

## CHAPTER 3. - GASIFICATION OF COAL

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Both of the general processes for producing liquid fuels from coal (as used previously in Germany) depend upon gases obtained by gasifying coal. The Fischer-Tropsch synthesis employs a mixture of carbon monoxide and hydrogen, after suitable purification of raw synthesis gas to remove carbon dioxide and sulfur compounds. For the hydrogenation process, the gas produced from coal, after purification, is reacted further with steam to furnish additional hydrogen. With both processes, improvement of the gasification step offers a means for appreciably reducing the final cost of the synthetic liquid fuel product. In the Fischer-Tropsch and methanation processes, the cost of supplying the purified synthesis gas is around 70 to 80 percent of the total synthesis cost, and in hydrogenation the cost of hydrogen and its compression is around 40 to 45 percent.

The gasification portion of the synthetic liquid fuels program was then aimed at development of methods for making gas for use in the synthesis processes. The major work on gasification, considered in this section, was concerned with conventional aboveground processes. The special topic of gasification of coal belowground is considered later in chapter 4.

Status of Coal Gasification in 1944

Before 1944, large-scale coal gasification in the United States was limited to two basic processes. Coal carbonization was in use by some utilities and steel plants to produce a gas of about 500 to 600 B.t.u. per cubic foot<sup>6/</sup> heating value. The economic feasibility of this process depended on the production of marketable coke for use in domestic heating or reduction of

<sup>6/</sup> Throughout this chapter, volumes of gas were calculated, according to a gas-industry standard, as dry gas at 60° F. and 30 in. Hg.

iron ore, so such carbonization was always limited to the higher rank, coking, bituminous coals. The gas produced by this process was not suitable for synthesis of liquid fuels or chemicals, although hydrogen might be obtained from it by a liquefaction process.

The second process was the production of "water gas" by blowing air and steam, alternately, through a bed of the coke made in the first process or of certain noncoking bituminous coals or anthracite. The gas made by using coke consisted mainly of carbon monoxide and hydrogen and could be used for synthesis processes. The gas made from coal, on the other hand, contained substantial amounts of methane and other hydrocarbon gases and, with some carburetion, was suitable for the domestic market.

The gas-utility industry was already changing over to the distribution of natural gas before World War II, and this trend accelerated rapidly after the war. Because of the large supplies of natural gas available at low prices in the Southwest, the chemical industry also converted for the most part to the production of its synthesis gas (for example, for producing ammonia and alcohol) from natural gas instead of coal.

A third gasification process, in use before 1944 on a comparatively small scale, was the producer-gas process, which makes a low-heating-value gas by gasifying coal or coke continuously in a fixed bed with air and steam. Because of its high nitrogen content, this gas is used only for heating, as in open-hearth furnaces, glass furnaces, and coke ovens.

The large supplies of good coking coal in this country before the war retarded development of other possible gas-production processes with lower grade coals. Although comparatively minor improvements were made in the high-temperature coal-carbonization processes, a continuous process for producing water gas or synthesis gas by blowing a mixture of oxygen and steam through fixed beds of noncaking fuel was developed to replace the classic cyclic process. However, the economics of this process was not good, because of the comparatively high cost of the coke and oxygen, and the competition of the increasing supplies of natural gas.

Over a period of years before 1944, there had been only sporadic attempts to develop other processes, for example, low-temperature carbonization and certain gasification processes for lower rank fuels such as lignite. The Bureau of Mines was a pioneer in studying the complete gasification of lignite. As has been mentioned, the availability of natural gas after the war deterred the chemical and utility industries from process-development work in this field.

Two reviews of the status of coal gasification at that time have been published (22, 26). The patent literature, before 1944, shows that a large number of complete coal-gasification processes had been proposed, which had never reached the development stage in this country.

In Europe the situation was quite different. Without large supplies of coking coals or natural gas or petroleum, both the chemical and utility industries had been compelled to look to coal for fuel, and to its complete gasification to supply the domestic heating market as well as to produce synthesis

gas for making chemicals and liquid fuels. This development work, centered in Germany, was accelerated by the artificial demand for liquid fuels caused by the buildup of the German war machine (25, 27). Processes developed, or in development during 1930-44, aimed at utilization of the noncoking bituminous coals and brown coals which are available in large quantities on the continent.

The Lurgi, Winkler, Wintershall-Schmalfeldt, and Koppers are typical of these processes, which were designed for nonslagging operation. Some development work had been done on slagging gas generators in which steam and oxygen were the gasifying agents, and comparatively large 1-1/2- to 2-1/2-inch coke was the fuel.

The Lurgi process, operated at 20 to 30 atmospheres, had been used to gasify granular brown coals with oxygen and steam. The gas produced had a heating value of about 450 B.t.u. per cubic foot and could be used in synthesis processes, although it contained a fairly high percentage of methane. It was used for city gas in the Leipzig area. For the production of synthesis gas the Lurgi process has two disadvantages, the high methane content of the gas and the need for a sized, noncoking fuel with ash of a high melting point.

The Winkler process was used to gasify brown-coal char in a fluidized bed with oxygen and steam. The gas is suitable for synthesis processes. Any noncaking bituminous coal, including the finer sizes, can be gasified by this process, but fuel with ash of high melting point is required. The dust-removal problem is serious.

The Wintershall-Schmalfeldt process was employed to gasify brown coals in suspension in a recycling stream of superheated gas and steam. In later developments some oxygen was used to augment the heat supply for the steam-carbon reaction. This process required a large investment in equipment; it operated below slagging temperatures, and conversion rates were low.

The Koppers process was proposed to gasify finely divided fuels (brown coals, lignites, or bituminous coals) in a stream of oxygen and highly superheated (1,800° to 2,000° F.) steam. This process was designed for nonslagging operation and is better suited to the more reactive lower rank fuels.

#### Bureau of Mines Contributions to Coal Gasification, 1944-55

##### Economics of Coal-Gasification Processes

The preceding review of technical developments to 1944 indicated the need for developing a process for producing, directly from coal, a low-cost gas suitable for preparing synthetic liquid fuels.

The basic problem was to find a low-cost fuel and a low-cost method of supplying the heat for the steam-carbon reaction. To reduce costs further, the process should be capable of large outputs per unit of equipment size, that is, per cubic foot of reaction space. Also, the raw gas should not require expensive methods for removing impurities, such as sulfur, carbon dioxide, and dust or unreacted fuel.

The Bureau examined several approaches to the problem of finding a low-cost fuel in the early phases of the program. One approach was to find a method for upgrading low-grade fuels so that they could be used in a standard water-gas machine. This could be done by briquetting certain fines then considered waste products and available at low cost. However, consideration of ~~all possibilities of this approach indicated no hope for an economical solution.~~

Another approach was to adjust the process to the fuel. Available low-cost fuels were characterized by high ash content and fine size (screenings, etc.). It was also apparent that, to produce large outputs of coal at low cost, future mining probably would tend toward increased production of fines of higher ash content.

Consequently, it was decided that a process capable of gasifying a pulverized fuel offered the best chance to cut fuel costs and probably offered the best possibilities of being applicable to American coals of all ranks, from lignite to anthracite, without serious modifications of equipment. A related requirement would be that the process operate at temperatures favorable for the less reactive fuels and above the ash melting point to simplify ash handling and disposal. This would allow removal of all or a considerable part of the ash as molten slag.

Bureau of Mines personnel allocated to this program was engaged until late in 1945 with the Technical Oil Mission (6). The Mission survey teams obtained information on various German processes, particularly the Lurgi and the Winkler, as well as on methods for removing dust and sulfur.

The major Bureau of Mines research on gasification was carried on at the Morgantown, W. Va., experiment station. Early in 1946 facilities for coal preparation were installed, a laboratory-scale gasifier was constructed, and laboratory studies were started on sulfur compounds present in gases made from coal. Literature studies were made of available data (7) on American coals and on the reactivities of carbon and carbon-containing materials.

### Basic Research

#### Kinetic Study of the Steam-Carbon Reaction

In planning industrial processes based on chemical reactions, such as the gasification of pulverized coal, knowledge of the rates of reaction is essential. Data were obtained on the gasification of 200- to 230-mesh petroleum coke and electrode graphite in a large excess of steam, within an electrically heated 6-1/2-foot-long partly isothermal reactor tube (19).

Carbon particles, preheated to the reactor temperature, were dropped into the reactor at a constant rate of 4 to 5 grams per hour. An excess of preheated steam flowed with the carbon particles to the reactor exit. Partly gasified particles were collected in a receiver. The residual carbon was weighed and again passed through the reactor at the same rate. Each run consisted of four or five such passes. The residence time of the particles under gasification conditions was determined for each run.

Initial reaction rates were determined graphically. The initial reaction rate of graphite was much higher than that of petroleum coke, but the rates approached each other as contact time increased with repeated recycling of the particles. Petroleum coke reacted at nearly uniform rate with respect to time, but the initially high reaction rate of graphite decreased rapidly (see fig. 5). Further Arrhenius graphs of the data yielded straight lines for reaction rate versus the reciprocals of absolute temperatures with both graphite and petroleum coke; surface reaction rates were apparently the controlling influence throughout the temperature range of 1,800° to 2,500° F.

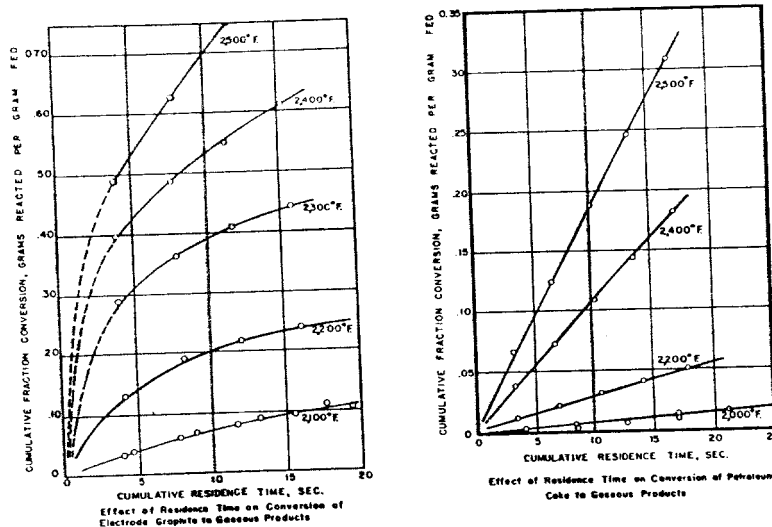


FIGURE 5. - Rates of Reaction of Electrode Graphite (left) and Petroleum Coke (right) With Steam at Temperatures up to 2,500° F. and Residence Time up to 25 Seconds.

A preliminary study was made of the effect of oxygen on the rate of the steam-carbon reaction (15, pp. 51-52). Extrapolation of carbon conversions to zero oxygen concentration, and comparison of the initial rates thus obtained with those determined in the absence of oxygen, were used to indicate the effect of oxygen. Runs were made with petroleum coke at temperatures from 1,800° to 2,500° F. and with volume concentrations of oxygen in steam from 0 to 1 percent. The initial reaction rates accelerated rapidly above 2,200° F. and appeared to increase with the oxygen-input ratio.

The effects of particle sizes and geometric surface areas on the steam-carbon reaction were studied with graphite particles that ranged from 60- to 230-mesh in size (16). The rate of reaction of graphite with steam at 2,400° F. was found to be a straight-line function of particle diameter, the rate increasing as particle diameter decreased.

## Flashing and Gasification of Coal-Water Slurries in Heated Coils

### Micronizing Coal in Steam

The object of this work was to develop a method for flashing coal-water slurries, in an externally heated alloy-steel tube coil, into a suspension of fine coal in steam for the purpose of feeding it into gasifiers at uniform rate. A practical apparatus was constructed (see figs. 6 and 7) for evaluating the essential variables in micronization of coals.

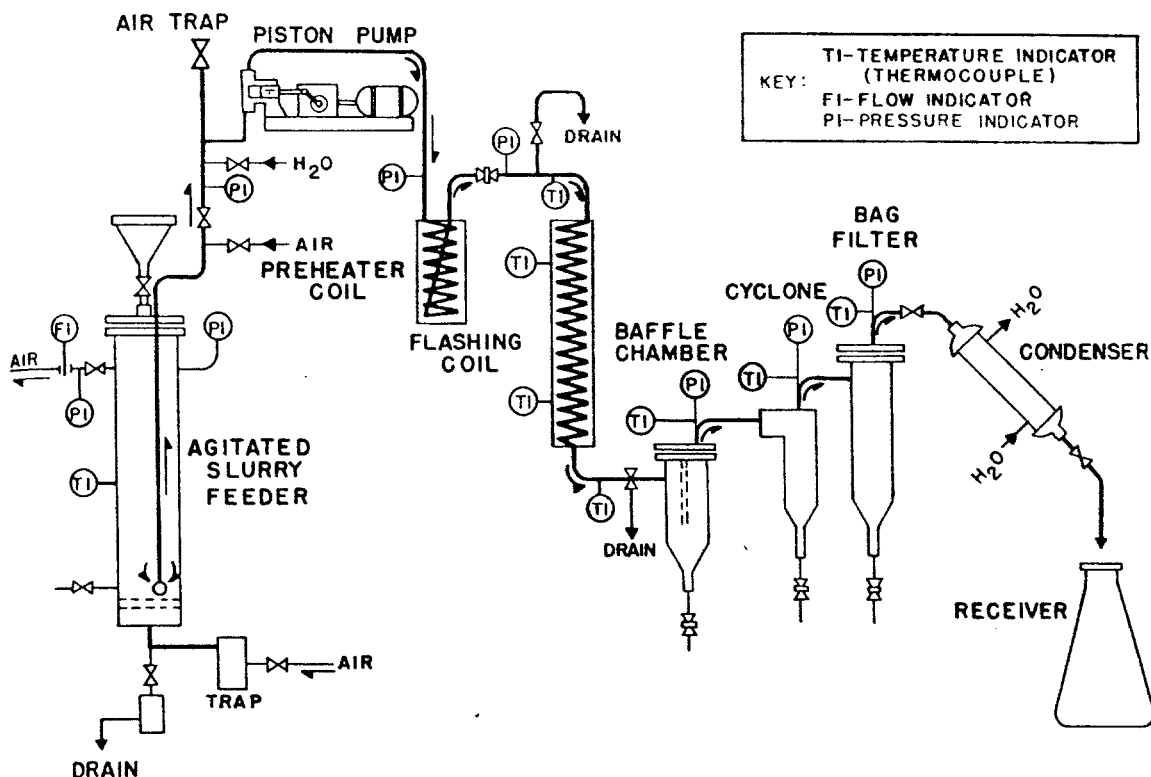


FIGURE 6. - Improved Metallic Bench-Scale Apparatus for Flashing Coal-Water Slurries.

With anthracite and a lignitic-subbituminous coal, pressures of flashing up to 150 p.s.i.g. and temperatures up to 750° F. were found to have no effect on particle size except as they changed the velocity of steam through the 50-foot-long, 3/8-inch tube coil. The micronization observed was due to impact and abrasion, which tended to increase with the velocity. Considerable increase in flow velocity has been achieved by decreasing the pressure at the coil outlet. The degree of micronization for several runs is shown graphically in figure 8.

### Gasification of Coal Slurries in an Externally Heated Metallic Tube-Coil Reactor

Efforts have been made recently to gasify powdered coals without using oxygen. This is especially desirable when waste heat at 1,600°-1,800° F. or low-cost heat from nuclear fission might be available. To determine the feasibility

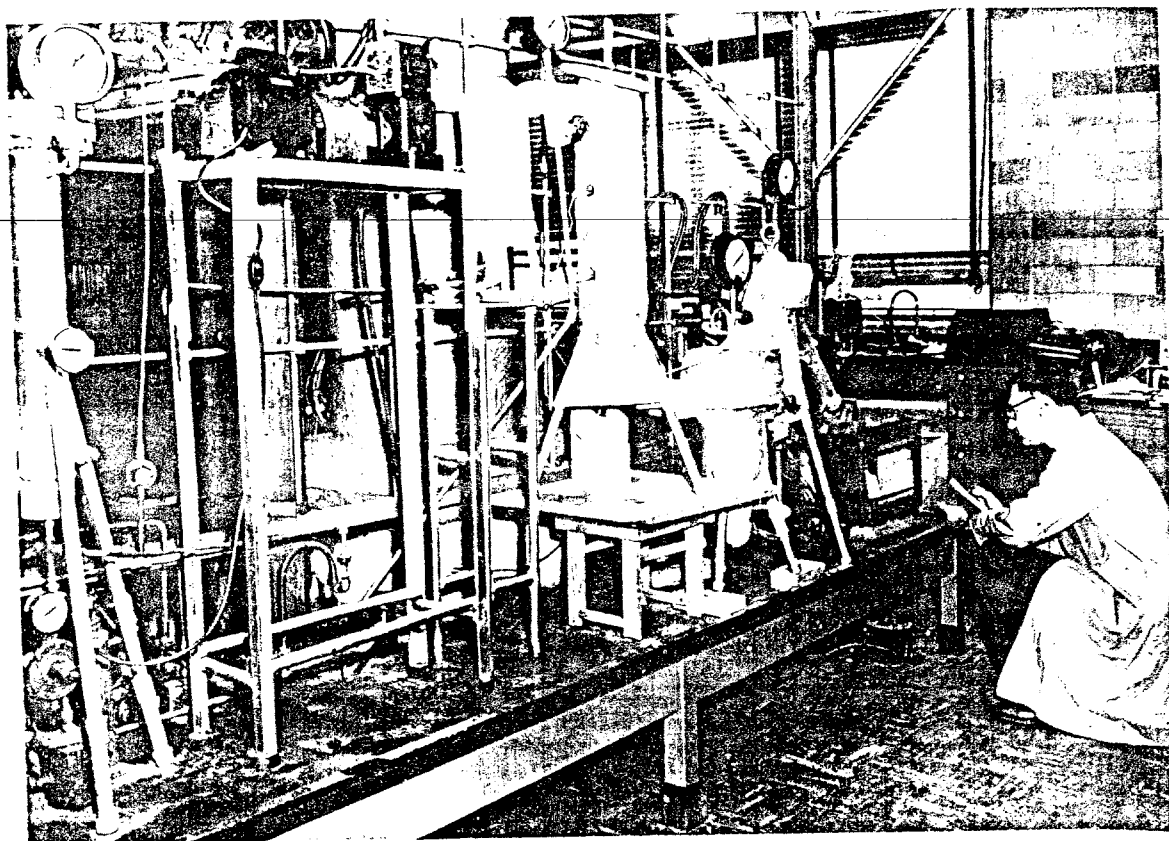


FIGURE 7. - Apparatus for Micronization of Coal by Flashing Coal-Water Slurries in Heated Coils.

of gasifying a coal slurry in an externally heated metallic tube coil, a laboratory-size coil reactor (see fig. 9) was constructed. Comparatively coarse granular coal reacts with steam in a gasifier of such design at relatively low temperatures for two reasons:

1. Flashing of the slurry into a suspension of coal in high-velocity steam micronizes the granular particles. The enormously increased surface area increases the rate of reaction between carbon and steam at lower temperatures.
2. The rate of diffusion between the reactant steam and gaseous products increases considerably due to the turbulence of the flow.

The coal-water slurry is pumped through a 117-foot-long, electrically heated, 3/8-inch, alloy-steel tube coil having an inside diameter of 0.315 inch. Argon is passed slowly through the insulation of the furnace to protect the coil from external corrosion. Reaction products and excess steam pass through a condenser and scrubbing vessels; the gases are then sampled, metered, and vented.

In preliminary runs coal slurries were gasified at 1,350°-1,850° F. with coal feed rates of 1-5 pounds per hour. About 65 percent of the carbon in Lake deSmet, Wyo., coal gasified in about one second at 1,850° F. temperature. The



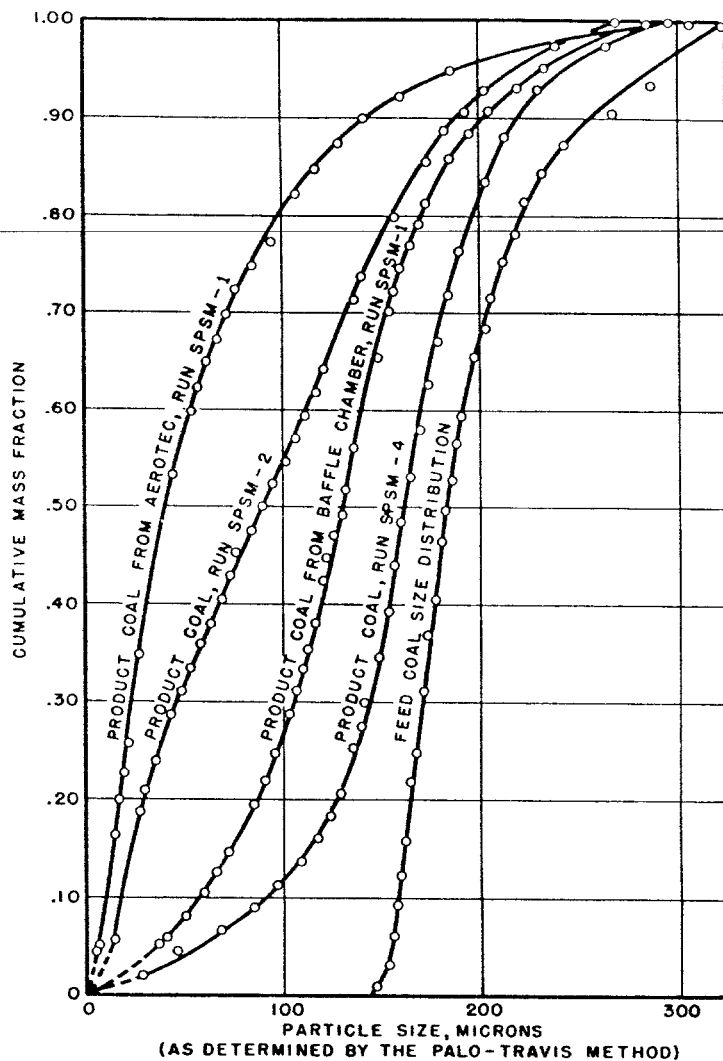


FIGURE 8. - Size Distributions of Feed and Products From Micronization of Coal.

gas produced contained about 16 percent carbon monoxide, 60 percent hydrogen, 20 percent carbon dioxide, and 3-1/2 percent methane. This was a high rate of carbon conversion, in view of the extremely short residence time. Further work is in progress.

#### Study of Metal Chelates as Oxygen Carriers

Investigation of metal chelates as oxygen carriers is aimed toward developing a process for producing tonnage oxygen based on the selective sorption of oxygen from air and subsequent recovery by desorption. Such a process should satisfy the following conditions:

1. Relatively low temperature difference between absorption and desorption steps.
2. Low heats of reaction for the oxygenation and reduction steps.