COAL GASIFICATION

1. Introduction

Coal gasification is the process of reacting coal with oxygen, steam, and carbon dioxide to form a product gas containing hydrogen and carbon monoxide. Gasification is essentially incomplete combustion. The chemical and physical processes are quite similar; the main difference being the nature of the final products. From a processing point of view the main operating difference is that gasification consumes heat evolved during combustion. Under the reducing environment of

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gasification the sulfur in the coal is released as hydrogen sulfide rather than sulfur dioxide and the coal's nitrogen is converted mostly to ammonia rather than nitrogen oxides. These reduced forms of sulfur and nitrogen are easily isolated, captured, and utilized, and thus gasification is a clean coal technology with better environmental performance than coal combustion.

Depending on the type of gasifier and the operating conditions gasification can be used to produce a fuel gas suitable for any number of applications. A low heating value fuel gas is produced from an air blown gasifier for use as an industrial fuel and for power production. A medium heating value fuel gas is produced from enriched oxygen blown gasification for use as a synthesis gas in the production of chemicals such as ammonia, methanol, and transportation fuels. A high heating value gas can be produced from shifting the medium heating value product gas over catalysts to produce a substitute or synthetic natural gas (SNG).

Coal gasification is presented by first describing the chemistry of the process and the coal characteristics that affect the processes. Coal gasification processes have been tailored to adapt to the different types of coal feedstocks available. The development of gasification is then presented from an historical perspective. This leads into the discussion of the types of gasifiers most commonly used and the process improvements made to meet the changing market needs. Complete gasification systems are then described including typical system configuration, required system attributes, and environmental performance. The current status, economics of gasification technology, and future of gasification are also discussed.

2. Coal Gasification Chemistry

In a gasifier, coal undergoes a series of chemical and physical changes as shown in Figure 1.

Each of the steps is described in more detail below. As the coal is heated most of the moisture is driven out when the particle temperature is $\sim 105^{\circ}$ C. Drying is a rapid process and can be essentially complete when the temperature reaches 300°C (1) depending on the type of coal and heating method used.

Devolatilization or pyrolysis accounts for a large percentage coal weight loss and occurs rapidly during the initial stages of coal heat up. During this process, the labile bonds between the aromatic clusters in coal are cleaved, generating

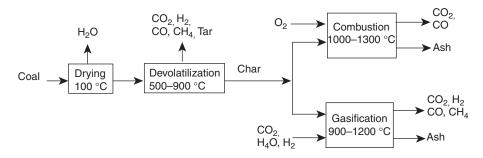


Fig. 1. Chemical and physical changes of coal.

fragments of molecular weight much smaller than coal. Fragments with low molecular weights vaporize and escape from the coal particle to constitute light gases and tar. The fragments with high molecular weight, and hence low vapor pressures, remain in the coal under typical devolatilization conditions until they reattach to the char lattice. These high molecular weight compounds plus the residual lattice are referred to as metaplast (2). During this period some coals swell to a degree, depending on its swelling index and the heating conditions (1). The metaplast further depolymerizes to yield char and volatiles.

The volatile yield and composition depends on the heating rate and final temperature. At slow-heating rates (<1°C/s) the volatile yield is low due to repolymerization. Then the total volatile yield will be equal to the volatile matter content determined from the ASTM Proximate Analysis, which is an analysis done at a slow heating rate. Under rapid-heating rate (500–10⁵°C/s) the volatile yield is 20–40% more than that at slow-heating rates (1). At any given temperature only a certain fraction of the volatiles is released. Significant devolatilization begins when the coal temperature is about 500°C. As the temperature is increased more volatiles are released. The maximum volatile yield occurs when the temperature is >900°C, the temperature at which the American Society for Testing and Materials (ASTM) Proximate Analysis for volatile matter is conducted.

The fraction of the devolatilization gas that condenses at room temperature and pressure is called tar. It is a mixture of hydrocarbons with an average molecular weight ranging from 200–500 g/mol (2). The yield of tar depends on the coal rank; higher rank coals produce lesser amounts of tar. Higher gasifier temperature also reduces the amount of tar in the gasifier products because of increased cracking of tar into lighter gases. The amount of tar also decreases with increasing pressure and decreasing heating rates.

The devolatilization gas that does not condense at room temperature and pressure consists mainly of CO, CO_2 , CH_4 , H_2 , and H_2O . The predominant source of CH_4 in the gasifier-product gas is the devolatilization process, and its production is favored by low temperature and high pressure. Therefore, the amount of methane in the product of moving bed gasifiers, which operates at a low temperature, is higher than that in typical fluidized bed and entrained bed gasifiers (3).

The solid product left over from devolatilization is char. During devolatilization the porosity changes from 2 to 20%, typical of coal, to >80%. The nitrogen surface area increases from $10-20 \text{ m}^2/\text{g}$ (coal) to $200-400 \text{ m}^2/\text{g}$ (1). The increased surface area increases the reactivity of char. The reactivity of char depends on properties of coal minerals, pyrolysis conditions, and gasification conditions. If the char porosity reaches a critical porosity (70–80%) the char will fragment into fine solids, which also increases the reactivity of char.

Char in an oxygen atmosphere undergoes combustion. In gasifiers partial combustion occurs in an oxygen-deficient, or reducing, atmosphere. Gasifiers use 30-50% of the oxygen theoretically required for complete combustion to carbon dioxide and water. Carbon monoxide and hydrogen are the principal products, and only a fraction of the carbon in the coal is oxidized completely to carbon dioxide. The combustion reaction is written in a general form as follows:

$$(1 + \lambda) C + O_2 \rightarrow 2\lambda CO + (1 - \lambda) CO_2 \qquad \Delta H^{\circ}_{298 K} = 172.5 \ \lambda - 393.5 \ kJ/mol$$

Reaction	Relative rate	
$\begin{array}{c} C + O_2 \\ C + H_2 O \\ C + C O_2 \\ C + H_2 \end{array}$	$10^{5} \\ 3 \\ 1 \\ 10^{-3}$	

Table 1. Relative Gasification Rates at 10 kPa and 800°C

where λ varies from 0 (pure CO₂ product) to 1 (pure CO product). The value of λ depends upon the gasification conditions and is usually close to 1. Under typical gasifier conditions this reaction rate is controlled by diffusion limitation. For a particle size of 90 µm the rate is limited by diffusion rate for temperatures >475°C. For smaller particles, diffusion rate becomes limiting only at a higher temperature (1325°C for 20 µm particles). The heat released by the partial combustion provides the bulk of the energy necessary to drive the endothermic gasification reactions.

The oxygen is rapidly consumed in the combustion zone, which occupies a small volume of the reactor. Further conversion of char occurs through the much slower, reversible gasification reactions with CO_2 , H_2O , and H_2 .

$$\begin{array}{ll} \mathrm{C} + \mathrm{CO}_2 \leftrightarrow 2 \, \mathrm{CO} & \Delta H^\circ_{298 \ \mathrm{K}} = 172.5 \ \mathrm{kJ/mol} \\ \mathrm{C} + \mathrm{H}_2\mathrm{O} \leftrightarrow \mathrm{CO} + \mathrm{H}_2 & \Delta H^\circ_{298 \ \mathrm{K}} = 131.3 \ \mathrm{kJ/mol} \\ \mathrm{C} + 2\mathrm{H}_2 \leftrightarrow \mathrm{CH}_4 & \Delta H^\circ_{298 \ \mathrm{K}} = -74.8 \ \mathrm{kJ/mol} \end{array}$$

The rate of gasification reaction depends upon the char properties and the gasification conditions. Table 1 (4) gives the typical orders of magnitude of various reactions: Another important chemical reaction in a gasifier is the water–gas shift reaction:

$$\mathrm{CO} + \mathrm{H}_2\mathrm{O} \leftrightarrow \mathrm{CO}_2 + \mathrm{H}_2 \qquad \Delta H^\circ_{298 \ \mathrm{K}} = -41.2 \ \mathrm{kJ/mol}$$

Mineral matter in the coal catalyzes this gas-phase reaction. Other gasphase reactions are the combustion of CO, H_2 , and CH_4 and tar cracking.

The physical and chemical changes in the sorbent material are depicted in Figure 2

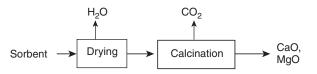


Fig. 2. Physical and chemical changes in solvent materal.

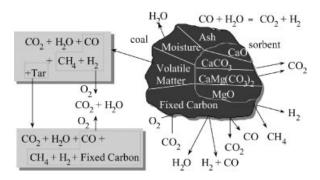


Fig. 3. Graphical representation of chemical reactions in coal, sorbent, and the gas phase during coal gasification (5).

When limestone is heated in the gasifier, initially the moisture is driven out. Further heating decomposes or calcines the limestone:

 $CaCO_3 \leftrightarrow CaO + CO_2$ $\Delta H^{\circ}_{298 \text{ K}} = 183 \text{ kJ/mol}$

The calcium oxide removes some of the sulfur gases as calcium sulfide.

Trace elements, such as sulfur and nitrogen, are also involved in the gasification reactions. Sulfur in coal is converted primarily to H_2S under the reducing conditions of gasification. Approximately 5–15% of the sulfur is converted to COS, whereas the coal nitrogen is converted primarily to N_2 ; trace amounts of NH₃ and HCN are also formed.

The heating, drying, devolatilization, combustion, and gasification of coal in the presence of a sorbent are represented graphically in Figure 3. The reactions portrayed in Figure 3 have been incorporated into computer simulations to describe the behavior of coal during various coal gasification processes (5).

High temperature favors endothermic reactions (increases the products on the right hand side). High temperature favors reactions in which there is a reduction in the number of moles, as in the methanation reaction (2 mol of hydrogen gives 1 mol of methane). Hydrogen and carbon monoxide production increases with decreasing oxygen in the feed, with decreasing pressure, and with increasing temperature. Hydrogen production increases and carbon monoxide production decreases with increased steam rate. Methane production increases with decreasing temperature and increasing pressure. The product gas of air blown gasifiers is diluted by nitrogen. Upon heating bituminous coals become sticky and swell, which can cause problems in fixed-bed gasifiers. Such coals are more easily handled in fluidized and entrained bed gasifiers.

3. Syngas Chemistry

Whereas near-term application of coal gasification is expected to be in the production of electricity through combined cycle power generation systems, longer term applications show considerable potential for producing chemicals from coal using syngas processing (7). Products include ammonia, methanol, synthetic natural gas, and conventional transportation fuels.

The cost and availability of oil and natural gas influence the competitiveness of coal gasification, but coal is expected to continue to play an ever-increasing role as a significant resource base for both energy and chemicals.

3.1. Ammonia. Ammonia is produced through the reaction of hydrogen and nitrogen. In a coal-to-ammonia facility, coal gasification produces the hydrogen and an air separation plant, which also provides oxygen for coal gasification, supplies the nitrogen. Because coal gasification produces a mixture of hydrogen and carbon monoxide, the CO is combined with steam in a water gas shift reactor to produce carbon dioxide and H_2 . Following CO_2 removal, the hydrogen stream is fed to an ammonia synthesis reactor where it reacts with molecular nitrogen to produce ammonia.

The water gas shift reaction is used to increase the amount of hydrogen in the gas. For shifting coal-derived gas, conventional iron-chromium catalysts can be used. Because coal gas has a significantly higher concentration of carbon monoxide than is found in gas streams in conventional refineries, the catalyst must be able to withstand high thermal loads. However, potential catalyst poisons such as phenol and other hydrocarbons are not a concern in entrained-bed gasifiers.

3.2. Methanol. Methanol is produced by stoichiometric reaction of CO and H_2 . The syngas produced by coal gasification contains insufficient hydrogen for complete conversion to methanol, and partial CO shifting is required to obtain the desired concentrations of H_2 , CO, and CO₂. These concentrations are expressed in terms of a stoichiometric number, $(H_2 - CO)/(H_2 + CO_2)$, which has a desired value of 2. In some cases CO₂ removal is required to achieve the stoichiometric number target. Both CO and H_2 are combined to form methanol in a catalytic methanol synthesis reactor.

The exothermic reaction

 $\mathrm{CO} + 2\mathrm{H}_2 \rightarrow \mathrm{CH}_3\mathrm{OH}$ $\Delta H_\mathrm{R} = -109 \mathrm{~kJ} (-26 \mathrm{~kcal})$

is enhanced by high pressures and low temperatures. Catalysts used in the reactor are based on copper, zinc, or chromium oxides (8), and reactors are designed to remove the exothermic heat of reaction effectively.

Mobil Oil Corporation has developed a process on a pilot scale that can successfully convert methanol into 96-octane gasoline. Although methanol can be used directly as a transportation fuel, conversion to gasoline would eliminate the need to modify engines and would also eliminate some of the problems encountered using gasoline—methanol blends (see Alcohol Fuels; Gasoline and other motor fuels)

3.3. Synthetic Natural Gas. Another potentially very large application of coal gasification is the production of SNG. The syngas produced from coal gasification is shifted to produce a H_2/CO ratio of approximately 3:1. The carbon dioxide produced during shifting is removed, and CO and H_2 react to produce methane (CH₄), or SNG, and water in a methanation reactor.

The following reactions can occur simultaneously within a methanation reactor:

$$\begin{array}{l} \mathrm{CO} + 3\mathrm{H}_2 \rightarrow \mathrm{CH}_4 + \mathrm{H}_2\mathrm{O} \\ \mathrm{CO}_2 + 4\,\mathrm{H}_2 \rightarrow \mathrm{CH}_4 + 2\,\mathrm{H}_2\mathrm{O} \\ \mathrm{CO} + \mathrm{H}_2\mathrm{O} \rightarrow \mathrm{CO}_2 + \mathrm{H}_2 \\ 2\,\mathrm{CO} \rightarrow \mathrm{CO}_2 + \mathrm{C} \end{array}$$

The heat released from the $CO-H_2$ reaction must be removed from the system to prevent excessive temperatures, catalyst deactivation by sintering, and carbon deposition. Several reactor configurations have been developed to achieve this (9).

The tube wall reactor (TWR) system features the use of catalyst-coated tubes. The Raney nickel catalyst is flame-sprayed onto the inside surface of the tubes, and the tubes are immersed in a liquid, such as Dowtherm, which conducts the heat away. Some quantity of recycle gas, in the ratio from 0 to 5, may also be used. In fluidized-bed catalyst systems, Raney or thorium nickel catalysts operate under moderate pressure, and heat is quickly removed from the system by the off-gas stream. In the liquid-phase, methanation system developed by Chem Systems, an inert liquid is pumped upward through the reactor, operating at 2–7 MPa (300–1000 psi) and 300–350°C, at a velocity sufficient to fluidize the catalyst and remove process heat. At the same time, the coal gas is passed up through the reactor, where methanation occurs in the presence of the catalyst. It has been found that catalyst attrition is substantially reduced over that in gas-fluidized beds because of the cushioning effect of the liquid. Processes have also been developed for hydrogasification that maximize direct conversion of coal to methane. A good example is the HYGAS process, which involves the direct hydrogenation of coal in the presence of hydrogen and steam, under pressure, in two fluidized-bed stages. Additional developments have been pursued with catalysts, such as Exxon's catalytic gasification process, but these processes have not been commercialized.

A coal-to-SNG facility can be built at a coal mine-mouth location, taking advantage of low cost coal. SNG can then be pipelined to local distribution companies and distributed through the existing infrastructure. This approach is used in the Great Plains Coal Gasification Project in Beulah, North Dakota, which employs Lurgi gasifiers followed by shift and methanation steps. SNG has the advantage that it can directly displace natural gas to serve residential, industrial, and utility customers reliably.

Another technology that is being pursued for fuel utilization of coal is mild gasification. Similar to pyrolysis, mild gasification is performed at atmospheric pressure at temperatures <600°C. By drying and heating under controlled conditions, the coal is partially devolatilized and converted to gases and a solid residue. The gases can be used as fuel and partially condensed to produce a liquid fuel similar to residual fuel oil. The solid product is similar to low moisture, high heating value coal. A demonstration project for Powder River Basin coal was tested by ENCOAL in Wyoming.

3.4. Conventional Transportation Fuels. Synthesis gas produced from coal gasification or from natural gas by partial oxidation or steam reforming

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can be converted into a variety of transportation fuels, such as gasoline, aviation turbine fuel (see AVIATION AND OTHER GAS TURBINE FUELS), and diesel fuel. The Fischer-Tropsch process that converts synthesis gas into largely aliphatic hydrocarbons over an iron or cobalt catalyst is widely used for this application. The process was operated successfully in Germany during World War II and is used commercially at the Sasol plants in South Africa.

More recently, Shell developed proprietary technology for converting syngas into liquid hydrocarbons (10). This technology is particularly well suited for producing high quality distillate fractions and is therefore referred to as the Shell Middle Distillate Synthesis (SMDS) process. This is a modernized version of the classical Fischer-Tropsch technique. In the first step, the synthesis gas components, hydrogen and carbon monoxide, react to form predominantly long-chain paraffins that extend well into the wax range.

Underlying the Fischer-Tropsch reaction is a chain-growth mechanism. The product distribution is in accordance with Schultz-Flory polymerization kinetics and can be characterized by the probability of chain growth; the higher the probability of chain growth, the heavier the waxy product. In the development of the SMDS process, proprietary catalysts have been developed with a high selectivity toward heavier products and, therefore, with a low yield of products in the gas and gasoline range. Much attention has been paid to the selection of a reactor for this very highly exothermic process. In principle, three different types of reactors can be used for the synthesis: a fixed-bed reactor, an ebulliating or fluidized-bed reactor, and a slurry reactor.

The use of a fluidized-bed reactor is possible only when the reactants are essentially in the gaseous phase. Fluidized-beds are not suitable for middle distillate synthesis, where a heavy wax is formed. For gasoline synthesis processes like the Mobil MTG process and the Synthol process, such reactors are especially suitable when frequent or continuous regeneration of the catalyst is required. Slurry reactors and ebulliating-bed reactors comprising a three-phase system with very fine catalyst are, in principle, suitable for middle distillate and wax synthesis, but have not been applied on a commercial scale.

For the Fischer-Tropsch reaction in the first stage (heavy paraffin synthesis, or HPS) of the SMDS process, a tubular fixed-bed reactor has been chosen for its inherent simplicity in design and operation and also for its proven technology in other processes, such as methanol synthesis. The catalyst is located in the tubes, which are cooled by boiling water around them, and considerable heat can thus be removed by boiling heat transfer. The good stability of the SMDS catalyst makes it possible to use a fixed-bed reactor. In the next step, heavy paraffin cracking (HPC), the long-chain waxy paraffins are cracked to desired size under mild hydrocracking conditions using a commercial Shell catalyst. In the final step, by selection of the corresponding cut points, the product stream is split into fractions of the required specification. The products manufactured in the SMDS process are predominantly paraffinic and free of impurities such as nitrogen and sulfur.

4. Coal Characteristics Affecting Gasification

Developers of coal gasification technology have studied the impact of key coal properties on different parts of the gasification process. These tests have

	Composition	n, wt%	
Constituent	High	Low	
ash	24.5^a	0.5^b	
	(up to 35)		
oxygen	16.3^{c}	0.1^b	
sulfur	5.2^b	0.3^d	
chlorine	0.41^e		
moisture	30.7^a		
Na ₂ O	3.1^d		
$K_2 \overline{O}$	3.3^{f}		
CaO	23.7^c	0.8^b	
$CaO + MgO + Fe_2O_3$		10.2^g	
Fe ₂ O ₃	27.8^h		
SiÕ ₂	58.9^i		
Al_2O_3	32.6^{j}		

Table 2. Feed Property Extremes as Tested in SCGP-1

^{*a*}Texas lignite.

^b Petroleum coke.

^c Buckskin, Powder River Basin, Wyo.

^d SUFCo.

^e Pyro No. 9.

^fPike County. ^gNewlands.

 h R&F.

^{*i*} El Cerrejon.

^jSkvline.

provided a good understanding of the influence of coal properties and have led to the development of process and equipment options. For example, a comprehensive demonstration program conducted on the Shell Coal Gasification Process (SCGP) at the demonstration plant near Houston (SCGP-1) included 18 different feeds including petroleum coke and a very broad range of coals (11). The property extremes are shown in Table 2.

4.1. Reactivity. Reactivity is used to describe the relative degree of ease with which a coal undergoes gasification reactions. The primary property affecting the ease of conversion is the coal rank, which in turn reflects its volatile matter content, oxygen content, level of maturity, extent of aromatic ring condensation, and porosity. The lower the rank the higher the volatile matter content and the more open the pore structure. Also, lower rank coals have more heteroatoms (oxygen, nitrogen, and sulfur) within the organic structure and the aromatic structures are poorly aligned. Such an amorphous and open structure contains more active sites making reaction with oxygen and steam easier. As the rank increases the carbon lattice becomes better aligned and the porosity reduces until, in anthracite coals the carbon structure becomes less reactive developing the flat basal structures found in graphite. Reactivity varies dramatically with rank with some low rank coals being several orders of magnitude more reactive than high rank coals (6).

Other factors that have impact on reactivity are maceral distribution and the content of some catalytic mineral components. Vitrinites is the most common

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The cause of the higher reactivity in low rank coals is the higher porosity, the larger number of active sites, and higher content and dispersion of catalytic metals such as calcium, potassium, and sodium. The organic matrix in low rank coals have carboxylic acid and other heteroatomic (O, S, and N) functional groups that have exchanged hydrogen ions with these cations producing ideal catalytic sites lowering the activation energy for gasification reactions. From Table 2, it can be seen that anthracite coal and petroleum coke that have the lowest reactivity also have low oxygen content [0.1% moisture free (MF)], whereas subbituminous coal from the Powder River basin, named Buckskin, has the highest reactivity (an oxygen content of 16.3% MF). It has been shown that the high degree of dispersion of catalytic minerals in low rank coals, like the Powder River basin coal, account for its high reactivity (12). Ion-exchangeable cations are attributed to this coal's superior performance in the Transport gasifier (13).

4.2. Moisture and Oxygen Content. The moisture present in the coal is primarily a diluent. Although steam is used for gasification, there are several sources of water for this steam and only a small amount of the total water available is actually converted in the steam-carbon reaction. Steam is introduced with air and oxygen to moderate the temperature in the combustion zone of the gasifier. Some entrained gasifiers introduce coal into the reactor with the aid of water as a coal–water slurry. In addition, all coals have moisture content. Lower rank coals, which have been exposed to geological maturation over shorter periods of time and at lower temperatures, have more moisture than higher rank coals. As a result, the feed rate of lower rank coals must be increased to obtain a gas product of equivalent heat value.

Similarly, the higher oxygen content of lower rank coals reduces the heating value of these coals relative to higher rank coals. To offset the effectively higher oxidation state of the low rank coals a higher coal feed rate is required to obtain similar product gas quality.

In addition, the integrity of the coal structure becomes weaker and the coal particle becomes more friable as the oxygen and moisture contents increase. Low rank coals can exhibit serious particle decrepitation, dusting, and even spontaneous combustion when exposed to drying conditions upon storage, crushing, and handling.

Fixed-bed gasifiers can accommodate coal with moisture contents as high as 35% as long as the ash content is <10%. Entrained and fluid beds generally prefer less moisture to aid in forming the coal water slurry and reducing the extent of solids losses via elutriation and entrainment into the product gas stream. Because of the large heat of vaporization of water, excessive moisture can affect the gasifier temperature and impede normal operations.

4.3. Caking Properties. When bituminous coals are heated to $300-350^{\circ}$ C the particles tend to swell and agglomerate producing a consolidated cake. The handling of this caking char and the heavy tars that accompany it has been critical to the development of gasification processes. The agglomerate

that forms in a fixed or fluid bed disrupts gas flow patterns and lowers thermal efficiency.

It is useful to understand the nature of this caking phenomenon. Compared to low rank coals bituminous coals have undergone a greater degree of metamorphism: the organic structure has fewer heteroatomic cross-links, the aromatics are more highly condensed and aligned, and the pore structure is less open and more microporous. Upon drying bituminous coal particles retain their strength rather than becoming weak and friable as in low rank coals. However, upon heating to the point where organic constituents begin to decompose, organic volatiles form and partially dissolve the more cross-linked features. The result is essentially a plastic melt. This plastic transformation of the bituminous coal structure is referred to as metaplast and results in a sticky, particle-agglomerating phase during the heating process. Once the volatile matter content of the coal falls below $\sim 20\%$ the caking tendencies are reduced.

In addition, to formation of cake, bituminous coals also produce high molecular weight tars. To handle the tars formed in coal process units, the downstream gas cleanup system must be engineered to avoid plugging and fouling of lines, heat exchangers, and filters.

4.4. Mineral Composition. The mineral content affects gasifier performance, especially for most slagging gasifiers, because minerals melt to form of slag and provide an insulating coverage on the wall of the gasifier, which reduces the heat transferred during the gasification reaction (14). Mineral content also influences the requirements of the slag tap and the slag handling system. A related parameter is the slagging efficiency, which is the percentage of mineral solids recovered as slag out of the bottom of the gasifier relative to the total mineral solids produced by the process. As shown in 2, the feeds that were gasified at SCGP-1 had mineral producing ash contents ranging from 0.5% MF for petroleum coke to 24.5% MF for Texas lignite.

Iron sulfide, or pyrite, has a high density and relatively low melting point and is often found in the clinkers produced by temperature excursions in fluidbed gasifiers. Fluxing agents, such as sodium and calcium, particularly when combined with the ubiquitous clays and pyrite reduce the ash melting temperatures and are often identified as the cause for the initiation of process clinkers. As a result, coal mineral composition is monitored as closely as the heating value in an attempt to avoid operational problems. These minerals are generally evaluated by analyzing the oxide residue or ash produced upon burning the coal.

Ash Melting Point/Slag Viscosity. For gasification technologies utilizing a slagging gasifier, slag flow behavior is an important parameter. To determine slag viscosity, the viscosity of coal ash is measured in a reducing atmosphere. Coals having a wide range of ash fusion temperatures were tested at fluid temperatures ranging from 1190°C for Illinois No. 5 coal to 1500°C and higher for several Appalachian coals, such as Skyline, Robinson Creek, and Pike County. Coal ash viscosity plots versus temperature are shown in Figure 4 for a variety of coals. Slag viscosity varies over several orders of magnitude for the different coals at representative gasifier temperatures. For example, Buckskin, which is a subbituminous coal from the Powder River Basin in Wyoming, has a much lower viscosity than an Appalachian coal such as Blacksville No. 2. For coals having

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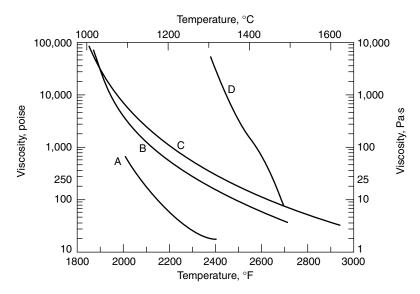


Fig. 4. Viscosity-temperature profiles of coal ash samples, where A represents Buckskin; B, pyro No. 9; C, Blacksville (Appalachia) No. 2; and D, Drayton.

high slag viscosities, slag behavior can be modified by the addition of a flux such as limestone (calcium carbonate).

Fouling Precursors. Fouling of heat transfer surfaces can result from constituents such as chlorine, sodium, potassium, and calcium. Most fouling indexes are based on experience with pulverized coal boilers. This information is often used to select conventionally high fouling coals to obtain fouling data on gasification units. In order to establish fouling indexes, the coal selection criteria in demonstration programs have included a wide range of fouling agents. As shown in 2, the calcium oxide content ranged from 0.8% for petroleum coke to 23.7% for Buckskin coal.

Corrosive Components. The primary coal properties affecting corrosion are sulfur and chlorine levels. Upon gasification these elements form acidic gases, hydrogen sulfide, and hydrogen chloride, which are responsible for corrosion of metals and other materials. The formation of these corrosive species is dependent on their content in the feed coal. The range of sulfur levels in Table 2 shows a low of 0.3% for SUFCo and a high of 5.2% for petroleum coke. Coals in the U.S. interior basin (including Illinois, Indiana, and Ohio typically have the highest sulfur levels, although the Appalachian region coals have high levels as well. R&F coal has 4.2% sulfur. Chlorine is less abundant and not uniformly distributed through the geologic basins. The highest chlorine level in this group of coals is 0.41% for Pyro No. 9 coal.

5. History

5.1. Early Coal Use and Gasification. Coal has been used for centuries (see Table 3). The very first recorded use of coal was in China between

Year AD	Event
589	First recorded use of coal in China
852	Coal first mentioned in the "Saxon Chronicle" of the Abbey of Petersborough
1180	Coal systematically mined in England
1250	Coal recognized as a commercial commodity
1316	Royal Proclamation forbidding use of coal in London due to its "noisome smell"
1609	Van Helmont identifies gas production from coal combustion.
1659	Shirley investigated "natural gas" released from a well in Lancashire England
1675	Coal was distilled for the production of tar
1780	Fontana proposes making "blue water gas" by passing steam over incandescent carbon.
1792	Murdoch lights his Scotch home with gas from coal heated in an iron retort
1803	Huge gas powered lamp installed on 40 ft. high tower on Main Street, Richmond Virginia
1812	London Streets illuminated by the London and Westminster Gas Light and Coke Co.
1859	Drake drills first oil well near Titusville, Pennsylvania
1872	Lowe invents carbureted gasifier
1880	Development of modern day fixed bed coke ovens
1920	Fischer and Tropsch develop catalysts to convert coal synthesis gas to liquids.
1926	Rheinbraun develops fluid bed gasifier
1936	Development of the modern day entrained bed Koppers-Totzek gasifier
1950	Production of gasoline and diesel fuel using Lurgi gasifier in South Africa
1970	Clean Air Act
1973	Arab oil embargo
1983	Syngas production for chemical production using Texaco gasifier at Tennessee Eastman
1984	Production of synthetic natural gas at Dakota Gasification Plant using Lurgi gasifiers
1996	Clean coal demonstration power production plants using Texaco and E-Gas gasifiers
2001	Power shortages in California

Table 3. Significant Events Related to Coal

220-589 AD. As noted in the Saxon Chronicle for the Abbey of Petersborough, coal was being used in Europe as early as 852 AD. However, the association of coal with gas production was not made until 1609. It was in Jean Baptist van Helmont's alchemy laboratory that coal was heated and the release of gas was first noted. He wrote how the coal "did belch forth a wild spirit or breath... not susceptible of being confined in vessels, nor capable of being reduced to a visible body". Helmont was likely frightened by this spirit and quite naturally called this spirit "gas" as derived from the Dutch word "Geest" for ghost (15).

To understand the development of coal conversion technologies, such as coal gasification, one must understand the factors that drove its development. Coal was the energy source that fueled the Industrial Revolution. Coal has a higher heating value than wood making it less bulky and easier to store and transport to the marketplace. It was found in concentrated seams underground and as such was systematically mined in England as early as 1180. These facts coupled with its widespread abundance in Europe made coal the preferred fuel in cities and towns.

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In 1272, coal was first used in London; however, a Royal Proclamation in 1316 outlawed its use because of the "noisome smell". With the population growth around key cities the uncontrolled use of coal for home heating and the workshops created the first recorded air pollution. Again, in 1580 Queen Elizabeth forbade the use of coal in London when Parliament was in session because "the health of the knights of the shires might suffer during the abode in the Metropolis." The use of coal continued unabated so that in 1662 King Charles II raised a huge sum of 200,000 lb from a "health tax" imposed on fireplaces (15).

As the world emerged from the Middle Ages coal provided more than just the fuel for innovation. Coal gas also played a key role in innovations in developments in materials and lighting. Development of industry required better materials for tools and machines. The first record of a Blast Furnace for the reduction of iron ore to iron was in the England in 1496. It was 1709 when Abraham Darby introduced coke to the blast furnace in place of charcoal, allowing the furnace size to be increased making the production of large amounts of cheap iron possible. Prior to this time charcoal would collapse under the extra weight of the charge of iron ore in a large furnace (see Fig. 5).

5.2. Coke Manufacturing Processes. These processes were developed to convert the softer bituminous coals into a strong hard coke ideally suited for iron making. It was found that when bituminous coals are heated slowly in the absence of air and then cooled the solid coke is both hard and inherently strong. The *coking process* requires heating to temperatures approaching 1000°C. This process heat is provided either by burning a portion of the coal directly and allowing the hot products of combustion to pass through a bed of coal, or by indirectly heating the exterior walls of a vessel containing the coal. The indirect method of heating a coke oven is still the most commonly used coking process used today because it maximizes the formation and quality of the coke product. In both heating methods, a flammable coal gas is produced that were readily put to use in a variety of applications.

The first coal *gasifier* was used by Fontana in 1780 when he directed a flow of water (steam) over red-hot, or incandescent, coal that was previously heated in an air blast. This process can be simply described using two chemical reactions: char combustion for producing heat and steam gasification for producing synthesis gas, a combustible mixture of carbon monoxide and hydrogen.

Fontana called the resulting coal gas "blue water gas", because it produced a pale blue flame when burned in air (4).

5.3. Manufacture of "Blue Water Gas" and "Town Gas". At the time of the French and American Revolutions houses were lit using candles and whale oil and heated with wood fireplaces or coal burners. Textile factories and mills were poorly lit and cold places to work. Thomas Edison did not discover the light bulb until late in the nineteenth century, 1879. The first electric power plant was not built until 1882. However, as early as 1792 a Scotch engineer named Murdock, used Fontana's process to produce a fuel gas and light his house. Later, James Watt, the inventor of the steam engine, employed Murdock to light one of his foundries with the newly discovered coal gas (16).

The first recorded use of coal gas in America was to light a street lamp mounted on a 40-ft tower on Main Street in Richmond, Virginia in 1803. The

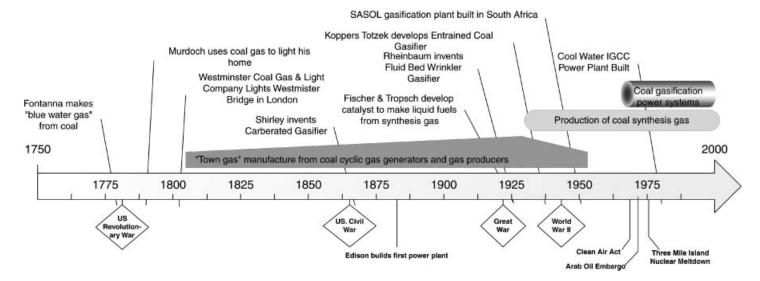


Fig. 5. Timeline for coal gasification development.

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Westminster and London Gas Light and Coke Co. was given a royal grant to provide lighting for London streets in 1812 becoming the first company to produce coal gas commercially. In spite of the hostilities in the War of 1812, the British influence in America was evident from the fact that just five years later the Gas Light Company of Baltimore became America's first gas company. In all of these cases the fuel gas was manufactured from coal gasification. However, it was not until 1865 that the use of coal gas for lighting became common practice.

During the nineteenth century, gas manufactured from coal was the only source of fuel gas. Although Thomas Shirley first described a discovery of natural gas from a well in Lancashire in 1659, but this resource was not commercialized until the early 1900s. Coal gasification was used for lighting and for heating in industrial processes. Later coal gas served as a rich source of chemicals and liquid fuels as well.

Shortly after the American Civil War, by 1875, every large and medium sized city in industrialized Europe and America developed coal gasworks and gas distribution networks. Gasworks consisted of a large number of individual gasifiers operating in various stages of the process: some being loaded, some being heated under air blast, others producing gas under a steam blast, and still others being cleaned out or emptied. Loading and unloading was typically done by hand (see Fig. 6).

The "blue water gas" produced from *cyclic gas generators* of the type used by Fontana were not strictly suitable for street or household lighting. The first coal

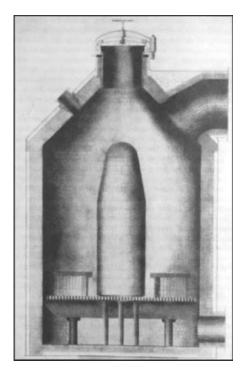


Fig. 6. Water-gas generator with central pier for improved heat distribution (17).

gasifiers built for this purpose undoubtedly suffered several setbacks. Although the heating value was high enough to sustain combustion, the blue flame was not sufficiently bright to illuminate a street or room in a house. Early developers learned that the coal gas needed to contain components called "illuminates" to provide a bright luminous yellow flame. These components consisted of higher hydrocarbons with hydrocarbon chain lengths of 2 or more. However, a second problem was observed if too many or the wrong types of higher hydrocarbons were added; condensable species, tars and naphthalene caused fouling and plugging in the piping and transfer lines. In addition, noxious and poisonous gases were also produced. These impurities included primarily hydrogen sulfide, hydrogen cyanide, and ammonia.

The solution to produce a brighter yellow flame was to simply add "illuminates" as the product gases left the gasifier. In the *carbureted gasifier process* developed in 1872, Professor Thaddeus Lowe produced both an "oil gas" and "blue water gas". The "oil gas" contained condensable coal tars or cheap petroleum distillate oil and was sprayed onto a hot brick matrix or checker work. After the "blue water gas" was formed it was passed over this checker work and mixed with the volatile products of "oil gas" decomposition. The high temperature of the brick and reducing nature of the product gas leaving the gasifier would cause cracking and vaporization of the coal tars and oils (15).

Solutions were readily developed to separate and clean the undesirable components from the coal gas. Cooling, direct water spray, and separation methods such as filtering of the gases were usually necessary to remove condensables and impurities. The carbureted gas was commonly called "town gas" because this gas was distributed through a network of pipelines for use in lighting streets and eventually homes in many of the late nineteenth century cities in America and Europe.

As is still true today, the coal gas in those early gasifiers was produced in several different reactors depending on the desired products and end use. Coal technologies besides combustion included: *pyrolysis, coking, cyclic gas generators*, and *gas producers*. All of these processes are heated with insufficient air to convert the coal to the final products of combustion. The products of all of these processes are a solid fuel, condensables and tars, and flammable gases. Both the quality and quantity of these products depends on how the coal is heated, the gas atmosphere used during the process, and the temperature. A comparison of the various products generated from these coal conversion processes is presented in Table 4.

Pyrolysis, or heating coal in the absence of oxygen, was conducted principally for the manufacture of chemicals. This conversion process is geared toward the production of condensable products such as benzene, toluene, naphthalene, phenols, creosote, and pitch. *Coking* is another process in which coal is heated in the absence of oxygen, but unlike pyrolysis the principal product of interest is the solid coke. In *pyrolysis*, the coal beds are thin and temperatures are low and near the melting, or the decomposition, temperature of coal to promote production of liquids and gases. While in *coking* the coal beds are deep and the temperatures are much higher to promote resolidification of the decomposing coal. In both of these processes, fuel gas is formed in significant quantities and of high quality as measured by the gas heating value in terms of British thermal units

Product composition			Gas p	roducers	Entrain		
	Blue water gas	Town gas	Fixed bed	Fluid bed	Fixed bed	Fluid bed	Coke oven
oxidant	steam	steam	air, steam	oxygen, steam	air, steam	oxygen, steam	none
fuel	bit. coal	bit. coal	bit. coal	brown coal	subbit. coal	bit. coal	bit. coal
exit temp, °C			142	482	1093	1093	982
Gases, %wt							
H_2	50.5	40.5	14.5	36.0	12.9	35.8	46.5
CŌ	38.5	34.0	25.0	44.4	23.5	50.7	6.3
CH_4	1.0	10.2	3.1	1.6	0.02	0.1	32.1
illuminates ^b		8.0		0	0	0	4.0
N_2	3.5	2.9	52.7	0.8	60.3	0.2	8.0
\tilde{CO}_2	6.0	3.0	4.7	15.7	3.1	13.1	2.2
char, wt%			19.5	20.0	11.8	11.4	75
gas HHV, Btu/scf	300	550	167	267	117	280	584

Table 4. Operating Conditions and Product Distribution for Various Coal Conversion Processes Using Bituminous Coal^a

 a From Refs. (15,18). b Noncondensable gases consisting of hydrocarbons with carbon chains between C2–C4.

per standard cubic foot of gas (Btu/scf). Where possible this gas was used for process heat, but any excess gas could be sold.

There were two types of coal gasifiers in the mid-1800s cyclic gas generators similar to those first used by Fontana and gas producers. The cyclic gas generators produced a high quality fuel gas. This high heat value gas was made possible by collecting only the products during the steam blast and venting the products of combustion during the air blast. As we noted above the carbureted gasifier produced "town gas" that was particularly well suited for lighting. The original cyclic gas generators produced "blue water gas" that was an ideal industrial fuel used in kilns, boilers, brick ovens, and for curing materials, particularly where particulate impurities were to be avoided. However, it was readily seen that the production rate could be improved by making this cyclic process continuous.

Gas manufacturers in the early 1800s learned that they could improve both the rate and efficiency of the gas making process by introducing steam and air together into the coal. In this way, the steam was available to gasify the carbon at the same time that heat was being produced from coal combustion. The result was the ability to substantially increase the overall conversion of coal to gas. Thus, modern *gas producers* were developed. Gasifiers were batch units that were loaded by hand, and the ash or coke, often still glowing hot, had to be removed by hand usually with nothing more than a shovel. As late as 1936 the Federal Trade commission reported that there were 3800 machine fed and ~1000 hand-fed *gas producers* in the United States alone (1).

The development of gas producers did not, however, displace the cyclic gas generators. By firing air and steam simultaneously the product gas in these producers now contained the unreactive nitrogen that is present in the air stream fed to the gasifier. Air is 79% N₂ and only 21% O₂ by volume. This added nitrogen is merely a diluent and plays no role in the combustion or gasification reactions. In addition, this diluent must be heated both during the gasification process and later as the fuel is burned. As a result the producer gas has only about one-half of the heating value compared to "town gas". There are several issues for using this low quality gas fuel. The large gas volume and lower heat value reduce the feasibility to economically transport producer gas for more than a short distance. Upon combustion the flame is longer, cooler, and more luminous. However, producer gas found many industrial applications to take advantage of the flame properties of this low Btu gas. For example, in glass making the relatively low temperatures and long flame are useful to avoid hot spots and nonuniformities in the glass.

By 1930, there were over 11,000 coal gasifiers operating in the United States and in the early 1930s over 11 million metric tons of coal were gasified annually. Fredersdorff and Elliott (19) described "the ideal complete gasification process as a single stage, continuous process which uses air as the oxidizing medium and converts any type of coal into a combustible or synthesis gas low in inerts". The development of the modern-day gasifier strives to meet this goal while achieving the most desirable mix of products depending on the end-use or process application. Three modern coal gas producer types are shown schematically in Figure 7, called Fixed Bed, Fluid Bed, and Entrained-Flow gasifiers.

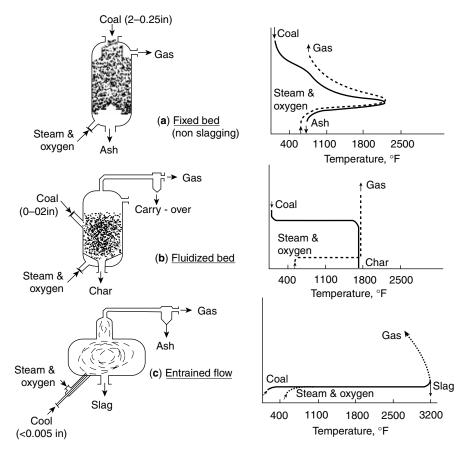


Fig. 7. Types of gasifiers: (**a**) moving-bed (dry ash), (**b**) fluidized bed, and (**c**) entrained-flow (20).

Each of these gasifiers has advantages, disadvantages, and potential for process improvement. Each type of gasifier converts certain coal feedstock over unique time-temperature profiles resulting in various product distributions. In Figure 5, a typical temperature profile is also portrayed for each reactor system, highlighting the process differences for these technologies. Typical product distributions for each are presented in Table 4 and compared to the early cyclic gas generators. The prime developments in the modern gasifiers were the variety of different coals that could be gasified, the coal throughput or capacity, and the overall coal conversion. The sacrifice made to achieve these improvements was the reduced quality of the fuel gas that was often lower, particularly when gasifying coal in air and steam. However, the composition of hydrogen and carbon monoxide produced in the newer generation of gasifier was sufficient for new applications whether they were liquid production, chemical synthesis, or power generation.

5.4. Gasification for Liquid and Chemical Feedstock. Although Drake discovered oil in Titusville, Pennsylvania in 1859, petroleum products

did not make inroads into the gas manufacturing markets until the 1930s. At that time oil and natural gas began to effectively displace manufactured gas from pipeline distribution networks. However, coal gasification remained a strong industry as a result of the emerging new transportation and chemical industries.

During the first quarter of the twentieth century coal was king, providing: heat for homes and industry, fuel gas for town lighting, process gas for industry, coke for the iron industry, and fuel for steam locomotives and shipping. By 1850, with the completion of the transcontinental railroad in the United States coalfired steam locomotives tied the nation together. By 1900, electric power was beginning to compete with the coal gas industry for public lighting. The electric power industry was also coal based, although large hydroelectric power plants from dams would be built between 1900 and 1930. With the development of the internal combustion engine and Ford's innovations in the automobile industry, it was natural for scientists to look toward coal for the liquid fuel that was needed.

During the 1930s new cheaper and cleaner fuels began to emerge—natural gas and oil industries were born at this time. However, this did not happen overnight. Even after the Second World War, 5 million tons of coal was consumed per year through the manufacture of carbureted gas.

The first liquid fuels developed from coal can be traced to Bergius in Germany in 1913. He used a liquid extraction process heating pulverized coal with oil under high pressures and with hydrogen. This is called direct hydrogenation or direct liquefaction of coal.

In 1920, two other German researchers, Fischer and Tropsch, developed a catalyst to convert the hydrogen and carbon monoxide from coal gasifier gas to hydrocarbon liquids. The Fischer-Tropsch process became known as indirect coal liquefaction since coal was first gasified before the gases were reduced to liquids.

By the mid-1930s Germany had plants to produce gasoline and oil from both direct and indirect liquefaction technologies, ~ 1 million and 0.5 million gal/year, respectively (21). Individual reactors in the early Fischer-Tropsch plants yielded ~ 5000 L of gasoline per day. Literally 100 such gasifiers were used in the first German plants (Fig. 8). But because the Bergius process plants were further developed and could be more readily scaled up to larger size, that was the technology chosen in 1939 when Germany expanded production to fuel the Nazi war machine. Direct hydrogenation reactors were eventually built to process up to 350 tons of coal per day yielding 250,000 L of gasoline. The capacity for oil and gasoline manufacture from direct liquefaction plants quadrupled during the war years in Germany. Elsewhere, however, oil resources were sufficient to meet demands and coal was not extensively used to produce liquids. After the war the technology used in the German synthetic fuels industry was studied, but considered uneconomical given the more competitive costs of natural gas and oil production.

A unique economic climate developed in South Africa, where the South African Coal, Oil, and Gas Company has operated the SASOL plant to produce liquid transportation fuels. SASOL is a coal gasification plant operating since 1955 using the Fischer-Tropsch process for the production of liquid fuels. South Africa was economically sanctioned for their social policies of Apartheid,

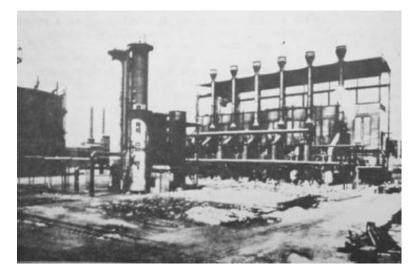


Fig. 8. Fischer-Tropsch synthesis for gas generators, Ruhrchemie plant (21).

and had no known or available oil resources, but ample coal supplies. The individual reactors in these plants are 100 times larger than the plants used in Germany during the Second World War. SASOL had a capacity of nearly 2.5 million gal of oil and gasoline per day in 1982 using Lurgi fixed-bed gasifiers.

6. Gasifier Types

6.1. Fixed-Bed Gasification. As described above, the early gasification processes were developed using a countercurrent, *fixed bed gasifier*. Within the bed the fuel is not actually fixed but in fact moves, by gravity flow, as the combusted ash is withdrawn from the gasifier. Typically, the air and steam are introduced at the bottom and travel upward through the coal bed. The coal is fed onto the top of the bed and travels downward countercurrent to the flow of gases. The gas outlet and coal feed inlets fix the upper level of the bed, while the bottom of the bed is most commonly fixed by the presence of a rotating grate.

The *fixed bed* was long considered to be the most efficient method of converting carbonaceous fuels to fuel gas. A schematic of the processes taking place within this *fixed bed gasifier* are represented in Figure 9 (22). The fuel bed is generally divided into different temperature zones corresponding to the following:

- 1. The topmost layer where coal is dried and preheated and volatile hydrocarbons are released.
- 2. The reduction, or gasification, zone where the hot char reacts with steam and carbon dioxide to produce hydrogen and carbon monoxide.
- 3. The oxidation zone where the residual carbon reacts with oxygen producing heat for gasification reactions.
- 4. The ash cooling and air preheat zone at the bottom of the gasifier.

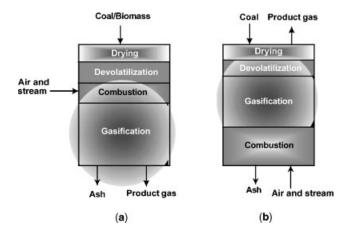


Fig. 9. Schematic for fixed- or moving-bed gas producer (22).

Fixed beds have several advantages. The flow of the hot gases up from the combustion zone preheats the coal leading to maximum heat economy. High carbon conversion is assured by plug flow of solids through the gasification and combustion zones and the relatively long residence times of the fuel in the vessel. The product gas exits relatively cool temperatures and without contamination of solids.

Fixed-Bed gasifiers have been developed to handle a variety of solid fuels. The factors important in affecting a fuels performance in a fixed-bed gasifier are particle size and size distribution, tendency for coal to melt and form an agglomerated mass, the temperature at which the ash melts and fuses, and the reactivity of the coal. The most suitable coals are uniformly sized crushed particles without tendency to agglomerate and with a minimum of fines but having reasonable mechanical strength. With such fuels the flow of gases through the bed is distributed uniformly through the bed resulting in uniform temperature distribution and stable reaction zones. Reactivity of the coal or coke feedstock affects the size of the reaction zones. Less reactive coals need larger bed depths, higher reaction temperatures, and longer residence times to achieve complete carbon conversion.

The disadvantage of the *fixed-bed gasifier* is the inability to process caking coals. These coals, generally of bituminous coal rank have a tendency to swell and agglomerate upon heating. As a result of this behavior such coals cause mal-distribution of both gas and solids flows leading to process failure. In order to use these coals a pretreatment by either preoxidizing, or more commonly coking, to eliminate any caking tendencies in the coal. The production of tars in the heating zone can also lead to fouling of the gas pipelines when the gas is used as a chemical feedstock, or synthesis gas, or when distributing this gas to public utilities. Coals having >35% moisture content lose strength upon heating and are not suitable feedstocks for fixed-bed gasifiers.

In fixed-bed gasifiers the devolatilization products exit the gasifier with the syngas, because of low temperatures and lack of oxygen in the devolatilization zone. This causes increased amount of methane in the product gas, which



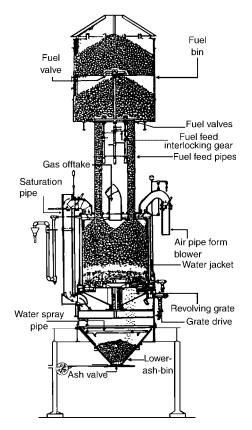


Fig. 10. Wellman-Galusha fixed-bed gasifier showing gravity coal feed system (18).

increases the heating value of the gas. But the low temperature and the countercurrent operation also allows the tar to escape, which is detrimental to the down stream equipment.

The Wellman-Galusha is a good example of an atmospheric *fixed-bed gasifier* with automated coal feed and a rotating grate for ash removal. The development of a coal feed system without mechanical parts provided a reliable maintenance free system as long as the coal feed size could be adequately controlled (Fig. 10).

The more critical performance limit for coal gasifiers, however, was the size, capacity, and coal throughput. The Lurgi gasifier was first commercialized in 1936. By 1950 the Lurgi brought significant technological advancements to fixed-bed gasification increasing the capacity and throughput. The primary improvements were to increase the operating pressure, temperature, and use of oxygen rather than air (Fig. 11).

Higher pressure increases the gasification rates per unit volume, decreases the heat losses, and reduces the number of reactor vessels to achieve a desired gas production rate. Higher reaction rates increase a reactor's capacity and throughput. Faster reactions also result in a shorter combustion zone. A shorter

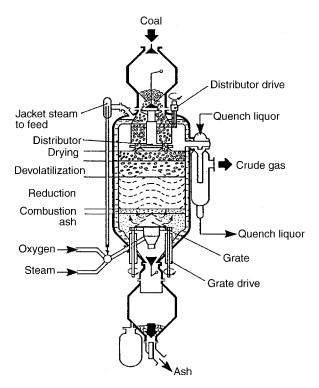


Fig. 11. The pressurized, oxygen blown, Lurgi gasifier showing with superimposed reaction zones (20).

high temperature combustion zone reduces heat losses and thus process inefficiencies.

The use of oxygen from an air separation facility also produces higher temperature in the combustion zone that, in turn, also increases the combustion and gasification rates. Using air the maximum combustion temperatures reach 1200–1300°C, but using only oxygen the temperatures reach 1500–1800°C. The use of oxygen in the gasifier also produces a higher quality gas fuel through the elimination of the nitrogen diluent in the produced gas.

The main technological hurdle to overcome with higher operating temperatures was the fact that the mineral impurities in coal, mainly sand and clays, melt and become sticky at these higher temperatures. This molten ash is known as slag. In a slagging gasifier this molten ash is kept fluid and drained (Fig. 12). Lurgi offered the first pressurized, oxygen-blown, fixed bed, slagging gasifier in the British Gas Lurgi gasifier in 1976. The slagging gasifier technology was developed at BGCs Westfield facility in Scotland, initially on a 275-tons/day pilot plant, and subsequently, a 550-tons/day demonstration unit.

A significant efficiency advantage is gained by reducing the steam requirement to only $\sim 15\%$ of that required by the dry-ash Lurgi gasifier. Compared with raw gas from the dry-ash Lurgi gasifier, the raw gas from the slagging gasifier has lower H₂O, CO₂, and CH₄ and higher CO content, primarily because of the

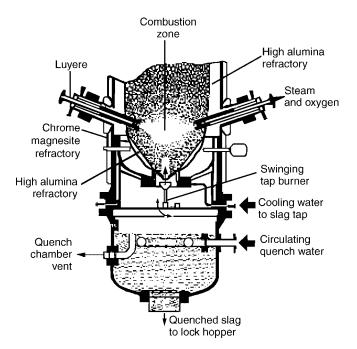


Fig. 12. The base of the British gas Lurgi slagging gasifier showing the hearth area and slag tapping (20).

lower steam consumption. Recycle of the tar and oil in the slagging gasifier increases the gas yield by reducing the net hydrocarbon liquid production to only naphtha and phenols.

6.2. Fluid-Bed Gasifiers. Another approach developed in the 1930s to overcome the size limitations and lack of fuel flexibility in the early fixed-bed gasifiers was the fluidized-bed gasifier. Rheinbraun developed the most commercially successful fluid bed gasifier in 1926, the Winkler gasifier. In this process, the reactor vessel was designed so that the air and steam flow required for gasification was sufficient to fluidize the bed of coal, char, and ash. Fluidization occurs when the gas flow velocity lifts the particles causing the gas–solid mixture to flow like a fluid. The coal feed to this gasifier is a finer crushed coal rather than the larger nuggets used in fixed beds (Fig. 13).

Fluidized-bed gasifiers provide better mixing and uniform temperatures that allow oxygen to react with the devolatilization products. These products also undergo thermal cracking, primarily on hot char surfaces, reacting with steam and H_2 . In dry fluidized-bed gasifiers, temperatures have to be maintained below the ash melting point, which leads to incomplete carbon conversion for unreactive coals. Agglomerating ash gasifiers operate at higher temperatures, near the ash softening point, which provides improved carbon conversion.

The primary advantage of the fluid-bed gasifier is the flexibility to use caking coals as well as low quality coals of high ash content. In addition, a fluid-bed gasifier is able to operate over a wide range of operating loads or outputs without significant drop in process efficiency. This fluid-bed process has a large inventory

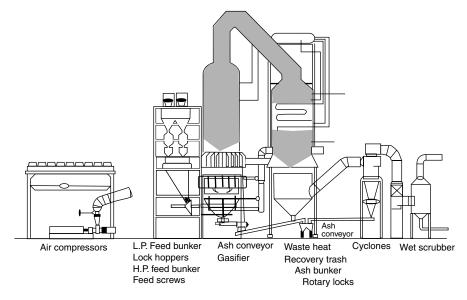


Fig. 13. Conceptual plant layout for the Winkler fluid bed gasifier process showing major auxilliary equipment (20).

of hot solids that stabilizes the temperature and eliminates the potential of oxygen breaking through and burning with the fuel gases in the event of an inadvertent loss of coal feed. Fluid beds also have high heat transfer rates and good solids and gas mixing. This minimizes the formation of localized hot spots that can produce molten ash agglomerates known as clinkers.

In addition, fluid-bed gasifiers can include inexpensive disposable sorbents, such as limestone, to absorb sulfur, reducing air emissions. The temperature regime is ideal for capture of hydrogen sulfide using limestone or dolomite.

One drawback, actually a result of good mixing in the fluid-bed gasifier, is the high temperature of the fuel gas at the exit of the reactor. When using cold gas clean up this high exit temperature represents a loss in process heat and thus process inefficiency. Likewise, solids drained from this type of wellmixed reactor also include a significant amount of carbon that must be utilized to avoid inefficiencies in carbon utilization. Solid particulates entrained in the product fuel gas are also high in carbon representing unreacted coal that must be recovered. Lower quality coals are more friable and result in higher loadings of such dust exacerbating the technical challenges.

In 1976, a vertical fluid bed gasifier was developed by Kellogg-Rust-Westinghouse (KRW) and the U.S. Department of Energy to overcome these drawbacks in the fluid bed gasifier. In the Kellogg Rust Westinghouse (KRW) gasifier, a central air or oxygen jet generates a hot zone at the base of the fluid bed to agglomerate ash. Unconverted char is recycled to this zone, which is hot enough to gasify the char and soften the ash. The ash particles stick together, growing in size and density, until they settle from the fluid bed and are separated from the char and removed from the bottom through dry pressure lock-hoppers. Agglomerated ash operation improves the ability of fluidized-bed processes to

gasify unreactive high rank coals and caking coals efficiently. In addition, this technology offers the potential to reduce the loss of carbon and the resulting process inefficiencies. This process permits the fluid bed to operate at higher temperatures (up to 1150° C), and thus increases gasification rates and coal throughput. The development of this process has culminated with the construction of a 300 MW Clean Coal Technology plant by Siearra Pacific Power Co. at Pinon Pine power plant near Reno, Nevada in 1998.

A Transport gasifier is being developed by KBR-Haliburton, Southern Co., and the U.S. Department of Energy in further attempts to decrease the capital costs of gasification by increasing the coal through-put (23). The transport reactor achieves more rigorous mixing than bubbling or jetting fluid beds through the aggressive recirculation of char and ash: 100 kg of char is recycled into the reactor for every kilogram of coal. The Transport reactor also exhibits the plug flow characteristics of entrained reactors without as high a gas exit temperature. The unique feature of the Transport gasifier is the large inventory of hot recycle solids stabilizing the reaction zone while moderating temperatures. The process is operated at elevated pressure of 10-15 atm. Powder River basin subbituminous coals are reported to react to 90% C conversion in <20 s at only 950°C (12). Analysis conducted confirmed that this type of coal could be gasified in the Transport reactor; however, less reactive coals were found to be only partially combusted and would produce hot spots if higher operating temperatures were attempted (12). Under air-blown operations S capture with the inherent Ca -containing minerals exceeds 95%; however, oxygen-blown attempts at the 100-kg/h facility have merely replaced nitrogen with steam and a shift in equilibrium results in only 20% S capture even with added limestone. Uncertainties that remain are the separation of solids from the product gas and the ability to convert a wider variety of coals.

6.3. Entrained Flow Gasifiers. Like fluidized beds, entrained coal gasifiers were developed to improve the gas production rate and operate with a wider range of fuel feedstocks. In an entrained gasifier the coal is introduced into air or oxygen in a dilute stream and heated to high temperatures, $1300-1475^{\circ}$ C, over only a very short period, 2-3 s. Entrained-bed slagging gasifiers provide uniform high temperatures, resulting in complete conversion of all coals to hydrogen, carbon monoxide, and carbon dioxide, and producing no tars, oils, or phenols. As a result the throughput and capacity of the entrained reactor is the highest of all gasifiers. Coal friability does not affect operations since the coal must be pulverized for the entrained flow gasifiers. Likewise, coal swelling and agglomeration do not influence gasification performance since the particles are separated from each other in the flowing gas stream. The product stream contains no tars and very little methane because the heavy volatiles are rapidly released and cracked at the high temperature and within the short time available in the reactor.

Koppers Totzek began developing the first entrained coal gasifier in 1938 (Fig. 14). The co-current flow of gases and solids results in a high gas exit temperature. This requires gas cooling and means loss of process heat or relatively low process efficiency.

The Texaco coal gasification process (TCGP) is the most widely employed commercial entrained flow gasifier. The Texaco gasifier has been licensed several times for use in chemical manufacture. The first license was issued to Tennessee

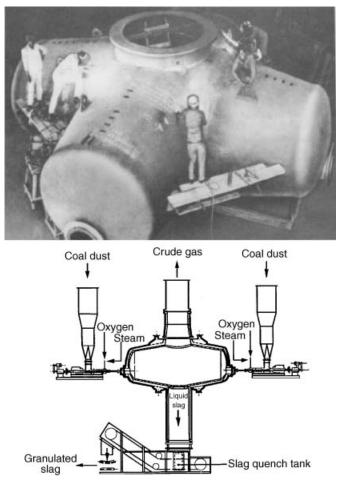


Fig. 14. Schematic Koppers-Trotzek entrained flow gasifier and photograph of gasifier with four burner heads (20).

Eastman for an 800 t/d plant, which was started up in 1983 and is used in the production of acetic anhydride. In 1984, a second Texaco coal gasification plant having a coal capacity of 1650 ton /day was built in Japan to produce ammonia. Since 1986 a third commercial plant, having a coal capacity of 800 t/d, was operated in Germany for use in oxo-chemicals manufacture.

Shell developed oxygen blown, pressurized, slagging, entrained coal gasification process. In 1993, the largest and most integrated coal gasification cycle plant in the world, a 253-MW power plant, was built in the Netherlands. The net efficiency at full load is ~43% based upon the lower heating value (LHV) of the coal. The level of sulfur removal is ~98% minimum (75 mg/m³), and a maximum NO_x emission from the gas turbine of 95 grams/gigajoule (g/GJ). By-products are slag, fly ash, sulfur, and salt. The coal consumption at full load is about 2000-ton/day (585 MW equiv) using dry ground coal. The gasification pressure is ~2.8 megapascals (MPa or 28 bar); reaction temperature, about $1500\,^\circ\text{C};$ steam pressures of 12.5, 4, and 0.8 MPa (125, 40, and 8 bar); and sulfur production, ${\sim}5000$ tons/year.

7. Gasifier Performance

Entrained-flow gasifiers can process a wide variety of coals, and data for several SCGP-1 coals are shown. In Table 5 compositional analyses; Table 6 shows ash minerals; and Table 7 presents a summary of gasification parameters for the different feeds. These feeds range from high rank bituminous coals from Appalachia, eg, Pike County coal, to low rank Texas lignite and petroleum coke (24–31).

Operating Parameters. The primary gasifier operating parameters are coal composition, coal throughput, oxygen/coal ratio and steam/oxygen ratio. The amount of oxygen and steam fed to the gasifier depends on the coal composition. In general, low rank coals are very reactive and require less oxygen and little to no steam, whereas high rank coals are relatively unreactive, requiring more oxygen and a moderate amount of steam. Steam provides an alternative source of oxygen for the gasification reaction and helps to moderate the gasification temperature. As a source of hydrogen, steam also helps to balance the H_2/CO ratio, giving a constant syngas composition for all coals including petroleum coke. Gasifier performance is evaluated in terms of syngas production and composition, carbon conversion, and cold gas efficiency (see Table 7).

Cold Gas Efficiency (CGE). Cold gas efficiency, a key measure of the efficiency of coal gasification, represents the chemical energy in the syngas relative to the chemical energy in the incoming coal. Cold gas efficiency on a sweet gas basis is calculated as the percentage of the heating value in coal that is converted to clean product syngas after removal of H_2S and COS.

Carbon Conversion. Carbon conversion on a once-through basis is a function of the coal composition and is strongly influenced by the oxygen/coal ratio. For some coals, the level of steam in the blast also affects the conversion pattern. Another factor is fly ash recycle, which raises the carbon conversion by recycling the unconverted carbon, most of which resides on the fly ash. This results in an overall carbon conversion greater than 99%.

Gas Composition and Heating Value. In Table 7 the syngas composition for a number of feedstocks is presented. These numbers reflect the composition of the gasifier off-gas on a dry basis. The primary gas components are CO and H₂, ranging from 59 to 67% and from 25 to 31%, respectively. Generally the gas composition is constant within a fairly narrow band for all coals including petroleum coke. The moderate variation is primarily because of variation in the CO₂ concentration caused by different steam/oxygen levels in the blast and oxygen/moisture and ash-free (MAF) coal ratios. The HHV of the product syngas after removal of H₂S, COS, and CO₂ is typically 12 MJ/m³ (300 Btu/ft³) and does not change significantly with changes in feedstock or gasifier conditions. The product syngas, also called medium-Btu gas (MBG), makes an excellent fuel for commercial gas turbines.

Heat Balance. Mass and heat balances are calculated around the gasification block, which includes the gasifier, quench, syngas cooler, and solids removal systems. A typical heat balance for Pike County coal is shown in

Component	Texas lignite	Pike County b	${\bf Pike \ County}^c$	Dotiki	Newlands	El Cerrejon	Skyline	Robinson Creek	R
					Samples as r	eceived			
moisture	30.70	8.55	6.04	6.71	7.01	11.86	7.95	5.61	5.7
ash	16.96	6.87	10.71	8.36	14.14	7.75	8.80	7.23	13.
volatile	30.19	32.13	30.80	34.61	25.30	33.30	33.36	32.63	35.
fixed carbon	22.15	52.45	52.45	50.32	53.55	47.09	49.89	54.53	46.
total	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100
					Dry basis san	nples			
ash	24.48	7.51	11.40	8.96	15.20	8.79	9.56	7.66	13.
carbon	56.22	79.40	75.24	74.47	71.49	74.71	74.44	78.31	69.
hydrogen	4.36	5.18	4.67	5.23	4.28	4.99	4.83	5.09	4.5
nitrogen	1.13	1.59	1.49	1.58	1.61	1.53	1.51	1.43	1.4
chlorine	0.08	0.20	0.12	0.23	0.09	0.02	0.02	0.16	0.1
sulfur	1.67	0.67	0.79	3.10	0.63	0.98	1.11	1.15	4.1
oxygen	12.06	5.46	6.29	6.44	6.70	8.99	8.52	6.20	6.2
total	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100
					Other proper	ties			
$\mathrm{HHV}^d \mathrm{kJ/kg}^e$	22839	32586	30749	31439	29225	30918	30523	32567	293
Hardgrove grindability	63	47	49	55	57	53	36	43	60

Table 5. Analyses of SCGP-1 Feedstocks^a

^a Composition is given in units of wt% unless other units are indicated.
^b Washed samples.
^c Run-of-mine samples.
^d HHV = higher heating value, on dry basis.
^e To convert kJ to kcal, divide by 4.184.

Component	Texas lignite	Pike County ^b	Pike County ^{cd}	Dotiki	Newlands	El Cerrejon	Skyline	Robinson Creek	R&D
					Ash n	nineral			
P_2O_5	0.16	0.06	0.31	0.25	1.36	0.20	0.30	0.28	0.3
SiO_2	48.82	52.30	53.22	51.10	50.89	58.94	51.20	50.92	471
Fe_2O_3	7.26	5.89.	728	13.31.	7.59	9.04	9.09	11.05	277
$A1_2O_3$	15.12	31.00	28.70	21.50	31.62	17.45	32.61	29.06	190
TiO_2	1.05	1.33	1.49	1.30	1.61	0.78	1.59	1.38	0.8
CaO	11.67	4.30	1.23	4.46	218	4.16	1.19	1.63	1.1
MgO	2.05	1.04	1.10	0.69	0.43	2.45	0.70	1.04	0.6
SO_3	10.89	1.18	0.75	4.05	0.94	3.97	0.94	1.05	0.9
K_2O	1.06	2.24	3.32	2.34	0.51	1.83	1.85	2.74	1.6
Na ₂ O	0.42	0.27	0.36	0.44	0.12	0.53	0.21	0.52	0.1
Total	99.49	99.61	97.86	99.44	97.23	99.36	99.66	99.67	99.8
					Ash content				
ash (MF), mean%	24.48	7.50	hAG	8.96	15.20	8.79	9.56	7.66	13.8
standard deviation	3.31	0.72	1.09	0.42	0.99	1.18	2.97	2.17	1.9
standard deviation, %	13.52	9.60	9.56	4.69	6.51	13.42	31.07	28.33	144

Table 6. Ash Minerals Variability of SCGP-1 Feedstocks^a

^a Composition is given in units of wt%.
^b Washed sample.
^c Run-of-mine sample.
^d Sample also contains 71.77 wt% V₂O₅ and 7.37 wt% NiO.

Parameter	Texas lignite ^{a}	${\rm Texas}\ {\rm lignite}^b$	Pike County ^c	$\operatorname{Pike}\operatorname{County}^d$	Dotiki	Newlands	El Cerrejon	Skyline
coal to plant, t/d	335	248	154	175	166	171	246	179
oxyge/MAF-coal ratio	0.877	0.865	1.006	0.974	0.970	0.986	0.922	0.955
burner steam/oxygen ratio			0.141	0.108	0.128	0.089	0.141	0.122
gasifier off-gas, vol % ^e								
Co	60.59	61.82	63.08	64.43	62.05	65.30	63.41	63.17
H_2	28.20	28.01	29.81	30.14	30.33	26.90	30.78	29.24
CO_2	5.38	4.47	2.59	0.68	2.47	2.26	1.68	1.99
$\mathrm{H_2S} + \mathrm{COS}$	0.71	0.80	0.24	0.36	0.90	0.28	0.27	0.34
$\overline{\mathrm{N}_2^{-}} + \mathrm{Ar} + \mathrm{CH}_4$	5.08	4.83	4.18	4.34	4.18	5.19	3.83	5.21
sweet syngas, ka/t	12062	10381	12213	13230	12275	12116	16707	13437
HHV ^f energy basis, GJ/h ^g	155.2	130.9	158.6	173.0	161.7	153.2	227.2	172.0
sulfur removal, % ^h	99.1	99.8	99.7	99.8	99.8	99.5	98.6	99.5
carbon conversion, %	99.7	99.4	99.9	99.1	99.9	99.7	99.6	99.9
cold gas efficienty, % HHV ^f (sweet gas basis)	78.8	80.3	80.9	83.0	80.1	80.3	83.4	82.4

Table 7. Summary of SCGP-1 Gasification Performance

^{*a*} High ash content. ^{*b*} Low ash content.

^cWashed samples.

^d Run-of-mine samples.

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^e Dry gas. ^f HHV = higher heating value. ^g To convert J to cal, divide by 4.184.

^h From syngas.

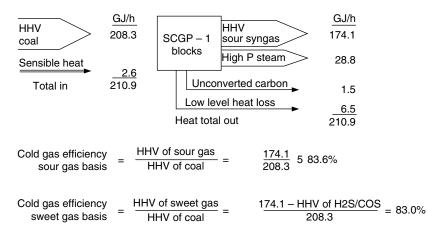


Fig. 15. SCGP-1 heat balance for run-of-mine Pike County coal. To convert GJ/h to Btu/h, multiply by 9.48'105.

Figure 15. Input streams are HHV of the incoming coal and sensible heat of the coal, oxygen and steam blast. The output streams are HHV of the sour syngas obtained from gas analysis, HHV of unconverted carbon obtained from analysis and weights of solids, heat recovered in the steam system in the gasifier and syngas cooler, and low level heat representing unrecovered sensible heat in the syngas. The low level heat is calculated by difference, thus forcing the heat balance to 100%, and it is typically 3-4% for all feedstocks. Thus in most cases at least 95% of the energy of the feed streams, mostly heating value of the coal, is converted to usable energy in the form of syngas and high pressure steam.

8. Gasification Systems

8.1. System Configurations. The flexibility of gasification technology allows it to be integrated into a variety of system configurations to produce electrical power, thermal energy, fuels, or chemicals (Fig. 16). The heart of the system is the gasifier. It converts a carbonaceous feedstock (such as coal) in the presence of steam and oxygen (or air) at high temperatures and moderate pressure, into synthesis gas, a mixture of carbon monoxide and hydrogen (with some carbon dioxide and methane). Minerals (ash) in the feedstock separate and leave the bottom of the gasifier either as an inert glass-like slag or other marketable solid product. A small fraction of the ash becomes entrained in the syngas and requires downstream particulate removal. Any sulfur in the feedstock gets converted to hydrogen sulfide and along with ammonia, hydrogen chloride or other contaminants, needs to be removed to meet pollutant emission limits or predetermined levels necessary for further downstream processing. The cleaned synthesis gas is then combusted in a high efficiency gas turbine/generator to produce both electrical power and supply compressed air to the air separation unit that generate oxygen for the gasifier. The hot combustion gas from the turbine is sent to a heat recovery steam generator (HRSG), which in turn, drives a

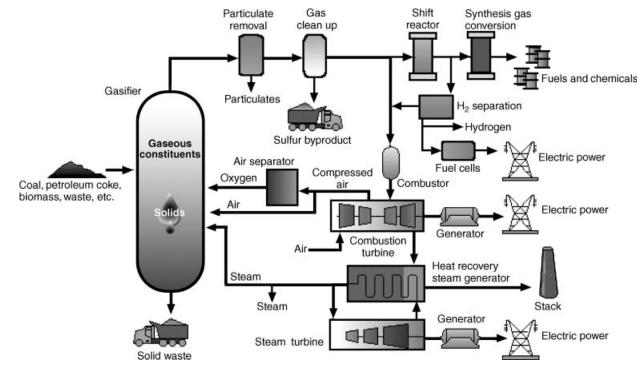


Fig. 16. Integrated gasification system configurations.

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steam turbine/generator to produce additional electrical power. These plants are referred to as IGCC. This combined use of combustion and steam turbines significantly boosts plant efficiencies over single cycles. In this mode of operation, approximately two-thirds of the electricity is produced in the combustion gas turbine/generator. A variation of this concept, cogeneration, can produce highgrade heat or steam for specific applications.

The other significant mode that this technology can be configured into is coproduction. This term refers to the coproduction of power, fuels, or chemicals. Products can be produced by either processing the feedstock prior to gasification to remove valuable components or by converting the feedstock into synthesis gas and later into products. Although not highlighted in Figure 14, feedstocks such as coal can be extracted to produce valuable precursors for the manufacture of high strength, lightweight carbon fibers and anode coke for the manufacturing industry. The rejected carbon-containing materials are then gasified and converted to power or other products. In the second option, raw feedstocks are directly gasified and cleaned to produce synthesis gas that can be routed through the combined cycle and/or be catalytically converted into fuels or chemicals.

8.2. Attributes of Gasification Technology. Gasification has many positive attributes that make it a desirable technology for the production of power, fuels, and/or chemicals. Some of those attributes that have helped to stimulate the current market and provide for a promising future are as follows:

Fuel Flexibility. In general, gasification has the ability to utilize all carbon-containing feedstocks. In addition to primary fuels such as coal, gasification can process hazardous wastes, municipal solid waste, sewage sludge, biomass, etc, after proper preparation to produce clean synthesis gas for further processing. Because of its ability to use low-cost feedstocks, gasification is the technology of choice for many industrial applications such as the gasification of petroleum coke in refineries. The ability to cofeed opportunity fuels (low cost) gives gasification good market adaptability.

Product Flexibility. Gasification is the only technology that offers both upstream (feedstock flexibility) and downstream (product flexibility) advantages. Integrated gasification combined cycle, and gasification processes in general, is the only advanced power generation technology capable of coproducing a wide variety of commodity and premium products (eg, methanol, higher alcohols, diesel fuel, naphtha, waxes, hydrogen...) in addition to electricity, to meet future market requirements. It is this ability to produce value-added products that has made gasification economical in selected situations and will be a key driver in a deregulated power market.

Cleanup. Because gasification operates at high pressure with a reducing atmosphere, the products from the gasifier are more amenable to cleaning to reduce ultimate emissions of sulfur and nitrogen oxides as well as other pollutants than those from combustion processes. In general, the volume of the fuel gas processed in an IGCC plant for contaminant removal is typically one-third that from a conventional combustion power plant. Processing lower volumes of gas translates to lower capital cost for pollution prevention. The removal of sulfur, nitrogen, and other contaminants from the reducing gas is also much easier than from combustion systems. This results in sulfur and nitrogen oxide emissions being more than an order of magnitude less than those of conventional

combustion processes. Gasification plants can also be configured to reach nearzero levels of emissions when required.

By-Product Utilization. Unlike that from combustion processes, the byproduct ash and slag from the gasification technologies have also been shown to be nonhazardous. The material can be readily used for landfill without added disposal cost or can be used in construction materials or further processed to produce value-added products, leading to a zero discharge plant. Sulfur can also be readily removed and converted into elemental sulfur or sulfuric acid as a saleable product.

Efficiency. Compared to combustion systems, gasification is the most efficient and environmentally friendly technology for producing low cost electricity from solid feedstocks, and IGCC can be made to approach the efficiency and environmental friendliness of natural gas combined cycle plants. Further increases in efficiency can be achieved through integration with fuel cells and other advanced technologies. These higher efficiencies translate to lower operating costs, resource conservation, and lower carbon dioxide emissions (a global warming issue). In addition, the gasification process can be readily adapted with advanced technologies for the concentration of CO_2 with minimized impact on cost and thermal efficiency. The ability of a technology to achieve higher efficiencies are major factors in technology selection for future energy plants.

System Flexibility. Gasification technology can be configured into a wide variety of systems to maximize efficiency, achieve fuel/product flexibility, or emphasize environmental performance. Although current cost for green field sites is high, gasification processes can be economically integrated into existing refineries and chemical plants. With proper integration and the use of existing infrastructure, the overall cost of a project can be significantly reduced. Through deployment in such environments, additional knowledge and experience will be gained, thereby reducing capital, operating, and maintenance costs for future facilities.

9. Environmental Performance

One advantage of modern IGCC systems is excellent environmental performance. Not only are regulatory standards met, but also emissions and effluents are well below acceptable levels (13,33,34). Regulations regarding pollutant emissions are becoming increasingly stringent. The need to reduce emissions of SO_2 , NO_X , particulate matter, and other pollutants is principally governed by the Clean Air Act Amendment (CAAA) of 1990, which is a very complex law to interpret and is site specific. Some of the more major sections of the CAAA are as follows:

1. Clean Air Act (Including 1990 Amendments)

Title I—Air Pollution Prevention and Control Part A—Air Quality and Emission Limitations Section 109—National Ambient Air Quality Standards In July 1997 EPA promulgated new standards for particulate matter finer than 2.5 μm (PM2.5) and revised the ambient ozone standards.

Section 111—Standards of Performance for New Stationary Sources

Part C—Prevention of Significant Deterioration of Air Quality

Section 169A&B—Visibility protection for Federal Class I Areas

(EPA has issued new requirements to improve the visibility in National Parks and other Class I areas primarily through the reduction of fine particles).

Title III—Hazardous Air Pollutants

Section 3.02—Listing of 189 Hazardous Air Pollutants (HAPs)

Several HAPs are released as particles (eg, arsenic, cadmium, chromium, lead) or as acid gases (eg hydrochloric acid) during the combustion of coal. Section 3.04—Promulgation of Maximum Achievable Control Technology standards

Section 3.11—Atmospheric Deposition to Great Lakes and Coastal Waters Section 3.12—Specific Studies

Hazardous Air Pollutant Report to Congress

Hydrofluoric Acid and Uses

Section 3.15—Prevention of Accidental Release

Includes reporting of sulfur trioxide releases

Title IV—Acid Deposition Control

Section 404—Phase I Sulfur Dioxide Requirements

Section 405—Phase II Sulfur Dioxide Requirements

Section 407-Nitrogen Oxides Emissions Reductions Program

Another significant environmental issue is global warming. World carbon emissions are expected to reach 8 billion metric tons by 2010. Emissions from developing countries were about 60% of those from the industrialized countries in 1990, but by 2010, will surpass them with respect to carbon emissions. The sharp increase is expected to result from economic expansion, accompanied by increasing power demand, and by continued heavy reliance on coal, especially in Asia. There is worldwide concern over greenhouse gas production and general agreement that reduction would be prudent. However, reduction goals and mechanisms for reduction implementation have not yet been established.

9.1. Acid Rain Emissions. Integrated gasification combined cycle represents a superior technology for controlling SO_2 and NO_x emissions. Emissions are much lower than those from traditional coal combustion technologies (34). During gasification, the sulfur in the coal is converted to reduced sulfur compounds, primarily H_2S and a small amount of carbonyl sulfide, COS. Because the sulfur is gasified to H_2S and COS in a high pressure concentrated stream, rather than fully combusted to SO_2 in a dilute-phase flue gas stream, the sulfur content of the coal gas can be reduced to an extremely low level using well-established acid gas treating technology. The sulfur is recovered from the gasification plant as salable, elemental sulfur. A small quantity of sulfur can also be captured in the slag as sulfates.

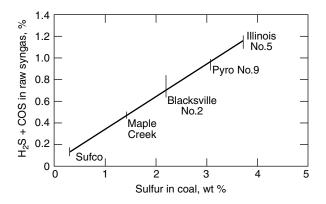


Fig. 17. Sulfur profile for SCGP-1 feedstocks. Overall sulfur removal is > 99.4% and the sulfur in the sweet syngas is < 20 ppm.

The gas treating and sulfur recovery processes employed in coal gasification have been broadly applied and operated for decades in refinery and petrochemical facilities and in natural gas sweetening plants. Operating experience from SCGP-1 (13,30) has confirmed that overall sulfur removal efficiencies of 99.4% from the raw syngas are achievable, independent of coal sulfur content for a variety of coals (Fig. 17). Modern pulverized coal (PC) plants generally have flue gas desulfurization (FGD) units capable of 95% sulfur capture. See Figure 18 for a comparison of emissions. New technologies are being developed for removing sulfur and other contaminants at high temperature. One hot-gas cleanup process uses metal oxide sorbents to remove $H_2S + COS$ from raw gas at high (>500°C) temperature and system pressure.

During coal gasification the nitrogen content of coal is converted to molecular nitrogen, N_2 , ammonia, NH_3 , and a small amount of hydrogen cyanide, HCN. In moving-bed gasifiers, some of the nitrogen also goes into tars and oils. The NH_3 and HCN can also be removed from the coal gas using conventional (cold) gas treating processes. Other techniques are being investigated in hot-gas

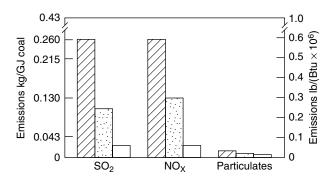


Fig. 18. Environmental emissions, where \square represents new source performance standards (NSPS) requirements; \square represents a pulverized coal (PC) plant; and \square represents SCGP-1.

cleanup technologies. After removal of HCN and NH_3 , combustion of the coal gas in the gas turbine produces no fuel-based NO_x . Only a small amount of thermal NO_x is formed and this can be controlled to low levels through turbine combustor design and, if necessary, steam or nitrogen addition. Based on tests using SCGPtype coal gas fired in a full-scale GE-frame 7F combustor (35), a NO_x concentration of no >10 ppm in the gas turbine flue gas is attainable. See Figure 16 for a comparison of NO_x emissions from a PC plant equipped with low NO_x burners.

9.2. Criteria Air Pollutants. Moving-bed gasifiers produce tars, oils, phenols, and heavy hydrocarbons, the concentrations in the gas product are controlled by quenching and water scrubbing. Fluidized-bed gasifiers produce significantly lower amounts of these compounds because of higher operating temperatures. Entrained-flow gasifiers operate at even higher temperatures, typically in excess of 1650°C. SCGP-1 experience has confirmed that carbon conversions of > 99.5% are easily attainable for any coal and that essentially no organic compounds heavier than methane are produced (35). Emissions of volatile organic compounds (VOC) from a IGCC plant are expected to be \sim 300 times lower than those from a similarly sized coal-fired steam plant equipped with low NO_x burners and an FGD unit.

The product gas after cleanup consists of primarily CO and H₂. Combustion of coal gas in high firing-temperature gas turbines converts virtually all of the CO to CO₂, and gas turbine exhaust is expected to contain no >10 ppm CO when operating at design conditions. Carbon monoxide emissions from an IGCC plant are thus expected to be around one-tenth those of a modern coal-fired plant equipped with low NO_x burners.

Particulate removal from the coal gas is effected either through a series of dry-solid and wet-solid removal steps or through the use of dry solids filters, so that the gas fed to the combustion turbine is essentially free of suspended particulates. The emissions of total suspended particulates (TSP) from an IGCC plant are about one-third those from a comparable pulverized coal plant equipped with a fabric filter and FGD unit.

9.3. Hazardous Air Pollutants. A number of the metals present in coal have the potential to be released as toxic air emissions. In moving- and fluidized-bed gasifiers, these metals are captured in the water. In entrained-bed gasifiers, a majority of these metals are captured in the slag. Because the coal ash in entrained gasifiers becomes vitrified at the high gasifier temperatures, the resultant glasslike slag encapsulates the metals in nonleachable form. In conventional treating systems using cold-gas cleanup, the small fraction of metals released to the gas phase is captured effectively in the gas cooling and gas treating steps. The combination of gas cooling and multistage gas—liquid contacting reduces very substantially the potential for airborne emissions of volatile metals such as lead, beryllium, mercury, or arsenic.

The total emissions of hazardous air pollutants from an IGCC plant having wet cleanup are expected to be at least an order of magnitude lower than those achievable from a modern coal-fired steam plant (37). Metals removal in hot-gas cleanup systems is still under development.

9.4. Water Consumption and Effluent Characterization. Another advantage of IGCC power generation is derived from lower water requirements. Because more than one half of the power generated in a IGCC plant comes from

the gas turbine, the water requirement is only 70-80% of that required for a coal-fired power plant, where all of the power is generated from steam turbines.

Whereas moving-bed gasifiers require complex water-treatment systems to address tars, phenols, and metals, this complexity is mostly alleviated for fluidized-bed gasifiers and is eliminated for entrained-flow gasifiers. The exiting water streams of SCGP-1 contain no detectable amounts of volatile or semivolatile organics. The effluent from an IGCC facility can be biotreated to meet National Pollutant Discharge Elimination System (NPDES) standards (38). Biological treatment provides oxidation for the small amounts of inorganic nitrogen and sulfur species that remain in the water. Effluent from SCGP-1 has pH 7.8 and contains (see also Table 8).

Both acute and chronic toxicity testing of the treated effluent on daphnia shrimp and fathead minnows have indicated that the effluent is completely suitable for discharge into receiving waters with no adverse impact (38).

9.5. Solid By-Products. Coal gasification power generation systems do not produce any scrubber sludge, a significant advantage over both direct coal combustion processes that use limestone-stack gas scrubbers and fluidized-bed

very Low Concentrations of Trace Metals		
Chemical analysis	ppmwt	
oil and grease	<1	
phenols	$<\!0.1$	
ammonia	${<}0.5$	
nitrite	${<}0.5$	
nitrate	50	
total cyanide	< 0.27	
thiocyanate	0.1	
formate	$<\!0.1$	
thiosulfate	$<\!0.1$	
sulfate	109	
sodium	470	
chloride	510	
pH	7.8	
Metal	ppmwt	
antimony	0.01	
arsenic	0.02	
beryllium	$<\!0.01$	
cadmium	$<\!0.03$	
chromium	$<\!0.01$	
copper	$<\!0.01$	
lead	$<\!0.03$	
mercury	$<\!0.001$	
nickel	$<\!0.02$	
selenium	0.35	
silver	0.03	
thallium	$<\!0.001$	
zinc	0.03	

Table 8. The Biotreated SCGP-1 EffluentContains Fully Oxidized Products andVery Low Concentrations of Trace Metals

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combustion processes that use solid absorbents for sulfur capture. In coal gasification, the sulfur in the coal is recovered as bright yellow elemental sulfur for which there are several commercial applications, the largest being in the phosphate fertilizer industry (see FERTILIZERS). Elemental sulfur is a commodity traded worldwide, with 1990s prices in excess of \$100/ton.

The ash in the coal is converted to slag, fly slag, or fly ash. Moving-bed and fluidized-bed gasifiers produce fly ash, which may be disposed of in a manner similar to that used for conventional power plant fly ash. In slagging gasifiers, the coal ash is mostly converted to a glasslike slag that has very low leach-ability. Environmental characterization of SCGP-1 slag and fly slag was performed for several coals using the extraction procedure (EP) toxicity tests and the toxicity characteristic leaching procedure test (TCLP), confirming that toxic trace metal concentrations in the leachate were well below Resource Conservation and Recovery Act (RCRA) requirements (39). Many of the elements, if present, were even below the detection limits. Additionally, the runoff from the slag storage area was collected and analyzed for comparison with the National Interim Primary Drinking Water Standards. The results of this comparison show that the measured values are typically much less than those allowed by the stringent national standard (39).

As part of a solids utilization program at SCGP-1, gasifier slag has been used as a principal component in concrete mixtures (Slagcrete) to make roads, pads, and storage bins. Other applications of gasifier slag and fly slag that are expected to be promising are in asphalt aggregate, Portland cement kiln feed, and lightweight aggregate (see CEMENT) (40). Compressive strength and dynamic creep tests have shown that both slag and fly slag have excellent construction properties.

9.6. CO₂ Emissions and Global Warming. The high coal-to-busbar efficiency of an IGCC system provides a significant advantage in responding to CO_2 emissions and thus to global warming concerns. High efficiency translates to lower coal consumption and lower CO_2 production per unit of electricity generated. The average existing PC unit has a heat rate of more than 10,550 kJ/kWh (10,000 Btu/kWh) on a higher heating value basis, which means that associated CO_2 emissions for a nominal 450-MW plant are well over 4 million tons/yr. The most efficient IGCC units offer heat rates of 8650 kJ/kWh (8200 Btu/kWh) and reduce CO_2 emissions by ~15-20% relative to the emissions from a PC unit. Current IGCC units such as the Buggenum, The Netherlands, IGCC plant has reported even higher efficiency than this with a heat rate of 7935 Btu/kWh or 43% efficiency.

10. Economics

10.1. Gasification in the Evolving World Energy Market. Today's energy picture is colored by several recent world events. Western economic development has proved to have an enormous appetite for energy with the United States being the largest energy producer and consumer. Requirements for transportation fuel has outstripped domestic oil supplies, but a coal gas/liquid industry does not exist to offset any increases in demand. As a result secure

international supplies of oil are considered critical to economic health. Thus, situations in oil-producing countries such as the Arab–Israeli wars in 1967 and 1973, the oil embargo in 1973, the Islamic revolution in Iran in 1979, the Iran–Iraq war in 1980, and the Iraqi attack of Kuwait 1990 have all resulted in keen interest from western countries and particularly responses from U.S. government. The continued development of a coal gasification industry, utilizing abundant U.S. domestic coal resources, could help to temper our responses to these threatening economic situations.

The power industry has also undergone dramatic changes over the past 50 years. The production of electric power has been dominated by coal-fired power plants. At the beginning of the twentieth century the power industry was initially developed as a public service through local, municipal, and regional power companies. By 1955 the power industry was well established. Power companies with approval of the Public Utilities Commission were able to build large power plants with scheduled costs and prices. The capacity and distribution of power was planned and orchestrated to achieve conservative margins of safety. The Clean Air and Water Legislation in the 1970s and 1980s have changed the way Electric Power is produced. Strong financial incentives and penalties were provided to develop cleaner power systems. Large public power utilities were slow to convert their existing plants to meet these new stricter restrictions in sulfur oxides, nitrogen oxides, and particulate emissions. There was public resistance to the huge costs associated with the cleanup systems required. As a result, new construction of power plants slowed in favor of retrofitting existing plants and excess capacity dropped to dangerously low levels. With deregulation of the power industry this situation continued unabated until the blackouts in California in the summer of 2001.

Nuclear power was expected to be the answer to clean and inexpensive electricity. However, scientist began to understand the long lasting hazards of radiation and costs for these power plants began to rise to meet safety regulations. Finally, a nuclear meltdown at the Three Mile Island power plant in 1979 and an explosion at Chernobyl in 1986 essentially halted further expansion of nuclear power plants. In addition, these incidents along with finding and cleanup of industrial waste sites raised the general public awareness of potential hazards and environmental concerns about pollution from all types of power plants.

To encourage development of more economic and cleaner power plants, the power industry was deregulated in the 1990s allowing independent power producers to compete with public utilities. Combined cycle plants, coal waste burners, and cogeneration facilities were now economically competitive and were built in the 1990s on a smaller scale to take advantage of special situations in various localities. However the changing nature of environmental legislation and the uncertainty that this produces have stifled the construction of new power plants. As a result, by the year 2000 the excess capacity of electric power generating facilities in the United States has fallen to <5%. The combined effects of deregulation and lack of power generating facilities and inadequate transmission lines led to California's electric power outages in the summer of 2001.

Coal power systems historically have raised steam and expanded it through a steam turbine to produce electricity. This process is limited in its efficiency because of the high heat requirement necessary to convert water from a liquid to steam. This heat of vaporization amounts to 540 cal/g of water. Nearly twothirds of the heat needed for the steam cycle is lost to raising steam and cannot be recovered as useful power. However gas turbines, which can use synthesis gas to generate power, do not have this inherent inefficiency.

Gas turbines, originally built for jet engines, have been improved to the efficient high temperature machines of today. In the gas turbine the release of chemical energy by the production of gases and thermal expansion of those gases under pressure is more completely recovered than from a steam turbines. Fuel cells, which can use synthesis gas or hydrogen, are also being developed and promise to provide the most efficient conversion of chemical energy to electricity. These technologies will provide the driving force for the next generation of highly efficient electric power plants. Coal gasification is uniquely posed to take advantage of these new technologies based upon the reliable abundant coal supplies.

Coal, the primary fuel for electricity generation in the United States and other countries, is expected to have an increasing role in the future. Conventional coal-fired electricity generation has resulted in numerous environmental problems, notably emissions of sulfur and nitrogen compounds, both of which have been linked to acid rain, and emissions of particulates (see AIR POLLUTION). Conventional coal-combustion technologies only partially solve these problems. Modern coal gasification combined cycle power generation technologies, also known as IGCC systems, present electric power producers with important options and opportunities to improve efficiency, environmental performance, and overall cost effectiveness.

10.2. Increasing Power Demands. Possibly the single largest driver for the demand of gasification technology is electrical power. In the United States, the Energy Information Administration (EIA) predicts that electricity demand in the commercial and industrial sectors will grow by 2.0 and 1.4%per year, respectively, between 1999 and 2020 (41). This translates to upward of 700 GW of new capacity, repowering, and replacement of existing power plants. It is likely that a large majority of this power (estimated at >85%) will come for natural gas-fired turbine generators due to their low capital cost and high efficiency. However, over half of all electricity in the United States is produced by coal-fired power plants (41). A great opportunity exists to retrofit and repower many of these existing plants with clean coal technology. It has been estimated that 40,000 MW of increased electricity production is possible over the next 3 years by retrofit and repowering of the existing coal fleet with state-of-the-art technologies (42).

Internationally, the market for power generation is somewhat different. By 2020, worldwide electricity demand is expected to be more than triple to 23 trillion kWh. The greatest gains are expected in developing Asia and in Central and South America. In Asia, electric power production cannot keep pace with economic growth and electricity shortages are common. Because these nations have large indigenous coal supplies, economical and environmentally acceptable coal-fired power plants have tremendous market potential with the initial commercial units being mostly conventional technology. As environmental requirements become more stringent and more capital becomes available to those markets, opportunities will be available for advanced coal-based technologies. In contrast to the vast coal resources in Asia, Central and South America

currently rely heavily on hydropower for electricity generation, and new capacity will likely be fueled by the large reserves of natural gas in the region.

There are also several niche industrial markets that require power in which gasification could play a predominant role. For example, the refining industry generates large quantities of high Btu content waste streams that could be gasified to generate power, steam, and synthesis gas. Refineries are heavy users of power and steam, are large users of hydrogen and are often located in close proximity to large chemical complexes (potential customers for both the power and the synthesis gas coproduct). One estimate places the worldwide refining market for gasification over the next 5 years as high as 4000 MW (\sim 2500 MW of power and the remainder to synthesis gas). The near-term market is in China and Japan where there is growth in new refining capacity and an expanding need for more residual upgrading. The need to replace base-load power generation capacity creates a domestic market for gasification refinery applications \sim 2010.

Another niche market example is in the pulp and paper industry. Much of the black liquor and biomass-fueled steam turbine cogeneration capacity in the U.S. pulp and paper industry will need to be retired and replaced over the next 20 years. Gasification could be employed to efficiently convert the biomass waste into steam and power and to recover chemicals. In 1994, 1.2×10^{18} J (or 38,000 MW) of black liquor were consumed by the U.S. pulp and paper industry. If biomass gasifiers were coupled with the Tomlinson boilers to cogenerate power and steam from this amount of black liquor, an additional 22 billion kw-h/year (or nearly one-half of the 51 billion kWh that the total industry purchased in 1994) could be generated. Gasification must compete against FBC in this market; however, gasification has the advantage of generating synthesis gas fuel for kiln operation in addition to being able to destroy hazardous chemicals and reduce sulfur emissions.

10.3. Deregulation. In the United States, deregulation is expected to completely restructure the U.S. electric utility industry. New capacity additions will likely favor low capital-cost power generation technologies. Competition will force utilities to increase utilization rates at existing plants, maximize efficiency, and minimize operating costs. Aging plants nearing retirement may be kept open if their operating costs are proven to be competitive, and plants with high fixed costs will be forced to close. Traditional "electric only" utility generators will see their market share dwindle as energy firms (firms that produce a variety of energy products such as steam, chemicals, fuels and synthesis gas) capture an increasing volume of electricity sales. Many niche industrial applications that integrate electric power generation with industrial processes to provide gains in productivity, environmental performance, and capital utilization will become attractive for gasification technologies.

Unless fuel price or availability at specific locations dictates otherwise, natural gas-fired turbine generators are the preferred low-cost technology. The capital cost for a natural gas-fired combined cycle plant is \$400–600/kW, about one-half the cost of an IGCC plant that gasifies coal. IGCC is capital intensive; it needs economies of scale to be an attractive investment option. However, IGCC costs can be improved by integrating in a synergistic way with various industrial applications. For example, gasification can operate on low cost opportunity feedstocks, it can be used to convert hazardous waste into useful products reducing or eliminating waste disposal costs, and it can coproduce power, steam, and high value products for use within the host plant or for export.

10.4. Fuel Reliability and Affordability. Numerous factors threaten the reliability and affordability of electricity throughout the United States. In the past year alone, rolling blackouts have become commonplace in California and skyrocketing electricity prices have shocked the state's \$1.3 trillion dollar economy—the sixth largest on earth (43). Other parts of the country are not immune from these effects. The demographics of population growth, demand increases, transmission bottlenecks, supply gaps, and an aging infrastructure are creating reliability problems in power generation markets across the country. The likelihood of power outages continues to increase and rolling blackouts like those experienced in California could occur elsewhere. Additionally, current environmental regulatory constraints and anticipated new, more stringent regulations pose potential hurdles and longer lead times for siting and permitting of new power plants.

Fuel diversity is the best way to insure reliable and affordable electricity. A diverse fuel mix helps keep electricity prices low by insulating companies and consumers from issues related to fuel variability, fuel price fluctuations, regional shortages, regional disruptions and changes in regulatory policies. Regional reserves, fuel transportation infrastructure and fuel cost often dictate the best fuel mix for a given region. These factors have contributed to substantial regional fuel diversity throughout the United States, which is recognized by power producers as an important aspect to reliable and affordable electricity throughout the United States (44). Technologies must be available to accommodate a diverse fuel mix and coal plays a dominant role in the fuel mix throughout the United States.

Figure 19 depicts projected fuel prices through 2020 (41). The cost for coal is projected to be very stable or slightly lower throughout that period, while natural gas and fuel oil are projected to increase in cost. To evaluate the potential impact of fuel price on technology selection, a recent study was sponsored by the Department of Energy. Figure 20 illustrates the effect of increasing natural gas costs on the cost of electricity for natural gas combined cycle and coal-fired power plants.

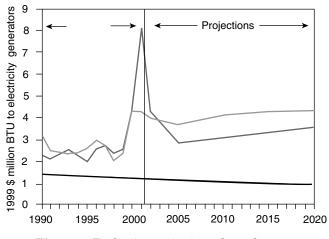


Fig. 19. Fuel price projections through 2020.

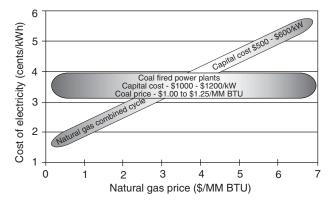
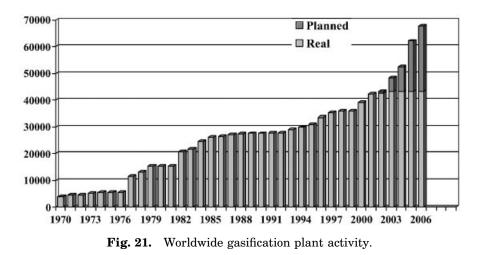


Fig. 20. Effect of natural gas price on cost of electricity.

10.5. Status of Gasification Technology. The worldwide activity in gasification technology has increased significantly over the past 30 years (Fig. 21). Nearly all of the gasification capacity through the mid-1970s can be attributed to the 19 Lurgi gasifiers operating at Sasol in South Africa. The relatively large increases in capacity in the latter part of the 1970s and the early 1980s represent the startup of 80 gasifiers associated with Sasol II and III, representing a combined increase of nearly 8300 mega Watt thermal (MWth) of synthesis gas capacity. A lesser, but notable, increase in capacity also occurred in the early 1980s with the commissioning of 14 Lurgi gasifiers at the Dakota Gasification plant in Buelah, North Dakota, adding another 1500 MWth of capacity. Following this, capacity remained relatively flat for over a decade. However, within a few short years due to deregulation and an increased need to more fully utilize the resources available, capacity increased by almost 50% and is expected to grow by nearly 60% in the next 5 years. Currently, there are >385 gasifiers in operation or under construction at 138 sites located in 22 nations in North and South America, Europe, Asia, Africa, and Australia. The largest 30 gasification plants in the world are listed in Table 9. This table also lists the principal product for each.



Plant owner	Country	Gasifier	Year	MWthOut	Fuel	Product
Sasol Chemical Ind. (Pty.) Ltd./Sasol Ltd.	South Africa	Lurgi	1977	5,090.0	bit.coal	FT liquids
Sasol Chemical Ind. (Pty.) Ltd./Sasol Ltd.	South Africa	Lurgi	1982	5,090.0	bit.coal	FT liquids
Dakota Gasification Co.	United States	Lurgi	1984	1,900.3	ligruite and ref. resid.	SNG and CO_2
SARLUX srl	Italy	Texaco	2001	1,216.7	visbreaker residue	$\begin{array}{c} ext{electricity, H}_2 ext{ and} \\ ext{steam} \end{array}$
Shell MDS (Malaysia) Sdn. Bhd.	Malaysia	Shell	1993	1,032.4	natural gas	mid-distillates
Mitteldeutsche Erdoöl-Raffinerie GmbH	Germany	Shell	1985	984.3	visbreaker residue	H ₂ , methanol and elec.
ISAB Energy	Italy	Texaco	2000	981.8	ROSE asphalt	electricity, H ₂ and steam
Sasol Chemical Ind. (Pty.) Ltd./Sasol Ltd.	South Africa	Lurgi	1955	970.6	bit. coal	FT liquids
Global Energy, Inc.	Germany	Lurgi	1964	848.3	municipal waste	electricity and methanol
Nippon Petroleum Refining Co.	Japan	Texaco	2003	792.9	vac.residue	eletricity
Millenium (Quantum)	United States	Texaco	1979	656.2	natural gas	methanol and CO
Hydro Agri Brunsbuüttel	Germany	Shell	1978	642.5	heavy vis. residue	ammonia
Shell Nederland Raffinaderij BV	Netherlands	Shell	1977	637.3	visbreaker residue	H ₂ and electricity
Sokolovska Uhelna, A.S.	Czech Republic	Lurgi	1996	636.4	liguite	electricity and steam
Global Energy, Inc.	United States	E-Gas	1995	590.6	petcoke	electricity

Table 9. Thirty Largest Gasification Plants Worldwide

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VEBA Chemie AG	Germany	Shell	1973	587.8	vac. residue	ammonia and methanol
Elcogas SA	Spain	PRENFLO	1997	587.8	coal and petcoke	electricity
Motiva Enterprises LLC	United States	Texaco	2001	519.5	fluid petcoke	electricity and steam
api Energia S.p.A.	Italy	Texaco	2001	496.2	visbreaker residue	electricity and steam
Chemopetrol a.s.	Czech Republic	Shell	1971	492.1	vac.residue	methanol and ammonia
Demkolec BV	Netherlands	Shell	1994	465.9	bit. coal	electricity
Ultrafertil S.A.	Brazil	Shell	1979	451.1	asphalt residue	ammonia
Tampa Electric Co.	United States	Texaco	1996	451.1	coal	electricity
Shanghai Pacific Chemical Corp.	China	GTI	1994	410.1	bit.coal	fuel gas and town gas
Gujarat National Fertilizer Co.	India	Texaco	1982	405.3	ref.residue	ammonia and methanol
Esso Singapore Pty. Ltd.	Singapore	Texaco	2001	363.6	residual oil	electricity, H ₂ and steam
ExxonMobil	United States	Texaco	2000	347.2	deasphalter pitch	syngas
BASF AG	Germany	Texaco	1974	341.8	vac. resid. and fuel oil	methanol
China National Petrochem. Corp./Sinopec	China	Texaco	1988	341.8	visbreaker residue	gases
Quimigal Adubos	Portugal	Shell	1984	328.1	vac.residue	ammonia

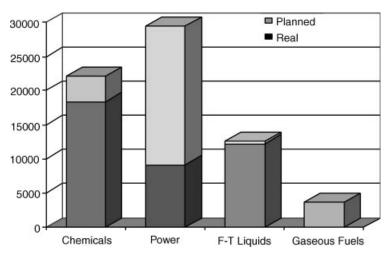


Fig. 22. Gasification by application.

A tremendous rise in capacity is expected to continue beyond 2005. Most of the capacity growth for gasification technology over the next several years appears to be power-based (Fig. 22). Both deregulation in the United States and demand growth worldwide appear to be the principal drivers. With this capacity increase, the use of synthesis gas for the production of electricity is approaching that for the production of chemicals.

To meet this need, an additional 33 plants with 48 gasifiers are expected to be constructed in the next 5 years, which would add another 18,000 MWth of synthesis gas capacity (45). A significant fraction of this expected growth will occur in the developing nations in the Pacific Rim as the need for further electrification of these nation's economies grow (Fig. 23). Western Europe will also see large generation increases where refineries need to fully utilize available

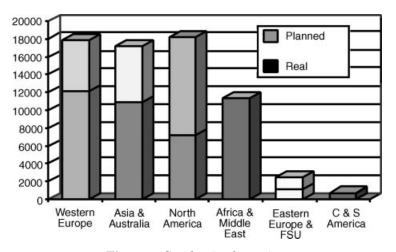


Fig. 23. Gasification by region.

feedstocks while reducing fuel oil production. Major capacity addition is expected in North America and will be concentrated in the refining industry as well. Very little or no growth is anticipated to occur in Africa or other regions of the world (see Table 9).

With regard to fuel, coal and petroleum residuals are by far the dominant feedstocks and account for >70% of the synthesis gas capacity. The major users of coal for synthesis gas production are Sasol, Dakota Gasification, and the current IGCC demonstration projects being sponsored by the U.S. Department of Energy. Natural gas is also an important feedstock for synthesis gas production, accounting for $\sim 20\%$ of today's capacity, and is used almost exclusively in the production of chemicals. Over the next several years, most of the growth in capacity will be from the gasification of coal and petroleum residuals, with a small fraction from petroleum coke. The growth in these feedstocks, however, will be used primarily to produce electricity. Coal, petroleum, and petcoke are the predominant feedstocks (Fig. 24) as indicated by the projected plants. No capacity additions for synthesis gas production from natural gas are projected.

10.6. Future Gasification Technology Development. Power Generation. For many years, government and industry have worked to develop the concept of integrating coal gasification with clean, efficient gas and steam turbines to create IGCC and various hybid power systems. In the United States, IGCC technologies are now being demonstrated as part of the U.S. Department of Energy's (DOEs) Clean Coal Technology (CCT) Demonstration Program. The significant strides in this program together with those from plants operating in the Netherlands and Spain have successfully demonstrated the performance of these coal-based power generation facilities. However, the capital cost of such advanced new plants and the risks involved with first-of-a-kind facilities and

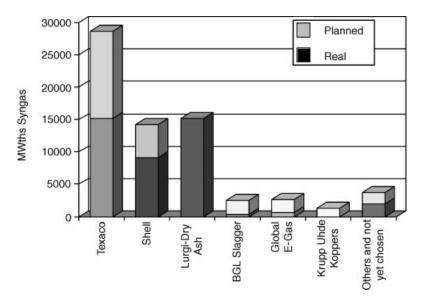


Fig. 24. Gasification by primary feed.

Table 10. Impact of Te	childingy Auva			lice
Gasifier "gas turbine" plant start-up	Destec "F" (2000)	Destec "G" (2005)	Destec "H" (2010)	Transport 2010
coal feed, t/day	4319	2449	2781	2552
oxygen feed, 95%, t/day	3592	1842	2027	N/A
no. of gasifiers trains	2	1	1	2
gas cleanup	amine	transport HGD	transport HGD	In situ desulf. transport/HG polisher
gas turbine (number)	GE7001Fa (2)	Westinghouse $G(1)$	GE "H" (1)	GE "H" (1)
gas turbine output, MWe	394	263	335	271
steam turbine output	255	141	155	140
net power, MWe	543	349	427	398
efficiency, HHV, %	40.1	45.4	49.0	49.7
total plant cost, KS, \$/kW	1241	1229	1087	961
production cost, mills/k Wh	14.7	14.5	13.3	13.0
cost of electricity, mills/k Wh	38.8	37.6	33.8	31.2

Table 10. Impact of Technology Advances on IGCC Cost and Performance

technologies will likely delay the widespread application of IGCC for power generation until the next decade.

Although estimates for IGCC applications range from \$1200-1500/kW, there is promise that costs may be reduced to below \$1000/kW making coalfired IGCC a more attractive alternative to gas-fired gas turbine generators. A recent study sponsored by the DOE analyzed the impact of technology improvements on overall cost and performance of an IGCC system (46). In Table 10 the impact is presented in terms of the evolutionary improvements with time in gas turbine technology (ie, F-, G-, and H-class turbines), gasification technology (Destec, renamed E-Gas, and transport gasifiers) and integration improvements. As apparent by inspecting Table 10, these evolutionary improvements have a significant impact on thermal efficiency and capital and O&M costs, ultimately reducing the cost of electricity. The nearly 10% increase in thermal efficiency due to the use of hot gas desulfurization and advanced turbines results in a 20% reduction in the emission of carbon dioxide per kWe generated (47).

The costs presented in Table 10 represent those for the first plants built with the indicated technologies and are likely to be somewhat higher than that for subsequent plants using the same technologies. Cost reductions and performance improvements are being made and will continue to be made as increased operating experience leads to improved, optimized process designs. Figure 25 provides an indication of what cost reductions might be expected from deployment of successive plants. The slope of the "learning curves" is representative of cost reductions associated with the deployment of other advanced technologies in the marketplace in the recent past. This figure shows that through successive deployment of the technology, costs may be reduced to <\$1000/kWe. As shown by

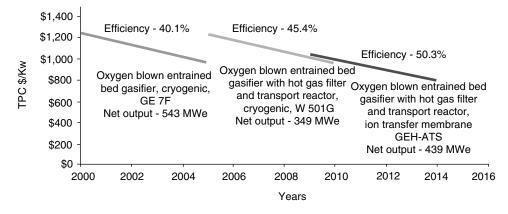


Fig. 25. Gasification cost and efficiency projections for various process improvements.

the curve on the right, potential exists for achieving a cost of about \$850/kWe, that which is considered by industry to be competitive with Natural Gas Combined Cycle (NGCC).

Coproduction. In the absence of niche market applications such as the petroleum industry or in the case of very high natural gas prices, gasification for power production will likely not occur to an appreciable extent until capital costs are significantly lowered. Identifying innovative opportunities will be critical to gasification's near-term market acceptance. For example, there continues to be a high demand for peaking and intermediate load capacity. Combining coproduction of chemicals and power in a gasification facility allows the flexibility to maximize power generation during peak demand periods and maximize chemicals production during off-peak periods, making gasification more attractive to the existing power market. It also allows flexibility in geographical location based on local grid peak-shaving needs. Also, combined power and methanol plant requires less capital than separate power and methanol processes. Locating the integrated facility near a chemical complex provides a means of improving gasification economics by ensuring full utilization of the gasifier and exploiting synergies between the processes. Utilizing cofeeding of opportunity fuels or "waste" with coal while coproducing power and fuels and/or chemicals is another way to avoid waste disposal costs and diversify fuel supply mix as a hedge against volatile natural gas prices.

An analysis was conducted for the DOE that studied the economics of coproduction, and demonstrated the advantage of multiple revenue streams, ie, from sale of electric power, liquid fuels, and chemicals. In one scenario, it is assumed that the power generated by coproduction is sold at the same price as that generated by natural gas combined cycle. Thus, the price of natural gas sets the cost of power and establishes a benchmark for economic comparison. In addition, liquid fuels are valued at an assumed premium of \$8/barrel over the reference world oil price of \$21/barrel in 2010 as predicted by the U.S. Energy Information Agency. Based on a fixed return on equity (ROE) of 15% and a coal price of \$30/ ton, IGCC is competitive with natural gas at ~\$3.75/million Btu. For comparison, by generating additional revenue, coproduction attains the same ROE at a lower natural gas price of about \$3.25/million Btu. These figures illustrate the improved profitability of coproduction (48). Yet another study concluded that through proper integration, coproduction can offer higher process efficiencies with little added capital (49).

Technology Development Needs. To reduce capital costs, increase efficiency and decrease emissions, technology development is required. The U.S. Government is undertaking a variety of research, development and demonstration projects with industry to accomplish those goals. Some of the general efforts being undertaken are as follows:

Gasification. Advances in the gasifier itself to enhance efficiency, reliability, and feedstock flexibility and economics are crucial for gasification system improvements. Research is being conducted on advanced gasifiers, such as a high throughput transport gasifier, so higher performance goals can be reached and the variety of possible feedstocks can be further expanded. Advanced refractory materials and new process instrumentation are being developed to improve system reliability and availability, operational control, and overall system performance. Studies of alternative feedstocks (biomass and waste from refineries, industries, and municipalities) are being conducted to improve gasifier flexibility and utility. Data from fluid dynamic models are being used to develop and improve advanced gasification.

Gas Cleanup and Conditioning. In the gas cleaning and conditioning area, the goal is to achieve near-zero emissions while simultaneously reducing capital and operating costs. Novel gas cleaning and conditioning technologies are being developed to reach this goal. Processes that operate at mild to high temperatures and incorporate multicontaminant control to parts per billion levels are being explored. These include a two-stage process for H₂S, trace metals, HCl, and particulates removal; membrane processes for control of H₂S, Hg, and CO; and sorbents/catalysts for NH₃and H₂S control. Investigation of technologies for mercury removal is also underway. For removal of particulates, both ceramic and metallic filters are being assessed.

Gas Separation. Advanced gas separation research offers the potential for substantial improvement in environmental and cost performance. It has been estimated that the cost of a conventional cryogenic air separation subsystem of an oxygen-blown gasification plant is >15% of the total plant cost. Parasitic power losses for air compression are also quite high. Development efforts are underway on oxygen ion separation membranes. This technology will also potentially enhance process efficiency as well. Improved hydrogen recovery and CO_2 removal are also important. Efforts are underway for developing high-temperature ceramic membranes for hydrogen recovery from syngas streams, as well as low-temperature approaches for hydrogen recovery and carbon dioxide removal.

 $Product / By \ Product \ Utilization.$ Markets and applications are being assessed for utilization of gasifier bottoms as a saleable by-product. Processes for near-zero discharge of wastes are being evaluated. Post-H₂S removal processes for the by-product production of sulfur and/or sulfuric acid are being researched. A single-step sulfur removal and by-product production process is also being evaluated.

Systems Analysis and Technology Integration. Plant design and system configurations for various market applications are being assessed. Novel approaches to technology integration, flexible fuel supplies, and product yields are being evaluated to achieve improved plant economics for gasification systems.

Gasification technologies will continue to play a major role in satisfying the increasing worldwide energy demand during this cmentury. Market forces and environmental concerns are resulting in the expanded use of integrated gasification combined cycle plants and hybrid systems (combined gasification and combustion). The gasifier vastly expands the fuel base beyond natural gas to include abundant and lower cost feedstock such as coal, biomass, and agricultural, forestry, and refinery wastes. It enables the separation of pollutants from the product gas and the reduction of greenhouse gases. These facilitate the clean and efficient generation of electricity and the production of chemicals and clean liquid fuels. The capability to coproduce electricity, chemicals, and liquid fuels makes the technology economically attractive to a broad range of industrial applications. Feedstock and product flexibility, high efficiency and near zero pollutant emissions make gasification the technology of choice for electricity generation and chemicals and liquid fuel production in the twenty-first century. Table 11 provides a list of upcoming or planned gasification plants in the near future.

Plant Owner	Country	Gasifier	Year	MWth	Fuel	Product
Global Energy, Inc./Fife Electric	United Knigdom	BGL	2002	732.5	coal and sludge	electricity
IBIL energy Sys- tems Ltd. (IES)	India	GTI U-GAS	2002	109.1	lignite	electricity and Steam
Global Energy, Inc.	United States	BGL	2003	727.0	coal and MSW	electricity and diesel
Global Energy, Inc.	United States	BGL	2003	727.0	coal and MSW	electricity and H ₂
Indian Oil Corp. Ltd.	India	Shell	2003	888.6	petcoke	H ₂ and electricity
Sokolovska Uhelna, A.S.	Czech Republic	HTW	2003	787.4	lignite	electricity
Sinopec/Shell	China	Shell	2003	466.2	coal	ammonia
AGIP Raffinazione S.p.A.	Italy	Shell	2003	456.6	visbreaker resid.	electricity and H ₂
Calla Energy Partners, LLC	United States	GTI U-GAS	2003	80.7	biomass	electricity
Sistems de Ener- gia Renovavel	Brazil	TPS	2003	68.4	biomass	electricity
ATI Sulcis	Italy	Shell	2004	956.9	coal	electricity
Sinopec/Shell	China	Shell	2004	956.9	coal	electricity and syngas
Unspecified Uti- lity Consortium	Japan	ICGRA	2004	585.4	coal	electricity
Sinopec/Shell	China	Shell	2004	478.5	coal	ammonia
Sinopec/Shell	China	Shell	2004	466.2	coal	ammonia
Waste Manage- ment & Proces- sors, Inc.	United States	Texaco	2004	410.1	ant. culm	diesel and electricity

Table 11. Planned Future Gasification Plants Worldwide

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Plant Owner	Country	Gasifier	Year	MWth	Fuel	Product
Koa Oil Co. Ltd.	Japan	Texaco	2004	287.1	petcoke Electri- city	
Boise Cascade Corp.	United States	GTI U-GAS	2004	102.5	biomass	electricity
Port of Port Arthur/Sabine Power I, Ltd.	United States	E-GAS	2005	2,029.4	petcoke	electricity
Petronor (Repsol- YPF)/Iberdrola (PIEMSA)	Spain	Texaco	2005	1,654.1	vac. residue	electricity and H ₂
TECO Power Services Corp./ Citgo/Texaco	United States	Texaco	2005	1,406.7	petcoke	elect, H ₂ and steam
Eagle Energy (TECO Power Services/ Texaco)	United States	Texaco	2005	1,367.1	petcoke	electricity
Proj. IGCC Nor- mandie (Total- FinalElf/EdF/ Texaco)	France	Texaco	2005	1,043.1	fuel oil	elect, H ₂ and steam
Hindustan Petro- leum Corp. Ltd.	India	Texaco	2005	649.3	petcoke	electricity
Refineria Gdanska SA	Poland	Texaco	2005	496.2	visbreaker resid.	elect, H ₂ and steam
Unspecified owner	Unspec. Eur. Country	Shell	2005	478.5	residue	electricity
Agip Raffinazione S.p.A.	Italy	Texaco	2005	409.1	visbreaker resid.	electricity
Unspecified Owner	United States	Texaco	2006	2,761.4	coal	electricity
Shell Deer Park Refining Co.	Untied States	Texaco	2006	1,400.0	petcoke	elect, H ₂ and steam
Dong Ting	China	Texaco	2006	1,170.8	coal	syngas
Beijing Coking	China	Texaco	2006	286.6	fuel oil	methanol
Netherlands Refining Co. BV	Nether- lands	Texaco	2006	47.6	waste plastics	electricity and CO

Table 11 (Continued)

11. Glossary

Agglomerating ash gasifier	Gasifier that maintains a sufficiently high tempera-
	ture to promote ash agglomeration (eg, U-GAS)
Ash fusion temperature	Temperature at which the ash melts ($^{\circ}$ C)
ASTM	ASTM
BGC	British Gas Company
Bituminous coal	ASTM coal ranking for coal with heating value
	>10,500 BTU/lb and Volatile Matter content $>$ 14%

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Blue Water gas	Product of cyclic gas generators that produce a blue
Caking coal	Coal that become sticky upon heating
Clean Air Act	A federal law enacted in United States in 1990 that entrusts the EPA to set limits on how much of a pollutant can be in the air anywhere in the United States.
CAAA	Clean Air Act Amendment
CCT	Clean Coal Technology Demonstration Program: A
Clean Water Act	U.S. department of energy initiative. A federal law enacted in United States in 1972 to protect the nation's waters, including lakes, rivers, aquifers and coastal areas.
Cold Gas Efficiency	The ratio of the chemical energy in the syngas relative to the chemical energy in the incoming coal.
Cogeneration	Production of high grade heat or steam in addition to electric power.
Co-production	Production of fuels or chemicals in addition to elec-
	tric power.
Criteria Air Pollutants	VOC
Cyclic Gas generator	Gas generators cyclically operated in combustion (with air) and gasification (with steam) modes.
Destec gasifier	Entrained flow gasifier originally developed by Shell Oil Co.
Deregulation	Changes enacted by several U.S. states to make electric industry open to competition, giving consu- mers the power to choose their electricity provider.
Devolatilization	The process of volatile matter release from coal as it is heated.
Direct hydrogenation	Obtaining liquid fuels from coal by heating pulver- ized coal with oil under high pressures and with hydrogen.
Direct liquefaction	Same as direct hydrogenation.
Dry ash gasifier	Gasifier in which the ash temperature is main- tained below ash fusion temperature.
E-Gas	Same as Destec gasifier
EIA	U.S. Energy Information Administration
ENCOAL	Mild Coal Gasification Project
Entrained flow gasifier	Gasifier in which the coal particles are entrained with the gasifying agent to react in a cocurrent flow.
EPA	U.S. EPA
EP	Extraction Procedure (toxicity tests)
FBC Fisher-Tropsch synthesis	Fluidized bed combustor Chemical process for producing liquid fuels from
	syngas
Fixed bed gasifier	A gasifier with a slowly moving packed bed of coal with a fixed height. Also known as moving bed gasifier.

Fusinite	Coal maceral derived from biodegraded or charred
ECD	wood.
FGD Flacidized had Cariforn	Flue gas desulfurization.
Fluidized bed Gasifier	A gasifier in which the bed of coal, char, and ash are
TLAD	fluidized.
HAPs	Hazardous Air Pollutants
HHV	Higher heating value
High rank coal	Coals with heating value greater than or equal to
IIDO	that of bituminous coal (>10,500 btu/lb)
HPC	Heavy paraffin cracking
HPS	Heavy paraffin synthesis
HRSG	Heat recovery steam generator
HYGAS process	Direct hydrogenation of coal in the presence of
	hydrogen and steam, under pressure, in two
	fluidized-bed stages
Hybrid power systems	Combined power systems employing both gasifica-
	tion to drive gas turbines and combustion steam
	boilers to fuel steam turbines.
IGCC	Integrated gasification combined cycle (steam and
	gas cycles)
Indirect liquefaction	Process of producing syngas from coal and then con-
	verting the syngas to liquid fuels
Koppers-Totzek gasifier	A commercialized entrained bed gasifier
KRW gasifier	Kellogg, Rust, Westinghouse gasifier. A fluidized
	bed gasifier tested in a pilot plant.
Liptinite	Coal macerals derived from hydrogen-rich
	sources such as spores, leaf cuticles, and algal
	colonies.
Low rank coal	Nonagglomerating coals with heating value less
	than that of bituminous coal (<10,500 btu/lb)
LHV	Lower heating value
Lurgi Gasifier	A commercialized fixed bed gasifier.
NPDES	National Pollutant Discharge Elimination System
	standards
NGCC	Natural Gas Combined Cycle
MAF	Moisture and ash free (analysis of coal)
MBG	Medium BTU gas (250–500 Btu/cu ft)
MF	Moisture free (analysis of coal)
Moving bed gasifier	Same as fixed-bed gasifier
MTG	Methanol to gasoline process
O&M costs	Operation and maintenance costs
Opportunity fuels	Low cost fuels readily available in certain markets
	(eg, biomass, municipal solid waste, sewage sludge,
	etc)
PC plants	Pulverized coal plants
Public Utilities	State regulatory agencies for electric utility
Commission	companies
Pyrolysis	Decomposition of coal with heat

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Rank	A classification of coals describing the relative ma- turity of the coal; higher ranks being further evolved having less volatiles, higher carbon, and less hetero-atoms.
RCRA	Resource Conservation and Recovery Act estab- lishes a framework for U.S. national programs to achieve environmentally sound management of both hazardous and nonhazardous wastes.
ROE	Return on equity
Repowering	Modification or enhancements to existing power plant to replace aging or outdated components or systems.
Retrofitting	Upgrading or improving an outdated technology using modern developments.
SASOL process	Synthetic liquid fuels plant operated by the South African Coal, Oil, and Gas Co.
SCGP	Shell coal gasification process
Slagging gasifier	A gasifier designed to melt and remove the coal minerals
Slagcrete	A concrete mixture with gasifier slag as a principal component.
SNG	Synthetic or substitute natural gas, consisting mainly of CH_4
Synthol process SMDS	A Fischer-Tropsch process developed at SASOL Shell Middle Distillate Synthesis process
Softening point	Temperature required for the onset of melting the coal ash
Sour syngas	Product gas containing acidic components such as H_2S
Steam gasification	Reaction of water and carbon to produce CO and hydrogen
Subbituminous coal	ASTM coal ranking for nonagglomerating coal with heating value >8,100 and <10,500 BTU/lb
sweet syngas	Product syngas with acidic components removed.
Syngas	A mixture of carbon monoxide and hydrogen (with some carbon dioxide and methane).
Tar	Complex condensable hydrocarbons produced from the heating of coal
TCLP	The standard test: toxicity characteristic leaching procedure test
TEXACO gasifier	Entrained flow gasifier commercialized by Texaco Oil Co.
TSP	Total suspended particulates
TWR	The tube wall reactor, a reactor that uses catalyst- coated tubes
Town Gas	Synthetic natural gas distributed through pipeline network to cities and town for street lighting and domestic uses.

Vitrinite	The most common coal maceral derived from woody
	tissue
VOC	Volatile organic compounds
Wellman-Galusha gasifier	A commercialized fixed bed gasifier
Winkler Gasifier	A commercialized fluidized bed gasifier

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