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Catalytic Coal Gasification: An Emerging Technology

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The production of gaseous fuel from coal has been practiced for hundreds of years. In 1670 it was discovered that simply heating coal in a closed vessel produces a gas that burns with a luminous flame. The first practical use of this phenomenon may have occurred in 1792, when a Scotsman named William Murdoch used coal gas to light his home. A manufactured gas industry began in England in 1810 and in the United States in 1816.

Summary. Catalytic coal gasification is being developed as a more efficient and less costly approach to producing methane from coal. With a potassium catalyst all the reactions can take place at one temperature, so that endothermic and exothermic reactions can be integrated in a single reactor. A key aspect of the concept involves continuous recycling of product carbon monoxide and hydrogen to the gasifier following separation of methane. Development of the process has advanced steadily since the basic concept was proposed in 1971. A 23-day demonstration run was recently completed in a process development unit with a coal feed rate of 1 ton per day. The next major step in the program will be to design and construct a large pilot plant to bring the technology to commercial readiness in the late 1980's.

In the later 1800's, the water gas process was developed to make large quantities of burnable gas for lighting applications by reacting steam with hot coal. The product had to be enriched by adding volatile hydrocarbons to bring the gas to the illuminating power of gas "distilled" directly from coal. Although the approach was inefficient and cumbersome by today's standards, "carbureted" water gas accounted for 57 percent of U.S. manufactured gas production at its peak in 1947. The manufactured gas industry was gradually replaced by a new modern gas industry supplied by natural gas during the 1950's and 1960's (*1*). Carbureted water gas is a mixture of methane, hydrogen, carbon monoxide, carbon dioxide, nitrogen, ethane, oxygen, and illuminants. Commercially distributed natural gas is essentially methane with small amounts of other light hydrocarbons and various impurities.

During the 1970's, it came to be realized that the world's supplies of inexpensive petroleum and natural gas are limited and that alternative forms of these fuels will be required in the future. One such option involves the production of synthetic or substitute natural gas (SNG) from coal. There are several reasons for interest in this option:

1) Natural gas and SNG burn cleanly.

2) A vast array of pipelines and end-use apparatus is in place in many of the developed countries for the use of gaseous fuels.

3) World coal reserves are enormous, and it is now well established that coal can be readily gasified.

4) Projections indicate that SNG from coal should be an economically attractive substitute fuel in many areas of the world in the 1990's.

There are a number of coal gasifiers that have been demonstrated at large scale and are considered available for commercial use today. The most widely used of these so-called first-generation concepts is the Lurgi gasifier. Also available are the Koppers-Totzek and Winkler systems. In advanced development are a number of second-generation systems, including Texaco, Shell, and slagging Lurgi. All of these approaches involve the gasification of coal with steam and oxygen. Differences are related to configuration, product yields and distributions (*2*), complexity, operating temperatures and pressures, and ability to handle different types of coals.

Our laboratory has been interested in improved coal gasification concepts since the late 1960's. In the early 1970's R. E. Pennington, N. C. Nahas, L. W. Vernon, and K. K. Koh conceived a method for gasifying a wide range of coals at a relatively low temperature to produce an almost pure stream of methane (*3*). This third-generation catalytic coal gasification (CCG) process (*4*) is now in development and is the subject of this article.

Chemistry of Coal Gasification

Coal is a nonhomogeneous, solid material composed of carbon (55 to 95 percent), hydrogen (3 to 6 percent), water (1 to 30 percent), and mineral matter (1 to 20 percent), which is largely silicon, alu-

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minum, calcium, magnesium, and iron (the wide ranges indicate that coal compositions vary dramatically). Coal combines with steam at high temperatures (550° to 1650°C) to form carbon monoxide and hydrogen as follows:

$$C + H_2O \rightarrow H_2 + CO \quad (1)$$

Here coal (nominally $CH_{0.8}$) is approximated as pure carbon. The energy required for this reaction is 134 kilojoules per mole.

Methane (SNG) is a gas with a high energy content—about 37 megajoules per cubic meter (1000 Btu's per cubic foot) at standard temperature and pressure. A mixture of CO and H_2 has a lower energy content (about 11 MJ/m^3), but it can be methanated at roughly 425°C as follows:

$$3H_2 + CO \rightarrow CH_4 + H_2O \quad (2)$$

This reaction liberates 226 kJ/mole. Overall, the reaction is thus

$$2C + 2H_2O \rightarrow CH_4 + CO_2 \quad (3)$$

Because the proportions of CO and H_2 in Eqs. 1 and 2 are different, an intermediate step called the "water gas shift" is required:

$$CO + H_2O \rightarrow H_2 + CO_2 \quad (4)$$

This reaction occurs at 400° to 500°C and liberates 33.5 kJ/mole.

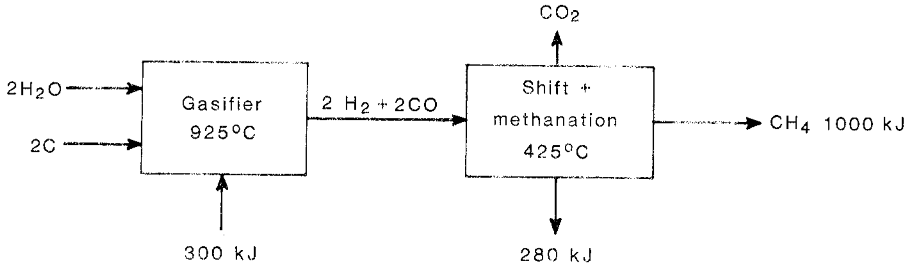


Fig. 1. Thermal coal gasification to methane. Coal and steam are combined in a high-temperature gasifier to produce CO and H_2, which must subsequently be shifted and methanated at a lower temperature.

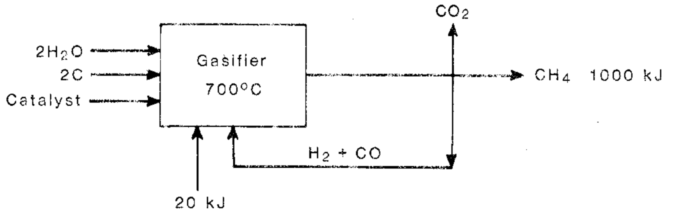


Fig. 2. Catalytic coal gasification to methane. Catalyzed coal, steam, and recycled CO + H_2 react in a single vessel.

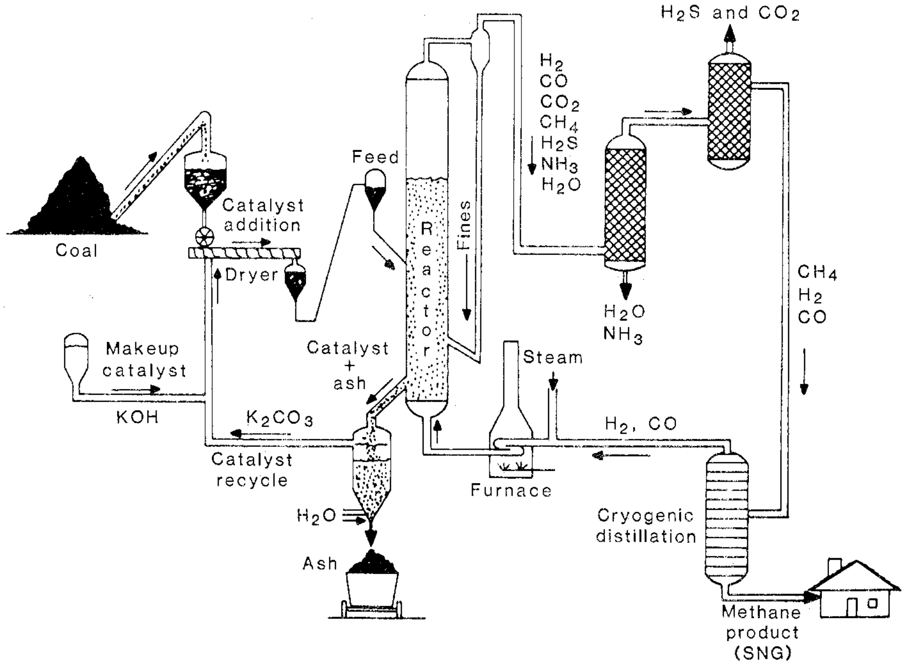


Fig. 3. Schematic of the catalytic gasification process.

On the basis of this chemistry, so-called thermal gasification systems operate as indicated in Fig. 1, where heat inputs are scaled to 1000 kJ of product methane for simplicity. Thermal gasifiers normally operate at 900° to 1600°C. Coal, steam, and oxygen are the feeds, the oxygen burning some of the coal to liberate the high-temperature thermal energy required for the process. Oxygen is preferred over air for making intermediate or high Btu gas because it avoids nitrogen contamination of the product gases.

Shift and methanation occur at about 425°C, the temperature at which heat is removed from the second stage. Because of this lower temperature, the heat liberated by methanation cannot be used to provide the high-temperature heat required by the gasifier, resulting in a significant inefficiency.

Catalytic Coal Gasification

One opportunity to improve the coal-to-methane process involves heat integration between the gasifier and the shift-methanator. This would require lowering the gasifier operating temperature and increasing shift-methanation temperatures. One means for accomplishing these changes involves the use of a coal gasification catalyst.

It had been known for some time that alkali metal salts of weak acids (for instance, K_2CO_3, Na_2CO_3, K_2S, and Na_2S) can catalyze the steam gasification of coal. The literature on the catalysis of carbon gasification is extensive [see reviews by Walker *et al.* (*5*) and Johnson (*6*)]. Taylor and Neville (*7*) tested the catalytic effects of a number of alkali salts and found K_2CO_3 and Na_2CO_3 particularly active, with K_2CO_3 showing the greatest activity or gasification rate. Kroger (*8*) extended the list of alkali metal salts and transition metals as gasification catalysts and he also studied mixtures of these compounds. Frequent publications on this topic demonstrate that it remains an active area of research.

In the early 1970's at our laboratory, researchers confirmed that at catalyst concentrations on the order of 10 to 20 percent by weight K_2CO_3 on Illinois bituminous coal, commercially acceptable gasification rates could be obtained at 700°C, compared to a required temperature of about 925°C without catalyst. More significantly, they discovered that potassium on coal or char (coal that has been partly depleted of carbon and hydrogen by gasification) was a good methanation catalyst at gasification temperatures. It was later found that potassium hydroxide could be easily applied to coals in a water solution and recovered as K_2CO_3 from the spent coal char with a water wash.

In Exxon's CCG concept the gasification, shift, and methanation reactions are all made to occur at the same temperature through the use of a potassium catalyst. This permits virtually complete heat integration of the gasification and methanation steps, significantly improving overall efficiency. At the 700°C operating temperature, thermodynamics does not allow complete methanation, so product CO and H_2 must be recycled back to the reactor. The process is shown schematically in Fig. 2.

The embodiment of this concept in a practical process is shown in Fig. 3. Beginning at the left, coal is crushed to approximately 1 to 2 millimeters (8 to 16 mesh) and slurried with an aqueous solution of KOH and K_2CO_3 recycle catalyst. The coal is then dried and fed through a series of lock hoppers to a fluidized bed reactor, which operates at 700°C and 3.5 megapascals (500 pounds per square inch). The catalyzed coal is fluidized with a mixture of steam and recycled CO and H_2 moving at about 30 centimeters per second.

At a solids residence time of 10 to 12 hours, approximately 90 percent of the carbon in the coal is gasified, and any pyrolysis liquids or lighter hydrocarbons that may be formed are completely converted to gas within the reactor. Residual solids are removed through a series of valves and washed with water to recover soluble potassium in the carbonate form. The recovered catalyst solution is recycled and the washed material sent to landfill or possibly used for constructive purposes.

Exiting from the reactor is a combination of gases and fines (very small coal particles that are produced in crushing or from various phenomena in the reactor). The fines are separated in a cyclone and returned to the bed for further gasification or sent directly to the catalyst recovery system. The gases are sent through a series of gas separators to remove water, ammonia, hydrogen sulfide, and CO_2. The resultant mixture of CH_4, CO, and H_2 is then separated by cryogenic distillation to recover the product methane. The remaining CO and H_2 are compressed, sent to a furnace that provides modest preheat, and then returned to the reactor, where they undergo an exothermic reaction (Eq. 2) to form methane and supply most of the energy required for the gasification process.

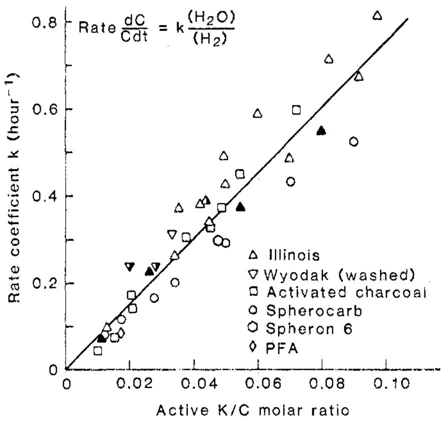


Fig. 4. Gasification rate as a function of potassium loading on a variety of carbonaceous materials. *PFA*, polyfurfuryl alcohol coke.

The potential advantages of the CCG process are as follows:

1) The catalyst, which promotes both gasification and methanation, allows efficient heat integration and minimizes heat input requirements.

2) There is no need for oxygen, which is expensive and can cause coal ash slagging with associated operating problems.

3) No pyrolysis liquids are produced.

4) Separate shift and methanation steps are not required.

5) The use of catalyst results in moderate operating temperatures.

6) Fluidized bed reactors can be relatively easily scaled to very large sizes, so that comparatively few reactors (three or four) would be required in a commercial plant.

Along with these advantages there are some problems. First, potassium catalyst is not inexpensive, and it must be recovered and recycled for the process to be economical. A water wash recovers 65 to 70 percent of the potassium, primarily as K_2CO_3, which is an effective recycle catalyst. Second, at 90 percent carbon conversion the amount of residue (char or ash) to be disposed of is greater than for alternative processes. Disposal of solids is a concern in any coal conversion process. In the case of CCG the solid residue consists of normal ash materials, which should not represent a toxicological problem; potassium in a nonsoluble state (the char will have been water-washed); and unconverted carbon. Detailed analyses of disposal issues have not yet been performed, but the presence of unconverted carbon, potassium, and ash minerals suggests that use for brick, cement, or other applications may be feasible, in which case the expected disposal debit could be reduced or eliminated.

Basic Research

Although the literature on alkali-promoted gasification spans more than a century, diverse and sometimes conflicting mechanisms have been proposed to explain the observed phenomena. As CCG has advanced through the predevelopment and development phases (discussed below), a supporting basic research program aimed at developing better understanding of the reaction mechanisms of potassium-catalyzed gasification has been maintained. Many of the effects of potassium on coal have been studied (*9*).

As an example, Fig. 4 shows the gasification rate coefficient, *k*, plotted as a function of the active potassium/carbon molar ratio. The K/C ratios for coal and coal chars have been adjusted to reflect only active potassium by subtracting the amount that is bound in inactive forms by certain ash minerals. As shown in Fig. 4, the gasification rate increases linearly with the catalyst loading. The form of the carbon appears to have little effect on the gasification activity. The figure contains data for several coal chars, an activated charcoal derived from coconut shells, and several very pure amorphous carbons.

The method of catalyst impregnation also seems to have little effect on the gasification rate. The open data points in Fig. 4 characterize samples that were impregnated with an aqueous solution of K_2CO_3 or KOH; the shaded points are for samples that were mixed with dry powdered K_2CO_3; and the half-shaded points represent samples that were subjected to ion exchange with potassium. These findings are consistent with the hypothesis that active sites form on the surface of coal. All these experiments were performed in a small flow reactor (mini-gasifier) at a total pressure of 1 atmosphere.

Although the chemical state at the active catalytic sites is not understood, it appears that K_2CO_3 may combine with the char at the site where CO_2 is released. This mechanism appears consistent with experiments in which K_2CO_3 was labeled with carbon-14, which was subsequently liberated when reacted with coal. Further, x-ray diffraction studies have shown the disappearance of K_2CO_3 as it interacts with coal. Kinetic studies with isotope tracers have shown that the decomposition of water on the active sites is a fast, reversible reaction resulting in the formation of surface oxides. Rate inhibition arises from the reversal of this reaction by the hydrogen product. A subsequent rate-determining step involves a critical surface oxide which leads to liberation of carbon monoxide.

Some critical steps in the proposed gasification mechanism that is evolving from these studies are potassium-carbon complex formation

$$\text{Char} + K_2CO_3 \rightarrow \text{K-char} + CO_2 \quad (5)$$

water activation

$$\text{K-char} + H_2O \rightleftarrows \text{K-char-O} + H_2 \quad (6)$$

and gasification

$$\text{K-char-O} \rightarrow \text{K-char} + CO \quad (7)$$

These mechanism studies have already proved useful in the formulation and evolution of kinetic models used to design the gasification reactor (*10, 11*). They may also be of value in developing and understanding other catalyst systems that could lead to an improved process.

The methanation reaction has also been studied extensively with small, fixed-bed reactors. Figure 5 shows the conversion of CO and H_2 to methane as a function of temperature in an experiment in which CO and H_2 are passed over an Illinois bituminous coal char impregnated with K_2CO_3. At temperatures above about 700°C, equilibrium conversions were obtained even at very high gas velocities. At lower temperatures conversions fall below equilibrium levels due to kinetic limitations. Recent experiments have shown that very high carbon conversion (approaching 100 percent) affects the methanation rate. These data have been factored into the kinetic reactor model, enabling more accurate predictions of methane production in reactors with highly converted chars.

Process Development

A process development program is designed to bring a technology from the small-scale laboratory phase to the commercial plant stage in the shortest time and at the lowest cost possible; both time and cost are a function of the incentives and risks involved. The steps that we follow in developing synthetic fuel processes involve four major phases: predevelopment, development, precommercialization, and commercialization. In each phase there are various degrees of relatively fundamental research, laboratory testing, and engineering. Laboratory research involves the design and operation of experiments in both bench apparatus and small pilot plants to obtain relevant data. In general, engineering studies include (i) development of study designs for commercial systems, which can be used to identify technical issues and to quantify process debits and credits uncovered in the research; (ii) design of pilot plants; (iii) planning and design of pioneer commercial plants; and (iv) hardware development. Engineering also provides critical guidance to laboratory research by identifying data needs and practical and economical commercial operating conditions and configurations. In the development of CCG there is a close interplay between laboratory and engineering activities.

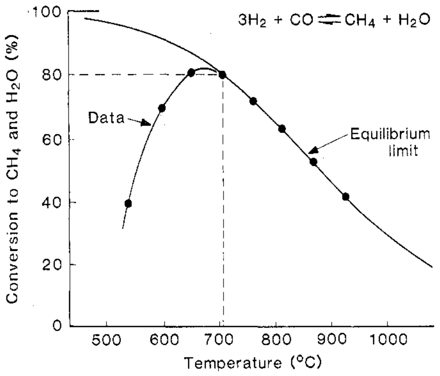


Fig. 5. Conversion to methane as a function of temperature. Conditions: 20 percent K_2CO_3, 80 percent char, 3.5 MPa, and a space velocity of 2300 volume/hour/volume of bed.

The predevelopment phase of CCG research consisted of work in bench-scale minigasifiers (on the order of grams of coal) and in small pilot plants (25 to 100 kilograms per day). The objective was to identify preferred ranges of operating conditions in order to establish the basis for design of a larger unit and the key data needs and incentives for the process development program.

The centerpiece of the current CCG process development phase is a process development unit (PDU) operating at a nominal 900 kg (1 ton) per day. The purpose of this unit is to establish the data base and incentives needed for the precommercialization phase.

The precommercialization phase of CCG development is envisioned to involve the operation of a large pilot plant as well as supporting operation of the PDU, bench research, and engineering studies. The objective of this phase will be to provide sufficient scale-up data that the pioneer commercial plant (the first commercial-scale unit) can be designed with acceptable technical risk. The size or capacity of the large pilot plant must therefore be set at the minimum judged necessary to obtain representative scale-up data at the lowest cost.

A pioneer commercial plant is a standalone facility, whose function is to operate as a profitable commercial venture. It should contain all of the equipment characteristic of full commercial size. However, the pioneer plant could contain only a single train of equipment in some or all of the plant sections, where a commercial plant might have a number of trains—for instance, parallel coal preparation sections and parallel reactors.

Process Development Unit

The catalytic gasification process is now in the process development phase, with work being carried out in the PDU. This unit is large enough to permit continuous feed (900 kg/day) and withdrawal, and it can operate under projected commercial operating conditions. On the other hand, the unit is small enough so that modifications can be made relatively quickly and at reasonable cost.

The PDU includes complete facilities for coal preparation, gasification, product gas separation, and catalyst recovery. The fluidized bed reactor has an inside diameter of 25 centimeters and is approximately 25 meters tall. During operation, the bed extends to the 15- to 17-m level. The reactor and feed equipment are contained in a 12-story tower. Figure 6 shows the building with its relatively slender reactor during the construction phase in 1978. The product gas separation and catalyst recovery sections are contained in separate adjacent facilities.

The PDU is highly automated, including a programmable controller for sequencing and cyclic operations, safety interlocks, and emergency shutdown operations. Data acquisition is facilitated by a minicomputer connected to an analog-to-digital converter. Approximately 800 process variables are continuously monitored and reported at varying intervals. Off-line data reduction and reconciliation programs utilizing the process variable data, as well as laboratory solids analyses, provide the complete data work-up. Statistical analyses are used to adjust the data to satisfy numerous material balance constraints.

The unit began operation during the first half of 1979. Early in the operations several problems were encountered that had not been apparent in the bench-scale and small pilot-scale research (*12*). The most significant problems were agglomerate formation in the gasifier, a low gasifier fluidized bed density, and lower than anticipated methane production. These problems and their solutions are discussed in detail below.

Agglomerate Formation in Gasifier

During start-up of the PDU, coal feeding and char removal required a great deal of attention because the feed and withdrawal lines had a tendency to plug due to agglomeration of the feed coal. As operating conditions were narrowed to approach design targets, problems of large agglomerate formation were encountered in the bottom of the gasifier. The agglomerates were observed to be of two types. The first type was black, insoluble in water, and consisted of low-conversion coal particles cemented together by hydrocarbon material that had gone through a plastic stage. The second type was light in color, contained essentially no hydrocarbons, and disintegrated when placed in water; analysis showed that this agglomerate consisted of ash particles cemented together by water-soluble potassium carbonate.

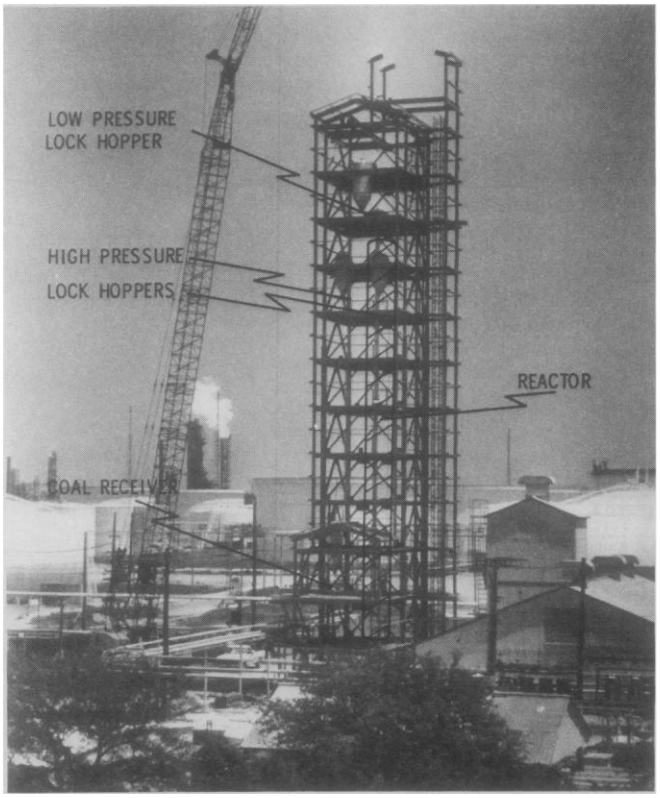


Fig. 6. Photograph of the PDU under construction. The slender pipe in the center of the tower is the fluidized bed reactor.

The problem with agglomerates was related to the fact that if they were allowed to accumulate in the bottom of the gasifier in the presence of steam for several hours, they would form a bridge or plug that would prevent char withdrawal. Process variable studies showed that agglomerates seemed to be formed near the point where coal first enters the fluidized bed. Subsequent studies indicated that the agglomeration could be eliminated by increasing the gas velocity in the coal feed line so as to minimize the opportunity for freshly fed coal particles to stick together before they are dispersed in the fluidized bed.

During the early phases of PDU operations, agglomerates would shut the unit down in a matter of hours or days. Once the problem was understood and proper precautions were taken, runs of 20 to 30 days could be made without any buildup of agglomerates. More than 5000 total hours of gasifier operation have been logged in the last 2 years.

Fluidized Bed Density

After the problem of agglomerate formation was solved and long continuous gasification runs were achieved, it became apparent that the gasifier had a lower than anticipated fluidized bed density. During the initial PDU operations, fluidized bed densities of 75 to 100 kg/m^3 (5 to 7 lb/ft^3) were obtained. Analysis of the bed contents showed the particle size distribution to be much smaller than observed in smaller pilot plant operations. It was theorized that the Illinois No. 6 coal being fed to the PDU was becoming plastic when heated in the gasifier, evolving gases upon devolatilization while still plastic, puffing up like popcorn during the gas evolution, and then breaking down to finer particles by attrition in the gasifier. This mechanism is illustrated in Fig. 7. This problem was considered to be significant because if unsolved it would result in very large commercial gasifiers, excessive fines losses from the bed, and more difficult separation of char from the recycled catalyst solution.

The first step in solving this problem was to review the experience from earlier small pilot-scale experiments. During the predevelopment phase, a small pilot unit called the fluidized bed gasifier (FBG) was operated. This unit had a coal capacity of 100 kg/day, was limited to an operating pressure of 0.8 MPa, and obtained fluidized bed densities of 150 to 300 kg/m^3. Analysis of FBG bed densities showed that during early runs the density was 300 kg/m^3 and that over a year of operation it gradually decreased to 150 kg/m^3. During the same period, the catalyst addition system had been changed in stages, in both design and operation, to exclude air from the system. These observations seemed to correlate with experience in other coal gasification development programs where oxidative pretreatment was used to permit the feeding of caking coals to fluid bed gasifiers. That experience suggested that the presence of the catalyst might permit oxidation at very mild conditions and at low severity. This led to the testing of oxidative pretreatment as a means of solving the bed density problem.

Testing of this proposed solution was implemented by parallel bench- and pilot-scale programs. A laboratory bench technique was developed for rapidly heating a small amount of coal under pressure to simulate experience in the PDU. A swelling index was defined by this test as the ratio of the final volume of the char after heating to the initial volume of the coal. Figure 8 shows the results obtained with a small laboratory fluidized bed oxidizer with a residence time of 6 hours and 6 percent oxygen in nitrogen. The data indicate that there is a narrow temperature range around 200°C that is characterized by a low swelling index. Other experiments showed that the optimum temperature varies with oxygen concentration. Detailed analysis (*13*) showed that the chemical change that occurred during oxidation was the addition of carboxylic acid groups to the coal structure. The number of such functional groups was greatest for samples oxidized at the optimum temperature.

In parallel with this bench-scale effort, an off-line fluidized bed dryer was used to oxidize catalyst-impregnated coal in batch fashion for testing in the PDU. Tests of this coal showed that oxidation had the potential to produce significantly higher bed densities in the PDU.

On the basis of the bench-scale research and batch pilot tests, a continu-

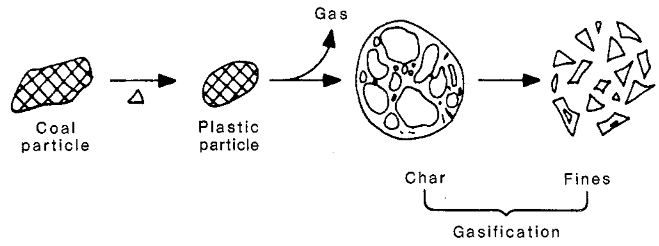
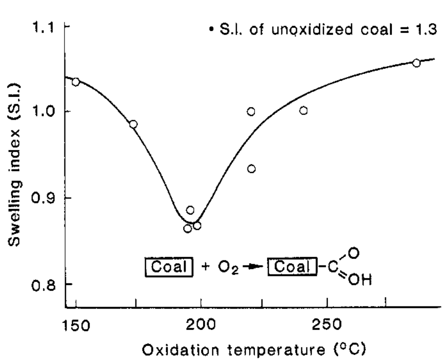


Fig. 7. Schematic of proposed mechanism for coal swelling. It is postulated that coal particles puff up like popcorn as they go through the plastic phase due to internal release of volatile gases. Both the resulting char and the fines from char breakage have lower densities than the input coal.



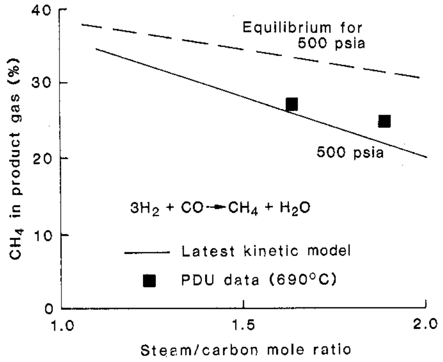


Fig. 8 (left). Swelling index as a function of coal preoxidation temperature from a bench-scale experiment with 6 percent O_2 in N_2 and a 6-hour residence time. Fig. 9 (right). Amount of methane in the product gas as a function of the steam/carbon ratio at 3.5 MPa (500 pounds per square inch absolute).

ous flow, fluidized bed preoxidizer was installed on the PDU. Process variables were studied at different preoxidation temperatures and coal residence times. At residence times of 4 to 11 hours and temperatures of 175° to 200°C, gasifier fluidized bed densities between 200 and 450 kg/m^3 were achieved. Engineering studies showed that a fluidized bed dryer can serve the dual function of drying and preoxidizing the feed coal with only a moderate effect (~ 5 percent) on the cost of the gas.

Methanation Rate

Another problem identified by the early PDU operations was that of lower than anticipated methane production in the gasifier. This is important because a key function of potassium in the CCG process is catalysis of the reaction of carbon monoxide and hydrogen to form methane (Eq. 2).

The kinetic model described previously was used to predict the methanation activity of the catalyst in a commercial fluidized bed gasifier operating at 3.5 MPa. This resulted in the prediction that the product gas leaving the gasifier would be very close in composition to that expected at methanation equilibrium, that is, about 31 mole percent methane in the dry product gas. The methanation activity of the catalyst determines the number of moles of carbon monoxide and hydrogen that must be recycled to the gasifier per mole of methane produced. This ratio was initially predicted to be 1.5 for the commercial gasifier.

Initial PDU operations yielded a gasifier output stream containing only 27 mole percent methane in the product gas. If left unchanged, this lower methanation rate would require larger gas separation equipment to handle the higher gas flows. Further, larger gasifiers would be required because of the extra carbon monoxide and hydrogen recycled to the gasifier and their inhibiting effect on carbon gasification.

The gasifier model was updated to reflect the performance demonstrated in the PDU and to identify avenues for raising the methane content of the product gas. The results of this analysis are shown in Fig. 9, which is a graph of mole percent methane in the product gas versus the ratio of steam and carbon fed to the gasifier. Figure 9 shows equilibrium predictions and two actual data points for PDU operation. As the steam/carbon ratio is lowered, the methane content of the product gas increases. A second approach to achieving a higher methane content would be to use a larger gasifier to increase residence time and allow a closer approach to methanation equilibrium.

Why methane equilibrium is not achieved in the PDU is under investigation. Future PDU runs are planned to verify the kinetic model predictions for the methane concentration at lower steam/coal ratios. Also, economic studies are under way to define the optimum methane level, considering the economic trade-offs involved with steam rate, reactor size, and recycle gas rate.

Gasification Demonstration Run

In the course of working on the problem areas outlined above, 27 PDU run periods were completed, the longest being 33 days. During these periods, numerous process variable studies were conducted. There were 53 material balance periods (*14*), each representing 24 hours of stable conditions, covering gasifier operating pressures from 0.7 to 3.5 MPa, gasifier operating temperatures from 650° to 700°C, carbon conversions from 35 to 97 percent, steam conversions from 20 to 45 percent, and fluidized bed densities from 80 to 480 kg/m^3. These material balance periods provided the basis for setting the conditions for a gasifier demonstration run, needed for generating data to set the design basis for the CCG large pilot plant. To this end, it was necessary to operate the gasifier at target commercial conditions at a steady state for a sustained period of time.

After a number of minor problems, including plugging of the char withdrawal line and valve failures, a successful 23-day PDU gasifier demonstration run was conducted during March and April 1981. Operations were generally stable and steady, and 14 material balance periods were obtained. In general, the coal and steam rates were maintained near the target values and under good control. The gasification pressure was 3.5 MPa and the temperature was controlled at about 690°C. The fluidized bed density was about 225 kg/m^3, steam conversion was 35 percent, and carbon conversion was 85 to 90 percent. Methane content of the product gas was generally 20 to 25 percent, somewhat lower than the target value of 25 percent, but this level was consistent with the high steam/coal ratio used in this run. Material balance closure was generally ± 5 percent, and no plugging of the gasifier was observed. The run was thus considered a major success.

Further analyses and correlations of the demonstration run data are being made, but it is clear that an adequate data base now exists for the design of the gasification section of a large pilot plant. The recent demonstration run was a major milestone in the CCG development program because it provided confidence in our ability to achieve commercial operating conditions in the PDU.

Concluding Remarks

Research and development on catalytic coal gasification for methane production is proceeding well. The operation of a process development unit at 900 kg/day has demonstrated operability and identified a number of problems for which solutions have been found. An essential aspect of CCG development is the close coupling between basic, bench-scale, and PDU research and engineering studies. The next major step is the design, construction, and operation of a large pilot plant (90,000 kg/day) in the middle 1980's. Success in future research and development could bring CCG to commercial readiness in the later 1980's.

References and Notes

1. *National Gas Survey* (Federal Power Commission, Washington, D. C., 1975), vol. 1.
2. Gasification of coal produces methane as well as other hydrocarbon gases, pyrolysis liquids, hydrogen, carbon monoxide, carbon dioxide, oxygenated hydrocarbons, and so forth. Relative yields are dependent on a variety of gasifier parameters.
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13. R. Liotta, personal communication.
14. Material balance periods in our program are 24-hour intervals of stable operation during which feed and product stream data are collected. The information is later reconciled to mass balance the reactants versus the products.
15. We acknowledge and greatly appreciate the efforts of the large group of scientists, engineers, technicians, and support personnel who have contributed to the success of the CCG program. We particularly thank C. A. Euker and N. C. Nahas, who headed the CCG Process Development Section at Baytown, Texas, during the formative periods of the program. While Exxon funding was used to initiate CCG research and to construct the PDU, funding from the Department of Energy and the Gas Research Institute carried the program through the predevelopment and early development phases. Their support and encouragement are gratefully acknowledged.

Regional Differences in the Growth of Normal and Neoplastic Cells

Robert Auerbach and Wanda Auerbach

In 1936, when J. M. Twort and C. C. Twort painted mice with a synthetic tar dissolved in chloroform, they discovered that the onset of visible tumors was markedly delayed in those mice treated with the carcinogen more posteriorly in the sacral region compared to those treated anteriorly in the scapular region (*1*). This was not a casual observation, because these investigators in the course of their many studies reported on some 15,000 tumors induced by a wide variety of carcinogens administered to over 100,000 mice. Even the particular experiments cited, on the effects of synthetic tar, involved several hundred tumors, and the records included tumor onset, total incidence over time, and the extent of malignancy. The study was convincing, the results were striking, and there was no doubt that the carcinogen was more effective in the anterior than in the posterior site. Yet the work attracted little attention and was seemingly forgotten.

In 1973 Vaage published a description of techniques used in tumor transplantation (*2*). He mentioned that there are significant differences in the growth of tumor cells inoculated anteriorly compared to those injected posteriorly under otherwise identical conditions. He recognized the importance of precision in the development of injection protocols, and, like Twort and Twort, made a strong plea for consistency in the application of experimental procedures to cancer research. But the discussion of anteroposterior differences was brief and his comments were not generally noticed.

In 1975 Kobayashi (*3*), working at a marine laboratory in Japan, compared the response of the mouse to experimental wounding of the dorsal skin at various levels of the trunk. He observed that there was a higher mitotic index in the epidermis surrounding wounds made anteriorly than around wounds made more posteriorly. But the report was an isolated one, not placed in the perspective of tumor growth, and received little subsequent discussion.

Our own studies came about because of seemingly capricious results obtained in the course of investigating the response of transplanted tumor cells to immune regulation (*4–6*). As we puzzled over the data we eventually discovered that regional differences due to location of the tumor inoculum—even a few millimeters distance within the trunk skin—were of such major importance that they were obscuring any possible differential between control and experimental groups. In trying to gain understanding of this phenomenon we searched the scientific literature. We found here and there anecdotal or modestly documented observations, often incidental to the work being presented. Occasionally we found, sometimes by serendipity alone, substantive works such as those cited above. These works, however, offered no links to the earlier literature and were therefore difficult to place in any historical context. We expanded both our literature search and our own experiments to include normal tissue transplants, drug efficacy, and a variety of physiological parameters ranging from blood flow to the aging process. An underlying pattern of regional influences began to appear.

Even if the various studies of anteroposterior differences in carcinogenesis,

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