COAL

1. Introduction

The use of coal, known as the rock that burns, was recorded in China, Greece, and Italy >2000 years ago. Coal mining began in Germany around the tenth century AD and enough coal was mined in England for export in the thirteenth century. Coal mining began in the United States in ~1700.

Coal is usually a dark black color, although geologically younger deposits of brown coal have a brownish red color (see LIGNITE AND BROWN COAL). The color, luster, texture, and fracture vary with rank, type, and grade. Coal is the result of combined biological, chemical, and physical degradation of accumulated plant matter over geological ages. The relative amounts of remaining plant parts leads to different types of coal, which are sometimes termed banded, splint, nonbanded (cannel and boghead); or hard or soft; or lignite, subbituminous, bituminous, or anthracite. In Europe, the banded and splint types are generally referred to as ulmic or humic coals. Still other terms refer to the origins of the plant parts through maceral names such as vitrinite, liptinite, and inertinite. The degree of conversion of plant matter or coalification is referred to as rank. Brown coal and lignite, subbituminous coal, bituminous coal, and anthracite make up the rank series with increasing carbon content. The impurities in these coals cause differences in grade.

Coal consists primarily of carbon, hydrogen, and oxygen, and contains lesser amounts of nitrogen and sulfur and varying amounts of moisture and mineral matter. The mode of formation of coal, the variation in plant composition, the microstructure, and the variety of mineral matter indicate that there is a mixture of materials in coal. The nature of the organic species present depends on the degree of biochemical change of the original plant material, on the historic pressures and temperatures after the initial biochemical degradation, and on the finely divided mineral matter deposited either at the same time as the plant material or later. The principal types of organic compounds have resulted from the formation and condensation of polynuclear and heterocyclic ring compounds containing carbon, hydrogen, nitrogen, oxygen, and sulfur. The fraction of carbon in aromatic ring structures increases with rank.

Nearly all coal is used in combustion and coking (see COAL CONVERSION PROCESSES). At least 80% is burned directly in boilers for generation of electricity (see MAGNETOHYDRODYNAMICS; POWER GENERATION) or steam for industrial purposes. Small amounts are used for transportation, space heating, firing of ceramic products, etc. The rest is essentially pyrolyzed to produce coke, coal gas, ammonia (qv), coal tar, and light oil products from which many chemicals are produced (see FEEDSTOCKS,COAL CHEMICALS). Combustible gases and chemical intermediates are also produced by the gasification of coal (seeFUELS,SYNTHETIC), and different-carbon (qv) products are produced by various heat treatments. A small amount of coal is used in miscellaneous applications such as fillers (qv), pigments (qv), foundry material, and water (qv) filtration (qv).

In 1991, the annual coal production was $\sim 1.1 \times 10^9$t in the United States and 4.7×10^9t worldwide, the latter essentially unchanged since 1991 (1,2).

World reserves of bituminous coal and anthracite are $\sim 5.6 \times 10^{12}$t of coal equivalent, ie, 29.3 GJ/t (12.6×10^3Btu/lb), and subbituminous and lignite are 2.9×10^{12} t of coal equivalent (see FUEL RESOURCES). For economic and environmental reasons coal consumption has been cyclic.

2. Origin of Coal

Coal evolved from partially decomposed plants in a shallow-water environment. Various chemical and physical changes occurred in two distinct stages: one biochemical and the other physicochemical (geochemical) (3–7). Because some parts of plant material are more resistant to biochemical degradation than others, optical variations in petrologically distinguishable coals resulted. The terms vitrain and clarain refer to bright coals; durain is a dull coal, and fusain is structured fossil charcoal. Exposure to pressure and heat during the geochemical stage caused the differences in degree of coalification or rank that are observable in the continuous series: peat, brown coal and lignite, subbituminous coal, bituminous coal, and anthracite. The carbon containing deposits in which the inorganic material predominates, such as in oil shale (qv) and bituminous shale, are not classified as coal.

Complete decay of plant material by oxidation and oxygen-based bacteria and fungi is prevented only in water-logged environments such as swamps in regions where there is rapid and plentiful plant growth. Peat is formed in such swamps from plant debris such as branches and twigs, bark, leaves, spores and pollen, and even tree trunks that are rapidly submerged in the swamp water. A series of coal seams have been formed from peat swamps growing in an area that has undergone repeated subsidence followed by deposition of lacustrine or marine intrusion material. Periods during which vegetation flourished and peat accumulated were followed by rapid subsidence resulting in submergence of the peat swamp and covering of the deposit with silt and sand. It has been suggested that in the United States the Dismal Swamp of Virginia and North Carolina, which is gradually being flooded by Lake Drummond, is an area undergoing active subsidence (8).

According to the autochthonous, *in situ*, theory of coal formation, peat beds and subsequently coal were formed from the accumulation of plants and plant debris in place. According to the allochthonous theory, the coal-producing peat bogs or swamps were formed from plant debris that had been transported, usually by streams or coastal currents, to the observed burial sites.

3. Biochemical Stage

The initial biochemical decomposition of plant matter depends on two factors: the ability of the different plant parts to resist attack and the existing conditions of the swamp water. Fungi and bacteria can cause complete decay of plant matter that is exposed to aerated water or to the atmosphere. The decay is less complete if the vegetation is immersed in water containing anaerobic bacteria. Under these latter conditions, the plant protoplasm, proteins, starches, and to a lesser extent the cellulose (qv) are easily digested. Lignin (qv) is more resistant. The most decay-resistant plant parts, for both anaerobic and aerobic decomposition, are the waxy protective layers, ie, cuticles, spore, and pollen walls, and the resins. Vitrain results from the partial decay of lignin and cellulose in stagnant water. The original cell structure of the parent plant tissue can be recognized in many samples.

The clarain (9) and bright attritus (8) are finely banded bright parts of coal that evolved from the residues of fine woody material such as branches, twigs, leaves, spores, bark, and pollen. In aerated waters, the plant parts were more decomposed and show a higher concentration of resins, spores, and cuticles. Dull coal, called durain (9), was formed under these conditions and occurs commonly in Pennsylvanian coals. In the United States, it is known as splint or block coal. More selective chemical and biochemical activity, probably in a drier environment, led to the formation of soft, charcoal-like fusain from woody plant material. The conversion was rapid and probably complete by the end of the peat formation stage. Cannel coal is believed to have formed in aerated water, which decomposed all but the spores and pollen. The name is derived from its quality of burning in splints with a candlelike flame. Boghead coal closely resembles cannel coal but was derived from algae instead of plant spores.

3.1. Geochemical Stage. The conversion of peat to bituminous coal is the result of the cumulative effects of temperature and pressure over a long time. The sediment covering the peat provides the pressure and insulation so that the earth's internal heat can be applied to the conversion. The temperature increase is ~2–5°C for each 100 m of depth. The changes in plant matter are termed normal coalification.

Moisture is lost and the chemical composition changes during coalification. Oxygen and hydrogen decrease and carbon increases. These compositional changes are accompanied by decreases in volatile matter and increases in calorific value. The volatile matter and calorific content are the main criteria used for commercial classification in the United States and for the International Classification.

The change in rank from bituminous coal to anthracite involves the application of significantly higher pressures, ie, as in mountain building activity, and temperatures, ie, as in volcanic activity. The more distant the coal from the disruption, the less proportionate the alteration. Tectonic plate movements involved in mountain building provide pressure for some changes to anthracite. As a general rule, the older the coal deposit, the more complete the coalification and the higher the rank of coal. Most commercial bituminous coal fields were deposited during the Pennsylvanian (~285–320 million years ago), Upper Cretaceous (~65–100 million years ago), and early Tertiary (~20–65 million years ago) ages. The lower rank coals come primarily from the Tertiary and Upper Cretaceous ages, and peat deposits are relatively recent, <1 million years old. However, age alone does not determine rank. The brown coal of the Moscow basin is not buried deeply, and although it was deposited during the Lower Carboniferous or Mississippian age (~320–360 million years ago), there was not enough heat and pressure to convert it further.

4. Coal Petrography

Careful examination of a piece of coal shows that it is usually made up of layers or bands of different materials that are distinct entities upon microscopic examination, which are distinguishable by optical characteristics (10–12). The study of the origin, composition, and technological application of these materials is called coal petrology, whereas coal petrography involves the systematic quantification of the amounts and characteristics by microscopic study. The petrology of coal may involve either a macroscopic or microscopic scale.

On the macroscopic scale, two coal classifications have been used: humic or banded coals and sapropelic or nonbanded coals. Stratification in the banded coals, which result from plant parts, is quite obvious; the nonbanded coals, which derive from algal materials and spores, are much more uniform. The physical and chemical properties of the different layers in a piece of coal or a seam can vary significantly. Therefore the relative amounts of the layers are important in determining the overall characteristics of the mined product. Coal petrography has been widely applied in cokemaking and is important in coal liquefaction programs.

If the mineral matter in the coal exceeds ~40%, then the material is referred to as a coaly or carbonaceous shale. If the mineral matter is a finely divided clay, well dispersed in the coal, then the material may be described as a stony coal or bone coal.

4.1. Macerals. Coal parts derived from different plant parts, are referred to as macerals (13). The maceral names end in “-inite”. The most abundant (~85%) maceral in U.S. coal is vitrinite, derived from the woody tissues of plants. Another maceral, called liptinite, is derived from the waxy parts of spores and pollen, or algal remains. The liptinite macerals fluoresce under blue light permitting a subdivision based on fluorescence. A third maceral, inertinite, is thought to be derived from oxidized material or fossilized charcoal remnants of early forest fires.

A number of subdivisions of the maceral groups have been developed and documented by the International Commission on Coal Petrology (14). Table 1 lists the Stopes-Heerlen classification of higher rank coals. Periodic revisions include descriptions of the macerals, submacerals, morphology, physical properties, and chemical characteristics. Theories on the mode of formation of the macerals and their significance in commercial applications are also included in (14).

The macerals in lower rank coals, eg, lignite and subbituminous coal, are more complex and have been given a special classification. The term huminite has been applied to the macerals derived from the humification of lignocellulosic tissues. Huminite is the precursor to the vitrinite observed in higher rank coals.

The elemental composition of the three maceral groups varies. The vitrinite, which frequently is ~85% of the sample in the United States, is similar to the parent coal. The liptinites are richer in hydrogen, whereas the inertinites are relatively deficient in hydrogen and richer in carbon. The liptinites also contain more aliphatic materials; the inertinites are richer in aromatics. The term inertinite refers to the relative chemical inertness of this material, making it especially undesirable for liquefaction processes because it tends to accumulate in recycled feedstock streams.

Table 1. **Stopes-Heerlen Classification of Maceral Groups, Macerals, and Submacerals of Higher Rank Coals**[a]

Maceral Group	Maceral Subgroup	Maceral (ICCP, 1995)	Maceral (ASTM D 2799)
Vitrinite	Telinite	Telinite Collotelinite	Vitrinite
	Detrovitrinite	Vitrodetrinite Collodetrinite	
	Gelovitrinite	Gelinite Corpogelinite	
Liptinite			Sporinite Cutinite Resinite Alginite
Inertinite	Telo-Inertinite		Fusinite Semifusinite Sclerotinite
	Detro-Inertinite		Inertodetrinite Micrinite
	Gelo-Inertinite		Macrinite

[a] See Ref. 9.

4.2. Vitrinite Reflectance. The amount of light reflected from a polished plane surface of a coal particle under specified illumination conditions increases with the aromaticity of the sample and the rank of the coal or maceral. Precise measurements of reflectance, expressed as a percentage, are used as an indication of coal rank.

Precise reflectance measurements are carried out using incident light having wavelength of 546 nm (green mercury line), and carefully polished coal specimens immersed in an oil having a refractive index of 1.518 at 23°C. Comparison is made to a calibrated standard, using a photomultiplier system. Coal is an anisotropic material, so the reflectance varies according to the orientation of the particle. A typical procedure involves making many measurements on the vitrinite particles in a coarsely ground sample to obtain a range of values that are then used to determine a maximum vitrinite reflectance that correlates with coal rank. Minimum values can also be correlated. Figure 1 illustrates the relationship between reflectances and the carbon content (12). The reflectance of liptinite macerals is less than that for vitrinite, and the petrographer can distinguish the two for low rank coals. However, as measurements are made on progressively higher rank coals, the reflectivities of the liptinites and vitrinites become similar and are the same for medium volatile bituminous coals. For inertinites, the distinctions between reflectivities for vitrinite persist into the anthracite range. Table 2 indicates the vitrinite reflectances for the various coal ranks (12).

4.3. Application of Coal Petrology and Petrography. Petrographic analysis is frequently carried out for economic evaluation or to obtain geologic information. Samples are usually lumps or more coarsely ground material that have been mounted in resins and polished. Maceral analysis involves the examination of a large number (usually >500) of particles during a traverse of a

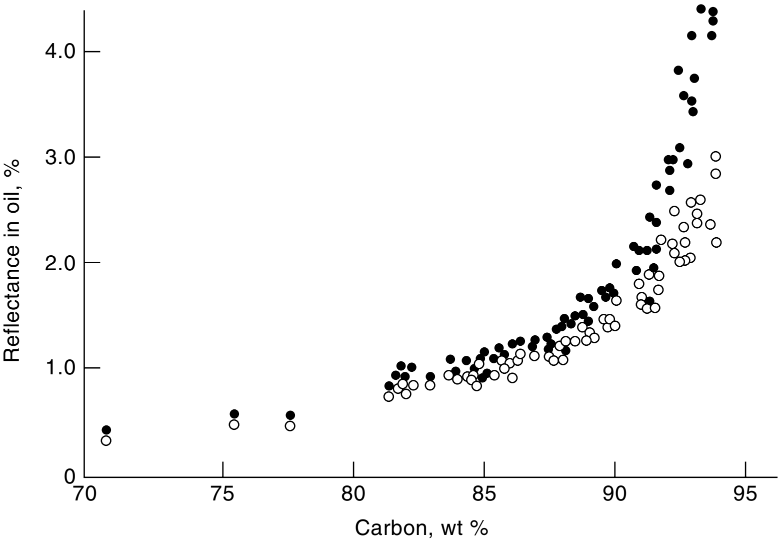


Fig. 1. Relation of vitrinite reflectance (R_o) in percent to maximum temperature (T_{max}) and effective heating time (t_{eff}) where E_A = activation energy in kJ and t_{eff} is within 15°C of T_{max}. To convert kJ to kcal, divide by 4.184 (11).

polished surface to identify the macerals at specified intervals. A volume percentage of each of the macerals present in a sample is calculated.

Seam correlations, measurements of rank and geologic history, interpretation of petroleum (qv) formation with coal deposits, prediction of coke properties, and detection of coal oxidation can be determined from petrographic analysis. Constituents of seams can be observed over considerable distances, permitting the correlation of seam profiles in coal basins. Measurements of vitrinite reflectance within a seam permit mapping of variations in thermal and tectonic histories. Figure 2 indicates the relationship of vitrinite reflectance to maximum temperatures and effective heating time in the seam (11,15).

Table 2. **Vitrinite Reflectance Limits, Taken in Oil, and ASTM Coal Rank Classes**[a]

Coal rank	Maximum reflectance, %
subbituminous	<0.47
high volatile bituminous	
C	0.47–0.57
B	0.57–0.71
A	0.71–1.10
medium volatile bituminous	1.10–1.50
low volatile bituminous	1.50–2.05
semianthracite	2.05–3.00[b]
anthracite	>3.00[b]

[a] Ref. 11.

[b] Approximate value.

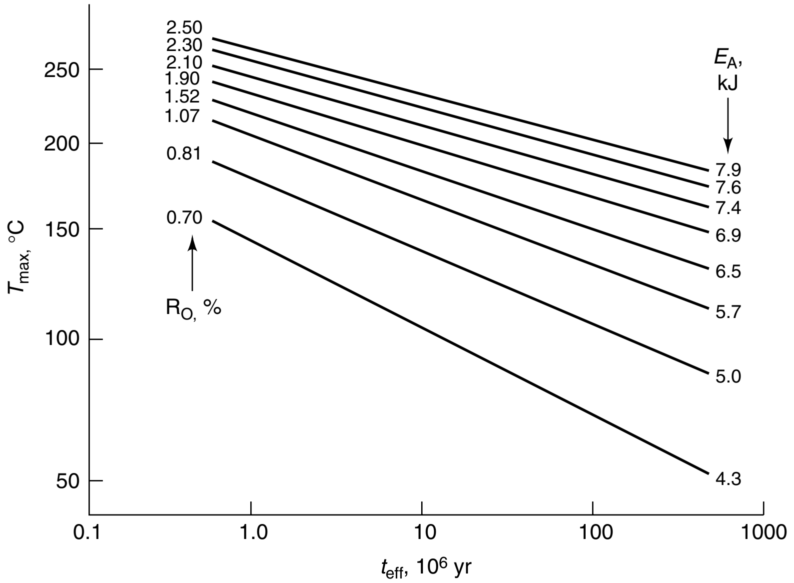


Fig. 2. Relation of vitrinite reflectance (R_o) in percent to maximum temperature (T_{max}) and effective heating time (t_{eff}) where E_A = activation energy in kJ and t_{eff} is within 15°C of T_{max}. To convert kJ to kcal, divide by 4.184 (11).

The coking behavior of coal depends on the rank of the coal, the properties of the individual constituents, and their relative amounts (3–7,10). For some purposes, a blend of coals can be selected to achieve desired coking properties. The maceral groups behave differently on heating: vitrinite from most medium rank coal (9–33% volatile matter) has good plasticity and swelling properties and produces an excellent coke; inertinite is almost inert and does not soften on heating, and exinite becomes extremely plastic and is almost completely distilled as tar. By careful control of the petrological composition and the rank of a coal blend, behavior during carbonization can be controlled. Additionally, coking behavior can be reasonably predicted using petrography and maceral breakage (10). Oxidation reduces the coke-forming properties of a given coal and can also be detected by petrographic techniques (16).

5. Classification Systems

Prior to the nineteenth century, coal was classified according to appearance, eg, bright coal, black coal, or brown coal. A number of classification systems have since been developed. These may be divided into two types, which are complementary: scientific and commercial. Both are used in research, whereas the commercial classification is essential industrially. In the scientific category, the Seyler chart has considerable value.

5.1. The Seyler Classification. The Seyler chart, shown in Figure 3, is based on the carbon and hydrogen content of coals determined on a dry

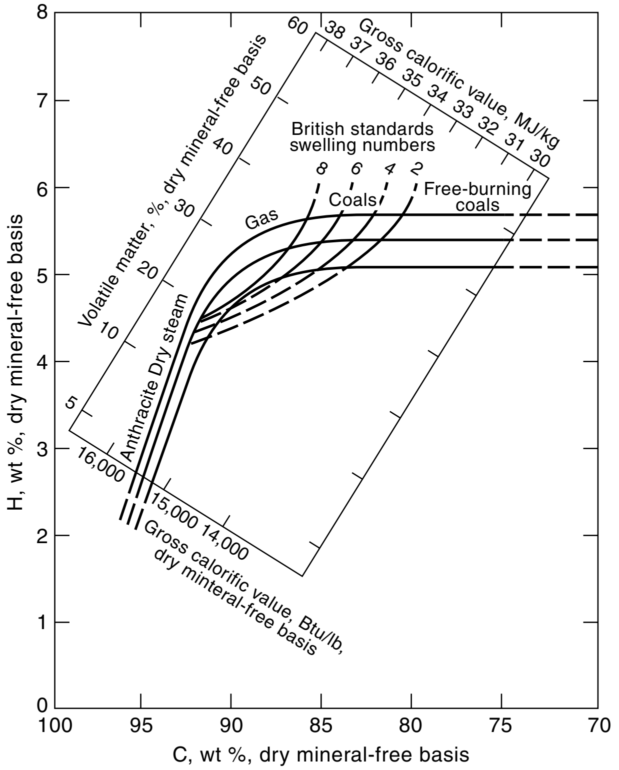


Fig. 3. Simplified form of Seyler's coal classification chart (17). An updated version of Seyler's coal classification is described in Reference6. Note that ASTM uses the free-swelling index (18).

mineral-matter-free basis (17). Points representing different coal samples lie along a broad band. The center band on the chart shows the properties of coal rich in vitrinite. The location of the band indicates the range and interrelationship of the properties. Coals above the band are richer in hydrogen, eg, cannel and boghead coals, and the liptinite macerals in the usual coals. Coals below the band are represented by the maceral inertinite. Other properties, such as moisture and swelling indexes, also fit into specific areas on this chart. The curve in the solid band represents a composition range where the properties of the coal change rapidly. Swelling indexes, coking power, and calorific values are maximized, and moisture is minimized. The lowest rank coals lie on the right side of this curve; the highest rank coals are on the left at the lower part of the band.

5.2. The ASTM Classification. The ASTM classification system was adopted in 1938 as a standard means of specification. This system is used in the United States and in many other parts of the world, and is designated D388 in the ASTM Standards (18). The higher rank coals are specified by fixed carbon $\geq$69%, or for volatile matter $\leq$31%, on a dry, mineral-free basis. Lower rank coals are classified by calorific value on the moist, mineral-matter-free basis. These parameters are given in Table 3. Calorific value depends on two

Table 3. **Classification of Coals by Rank**

Coals	Fixed carbon, %[a]		Volatile matter, %[a]		Gross calorific value, kJ/kg		Agglomerating character
	≥	<	>	≤	≥	<	
Anthracitic							
meta-anthracite	98			2			nonagglomerating
anthracite	92	98	2	8			
semianthracite	86	92	8	14			
Bituminous							
low volatile	78	86	14	22			commonly agglomerating[e]
medium volatile	69	78	22	31			
high volatile							
A		69	31		32,500[d]		
B					30,200[d]	32,500	
C					26,700	30,200	
					24,400	26,700	agglomerating
Subbituminous							
A					24,400	26,700	
B					22,100	24,400	
C					19,300	22,100	
Lignitic							
A					14,600	19,300	nonagglomerating
B						14,600	

[a] Dry, mineral-matter-free basis.

[b] To convert from kJ/kg to Btu/lb, multiply by 0.4302; moist mineral-matter-free basis, ie, contains inherent moisture but not water visible on the surface.

[c] If agglomerating, classify in low volatile group of the bituminous class.

[d] Coals having 69% or more fixed carbon on the dry, mineral-matter-free basis are classified according to fixed carbon, regardless of gross calorific value.

[e] There may be nonagglomerating varieties in the groups of the bituminous class, and there are notable exceptions in high volatile C bituminous group.

properties: moisture absorbing capacity and the calorific value of the pure coal matter. When some overlap between bituminous and subbituminous coals occurs, it is resolved on the basis of the agglomerating properties.

5.3. National Coal Board Classification for British Coals. The classification proposed in 1946 by the U.K. Department of Scientific and Industrial Research led to the system in use by the National Coal Board for coals in the United Kingdom. There are two parameters: the quantity of volatile matter determined on a dry, mineral-matter-free basis, and the Gray-King coke-type assay, a measure of coking power as designated in the British Standards (18). This latter assay is used as a primary means of classification for lower rank coals. The classification applies to coals having <10% ash. High ash coals are cleaned before analysis by a float-sink separation to reduce the ash content <10%.

5.4. International Classification. *Hard Coal.* The amount of coal in international commerce since ~1945 necessitated an international system of coal classification and in 1956 the Coal Committee of the European Economic Community agreed on a system designated the International Classification of Hard Coal by Type (3). Volatile matter and gross calorific value on a moist, ash-free basis are among the parameters considered. Table 4 shows the various classes of the international system and gives the corresponding national names used for these coals.

A three-digit classification is employed in the international system (10), where the first digit indicates the class or rank, such that higher digits correspond to lower ranks; the second digit indicates the group indicated by caking properties such as the free-swelling index or the Roga index; and the third digit defines a subgroup based on coking properties as measured using a dilatometer or the Gray-King assay. Coals having volatile matter up to 33% are divided into classes 1–5; coals having volatile matter <33% are divided into classes 6–9. The calorific values are given for a moisture content obtained after equilibrating at 30°C and 96% rh. The nine classes are then divided into four groups as measured through either the free-swelling index (17) or the Roga index. These tests indicate properties observed when the coal is heated rapidly.

Brown Coal and Lignite. The brown coals and lignites, defined as coals having heating values that are <23,860 kJ/kg (10,260 Btu/lb, 5700 kcal/kg), are classified separately (see LIGNITE AND BROWN COAL). A four-digit code is used for classification. The first two digits (class parameter) are defined by total moisture content of freshly mined coal on an ash-free basis. The third and fourth digit are defined by the tar yield on a dry, ash-free basis.

6. Composition and Structure

The constitution of a coal involves both the elemental composition and the functional groups that are derived therefrom. The structure of the coal solid depends to a significant extent on the arrangement of the functional groups within the material.

6.1. Composition. The functional groups within coal contain the elements C, H, O, N, or S (3,4,5,19). The significant oxygen-containing groups

Table 4. **The International and Corresponding National Systems of Coal Classes**[a]

International system			National classifications							
Class no.	Volatile matter, %	Calorific value, kJ/g[b,c]	Belgium	Germany	France	Italy	The Netherlands	Poland	United Kingdom	United States
0	0–3					antraciti speciali		*meta*-antracyt		*meta*-anthracite
1A	3–6.5		maigre	Anthrazit	anthracite	antraciti communi	anthraciet	antracyt	anthracite	anthracite
1B	6.5–10							polantracyt		
2	10–14		1/4 gras	Mager-kohle	maigre	carboni magri	mager	chudy	dry steam	semianthracite
3	14–20		1/2 gras 3/4 gras	Esskohle	demigras	carboni semigrassi	esskool	polkoksowy *meta*-koksowy	coking steam	low volatile bituminous
4	20–28			Fettkohle	gras à courte flamme	carboni grassi corta fiamma	vetkool	*orto*-koksowy	medium volatile coking	medium volatile-bituminous
5	28–33		gras	Gaskohle	gras proprement dit	carboni grassi media fiamma		gazowo koksowy		high volatile bituminous A
6	>33 (33–40)	32.4–35.4				carboni da gas	gaskool			
7	>33 (32–44)	30.1–32.4			flambant gras	carboni grassi da vapore	gasvlam-kool	gazowy	high volatile	high volatile bituminous B
8	>33 (34–46)	25.6–30.1		Gas flamm-kohle	flambant scc	carboni secchi	vlamkool	gazowo-plomienny		high volatile bituminous C
9	>33 (36–48)	<25.6						plomienny		subbituminous

[a] Ref. 3.

[b] Calculated to standard moisture content.

[c] To convert kJ/g to Btu/lb, multiply by 430.2.

found in coals are carbonyl, hydroxyl, carboxylic acid, and methoxy. The nitrogen-containing groups include aromatic nitriles, pyridines, carbazoles, quinolines, and pyrroles (20). Sulfur is primarily found in thiols, dialkyl and aryl–alkyl thioethers, thiophene groups, and disulfides. Elemental sulfur is observed in oxidized coal (20).

The relative and absolute amounts of the various groups vary with coal rank and maceral type. The principal oxygen-containing functional groups in vitrinites of mature coals are phenolic hydroxyl and conjugated carbonyls as in quinones. Spectroscopic evidence exists for hydrogen bonding of hydroxyl and carbonyl groups. There are unconjugated carbonyl groups such as ketones in exinites. The infrared (ir) absorption bands are displaced from the normal carbonyl range for simple ketones by the conjugation in vitrinites. Interactions between the carbonyl and hydroxyl groups affect the normal reactions.

A range of quantitative organic analytical techniques may be used to determine functional group concentrations. Acetylation and *O*-alkylation are used to determine hydroxyl groups, whereas carbonyl groups are difficult to quantify using simple procedures. A variety of instrumental techniques has also been used to aid in the understanding of coal structure and constitution. Magnetic resonance techniques have been particularly helpful in determining relative amounts of different carbon species within a coal. Table 5 contains data obtained using these techniques (21).

Aromaticity of coal molecules increases with coal rank. Calculations based on several models indicate that the number of aromatic carbons per cluster varies from 9 for lignite to 20 for low volatile bituminous coal, and the number of attachments per cluster varies from 3 for lignite to 5 for subbituminous through medium bituminous coal. The value is 4 for low volatile bituminous (21).

Reaction of coals and mild selective oxidizing agents such as benzoquinone (20,22) causes the coals to lose much of the hydrogen content. Similarly, a palladium catalyst can cause the evolution of molecular hydrogen (23,24). These methods may give an indication of the minimum amount of hydrogen in the coal that is involved in hydroaromatic rings. This amount is close to the total nonaromatic hydrogen determined for lower rank coals. Other hydrogen determining methods involve dehydrogenation using sulfur (25) and using halogens (26). The values obtained by these last methods are somewhat lower than that of benzoquinone.

Hydrogen can be added to the aromatic structures converting them to hydroaromatic rings. The hydrogen addition and removal is generally but not entirely reversible (24).

High resolution mass spectrometry (ms) (qv) has been used with extracts of a series of coals to indicate the association of different heteroatoms (27). Various types of chromatography (qv) have also been used to identify the smaller species that can be extracted from coal.

6.2. Coal Structure. *Bonding in Macromolecules.* Conclusions regarding the chemical structure of the macromolecules within coal are generally based on experimental measurements and an understanding of structural organic chemistry (3,4,20,28). The description given herein refers to vitrinites.

Several requirements must be met in developing a structure. Not only must elementary analysis and other physical measurements be consistent, but limita-

Table 5. **Carbon Structural Distribution of the Argonne Premium Coals Based on NMR Measurements**[a]

	Fraction of carbon type[b]											
Coal[c]	f_a	$'f_a$	$f^C{}_a$	$f^H{}_a$	$f^N{}_a$	$f^P{}_a$	$f^S{}_a$	$f^B{}_a$	f_{al}	$f^H{}_{al}$	$f_{al}*$	$f^O{}_{al}$
North Dakota (L)	0.61	0.54	0.07	0.26	0.28	0.06	0.13	0.09	0.39	0.25	0.14	0.12
Wyodak (SB)	0.63	0.55	0.08	0.17	0.38	0.08	0.14	0.16	0.37	0.27	0.10	0.10
Blind Canyon (HVB)	0.65	0.64	0.01	0.22	0.42	0.07	0.15	0.20	0.35	0.22	0.13	0.04
Illinois no. 6 (HVB)	0.72	0.72	0.00	0.26	0.46	0.06	0.18	0.22	0.28	0.19	0.09	0.05
Pittsburgh (HVB)	0.72	0.72	0.00	0.27	0.45	0.06	0.17	0.22	0.28	0.13	0.15	0.03
Lewiston-Stockton (HVB)	0.75	0.75	0.00	0.27	0.48	0.05	0.21	0.22	0.25	0.14	0.11	0.04
Upper Freeport (MVB)	0.81	0.81	0.00	0.28	0.53	0.04	0.20	0.29	0.19	0.09	0.10	0.02
Pocahontas (LVB)	0.86	0.86	0.00	0.33	0.53	0.02	0.17	0.34	0.14	0.08	0.06	0.01

[a] Ref. 21.

[b] The symbols f_a and f_{al} correspond to total fraction of sp^2 and sp^3 hybridized carbon, respectively. f'_a represents the fraction of sp^2 carbon in aromatic rings; $f^C{}_a$ = the fraction in carbonyls, $\delta > 165$ ppm; $f_a{}^H$ = the aromatic fraction that is protonated; $f_a{}^N$ = the aromatic fraction that is nonprotonated; $f_a,{}^P$ = the phenolic or phenolic ether carbon, $\delta = 150–165$ ppm; $f_a{}^S$, the alkylated aromatic carbon, $\delta = 135–150$ ppm; $f_a{}^B$, the aromatic bridgehead carbon; $f_{al}{}^H$ represents the fraction of CH or CH_2 aliphatic carbon; $f_{al}{}^*$ = the CH_3 or nonprotonated aliphatic carbon; and $f_{al}{}^O$ = the aliphatic carbon bound to oxygen, $\delta = 50–90$ ppm.

[c] L = lignite; SB = subbituminous; HVB = high volatile bituminous; MVB = medium volatile bituminous; and LVB = low volatile bituminous.

tions of structural organic chemistry and stereochemistry must also be satisfied. Mathematical expressions have been developed to test the consistency of any given set of parameters used to describe the molecular structure of coal and analyses of this type have been reported (4,6,19,20,29,30).

Evidence suggests that the structure for vitrinites in bituminous coals and anthracite has the following characteristics: (*1*) the molecule contains a number of small aromatic nuclei or clusters, each usually having from one to four fused benzene rings. The average number of clusters in the molecule is characteristic of the coal rank. This average increases slightly to 90% carbon and then increases rapidly; (*2*) the aromatic clusters are partly linked together by hydroaromatic, including alicyclic, ring structures. These latter rings also contain six carbon atoms and thus, upon loss of hydrogen can become part of the cluster, increasing the average cluster size; (*3*) other linkages between clusters involve short groupings such as methylene, ethylene, and ether oxygen ; (*4*) a significant amount of hydrogen sites on the aromatic and hydroaromatic rings have been substituted by methyl and sometimes larger aliphatic groups; (*5*) the oxygen in vitrinites usually occurs in phenolic hydroxyl groups and ethers (19), substituting for hydrogen on the aromatic structures; (*6*) a small fraction of the rings, both aromatic and hydroaromatic, have oxygen substituted for a carbon atom. Some of these heterocyclic rings may be five membered; (*7*) a lesser amount of oxygen than that occurring in phenolic groups appears in the carbonyl moieties, ie, in quinone form on aromatic rings or as ketones attached to hydroaromatic ones. In both cases, the oxygen is apparently hydrogen bonded to adjacent hydroxyl groups: the reactivity and peak location for the characteristic ir absorptions are different from those in the typical quinones and ketones; (*8*) nitrogen is less abundant in vitrinites than oxygen. It is usually present as a heteroatom in a ring structure or as a nitrile (19); (*9*) most of the sulfur in as-mined coal, especially if the S content exceeds 2 wt%, is associated with inorganic material. However, for very low sulfur coals the organic sulfur which occurs as both aliphatic and aromatic usually exceeds the inorganic. Sulfide and disulfide groups may also link clusters (19,20); (*10*) a given piece of coal contains a variety of molecules having different proportions of a given structural feature and varying molecular weight. These molecules are composed of planar fused ring clusters linked to nonplanar hydroaromatic structures. The overall structure is irregular, open, and complex. Entanglement between molecules occurs, as do cross-links of hydrogen-bonded species, leading to difficulties in molecular weight determinations for extracts and to changes in properties on heating. The different molecular shapes and sizes in a piece of coal lead to irregularities in packing and hence the amorphous nature and the extensive ultrafine porosity; and (*11*) some of the evidence suggests that the molecular weights of extracts representing 5–50 wt% of the coal average 1000–3000. Larger transient units also exist owing to aggregation and it is probable that unextracted coal contains even larger molecular units (19).

Figure 4 gives a representation of the coal molecule (28) that correlates with products obtained from liquefaction. Heating >400°C or mild chemical oxidation changes some hydroaromatic into aromatic structures. Hydroaromatic structures appear to be involved in tar formation (31); however, the tar probably contains some of the same smaller aromatic structures also found in the coal. Tar

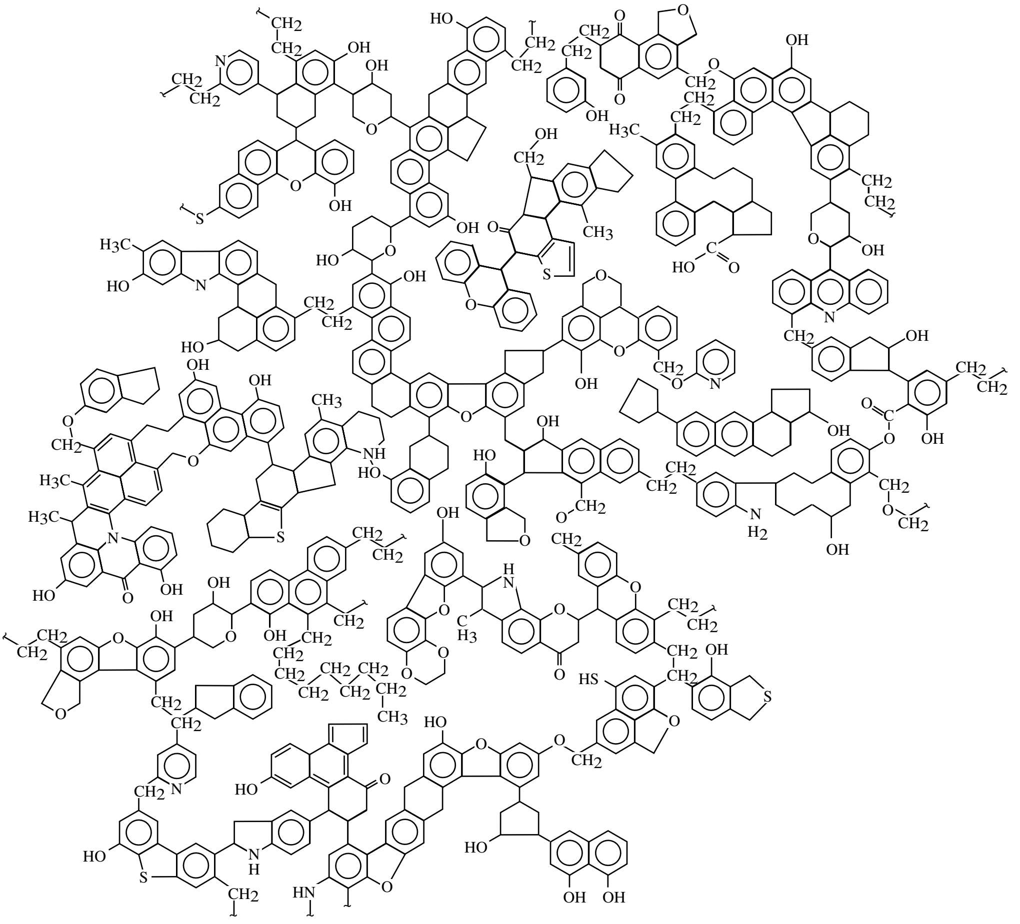


Fig. 4. Model of bituminous coal structure (28).

and gas production, including changes with rank, is consistent (3,4) with the structural model described. Most of the larger units remain in the char or coke produced by heating. The average size of the molecular units is increased by polymerization and conversion of hydroaromatic links to aromatic rings.

Bonding between Macromolecules. The macromolecules that make up the coal structure are held together by a variety of forces and bonds (32). The coal network model is one approach to describing the three-dimensional (3D) structure of the solid. Aromatic clusters are linked by a variety of connecting bonds, through oxygen, methylene or longer aliphatic groups, or disulfide bridges, and the proportions of the different functional groups change as the rank of the coal is progressively increased. For example, oxygen is diminished. Acid groups decrease early in the rank progression and other groups follow leaving ether moieties that act as cross-links between the larger clusters in the high rank coals. Another type of linkage involves hydrogen bonds which, eg, hold hydroxy and ketogroups together in the solid. A review of coal models, the mechanical properties of the network, and the glass-transition temperature corresponding to the change from a fluid to a rigid amorphous solid are available (32).

A model for coal fluidity based on a macromolecular network pyrolysis model has been developed (33). In that model, bond breaking is described as a first-order reaction having a range of activation energies. A variety of lattices have also been used to describe the bonding in coal. In turn, these structures have been used to describe devolatilization, combustion, and char formation (34). The form of the macromolecule in a liquid extract tends to be spherical as a result of surface energy considerations, hydrogen bonds, and van der Waals forces (32).

6.3. Coal Constitution. Chemical composition studies (35,36) indicate that brown coals have a relatively high oxygen content. About two-thirds of the oxygen is bonded carboxyl, acetylatable hydroxyl, and methoxy groups. Additionally, unlike in bituminous coals, some alcoholic hydroxyl groups are believed to exist.

Anthracites. The anthracites, which approach graphite in composition (see CARBON,GRAPHITE), are classified higher in rank, have less oxygen and hydrogen, and are less reactive than bituminous coals. Anthracites are also insoluble in organic solvents. These characteristics become more pronounced as rank increases within the anthracite group. The aromatic carbon fraction of anthracites is at least 0.9, and the number of aromatic rings per cluster is greater than that for the low volatile bituminous coals, with a value of $\sim$10 for anthracite having 95 wt% C. There is X-ray diffraction evidence (37) to indicate that the aromatic rings are more loosely and variably assembled than those in bituminous coal clusters. The anthracites have greater optical and mechanical anisotropy than lower rank coals, and the internal pore volume and surface increase with rank after the minimum below $\sim$90 wt% C.

6.4. Mineral Matter in Coal. The mineral matter (7,38) in coal results from several separate processes. Some comes from the material inherent in all living matter; some from the detrital minerals deposited during the time of peat formation; and a third type from secondary minerals that crystallized from water that has percolated through the coal seams.

A variety of instrumental techniques may be used to determine mineral content. Typically the coal sample is prepared by low temperature ashing to remove the organic material. Then one or more of the techniques of X-ray diffraction, ir spectroscopy, differential thermal analysis, electron microscopy, and petrographic analysis may be employed (7).

The various clay minerals are the most common detrital mineral (see CLAYS); however, other common ones include quartz, feldspar, garnet, apatite, zircon, muscovite, epidote, biotite, augite, kyanite, rutile, staurolite, topaz, and tourmaline. The secondary minerals are generally kaolinite, calcite, and pyrite. Analyses have shown the presence of almost all elements in at least trace quantities in the mineral matter (39). Certain elements, ie, germanium, beryllium, boron, and antimony, are found primarily with the organic matter in coal, whereas zinc, cadmium, manganese, arsenic, molybdenum, and iron are found with the inorganic material. The primary elemental constituents of mineral matter in coal are aluminum, silicon, iron, calcium, magnesium, sodium, and sulfur.

The relative concentrations depend primarily on the geographical location of the coal seam, and vary from place to place within a given field. In the eastern United States, the most abundant mineral elements are silicon, aluminum, and iron and there are much lower amounts of alkali and alkaline earth elements. West of the Mississippi River the relative amounts of silicon, aluminum, and iron are much less and the alkaline earth and alkali elements are much greater.

7. Properties

Pieces of coal are mixtures of materials somewhat randomly distributed in differing amounts. The mineral matter can be readily distinguished from the organic, which is itself a mixture. Coal properties reflect the individual constituents and the relative proportions. By analogy to geologic formations, the macerals are the constituents that correspond to minerals that make up individual rocks. For coals, macerals, which tend to be consistent in their properties, represent particular classes of plant parts that have been transformed into coal (40). Most detailed chemical and physical studies of coal have been made on macerals or samples rich in a particular maceral, because maceral separation is time consuming.

The most predominant maceral group in U.S. coals is vitrinite. The other important maceral groups include inertinite, including fusinite, a dull fibrous material similar to charcoal, and the liptinite group, including sporinite, which is relatively fusible and volatile. Differences in macerals are evident over the range of coal rank, ie, from brown coal or lignite to anthracite. The definition of rank is that generally accepted as the wt% C, on a dry, mineral-free basis, in the vitrinite associated with the given coal in the seam. The range of ranks in which differences between macerals are most significant is 75–92 wt% C content of the vitrinite. These coals are bituminous.

In the United States the commercial classification of coals is based on the fixed carbon (or volatile matter) content and the moist heating value. One correlation is made by plotting the hydrogen content, on a mineral-free basis, against the corresponding carbon content. A similar plot, made using the commercial criteria of volatile matter and heating placed on axes at an appropriate angle to the % C, % H axes, forms the Seyler coal classification chart (17). Both are illustrated in Figure 3. Table 6 indicates the usual range of composition of commercial coals of increasing rank.

7.1. Physical Methods of Examination.

Physical methods used to examine coals can be divided into two classes that, in the one case, yield information of a structural nature such as the size of the aromatic nuclei, ie, methods such as X-ray diffraction, molar refraction, and calorific value as a function of composition; and in the other case indicate the fraction of carbon present in aromatic form, ie, methods such as ir and nuclear magnetic resonance (nmr) spectroscopies, and density as a function of composition. Some methods used and types of information obtained from them are (41).

The scattering of X-rays (6,37,42) gives information on the average distances between the carbon atoms in coal and insight into the bonding between these atoms. Because X-ray scattering depends on the number of protons in the

Table 6. **Composition of Humic Coals**

	Composition, wt%[a]						
Type of coal	C	H	O	N	Moisture as found	Volatile matter	Calorific value, kJ/g[b]
peat	45–60	3.5–6.8	20–45	0.75–3.0	70–90	45–75	17–22
brown coals and lignites	60–75	4.5–5.5	17–35	0.75–2.1	30–50	45–60	28–30
bituminous coals	75–92	4.0–5.6	3.0–20	0.75–2.0	1.0–20	11–50	29–37
anthracites	92–95	2.9–4.0	2.0–3.0	0.5–2.0	1.5–3.5	3.5–10	36–37

[a] Dry, mineral-matter-free basis except for moisture value.

[b] To convert kJ/g to Btu/lb, multiply by 430.2.

nucleus, carbon is much more effective in scattering X-rays than hydrogen (see X-RAY TECHNOLOGY). The ultraviolet (uv) and visible spectra (6) of coal and various solvent extracts show decreasing absorption with increasing wavelength and lack features to aid in interpreting structure except for one peak ~270 nm, which is believed to result from superposition of effects from many similar species. In studies of specific features, comparisons are usually made between coal or coal-derived samples, and pure, usually aromatic, compounds indicating probable presence of particular structures or functional groups. Similar statements can be made concerning reflectance and refractive index (3,4). The derived optical anisotropy is especially evident in coals having carbon contents that exceed 80–85 wt%. Measurements perpendicular and parallel to the bedding plane give different results for optical and some other characteristics (see SPECTROSCOPY).

A significantly greater amount of information concerning functional groups such as hydroxyl can be obtained from ir absorption (3,4,6); however, this is less specific than the information obtained from an individual organic compound (Fig. 5) (6). An estimate of the relative amount of hydrogen attached to aromatic and nonaromatic structures can, however, be made by using this method (see INFRARED AND RAMAN SPECTROSCOPY). Studies may be carried out on raw coal or products derived from the coal. Physical separation is used to separate fractions of extract and aid in the deduction of the parent coal structure. A method of characterizing coal liquids in terms of 10 fractions of different functionality has been described (43).

Magnetic resonance spectra (^{1}H and ^{13}C) (6,21) also yield information on bonding for hydrogen and carbon, including the distribution between aromatic and nonaromatic structures, as well as bonding to various heteroatoms. Additional estimates may be made of hydrogen in CH, CH_2, and CH_3 groups. Developments in solid-state ^{13}C nmr spectroscopy, coupled with cross-polarization (cp), magic angle spinning (mas), and dipolar-decoupling techniques have made these estimates somewhat more quantitative than those from ir measurements (see MAGNETIC SPIN RESONANCE). Quantitation has been somewhat limited because of a fraction of the atoms that are not observed owing to paramagnetic centers or spin dynamics of the system (21) (see Table 5).

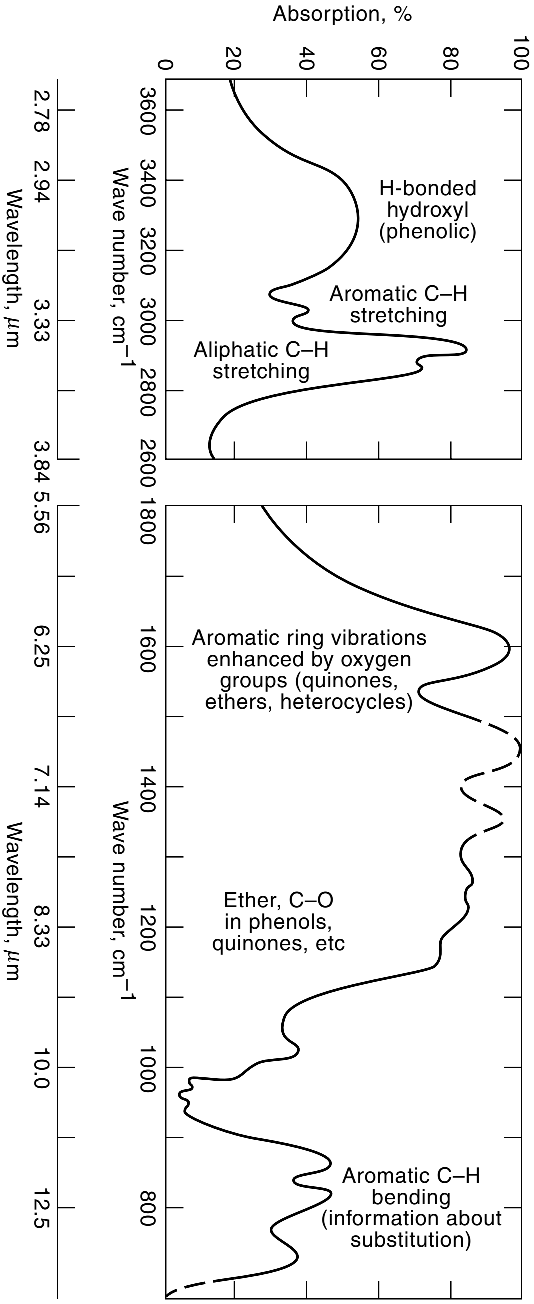


Fig. 5. Infrared spectrum of a medium rank coal where the dashed lines represent variations that occur as a result of differing maceral content or because of functional group conjugation.

Electron spin resonance (esr) (6,44) has had more limited use in coal studies. A rough estimate of the free-radical concentration or unsatisfied chemical bonds in the coal structure has been obtained as a function of coal rank and heat treatment. For example, the concentration increases from 2×10^{18}radicals/g at 80 wt% carbon to a sharp peak of $\sim 50 \times 10^{18}$radicals/g at 95 wt % carbon content and drops almost to zero at 97 wt% carbon. The concentration of these radicals is less than that of the common functional groups such as hydroxyl. However, radical existence seems to be intrinsic to the coal molecule and may affect the reactivity of the coal as well as its absorption of uv radiation. Measurements from room temperature to 900 K indicate that the number of electron spins/g increases sharply above ~600 K and peaks from 773 to 850 K, with increasing values for higher rank coals. Oxidation increases the number of radicals by a factor of 3 over 8 days (44).

The other physical measurements (4,6), except for diamagnetic susceptibility (4) and possibly density (4), are primarily of interest for determining chemical structural properties of coal.

7.2. Physical Properties. Most of the physical properties discussed herein depend on the direction of measurement as compared to the bedding plane of the coal. Additionally, these properties vary according to the history of the piece of coal. Properties also vary between pieces because of coal's brittle nature and the crack and pore structure. One example concerns electrical conductivity. Absolute values of coal sample specific conductivity are not easy to determine. A more characteristic value is the energy gap for transfer of electrons between molecules, which is determined by a series of measurements over a range of temperatures and is unaffected by the presence of cracks. The velocity of sound is also dependent on continuity in the coal.

The specific electrical conductivity of dry coals is very low, specific resistance 10^{10}–10^{14} Ωcm, although it increases with rank. Coal has semiconducting properties. The conductivity tends to increase exponentially with increasing temperatures (4,6). As coals are heated to above ~600°C the conductivity rises especially rapidly owing to rearrangements in the carbon structure, although thermal decomposition contributes somewhat below this temperature. Moisture increases conductivity of coal samples through the water film.

The dielectric constant is also affected by structural changes on strong heating. Also the value is very rank dependent, exhibiting a minimum at ~88 wt% C and rising rapidly for carbon contents >90 wt% (4,6,45). Polar functional groups are primarily responsible for the dielectric of lower ranks. For higher ranks the dielectric constant arises from the increase in electrical conductivity. Information on the freedom of motion of the different water molecules in the particles can be obtained from dielectric constant studies (45).

Magnetic susceptibility measurements indicate that the organic part of the coal is diamagnetic, having traces of paramagnetic behavior resulting from free radicals or unpaired electrons (6).

Density values (4,6) of coals differ considerably, even after correcting for the mineral matter, depending on the method of determination. The true density of coal matter is most accurately obtained from measuring the displacement of helium after the absorbed gases have been removed from the coal sample. Density values increase with carbon content or rank for vitrinites. Values are

1.4–1.6 g/ml >85 wt % carbon where there is a shallow minimum. A plot of density versus hydrogen content gives almost a straight-line relationship, and if the reciprocal of density is plotted, the linear relationship is improved. Values for different macerals as well as for a given maceral of different ranks are almost on the same line.

Thermal conductivity and thermal diffusivity are also dependent on pore and crack structure. Thermal conductivities for coals of different ranks at room temperature are in the range of 0.230.35 W/(m × K). The range includes the spread owing to crack variations and thermal diffusivities of $(1–2) \times 10^{-3}$ cm ^{2}/s. At 800°C, these ranges increase to 1–2W /(m × K) and $(1–5) \times 0^{-2}$ cm ^{2}/s, respectively. The increase is mainly caused by radiation across pores and cracks.

The specific heat of coal can be determined by direct measurement or from the ratio of the thermal conductivity and thermal diffusivity. The latter method gives values decreasing from 1.25J/(g × K)[0.3cal/(g × K)] at 20°C to 0.4J/[g × K)(0.1cal /(g × K)] at 800°C. The specific heat is affected by the oxidation of the coal (46).

7.3. Ultrafine Structure. Coal contains an extensive network of ultrafine capillaries (3,4,6,47) that pass in all directions through any particle. The smallest and most extensive passages are caused by the voids from imperfect packing of the large organic molecules. Vapors pass through these passages during adsorption, chemical reaction, or thermal decomposition. The rates of these processes depend on the diameters of the capillaries and any restrictions in them. Most of the inherent moisture in the coal is contained in these capillaries. The porous structure of the coal and products derived from it have a significant effect on the absorptive properties of these materials.

A range of approaches has been developed for studying the pore structure. For example, heat of wetting by organic liquids is one measure of the accessible surface. The use of liquids having different molecular sizes gives information about restrictions in the pores. Measurements of the apparent density in these liquids give corresponding information about the volume of capillaries. Measurement of the adsorption of gases and vapors provide information about internal volume and surface area. Pores have been classified into three size ranges: (*1*) micropores (<0.4–2.0 nm) measured by CO_2 adsorption at 298 K; (*2*) mesopores (2.0–50 nm) from N_2 adsorption at 77 K; and (*3*) macropores (>50 nm) from mercury porosimetry. For coals having <75 wt% C, macropores primarily determine porosity. For 76–84 wt% C, ~80% of the pore volume primarily results from micro and transitional pores. For the higher rank coals porosity is caused primarily by micropores (48).

Bituminous coals appear to have specific internal surfaces in the range of 30–100 m^2/g arising almost entirely from ultrafine capillaries of <4-nm diameters. The surface area of the very fine capillaries can be measured accurately by using methods not too far below room temperature, depending on the gas or vapor used (49). Diffusion into the particle is very slow at low temperatures. Therefore, measurements at liquid nitrogen temperature (77 K) relate to the external surfaces and macro- as well as mesopores, and may yield areas that are lower than ambient temperature measurements by factors of 100. Sorption by neon or krypton near room temperature and heat of wetting in methanol have given surface area values. The methanol method is affected, up to a factor

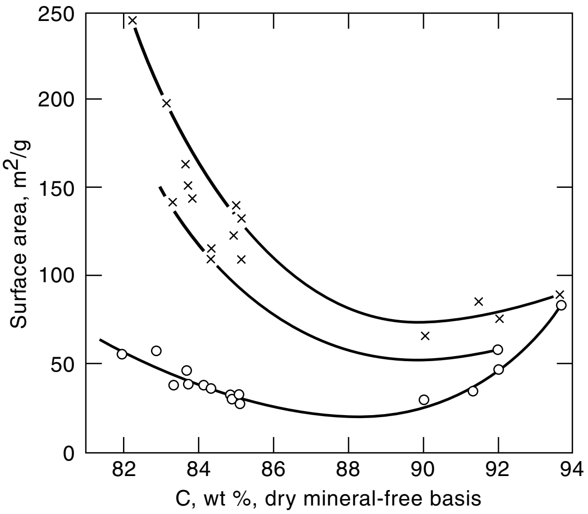


Fig. 6. Surface area of coals as estimated from (o) neon sorption at 25°C and (x) methanol heat of wetting.

of 4, by polar groups, but it is faster. Pore characteristics may also be determined by nmr measurements, as with xenon (50). Measurements of the change in the nmr chemical shift with varying Xe pressures permit the calculation of pore diameters. Total porosity volumes of bituminous and anthracite coal particles are ~10–20%, and ~3–10% are in the microphase range. There are shallow minima in plots of internal area as can be seen from Figure 6 and in plots of internal porosity against coal rank in the range of vitrinite carbon content of 86–90% (6). It is possible to use low angle scattering of X-rays to obtain a value of internal surface, but this does not distinguish between accessible capillaries and closed pores (51).

Different coals have been observed in the electron microscope when two pore-size ranges appear, one of >20 nm and the other <10 nm (52). Fine pores from 1–10 nm across have been observed using a lead impregnation procedure (53). Effectiveness of coal conversion processes depends on rapid contact of gases with the surface. Large internal surfaces are required for satisfactory rates (54).

7.4. Mechanical Properties. Mechanical properties (4,6,55) are important for a number of steps in coal preparation from mining through handling, crushing, and grinding. The properties include elasticity and strength as measured by standard laboratory tests and empirical tests for grindability and friability, and indirect measurements based on particle size distributions.

Deformation Under Load. The mechanical behavior of coal is strongly affected by the presence of cracks, as shown by the lack of proportionality between stress and strain in compression tests or between strength and rank. However, tests in triaxial compression indicate that as the confirming pressure is increased different coals tend to exhibit similar values of compressive strength perpendicular to the directions of these confining pressures. Except for anthracites, different coals exhibit small amounts of recoverable and irrecoverable strain under load.

Dynamic tests have been used to measure the variation of elastic properties with coal rank. Tests using vitrains suggested that coals were mechanically isotropic up to 92 wt% C, with anisotropy increasing above this value. Dynamic tests were used to measure internal energy losses in vibration and to study the fluidity changes of coking coals on oxidation. The Young's modulus for median rank coals has been found to be about 4 GPa (4×10^{10}dyn/cm^2) (6). Sharp increases in the Young's and shear moduli have been found in vitrinites having increase in carbon content >92 wt%.

Strength. The strength of a coal as measured in the laboratory may not be relevant to mining or size reduction problems where the applied forces are much more complex. There are indications that compressive strength, measured by compression of a disk, may give useful correlations to the ease of cutting for different kinds of equipment. Studies of the probability of survival of pieces of different size suggest that the breaking stress S should be most closely related to the linear size x rather than the area or volume of the piece. The results of a number of studies (6) indicate that S is proportional to x^{-r} where r frequently has the value 1–2.

The effects of rank on both compressive and impact strength have been studied, and usual minima were found at 20–25% dry, ash-free volatile matter (88–90 wt% carbon). Accordingly, the Hardgrove grindability index exhibits maximum values in this area.

Size Distribution Relationships. Different models have been used to describe the size distribution of particles experiencing single and multiple fractures. A model based on fracture at the site of the weakest link and a distribution of weakest links in the system gave results that could be described as well by the Rosin-Rammler relation (56). The latter is based on the concept that fracture takes place at prexisting flaws that are distributed randomly throughout the particle.

Comminution. The size reduction of coal during handling of comminution results from many ill-defined forces. Grindability and friability tests are useful indicators of size reduction for any given coal having a specific energy consumption. The Hardgrove test yields an index that varies with coal rank, moisture content, and ash and maceral distribution. The higher the grindability index, the lower the energy requirements to achieve a given size reduction. These indexes are useful in establishing capacity factors for pulverizers. Grinding is easiest for coals having 75–80% dry, ash-free fixed carbon. Optimum moisture contents have been observed for the younger coals. High moisture contents lead to difficulty in grinding, and excessive drying causes the coal particles to be tougher than the optimum dryness.

A relationship between energy consumption and size reduction would be helpful for comminution processes, but none of the many attempts to develop this have been broadly applicable. One reason is that generation of new surface is only one of many phenomena in the size reduction (qv) process. The energy requirements of a comminution system may, however, be estimated from laboratory tests for given amounts of size reduction. For pulverized coal-fired boilers 70% <74mm size (−200mesh) is frequently used. Product size distributions of reproducible forms are obtained from a range of graded coal input sizes and careful control of crushing conditions in the laboratory. The Hardgrove test gives data for comparing ease of grinding different coals (57). The efficiency of pulverizers can then be calculated from the energy requirements for each product size from a series of tests. The relationship between a particle size and energy consumption obtained from plant data is frequently expressed in terms of Kick's or Rittinger's laws or some modification such as Bond's law (58). These empirical relationships do not provide much insight into the mechanism of the grinding process.

The development of a continuous grinding index was the focus of work in the late 1970s (59). The laboratory test equipment used is similar to that for the Hardgrove test but permits classifying the product and recycling the oversize material. An improved correlation is obtained that may, however, need to be corrected for the relative sizes of the test grinding balls versus those used in commercial scale equipment. The continuous grinding index is especially useful for lower rank coals.

7.5. Properties Involving Utilization.

Coal rank is the most important single property for application of coal. Rank sets limits on many properties such as volatile matter, calorific value, and swelling and coking characteristics. Other properties of significance include grindability, ash content and composition, and sulfur content.

Combustion. Most of the mined coal is burned to produce steam for electric power generation (qv). The calorific value determines the amount of steam that can be generated. However, the design and operation of a boiler requires consideration of a number of other properties (see FURNACES, FUEL-FIRED).

In general, high rank coals (high calorific value) are more difficult to ignite, requiring supplemental oil firing and slower burning in large furnaces to reach complete combustion. Greater reactivity makes lower rank coals better suited for cyclone burners, which carry out rapid, intense combustion to maximize carbon utilization and minimize smoke emission. The burning profile, a derivative thermogravimetric analysis of oxidation, is used to characterize coal for oxidation or combustion behavior (see COMBUSTION TECHNOLOGY). Volatile matter is important for ease of coal ignition. Because high rank coals have low volatile matter contents, they burn more slowly and with a short flame. They are primarily used for domestic heating, where heat is transferred directly from the fuel bed. For kilns, long hot flames are preferred and the coal should have medium to high volatile matter. The heating value released with the volatile matter for the various coals is given in Table 7 (6).

Table 7. **Rank and Heating Value in Volatile Matter**

Rank (ASTM)	Volatile matter, %	Total heat energy liberated in volatile matter, %
anthracite	<8	5–14
semianthracite	8–14	14–21
low volatile bituminous	14–22	21–28
medium volatile bituminous	22–31	28–36
high volatile A bituminous	>31	36–47

The swelling and caking properties of coal are not important for most boiler firing, such as pulverized coal-fired use. Some units, however, such as retort stokers, form coke in their normal operation. The smaller domestic heating units also require noncaking coal for satisfactory operation. For pulverized coal firing, a high Hardgrove index or grindability index is desired. The easier the coal is to grind, the lower the energy cost for pulverizing. The abrasiveness of the coal is also important because this determines the wear rate on pulverizer elements.

Coal moisture content, which affects both handling characteristics and freight costs, is most important for fine (<0.5 mm) particles. The lower rank coals have higher moisture contents. The moisture acts as a diluent, lowering flame temperatures and carrying sensible heat out with the flue gases. For pulverized coal firing the moisture content must be low to maximize grindability and avoid clogging. Thus dry run-of-mine coal having up to 30% ash may be more desirable than cleaning and drying coal.

The moisture content of peat or brown coal that is briquetted for fuel must be reduced to $\sim$15% for satisfactory briquetting. Mechanical or natural means are used because of the cost of thermal drying. Moisture is sometimes desirable. About 8% is necessary for prevention of combustible loss from a chain-grate stoker.

Ash content is also important. Ash discharge at high temperature, as molten ash from a slagging boiler, involves substantial amounts of sensible heat. However, the higher cost of washed coal of lower ($\sim$10%) ash content does not always merit its use. Ash disposal and extra freight costs for high ash coals also affect the coal selection. The use of continuous mining equipment produces coal having $\sim$25% ash content. The average ash content of steam coal burned in the United States is $\sim$15%. For some applications, such as chain-grate stokers, a minimum ash content of $\sim$7–10% is needed to protect the metal parts.

Ash fusion characteristics are important in ash deposition in boilers. Ash deposition occurring on the furnace walls is termed slagging, whereas accumulation on the superheater and other tubes is termed fouling. A variety of empirical indexes have been developed (60,61) to relate fouling and slagging to the ash chemical composition through parameters such as acidic and basic oxides content, sodium, calcium and magnesium, and sulfur.

A related property is the viscosity of coal ash. Ash viscosity affects the rate at which ash deposits may flow from the walls, and thus the requirements for ash removal equipment such as wall blowers and soot blowers. The preferred coal ash has a narrow temperature range through which it passes the plastic range, $\sim$25,000–1,000,000 mPa × s (=cP) (62).

Some minor constituents can interfere in firing. High ($>0.6\%$) chlorine is associated with high sodium and complex sulfate deposits that appear to be required to initiate deposition on superheater tubes, as well as initiate stress corrosion cracking of superheater tubes. Phosphorus ($>0.03\%$ of the coal) contributes to phosphate deposits where high firing temperatures are used. Sulfur forms complex sulfates, however, its most damaging effect is corrosion of the boiler's coolest parts through condensation of sulfur oxides as sulfuric acid. Control is achieved by setting flue gas temperatures above the acid dew point in the boiler areas of concern.

Sulfur content plays an important role in meeting air quality standards. In the United States, the EPA has set an emission limit for SO_2 of 516 g/10^6 kJ (490 g/ × 10^6 Btu) of coal burned. To meet this, steam coals have to contain <1% sulfur. Regulations resulting from the Clean Air Act of 1991 call for reduction of the total amount of sulfur oxide emissions by 8–9 million tons annually. One-half of this reduction was required by 1995, the remainder by the year 2000. A cap on the total emissions is given, and reductions in NO_x and particulate emissions are also mandated. Credits given for reductions of emissions that exceed the amount indicated for a given plant may be sold to other facilities unable to meet the requirements. This requirement is expected to force the addition of SO_2, NO_x, and particulate removal equipment to all boilers. Technology is being developed to control SO_x and NO_x through a combination of sorbent injection into the furnace and scrubbing and/or baghouse treatment to neutralize the acid gases and catalytically convert the NO_x to nitrogen (63).

Fluidized-bed boilers have been built with sizes up to 150 mw for commercial power generation and cogeneration units. This type of technology is displacing some stoker fired units at the low capacity (<200,000 kg/h of steam) boilers and smaller pulverized coal units at the large size of the fluidized-bed range. Bubbling and circulating bed designs are used, with operating temperatures in the 816–899°C range. Sulfur oxides are controlled using dolomite or limestone injection in the bed. Higher (2:1–5:1) Ca/S ratios are needed for fluid bed units than for wet scrubbing (1:1) or spray dryers (1.2–1.5:1). Nitrous oxide emissions are higher for fluid-bed units than for the other methods, possibly because of formation and oxidation of hydrogen cyanide (64,65) (see REACTOR TECHNOLOGY).

Coke Production. Coking coals are mainly selected on the basis of the quality and amount of coke that they produce, although gas yield is also considered. About 65–70% of the coal charged is produced as coke. The gas quality depends on the coal rank and is a maximum, measured in energy in gas per mass of coal, for coals of ~89 wt% carbon on a dry, mineral matter-free basis, or 30% volatile matter.

Coals having 18–32% volatile matter are used to produce hard metallurgical coke. Methods have been developed to blend coals having properties outside this range to produce coke. Several coals are frequently blended to improve the quality of the coke (6,66). Blending also affects the shrinkage required to remove the coke from the ovens after initial swelling. Lower rank coals having up to 40% volatile matter may be used alone or in blends at a gasmaking plant. This coke, which need not be as strong as metallurgical coke, is more reactive, and is used in the domestic market.

Coking coal is cleaned so that the coke ash content is not >10%. An upper limit of 1–2 wt% sulfur is recommended for blast furnace coke. A high sulfur content causes steel (qv) to be brittle and difficult to roll. Some coal seams have coking properties suitable for metallurgical coke, but the high sulfur prevents that application. Small amounts of phosphorus also make steel brittle, thus low phosphorus coals are needed for coke production, especially if the iron (qv) ore contains phosphorus.

Solvent Extraction. Coal partially dissolves in a number of solvents and this property has been used to aid in characterization of coal material, because the composition of extracts is sometimes similar to the coal. A wide range of organic solvents can be used (6,67), but dissolution is never complete and usually requires heating to temperatures sufficient for some thermal degradation or solvent reaction to take place, eg, ~400°C. Dissolution of up to 40 wt% can be achieved near room temperature and up to 90% near 400°C. At room temperature the best solvents are primary aliphatic amines, pyridine, and some higher ketones, especially when used with dimethylformamide (DMF). Above 300°C large amounts can be dissolved using phenanthrene, 1-naphthol, and some coal-derived high boiling fractions. Coals having 80–85% carbon in the vitrinite give the largest yields of extract. Very little coal having >90 wt%C dissolves. Ultrasonic enhancement of extraction increases the yield of product by about 2.5 times the nonirradiated material (68) (see ULTRASONICS). The increase occurs only in solvent mixtures that significantly swell coals that range in rank from lignite to high volatile bituminous coal.

When the concentration of dissolved coal exceeds ~5% of the solution by weight, the extracted material resembles the parent coal in composition and some properties. The extract consists of the smaller molecules within the range of the parent coal. Recovered extract is relatively nonvolatile and high melting. A kinetic study of coal dissolution indicated increasing heats of activation for increasing amounts of dissolved coal (69).

Gasification. Many of the coal selection criteria for combustion apply to gasification, which is typically a form of partial oxidation. Gasifiers are primarily described as fixed bed, fluidized bed, entrained, or rotating bed (70). The fixed bed involves an upward flow of reaction gas through a relatively stationary bed of hot coal. The gas velocity is slow enough to avoid blowing the coal out of the bed. The fluidized bed operates at higher gas velocities than the fixed bed and utilizes somewhat smaller particles. The entrained bed operates with parallel flows of reaction gas and finely pulverized coal particles to minimize reaction time and maximize throughput of product. The rotating bed is similar to a kiln that operates with the coal entering at the upper part of the inclined kiln. Rotation avoids clinkering and exposes fresh surfaces to enhance completion of the reaction. The range of coals that may be used vary from one gasifier type to another. Entrained flow gasifiers are able to handle the widest range of raw coals. Fixed-bed gasifiers require mildly caking or noncaking feedstocks for normal operation.

The Lurgi fixed-bed gasifier operates using lump coal of a noncaking type having an ash composition chosen to avoid a sticky, partly fused ash in the reactor. A slagging version of this gasifier has been tested in Westfield, Scotland. Other fixed-bed gasifiers have similar coal requirements.

The Shell-Koppers-Totzek gasifier is an entrained-bed type. It can gasify lignite and subbituminous or bituminous coal. The coal is fed as a pulverized fuel, usually ground to 70% <74 mm (−200 mesh) as used for pulverized coal fired boilers. Residence times are only a few seconds, therefore coal reactivity is important. The gasifier operates at >1650°C, so that coal ash flows out of the gasifier as a molten slag. Coal ash composition must permit continuing molten ash flow.

Fluidized-bed gasifiers typically require a coal feed of particles near 2–3 mm diameter. Caking coals are to be avoided because they usually agglomerate in the bed. This can be avoided using a pretreatment consisting of a surface oxidation with air in a fluidized bed. A useful flue gas is produced. Examples of this type include the commercially available Winkler, and the U-Gas technology developed at the Institute of Gas Technology in Chicago.

8. Chemistry

Coal reactions, which on heating are important to the production of coke and synthetic fuels, are complicated by its structure.

Mature ($>$75 wt % C) coals are built of assemblages of polynuclear ring systems connected by a variety of functional groups and hydrogen-bonded cross-links (Fig. 4) (3,4,7,21). The ring systems themselves contain many functional groups. These polynuclear coal molecules differ one from another to some extent in the coal matter. For bituminous coal, a tarlike material occupies some of the interstices between the molecules. Generally, coal materials are nonvolatile except for some moisture, light hydrocarbons, and contained carbon dioxide. The volatile matter produced on carbonization reflects decomposition of parts of the molecule and the release of moisture. Rate of heating affects the volatile matter content such that faster rates give higher volatile matter yields.

Coal composition is denoted by rank. Carbon content increases and oxygen content decreases with increasing rank. Table 8 gives a listing of the empirical formulas in terms of hydrogen, oxygen, nitrogen, and organic sulfur per 100 carbon atoms for a set of eight premium coal samples.

The surface of coal particles undergoes air oxidation, a process that may initiate spontaneous combustion in storage piles or weathering with a loss of heating and coking value during storage. Combustion produces oxides of sulfur and nitrogen as well as carbon dioxide and water vapor. The SO_x results from oxidation of both organic sulfur and inorganic forms such as pyrite. Nitrogen

Table 8. **Empirical Composition of Argonne Premium Coal Samples**[a]

Coal		Composition, atoms/100 atoms C			
Type	From	H	O	N	S
	Nonbituminous coals				
lignite	Beulah-Zap	79.5	20.9	1.4	0.4
subbituminous	Wyodak-Anderson	85.6	18.0	1.3	0.2
	Bituminous coals				
high volatile 1[b]		77.3	13.1	1.5	1.2
high volatile 2[c]		85.7	10.8	1.7	0.2
high volatile 3[d]		76.3	8.9	1.6	0.3
high volatile 4[e]		76.7	8.0	1.7	0.4
medium volatile	Upper Freeport	66.0	6.6	1.6	0.3
low volatile[f]		58.5	2.0	1.2	0.2

[a] Ref. 71.

[b] Illinois no. 6 coal.

[c] Blind Canyon coal.

[d] Lewiston-Stockton coal.

[e] Pittsburgh coal.

[f] Pocahontas no. 3 coal.

oxides are formed primarily from the nitrogen in the coal during high temperature combustion, rather than from the air used for combustion.

Partial oxidation as carried out in gasification produces carbon monoxide, hydrogen gas, carbon dioxide, and water vapor. The carbon dioxide reacts with hot carbon from the coal to produce carbon monoxide, and steam reacts with the carbon to produce carbon monoxide and hydrogen. The hydrogen can react with carbon through direct hydrogen gasification:

$$C + 2\,H_2 \rightarrow CH_4$$

at high hydrogen pressure, frequently 6.9 MPa (1000 psi) and moderate temperatures of 650–700°C. Methane may also be produced from

$$CO + 3\,H_2 \rightarrow CH_4 + H_2O$$

in a nickel-catalyzed reactor. This latter reaction is highly exothermic and is used to provide steam for the process. The correct 3:1 ratio of hydrogen to carbon monoxide is achieved using the water-gas-shift reaction:

$$CO + H_2O \rightarrow H_2 + CO_2$$

A mixture of CO and H_2, called synthesis gas, may also be used in other catalytic reactors to make methanol (qv) or hydrocarbons (qv):

$$CO + 2\,H_2 \rightarrow CH_3OH$$

$$n\,CO + 2n\,H_2 \rightarrow (CH_2)_n + n\,H_2O$$

Surface oxidation short of combustion, or using nitric acid or potassium permanganate solutions, produces regenerated humic acids similar to those extracted from peat or soil. Further oxidation produces aromatic acids and oxalic acid, but at least one-half of the carbon forms carbon dioxide.

Treatment with hydrogen at 400°C and 12.4 MPa (1800 psi) increases the coking power of some coal and produces a change that resembles an increase in rank. Hydrogenation using an appropriate solvent liquefies coal. Noncatalyzed processes primarily produce a tarlike solvent-refined-coal used as a boiler fuel. Catalysts and additional hydrogen were used in the H-Coal process developed by Hydrocarbon Research, Inc. to produce a higher quality liquid product. A 450 t/day plant was built in Catlettsburg, Kentucky, to demonstrate this process by making a coal-derived refinery feedstock. The reactor used a catalyst suspended in a process derived liquid or ebulated bed. Hydrogen reactions over short (0.1–2 s) times with very rapid heating produce a range of liquids such as benzene, toluene, xylene, and phenol. A less rapid heating and lower maximum temperatures permit removal of some sulfur and nitrogen from the coal (72). These efforts have not been commercialized.

Treatment of coal with chlorine or bromine results in addition and substitution reactions. At temperatures up to 600°C chlorinolysis produces carbon tetrachloride, phosgene, and thionyl chloride (73). Treatment with fluorine or chlorine trifluoride at atmospheric pressure and 300°C can produce large yields of liquid products.

Hydrolysis using aqueous alkali has been found to remove ash material including pyrite. A small pilot plant for studying this process was built at the Battelle Memorial Institute in Columbus, Ohio (74) and subsequently discontinued. Other studies have produced a variety of gases and organic compounds such as phenols, nitrogen bases, liquid hydrocarbons, and fatty acids totaling as much as 13 wt% of the coal. The products indicate that oxidation and other reactions as well as hydrolysis take place.

The pyritic sulfur in coal can undergo reaction with sulfate solutions to release elemental sulfur (see SULFUR REMOVAL AND RECOVERY). Processes to reduce the sulfur content of coal have been sought (75). The reaction of coal and sulfuric acid has been used to produce cation exchangers, but it was not very efficient and is no longer employed. Efforts have turned to the use of hot concentrated alkali in a process called Gravimelt.

Many of the products made by hydrogenation, oxidation, hydrolysis, or fluorination are of industrial importance. Concern about stable, low cost petroleum and natural gas supplies is increasing the interest in some of the coal products as upgraded fuels to meet air pollution control requirements as well as to take advantage of the greater ease of handling of the liquid or gaseous material and to utilize existing facilities such as pipelines (qv) and furnaces. A demonstration plant was built in North Dakota for conversion of coal to methane, also known as substitute natural gas (SNG) production. This plant, operated by Great Plains Gasification Associates and in use at this writing, may be converted to produce methanol instead of methane (see GAS, NATURAL). A chemistry based on the conversion of synthesis gas has been developed and applied extensively in South Africa to the production of liquid fuels and many other products. A small-scale production is used in the manufacture of photographic film materials from coal-derived synthesis gas in the Eastman Kodak plant in Kingsport, Tennessee. However, the principal production of chemicals from coal involves the by-products of coke manufacturing.

8.1. Reactions of Coal Ash. Mineral matter impurities have an important effect on the utilization of a coal. One of the constituents of greatest concern is pyrite because of the potential for sulfur oxide generation on combustion. The highest concentrations of pyrite are associated with coal deposition under marine environments, as typified by the Illinois Basin, including parts of Illinois, Indiana, and Kentucky. Additionally, the mineral matter has a tendency to form sticky deposits in a boiler. This tendency is most pronounced using mixtures that are rich in water-soluble alkalies such as are found in the Western Plains states. Coals from North Dakota, South Dakota, Wyoming, and Montana are typically low in the sulfur-bearing constituents and therefore otherwise desirable as fuels.

Coal deposits from east of the Mississippi River generally have acidic mineral constituents, ie, they are richer in silica and alumina and tend to produce higher melting ash mixtures. These materials do not soften until >1000°C and have limited problems with deposition on the inside walls of the boiler (slagging) or on the superheater tubes inside the boiler (fouling).

Coal ash passes through many reactors without significant chemical change. High temperature, exceeding the ash-softening temperature for the coal, permits reactions of the simpler ash constituents to form more complex species. Molten ash behavior affects slagging and ash removal. Correlations of viscosity have been made with a variety of chemical parameters, and descriptions based on acid–base chemistry appear to correlate with observed effects (74). Iron may be interconverted between the Fe(II) and Fe(III) states. Significant reduction in viscosity occurs as ferrous concentrations increase.

Corrosion of boiler tubes appears to be initiated in some cases with the formation of a white layer of general composition $(Na,K)_3Al(SO_4)_3$. Conditions for initiation of the deposit are favored by coals having high alkali and sulfur contents. The white layer bonds to the tubes and permits growth of ash deposits that insulate the layer and permit further corrosion.

8.2. Plasticity of Heated Coals.

Coals having a certain range of composition associated with the bend in the Seyler diagram (Fig. 3) and having 88–90 wt% carbon soften to a liquid condition when heated (4,6). These materials are known as prime coking coals. The soft condition is somewhat reversible for a time, but does not persist for many hours at 400°C, and is not observed above ~550°C if the sample is continuously heated as in a coking process. Continuous or lengthy heating result in degradation of coal matter, releasing vapors and resulting in polymerization of the remaining material. The coal does not behave like a Newtonian fluid and only empirical measurements of plasticity can be made (see RHEOLOGICAL MEASUREMENTS). About 10–30% of the coal becomes liquid, having a melting point <200°C, and this molten material plasticizes the solid matrix remaining.

The molten part of a vitrinite is similar to the gross maceral, and a part of the maceral is converted to a form that can be melted after heating to 300–400°C. The molten material is unstable and forms a solid product (coke) >350°C at rates that increase with temperature. The decomposition of the liquid phase is rapid for lower rank noncoking coals, and less rapid for prime coking coals. The material that melts resembles coal rather than tar and, depending on rank, only a slight or moderate amount is volatile.

The fluidity of coal increases and then decreases at a given temperature. This has been interpreted in terms of reaction sequence of coal–fluid coal–semicoke. In the initial step, a part of the coal is decomposed to add to that which normally becomes fluid. In the second step, the fluid phase decomposes to volatile matter and a solid semicoke. The semicoke later fuses accompanied by evolution of additional volatile matter to form a high temperature coke.

Formation of a true coke requires that the fluid phase persist long enough during heating for the coal pieces to form a compact mass before solidification occurs from the decomposition. Too much fluidity leads to an expanded froth owing to formation of dispersed bubbles from gas evolution in the fluid coal. Excess bubble formation results in a weak coke. The porous nature of true coke is caused by the bubble formation during the fluid phase. The strength of semicoke is set by the degree of fusion during the fluid stage and the thickness of the bubble walls formed during the frothing. In the final conversion to a hard high temperature coke, additional gas evolution occurs while the solid shrinks and is subjected to thermal stresses. The strength of the resultant coke and the size of the coke pieces are strongly affected by the crack structure produced as a result of the thermal stresses. Strong large pieces of coke are desirable to support the ore burden in blast furnaces.

Several laboratory tests (3,6) are used to determine the desirability of a coal or blend of coals for making coke. These are empirical and are carried out under conditions that approach the coking process. The three properties that have been studied are swelling, plasticity, and agglomeration.

Several dilatometers have been developed to determine the swelling characteristics of coals. The sample is placed in a cylindrical chamber with a piston resting on the coal surface. The piston motion reflects coal volume changes and is recorded as a function of temperature with a constant heating rate. When the coal first softens, contraction is caused by the weight of the piston on softened coal particles that deform to fill void spaces. Swelling then takes place when the particles are fused sufficiently to resist the flow of the evolving gases. The degree of swelling depends on the rate of release of volatile matter and the plasticity of the coal. The mass stabilizes at 450–500°C as the semicoke hardens. The shapes of the curves depend on the dilatometers. Curves obtained using the Hofmann apparatus have been classified into four main types that permit distinguishing coals having the optimum softening and swelling properties for production of a strong coke (76). Types A and C, and to some extent type D, can soften so that curves like those shown in Figure 7 can be obtained.

Free-swelling tests are commonly used to measure a coal's caking characteristics. A sample of coal is packed in a crucible or tube, without compaction, and heated at a fixed rate to ~800°C. Infusible coals distill without changing appearance or state of agglomeration. The fusible coals soften, fuse, and usually swell. The profile of the resultant coke is compared to a series of reference

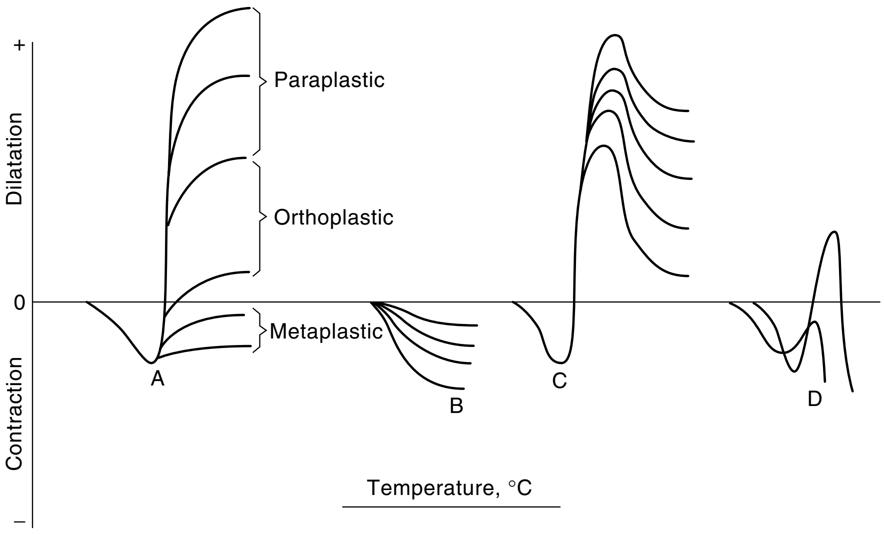


Fig. 7. Coal classification system according to Hofmann where A = Eu plastic; B = plastic; C = Per plastic; and D = Fluido plastic.Courtesy of Centre d'Etudes et Recherches des Charbonnages de France and Brennstoff-Chemie.

profiles so that a swelling index can be assigned. The profiles represent indexes between 0 and 9. The best cokes come from coals having indexes between 4 and 9.

The Gray-King assay, primarily carried out in Europe and the United Kingdom, is obtained from a similar test. The coal is heated to 600°C in a horizontal tube. Standard photographs are used to compare general appearance, profile, and size of the coke mass. Before testing, the more fusible coals are mixed with varying amounts of a standard electrode carbon of carefully selected size. A nonuniform scale termed A-F and G-G9 has been developed from the coke appearance for low swelling coals or from the amount of carbon required to give a standard appearance for the high swelling coals. The U.K. National Coal Board Rank Code Numbers are partly assigned on the Gray-King assay and partly on the volatile matter. The Gray-King assay procedure can also permit evaluation of yields of tar, gas, and liquor.

Plasticity can be studied using a device known as the Gieseler plastometer. A constant torque is applied to a shaft with rabble arms imbedded in coal in a crucible heated at a fixed rate. The rate of rotation of the shaft indicates the fluidity of the coal and is plotted as a function of the coal temperature. These curve, as shown in Figure 8, has a well-defined peak for coking coals usually near 450°C. Softening occurs at 350–400°C. At a normal heating rate of 3°C/min, the fluid hardening may be complete by 500°C.

Several agglutinating and agglomerating tests that indicate the bonding ability of the fusible components and depend on the crushing strength of a coke button produced, in some cases with addition of inert material, are also used. The Roga agglutinating test, developed in Poland, provides one of the criteria of the Geneva International Classification System. The coal sample is

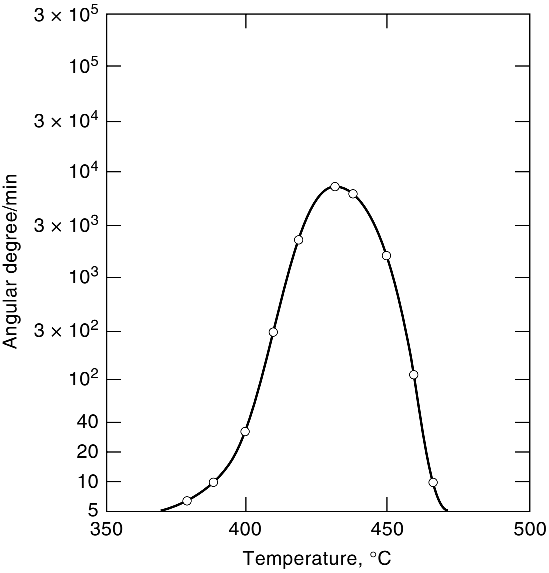


Fig. 8. Plasticity curve obtained using the Gieseler plastometer. Heating rate is 2°C/min. Courtesy of Centre d'Etudes et Recherches des Charbonnages de France.

mixed with carefully sized anthracite, compacted, and heated to 850°C in 15 min. The part of the product that passes through a 1 mm (~18 mesh) screen is weighed, and a rotating drum further degrades the product. Roga indexes from 0–70 have been determined.

A coherent plastic layer from a few millimeters to 2–3 cm thick separates the semicoke and coke from the unfused coal in the coke oven. Coking properties are assessed in Russia and some other countries by a measurement of the thickness of this plastic layer. A standardized test widely used in eastern Europe is the best known of this type (6) and involves a penetrometer used to measure the thickness of the plastic layer in a column of coal heated from the bottom. The various standard tests give results that are similar but do not give close correlations with each other.

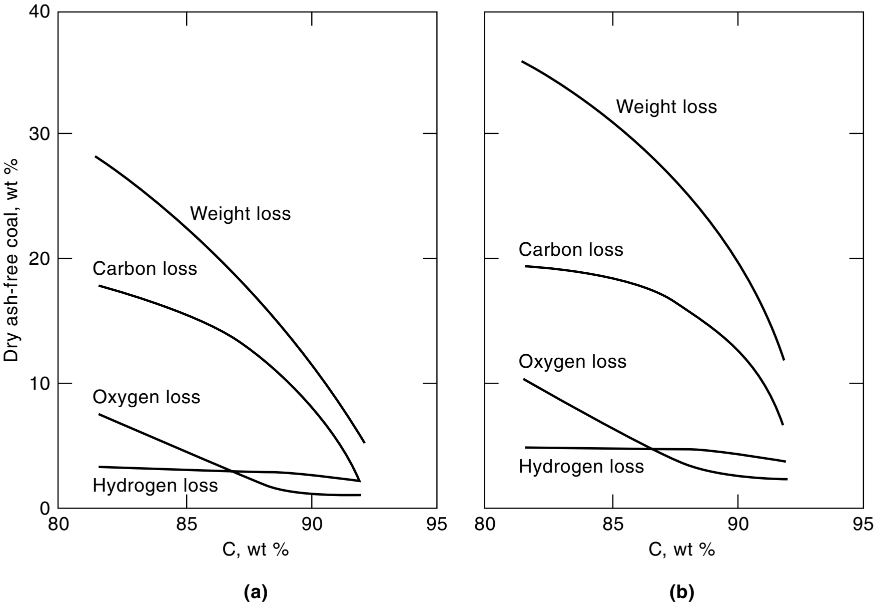
The behavior of different polymerizing and gas-relating materials has been used to relate the plastic behavior of coal with known kinds of chemical change (3). The plastic nature of coal matter is determined by the competition between the reactions that generate the liquid phase, and those that convert it to semicoke. In general for vitrinites the greater the heating rate, the greater the fluidity or plasticity and the dilatation. Inertinite essentially does not contribute to the plastic properties of the coal. Exinite becomes fluid when heated, but also rapidly devolatilizes instead of forming semicoke, has little value as a binder, and can increase the fluidity to an undesirable extent.

8.3. Pyrolysis of Coal. Most coals decompose below temperatures of ~400°C (5,6), characteristic of the onset of plasticity. Moisture is released near 100°C, and traces of oil and gases appear between 100 and 400°C, depending on the coal rank. As the temperature is raised in an inert atmosphere at a rate of 1–2°C/min, the evolution of decomposition products reaches a maximum rate near 450°C, and most of the tar is produced in the range of 400–500°C. Gas evolution begins in the same range but most evolves >500°C. If the coal temperature in a single reactor exceeds 900°C, the tars can be cracked, the yields are reduced, and the products are more aromatic. Heating beyond 900°C results in minor additional weight losses but the solid matter changes its structure. The tests for volatile matter indicate loss in weight at a specified temperature in the range of 875–1050°C from a covered crucible. This weight loss represents the loss of volatile decomposition products rather than volatile components.

A predictive macromolecular network decomposition model for coal conversion based on results of analytical measurements has been developed called the functional group, depolymerization, vaporization, cross-linking (FG-DVC) model (77). Data are obtained on weight loss on heating (thermogravimetry, tg) and analysis of the evolved species by Fourier transform infrared (FTir) spectrometry. Separate experimental data on solvent swelling, solvent extraction, and Gieseler plastometry are also used in the model.

Six factors form the basis of this model: (*1*) the decomposition of functional group sources in the coal yield the light gas species in thermal decomposition. The amount and evolution kinetics can be measured by tg/Ftir spectrometry and the functional group changes by Ftir and nmr; (*2*) the decomposition of a macromolecular network yields tar and metaplast. The amount and kinetics of the tar evolution can be measured by tg/Ftir and the molecular weight by field ionization mass spectrometry (Fims). The kinetics of metaplast formation and destruction can be measured by solvent extraction, by Gieseler plastometry, and by proton magnetic resonance thermal analysis (pmrta); (*3*) the molecular weight distribution of the metaplast depends on the network coordination number, ie, the average number of attachments on aromatic ring clusters. The coordination number can be determined by solvent swelling and nmr; (*4*) the network decomposition is controlled by bridge breaking. The number of bridges broken is limited by the available donatable hydrogen; (*5*) the network solidification is controlled by cross-linking. The changing cross-link density can be measured by solvent swelling and nmr. Cross-linking appears to occur with evolution of both CO_2 prior to bridge breaking and CH_4 after bridge breaking. Thus low rank coals, which form a lot of CO_2, cross-link prior to bridge breaking and are thermosetting. High volatile bituminous coals, which form little CO_2, undergo significant bridge breaking prior to cross-linking and become highly fluid. Weathering, which increases the CO_2 yield, causes increased cross-linking and lowers fluidity; and (*6*) the evolution of tar is controlled by mass transport in which the tar molecules evaporate into the light gas species and are carried out of the coal at rates proportional to their vapor pressure and the volume of light gases. High pressures reduce the volume of light gases and hence reduces the yield of heavy molecules having low vapor pressures. These changes can be studied using field ionization mass spectrometry.

Nature and Origin of Products. Volatile matter yields decrease with increasing coal rank. For slow heating, the final weight loss depends on the maximum temperature (Fig. 9). A variety of reactions take place and increasing temperatures provide the thermal energy required to break the stronger chemical



(a) **(b)**

Fig. 9. Composition of volatile matter as a function of rank (bright coals) at (**a**) 500°C and (**b**) 900°C. The wt% of C is on a dry ash-free basis of unheated coal. Courtesy of Institute of Fuel.

bonds. Much decomposition takes place in a short time (apparently $<$1s), but detection is limited by the rate of diffusion of the volatile products through the solid. The liquids result from initial decomposition and gases from decomposition of liquid material. Very rapid heating rates produce weight losses as high as 72% at 1900°C, suggesting that the intrinsic volatile matter is limited only by the vapor pressure of the initial pyrolysis fragments, and would be expected to increase with temperature and decreasing coal rank (78).

The residual solid or char heated to 500°C contains 3–3.5 wt% H and up to 5 wt% O. On further heating to 900°C the solid contains only 0.8 wt% H and up to 0.3 wt% O. An aqueous liquor is produced that comes from the moisture in the coal as well as hydroxyl and possibly other oxygen-containing groups. Phenols in the tar are probably derived from hydroxyl aromatic groups in the coal. The total tar yield appears to be proportional to the fraction of aromatic carbon in the coal (see TAR AND PITCH). Coke oven gas is obtained from a variety of reactions that include cracking some of the tar. The hydrogen in the gas is generated after the char is heated to 400°C, but most is evolved in the conversion of the fluid coal to semicoke or coke at 550–900°C. The steam in the ovens can also produce hydrogen on reaction with hot coke.

Pyrolysis Reaction Mechanisms. An overall picture of the pyrolysis process is generally accepted but the detailed mechanism is controversial. Information has been obtained from: the sequence of volatile material appearing in a coke plant as determined by gas chromatography; laboratory work simulating coking and minimizing secondary reactions by working in vacuum or sweeping with inert gas (79); laboratory studies using model organic compounds to determine the mechanism by which these materials are converted to coke, liquid, and gaseous products; and laboratory work with more complex materials, including specially synthesized polymers, to better provide a model of coal (4,80). Radioactive tracers have been used in the last two studies to follow the transformation to materials in the products (4). In the last study, gas generating materials were added to aid in simulating the swelling process. The dehydrogenation of coal, which can alter the distribution of products, also provides information regarding the formation mechanism (81). The mechanism of formation of metallurgical coke and its effect on coke properties has also been described (82).

The mechanism of coal pyrolysis has been discussed (77,79,82) and a table summarizing the various changes has been prepared (79). The early stages involve formation of a fluid through depolymerization and decomposition of coal organic matter containing hydrogen. Around 400–550°C aromatic and non-aromatic groups may condense after releasing hydroxyl groups. The highest yields of methane and hydrogen come from coals having 89–92 wt% C. Light hydrocarbons other than methane are released most readily $<$500°C; methane is released at 500°C. The highest rate for hydrogen occurs $>$700°C (77,79).

9. Resources

9.1. World Reserves.

Amounts of coal of some specified minimum deposit thickness and some specified maximum overburden thickness existing in the ground are termed resources. There is no economic consideration for resources, but reserves represent the portion of the resources that may be recovered economically using conventional mining equipment. The first inventory of world coal resources was made during the Twelfth International Geological Congress in Toronto in 1913. An example of the changes since 1913 can be seen from an examination of the coal resources for Canada. These were estimated to have been 1217×10^9 metric tons in 1913, based on a few observations and statistical allowance for all possible coalbeds to a minimum thickness of 0.3 m and to a maximum depth of 1220 m below the surface. In 1974, however, the estimate of solid fossil fuel resources (excluding peat) from the World Energy Conference gave the total resources as only 109×10^9 metric tons, and in 1986 the proven recoverable resources and estimated additional amount in place was given as 50×10^9 metric tons (83), <5% of the earliest figures (see FUEL RESOURCES).

Comprehensive reviews of energy sources are published by the World Energy Conference, formerly the World Power Conference at 6-year intervals (83). The 1986 survey includes reserves and also gives total resources. In 1986, the total proven reserves of recoverable solid fuels were given as 6×10^{11}metric tons. One metric ton is defined as 29.2×10^3MJ (27.7×10^6 Btu) to provide for the variation of calorific value in different coals. The total estimated additional reserves recoverable and total estimated additional amount in place are 2.2×10^{12} and 7.7×10^{12}metric tons, respectively. These figures are about double the 1913 estimates, primarily because significantly increased reserves have been indicated for Russia.

The part of the resource that is economically recoverable varies by country. The estimates made in the survey show that the proven recoverable reserves would last ~1200 years at the 1988 annual rate of production and that the estimated additional amount in place represent almost 1700 years at 1988 annual consumption.

In Table 9 (84), a somewhat different basis is used. The estimated total original coal resources of the world include beds 30-cm thick, and generally

Table 9. **Estimated Total Original Coal Resources of the World by Continents**[a]

Continent	Identified resources,[a] 10^9 t	Hypothetical[b] resources,[c] 10^9 t	Estimated total resources,[d] 10^9 t
Asia[c]	3,635[d]	6,362	9,997[e]
North America	1,727	2,272	3,999
Europe[f]	273	454	727
Africa	82	145	227
Oceania[g]	64	55	118
Central and South America	27	9	36
Totals	*5,808*	*9,297*	*15,104*

[a] Ref. 84.

[b] Original resources in the ground in beds 30 cm or more thick and generally <1299 m below surface but includes small amount between 1200 and 1800 m.

[c] Includes European Russia.

[d] Includes about 2090×10^9 metric tons in Russia.

[e] Includes $\sim 8600 \times 10^9$ in Russia.

[f] Includes Turkey.

[g] Australia, New Zealand, and New Caledonia.

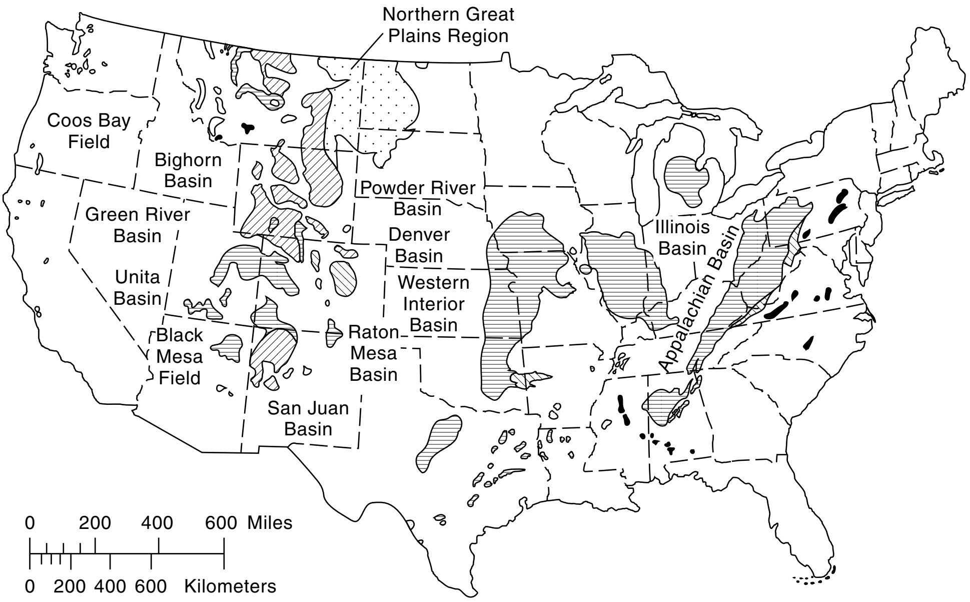


Fig. 10. Coal fields of the conterminous United States where ■ represents anthracite and semianthracite; ▨, low volatile bituminous coal; ≡/\, medium and high volatile bituminous coal; ▤, subbituminous coal; and ⊡, *lignite*(84).

<1220 m below surface but also include small amounts between 1229 and 1830 m. The data from column 1 are from earlier World Power Conference Surveys, whereas the figures for hypothetical resources (col. 2) and total estimated resources (col. 3) may be less reliable. This estimate represents about one-third more than the World Energy Conference Survey.

Reserves in the United States. Coal is widely distributed and abundant in the United States as indicated in Figures 10 and 11. A large portion of the coal fields contain lignite and subbituminous coal, however, and another portion of the coal is contained either in thin or deep beds that can be mined only with difficulty or great cost. Reserve estimates for the United States as of 1974 were on the order of 1.6×10^{12}t (84). The U.S. Geological Survey is currently completing a thorough revision of the previous estimates. This information is useful for showing the quantitative distribution of reserves, selecting appropriate areas for further exploration of development, and in planning coal-based industrial activity.

The reserves of 21 states have been classified by overburden thickness, reliability of estimates, and bed thickness. This coal represents ~60% of the total identified tonnage. Of this, 91% is <305 m from the surface, 43% is bituminous, and 58% is in beds thick enough to be mined economically.

On a uniform calorific value basis, coal constitutes 69% of the total estimated recoverable resources of fossil fuel in the United States. Petroleum and natural gas are ~7% and oil in oil shale, which is not as of this writing used as a fuel, is ~23%. The 1989 total recoverable reserves of coal are ~500 times the 1989 annual production (2), whereas the reserves of oil and gas are smaller,

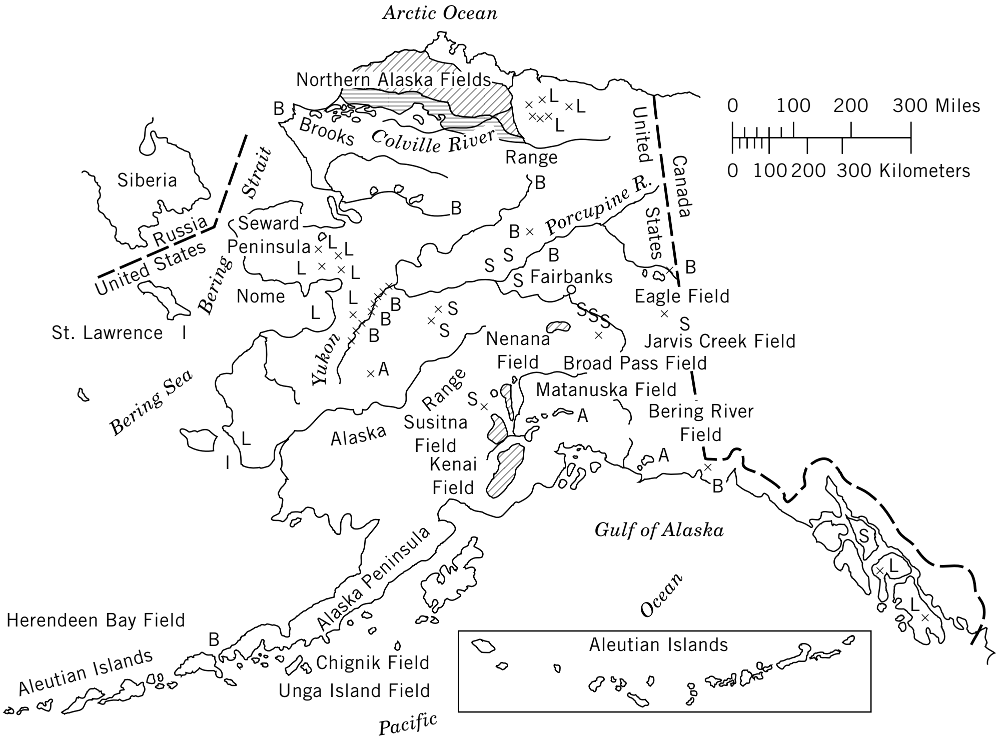


Fig. 11. Coal fields of Alaska where ▤ represents bituminous coal; ▨, subbituminous coal and lignite; and X is an isolated occurrence of coal of unknown extent. A = anthracite; B = bituminous; S = subbituminous; and L = lignite (84).

the production and consumption rate of oil and gas in the United States is three times that of coal.

9.2. Coal Production. In 1860 world coal production was 122×10^6t / year. Production increased to 1140×10^6t in 1913, giving a 4.2% annual average rate of increase. The rate has slowed and been erratic since that time. Statistical data on world coal production from 1860 to 1960 is given in (81). World coal and lignite production rose to $\sim 4.7 \times 10^9$t in 1988 (1).

United States Coal Production. Coal production in the United States, which dates back to 1702, started in earnest ~1820. It has increased with fluctuations to 617×10^6t in 1976, 755×10^6t in 1982, and 1099×10^6t in 1999 (2). In 1999 the United States produced 23.2% of the world's coal supply (1). U.S. coal production by state is given in Table 10 (2). Coal shipments in 2000 totaled 1048×10^6t to United States destinations. Of these, electric utilities received 894×10^6t; coke and gas plants 28×10^6t; and industrial, commercial, retail, and transportation 65×10^6t (2).

The demand for energy is continually increasing and the highest energy consumption in the world occurs in the United States. In 1989, consumption totaled 8.6×10^{13} MJ (81.3×10^{15}Btu) or 11.7 metric tons of coal-equivalent per

Table 10. **Coal Production by State, 1991, 1996–2000 (Thousand Short Tons)**

Coal-producing state and region	2000	1999	1998	1997	1996	1991	Percent change 1999–2000	Average annual percent change 1996–2000	Average annual percent change 1991–2000
Alabama	19,324	19,504	23,013	24,468	24,637	27,269	−0.9	−5.9	−3.8
Alaska	1,641	1,565	1,344	1,450	1,481	1,436	4.8	2.6	1.5
Arizona	13,111	11,787	11,315	11,723	10,442	13,203	11.2	5.8	−.1
Arkansas	12	22	24	18	21	52	−47.3	−13.7	−15.3
California						57			
Colorado	29,137	29,989	29,631	27,449	24,886	17,834	−2.8	4.0	5.6
Illinois	33,444	40,417	39,732	41,159	46,656	60,258	−17.3	−8.0	−6.3
Indiana	27,965	34,004	36,803	35,497	29,670	31,468	−17.8	−1.5	−1.3
Iowa						344			
Kansas	201	409	341	360	232	416	−50.8	−3.5	−7.8
Kentucky	130,688	139,626	150,295	155,853	152,425	158,980	−6.4	−3.8	−2.1
Total									
Eastern	*104,901*	110,043	*116,654*	*120,918*	*116,951*	*117,220*	*−4.7*	*−2.7*	*−1.2*
Western	*25,787*	*29,583*	*33,641*	*34,936*	*35,474*	*41,760*	*−12.8*	*−7.7*	*−5.2*
Louisiana	3,699	2,953	3,216	3,545	3,221	3,151	25.3	3.5	1.8
Maryland	4,546	3,837	4,060	4,160	4,093	3,773	18.5	2.7	2.1
Mississippi	902	18					nm[a]		
Missouri	436	392	372	401	710	2,304	11.3	−11.4	−16.9
Montana	38,352	41,102	42,840	41,005	37,891	38,237	−6.7	0.3	[bc]
New Mexico	27,323	29,156	28,597	27,025	24,067	21,518	−6.3	3.2	2.7
North Dakota	31,270	31,135	29,912	29,580	29,861	29,530	.4	1.1	.6
Ohio	22,269	22,480	28,048	29,154	28,572	30,569	0.9	−6.0	−3.4
Oklahoma	1,588	1,661	1,661	1,621	1,701	1,841	−4.4	−1.7	−1.6
Pennsylvania	74,619	76,399	81,036	76,198	67,942	65,381	−2.3	2.4	1.5
Total									
Anthracite	*4,572*	*4,753*	*5,231*	*4,678*	*4,751*	*3,445*	*−3.8*	*−.9*	*3.2*
Bituminous	*70,046*	*71,646*	*75,805*	*71,520*	*63,190*	*61,936*	*−2.2*	*2.6*	*1.4*
Tennessee	2,669	3,037	2,696	3,300	3,651	4,290	−12.1	−7.5	−5.1

Texas	49,498	53,072	52,583	53,328	55,164	53,825	−6.7	−2.7	0.9
Utah	26,656	26,373	26,075	26,683	27,507	21,945	1.1	9.8	2.2
Virginia	32,834	32,294	33,747	35,837	35,590	41,954	1.7	−2.0	−2.7
Washington	4,270	4,101	4,638	4,495	4,565	5,143	4.1	−1.6	−2.0
West Virginia	158,257	157,978	171,145	173,743	170,433	167,352	0.2	−1.8	9.6
Total									
Northern	*37,601*	*38,788*	*44,618*	*42,802*	*45,910*	*52,155*	*−3.1*	*−4.9*	*−3.6*
Southern	*120,656*	*119,191*	*126,527*	*130,941*	*124,523*	*115,196*	*1.2*	*9.8*	*0.5*
Wyoming	338,900	337,119	314,409	281,881	278,440	193,854	0.5	5.0	6.4
Appalachian Total[b]	419,419	425,573	460,400	467,778	451,868	457,808	−1.4	−1.8	−1.0
Interior Total[b]	*143,531*	*162,530*	*168,374*	*170,863*	*172,848*	*195,418*	*−11.7*	*−4.5*	*−3.4*
Western Total[b]	*510,661*	*512,328*	*488,762*	*451,291*	*439,140*	*342,758*	*0.3*	*3.8*	*4.5*
East of Miss. River	507,517	529,594	570,576	579,369	563,668	591,294	−4.2	−2.6	−1.7
West of Miss. River	566,094	570,837	546,960	510,563	500,188	404,690	−.8	3.1	3.8
U.S. Total	*1,073,612*	*1,100,431*	*1,117,535*	*1,089,932*	*1,063,856*	*995,984*	*−2.4*	*0.2*	*0.8*

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[a] nm/Not meaningful as value is >500 %.

[b] Data round to zero.

[c] For a definition of coal-producing regions, see Appendix C.

[d] *Notes*: Coal production excludes silt, culm, refuse bank, slurry dam, and dredge operations except for pennsylvania anthracite. Totals may not equal sum of components due to independent rounding.

[e] *Sources*: Energy Information Administration, Form EIA-7A, “Coal Production Report”; State Mining Agency Coal Production Reports; and/or U.S. Department of Labor, Mine Safety and Health Administration, form 7000-2, “Quarterly Mine Employment and Coal Production Report”.

Table 11. **Energy Source Usage in the United States, % Energy Basis**[a]

Source	1942	1952	1962	1972	1988	1991	1999
coal	68.7	45.2	32.5	25.6	23.5	23.1	21.4
petroleum	17.1	25.4	23.6	22.8	42.7	40.1	39.7
natural gas	9.0	23.3	38.3	44.4	23.1	24.7	23.3
hydroelectric	5.2	6.1	5.5	6.0	3.3	3.8	3.7
nuclear			0.1	1.2	7.1	8.0	8.1

[a] Ref. 2.

capita (85). World recoverable reserves were ~120 times the annual coal production in 1988 and ~10 times that for the additional reserves believed to be in place (1). Estimated coal consumption reduces the known recoverable reserves at ~1%/year. Whereas the use of bituminous coal is expected to continue to increase in terms of tonnage, the percentage of coal used in the United States has stabilized as shown in Table 11.

10. Sample Sources

Basic coal research requires a variety of coal samples of different ranks that workers may access using a minimum of effort. Coal sample banks fill this need. Moreover, over the past decades it has become evident that the quality of samples degrades from atmospheric oxidation and the degradation has limited the ability of researchers to compare results. The U.S. Department of Energy Office of Basic Energy Sciences has sponsored the Argonne Premium Coal Sample Program to permit the acquisition of ton-sized samples of each of eight different coals representing a range of coal ranks, chemical composition, geography, and maceral content (71).

The samples were collected and processed in a manner to avoid exposure to oxygen and control loss of moisture; then they were pulverized to convenient sizes for research, ie, <149 μ (−100 mesh) and <840 μ (−20 mesh); the entire ton was thoroughly mixed; and packaged in sealed glass ampules under nitrogen. These samples have been well characterized and are available in quantities that are expected to last for decades (Table 8).

Other coal sample banks are also in existence. The Penn State Sample Bank at Pennsylvania State University has the most diverse collection of samples (86). The Illinois Basin Coal Sample Program at the Illinois State Geological Survey specializes in samples from the Illinois Basin (87). The European Center for Coal Specimens, located in Eygelshoven in The Netherlands, has a significant collection of samples from the entire world (88). Each makes samples available in kilogram quantities.

11. Mining and Preparation

11.1. Mining.

Coal is obtained by either surface mining of outcrops or seams near the surface or by underground mining depending on geological conditions, which may vary from thick, flat seams to thin, inclined seams that are folded and need special mining methods. Coal mining has changed from a labor intensive activity to one that has become highly mechanized. In 2001, the average output per person per hour in underground mines in the United States was 4.2 tons. For surface mines, the output was 11.2 tons per person per hour (2).

Strip or open-pit mining involves removal of overburden from shallow seams, breaking of coal by blasting or mechanical means, and loading of the coal. The development of very large excavating equipment, including draglines, bulldozers, front-end loaders, and trucks, has been responsible for significantly increased production from strip mining.

The two methods of underground mining commonly used are room-and-pillar and longwall. In room-and-pillar mining the coal is removed from two sets of corridors that advance through the mine at right angles to each other. Regularly spaced pillars, constituting about one-half of the coal seam, are left behind to support the overhead layers in the mined areas. This method is used extensively in the United States and other nations having large reserves. The pillars may later be removed, leading to probable subsidence of the surface. Longwall mining is used to permit recovery of as much of the coal as possible (87). Two parallel headings are made 100–200-m apart and at right angles to the main heading. The longwall between the two headings is then mined away from the main heading. The equipment provides a movable roof support system that advances as the coal is mined and allows the roof to collapse in a controlled manner behind it. This method also leads to subsidence of the overhead layers.

Another method used in Europe for steeply inclined seams is called horizontal mining. Horizontal shafts are cut through rock below the coal seams. Vertical connections are then made to the seam to permit coal removal.

The mechanical equipment used in room-and-pillar underground mining usually involves a series of specific operations with continuous mining equipment. Continuous miners use rotating heads equipped with bits to pick or cut through the coal without blasting and load it into a shuttle car for conveying to a belt system.

11.2. Preparation. Coal preparation is of significant importance to the coal industry and to consumers (6,55,90). Preparation normally involves some size reduction of the mined coal and the systematic removal of some ash-forming material and very fine coal. The percentage of mined coal that is mechanically cleaned in the principal coal producing countries has risen during the past 30 years. There are a number of reasons for this. The most important in the United States is the increased use of continuous mining equipment. The nature of this operation tends to include inorganic foreign matter from the floor and ceiling of the seams, thus run-of-mine coal includes ~25% mineral matter. The size consist of the mined coal is also smaller when produced using continuous mining equipment. The depletion of the better quality seams, which are low in ash and sulfur, in many coal fields necessitates cleaning of the remainder. Moreover, the economic need to recover the maximum amount of salable coal has led to cleaning of the finer sizes that had previously been discarded. Stringent customer demands for coal meeting definite specifications, regulations requiring the removal of pyrites to control air pollution (qv), increased freight rates, and ash disposal costs all contribute to the upsurge in coal cleaning (see COAL CONVERSION PROCESS, CLEANING AND DESULFURIZATION).

In earlier times the coal was hand-picked for removal of larger pieces of refuse, but higher labor costs have made this method uneconomical. Mechanical separation methods are used, most of which depend on the difference in density between the coal and refuse. The washability characteristics of a coal determine the extent to which the refuse may be removed. The laboratory float-and-sink analysis gives information on the percentages and quality of the coal material occupying different density ranges. From this information, graphs are constructed showing the composite quantity and coal quality that can be obtained by cleaning at different specific gravities. This information is considered with the economic factors involved in the sale of the washed coal to choose an optimum method of cleaning. Cleaning plants are usually designed to handle the output of specific mines and to clean for a specific market. The plant includes various types of cleaning methods designed to move the different fractions through several cleaning circuits to maximize recovery.

In some areas, run-of-mine coal is separated into three products: a low gravity, premium-priced coal for metallurgical or other special use, a middlings product for possible boiler firing, and a high ash refuse. The complete preparation of coal usually requires several processes.

Cleaning Methods. Jig washing is the most widely used of all cleaning methods. A bed of coal particles is subjected to alternate upward and downward currents of water causing a moving bed of particles to stratify. The lighter clean coal particles go to the top and the heavier refuse particles to the bottom (see MINERALS RECOVERY AND PROCESSING). The heavy medium process is a simple float-and-sink one that is widely used for coarse coal cleaning. The medium is usually a suspension of pulverized magnetite, which is mixed to the desired specific gravity. This method is also used in cyclones for a wide range of coal cleaning.

By using trough washers, the coal is fed to a trough in a stream of water that carries the coal particles forward but allows heavier particles to sink and be removed. Washing tables are used for cleaning fine coal. A coal suspension in water flows across a slanted table that oscillates at right angles to the direction of flow. The heavier refuse particles settle onto the table and are trapped by riffles or bars, while the lighter coal particles are carried over the riffles in the current. If dry cleaning is used, the coal passes over a perforated, oscillating table through which air is blown. This method creates a dust problem, although it eliminates the need for drying the coal. In countries using hydraulic mining or underground dust suppression with water, there is limited opportunity for dry cleaning.

Froth flotation (qv) is the most important method for cleaning fine coal because very small particles cannot be separated by settling methods. Air is passed through a suspension of coal in water to which conditioning reagents, usually special oils, have been added. The oils are selected so that the coal particles preferentially attach themselves to the bubbles and separate from the refuse that remains in suspension.

Dewatering. The coal leaving the cleaning plant is very wet and must be at least partially dried to reduce freight charges, meet customer requirements, and avoid freezing. Draining on screens removes substantial amounts of water from larger coal, but other dewatering (qv) methods are required for smaller sizes having larger surface areas.

Vibrating screens and centrifuges are used for dewatering. For very fine coal, such as that obtained from flotation, vacuum filtration with a disk or drum-type filter may be used. Flocculants may be added to aid filtration (see FLOCCULATING AGENTS). They are also used for cleaning wastewater and pollution control. If very low ($\leq$2–3%) moisture contents are required, thermal drying must be used. A number of dryer types are available including fluidized bed; suspension; and rotary- and cascade-dryers. All of these are expensive to operate, however (see DRYING).

Storage. Storage of the coal may be necessary at any of the various steps in production or consumption, ie, at the mine, preparation plant, or consumer location. Electric utilities have the largest amounts of coal in storage, having stockpiles that frequently are able to meet 60–100 day normal demand thus protecting against delays, shortages, price changes, or seasonal demands (2).

For utilities, two types of storage are used. A small amount of coal in storage meets daily needs and is continually turned over. This coal is loaded into storage bins or bunkers. However, long-term reserves are carefully piled and left undisturbed except as necessary to sustain production.

Coal storage results in some deterioration of the fuel owing to air oxidation. Moreover, if inadequate care is taken, spontaneous heating and combustion may result. As the rank of coal decreases, it oxidizes more easily and must be piled more carefully. Anthracite does not usually present a problem.

The surface of the coal particles oxidizes or weathers resulting in cracks, finer particles, and reduced agglomeration all of which may destroy coking properties. If spontaneous heating takes place, the calorific value of the coal is reduced. Hot spots must be carefully dug out and used as quickly as possible. Without spontaneous heating and with good compaction, calorific value losses $<$1%/year have been recorded.

Coal piles are carefully constructed to exclude air or to allow adequate ventilation. The latter requires larger sizes, graded as 4 cm+ without fines, for avoiding heating by ventilation. For exclusion of air, mixed sizes provide fines to fill the gaps between larger pieces. Pockets of large sizes must not be allowed because these provide access for air. The coal should be compacted to maximize the bulk density of the coal pile.

Several approaches have been effective for storage: (*1*) large compacted layered piles where sides and top are sealed using an oil or asphalt (qv) emulsion. Four liters of oil seals 1 m^2 of coal; (*2*) large compacted layered piles where sides and top are covered first with fines to seal the pile and then with coarse coal to protect fines from wind and weather. The sides may slope at angles $\geq 30°$; (*3*) piles of compacted layers in open pits having tight sides so that the air has access only at the top; (*4*) sealed bins or bunkers in which airtight storage can be provided for smaller amounts of coal for long times; and (*5*) underwater storage in concrete pits. This is expensive and rarely used but effectively prevents deterioration although it introduces other problems related to handling wet coal.

Large compacted storage piles should be located on hard surfaces and not subject to flooding. A layer of fines may be put down first to facilitate recovery. Each layer of coal should be compacted after it is deposited. The top of the pile should have a slight crown to avoid water accumulation. Excessive heights should be avoided to prevent air infiltration caused by wind. Coal removal should be done in layers followed by compacting and smoothing the surface. Piles should be limited to the same rank of coal depending on the intended use.

12. Transportation

The usual means of transporting coal are railroad, barge, truck, conveyer belt from mine to plant, and slurry pipelines (2,4) (see TRANSPORTATION). In 2000, 1038×10^6t of coal was transported to United States destinations; of this, 65.5% was shipped by railroad, 14.3% by barge, 10.6% by truck, and 9.6% by conveyer, slurry pipeline, and other methods (2). Electric utilities consumed 80.04% of the coal transported in the United States in 2000 (2).

The unit train handles about one-half of rail transportation (71). Most unit trains consist of ~100 rail cars and are dedicated to coal haulage from the mine to the consuming plant. Almost no time is wasted at either the loading or unloading site as a result of efficient loading and unloading equipment. Diesel and diesel-electric trucks having capacities up to 320 t (2) handled the off-highway transport of coal.

A 437-km slurry pipeline, 46 cm in diameter, was started in 1970 to move coal from Arizona to southern Nevada. The coal is crushed and ground to the fineness needed for proper viscosity and settling. About 18–20% is −325 mesh (<44 μ), 35–45% is −100 mesh (<149 μ), and 0–2% is +14(<1070 μ) mesh. The solids content of the slurries has approached 70% using additives to stabilize the mixture. The slurry is dewatered with centrifuges before combustion of the coal.

For shipment in cold climates, a freezeproofing treatment using inorganic chemicals or oil spray is used. An oil spray on the inside of the coal cars is also effective at 3.8–5.7 L/car for four round trips of 1280 km each. Oil treatment has also been used for dustproofing; wind loss can be prevented by use of an asphalt emulsion on the top of rail cars (6).

Coal pipelines have been built in countries such as France (8.8 km), and Russia (61 km), and pipelines are also used for transporting limestone, copper concentrates, magnetite, and gilsonite in other parts of the world. The first coal pipeline, built in Ohio, led to freight rate reductions. The pipeline stopped operation after introduction of the unit train, used exclusively to transport coal from the mine to an electric power generation station.

Hydraulic transport is used in mines and for lifting coals to the surface in Russia, Poland, and France. Pneumatic transport of coal is used over short distances in power plants and steel mills. The longest (14.5 km) single flight conveyer belt in the world near Uniontown, Kentucky, has a capacity of 1360 t/h.

13. Economic Aspects

Table 12 gives the estimated destinations for United States bituminous coal exports in 1976, 1989, and 2000. About one-third of the bituminous coal exported from the United States in 1976 went to Europe, another third to Asia, and the remainder to North and South America. The pattern shifted to ~42% to North and South America, ~43% to Europe, 11% to Asia, and the balance to Africa (1).

Of the 1989 total, 65,128,000 t were metallurgical coal and 34,910,000 t were steam coal. Exports of coke from the United States in 1989 were 1,169,120 t, whereas anthracite exports were 745,749 t. Lignite exports were 163,628 t. In 1989 Canada produced 77,727,000 t, imported 16,160,000 t and exported 36,094,000 t. Japan is the principal recipient of Canadian exports. Selected coal exports and imports are given in Table 13.

The weighted average values for underground and surface mines decreased from \$28.24/t in 1984 to \$23.99/t in 1990 (2). Underground mine prices decreased from \$36.66/t to \$31.51/t, whereas surface mine prices dropped from \$22.70/t to \$18.72/t.

14. Analysis

Most countries have an official national organization, which is responsible for developing and maintaining standards for testing and analysis. The ASTM serves this purpose in the United States as does the British Standards (BS) Organization in the United Kingdom. In Geneva, the International Organization for Standardization (ISO) formed a committee (T.C.27), which is responsible for developing international standards. Each organization issues periodic updates of their standards. ASTM does this on an annual basis. The ASTM coal standard methods are each assigned a number, preceded by a D. The methods are periodically reviewed and revised. A two-digit number may follow the method number to indicate the year of the revision. BS standards of similar type also exist for most methods described. Details of an individual test may be found in the compilation for the respective source organization.

14.1. Sampling. The procedures for taking a sample, reducing the particle size of the sample, and separation of a smaller portion for later analysis are given in ASTM D2234 and D2013 (18) and BS1017. The procedures describe the minimum amount of sample needed to maintain a representative sample for analysis.

14.2. Size Analysis. ASTM and BS (18) provide a number of methods dealing with the size specifications and size analysis procedures including D197, D410, D311, and D431.

14.3. Moisture Holding Capacity. The bed or equilibrium moisture is the amount of moisture retained after equilibration at 96–97% rh at 30°C (D1412) (18). Total moisture is determined by air drying, crushing to smaller particle sizes and heating at 107°C to constant weight (D3302).

14.4. Analysis. The proximate analysis is based on determinations of volatile matter, moisture, and ash for a coal sample. Fixed carbon is then

Table 12. **U.S. Coal Exports by Destination, 1991, 1996–2000 (Thousand Short Tons)**

Continent and country of destination	2000	1999	1998	1997	1996	1991	Percent change 1999–2000	Average Annual percent change	
								1996–2000	1991–2000
North America									
Canada[a]	18,769	19,826	20,654	14,975	12,029	11,178	–5.3	11.8	5.9
Mexico	819	1,411	1,543	1,899	1,509	92	–41.9	–14.2	27.5
Other[b]	24	7	119	73	72	184	228.4	–23.8	–20.1
Total	*19,612*	*21,244*	*22,316*	*16,947*	*13,609*	*11,454*	*–7.7*	*9.6*	*6.1*
South America									
Argentina	204	3	324	325	304	429	NM	–9.4	–7.9
Brazil	4,536	4,442	6,475	7,455	6,540	7,052	2.1	–8.7	–4.8
Chile	53	43	51	146	574	135	21.7	–45.0	–9.9
Other[b]	30	32	185	288	87	45	–5.3	–23.4	–4.3
Total	*4,823*	*4,521*	*7,034*	*8,214*	*7,505*	*7,661*	*6.7*	*–10.5*	*–5.0*
Europe									
Belgium & Luxembourg	2,890	2,073	3,195	4,319	4,569	7,464	39.4	–10.8	–10.0
Bulgaria	919	522	989	1,114	1,387	946	75.9	–9.8	–.3
Denmark	77		274	350	1,316	4,658		–50.8	–36.6
Finland	317	233	463	662	704	530	36.2	–18.0	–5.5
France	3,044	2,522	3,192	3,398	3,852	9,509	20.7	–5.7	–11.9
Germany, FR	976	573	1,247	870	1,055	1,742	70.3	–1.9	–6.2
Hungary	72			[c]					
Iceland	53	51	39	54	62	45	4.1	–3.6	1.9
Ireland	502	868	1,150	637	765	1,313	–42.1	–10.0	–10.1
Italy	3,711	4,014	5,317	7,019	9,204	11,274	–7.5	–20.3	–11.6
Netherlands	2,623	3,432	4,516	4,825	7,058	9,625	–23.5	–21.9	–13.4
Norway	130	86	93	96	85	200	50.1	11.1	–4.7
Portugal	596	745	746	1,470	1,803	1,629	–20.0	–24.2	–10.6
Romania	489	322	1,097	2,244	1,512	1,147	52.0	–24.6	–9.0
Spain	2,686	2,472	3,156	4,134	4,093	4,694	8.6	–10.0	–6.0

Sweden	708	638	757	834	1,070	1,239	10.8	–9.8	–6.0
Turkey	1,809	795	1,592	2,092	2,167	2,186	127.6	–4.4	–2.1
United Kingdom	3,294	3,162	5,947	7,185	6,196	6,171	4.2	–14.6	–6.7
Yugoslavia, FR	73					979			–25.1
Other[b]	[c]	1	3	29	296	169	–82.3	–84.1	–53.0
Total	*24,969*	*22,508*	*33,773*	*41,331*	*47,193*	*65,520*	*10.9*	*–14.7*	*–10.2*
Asia									
China (Taiwan)	386	1,215	1,519	2,241	2,441	4,547	–68.3	–36.9	–24.0
Israel	62	603	527	593	1,202	651	–89.8	–52.4	–23.0
Japan	4,446	4,953	7,734	7,974	10,529	12,269	–10.2	–19.4	–10.7
Korea, Republic of	1,768	2,365	2,453	3,489	3,773	3,711	–25.2	–17.3	–7.9
Other[b]	40	21	78	201	36	611	91.7	3.0	–26.1
Total	*6,702*	*9,157*	*12,311*	*14,498*	*17,980*	*21,788*	*–26.8*	*–21.9*	*–12.3*
Oceania & Australia									
Other[b]	[c]	[c]	5	1	1	[c]	216.2	–27.4	7.4
Total	[c]	[c]	*5*	*1*	*1*	[c]	*216.2*	*–27.4*	*7.4*
Africa									
Algeria	296	317	343	264	177	522	–6.6	13.8	–6.1
Egypt	753	260	891	1,130	1,038	769	189.8	–7.7	–.2
Morocco	909		68	142	1,650	1,013	[c]	–13.8	–1.2
South Africa, Rep of	424	469	1,299	987	1,320	239	–9.5	–24.7	6.6
Other[b]	1	[c]	8	31	[c]	2	[d]	[c]	–13.2
Total	*2,383*	*1,046*	*2,609*	*2,554*	*4,184*	*2,545*	*127.9*	*–13.1*	*–.7*
Grand Total	*58,489*	*58,476*	*78,048*	*83,545*	*90,473*	*108,969*	[c]	*–10.3*	*–6.7*

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[a] Based on the U.S. Canada Free Trade Agreement, as of January 1990, the U.S. Department of Commerce began reporting statistics on U.S. exports to Canada based on information on imports provided monthly by the Canadian government.

[b] Includes countries with exports less than or equal to 50,000 short tons in 2000.

[c] Data round to zero.

[d] Not Meaningful as value is >500 %.

N77ote: Totals may not equal sum of components due to independent rounding.

Source: U.S. Department of Commerce, Bureau of the Census, “Monthly Report EM 545.”

Table 13. **World Coal Supply and Disposition, 1998 (Trillon Btu)**

Region/Country	Production	Imports	Exports	Apparent consumption
North America				
Canada	1,989	526	988	1,526
Mexico	211	68	0	275
United States	23,719	313	2,076	21,660
Total	*25917.72*	*906.98*	*3064.24*	*23460.63*
Central and South America				
Argentina	7	39	5	38
Brazil	67	509	0	568
Chile	7	132	1	147
Colombia	914	0	797	157
Peru	1	17	0	16
Venezuela	228	0	201	0
Other	0	24	0	23
Total	*1223.28*	*720.37*	*1002.97*	*949.85*
Western Europe				
Austria	12	122	0	130
Belgium	8	402	35	365
Bosnia and Herzegovina	16	0	0	16
Croatia	1	8	0	10
Denmark	0	203	4	233
Finland	0	131	0	145
France	155	496	15	666
Germany	2,585	724	10	3,395
Greece	339	37	2	372
Italy	1	456	3	461
Luxembourg	0	4	0	4
Macedonia, TFYR	72	10	0	81
Netherlands	0	551	226	348
Norway	9	43	8	45
Portugal	0	131	2	128
Serbia and Montenegro	387	12	0	399
Slovenia	56	3	0	59
Spain	335	327	14	658
Sweden	0	92	2	94
Turkey	572	312	0	893
United Kingdom	1,065	617	32	1,675
Other	0	85	0	87
Total	*5614.43*	*4767.92*	*353.99*	*10262.72*
Eastern Europe & Former U.S.S.R.				
Bulgaria	279	97	(0)	373
Czech Republic	961	44	113	901
Hungary	127	32	5	153
Poland	3,158	102	722	2,569
Romania	209	97	4	312
Slovakia	48	129	1	185
Belarus	0	18	0	18
Estonia	0	13	1	13
Kazakhstan	873	39	438	474
Kyrgyzstan	6	15	0	21
Latvia	0	3	0	3
Lithuania	0	6	0	6

Table 13 (*Continued*)

Region/Country	Production	Imports	Exports	Apparent consumption
Moldova	0	9	0	11
Russia	3,998	405	483	3,970
Tajikistan	0	2	0	2
Ukraine	1,613	215	49	1,781
Uzbekistan	43	0	0	43
Other	1	0	0	1
Total	*11317.77*	*1228.52*	*1815.28*	*10834.33*
Middle East				
Iran	24	18	0	41
Israel	0	251	0	242
Other	0	6	0	6
Total	*23.62*	*275.15*	*0.31*	*289.41*
Africa				
Algeria	1	16	0	20
Botswana	24	0	0	24
Congo (Kinshasa)	2	5	0	7
Egypt	9	48	11	47
Kenya	0	3	0	3
Malawi	1	0	0	2
Mauritius	0	2	0	2
Morocco	6	88	0	94
Mozambique	1	0	0	2
Niger	4	0	0	4
Nigeria	2	0	0	2
South Africa	5,273	31	1,622	3,396
Swaziland	7	0	0	7
Tunisia	0	2	0	2
Zambia	4	0	0	4
Zimbabwe	112	0	5	107
Other	0	1	0	1
Total	*5446.98*	*198.24*	*1638.54*	*3723.00*
Far East and Oceania				
Afghanistan	0	0	0	0
Australia	6,043	0	4,147	2,015
Bangladesh	0	4	0	4
Bhutan	0	1	0	0
Burma	1	0	0	2
China	24,595	33	1,025	23,781
Hong Kong	0	182	0	200
India	5,694	248	0	5,952
Indonesia	1,546	12	1,199	376
Japan	85	3,181	69	3,194
Korea, North	1,072	58	9	1,121
Korea, South	82	1,248	0	1,330
Malaysia	10	55	0	62
Mongolia	49	3	2	50
Nepal	0	3	0	3
New Caledonia	0	4	0	4
New Zealand	67	0	32	38
Pakistan	59	26	0	85
Philippines	23	81	0	101

Table 13 (*Continued*)

Region/Country	Production	Imports	Exports	Apparent consumption
Singapore	0	0	0	0
Taiwan	2	1,017	0	1,008
Thailand	242	43	0	290
Vietnam	250	1	75	121
other	0	1	0	1
Total	*39819.50*	*6199.75*	*6559.00*	*39737.60*
World Total	*89363.31*	*14296.93*	*14434.33*	*89257.53*

calculated by difference. Volatile matter is determined empirically by measuring the weight loss when coal is heated in a covered crucible at either 950°C (ASTM D3175 or D5142) or 900°C (BS).

The ultimate analysis gives the elemental composition in terms of C, H, N, S, and O (D3176, D3177, or D4239, and D3178) (18). The C and H analyses are based on oxidation of the sample in a tube and reaction of the gaseous products with absorbents to permit calculation of C and H content. The N analyses involves a Kjeldahl determination and the S involves oxidation and detection of SO_2 by ir detection, titration with base, or conversion to an insoluble sulfate to determine total S. Oxygen is determined by difference. Oxygen in the organic material is calculated by subtracting the dry, ash-free percentage of C, H, N, and organic S from 100. Sulfur forms are determined in a separate procedure. Sulfate and pyritic sulfur are determined separately and subtracted from total S to give organic S.

Other Elements. To determine chlorine, the sample is mixed with Eschka's mixture and burned to convert the chlorine to chloride or decomposed in an oxygen bomb (18). Chloride is determined by titration (D2361) or using a chloride selective electrode (D4208) (18) (see ELECTROANALYTICAL TECHNIQUES).

Phosphorus determination involves the conversion of phosphorus to soluble phosphate by digesting the coal ash with a mixture of sulfuric, nitric, and hydrofluoric acids (18). Phosphate is precipitated as ammonium phosphomolybdate, which may be reduced to give a blue solution that is determined colorimetrically or volumetrically (D2795) (18).

14.5. Calorific Value. To determine calorific value, a sample is placed in a bomb, pressurized with oxygen, and ignited. The temperature rise in the water bath of the calorimeter surrounding the bomb is used to determine the calorific value (D2015, D3286, or D1989) (18).

14.6. Ash Fusibility. A molded cone of ash is heated in a mildly reducing atmosphere and observed using an optical pyrometer during heating. The initial deformation temperature is reached when the cone tip becomes rounded; the softening temperature is evidenced when the height of the cone is equal to twice its width; the hemispherical temperature occurs when the cone becomes a hemispherical lump; and the fluid temperature is reached when no lump remains (D1857) (18).

15. Swelling and Coking Tests

For the free swelling index which is also known as the crucible swelling number (ASTM D720), a coal sample is rapidly heated to 820°C in a covered crucible. Then the profile of the resulting char is compared to a series of standard numbered profiles (18). For the Roga index weighed amounts of coal and standard anthracite are mixed and carbonized, and the product coke is tested in a Roga drum for its resistance to abrasion (89).

For the Gray-King coke-type assay test (91,92) coal is heated in a retort tube to 600°C and the product coke is compared to a series of standard cokes. For a strongly swelling coal, enough anthracite or electrode carbon is added to the coal to suppress the swelling. This method is primarily used in Europe.

In the Audibert-Arnu dilatometer test (91), a thin cylinder of compressed powdered coal contacting a steel piston is heated at a rate not $>5°$C/min. The piston movement is used to calculate the percent dilation.

15.1. Hardgrove Grindability Index and Strength Tests. A specially sized coal sample is ground in a specifically designed ball and race grinding mill (D409). The index is determined from the amount of coal remaining on a 74 mm (200 mesh) screen (18). The higher the index, the easier it is to grind the coal.

The drop shatter test indicates the resistance of a coal or coke to breakage on impact (see D440). A sample is dropped in a standard way a number of times from a specified height. For the tumbler test or abrasion index (ASTM D441), the coal or coke is rotated in a drum to determine the resistance to breakage by abrasion (18).

16. Health and Safety Factors

Coal mining has been a relatively dangerous occupation (2,91–93). During the period from 1961–1967 the average fatality rate in the United States for each million person hours worked was 1.05. In the 7 years after the passage of the Federal Coal Mine Health and Safety Act of 1969, the average fatality rate decreased to 0.58, and by 1989 the rate was 0.25 (2).

The rates of occupational injuries are reported per 200,000 employee-hours which correspond to ~100 employee-years. In 1989, the total for all mines was 11.84 or 11.84%. Over the 3-year periods before and after passage of the 1969 act, the rates for underground mining were 48.60% and 40.07%, respectively. The principal causes of fatalities are falling rock from mine roofs and faces, haulage, surface accidents, machinery, and explosions. For disabling injuries the primary causes are slips and falls, handling of materials, use of hand tools, lifting and pulling, falls of roof rock, and haulage and machinery (2).

16.1. Gases and Coal Dust Explosions. Gases can be hazardous in coal mines. Methane is of greatest concern, although other gases including carbon monoxide and hydrogen sulfide may be found in some mines. Methane must be detected and controlled because mixtures of air and 5–15% of methane are explosive.

The U.S. Mine Health and Safety Act of 1969 requires that a mine be closed if there is 1.5% or more methane in the air. The use of an electrical methane detection device is required. High capacity ventilation systems are designed to sweep gases from the cutting face and out of the mine. These systems remove all gases before they become harmful.

Whereas an explosion from methane tends to be localized, it may start coal dust explosions resulting in more widespread injury and loss of life. All coal breaking operations result in formation of fine coal particles; some are controlled with water during the mining operation. Breakage associated with hauling disperses dust, and dust accumulations can be made safe by rockdusting. Powdered limestone is spread over the mine surfaces to cover the dust.

16.2. Drainage. Some mines are located beneath subsurface streams, or the coal seams may be aquifers. These mines may become flooded if not continually pumped. In Pennsylvania anthracite mines as much as 30 tons of water may be pumped for each ton of coal mined (94).

Air or biological oxidation of pyrite leads to sulfate formation and dilute sulfuric acid in the mine drainage. This pollutes streams and the water supplies into which the mine water is drained. Means of controlling this problem are under study.

16.3. Other Hazards. Rocks falling from the roofs of mines used to cause the largest number of accidents. Roof bolts are placed in holes drilled into the roofs of working areas to tie the layers of rock together and thus prevent rock falls. A disease called pneumoconiosis, also called black lung, results from breathing coal dust over prolonged periods of time. The coal particles coat the lungs and prevent proper breathing.

16.4. Regulations. The U.S. Bureau of Mines, Mining Enforcement and Safety Administration (MESA) studies hazards and advises on accident prevention. MESA also administers laws dealing with safety in mines. Individual states may also have departments of mines to administer state standards.

The Federal Coal Mine Health and Safety Act set standards for mine ventilation, roof support, coal dust concentrations levels, mine inspections, and equipment. As a part of this comprehensive act, miners must receive medical examinations at employer expense, and payments are made from the U.S. government to miners who cannot work because of black lung disease.

17. Uses

17.1. Coal As Fuel. Coal is used as a fuel for electric power generation, industrial heating and steam generation, domestic heating, railroads, and coal processing. About 87% of the world's coal production is burned to produce heat and derived forms of energy. The balance is practically all processed thermally to make coke, fuel gas, and liquid by-products. Other uses of coke and fuel gas also contribute to coal consumption for heat. In the United States, coal use for power generation has increased to 86.1% in 1988, whereas coking coal use has dropped to 4.7% and the industrial/retail market declined to 9.2% (2).

Electric Power Generation. Coal is the primary fuel for thermal electric power generation. Since 1940 the quantity of bituminous coal consumed by electric utilities has grown substantially in each successive decade, and this growth is expected to continue for many years. Coal consumed by electric utilities increased from ~536×10^6t in 1981 to 991×10^6t in 2000 (2). The reasons for increased coal demand include availability, relative stability of decreasing coal prices, and lack of problems with spent fuel disposal as experienced in nuclear power plants (see NUCLEAR REACTOR TYPES).

The overall efficiency of electric power plants consisting of coal-fired boilers and steam turbines has plateaued at ~39%. The addition of pollutant control equipment has increased the internal power use on the stations and lowered the effective efficiency of the plant. The increased efficiencies have been achieved through use of larger units (up to 1500 MW) and higher pressures to 24.1 MPa (3500 psi) and reheat, but concerns about reliability and ability to match power generation and demand have kept plant sizes below these values. Maximum temperatures have not been increased because of the difficulties of corrosion owing to coal ash constituents, materials properties, and costs of better alloys. The advent of any future increases in efficiency depends on development of new systems of power generation, which might include fluid-bed boilers, gasification of coal to power a gas turbine having hot exhaust directed to a waste heat boiler in a combined cycle (gas turbine and steam turbine), or use of magnetohydrodynamics (qv) (see FURNACES,FUEL-FIRED).

Almost all modern large coal-fired boilers for electric power generation use pulverized coal. The cyclone furnace, built mainly for use in Germany and the United States, uses coarser pulverized coal. The ash is removed primarily as a molten slag from the combustor. Apparently, this design is no longer offered in the United States. This method of firing has not been accepted in the United Kingdom because of the higher softening temperature of the ash of the British coals. Stoker firing is generally limited to the smaller obsolete stand-by utility plants and generation plants used by industrial companies.

One significant advantage of pulverized coal boilers is the ability to use any kind of coal, including run-of-mine or uncleaned coals. However, with the advent of continuous mining equipment, the ash content frequently is ~25%, and some preparation is frequently practiced. There were 931 coal preparation plants in the United States in 1988, mainly in Kentucky, West Virginia, and Pennsylvania.

The advent of fluidized-bed boilers has enabled the size of units to go to 150 MW for commercial power generation and cogeneration in the last decade. This technology is displacing some stoker fired units at the low capacity applications and smaller pulverized coal units at the large size of the fluidized-bed range. Bubbling and circulating bed designs are used, and operating temperatures are in the 815–900°C range. Sulfur oxides are controlled using dolomite or limestone injection in the bed. Higher (2:1–5:1) calcium to sulfur ratios are needed for fluid-bed units than for wet scrubbing (1.0:-1) or spray dryers (1.2–1.5:1). Nitrous oxide emissions are higher than for other methods, possibly as a result of formation and oxidation of hydrogen cyanide (64). Several processes are being developed to reduce nitrogen oxides emissions. For example, three pressurized fluid-bed combustors are being demonstrated under the U.S. Department of Energy's Clean Coal Technology Program (15).

Integration of coal gasification and a combination of a gas turbine for power generation and a waste heat boiler for power generation is termed integrated gasification combined cycle (IGCC). Efficiencies are currently ~42% and promise to be higher as gas turbine technology improves (63). As of 1992 five plants using this technology have been announced in the United States. The IGCC technology uses sulfur gas removal techniques that result in higher removal rates than conventional scrubbers, in part because of the improved efficiency of scrubbing the more concentrated gases.

A primary concern in coal-fired power generation is the release of air pollutants. Limits on SO_2 output, 0.52 g/MJ equivalent of coal input to a new plant, have been established. For a bituminous coal of 27.9 MJ/kg, there is thus an upper limit of 0.72% sulfur content. Relatively few coals can meet this requirement. The U.S. Department of Energy indicated recoverable reserves of 420×10^9t in 1987 (2) that were categorized by sulfur content: 33.5% had 0.6% S or less, 15.4% had between 0.61% and 0.83% S, 16.1% had between 0.84 and 1.67% S, 12.4% had between 1.68 and 2.50% S, and 22.6% had >2.5% S. The lowest sulfur coal, ~86%, is found west of the Mississippi River, mainly in Montana and Wyoming, quite distant from the electric power demand centers in the East. A trend to utilization of the western coals has developed.

Industrial Heating and Steam Generation. The principal industrial users of coal include the iron (qv) and steel (qv) industry and the food, chemicals, paper (qv), engineering, bricks, and other clay products, and cement (qv) industries, and a group of miscellaneous consumers such as federal and local government installations, the armed services, and small industrial concerns. Most of the coal is burned directly for process heat, ie, for drying and firing kilns and furnaces, or indirectly for steam generation for process needs or for space heating, and for a small amount of electric power generation. Industrial coal usage in the United States has diminished significantly in past decades, especially among small users, because of the greater convenience in storing and handling gaseous and liquid fuels and the higher initial cost of coal-fired equipment.

Several developments are being pursued to utilize coal directly, ie, automation of controls, coal and ash handling equipment for smaller stoker and pulverized coal-fired units, design of packaged boiler units, and pollution control equipment. In the cement industry coal firing has been used, because the sulfur oxides react with some of the lime to make calcium sulfate in an acceptable amount.

17.2. Coal Processing to Synthetic Fuels and Other Products. The primary approaches to coal processing or coal conversion are thermal decomposition, including pyrolysis or carbonization (5,6), gasification (6), and liquefaction by hydrogenation (6). The hydrogenation of coal is not currently practiced commercially.

In the United States, the Clean Coal Technology program was created to develop and demonstrate the technology needed to use coal in a more environmentally acceptable manner. Activities range from basic research and establishing integrated operation of new processes in pilot plants through demonstration with commercial-scale equipment.

High Temperature Carbonization. High temperatures and long processing times are used in carbonizing coking coals in coke ovens or gas retorts. Besides metallurgical or gas coke the products include fuel gas, crude tar, light oils (benzene, toluene and xylene, referred to as BTX, and solvent naphtha), and ammonia gas (see COAL CONVERSION PROCESSES, CARBONIZATION).

Most coal chemicals are obtained from high temperature tar with an average yield over 5% of the coal which is carbonized. The yields in coking are ~70%

of the weight of feed coal. Tars obtained from vertical gas retorts have a much more uniform chemical composition than those from coke ovens. Two or more coals are usually blended. The conditions of carbonization vary depending on the coals used and affect the tar composition. Coal-tar chemicals include phenols, cresols, xylenols, benzene, toluene, naphthalene, and anthracene.

The largest consumer of coke is the iron and steel industry. In the United States, ~600 kg of coke is used to produce a metric ton of steel. Japanese equipment and practice reduce the requirement to 400–450 kg. Coke is also used to gas from the char in one vessel. The reducing gas converts iron oxide to iron in the upper two stages of a second vessel. Steam is converted to hydrogen and reoxidizes the iron in two stages in the lower half of the vessel.

None of these second- or third-generation processes has been commercialized, largely because of the relatively low price of available liquid and gaseous fuels.

A large commercial plant was completed in 1981 by a consortium of American Natural Gas and Peoples Gas, Light and Coke, and others for Mercer County, North Dakota. This plant has a design capacity of $3.7 \times 10^6 m^3$ (137.5×10^6 standard cubic feet [scf] of methane per day. The plant uses 14 Lurgi gasifiers and 12,700 t/day of lignite, 2,585 t/day oxygen, and 12,383 t/day of steam. The air separation plant is the largest in the hemisphere (see CYROGENICS; NITROGEN). The Phosam process is used for recovery of 113 t/day of ammonia, and the Stretford process was initially used for the recovery of 106 t/day of sulfur. The other products are used primarily as boiler fuels and include tar oil, naphtha, and crude phenol. The coal supply is Beulah-Zap lignite produced at an adjacent mine. The fine coal is removed before gasification and is sold to the neighboring electric utility (Basin Electric Power Cooperative) for use in the adjacent power plant and another plant about 48 km away. The cost of the gas was subsidized in the initial decade of operation. Future plans include the production of SNG and the use of the site for demonstration of a coal to methanol plant (95). In 1988 the ownership of the plant was transferred to the Dakota Gasification Co. of Bismarck. The agreement calls for operation of the plant until 2009 as long as revenues exceed expenses.

Processes for intermediate-Btu gas, ie, 9.3–18.6 $MJ \times m^3$ (250–500 $Btu \times ft^3$), or synthesis gas production were also developed. In the IGT U-Gas, or a similar Westinghouse process, crushed coal is fed into a fluidized-bed gasifier. Steam and oxygen enter the base of the bed. A part of these gases carry unreacted fines into a hot spout, which accelerates gasification and permits the ash to soften and particles to agglomerate. Ash agglomerates discharge below the spout. Product gases can be cleaned and pipelined as industrial fuel gas near 11.2 $MJ \times m^3$ (300 $Btu \times ft^3$). This technology has been offered by a consortium of Stone & Webster and Tampella Keeler. This group uses air-blown gasification and hot gas cleanup to lower the capital costs. The modification provides a lower heating value product.

Several plants use the Texaco partial oxidation gasifier developed as a modification of Texaco's oil consuming partial oxidation process. Pulverized coal falls through the reactor at high pressure and temperature to produce the gas which is then cleaned. The ratio of carbon monoxide to hydrogen can be adjusted by the water gas shift reaction as needed for a variety of chemical intermediates. This design was used in a plant to make chemical intermediates for the Eastman Kodak Co. in Kingsport, Tennessee. The Coolwater IGCC (integrated gasification-combined-cycle) demonstration plant in Southern California used this gasifier to provide fuel gas for boilers for electric power generation or for gas turbines for combined cycle power generation. The plant was technically successful but not able to compete economically. The scrubbing system removes a very high amount of the sulfur in the coal (96,97).

The high capital cost, ~$1500 × kW, is the principal deterrent to growth of the IGCC concept. The ability to remove up to 99% of the sulfur species from the combustion products make the IGCC an environmentally desirable option as make calcium carbide (see CARBIDES), from which acetylene is made. Synthesis gas for methanol and ammonia production is also made from gasification of coke.

Considerable research has been carried out to produce metallurgical grade coke from low rank bituminous and subbituminous coal, which is especially true in areas where coking coal reserves are becoming significantly depleted or are unavailable. The leading countries in this area of research are the United States (FMC Formcoke), Japan (Itoh process), and Germany (BFL process). These processes generally involve carbonization of crushed coal in fluidized beds, agglomerating the semicoke into conveniently sized balls with a binder, and calcining. The advantages of this technology include better heat transfer, shorter carbonizing time, continuous operation, and utilization of a much broader range of coals.

Low Temperature Carbonization. Lower temperature carbonization of lump coal at ~700°C, primarily used for production of solid smokeless fuel, gives a quantitatively and qualitatively different yield of solid, liquid, and gaseous products than does the high temperature processes.

Although a number of low temperature processes have been studied, only a few have been used commercially. These have been limited in the types of coal that are acceptable, and the by-products are less valuable than those obtained from high temperature processing. The Disco process is used in the United States to supply a limited amount of fuel to meet requirements of smoke ordinances. The British Coalite and Rexco processes produced substantial amounts of domestic smokeless fuel. Development of fluid-bed methods of carbonizing finer coal at ~400°C has been studied in the United Kingdom. A reactive char is briquetted without a binder to produce a premium open-fire smokeless fuel.

17.3. Gasification. Gasification of coal is used to provide gaseous fuels by surface and underground applications, liquid fuels by indirect liquefaction, ie, catalytic conversion of synthesis gas, and chemicals from conversion of synthesis gas. There are also applications in steelmaking (see COAL CONVERSION PROCESSES, GASIFICATION).

Gasifier Designs. A number of gasifiers are either available commercially or in various stages of development. These are described as fixed bed, fluidized bed, and entrained or rotating bed. The fixed bed involves an upward flow of reaction gas through a relatively stationary bed of hot coal. The gas velocity is slow enough to avoid blowing the coal out of the bed. The fluidized bed operates at higher gas velocities than the fixed bed and utilizes somewhat smaller particles. The entrained bed operates with parallel flows of reaction gas and finely pulverized coal particles to minimize reaction time and maximize throughput of product. The rotating bed is similar to a kiln, which operates with the coal entering at the upper part of the inclined kiln. Rotation avoids clinkering and exposes fresh surfaces to enhance completion of the reaction. The range of coals that may be used vary from one gasifier type to another with entrained flow gasifiers able to handle the widest range of raw coals. Fixed-bed gasifiers require mildly caking or noncaking feedstocks for normal operation.

The Lurgi fixed-bed gasifier operates using lump coal of a noncaking type with an ash composition chosen to avoid a sticky, partly fused ash in the reactor. A slagging version of this gasifier has been tested in Westfield, Scotland. Other fixed-bed gasifiers have similar coal requirements.

Fluidized-bed gasifiers typically require a coal feed of particles near 2–3 mm in diameter. Caking coals are to be avoided, because they usually agglomerate in the bed. This can be avoided using a pretreatment consisting of a surface oxidation with air in a fluidized bed. A useful fuel gas is produced. Examples of this type include the commercially available Winkler, and the U-Gas technology developed at the Institute of Gas Technology in Chicago. The latter is offered by a joint venture of Stone & Webster and Tampella Keeler. This system uses air-blown gasification and hot gas cleanup.

The Texaco gasifier and a similar unit developed by The Dow Chemical Company are pressurized entrained gasifiers. At the top, pulverized coal is mixed with reaction gas and is blown down into the gasifier. The reaction products leave from the side, and ash is blown down to a water pool where it is quenched. These units have operated at an Eastman Kodak facility in Kingsport, Tennessee and at the Coolwater power station in California for an integrated combined cycle power plant.

Pulverized coal is used in several entrained gasifiers and was studied in Germany before World War II. The Koppers-Totzek gasifier has been used commercially in different parts of the world. The original design used multiple (2 or 4) heads to feed coal, air or oxygen, and steam into an entrained atmospheric pressure reactor. Molten slag is discharged. The Babcock and Wilcox company also built an entrained bed gasifier for the DuPont Company at Belle, West Virginia, for chemical feedstock.

The Shell-Koppers-Totzek gasifier is also an entrained type. It can gasify lignite and subbituminous or bituminous coal. The coal is fed as a pulverized fuel, usually ground to 70% <74mm (−200 mesh) as used for pulverized coal fired boilers. Residence times are only a few seconds, therefore coal reactivity is important. The gasifier operates at >1650°C, at 2.2 MPa (22 atm) so that coal ash flows out of the gasifier as a molten slag. Coal ash composition must permit continuing molten ash flow. The joint development of the Shell Oil Co. and Koppers-Totzek led to a demonstration plant in The Netherlands having a gasifier for a 250-MW (50 cycle) integrated gasification combined cycle scheduled to begin operation in 1993. This is to be one of the first of the new generation of these plants to operate (95).

Surface Gasification Technology. Gasification of coal for fuel gas and chemical intermediate production has been developed commercially, and improvements in technology are being studied in a number of facilities. In the United States, the purpose of a number of programs has shifted from production of a substitute natural gas (methane) to electric power generation (qv) through the integrated IGCC plants. The interest in this use of coal results from the low emission levels that can be achieved and the potential for higher power generation efficiency.

Efficiencies of ~42% from natural gas to electricity have been indicated and can improve as the high temperature capabilities of turbines improve. Coal gasification would lower the overall energy efficiency but still give efficiencies greater than those with conventional coal-fired plants having typical emission control systems. Conventional power plants are able to produce electricity having heat rates of ~10 MJ × kWh and 90% SO_2 removal. The heat rates for IGCC plants are expected to be from 8 to 9.5 MJ × kWh having 99% SO_2 removal (96).

The Lurgi process (6) is the most successful complete gasification process for converting weakly caking coals as well as noncaking ones. The gasification takes place with steam and oxygen at 2–3 MPa (20–30 atm) to produce a 13.0–14.9 $MJ \times m^3$ (350–400 $Btu \times ft^3$) gas, which may be enriched with hydrocarbons to meet town gas specifications. The reactor is a slowly moving bed and is federal lump coal. Fine coal particles are usually removed before feeding to the gasifier.

The first commercial operation of the Lurgi process was in Germany in 1936 using brown coal. The reactor was modified to stir the coal bed to permit utilization of bituminous coal. One plant was built at the Dorsten Works of Steinkohlengas AG, and the Sasol plants were built in South Africa to provide synthesis gas for liquid fuels.

The gasifier for the 250 MW IGCC project in The Netherlands, scheduled to begin operation in 1993, is a 55 MW gas turbine with the balance of the power from a steam turbine. An Australian coal is to be used, and sulfur removal is expected to be 98.5% (96).

In the 1970s, a combined U.S. Federal government–American Gas Association program supported the development of second generation processes for making pipeline quality gas. In these processes coal is prepared, gasified, the gas is cooled, shifted if necessary to adjust the H_2/ CO ratio to ~3:1, the acid gases (H_2S and CO_2) removed, and then catalytic conversion to methane is carried out. Under this program the Institute of Gas Technology in Chicago developed the Hygas process in a 68 t/day pilot plant in which the gasifier at 6.9 MPa (1000 psi) accepts a coal slurry, dries it, goes through the first stage hydrogenation at 650–730°C, and second stage at 815–930°C before steam-oxygen gasification of the char to obtain high carbon utilization. The process also produces some benzene, toluene, and xylene, which were used in the pilot plant to make up the slurry. This process has been operated successfully using lignite, subbituminous, and bituminous coals.

The CO_2 Acceptor Process was also developed under this program by Consolidation Coal Co. in a 36-t/day pilot plant at Rapid City, South Dakota. Heat to drive the gasification process was provided by the reaction of calcined dolomite (MgO–CaO) and CO_2 produced in gasification of lignite or subbituminous coal using steam at 1 MPa (10 atm) and 815°C. The spent dolomite is regenerated at 1010°C in a separate vessel and returned to the gasifier. The process has operated successfully using lignite and subbituminous coal.

Still another process, called BI–GAS, was developed by Bituminous Coal Research in a 73-t/day pilot plant in Homer City, Pennsylvania. In this entrained-bed process, pulverized coal slurry was dried and blown into the second stage of the gasifier to contact 1205°C gases at ~6.9 MPa (1000 psi) for a few seconds residence time. Unreacted char is separated and recycled to the first stage to react with oxygen and steam at ~1650°C to produce hot gas and molten slag that is tapped.

The Synthane process was developed by the DOE at the Pittsburgh Energy Research Center. This fluidized-bed process operated at a ~6.9 MPa (1000 psi) and 980°C to gasify coal and produce some char. It used subbituminous coal. A third-generation process called Steam-Iron was also developed by the Institute of Gas Technology at a pilot plant in Chicago. This plant generates hydrogen from char produced in any gasification process. A gas producer uses air to make a reducing the Clean Air Act Amendment of 1991 phases in and the increased efficiency reduces the carbon dioxide emissions.

A similar design has been developed using a 161-MW plant by The Dow Chemical Company in its Plaquemine, Louisiana location. Destec, Inc. is a power subsidiary of The Dow Chemical Company and has joined with Public Service Of Indiana to build a new 230-MW plant near Terre Haute, Indiana. Operation is projected for 1995 (96).

Future large gasification plants, intended to produce $\sim 7 \times 10^6 m^3$ standard (250 million scf) of methane per day, are expected to be sited near a coal field having an adequate water supply. It is cheaper to transport energy in the form of gas through a pipeline than coal by either rail or pipeline. The process chosen is expected to utilize available coal in the most economical manner.

Underground Coal Gasification (UCG). Underground coal gasification is intended to gasify a coal seam *in situ*, converting the coal into gas and leaving the ash underground. This approach avoids the need for mining and reactors for gasification. UCG is presently considered most interesting for deep coal or steeply sloping seams. This approach involves drilling holes to provide air or oxygen for gasification and removal of product gases and liquids (98).

A low calorific value gas, which includes nitrogen from air, could be produced for boiler or turbine use in electric power production, or an intermediate calorific value gas containing no nitrogen for an industrial fuel gas, or synthesis gas for chemical and methane production could be provided. This approach, which has been studied in Russia, Europe, Japan, and the United States, is still noncommercial in part because it is not economically competitive.

Although many environmental and safety problems can be avoided using UCG, there is some concern about groundwater contamination as a result of the process (see GROUNDWATER MONITORING).

In the United States a program, carried out near Hanna, Wyoming for the Department of Energy, examined different approaches to gasification, including use of air and oxygen. Other programs under government sponsorship included use of a longwall generator at the Morgantown, West Virginia Energy Technology Center.

Industrial testing programs have been carried out by Gulf Research and Development Co. in Western Kentucky (99) on a coal seam at a depth of 32.6 m and a thickness of 2.7 m. The coal seam was excavated for study after the gasification program. Another program using Russian technology is being carried out by Texas Utilities Services in an East Texas lignite deposit.

A joint Belgian–West German program is aimed at gasifying seams ≥ 1000 m underground and using the gases for combined cycle electrical generating plants. If initial efforts are effective, hydrogen is to be pumped into hot coal seams to make substitute natural gas from the exothermic carbon–hydrogen reaction. Work using underground coal gasification has been most extensive in Russia where the technology has been applied to produce gas for four or five electrical generation stations. An institute was established in 1933 to study this process and has primarily studied air-blown gasification that produced a gas of $\sim$3.35–4.20 MJ/m^3 (90–113 Btu/ft^3) heat content. Other work has produced synthesis gas suitable for chemical production.

The chemistry of underground gasification has much in common with surface gasification; however, many of the parameters cannot be controlled because the reaction occurs in a remote site. Heat energy to drive the gasification comes primarily from carbon combustion to produce CO and then CO_2. Because many coal seams are also aquifers there is a considerable amount of water intrusion, which leads to steam generation at the expense of the reaction energy. As a result the rate of air or oxygen passage through the injection wells and seam are adjusted to maintain a low level of moisture in the product gas. The steam is beneficial for char gasification and some is consumed in the water gas shift reaction to produce H_2 and CO_2 from H_2O and CO. Some H_2 reacts with C to produce CH_4, which enhances the calorific value of the gas.

UCG is started by drilling wells to serve as injection points for oxidant and steam as well as collection points for product gases. Permeability of the coal seam is achieved by directional drilling, countercurrent combustion, electrolinking or hydraulic fracturing. Permeability is needed to provide a high rate of production with a minimum of pressure drop through the reaction zone. Low rank, ie, lignite and subbituminous, coals crack and shrink during gasification, rendering the seams more permeable. The bituminous coals swell and plug gas channels unless carefully preconditioned with preliminary oxidation to avoid this.

Liquid Fuels and Chemicals from Gasification of Coal. Gasification of coal using steam and oxygen in different gasifiers provides varying proportions of carbon monoxide and hydrogen. Operations at increasing pressures increases the formation of methane. Because mixtures of CO and H_2 are used as the start of chemical synthesis and methane is not wanted or needed for chemical processes, the conditions favoring its formation are avoided. The product gases may then be passed over catalysts to obtain specific products. Iron-based catalysts are used to produce hydrocarbons in the Fischer-Tropsch process, or zinc or copper catalysts are used to make methanol.

The Fischer-Tropsch process has not been economical in competitive markets. The South African Sasol plant (100) has operated successfully using the Kellogg and German Arge (Ruhr Chemie Lurgi) modification of the Fischer-Tropsch process. The original plant was designed to produce 227,000 t/year of gasoline, diesel oil, solvents, and chemicals from 907,000 metric tons of noncaking high ash subbituminous coal. The Lurgi gasification process is used to make the synthesis gas. The capacity of this plant was expanded substantially with Sasol II and Sasol III commissioned in 1980 and 1983 to meet transportation fuel needs for South Africa. The combined annual production capacity of the three Sasol facilities is $8 \times 10^6 m^3$ (50 million barrels) of liquid products (97).

The success of the Sasol project is attributed to the availability of cheap coal and the reliability of the selected components. Plants using Lurgi or Koppers-Totzek gasifiers for making chemicals are located in Australia, Turkey, Greece, India, and Yugoslavia, among other countries.

A variety of pilot plants using fluid-bed gasifiers have been built in the United States, Germany, and elsewhere. The Winkler process is the only one that has been used on a large scale. It was developed in Germany in the 1920s to make synthesis gas at atmospheric pressure. Plans were being made to develop a pressurized version. Plants using bituminous coal have been built in Spain and Japan with the atmospheric pressure gasifier.

Gasification and Metallurgy. Some interesting combinations of these technologies include direct reduction of iron ore and direct injection of coal into the blast furnace. In direct reduction, a reducing gas mixture of methane or carbon monoxide and hydrogen reduces iron ore pellets into elemental iron by reaction at 1000–1200°C. These pellets may later be used to feed steelmaking processes. In 1983, 45 plants having a capacity of 15×10^6 metric tons were in operation. Pulverized coal has been successfully injected into the tuyeres of a blast furnace of the Armco Co. in Middletown, Ohio to supplement coke (see IRON BY DIRECT REDUCTION).

17.4. Liquefaction. Liquefaction of coal to oil was first accomplished in 1914. Hydrogen was placed with a paste of coal, heavy oil, and a small amount of iron oxide catalyst at 450° and 20 MPa (200 atm) in stirred autoclaves. This process was developed by the I. G. Farbenindustrie AG to give commercial quality gasoline as the principal product. Twelve hydrogenation plants were operated during World War II to make liquid fuels (see COAL CONVERSION PROCESSES, LIQUEFACTION).

Imperial Chemical Industries in Great Britain hydrogenated coal to produce gasoline until the start of World War II. The process then operated on creosote middle oil until 1958. As of this writing none of these plants is being used to make liquid fuels for economic reasons. The present prices of coal and hydrogen from coal have not made synthetic liquid fuels competitive. Exceptions are those cases, as in South Africa, where there is availability of cheap coal, and fuel liquids are very important.

The Pott-Broche process (101) was best known as an early industrial use of solvent extraction of coal but was ended owing to war damage. The coal was extracted at ~400°C for 1–1.5 h under a hydrogen pressure of 10–15 MPa (100–150 atm) using a coal-derived solvent. Plant capacity was only 5 t/h with an 80% yield of extract. The product contained <0.05% mineral matter and had limited use, mainly in electrodes.

Solvent extraction work was carried out by a number of organizations in the United States. Pilot plants for producing SRC were built and initially sponsored by the Southern Company Services and Electric Power Research Institute in Wilsonville, Alabama in 1973 and built with Department of Energy sponsorship near Tacoma, Washington in 1974 having capacities of 5 and 45 t/day of coal input, respectively. The Wilsonville plant was closed in 1992 after many modifications from the initial design; the Tacoma plant is closed.

In the SRC work, coal was slurried with a process-derived anthracene oil and heated to 400–455°C at 12.4–13.8 MPa (1800–2000 psi) of hydrogen for

0–1 h. A viscous liquid was extracted. The product stream contains some hydrocarbon gases, and H_2S. The residue is gasified to generate hydrogen for the process. The remaining filtrate is separated into solvent, which is recycled, and SRC, a low ash, tarlike boiler fuel.

Heating value of the product (SRC) is ~37 MJ/kg (16,000 Btu/lb). Sulfur contents have been reduced from 2 to 7% initially to 0.9% and possibly less. Ash contents have been reduced from 8–20% to 0.17% (102). These properties permit compliance with environmental protection aging (EPA) requirements for SO_2 and particulate emissions. The SRC is primarily intended to be used as a boiler fuel in either a solid or molten form (heated to ~315°C). The solid has a Hardgrove index of 150 (103). Boiler tests have been successfully carried out using a utility boiler.

A series of process improvements have been developed at Wilsonville to produce high quality transportation fuels. Two integrated stages of liquefaction separated the initial coal dissolution from the hydrogenation to upgrade the product. This was known as SRC-II. An intermediate step, critical solvent deashing, was added to remove mineral matter to extend the life of catalysts used in hydrogenation. Later efforts involved the use of an ebulated bed developed by Hydrocarbon Research, Inc. (HRI) and eliminated the mineral matter removal between stages. Temperatures were lowered to reduce contamination of catalysts, which were also added to the first stage. This approach has been called the integrated catalytic two-stage liquefaction process (104).

Several processes progressed to demonstration scales but have not been commercialized, primarily because of economic inability to compete with available petroleum products. The H-Coal process developed by Hydrocarbon Research, Inc. was demonstrated at Catlettsburg, Kentucky using a 545 t/day plant and Department of Energy (DOE) support. The Exxon donor solvent liquefaction process was not commercialized either.

Processes for hydrogen gasification, hydrogen pyrolysis, or coking of coal usually produce liquid coproducts. The Hygas process produces about 6% liquids as benzene, toluene, and xylene. Substitution of petroleum residuum for the coal-derived process oil has been used in studies of coal liquefaction and offers promise as a lower cost technology (104).

17.5. Bioprocessing and Biotreatment of Coal.

The use of biotechnology to process coal to make gaseous and liquid fuels is an emerging field (105). Bacteria and enzymes have been studied to establish the technical feasibility of conversion. The earliest work was done on microbial decomposition of German hard coals (106). Reactors have been designed to use a variety of bacteria and fungi to break down the large molecular structure into smaller units that may be useful as intermediates (solubilization) or as liquid and gaseous fuels (conversion). Efforts have focused on lower rank coals, lignite or brown coal and subbituminous coal, because of greater reactivity. The conversion processes frequently introduce chemically combined oxygen through hydrolysis or related reactions to make the solid soluble in the reaction mixture as an initial step. Further reaction involves biological degradation of the resulting material to form gases or liquids.

The large-scale processing of coal is expected to involve plants similar to sewage treatment facilities in the handling of liquid and solid materials (see

WATER, SEWAGE). The reaction rates are substantially lower than those achieved in high temperature gasifiers and liquefaction reactors requiring much larger systems to achieve comparable coal throughput.

Biological processes are also being studied to investigate ability to remove sulfur species in order to remove potential contributors to acid rain (see AIR POLLUTION). These species include benzothiophene-type materials, which are the most difficult to remove chemically, as well as pyritic material. The pyrite may be treated to enhance the ability of flotation processes to separate the mineral from the combustible parts of the coal. Genetic engineering (qv) techniques are being applied to develop more effective species.

17.6. Other Uses. The quantity of coal used for purposes other than combustion or processing is quite small (2,6). Coal, especially anthracite, has established markets for use as purifying and filtering agents in either the natural form or converted to activated carbon (see CARBON). The latter can be prepared from bituminous coal or coke, and is used in sewage treatment, water purification, respirator absorbers, solvent recovery, and in the food industry. Some of these markets are quite profitable and new uses are continually being sought for this material.

Carbon black from oil is the main competition for the product from coal, which is used in filters. Carbon for electrodes is primarily made from petroleum coke, although pitch coke is used in Germany for this product. The pitch binder used for electrodes and other carbon products is almost always a selected coal tar pitch.

The preparation of pelletized iron ore represents a substantial market for coke and anthracite for sintering. Direct injection into the blast furnace of an auxiliary fuel, coal, or oil is now practiced to provide heat for the reduction and some of the reducing agent in place of the more expensive coke that serves these purposes. Some minor uses of coal include the use of fly ash, cinders, or even coal as a building material (see BUILDING MATERIALS, SURVEY); soil conditioners from coal by oxidizing it to humates (see SOIL STABILIZATION); and a variety of carbon and graphite products for the electrical industry, and possibly the nuclear energy program. The growth of synthetic fuels from coal should also provide substantial quantities of by-products including elemental sulfur, fertilizer as ammonia or its salts, and a range of liquid products. The availability of ammonia and straight-chain paraffins may permit future production of food from fossil fuels.

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