

PRODUCTION OF LOW BTU GAS
INVOLVING COAL PYROLYSIS AND GASIFICATION

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INTRODUCTION

Coal burning is a "dirty" process, with the major pollution culprits now considered as being solid flyash particulates and sulfur dioxide. More than likely, nitrogen oxides and other undesirables will come under attack after the SO₂ and flyash problems have been satisfactorily controlled. (Jameson, 1972) Because coal is currently the most plentiful source of energy, the combined efforts of industries and governmental agencies are urgently needed to develop economical, efficient and acceptable methods to convert coal into clean electrical power.

In this paper, we are presenting a scheme to pyrolyze caking coal in a fluidized bed, some experimental data, a pyrolysis-gasification reaction model and a conceptual flowscheme for a process to convert coal into electricity via the production of low-Btu gas. Although the experimental data presented here are not comprehensive, we hope to explore some of the alternatives involved in such a manner as to recognize the most efficient ways to maximize the coal-utilization efficiency.

EXPERIMENTAL

Equipment

An experimental 15-inch diameter fluidized bed was used at West Virginia University for the study of the pyrolysis of coal and other carbonaceous compounds. The schematic of the pilot-plant fluidized bed reactor and its auxiliary equipment is shown in Figure 1.

There are three sections to the fluidized bed reactor. The hot bottom is a mixing and gas combustion chamber where the fluidizing gases are preheated by burning natural gas (over 90% methane) inside the L-shaped chamber. The fluidizing gases are composed of specific ratios of the combustion products of the natural gas and additional air.

Between the gas combustion chamber bottom and the reactor bed section is the high-temperature grid plate. This gas distribution plate is 1/4 inch thick, 18 inches in diameter and made of Type 310 stainless steel. There are 584 holes in a square pattern located on 1/4 inch centers. Each hole is 0.0960 inches in diameter.

In the middle of the reactor is the fluidized bed. This reactor section has a 28-inch outside diameter and a 15-inch inside diameter. The insulation lining is 4 1/2 inches of Type 1620-K fire brick (Babcock and Wilcox Company, Augusta, Georgia) and 2 inches of "Plicast Tuff-Mix" castable refractory (Plicast Company, Chicago, Illinois). The height of this section is eight feet. Above the fluidized bed section, the reactor diameter expands to a 35-inch outside diameter and a 22-inch inside diameter. This particle disengaging chamber is four feet high and is lined in the same manner as in the fluidized bed section.

After leaving the reactor, the effluent gases are cooled and then cleaned by passage through either a canister-type nylon-bag filter or a dry-gas cyclone (10 inches diameter and 22 inches in length). Before being finally exhausted to the building exterior, the effluent gases are scrubbed in a series of two wet scrubbers, the first being a tray-type and the second scrubber being packed with one-inch "Intalox Saddles" (U. S. Stoneware, Akron, Ohio).

The solids are fed into the fluidized bed by means of a screw conveyor having a specially designed feeder valve. The feed location is five inches above the gas distribution plate through a 6-inch flanged port in the wall of the reactor. The 2 1/2 inch screw is constructed of carbon steel with a Type 310 stainless steel coating on the wearing surfaces. The construction details of this feeder and the auxiliary solid feed hopper system have been described by Burton and Bailie (1973). The feeding unit has performed successfully in feeding 15 to 60 pounds of solids per hour smoothly into the fluidized bed.

The gases leaving the fluid-bed reactor are sampled every five minutes and analyzed by a Bendix "Chroma-Matic" Model 618 Process Gas Chromatograph (Process Instruments Division, The Bendix Corporation, Ronceverte, West Virginia). This unit quantitatively analyzes the gas for H₂, CO₂, CO, CH₄ and O₂/Argon. The O₂/Argon value is the additive sum of the oxygen and the argon compositions, since the gas chromatograph peaks of both components are identical when using a molecular sieve column. Periodically, grab-samples of the effluent gases were withdrawn and analyzed on a Beckman GC-2A Gas Chromatograph and a Fisher Scientific Company Gas Chromatograph for the gas components listed above, plus acetylene, ethylene, ethane and nitrogen.

Operating Procedure

The reactor is filled with 0.025 inch diameter sand to a collapsed bed height of 30 inches. The gas velocity through the bed is maintained at a level where a good fluidization of the sand is assured, and then the bed is heated up to the pre-selected temperature (1840°F) by the combustion of methane in the bottom section of the reactor. The operating conditions in the reactor are summarized in Table 1.

TABLE 1
OPERATING CONDITIONS FOR
FLUIDIZED BED PYROLYSIS REACTOR

OPERATING TEMPERATURE	1400 - 1900°F
OPERATING PRESSURE	0 - 10 psig
COLLAPSED BED HEIGHT	2.5 feet
EXPANDED BED HEIGHT	3.5 - 4 feet
AVERAGE PARTICLE SIZE OF SAND	0.025 inches
DENSITY OF SOLID SAND PARTICLE	100 pounds/cubic feet
SUPERFICIAL FLUIDIZING GAS VELOCITY	1.5 feet/second

After the bed reaches the desired temperature, the rate of air to the methane burner and the rate of inert gas flow are adjusted to give the oxygen level and gas flow rate specified in the experimental plan. The reactor system is then allowed to come to steady-state conditions as judged by a leveling of the effluent gas composition as read on the continuously operating process gas chromatograph and by constant bed temperatures.

The test begins by slowly introducing the solid feed into the bed via the screw feeder. The bed temperature immediately drops because of the sensible heat required to heat the solid to the reaction temperature plus the heat of pyrolysis. The solid feed rate is carefully adjusted so that the bed temperature does not drop below the desired 1400-1500°F range. The reaction system is then allowed to come to a new steady-state condition with a constant solids feed rate, and the feed rate of the solids is then determined by weight difference.

Experimental Data

The results of the coal and sawdust pyrolysis experiments are reported here. The composition of these two solid feed materials are listed in Table 2. Four tests were made with sawdust and one test using coal. In addition, several types of carbonaceous solids were pyrolyzed in the fluidized bed, including municipal solid waste, chicken and cow manure, rubber, plastic, and sewage sludge.

TABLE 2

<u>COMPOSITION OF SOLID FEED MATERIAL</u>	<u>SAWDUST</u>	<u>BITUMINOUS COAL</u>
MOISTURE (Weight %, Wet Basis)	2.62	3.42
ULTIMATE ANALYSIS (Weight %, Dry Basis)		
CARBON	47.20	73.3
HYDROGEN	6.49	5.34
OXYGEN	45.34	10.23
SULFUR	-	2.80
NITROGEN	-	0.70
ASH	0.97	7.57
HEATING VALUE (Btu/Dry Pound)	8114	13,097
PARTICLE SIZE (Harmonic Mean Diameter) in microns	603	504

As described previously in this paper, in the course of the reaction test there were two periods of steady-state reactor operation, the first period just before the solid is introduced into the fluid bed, and the second period happening during the constant-rate solids feeding. In the case of all five tests, the reactor operation just prior to feeding the solids were identical, as listed in Table 3.

TABLE 3

OPERATING CONDITION VALUES
DURING PYROLYSIS EXPERIMENTS

INLET FLOW RATE OF AIR: 35.26 SCFM
 INLET FLOW RATE OF NATURAL GAS: 3.40 SCFM
 REACTOR TEMPERATURE PRIOR TO FEEDING SOLIDS: 1840°F
 DRY COMPOSITION OF GAS EXITING REACTOR BEFORE SOLIDS FEED IS STARTED
 H₂ - 0.1123% CO₂ - 10.1674%
 O₂/Argon - 1.1808% CH₄ - 0.0726%
 remainder is assumed to be N₂

STEADY-STATE CONDITIONS DURING SOLIDS PYROLYSIS

	COAL <u>TEST</u>	<u>A</u>	SAWDUST <u>B</u>	TESTS <u>C</u>	<u>D</u>
Operating Time Under Steady-State Conditions (Minutes)	155	86	75	70	577
Reactor Temperature, °F	1430	1430	1460	1450	1500
Solids Feed Rate (Dry Pounds/Minute)	0.336	0.368	0.122	0.682	0.342

After the solids were fed into the reactor and after the system again reached steady-state conditions, the effluent gas was analyzed, with the resulting effluent gas composition values for each of the five experiments given in Table 4.

TABLE 4

PERCENT COMPOSITION OF EFFLUENT GAS (DRY)
DURING PYROLYSIS EXPERIMENTS

MEASURED BY PROCESS GAS CHROMATOGRAPH	COAL	SAWDUST		TESTS	
	<u>TEST</u>	<u>A</u>	<u>B</u>	<u>C</u>	<u>D</u>
H ₂	4.954	4.577	2.498	6.030	5.208
CO ₂	11.288	12.180	12.108	12.237	11.468
O ₂ /Argon	0.885	0.810	1.070	0.825	0.934
CH ₄	1.790	2.235	0.318	3.305	1.850
CO	2.244	7.543	2.206	11.498	7.565

MEASURED BY RESEARCH
GAS CHROMATOGRAPH

C ₂ H ₂	0.215	0.532	0.068	0.960	0.557
C ₂ H ₄	NM	NM	NM	0.073	0.047
C ₂ H ₆	0.113	0.113	0.035	0.159	0.055
N ₂	73.451	73.451	80.783	66.745	73.527

NM - Not Measured

The composition values of CO₂, O₂/Argon, CO, CH₄ and H₂ were averaged from the analysis readings of the process gas chromatograph, and the composition values of C₂H₂, C₂H₄, C₂H₆ and N₂ were averaged from the analyses by the research gas chromatographs of several grab samples taken during the duration of the test.

Using these experimentally measured gas analysis values, a mass balance was computed about the reactor system using the nitrogen flow rate as the calculation base. The mass balances were quite good considering the 2-5% accuracy of the flow-measuring meters and analytical instruments. The gas produced from the coal or sawdust pyrolysis is considered to be the net gas flow rate value, after subtracting the volumetric flowrate of the effluent gases prior to feeding the solids from the flowrates of the gases leaving the reactor during the solids pyrolysis reaction. These computed "Pyrolysis Gas Production Values" for the five experimental runs are listed in Table 5.

TABLE 5

COMPUTED PYROLYSIS GAS
COMPOSITIONS AND PRODUCTION RATES

PYROLYSIS GAS COMPOSITION (Volume % dry)	COAL	SAWDUST		TESTS	
	<u>TEST</u>	<u>A</u>	<u>B</u>	<u>C</u>	<u>D</u>
H ₂	46.876	25.645	37.493	23.592	30.022
CO ₂	11.685	14.965	24.256	14.075	11.105
CH ₄	16.630	12.427	3.724	11.880	10.483
CO	21.722	43.264	33.824	45.714	44.513
C ₂ H ₂	2.081	3.050	1.043	3.817	3.277
C ₂ H ₄	NM	NM	NM	0.290	0.277
C ₂ H ₆	1.007	0.648	0.537	0.632	0.324
 PRODUCTION RATE (SCF/Pound Dry feed)	 10.92	 18.25	 18.20	 15.95	 18.62
 GAS HEATING VALUE (Btu/SCF)	 435	 398	 286	 412	 399

NM - Not Measured

The results of these experiments indicate that coal can be pyrolyzed to yield 10.92 SCF of a 435 Btu/SCF gas per pound of dry feed. Similarly, one pound of dry sawdust can be pyrolyzed into 18.29 SCF of a 398 Btu/SCF pyrolysis gas.

ASPECTS OF A PYROLYSIS REACTION MODEL

Generalized Criteria for a Coal Pyrolysis Model

When a coal particle is pyrolyzed, the following products are generally found: gases such as CO, H₂, CH₄, C₂H₂, C₂H₆, CO₂, etc., condensable liquid hydrocarbons such as benzene, toluene, etc., aqueous compounds and solid char. When designing a coal conversion plant, one may design the reactor system to maximize the production of the gaseous hydrocarbon, the liquid hydrocarbon, or the char products. The slot-type coke oven is deliberately designed to maximize the char production by allowing the volatile gases to evolve slowly from the solid phase without exterior gas purging, thereby prolonging the gas-solid contact time.

Upon heating coal becomes softened and forms a metaplastic with simultaneous devolatilizations of vapor pushing surface bitumen from interior of particle. If heating rate is rapid, this phenomena is so

violent that the particle literally bursts and develops a solid with a large surface area solid mass. If the heating rate is slow, the products during pyrolysis tend to repolymerize into large, more thermally stable molecules of solid matter that are retained in the interstices of the residual char particle. At high temperatures, the products of pyrolysis are lower in molecular weight than those produced at lower temperatures.

The maximization of the condensible hydrocarbon production is reached when the evolved volatile product is quenched or cooled rapidly after leaving the solid phase, allowing a minimum of time for the larger molecules to thermally decompose into the lower molecular weight gases. In converse, the synthesis gas production is maximized if the volatile hydrocarbon products are held at a high temperature for a prolonged period of time. This exposure to high temperatures will crack the tars and other condensible molecules to lower chain aliphatics - CH_4 , C_2H_6 , C_3H_8 , etc. The pyrolysis reaction mechanism has been discussed by a number of investigators. (Burton and Bailie (1972), Peters and Berthing, (1964), Kertamus and Hill (1964), Jones (1964), Kirov and Stephens (1967), and Squires (1972). Squires cites experimental data reported by Schroeder (1962) in which coal, catalyzed with 1% molybdenum and in a hydrogen atmosphere at 800°C yielded a 42.2% liquid hydrocarbon fraction after a 5 second gas residence time, a 23% liquid fraction yield after a gas residence time of 10 seconds, and, after a 25 second residence time of the gas, the liquid fraction yield was only 9.9%.

Although the liquid fraction was not collected in the experiments while feeding coal or sawdust, a liquid fraction and a char fraction was collected while pyrolyzing a municipal solid waste mixture. The liquid fraction was analyzed as representing 7.0% and the char fraction was 13.5% (moisture and ash-free weight basis) of the inlet solid feed. This contrasts with the data reported by Sanner, et al. (1970), who destructively distilled a municipal refuse in a retort, constructed to simulate a coke oven process. They found that at 900°C , the liquid fraction from the refuse was about 47% and the char fraction was close to 9%. The equipment used by Sanner, et al., allowed the effluent gases to be cooled immediately after leaving the retort, while the exiting gases were held for more than 8-10 seconds at close to the reaction temperature after leaving the pyrolysis zone of the fluid bed reactor, thus accounting for the condensible hydrocarbon fraction from the fluid bed being a 40% smaller value.

In Figure 2, the final conversion of coal attainable as a function of overall heating rate is shown. This figure indicates that rapid heating of the coal avoids the polymerization reaction which can turn the coal to stable char before volatile matter is evolved. Since the objective is to "cream off" the coal to obtain as high heating value gas as possible, a temperature of approximately $1400\text{--}1500^\circ\text{F}$ was selected for experimentation.

Mathematical Model for Coal Pyrolysis Reaction

A mathematical model is formulated here based on an assumption that the weight loss during pyrolysis reaction is closely related to heating rate of the coal particle, while the products distribution is primarily determined by the vapor residence time.

A heat balance on a pyrolyzing coal particle may be written as:

$$k_s \left(\frac{\partial^2 T}{\partial r^2} + \frac{2}{r} \frac{\partial T}{\partial r} \right) - \frac{3}{R} (\text{rate}) (\Delta H) = \rho_s C_{ps} \frac{\partial T}{\partial t} \quad (1)$$

$$\text{where rate} = \frac{\rho_s R}{3} \frac{dX}{dt} = k(f - X)$$

The rate of pyrolysis is assumed to be proportional to the amount of unconverted portion of coal which will eventually be distilled off at the given condition. Thus, f , the final conversion attainable is a constant which depends on reactor temperature and type of coal employed, as shown in Figure 3. Equation (1) can be solved numerically based on the following boundary conditions:

$$\left. \begin{aligned} r = R, (\text{rate}) (\Delta H) + \frac{\rho_s C_{ps} R}{3} \cdot \frac{dT_s}{dt} &= h(T_b - T_s) \\ r = 0, \frac{\partial T}{\partial r} &= 0 \\ t = 0, T = T_0 \text{ and } X &= 0 \end{aligned} \right\} (2)$$

The heat transfer coefficient for the particle includes convective, conductive and radiative heat transfer. The heat of reaction for pyrolysis is normally rather small, roughly 300 Btu/pound coal. Thus, if the term associated with heat of pyrolysis is neglected, the solid temperature can be approximately related to time as:

$$1/2 \cdot \ln \frac{(T_b+T)(T_b-T_0)}{(T_b-T)(T_b+T_0)} + \tan^{-1} \frac{T_b(T-T_0)}{T_b^2+TT_0} = \frac{3h}{\rho_s C_{ps} R} t \quad (3)$$

When coal particles are blown into a hot fluidized bed, the heat transfer coefficient is so large (approximately 25 Btu/hr ft²°F) that the particle reaches bed temperature within a few seconds as shown in Figure 4.

The calculated temperature and time relation for fluidized bed pyrolysis at the bed temperature of 1450° ~ 1500°F is shown for bituminous coal of present study and Elkol coal reported by Jones, et al. (1964). The calculated conversion using a kinetic rate constant, $k = k_0 \exp(-E/RT)$, where $k_0 = 1.224 \times 10^3$ lb/(ft² hr) and $E = 2100$ Btu/lb-mole (4.6 Kcal/mole), is presented in Figure 5.

CONCEPTUAL FLOWSCHEMES OF LOW-BTU GAS FROM COAL AND SUBSEQUENT ELECTRICITY CONVERSION PROCESSES

A number of conceptual designs have already been proposed to convert a low-Btu gas and then coal into electrical power efficiently and cleanly. A modification of the Bituminous Coal Research's high-Btu "BI-GAS" Process two-stage gasifier has been proposed to utilize air instead of pure oxygen and to operate the gasifier at 300 psig. BCR concluded that an in-plant coal gasification process may compare favorably with other environmental control concepts (such as tail-end SO₂ removal), if the total coal-to-electricity process were to be re-designed into an optimal system, (Bituminous Coal Research (1971)).

In this section we intend to describe a conceptual process alternative based on the experimental data presented in the previous section, and to use this flowscheme to show that there will be a distinct advantage in considering a two-step coal gasification subsystem. In the first step, the coal is pyrolyzed to release the larger molecule hydrocarbons, such as methane, ethane, propane, etc., which Arthur Squires calls the "cream" of the decomposition products of the coal "molecule" (Squires (1972)). The second-step gasifier vessel reacts the residue pyrolysis char with steam and air to form the gas containing H₂, CO, CO₂ etc., that is needed to fluidize the pyrolyzer.

The two processes compared here are illustrated in Figures 6 and 7. In Figure 6, the "one-step" Coal Gasifier is illustrated, in which the raw coal is fed directly into the high-temperature (1900°F) synthesis gas generator operating at 150 psig. There, the coal is directly gasified with air and steam to produce a stream of H₂, CO, H₂O, CO₂, CH₄, H₂S and N₂. This product gas is cleaned of the H₂S and other impurities and is then burned in a combustion chamber. The effluent gases from the combustion chamber are then sent through a combined gas turbine - steam turbine cycle. The coal would have the same composition as was used in the present pyrolysis experiments (Table 2). In this model, the gasifier system was assumed to operate adiabatically, the gases - H₂, CO, CO₂, H₂O - are assumed to emerge from the reactor in the same composition ratio as the equilibrium composition of the water-gas-shift reaction, and the carbon-steam reaction products are at a 20% approach to thermodynamic equilibrium.

The "Two-Step Coal Pyrolysis-Gasifier" is illustrated in Figure 7. The raw coal is fed to the fluidized bed where the pyrolysis of the coal takes place at 1400°F. It is conservatively assumed that there are no chemical reactions between the fluidizing gases and the volatilized coal-pyrolysis product gases. It is also assumed that the product gases evolved from the coal pyrolysis reaction are produced at the same rate and in the same composition as was produced in the experimental bed described in the previous section. The char separated from the effluent gas is then reacted with air and steam to produce the fluidizing gases for

coal-pyrolyzer. A small amount of raw coal must be added to the char feed to the gasifier in order to maintain the 1900°F gasifier temperature and to produce enough gas to fluidize the incoming coal in the pyrolyzer. The gas produced in this two-stage gasification system is then purified to remove the sulfur and other undesirable compounds, and is then burned in the combustion chamber with the combustion gases processed to generate electricity in the same gas- and steam-turbine system as described previously.

In Table 6 effluent gas compositions from pyrolyzer and gasifier are listed respectively.

TABLE 6
COMPOSITIONS OF EFFLUENT GASES FROM PYROLYZER AND GASIFIER

	CO ₂	CO	H ₂	H ₂ O	CH ₄	C ₂ H ₂	C ₂ H ₆	H ₂ S	N ₂
GASIFIER	1.00	34.77	9.34	0.51	0.20	-	-	0.82	53.36
PYROLYZER	3.19	31.54	16.95	1.78	3.59	0.43	0.21	0.64	41.68

As can be seen in Figure 8, the "Two-Stage Pyrolyzer-Gasifier" System generates electricity with a 2% or better thermal efficiency than does the "Single-Step Coal Gasifier System". The thermal efficiency is defined as the heat equivalent of the product electricity generated divided by the heat of combustion of the inlet coal feed.

The electricity generating ability of the present day gas turbines are limited by the temperature of the inlet gases, the maximum allowable operating limit of around 2000°F governed by the thermal tolerance or the turbine construction metal. Figure 8 indicates the effect that a 200°F higher inlet gas temperature will make on the overall process efficiency.

A very important system design consideration is the degree of carbon utilization in the synthesis-gas generator. An increase of 5% carbon utilization in the gasifier implies an increase of approximately 2% overall plant thermal efficiency.

CONCLUSION

Experimental data of coal pyrolysis in a sand fluidized bed indicates that it is possible to extract considerable amounts of hydrocarbons from the caking coal by a rapid heating and subsequent cracking in the vapor

phase. This is done by blowing pulverized coal into a fluidized bed of hot sand and elutriating the char along with gaseous product from the fluidized bed. The extremely good heat transfer of the fluidized bed provides the rapid heating required for this process. In this manner, the valuable hydrogen in coal is extracted either as free hydrogen or as hydrocarbons in the gas phase product. A conceptual scheme is presented which utilizes the product char to generate synthesis gas by gasification with air and steam for use in the pyrolyzer. The thermal efficiency calculated based on the two stage process with the combined gas and steam turbine cycle shows that this scheme is a promising way to produce clean power from coal.

ACKNOWLEDGEMENTS

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NOTATION

C_{ps}	:	Heat capacity of solid	(Btu/lb)
E	:	Activation energy	(Btu/mole)
f	:	Final conversion attainable	-
h	:	Heat transfer coefficient, include convection and radiation	(Btu/ft ² -hr-°R)
$k(T)$:	Rate constant	(lb/ft ² -hr)
k_o	:	Frequency factor	(lb/ft ² -hr)
k_s	:	Thermal conductivity of particle	(Btu/ft-hr-°R)
R	:	Radius of particle	(ft)
\hat{R}	:	Gas constant	(Btu/mole-°R)
rate	:	Reaction rate	(lb/ft ² -hr)
r	:	Radial distance in particle	(cm)
T	:	Solid temperature at $r = r$	(°R)
T_o	:	Room temperature	(°R)
T_b	:	Bed Temperature	(°R)
T_s	:	Surface temperature of Solid particle	(°R)
t	:	Solid residence time	(hr)
X	:	Solid conversion, dry-ash-free basis	-
ρ_s	:	Solid density	(lb/ft ³)

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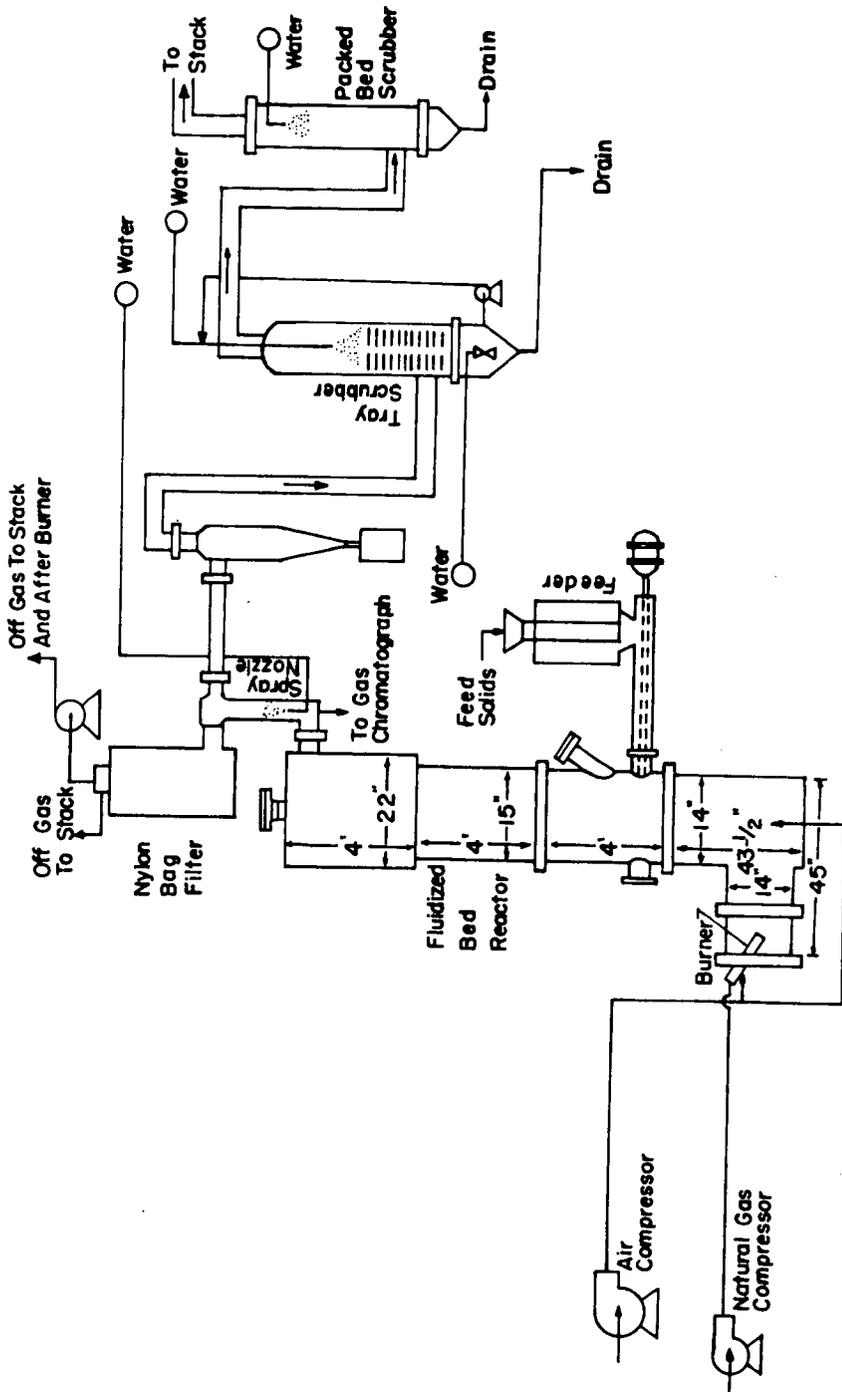


FIGURE 1 FLUIDIZED BED PYROLYSIS REACTOR SYSTEM

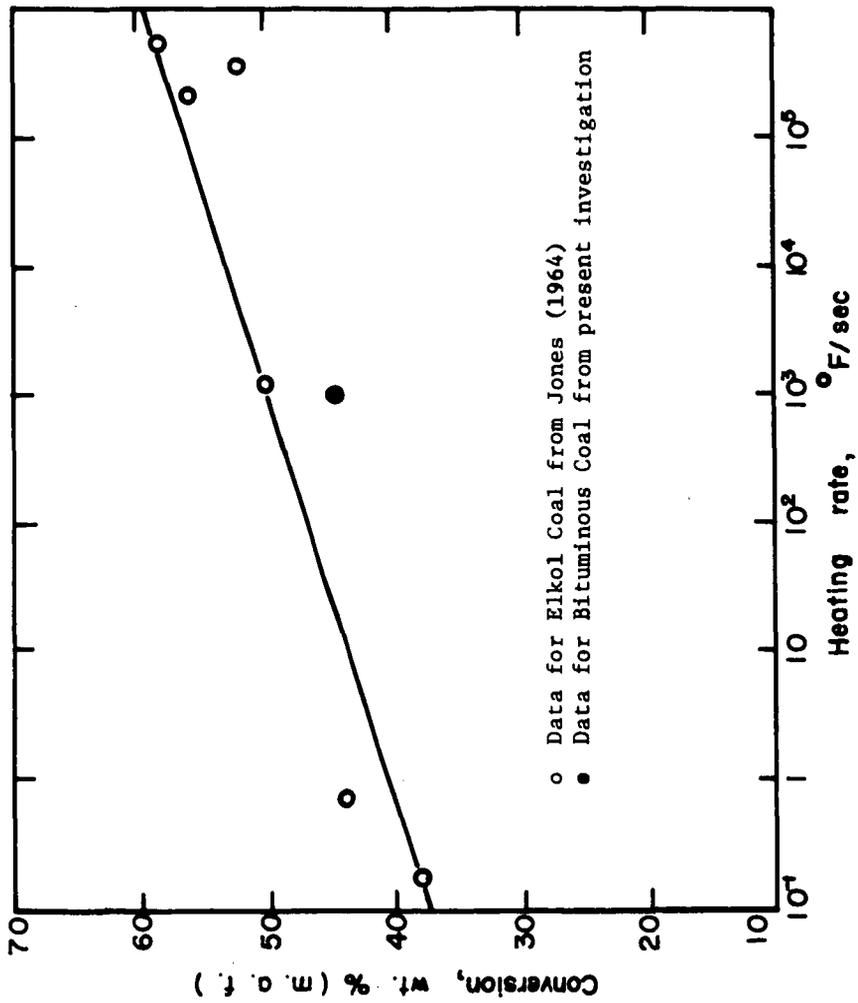


FIGURE 2

EFFECT OF OVERALL HEATING RATE ON DEVOLATILIZATION OF COAL

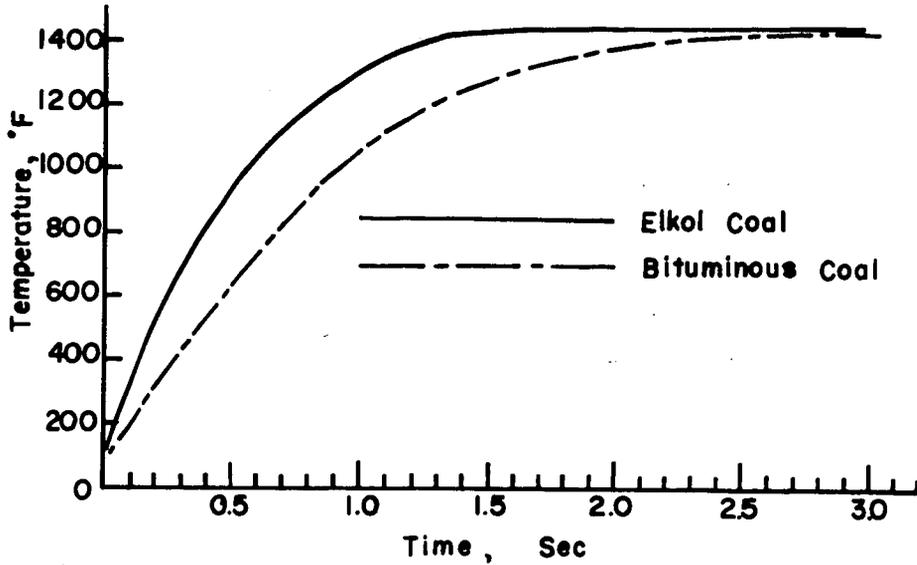


FIGURE 4

CALCULATED PARTICLE TEMPERATURE AS A FUNCTION OF TIME FOR PYROLYSIS REACTION IN FLUIDIZED BED SYSTEMS

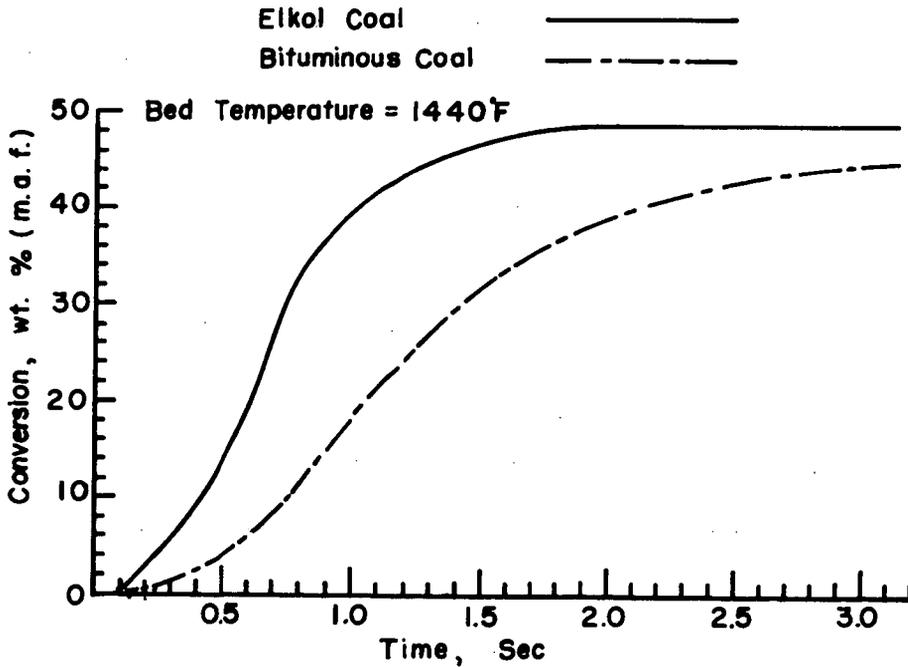


FIGURE 5

CALCULATED PARTICLE VOLATILE MATTER CONVERSION AS A FUNCTION OF TIME FOR PYROLYSIS REACTION IN FLUIDIZED BED SYSTEMS

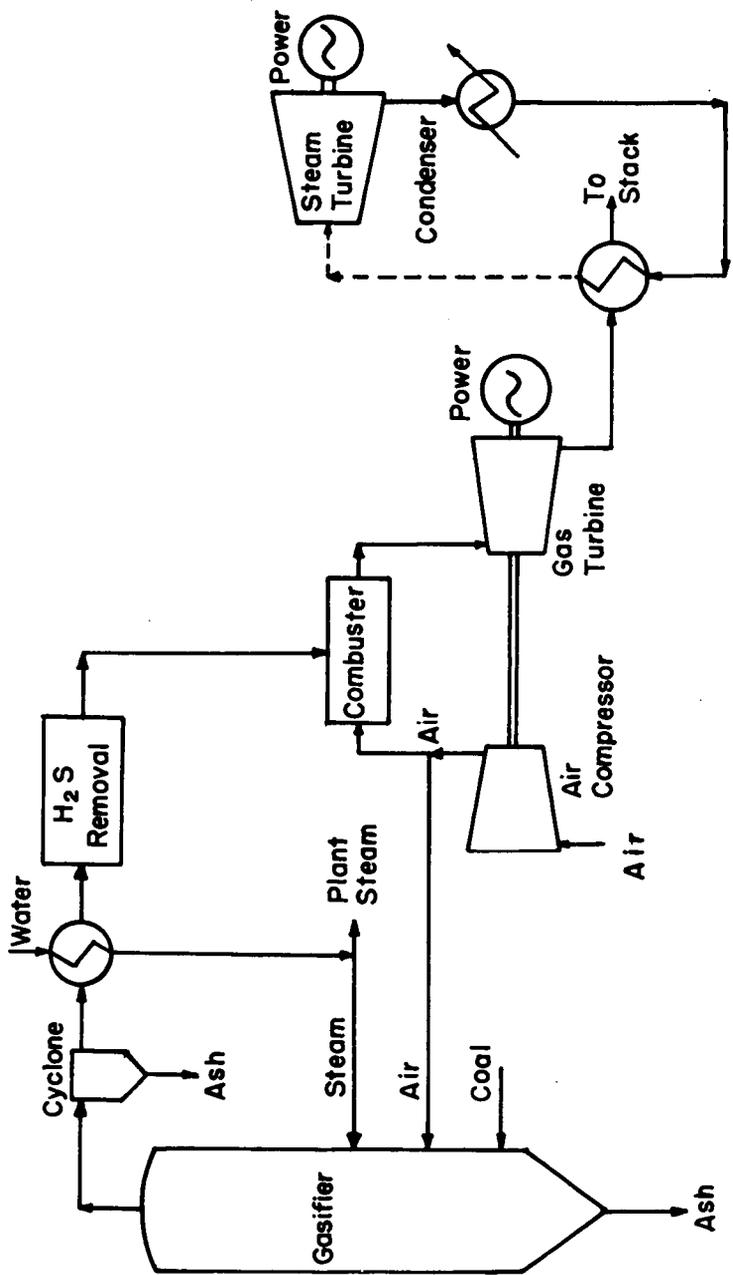


Figure 6 Power Generation Via Coal Gasification Combined Cycle
(One - Stage Coal Gasification System)

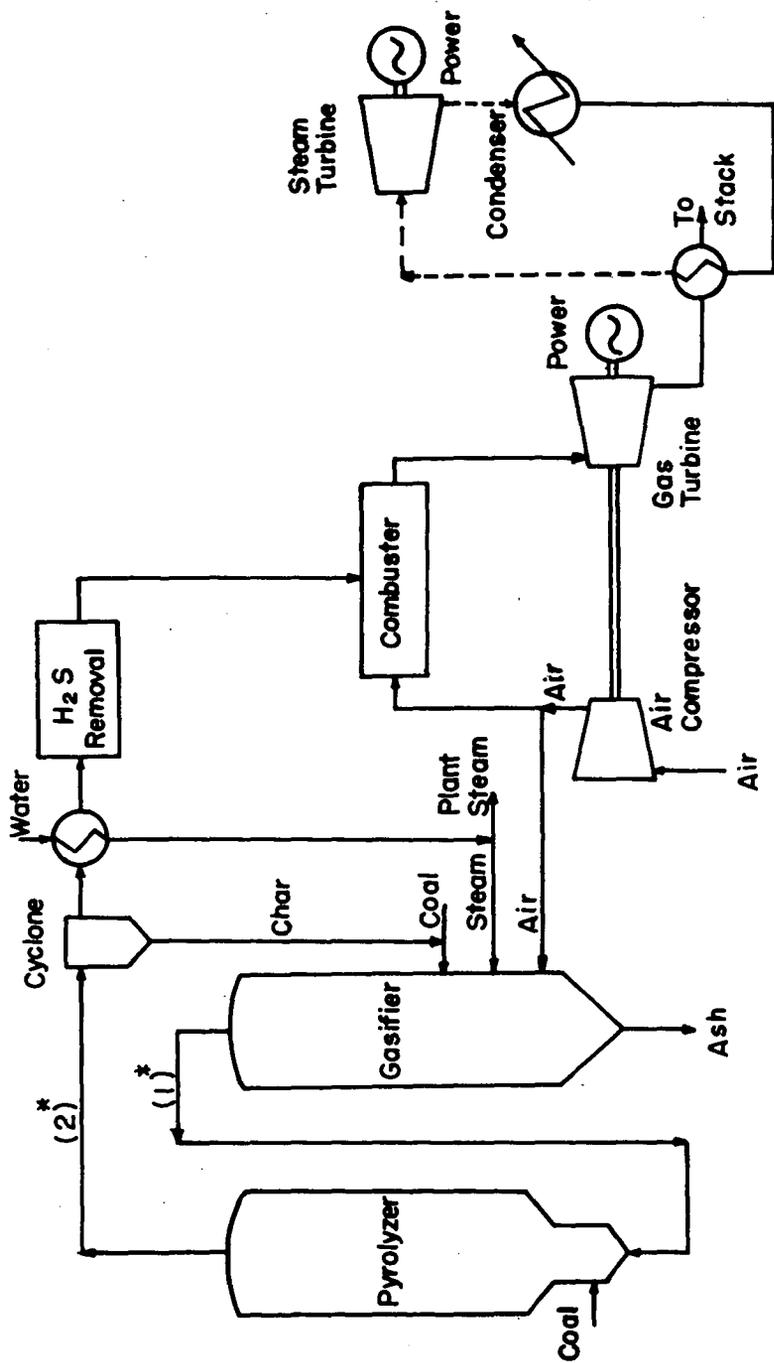


Figure 7 Power Generation Via Coal Gasification Combined Cycle
(Two-Stage Coal Pyrolysis Gasification System)

* Gas Compositions Are Given In Table 6 In The Text

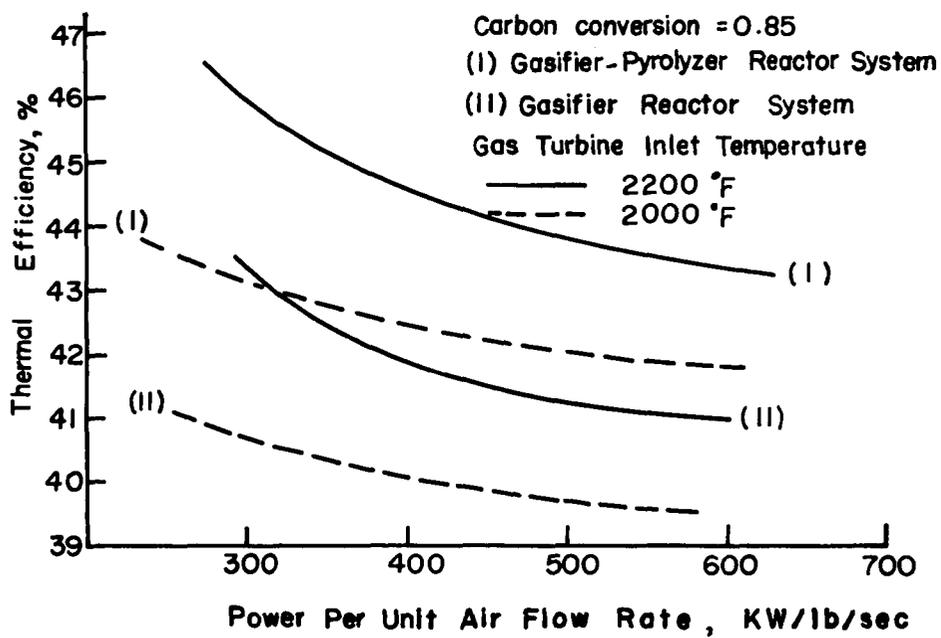


Figure 8 Thermal Efficiencies Of Two Processes Converting Coal To Electricity