] reflectance of the vitrinites
	0.5[d]			45[d]	8–10	29.3	volatile matter[c]
bituminous hard coal		exinite becomes markedly lighter in color[g]	≤ 80				x-ray diffraction (graphite lattice)
	2.5[d]	exinite no longer distinguishable from vitrinite in reflected light	≤ 90	30		36.0	hardness[c]
anthracite		reflectance anisotropy		10			
graphite	11.0		< 100	0			

[a] Ref. 9.
[b] In order of increasing rank.
[c] Dry ash-free.
[d] Values are estimated.
[e] Ash-free.
[f] To convert MJ/kg to Btu/lb, multiply by 430.2.
[g] This represents a coalification jump.

Other terms that have been used to differentiate types of lignitic coal include humic brown coals, referring to those having substantial amounts of extractable humic acids, and sapropelic coals, referring to more homogeneous coals often having a high concentration of individual plant components. Some additional methods of classification are under development that center on the use of lignite for combustion in utility boilers or electric power generation. Correlations based on the sodium concentration in the lignitic ash (10), or soluble Al concentration (11) are used. The classifications are often given in terms of the severity of boiler fouling.

3. Composition, Properties, and Analysis

3.1. Macroscopic Appearance

Lignitic coals vary from brown to dull black when moist, although the color may appear considerably lighter when the coal is dried. The freshly broken surface of the most common type, the unconsolidated humic variety, may be light reddish brown but darkens rapidly during oxidation. Breakage is easiest for the unconsolidated coals. Strength and toughness increase as coalification increases. Because of its weak structure and tendency to shrink and crack on drying, brown coal disintegrates more easily than more mature coals (12). Remains of plants can be seen in some of these coals.

3.2. Physicochemical Structure

Water-filled pores and capillaries of differing diameters permeate the organic gel material that makes up as-mined lignite. There is some retention of moisture on air drying (5). The void volume or porosity ranges up to about 44% for lignitic coals (13) and decreases as the rank increases. The pore diameters vary and include a significant amount of very small pores that limit the size of molecules that can enter or leave. Lignitic coals have properties of molecular sieves (qv) and the large pore volume is believed to be partially responsible for the high observed reactivity. The relative accessible internal surface area ranges from $100 - 200\ \mathrm{m}^2/\mathrm{g}$ for lignitic coal and is about half this value for bituminous coals. In general, the internal surface area of coal is associated with capillary systems having 4-nm pores linked by 0.5–0.8-nm passages. About 75% of this free volume in lignite is associated with the larger pores (14). Mineral matter including salts of the humic acids is nonuniformly distributed.

3.3. Properties

The apparent density of lignite is $0.8 - 1.35\ \mathrm{g/cm}^3$ (13), which is lower than values given for higher ranking coals. Therefore, greater volume is required for storage, transportation, and lignite reactors than is needed for an equivalent weight of more mature coals. Lignite generally has lower elasticity and greater plasticity than more mature coals. The plasticity index, ie, ratio of elastic energy to plastic energy, involved in compressing coals has been used to indicate the ease of briquette formation (14). Briquetting without a binder is possible only for softer, less mature coals.

Humic acids are alkali-extractable materials and total humic acid content is a term that refers to the humic acid content of coal that has had its carboxylate cations removed with sodium pyrophosphate. Values for some typical Australian brown coals range from 24–92% (13). Treatment of lignitic coals with mineral acid to release the alkali and alkaline cations may dissolve up to 20% of the coal. The naturally moist coals are slightly acidic and have a pH of 3.5–6.5.

Solvent extraction using nonreactive liquids, such as C_3- or C_4-alcohols, benzene, or benzene–alcohol mixtures, yields generally 5–20% wax or bitumen (15). The yield and composition of the product are determined primarily by the petrologic character of the coal, not its degree of coalification. Montan wax is extracted from suitable coals for a variety of purposes.

The tar yield is usually higher for lignite than for more mature coals. Tar yields are important in determining selection for carbonization and for liquid fuel production by pyrolysis.

3.4. Oxidation

The high reactivity of lignites with oxygen requires special care during mining, transportation, and storage to avoid spontaneous combustion from heat generation. Contact with basic $(\mathrm{pH} > 8)$ solutions or oxidizing agents results in slow oxidation and the subsequent formation of humic acids. Many lignitic coals are almost entirely soluble in alkali solution, providing a technique for distinguishing between lignitic and bituminous coal. Spontaneous ignition of briquettes and coal is observed after wetting and at freshly broken surfaces (16).

3.5. Analysis

Analyses of a number of lignitic coals are given in Table 3. Figure 1, a distribution plot of 300 U.S. coals according to ASTM classification by rank, indicates the broad range of fixed carbon values (18). According to the ASTM classification, fixed carbon for both lignite and subbituminous coals has an upper limit of 69%, but in practice this value rarely exceeds 61%.

The moisture content of freshly mined lignitic coals can be as high as 73%, but it is usually 30–65%. The more mature, consolidated coals have lower moisture contents and thus a higher heating value. Figure 2 shows

Table 3. Analyses of Lignitic Coals[a]

	Proximate analysis, %					Ultimate analysis, %					
Coal	Moisture	Ash	Volatile matter	Fixed carbon	Volatile matter[c]	C	H	S	N	O	Heat value, gross dry, MJ/kg[b]
United States											
North Dakota[d]	33.9–41.2	3.5–8.5	25.4–27.6	26.9–31.7	45.2–48.8	71.1–74.4	4.8–5.3	0.3–2.3	1.0–1.1	16.9–22.7	27.8–29.6
South Dakota	38.5	5.8	26.9	28.8	48.3	72.0	4.8	0.7	1.4	21.1	28.4
Australia											
Victoria											
Yallourn	66.3	0.7	17.7	15.3	53.4	67.4	4.7	0.3	0.5	27.1	25.9
Yallourn north	50.0	2.0	26.0	22.0	54.2	68.3	4.9	0.3	0.6	25.9	25.9
Germany											
Lower Rhine[e]	60.0	2.3	20.6	17.1	54.6	68.9	5.3	0.3	25.5		25.9
Geiseltal	50.4	6.3	27.3	20.0	57.7						26.8
Riebeck-Montan	49.2	6.7	26.5	17.6	60.1						28.9

[a] Ref. 17.
[b] To convert MJ/kg to Btu/lb, multiply by 430.2.
[c] Dry ash-free.
[d] Range for coals from five areas.
[e] Average for four similar coals.

moisture and ash contents, as well as net heating values for lignitic coals from the world's principal deposits (19).

Moisture content affects a number of applications. The grindability index, ASTM D409, measures the relative ease of pulverizing coals and theoretically helps determine the capacity of pulverizers. In practice, low values of grindability occur at moisture extremes and maximal grindability occurs at intermediate moisture content. A small pulverizer to test grinding conditions for design purposes has been developed (20).

Mineral matter content or ash yield varies widely, from $<6\%$ in thick deposits as found in Australia and Germany, to $>40\%$ in deposits in Turkey. The composition of these minerals varies with location and depth in the seam (21). In the United States, lignitic ashes tend to have higher CaO, MgO, and Na_2O contents than do the ashes of bituminous coals. Significant amounts of metals are also organically bound. For example, the uranium contents of some ashes from the northern Great Plains exceed 1000 ppm (22). The volatile matter of lignite and brown coals ranges from ca 40 to 55%, and hydrogen from ca 4.3 to 6.1%, both on a dry mineral–matter-free basis. The hydrogen-to-carbon atomic ratio for subbituminous coal in the ASTM classification is ca 0.8–1.0. For lignite, the range is from 0.8 to about 1.1. The oxygen-to-carbon ratio for subbituminous coals is ca 0.1–0.2, and for lignite it is 0.2–0.3 (23).

4. Resources and Production

The importance of a coal deposit depends on the amount that is economically recoverable by conventional mining techniques. The world total recoverable reserves of lignitic coals were 3.28×10^{11} metric tons at the end of 1990 (3), of which ca 47% was economically recoverable as of 1994 (Table 4). These estimates of reserves change as geological survey data improve and as the resources are developed.

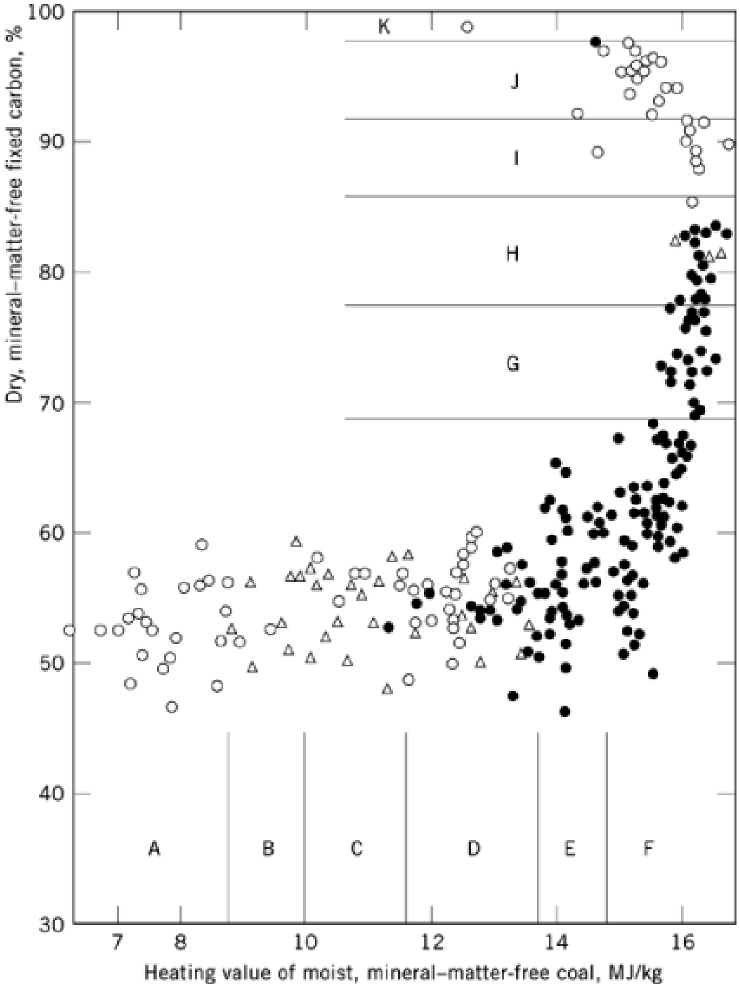


Fig. 1. Distribution plot for over 300 coals of the United States, illustrating ASTM classification by rank where (•) represents caking or agglutinating; (◦) noncaking; and (△) no information concerning caking properties. Region A corresponds to lignite; B, C, and D to subbituminous C, B, and A, respectively; D also corresponds to high volatile C bituminous; E corresponds to high volatile B bituminous; F, to high volatile A bituminous; G, to medium volatile bituminous; H, low volatile bituminous; I, semianthracite; J, anthracite; and K, *meta*-anthracite. To convert MJ/kg to Btu/lb, multiply by 430.2.

The production of lignite and brown coal for nations having large published reserves is also indicated in Table 4. Total world production, which was 6.6×10^8 t in 1961, increased to 8.1×10^8 t in 1972, and to 1.13×10^9 t in 1990 (24).

A comparison of available resources and production shows that Germany, ranked second in resources, was ranked first in production in 1990. Indeed a number of central and eastern European countries are producing proved recoverable reserves at a rate that should lead to exhaustion of local deposits before the end of the twenty-first century. On the other hand, the massive Russian reserves could allow production for a much longer time.

Rates of production of lignite have continued to increase since 1960. In 1980 374×10^6 tons of coal equivalent (tce) were produced. One tce is the amount of energy available from combustion of a metric ton

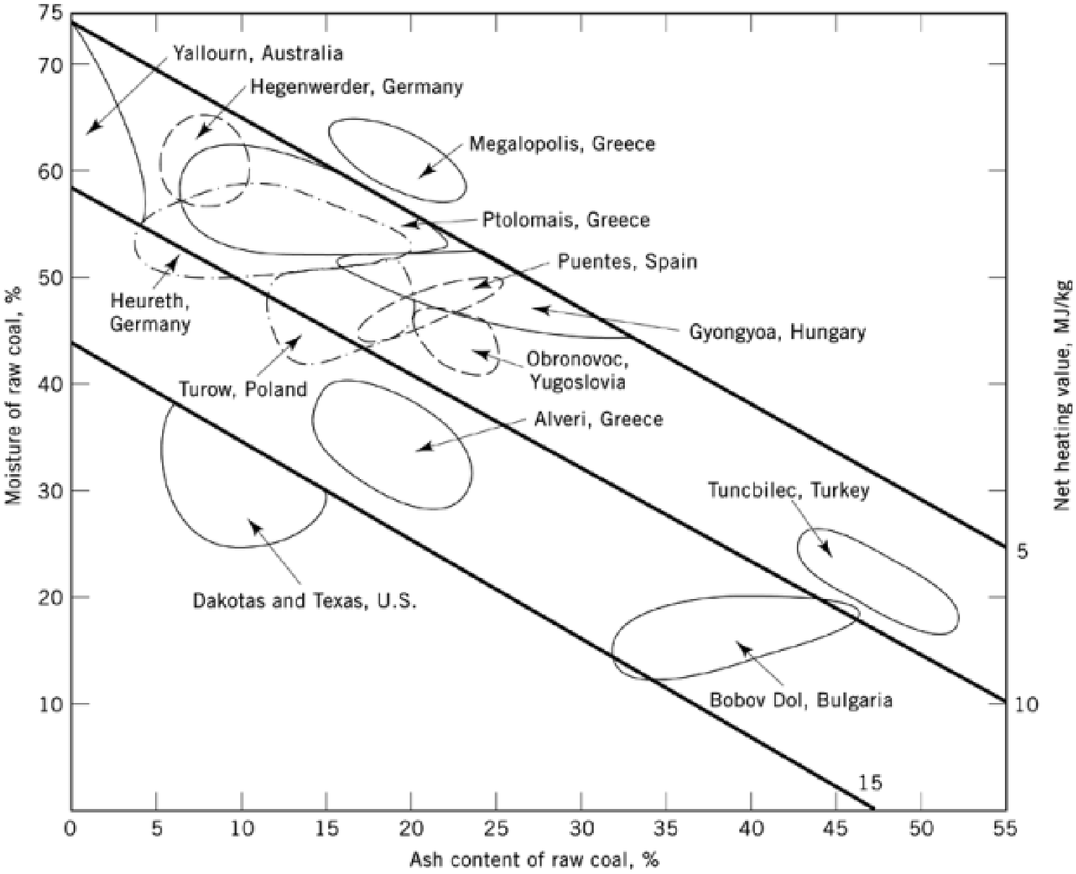


Fig. 2. Quality of lignitic coal. To convert MJ/kg to Btu/lb, multiply by 430.2.

of coal having a heat content of 29.3 GJ, ie, 29.3 MJ/kg (12, 600 Btu/lb) (3). In 1989 this figure had risen to 460×10^6 tce. This 23% increase is somewhat less than the 28% increase in hard coal production during this period (see Coal). In 1990 the 1130×10^6 metric tons of lignite produced worldwide represented 24% of the total coal production.

The extent of lignite production is generally not proportional either to total resources or to known economic reserves. Lack of energy alternatives is a strong motive to developments in lignite production. For example, after World War I, Germany's concern over secure fuel supplies and development of synthetic fuels accelerated studies with brown coal, and led to production of fuels based on synthetic fuel (synfuel) technology. South Africa, which has essentially no oil, has plentiful supplies of subbituminous coal, and synthetic fuel plants in Sasolburg and Secunda are based on coal. These synfuel plants are the largest in the world (see Fuels, synthetic). Other areas with limited alternatives are Victoria, Australia; southern India; and some eastern European countries.

When energy alternatives are available, a compromise between cost and quality is often realized. Blending of coals can be used to achieve more desirable qualities. For example, lignite from the former Yugoslavia has been blended with, and even substituted for, the highly caking Rasa coal used for coke production in the iron (qv) and steel (qv) industries.

Table 4. Worldwide Reserves and Production of Lignite[a]

	Resources, 10^6 tce[b], [c]			Production, 10^3 tce[c]			
Country	Proved in place	Proved recoverable reserves	Added recoverable reserves[b]	1980	1987	1989	1990[d]
Russia	110,000	100,000		79,970	82,500	82,000	188,000
Germany	102,000	56,150		116,141	123,017	121,356	357,000
Australia	46,000	41,900	183,000	10,629	13,509	15,604	48,000
United States	40,828	31,963		19,884	32,382	48,186	80,000
Indonesia		24,047					
China	37,200	18,600	34,700	9,359			
Yugoslavia	16,000	15,000		16,075	22,409	23,470	76,000
Poland	12,900	11,600	10,200	9,901	19,867	19,893	68,000
Mongolia	12,000			1,315	2,346	2,425	7,200
Turkey	7,705	6,986	235	6,197	18,648	16,228	43,000
Czech and Slovak FR	6,100	3,500	1,000	40,382	41,904	38,549	86,000
Hungary	5,465	2,883	1,124	8,152	7,368	6,440	5,400
Greece	5,312	3,000		4,341	8,527	9,640	52,000
Bulgaria	4,418	3,700		14,973	18,311	17,053	32,000
Romania	2,463	2,307	1,325	8,944	13,727	17,229	34,000
Iran	2,295						
India	2,100	1,900	3,932	1,501	2,762	3,267	9,500
Canada	1,615	2,827		10,239	15,967	17,666	9,400
New Zealand	1,556	9	28	137	43	80	200
Thailand	1,179	829	117	572	2,573	3,319	11,000
United Kingdom	1,000	500					
World total		*328,284*					*1,130,000*

[a] Refs. (3, 4, 24).

[b] Estimated values.

[c] 1 tce = 1 metric ton of coal equivalent = 29.3 GJ(2.78×10^7 Btu).

[d] Values are in units of 10^3 t.

In the United States and increasingly in other parts of the world, environmental regulations prohibit the combustion of all but very low sulfur-content coals without sulfur oxide emission controls. The cost of installing sulfur oxide control equipment together with concern about equipment reliability have led to the shipment of the lower rank low sulfur coals from up to 1600 km away from the mining site.

Especially in countries having planned economies, higher quality fuels are reversed for domestic heating, industrial fuels, and chemical feedstocks, and the lower quality lignitic coals for electric power generation. The high reactivity of lignites, coupled with relatively low cost at the mine, and the relatively lower cost of transporting product electricity or synfuels, favor the construction of facilities for lignite use that are adjacent to the mine site. In the United States, the Great Plains facility is an example (25).

4.1. Main Deposits and Production Areas

4.1.1. Europe and Russia

The eastern European reserves of lignitic coals provide the primary solid fuel for the eastern part of Germany, the former Czechoslovakia, Hungary, the former Yugoslavia, and Bulgaria. The importance of lignite as an energy source is great enough in Germany to permit long-range planning that includes removal and relocation of towns or villages situated on deposits in order to permit more complete recovery of the lignite resource. The mining is not considered complete until reclamation practices have demonstrated satisfactory crop production on the area that was mined (26).

Hard coal is more important in most of the western European countries with the exception of Austria and Italy. No lignitic coal production was indicated in 1989 for the Netherlands, Denmark, Belgium, Sweden, Norway, and the United Kingdom (24).

Much of the lignitic coal in Russia was laid down in the Lower Carboniferous age but was not covered deeply enough for the conversion to bituminous coal. Deposits in the Moscow basin, an area of ca 28,000 km^2, occur in discontinuous beds consisting of lenticular pockets of dull, laminated lignite. Some seams at the bottom of the deposit consist of a more consolidated bog-head coal. A wide range of coal ranks is found in the Kusnetz basin's largest field. Additional deposits are located in the eastern Urals (Sverdlov basin), in the central Urals (Kiselov basin), in the southern Ukraine, the Caucausus (Ahalzich district), the Bashkir Republic, the Kansk Atshinsky basin, the southern part of the Tungus basin, and the Far East (Artimov basin).

The German deposits are usually found in the southern part of the northern lowland and are mostly from the Eocene–Oligocene period. The two principal areas are the lower Rhineland and the central German fields. The deposits in central Germany are thick (ca 12 m) seams that are interconnected over large areas centered in the Leipzig–Halle area in the middle Elbe basin. The central German brown coal usually gives high yields of tar and coal extracts and is desirable for chemical processing. The coal in the Cottbus district has a lower bitumen and sulfur content and is used to make high temperature coke.

Coals of the western part of Germany usually occur in thinner seams and in more local areas. The main producing areas are west of Cologne with open-cast mines at Ville, Frechen, Garsdorf, and Frimmersdorf. This brown coal is important to electric power generation (27).

Reserves of high quality occur in Slovenia, Middle Bosnia, and Serbia. Larger quantities of lower quality coals are in West Slovenia (Velenj basin), northern Croatia (Zagorje basin), and eastern (Kolubara basin) and southern Serbia (Kosov basin).

Poland's deposits tend to be located primarily in the west as isolated, lens-shaped deposits. There are other deposits in the center of the country at Turow, Konin/Goslawice/Patnow, Turek/Adomow, and Rogozno. The production is used in power plants.

The highest quality lignite in central Europe is found in the former Czechoslovakia, where both open-cast and deep mining are used. The principal reserves are in the Eger Valley in northwest Bohemia, especially around Sokolov and Most. Other deposits are located at Grothau, Budweis, and Handlow. These deposits are being depleted at the greatest rate.

Hungary's coal production, which is mainly lignitic, is used for power generation. Coal is mined near Dorog in the western Bakony hills and in the northeastern hills. Bulgarian lignitic coal comes mostly from the Dimitrovo field (Pernik). Another field is near Dimitrovgrad (Maritsa basin). Greece's reserves, found mainly in the northwestern part of the country, are also used primarily for power generation.

4.1.2. North America

In the United States, lignite deposits are located in the northern Great Plains and in the Gulf states. Subbituminous coal is found along the Rocky Mountains. The western half of North Dakota has about 74% of the nation's resources, Montana 23%, Texas 2%, and Alabama and South Dakota about 0.5% each. The lignite resources to 914 m represent 28% of the total tonnage of all coal deposits in the United States. The lower cost and low sulfur content have contributed to rapid growth in production.

Overburden thicknesses in North Dakota range from 4–23 m, and seam thicknesses are 1–8 m. Clay partings split thick seams into several subseams. Some seams have been depleted by spontaneous combustion after exposure of the seam by erosion. Most of the U.S. lignite coal is woody, dark gray-black, splits readily along the bedding plane, and has bed moisture up to 40%, which dries to an equilibrium air-dried moisture of

15–20%. Sulfur content varies with location. Much of North Dakota lignite is low in sulfur and is used almost exclusively for electric power generation. Some, however, is used to fuel the Great Plains Gasification facility.

The lignite deposits of North Dakota and Montana extend into Canada as far as Saskatchewan. Canadian deposits are also located in Alberta, Yukon, the Northwest Territories, Ontario, and Manitoba. Production by open-cast mining, ca3.5×10^6 t in 1975, was 10.8×10^6 t in 1989.

4.1.3. Other Regions

In Australia, Victoria has the largest reserves, although smaller ones occur in southern Australia, western Australia, Tasmania, and Queensland (28). The main deposit consists of many thick seams in about 500 km^2 of the Latrobe Valley. The Yallourn coal field provides most of the fuel. The top coal seam is 65 m thick and is covered with 13 m of overburden. Most of this coal is used for electric power generation. It is very moist (55–72%), but has less than 5% ash. The lignitic coal found in South Australia is, for the most part, too deep for economic recovery. However, some of the better deposits are mined for power generation.

New Zealand's reserves are situated in the South Island. These are poorer and more dispersed than those in Australia. Production was about 80,000 t in 1989.

South America's reserves are small. In Chile, which has most of the South American reserves and production, only 22,000 t was produced in 1989.

In the Far East, total lignite production is a small percentage of the world total, but there are several significant areas. India's resources rank sixteenth in world size. The largest deposits are at Neyveli about 233 km from Madras. Poorer quality lignite is found in Kashmir. Pakistan's only reserve in production is at Jhimpir-Metig, about 97 km from Karachi. The largest producer in the Far East is the Democratic People's Republic of Korea (North Korea). The estimated North Korean production was 7.8×10^6 tce in 1989. Japan produced only 8000 tce in 1989. China's lignite resources are about 18.6×10^9 t, found mostly in the Northwest, not counting Mongolia. There are lesser amounts in the Southwest, and smaller amounts in the central southern region (3, 29).

No lignite production was reported in Africa. The only significant resources are in the Central African Republic and Nigeria. The latter deposit is unusual, having high hydrogen content, and coking provides unusually high yields of hydrocarbon-rich waxy tars.

4.2. Production

4.2.1. Mining

The mining or winning of lignitic coal typically involves deposits near the surface. The open-cast, open-cut, or strip-mining techniques employed involve mobile equipment built to provide a range of capacities to over $200,000\ m^3/d$. The rate of production can be increased rapidly, and the amount of labor per ton of coal mined is less than for underground mining. The quality of the coal, ratio of overburden thickness to seam thickness, stratigraphy, and distance to location of consumption are important in determining the cost to the consumer.

In modern practice, topsoil is first stockpiled for later application. The overburden, ie, sand, gravel, clay, etc, is then removed and the exposed coal is removed by bucket-wheel excavators, bucket-chain dredges, or draglines and shovels. Excavators having daily capacities of $2 \times 10^5\ m^3$ of overburden or coal have been built in Germany. These machines are 83 m high, 220 m long, and weigh 13,000 t. Plans for the Hambach mine near Cologne call for reaching a depth of about 500 m with an overburden:coal ratio of more than 6:1. The bucket-wheel excavator works with a stacker of similar capacity to move overburden. The coal usually is moved by a conveyor belt and later by electric locomotives in 2000 t lots (27).

Preparing for mining in the United States involves studying cores drilled initially on 1.6 km centers and later at $1.5 - 3$ cores/km^2 ($4 - 8$ cores/mi^2), depending on the occurrence of discontinuities. The actual mining takes into account the variation of deposit properties in terms of percent moisture, ash composition, ash content, and grindability as a function of moisture (9).

Seam thicknesses and depths vary tremendously. The most favorable deposits have shallow overburdens and thick seams that cover large areas. Acceptable stripping ratios, ie, overburden thickness to coal thickness, depend on the quality of the fuel. Ratios up to 10:1 have been used for bituminous coals, but lower ones are required for lignitic coals because of the lower heating value per unit weight.

A variety of measures must be taken to assure safe and continued operation. Because the natural water table is higher than the coal seams, or the seams are natural aquifers, it is necessary to pump water out of the pit or to drill wells around the mine and pump to reduce the water table. The Rheinische Braunkohlenwerke (Rheinbraun) pumps water at a rate of $1 - 1.2 \times 10^9\ \mathrm{m^3/yr}$. Part of this water is processed to provide drinking water for Neuss and Dusseldorf. The tendency of lignite to ignite spontaneously requires care in the amount of face that is exposed, especially in naturally dry, hot, windy climates.

4.2.2. Storage

Concern about spontaneous ignition has led some operators to try to match the mining and consumption rates, so that there is little if any reserve, as in minemouth power generation stations. When the coal must be stockpiled, careful stacking minimizes oxygen reaction and overheating. Uniform stacking in layers no more than 0.3 m thick avoids segregation of particle sizes, then compacting using earth-moving equipment, and covering the pile with finer material limits oxygen penetration, overheating, and ignition. By sloping (14°) the sides gradually, segregation is prevented and compaction is improved (30).

A smooth coal pile surface, coupled with the gradual slope, minimizes the differential wind pressures and consequent oxygen penetration. A $4 - 6 \times 10^6\ \mathrm{t}$ lignite stockpile from the excavation for the Garrison Dam in North Dakota has been stable for many years as a result of this storage method.

To limit drying, spraying with cold water is useful. The spraying can be coupled with a straw covering. Four Hungarian stacks covered with 10 cm of straw decreased in heat value only 0.4–6% after spraying periodically for 10 months, but lost 6–20% if unsprayed. Underwater storage is sometimes used in drier climates such as Australia. Processed fuels, eg, briquettes, can be stored without difficulty since they are less permeable to air, and, depending on process conditions, are less oxidizable.

4.2.3. Transportation

For short distances from the mine, transportation (qv) is by truck or conveyor belt. Rail transportation is generally used for greater distances. Slurry pipelines (qv) are being considered as an alternative. Rail transport over hundreds of kilometers results in loss of surface material in uncovered cars and a tendency to overheat in bottom-dumping rail cars owing to air infiltration around the cracks (31). Proper sealing and covers permit shipping over hundreds of kilometers.

4.2.4. Drying

In many cases, the high moisture content of young coals dictates significant drying (qv) before use. In some cases, partial removal of mineral matter, especially water-soluble species, is desirable.

Drying can be accomplished by evaporative, hydrothermal, or other thermal processes. Evaporative drying reduces the moisture content substantially, but increases the tendency to spontaneous combustion and decrepitation, ie, breaking up and cracking. Power plants use combinations of heated shafts and hot flue gas passed through the size reduction (qv) equipment to remove moisture (10). Hydrothermal, ie, nonevaporative, drying or dewatering (qv) was originally developed in the 1920s and has been continually refined. The Fleissner process is used commercially at a $250,000\ \mathrm{t/yr}$ plant in Kosovo, in the former Yugoslavia. This approach removes a large part of the water in a low rank coal in the liquid form, without using the energy necessary to evaporate this water. Also, a notable part of the water-soluble inorganic species, such as sodium salts, can be removed with the expressed water, reducing the tendency for fouling the boiler during combustion. The process water is acidic (pH 3–4), and rich (up to 4500 ppm) in phenols. Hydrothermal treatment is carried out at as low a temperature as possible to remove the water without increasing the need for water treatment.

Thermal drying has been studied in conjunction with a rail shipment of ca 1200 km from North Dakota to Illinois. Oil was applied at $6 - 8$ L/t to suppress dust loss, and cracks around the doors in the base of the car were sealed to prevent ignition. Stable shipment and stockpiling were then possible (31). Thermal drying may be carried out to further reduce the moisture content as required for briquetting or for more efficient pulverizing and combustion.

5. Health and Safety Factors

Because lignite mining is carried out by surface methods, the hazards associated with underground mining typically do not exist (see Coal). The principal hazards involve the tendency of the coal toward spontaneous combustion as the coal dries, especially at the exposed seam. Thus careful planning and continued reclamation efforts are required to cover faces. Adequate water for revegetation has been of some concern in the arid areas of the northwestern United States where vegetative growth is slow and reclamation is expected to take many years.

The lignitic coals of the northern United States tend to have low sulfur contents, making them attractive for boiler fuels to meet sulfur-emission standards. However, low sulfur content coals have impaired the performance of electrostatic precipitators. The ash of these coals tends to be high in alkaline earths (Ca, Mg) and alkalies (Na, K). As a result, the ash can trap sulfur as sulfites and sulfates (see Air pollution control methods).

Some North Dakota lignite ashes have also been observed to have above-average concentrations of uranium (21, 22), leading to interest in processing the ash for uranium recovery. However, this ash may be classified as hazardous.

6. Economic Aspects

The price of lignite per mined ton or per heat unit is lower than that for higher rank coals. The market for all coals is primarily as boiler fuel for electric power production. Prices are generally established by contracts between utility and supplier before mining begins.

Because of its low sulfur content, lignite is becoming more important. The U.S. Clean Air Act Amendments of 1990 have resulted in economic premiums for low sulfur coal corresponding to \$10/t for emission allowances at \$500/t of SO_2 (32).

7. Uses

Most of the world's coal supply is used for combustion to generate steam for electric power production. This is especially true of lignitic coals. Other uses for lignite, such as briquetting, for domestic and industrial fuels; carbonization, to provide coke and liquid by-products; gasification, to provide gaseous fuels; chemical feedstocks, for making fertilizers and other liquid fuels; and direct liquefaction are being developed.

7.1. Briquetting

Lignite briquettes have long been preferred over unprocessed brown coal for residential and industrial heating (10, 33, 34). The extrusion press has been used for the production of most of the briquettes made from lignite coals since 1858. Design improvements have provided for multiple pressing, and for different channels, feeding methods, and press drives, all of which have significantly increased output. The demand for briquettes has been dropping as other fuels are used for residential heating. About 75% of the carbon originally present in the lignite appears in the product briquettes. A typical briquetting plant produces surplus electric power equivalent to a few percent of the energy content of the original lignite.

Plants for briquette production exist in the eastern part of Germany, Australia, and India. German transport costs per unit of heating value are about 40% less for briquettes than for lignite.

Briquetting of coal may be carried out with or without the addition of a binder. Binderless briquetting, the predominant method, is restricted to the relatively soft, unconsolidated lignites. The coal size distribution and moisture content must be carefully regulated for successful agglomeration and pelletting. To obtain close packing of the material during compression, the coal is generally crushed to a size below 4 mm, where 60–65% is <1 mm. For maximum briquette strength, moisture contents are adjusted to optimum (between 11 and 18%) values which vary with the coal seam (5). At higher moisture, briquettes shrink and crack on equilibrating with the atmosphere, and at lower values the briquettes may swell and weaken.

After crushing, the lignite is dried thermally in a rotary dryer and cooled slowly to achieve uniform moisture distribution in the particles. When the temperature reaches the optimum range of 38–65°C, the lignite is pressed at ca 138 MPa (2000 psi). A reciprocating piston rams the coal through an increasingly restricted channel during the forward stroke and loads more lignite during the return strokes. A final gradual cooling is required. Pressing increases the briquette temperatures as much as 30°C internally and more at the surfaces. The commonly used Exter press forms $20 \times 6 \times 4$ cm briquettes, each weighing ca 550 g. The moisture in the coal is the binder and forms hydrogen bonds between the polar functional groups (5).

Harder, more mature coals generally have lower plasticity and greater elasticity. These coals are briquetted at higher pressures using a binder in ring-roll presses to give a harder carbonized product. Because briquettes have been used primarily for domestic heating and metallurgical and chemical processes, the binderless coking process has been predominant.

Briquettes must be transported carefully to avoid breakage. They are usually dumped into piles in sheds and frequently are screened to remove smaller broken particles. Hand-stacking significantly improves storage quality, and permits more material to be stored in a limited volume.

Briquettes have a heating value of $16.7 - 23.4$ MJ/kg $(7,200 - 10,000$ Btu/lb$)$ or from 2–3 times the value of a typical brown coal primarily because of the moisture loss.

7.2. Combustion

The combustion of lignite is used to generate energy steam for turbine generators to provide electric power. Most modern plants have a capacity of at least 100 MW, and frequently as much as 600 MW (35–38). A brown coal boiler needs to be 1.5 times larger than a bituminous coal boiler to produce the same amount of power because of the larger amount of inert materials therein, eg, water vapor and recycled flue gas. The fuel feed rates must be up to three times as great as for hard coal because of the high moisture content in the brown. The resulting higher capital costs are, however, offset by lower fuel costs.

The formerly common grate-supported combustion is used in some smaller units, but the dominant method of combustion involves suspension or pulverized coal firing. The high reactivity of lignite permits burning of coarser (65–70% mm (200 mesh)) particles than those of bituminous coal for a given furnace residence time. The average residence time of a coal particle in a large boiler may be 3–4 s, of which 0.25 s is required for combustion (see Furnaces, fuel–fired).

The relative reactivities of lignite and other coals have been the subject of many studies. The burning profile or rate of weight loss as a function of temperature or time for a coal sample heated at a constant rate in air is shown in Figure 3 for a number of coals (39). The initial weight loss owing to moisture release is followed by a more significant weight loss resulting from oxidation of the bulk of the organic substance. The onset of oxidation occurs at a lower temperature for lignite than for other coals. Thus, the oxidation reaction is complete

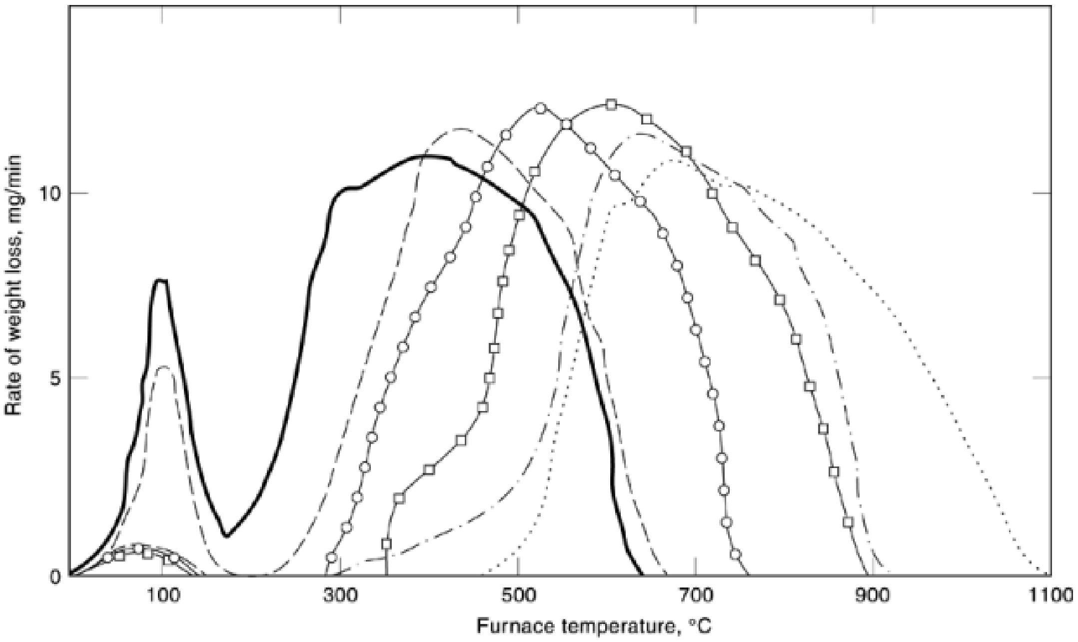


Fig. 3. Comparison of burning profiles for coals of different ranks where (…) represents anthracite; (—·) semianthracite; (□) LV bituminous; (∘) HV bituminous; (–) subbituminous; and (—) lignite. LV=low volatile; HV=high volatile.

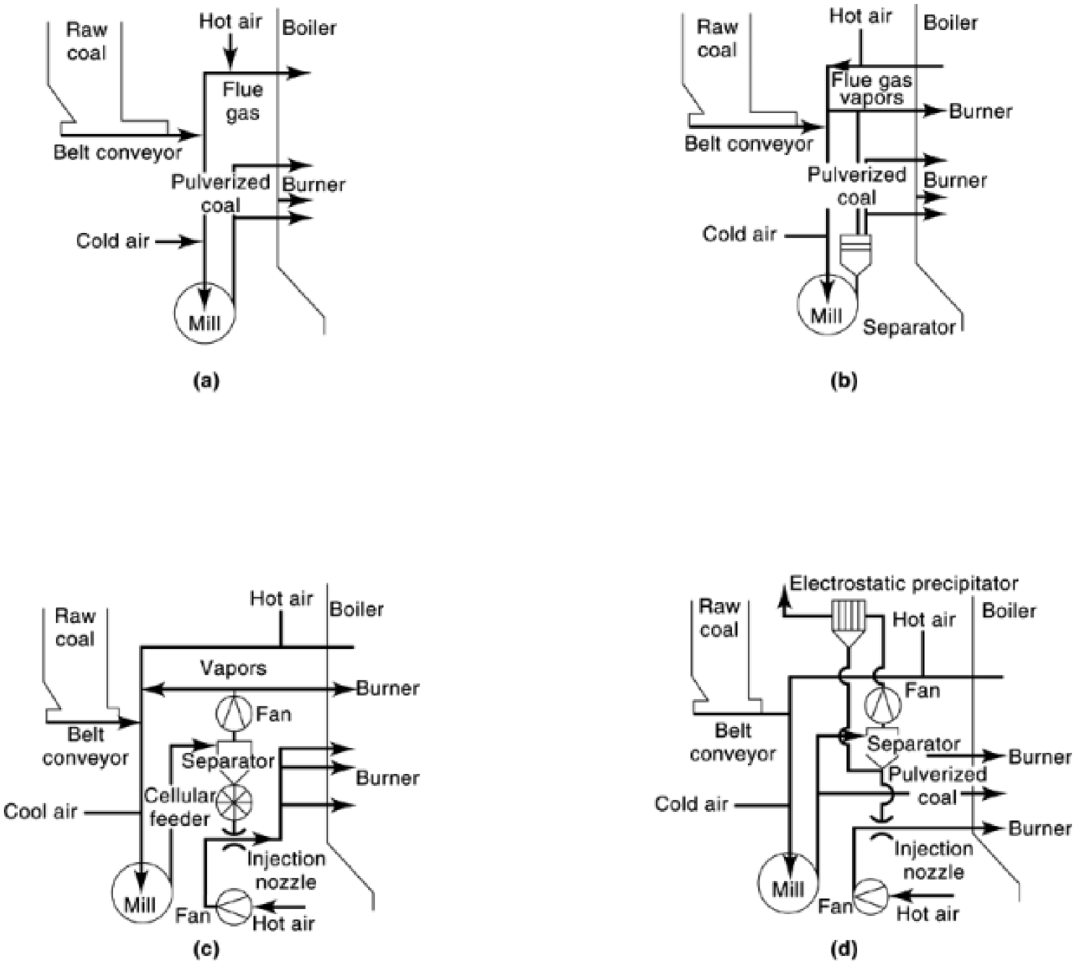
at a lower temperature because of the greater ease of lignite ignition. The completion temperature relates to residence time or size of the furnace cavity required for complete combustion.

Because of the wide variation in composition and properties of brown coal (see Table 3), efficient combustion of these fuels cannot be accomplished by a single system. The moisture content limits combustion efficiency because some chemical energy is required to convert liquid water to steam in the flue gases. The steam then increases the dew point of the gases, requiring higher temperatures to avoid condensation in the stack. For fuels up to 25% moisture content, 80% efficiency can be achieved. As the moisture content increases to 60%, the efficiency decreases to 70% and efficiency continues to decline about another 1% for each additional 1% moisture to 70%.

Most utility boilers are of the dry-bottom (solid ash) design, although a few cyclone-burner, slag-tap design stations are in operation in the United States. A variety of firing systems have been developed to partially dry the lignite before combustion (Fig. 4) (19). The Niederaussem plant (Fig. 4**a**) uses a stream of hot flue gas to dry the raw coal before pulverizing, then cold air is added through the pulverizer and all of the products are fed into the boiler. The Megalopolis station (Fig. 4**d**) uses hot flue gas to dry the lignite. A cyclone separator and electrostatic precipitator permit rejection of some of the water vapor to the atmosphere rather than to the boiler. Another drying method uses a vertical shaft, heated by combustion gases, for partial drying prior to grinding.

A high moisture content necessitates finer grinding of coal for rapid water release. For such coals, the classifier in the pulverizer is set to return more of the oversize material for further grinding.

Particulate removal following combustion is accomplished by electrostatic precipitators, which depend on the particles accumulating sufficient electric charge. Charge has been associated with the sulfur oxide content of the flue gas. The higher the SO_x, the more charge present. Larger precipitators must be used for low sulfur



(a) (b) (c) (d)

Fig. 4. Firing systems for lignitic coal. (**a**) Niederaussem, Germany: 600 MW; heating value (HV)=7.7–10.7 MJ/kg; moisture: 52–57%; ash content: 2–13%. (**b**) Yallourn, Australia: 350 MW; HV: 4.2–8.8 MJ/kg; moisture: 58–73%; ash content: max 3%. (**c**) Tuncbilec, Turkey: 150 MW; HV: 8.4 MJ/kg; moisture 22–24%; ash content: 42%. (**d**) Megalopolis, Greece: 125 MW; HV: 3.8 MJ/kg; moisture: 64%; ash content: 13%. To convert MJ/kg to Btu/lb, multiply by 430.2.

coals. Alternatively, an additive that can be adsorbed and readily ionized, eg, SO_3, can be injected upstream of the precipitator to improve the performance of existing units.

The ignition of moist lignitic coal requires up to five times the energy required for ignition of bituminous coal. This necessitates preheating, firing techniques, control of air supply, and preignition grates on chain-grate stokers. For grate firing, the lump coal must be evenly sized and distributed to allow uniform air flow and combustion. The disintegration of soft coal on heating produces a mixture of fine and lump coal, leading to flow resistant masses and some losses of incompletely burned fuel. The ash of some (German) coals tends to maintain the shape of the coal pieces; the small amount of ash from Australian (Victoria) coal does not.

Briquettes burn similarly to bituminous coal, although some tend to disintegrate on combustion. A low $(<6-7\%)$ ash content increases the possibility of disintegration. Normal combustion depletes the combined oxygen and volatile matter in the coal quickly, effectively changing its composition and combustion behavior, making control of combustion difficult.

During combustion, the alkali content (CaO, MgO, K_2O, Na_2O) of lignitic coals is released. Alkalies are normally sufficient to keep the sulfur in the coal during wet scrubbing (40). Sulfur capture is dependent primarily on the sodium, calcium, and the alkali:sulfur ratio (10, 41). Loss of retention occurs if the silica and alumina content is high, because an unreactive alkali aluminosilicate is formed, complexing the alkali. As sulfur emission standards become more stringent, supplemental limestone may be required in some cases.

7.3. Pyrolysis

Heating in the absence of oxygen releases moisture at low temperatures, carbon dioxide at temperatures $>200^\circ C$, and a variety of gaseous products at very high temperatures. Acid washing of the raw coal is used to remove extractable cations, followed by treatment with selected cations. Yields of CO_2, CO, CH_4, H_2, and H_2O depend on the amounts of inorganic species in the coal (42).

7.4. Carbonization

Low temperature carbonization of brown coal, usually as briquettes, was originally used to produce a tar and oil mixture that could be further processed to motor fuels, and later to produce a low heat–energy heating gas and a solid smokeless fuel. High temperature carbonization of lignite briquettes produces a coke that could be used to supplement coke from metallurgical-grade bituminous coal. Typically, the gas by-product was burned for steam generation and for some electric power generation (10, 43–46).

7.4.1. Low Temperature Carbonization

The Lurgi Spülgas process was developed to carbonize brown coal at relatively low temperatures to produce tars and oils (Fig. 5). A shaft furnace internally heated by process-derived fuel gas (Spülgas) is used. The product can range from a friable coke breeze to hard lump coal depending on the quality of the briquettes used in the feed. The briquettes, made in normal extrusion presses, break down into smaller sizes during carbonization.

The briquettes are distributed over bunkers and dried for about 5 h at 150°C (0.5% moisture), then descend to the lower carbonization zone for another 5 h at 500–800°C. Heating is provided by burning the purified product gas with air using from 35–48% of the gas. The rest usually is used for steam generation. The char (product) is cooled by circulation of product gas. The char reactivity increases with time and must be stored carefully.

When denser briquettes are used for carbonizing, the product has 10–12% volatile matter, up to 20% ash, and a heat value ca 25 MJ/kg (11, 000 Btu/lb). Overall yields for normal briquette carbonization are char 45%, tar 12.5%, and gas 130 m^3 (4600 ft^3) having a gross heating value of 8.4 MJ/m^3 (225 Btu/ft^3). The char was ca 30% above 20 mm, used for domestic and central heating; 50% from 6–20 mm, used for gasification; 30% less than 6 mm, used for steam generation and gasification. About 270 Lurgi Spülgas plants exist, mainly in Germany, Australia, India, and the Eastern European countries, and having a coal input of 10^5 t/d. Fluidized-bed and entrained carbonization of lignite have been studied but have not been commercialized (see Fluidization).

7.4.2. High Temperature Carbonization

Some lignitic coals can be blended with caking coals in coke production, primarily to produce a coke or coke breeze for iron ore reduction, or for carbide or phosphorus(qv) production (see Carbides). In Germany, small briquetted material under 1 mm is carbonized to 950°C for about 12 h. After dry cooling, the product coke is screened from 45 mm to <3 mm. A 24-retort battery has an output of $125 - 130$ t/d (42% yield), more than half of the coke is large and strong enough to be used in low shaft furnaces. The rest is used in gasifiers, lime producers, and carbide furnaces. The large coke is mixed with conventional coke for iron ore smelting. The CaO/SiO_2 ratio is high enough to reduce typical lime additions.

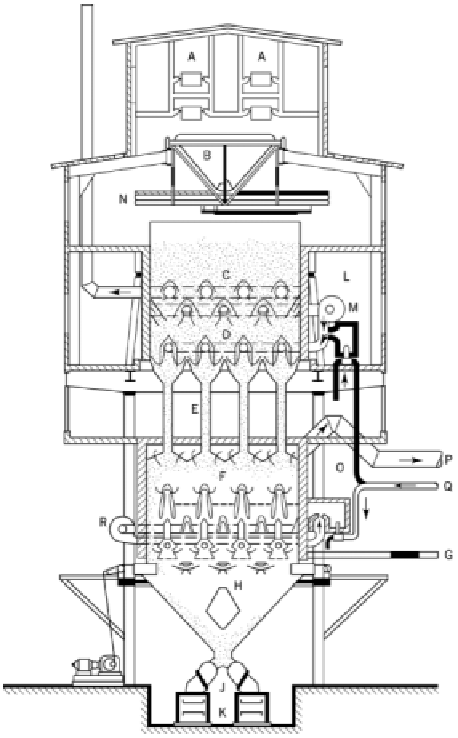


Fig. 5. Diagram of large Lurgi Spülgas carbonization plant: A, coal conveyor; B, movable distributor; C, coal bunker; D, drying zone; E, connecting shafts; F, carbonization zone; G, coke-cooling zone; H, coke extractors; J, coke hopper doors; K, coke conveyor; L, circulation fan for drying zone; M, combustion chamber for drying zone; N, stack for waste gases from drying zone; O, combustion chamber for carbonizing zone; P, offtake for mixture of carbonization gas and Spülgas; Q, intake for tar- and oil-free cooled Spülgas; R, fan for gas for coke quenching.

7.5. Gasification

Gasification converts solid fuel, tars, and oils to gaseous products such as CO, H_2, and CH_4 that can be burned directly or used in synthesis gas (syngas) mixtures, ie, CO and H_2 mixtures for production of liquid fuels and other chemicals (47, 48) (see Coal conversion processes, gasification; Fuels, synthetic–gaseous fuel; Hydrogen).

The Lurgi process has been the most commercially accepted gasification method since its commercialization in 1936, and is used in the large plants in South Africa, in modified designs in Germany, and in the United States for the Great Plains facility (25, 49, 50).

The gasification process includes coal preparation (crushing or pulverizing), charging into a gasifier (using special equipment if the gasifier is pressurized), quench of the hot product gas, shift reaction or adjustment of the $CO:H_2$ ratio (depending on the product requirement), gas purification (removal of sulfur-bearing species, typically H_2S, and often CO_2), and catalytic conversion if a special product such as substitute natural gas (SNG) or a liquid fuel is desired. For fuel-gas production, the shift, CO_2 removal, and catalytic conversion can be eliminated. The Lurgi gasifier shown in Figure 6 is a fixed-bed reactor into which coal is charged through lockhoppers at the top. Steam and oxygen mixtures enter at the bottom, react with the coal at ca 3.0 MPa (30 atm) pressure in a countercurrent mode. Product gas is removed at the top and quenched (25).

One of the largest plants began operation in 1984, near the largest (12.2×10^6 t/yr) coal mine in North Dakota by the Dakota Gasification Co. In 1988 average production was 4.2×10^6 m^3/d (158×10^6 ft^3/d) of

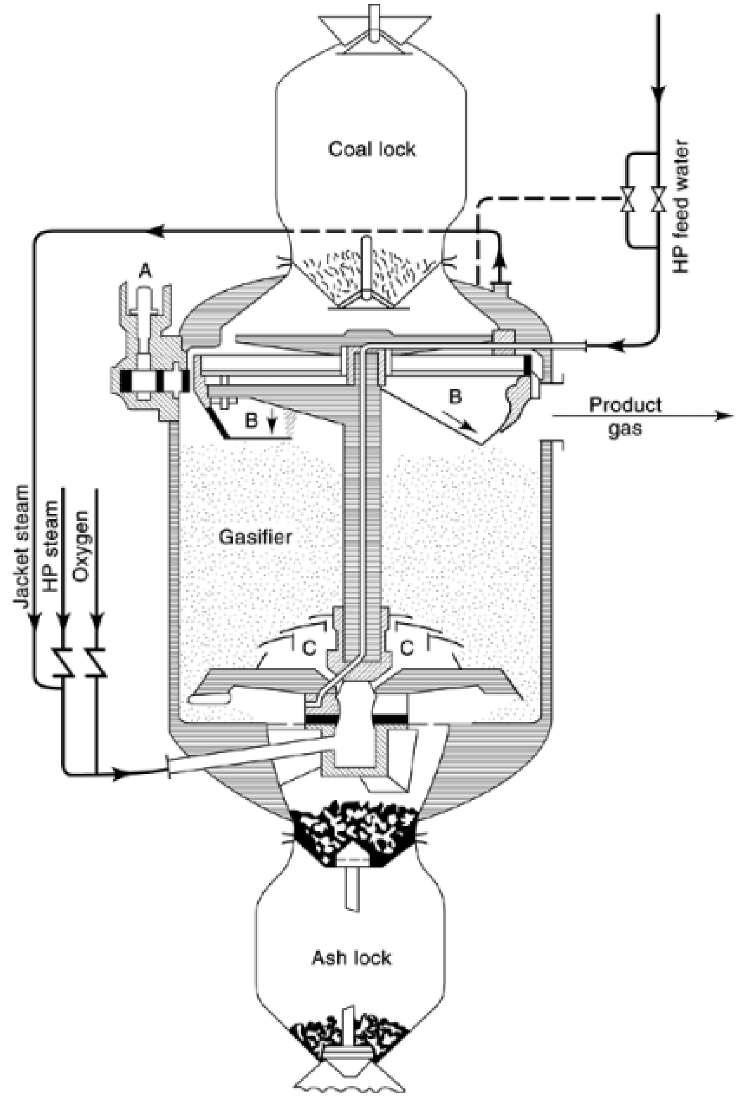


Fig. 6. Lurgi pressure gasifier: A=variable speed drive; B=coal distributor; C=rotating grate. HP=high pressure.

pipeline-quality gas (CH_4) (25). About 27, 500 t/d of lignite are mined and crushed to pass a 5 cm screen. Slightly more than half (56%) is fed to the gasifiers, and the remaining fines are sold for boiler fuel. The plant consumes 2, 800 t/d of oxygen and about 13, 000 t/d of steam. The accompanying oxygen (qv) plant is the largest in the country. In 1990 a plant to recover krypton and xenon was added.

Fourteen Lurgi Mark IV (4 m) gasifiers are in operation at 3.07 MPa (430 psig). About 30% of the gas stream is shifted to provide the 3:1 overall H_2:CO ratio needed for catalytic conversion to methane, CH_4. The acid gases, CO_2 and H_2S, are removed using a Rectisol unit. The gas stream is catalytically converted to methane, dried, and compressed to about 9.9 MPa (98 atm) with a gross heating value of 36.4 MJ/m^3 (977 Btu/ft^3). The Phosam process recovers 113 t/d of anhydrous ammonia (qv) which is sold, and the tars, oils, phenols, and naphtha are used for boiler fuel. The Sulfolin process (Linde AG) recovers 106 t/d of sulfur from the gaseous products (90% from the gases fed to the process).

The British Gas Corp./Lurgi slagging gasifier has been tested in Scotland (300 t/d unit). This method is designed to achieve higher throughputs. Lower costs are expected from operating at temperatures high enough to melt the coal ash, after which it flows out the bottom of the gasifier as a molten slag.

The first commercial fluidized-bed systems used Winkler units in which lignite and its char are gasified at atmospheric pressure with air or oxygen and steam mixtures. Improvements led to high temperature Winkler (HTW) units, being tested in a 1 t/h pilot plant in Germany. A number of fertilizer plants use lignite in the Koppers-Totzek gasifier, an entrained or suspension gasifier in which pulverized coal reacts with a steam–oxygen mixture to produce synthesis gas and ammonia. The ash is removed as a molten slag.

7.5.1. Methods in Development

The U.S. Department of Energy (DOE) and the American Gas Association (AGA) have sponsored the development of more efficient gasification processes, but as of this writing the plentiful supply of inexpensive natural gas has precluded commercialization.

A medium heat (11.2 MJ/m^3 (300 Btu/ft^3)) gas for industrial application has been produced at Westinghouse Electric's pilot plant in Waltz Mill, Pennsylvania (50), where lignite is gasified in a pressurized fluidized bed which uses oxygen and steam.

The HYGAS process has been developed by the Institute of Gas Technology under AGA/DOE sponsorship for SNG production in a 3 t/h pilot plant in Chicago, Illinois. The coal (lignite or higher rank coals), prepared as a recycle-oil slurry, is pumped into the top of the gasifier. The slurry is then dried, and the coal contacts a rising stream of H_2 and other gases to go through rapid-rate methanation. Operation at pressures to 7 MPa (1000 psi) permits over 50% of the methane to be made in the gasifier, reducing the downstream processing requirements. The residual char is further gasified in a lower fluidized bed. The resultant char is gasified using a steam–oxygen mixture in the lower fluidized bed. Coal-to-methane conversion efficiencies are about 75%, but this method has not been commercialized.

The CO_2 Acceptor process, also developed under AGA/DOE sponsorship, by the Consolidation Coal Co., uses steam to gasify lignitic coal. Heat is supplied by the exothermic reaction between CO_2 and calcined dolomite [17069-72-6]. The dolomite is calcined in a separate fluidized bed. This process operates in a 40 t/d pilot plant, but there are no plans for commercialization as of this writing.

Underground gasification of lower rank coals has potentially lower gas costs and reduced mining hazards. High pressure air is used to increase permeability. Tests of this technology have been carried out in Texas under sponsorship that included the Texas Utility Commission. Moscow-region coals have been gasified underground since 1947. Coal beds are up to 5 m thick, but these cannot be gasified when only 1 m thick.

7.6. Liquefaction

The synthesis gas or hydrogen required for liquid fuels is produced by a process known as indirect liquefaction; ie, gasification to produce synthesis gas followed by catalytic conversion to liquid products, or by direct hydrogenation of lignite (see Coal conversion processes, liquefaction). The Texaco partial oxidation process uses a slurry of coal in a high pressure gasifier (7 MPa (70 atm)) in the presence of oxygen. The slurry enters at the top of the reactor and is gasified as the coal descends. Ash is quenched in a water bath at the bottom. This gasifier has been successfully used in a number of plants.

The U-GAS process has been developed by the Institute of Gas Technology under DOE sponsorship with a 1 t/h pilot plant in Chicago. The gasifier is a single-stage fluidized bed able to accept lignitic or raw bituminous coal to produce a synthesis as at 1000°C. A unique feature is a venturi (short tube with tapering construction in the middle) at the base of the bed, which permits the discharge of ash agglomerates with high carbon utilization. The technology is being commercialized by Enviropower Inc. (Finland).

7.7. Liquid Fuels

Liquid fuels can be obtained as by-products of low temperature carbonization; by pyrolysis, solvent refining, or extraction; and gasification followed by catalytic conversion of either the coal or the products from the coal. A continuing interest in liquid fuels has produced activity in each of these areas (44–46). However, because crude oil prices have historically remained below the price at which synthetic fuels can be produced, commercialization awaits an economic reversal.

The conditions of pyrolysis either as low or high temperature carbonization, and the type of coal, determine the composition of liquids produced, known as tars. Humic coals give greater yields of phenol (qv) [108-95-2] (up to 50%), whereas hydrogen-rich coals give more hydrocarbons (qv). The whole tar and distillation fractions are used as fuels and as sources of phenols, or as an additive in carbonized briquettes. Pitch can be used as a binder for briquettes, for electrode carbon after coking, or for blending with road asphalt (qv).

Tars can be hydrogenated to produce liquid fuels. High hydrogen and low asphaltene, ie, benzene-soluble and pentane-insoluble, contents are desirable. The central German brown coals are attractive for this reason. The tars from the eastern part of Germany require much lower pressures and less hydrogen per unit of product than do brown coals near Cologne, which can require pressures up to 71 MPa (700 atm) (see Petroleum).

7.8. Integrated Projects

Large integrated complexes are employed to process lignitic coal into a variety of fuels and chemicals. Examples of these exist in South Africa, Germany, and India (43, 49, 51, 52) (see Fuels, synthetic). The South African complexes at Sasolburg and Secunda, in operation since 1982, were planned to provide liquid transportation fuels from local coals. These plants draw coal from a 300 km^2 coal field, which is expected to last until at least 2060. The subbituminous coal has a gross heating value (dry basis) of 23.9 MJ/kg $(10,300\ \mathrm{Btu/lb})$. The composition is ash 21.5%, sulfur 1.3%, carbon 79.67%, and hydrogen 4.3% on a dry ash-free basis. The seam is almost horizontal and has a range of thickness of 2–7 m at 100–200 m depth. It is mined using essentially continuous and longwall mining techniques. Belt conveyors take the coal to the adjacent complex. Total production from the six mines in the complex is more than 32×10^6 t/yr (49). Annual coal consumption for Sasol II is about 13×10^6 t, two-thirds of this for gasification, and the remainder for steam and electric power generation.

Fine coal is not acceptable as Lurgi-type gasifier (Fig. 6) input. Power generation capacity of 240 MW was selected to utilize the fines produced during crushing and handling of the coal. Oxygen (8600 t/d) is needed for the 36 Lurgi 4.0 m diameter gasifiers. High pressure steam requirements are 1230 t/h. Raw gas production is about 1.65×10^6 m^3/h $(1.4 \times 10^9\ ft^3/d)$. After quenching, this gas is fed to a Rectisol (cold methanol) purification plant to provide ca 1.2×10^6 m^3/h $(1 \times 10^9\ ft^3/d)$ of pure gas (0.07 ppm S). The pure gas composition is about 1.5% CO_2, 84.1% H_2 + CO, 13.5% CH_4, 0.5% N_2, and 0.4% C_nH_m. An adjacent oxygen plant consists of six units of 2300 t/d capacity, each at 3.45 MPa (500 psi). Steam generation involves six boilers producing 540 t/h of 430°C, 4 MPa (580 psi) steam each. The Rectisol plant discharges H_2S to a Claus unit which produces 99.97% pure sulfur (49) (see Sulfur removal and recovery).

The purified raw gas goes to a Synthol (Fischer-Tropsch) unit for catalytic conversion of CO and H_2 to liquid fuels. The tars and oils obtained from quenching the raw gas from the gasifiers go to a Phenosolvan plant to provide tar products for the refinery and ammonia for fertilizer. The Synthol plant has seven reactors, each with 1.9×10^6 m^3/h $(1.6 \times 10^9\ ft^3/d)$ gas feed. Annual plant production is 1.5×10^6 t motor fuels, 185×10^3 t ethylene, 85×10^3 t chemicals, 180×10^3 t tar products, 1×10^5 t ammonia (as N), and 9×10^4 t sulfur, for a total of 2.14×10^6 t/yr of valuable products.

Another integrated project is the VEB Gaskombinat Schwarze Pumpe plant in eastern Germany. This complex consists of three briquetting plants, three power stations, one brown coal high temperature coking plant, and one pressure gasification plant. Open-cast mines near Welzaw-Sud and Nochten produce soft brown coal. The overburden ratio is about 5.2:1. The equipment can remove up to 60 m of overburden and has a capacity of 20, 000 m^3/h (1.7 × 10^8 ft^3/d). The raw brown coal goes to both the briquetting plants and power stations. Some briquettes are produced directly for fuel, others go to the coke-oven plant to produce coke and liquid products or to the pressure gasification plant to produce gas and liquid products. The power station generates both electric energy and steam which is used on-site. The fuel gases from the coke-oven plant and the oil and coal gasification plants are blended, after purification, with product gases from the natural gas reformer to provide town gas. The capacity is estimated at 13 × 10^6 m^3/d (450 × 10^6 ft^3/d) town gas which has a heat content of 15.9 MJ/m^3 (ca 430 Btu/ft^3). The composition of the town gas is 1.3% CO_2, 12.3% CO, 0.84% O_2, 22.9% CH_4, 34.4% H_2, 26.2% N_2, 0.94% C_2H_6, 0.53% C_3H_8, 0.56% C_4H_{10}, and 0.03% C_2H_4. This gas is obtained by blending natural gas from eastern Germany and Russia, nitrogen (qv) from the adjacent air-separation plant, and reformed gas with the purified fuel gas stream from the plant.

Two air-separation plants provide oxygen to the gasifiers, 24 units with a 3.6 m internal diameter. These gasifiers, of a design similar to the Lurgi fixed-bed type, are located in an open-air structure for safety. After gasification of the briquettes, the raw gas is quenched to remove tars, oil, and unreacted fine particles. The product gas from the gasifiers and coke-oven plants is purified with refrigerated methanol. A Claus unit converts H_2S to elemental sulfur.

Operating parameters of this German plant, on the basis of one cubic meter of raw gas, include 0.139 m^3 O_2, 0.9 kg briquettes, 1.15 kg steam, 1.10 kg feed water, 0.016 kWh, and 1.30 kg gas liquor produced. Gasifier output is 1850 m^3/h and gas yield is 1465 m^3/t dry, ash-free coal. The coal briquettes have a 19% moisture content, 7.8% ash content (dry basis), and ash melting point of 1270°C. Thermal efficiency of the gas production process is about 60%, limited by the quality and ash melting characteristics of the coal. Overall efficiency from raw coal to finished products is less than 50%.

In the plant at Neyveli, India, the clay and sand overburden is used to make china clay, and the lignite is used for power generation, for gasification to provide feedstocks for fertilizer production, and for briquette production with by-product light oils. The mine area is 14 km^2 (5.41 mi^2), the recoverable lignite is ca 180 × 10^6 t, the overburden average thickness is 62 m, and the average lignite thickness is 13 m.

Power generation using pulverized coal produces up to 250 MW for export and 400 MW for internal consumption. Crushed lignite, dried to 8% H_2O, is gasified in Winkler generators with steam and oxygen. Ammonia is made from H_2 and N_2 from the air-separation plant. Further reaction of the NH_3 with CO_2 produces urea [57-13-6] for fertilizer. Extruded briquettes are formed by low temperature carbonization of crushed 12% H_2O coal. Lurgi Spülgas low temperature carbonization makes carbonized briquettes, light oils, and fuel for power generation.

7.9. Products from Synthesis Gas

Steam–oxygen gasification of coal produces syngas, a mixture of carbon monoxide [630-08-0] and hydrogen [1333-74-0]. Increasing pressure increases the amounts of methane [74-82-8] formed. This mixture can be used for a variety of products by Fischer-Tropsch synthesis. A 2:1::H_2:CO mixture over iron-based catalysts produces hydrocarbons and some alcohols, suitable for motor fuels. A similar gas mixture over zinc or copper-based catalysts yields methanol [67-56-1]. By the water gas shift reaction, a hydrogen stream can be obtained from the synthesis gas. The hydrogen can be used as a chemical feedstock in refineries or synthesis plants, such as those for ammonia. The Lurgi plants in South Africa are based on this technology (51, 52).

In 1974 a 1000 t/d ammonia plant went into operation near Johannesburg, South Africa. The lignitic (subbituminous) coal used there contains about 14% ash, 36% volatile matter, and 1% sulfur. The plant has six Koppers-Totzek low pressure, high temperature gasifiers. Refrigerated methanol (−38°C, 3.0 MPa (30 atm)) is used to remove H_2S. A 58% CO mixture reacts with steam over an iron catalyst to produce H_2. The carbon dioxide is removed with methanol (at −58°C and 5.2 MPa (51 atm)). Ammonia synthesis is carried out at ca 22 MPa (220 atm) (53) (see Ammonia).

7.10. Direct Hydrogenation

Direct hydrogenation of lignitic and other coals has been studied by many investigators. Lignite can be slurried with an anthracene-oil solvent, heated to a temperature of 460–500°C with 1:1 $CO:H_2$ synthesis gas at pressures to 28 MPa (280 atm) in a 2 kg/h reactor. The product liquids are separated, and in a commercial process, a suitable hydrogen-donor solvent would be recycled (54).

In a 1 t/d pilot plant utilizing the Exxon Donor Solvent process, coals from lignitic through bituminous are crushed, dried, and fed to the liquefaction reactor along with hydrogenated recycle solvent and hydrogen. The products are separated by distillation (qv). The recycle solvent is hydrogenated in a conventional reactor. The heavy bottoms are fed to a partial coker and gasifier to maximize liquid products and obtain a low heating value fuel gas. Hydrogen is generated by either steam reforming the gas or by gasifying unconverted coal (55).

British Coal Corp. is developing a gasoline-from-coal process at a facility at Point of Ayr (Scotland). This process involves treatment with liquid recycle solvents, digestion at 450–500°C, filtration to separate unconverted residues, and separation into two fractions. The lighter fraction is mildly hydrotreated, and the heavier one is hydrocracked (56).

Other methods have been tested by various companies and governmental agencies (57), but none have proven economically feasible.

7.11. Nonfuel Uses

7.11.1. Montan Wax

An important product of the direct extractive treatment of lignitic coals is montan wax [8002-53-7]. The term montan wax or Bergwachs refers strictly to the material obtained by solvent extraction of suitable German brown coals. The generic term for similar materials is montana wax. The small quantities made outside Germany are usually referred to as montan, prefixed by the country of origin (see Waxes).

Brown coals yield, on solvent extraction, 10–15% of a material that contains 60–90% light yellow or brown waxy substances. The remainder is a mixture of deep brown resinous and asphaltic substances. The yield may be increased by increasing the pressure during extraction, but this also adds dark colored dispersion products, and the resultant brown coal cannot be briquetted.

The crude wax is refined by extracting at 90–100°C with an azeotropic mixture of benzene and a mixture of alcohols, typically 85% benzene and 15% methanol (see Distillation, azeotropic and extractive). Distilling the solvent leaves a wax too darkly colored to be used without added refining.

Acid mixtures are used to oxidize and remove the dark materials. Proper control gives a series of bleached waxes. A white wax requires double refining and reduces the yield to about 30% of the crude wax input. A series of synthetic waxes is prepared by separating the acids and alcohols produced during saponification of the wax and reesterifying them with acids or alcohols selected to give desired properties of hardness, solubility, emulsification, and gloss.

In the United States, the most desirable lignites for wax production are those from California and Arkansas. A yield of ca 7% is obtained from these lignites; those from other states give only about 2%.

7.11.2. Miscellaneous

Activated carbon is made from low temperature char and superheated steam or by digesting brown coal with KOH, producing a hard product having a high surface area (57). Cation-exchange resins can be made by sulfonation or by nitration and reduction. Treatment with ethylene dichloride and ammonia gives an anion-exchange resin (see Ion exchange).

The German and Australian brown coals are also excellent humus sources (58). Reclaimed loess soils initially have only 0.4–0.5% humus, as compared to 1.5–2.0% for normal agricultural soils. The addition of sewage sludge or peat to brown coal improves its soil additive properties, both soil moisture retention and pH.

The reduction in normally alkaline pH releases phosphates and nitrogen compounds for plant use. Brown coal soil conditioners are being applied in reclamation and have become commercially available for gardens and vineyards (58). During World War II, and electrode carbon was produced in Germany from material extracted from lignite. The mineral matter was almost completely removed, and the extract was carbonized in coke ovens (59). Lignite ash used for cellular concrete blocks, as a concrete additive, soil stabilizer, cement raw material, and filler for construction and bricks was 2×10^6 t in 1967 (ca 4% of total production) (60).

Combinations of lignite flyash from North Dakota and hydrated lime can increase the strength and durability of soils. The lime content varies from 2–7% and lime:flyash ratio from 1:1 to 1:7 (61). Lignite flyash can also be used as a partial replacement for Portland cement to produce strong, durable concrete (62).

Low temperature (lt) tars of Fischer-Tropsch (FT) fractions provide reasonable substrates for growth of yeast for human or animal food supplements. Yeast growth yields were 99.8% (FT fraction), 95.2 and 84.2% (lt tar) of those from a petroleum-derived paraffin fraction (63) (see Foods, nonconventional).

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