

PROCEEDINGS OF THE 18th

IECEC '83

INTERSOCIETY ENERGY CONVERSION ENGINEERING CONFERENCE

VOLUME

2

OF 5 VOLUMES

FUEL UTILIZATION

18th
INTERSOCIETY
ENERGY CONVERSION
ENGINEERING
CONFERENCE

PROCEEDINGS
IN 5 VOLUMES

VOLUME 2
FUEL UTILIZATION

“Energy for the Marketplace”

SHERATON-TWIN TOWERS
ORLANDO, FLORIDA

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Intersociety Energy Conversion Engineering Conference

PURPOSE:

To reduce the overlapping and duplicative effort of the sponsoring societies in the field of advanced or non-conventional energy conversion. This conference is concerned with the engineering and application aspects of non-conventional energy conversion systems and devices as opposed to the details that are presented at various specialist conferences. Papers are screened for technical competence, clarity and brevity.

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A standing IECEC Steering Committee consisting of two members from each society coordinates all conference activities. Each year a society (this year AIChE) sponsors the conference. Session Organizers are appointed by the General Chairman and the Program Chairman. Session Organizers invite abstracts and receive abstracts from a general solicitation through the Program Chairman. Within limits set by the General and the Program Chairman the Session Organizers are responsible for the content of their sessions and appoint appropriate Session Chairmen.

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American Institute of Aeronautics and Astronautics
(1980 Conference, Seattle, WA)



American Society of Mechanical Engineers
(1981 Conference)



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(1982 Conference)

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Message from the General Chairman

Today throughout the world, there has been a de-emphasis in energy conversion projects simply to satisfy government requirements, and a realization that energy conversion must make sense in the marketplace. We as engineers, must consider how our projects are eventually going to be used, and how they will be eventually incorporated into products that people will buy and use.

For the last 18 years, the Intersociety Energy Conversion Engineering Conference (IECEC) has been a marketplace for ideas for potential products. Here, gathered together in one meeting, are the latest reports on technologies of diverse natures which will eventually compete for the marketplace. If your product is electricity for instance, you can evaluate the state of technology of several different types of heat engines, such as Stirling, Brayton Cycle, Rankine Cycle to operate different types of electric generators. You can see how photovoltaics, thermionics, and thermoelectric, MHD, and possibly other methods are doing in the direct conversion area. You can determine how wind energy, tidal energy, ocean thermal energy might fit into your needs. Possibly a new type of energy conversion will be discussed which you have never heard of before. As a marketplace for ideas, the Intersociety Energy Conversion Engineering Conference is ideal.

The utility of having one meeting for all use-related energy conversion technologies was recognized by the major engineering societies and related professional societies back in 1966. The result is the organization of the IECEC, to eliminate duplication of separately sponsored meetings. Each year, one of the sponsoring societies takes its turn to supervise and run this Conference. This year, the society is AIChE. The continuity of this Conference is provided by a Steering Committee composed of two representatives of each of the societies. The intent of the IECEC is to avoid subjects that are appropriate for established conferences on conventional power systems. It also intends to compliment all the specialist conferences that are meetings of experts in a limited area of energy conversion.

Orlando is an attractive vacation spot, and the meeting is scheduled before school starts. Therefore, we have not scheduled any late afternoon or evening activities at the conference, so that you may attend many of the internationally known attractions with your family or associates.

Since this meeting fulfills world-wide needs to learn how to do more with less by means of improved energy conversion, it has grown into an important international meeting. Particularly, we have important contributions from Europe and Japan in many technical areas. This year, the Japan Society of Mechanical Engineers is becoming one of the co-operating societies who help sponsor the IECEC. These co-operating societies do not take turns sponsoring one of the meetings, but do support the meetings with publicity to their members and do receive the privilege of registering at a reduced rate. However, whether you are a member of a sponsoring group or not, I am glad you were able to attend and partake of this meeting. If you are here as a specialist, we hope you will be interested to some degree in the whole field in energy conversion.

W. R. Martini
General Chairman

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Fossil Fuels

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P. L. Wilkinson, *American Gas Association, Arlington, VA*

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Oil Mining—An Old Power Technique—Now Economic

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Interfacial Phenomena in Foam Flooding Process for Heavy Oil Recovery

M. K. Sharma and D. O. Shah, *University of Florida, Gainesville, FL*

Effect of Staged Reactors on Yield Distribution for the SRC-I Process

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The process design for the SRC-I demonstration plant calls for two dissolvers operating in series. The objective of this study is to develop process data for operating the two dissolvers in the demonstration plant. Experiments were conducted using two different reactor configurations (series vs. parallel). In addition, the effect of varying the temperatures in the two reactors was also studied. The series operation of the dissolvers resulted in a decrease in the yield of preasphaltenes. Neither conversion nor the yields of oils, asphaltenes and gases are affected by reactor configuration. The staged temperature operation indicates that the temperatures in the two dissolvers in series can be optimized with respect to the yield of oils.

INTRODUCTION

International Coal Refining Company, a partnership of Air Products and Chemicals, Inc. and Wheelabrator-Frye Inc., under a cost-sharing agreement executed with the U.S. Department of Energy and the state of Kentucky, is in the process of designing a 6,000 tons per day coal feed SRC-I Demonstration Plant to be located in Newman, Kentucky. The Demonstration Plant Design will incorporate a first-stage liquefaction section (SRC-I) followed by an LC-Finer, referred to as Two-Stage Liquefaction (TSL). This paper is concerned with a specific design question on the first stage of the SRC-I Demonstration Plant. Details of the overall design can be found elsewhere (1,2).

In the first stage of the SRC-I demonstration plant, ground coal is mixed with a process derived solvent, and reacted at high temperature and pressure. A simplified flow scheme of the coal dissolution and reaction stages of the process is shown in Figure 1. At temperatures above approximately 300°C, coal begins to dissolve in the process derived recycle solvent. The first step is the absorption of

solvent into the coal and physical dissolution and solubilization of the coal macerals. This step occurs in the preheater wherein the residence time is small in comparison with the dissolver. Although a considerable amount of dissolution will occur in the preheater, a major amount of necessary chemical change will occur in the dissolver, namely sulfur removal, oil and distillate formation and solvent hydrogenation. The demonstration plant dissolver, which behaves like a bubble column with upward co-current flow of slurry and hydrogen gas, represents a considerable scale-up over the dissolvers at the Wilsonville and Ft. Lewis pilot plants which have been used as a design database for the SRC-I demonstration plant. The fluid dynamics in the dissolver is a function of the reactor size and the superficial slurry and gas velocities. Cold-flow studies have shown that the demonstration plant dissolver is completely backmixed with respect to the slurry at the proposed operating conditions (3).

Correlation of the pilot plant data established a basis for selecting the process parameters for the Demonstration Plant Dissolver (4). Reactor diameter was set by shipping considerations. Three options for reactor length were considered: a single vessel or either a series or parallel configuration of two half-size vessels. The smaller vessels were chosen to obtain the advantages of improved process flexibility.

The dissolvers can be operated either in series or in parallel. Series operation, which will be closer to plug flow than parallel operation, might offer the advantages of:

- o lower hydrocarbon gas yields
- o improved solvent quality
- o higher gas and liquid linear velocities
- o potentially lower sulfur in the products
- o staged temperature operation

Parallel operation, on the other hand, offers the advantages of:

- o more uniform temperature distribution across the reactor
- o higher solids accumulation in the reactor

Although series operation appears to be advantageous, the need to maintain an adequate solids level in the dissolver may be the major deciding factor. The work reported here was directed at determining whether series operation was superior in any way. Also, because the performance of dissolvers in series can be optimized by selecting the correct staged temperatures, another goal was to determine whether staged temperatures would impact demonstration plant operation.

Kinetic data on the SRC-I process had been developed earlier using a continuous stirred tank reactor (5). The yield data from a single CSTR was found to agree with data from the Wilsonville pilot plant. A model was developed using the kinetic data (5). Hence one can mathematically predict the yield distributions for two reactors in series based on data generated from a single CSTR. However, experimental data is not available in the open literature for two reactors in series. Hence, the objective of this program was to develop process data for operating the two dissolvers in the demonstration plant.

EXPERIMENTAL

To develop process data for operating the two dissolvers in the demonstration plant, experimental data were collected at conditions shown in Table 1. Comparisons were made in yield structure between series and parallel dissolver operations at three temperatures (415, 432, and 449°C). The same temperature was maintained in the two reactors operated in series at each level of temperature investigated. Parallel dissolver operation was represented by operating a single reactor at the same three temperature levels. Three additional conditions were examined each with a different set of temperatures in the two reactors in order to study the effect of staged temperatures on yield distribution. In Table 1, each operating condition is assigned a part number, which will be referred to in certain sections of this paper.

Materials: The same materials were used for all operating conditions. Washed Lafayette Kentucky #9 coal from a coal preparation plant was ground to 95% less than 0.074 mm size particles, air-dried, and then screened through a micron sieve before use. Detailed analysis of the screened coal sample is reported in Table 2. The feed solvent was a recycle solvent produced at the Ft. Lewis Pilot Plant during SRC-I operation. Analysis of the solvent is detailed in Table 3. When fractionated into individual fractions by solvent separation, the solvent contained 94.2 wt % oils and 4.8 wt % asphaltenes. The hydrogen content was 8.3%.

Operation: The experiments were conducted in a continuous 45 kg/day coal liquefaction unit

equipped with two 1000 cc continuous stirred autoclave reactors. The feed slurry (40 wt % Kentucky #9 coal, and 60 wt % Ft. Lewis solvent) was prepared immediately prior to use at room temperature, typically in 40-to-60-kg batches, and stored in a charge tank equipped with a turbine agitator and a recycle pump to maintain uniform suspension of the solids. Slurry handling was planned to minimize consumption of recycle solvent.

The feed slurry was pumped to the first continuous stirred tank reactor (CSTR) from a feed tank maintained at ambient temperature. The feed tank, also equipped with a recycle pump to mix the slurry, was refilled each hour from the charge tank. Whenever the feed slurry composition was changed, the feed tank was drained and then flushed several times with the new feed slurry. When reaction conditions were altered, at least 10 reactor volumes of slurry were processed through the reactor at the new conditions before any product samples were collected for analysis.

A continuous reactor was selected for these experiments so that the vapor composition in the reactor, including the partial pressure of light solvent components, would match that of an SRC-I dissolver. The stirred autoclave design of the CSTR ensures that the solids were mixed well and did not accumulate. Cold-flow experiments, conducted in a Plexiglas model, confirmed that the CSTR's flat-bladed turbine agitator kept the solids distributed throughout the reactor volume, so that they would not accumulate (6).

Because baffles tend to promote coking, they were not used in the CSTR. However, the presence of a thermowell reduced the size of the vortex with the void fraction in the reactor being about 0.18. Because all the sensible heat to bring the slurry to reaction temperature was provided by resistance heating of the reactor, the reactor wall was about 14°C hotter than the bulk slurry.

Before the reactor effluent passed to a gas/liquid separator, the product gas and slurry from the second CSTR was quenched at reactor pressure to 163°C to quench the reactions. The product slurry was throttled into the product receiver at a rate that maintained a constant, small inventory in the separator.

Water, light hydrocarbons, and gases, which⁵ flashed from the product slurry in the 3.8 X 10⁵ pascal receiver, are combined with the product gas stream from the separator. The combined gas stream was then cooled to about 33°C to condense the water and light organics, before the stream entered the gas chromatograph and dry test meter for product gas yields and volume measurement.

Feed and Product Analysis: Two sets of product samples were obtained at each operating condition to ensure that actual operating conditions

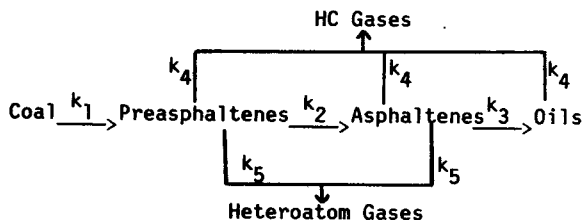
during sampling were close to planned conditions. One set of product samples was analyzed further.

The feed and product slurries were solvent-separated into four fractions: pentane-soluble material (oil), pentane-insoluble and benzene-soluble material (asphaltene), benzene-insoluble and pyridine-soluble material (preasphaltene), and pyridine-insoluble material. The latter fraction contains insoluble organic matter (IOM) and mineral residue. It is important to note that the fractions from solvent separation are defined operationally, not chemically. Thus, the end points of each fraction often vary, depending on the nature of individual samples as well as minute changes in procedural details. Product slurries were analyzed at least twice. Each fraction was further subjected to detailed elemental analysis. Product gases were analyzed by on-line gas chromatography up to C_4 hydrocarbon gases, including other nonhydrocarbon gases (H_2S , NH_3 , CO , CO_2 , etc.). The product water was determined indirectly by assuming that the water vapor exiting the process unit was in equilibrium with the condensed water.

Products from the process unit were sampled as gases, light condensate, and the main receiver product slurry. The light condensate was a mixture of water (about 80%) and oil. The quantity of oil was extremely small based on the total main receiver product.

Two of the organic condensate samples were subjected to elemental analysis, and the average of the two was used as the elemental analysis of the light oil condensate for each run.

Experimental Design: At the start of this program an attempt was made to predict the product yields that would result from two reactors operated in series using the following simplified version of the sequential kinetic model developed by Martin (5).



Individual rate constants in this model were determined using experimental data from various runs in which a Kentucky #9 (Pyro Mine) coal was processed. These rate constants were then used to predict product yields as a function of temperature for various numbers

of in-series reactors as well as a plugflow reactor. A constant space time of 2400 seconds was used in the calculations. In the case of multiple reactors, it was assumed that the total space time was 2400 seconds, with equal retention times in all reactors.

The predicted product yield structures, plotted in Figures 2-4, indicate that the oil yields do not differ significantly for various reactor configurations, but the asphaltene and preasphaltene yields depend on reactor configuration. It is important to point out that because differences in the asphaltene and preasphaltene yields between the single reactor and two in-series reactors are small, multiple analyses are required to distinguish these differences. Using the standard deviation calculated by Joshi (7) for analysis of asphaltenes and preasphaltenes, one can determine that four analyses will be necessary. Thus, in this program the product liquid samples collected at 840°F were analyzed at least four times and others were analyzed twice. Unit reproducibility was also checked.

Kentucky #9 coal (Lafayette) was selected for these studies because the demonstration plant planned to use Kentucky #9 coal. At the same time this program was underway other CPDU studies were simultaneously planned for this same coal.

Data were collected at three different temperature levels (415, 432 and 449°C) because the differences were expected to be small; multiple data points can detect a definite trend far more easily than a single data point.

Error Analysis: The experimental design called for multiple analyses of the product slurries. Results from these multiple analyses clearly indicated good data reproducibility, reflecting a marked improvement in our solvent separation technique. Because the differences in yield structure were small, determining the statistically permissible maximum differences between different analyses was extremely important. Consequently, an error analysis was conducted using the multiple analysis data generated during this program.

As part of this program five different samples were analyzed two or more times. By applying the pooled-variance statistical technique, one can use the data generated to determine standard deviations for each individual fraction measured by solvent separation (oil, asphaltene, preasphaltene, and residue). Tables 4-7 summarize the data gathered during this program for each fraction. The F-test for checking the homogeneity of the sum of square errors (SSE) was applied at a 95% confidence level to each of the data sets; nonhomogeneous samples are identified by an asterisk in Tables 4-7. Hence, the data are statistically homogeneous after the asterisked samples are omitted. Among all the data sets, only one set from Table 4 was omitted. The

estimate of the variance is shown at the bottom of each table.

Using the values of these variances, the statistically permissible maximum differences between two analyses of total product liquid were calculated and are shown in Table 8 for the duplicate and quadruplicate analyses of each sample. The statistically permissible maximum differences between two different yields based on maf coal basis (for experiments using 40 wt% KY #9 coal) is also shown in Table 8. Note that the values based on maf coal yields are magnified by about 3 times the values based on total product liquid. These results are used in the discussion below.

Analytical and Unit Reproducibility

Table 9 lists data that establishes the reproducibility of the analytical techniques. Data in Table 9 represent repeated analyses of two different samples (taken 10 hours apart from the CPDU at essentially identical operating conditions) performed by two operators. Sample III-1 was analyzed by the same operator twice and sample III-2 was analyzed by two different operators.

Results indicate that all of the data fall within the range of statistically permissible maximum differences established in Table 8. After the analytical precision of the data was established, we next checked the reproducibility of the CPDU. Yield distributions were calculated from two different samples taken 10 hours apart from the CPDU at the same operating conditions. The data (Table 10) clearly indicate that the unit is reproducible within the analytical precision established earlier.

RESULTS AND DISCUSSION

Series versus Parallel Operation: Because differences in yields were expected to be small, these experiments were conducted at three temperature levels. Results in Table 11 indicate that conversion decreased with increasing temperature. However, the reactor configuration did not seem to affect conversion at any temperature level.

Preasphaltene yields decreased at all temperature levels when the two reactors were operated in series. The decrease is outside the statistically permissible maximum differences established earlier (Table 8). At the highest temperature studied (449°C), the difference between the single reactor and two reactors in series was only 2.7% on an MAF coal basis; this value is statistically significant.

The reactor configuration did not change the sulfur contents of SRC significantly at 449°C, however, at 415 and 432°C, the series configuration resulted in a slight decrease

in the SRC sulfur content. Also the reactor configuration did not significantly influence asphaltene yields. At 415°C, the yield increased for two reactors operated in series, but at 432 and 449°C, no changes were noticed. Similarly, at 432°C the increased oil yield with the series configuration was statistically significant. However, the oil and asphaltene yields did not change at all temperature levels--only the preasphaltene yields showed the same trend at all temperature levels.

These results indicate that operating the dissolvers in series will decrease the preasphaltene yield. From a process viewpoint, this reduction should increase the amount of recoverable product. Recovery from the critical solvent deashing unit in the plant will increase when preasphaltene content decreases. Neither conversion nor oil, asphaltene, or gas yields are affected by reactor configuration.

Effect of Staged Temperatures: One of the advantages of series operation is that an added degree of flexibility is available by being able to vary the temperature in each dissolver. The objective of the study reported here was to determine the advantages, if any, of temperature staging. Operating conditions for experimental parts (VII-IX) are provided in Table 1.

Table 12 shows that oil yield was at a maximum when the first reactor was operated at 432°C and the second reactor at 449°C, compared to other temperature combinations. Yields of other fractions followed the expected pattern. For example, when the temperature of the first dissolver increased, hydrocarbon gas yields increased and preasphaltene yields decreased. Earlier results for Lafayette coal showed (Table 11) that increasing temperature decreased conversion; the same trend is observed here.

The data also indicate that operating the second dissolver at 432 and first one at 449°C reduced gas yields and increased oil yields. However, from a process viewpoint, operating the second dissolver at a lower temperature than the first would be impractical, requiring considerable quenching between the two.

Staged-temperature operation of the dissolvers has potential advantages. For the Lafayette Kentucky #9 coal, operating the first dissolver at 432°C and the second at 449°C resulted in a higher yield of oils than several other temperature combinations that were tested.

REFERENCES

1. Morris, S.M. and E.P. Foster, "The SRC-I Coal Refining Demonstration Plant," presented at the conference on Synthetic Fuels: Status and Directions, San Francisco, California, 14 October 1980.

2. Tao, J.C., Malhotra, R.K., and Sukel, T.M., Foster, E.P., and S.M. Morris, "Solvent Refined coal (SRC-I) Technology, Product Markets and Economics," presented at the 3rd International Coal Utilization Exhibition and Conference, Houston, Texas, November 1980.
3. Ying, D.H.S., Sivasubramanian, R., Moujaes, S.F., and E.N. Givens, "Gas Slurry Flow in Coal Liquefaction Processes," Final Technical Progress Report for Period 1 October 1979 - 31 March 1982, DOE-TIC 14801-T30, April 1982.
4. Thorogood, R.M. and C.L. Yeh, "Coal-Reactor Design for the SRC-I Process," presented at the 72nd Annual AIChE Meeting, San Francisco, California, November 1979.
5. Martin, C.J., July 1979. Kinetic model for coal slurry reactors. Phase Zero SRC Report.
6. Ying, D.H.S., 1982. Stirred reactor hydrodynamics. Pages 273-292 in SRC-I quarterly technical report, April-June 1982. DOE/OR/03054-8. International Coal Refining Co., Allentown, Pa.
7. Joshi, S.V., 1981. Coal process development unit (CPDU) data analysis. Pages 405-467 in SRC-I quarterly technical report, April-June 1981. DOE/OR/03054-4 Vol. 2. International Coal Refining Co., Allentown, Pa.

**TABLE 1
OPERATING CONDITIONS**

PRESSURE - 1.39×10^7 PASCALS
 H_2 FLOW - $0.87 \text{ m}^3/\text{kg COAL}$
 FEED COMPOSITION - 40% KY #9 COAL
 60% RECYCLE SOLVENT

REACTOR I			REACTOR II	
PART NUMBER	TIME, SECONDS	TEMP., °C	TIME, SECONDS	TEMP., °C
I	—	—	1716	415
II	—	—	1740	432
III	—	—	1686	449
IV	726	415	822	415
V	768	432	870	432
VI	768	449	846	449
VII	798	415	906	449
VIII	726	432	822	449
IX	750	449	852	432

**TABLE 2
ANALYSIS OF KY#9 COAL
(LAFAYETTE)**

PROXIMATE	
MOISTURE	1.61
VOLATILE MATTER	34.48
ASH	9.40
ULTIMATE ANALYSIS, MOISTURE FREE BASIS	
CARBON	72.93
HYDROGEN	5.00
OXYGEN	9.18
NITROGEN	1.54
SULFUR	2.94

**TABLE 3
ANALYSIS OF RECYCLE SOLVENT**

SOLVENT SEPARATION	
OILS	94.2
ASPHALTENE	4.8
PREASPHALTENE	0.8
RESIDUE	0.1
ELEMENTAL ANALYSIS	
CARBON	87.9
HYDROGEN	8.3
NITROGEN	1.0
SULFUR	0.4
OXYGEN	3.1

**TABLE 4
OIL (MULTIPLE ANALYSIS)**

SAMPLE NO.	WT %	DF	SSE
1	64.8, 65.8, 65.2, 65.4	3	0.52
2	66.1, 65.3	1	0.34
3	66.3, 64.9	1	0.98
4	63.7, 62.0	1	1.45
5	67.4, 63.1	1	9.42*
$S^2 = \frac{SSE}{DF}$ $= 3.29/6$ $= 0.548$ $S = 0.74$			

df: DEGREES OF FREEDOM
 SSE: SUM OF SQUARED ERRORS

**TABLE 5
ASPHALTENES
(MULTIPLE ANALYSIS)**

SAMPLE NO.	WT %	DF	SSE
1	11.2, 11.3, 12.5, 12.8	3	2.01
2	11.0, 11.4	1	0.08
3	11.5, 12.3	1	0.32
4	11.5, 13.6	1	2.21
5	12.2, 14.0	1	1.62
$S^2 = 6.24/7$ $= 0.891$ $S = 0.944$			

**TABLE 6
PREASPHALTENES
(MULTIPLE ANALYSIS)**

SAMPLE NO.	WT %	DF	SSE
1	13.2, 12.7, 13.4, 12.4	3	0.633
2	11.8, 11.9	1	0.005
3	11.8, 12.3	1	0.125
4	15.5, 15.7	1	0.020
5	13.6, 14.6	1	0.500
$S^2 = 1.283/7$ $= 0.183$ $S = 0.43$			

**TABLE 7
RESIDUE
(MULTIPLE ANALYSIS)**

SAMPLE NO.	WT %	DF	SSE
1	10.9, 10.3, 9.1, 9.5	3	1.95
2	11.0, 11.4	1	0.08
3	10.4, 10.0	1	0.08
4	9.3, 8.7	1	0.18
5	6.8, 8.3	1	1.13
S ² = 3.415/7			
= 0.488			
S = 0.698			

**TABLE 8
STATISTICALLY PERMISSIBLE
MAXIMUM DIFFERENCES
CONFIDENCE LEVEL - 95%**

	DIFFERENCES, WT % (2 ANALYSES) BASED ON		DIFFERENCES, WT % (4 ANALYSES) BASED ON	
	TPL*	MAF COAL	TPL*	MAF COAL
OILS	1.83	5.14	1.30	3.65
ASPHALTENES	2.23	6.27	1.59	4.47
PREASPHALTENES	1.02	2.87	0.72	2.02
RESIDUE	1.65	4.64	1.17	3.09

*TOTAL PRODUCT LIQUID

**TABLE 9
ANALYTICAL REPRODUCIBILITY**

OPERATOR	A	A	A	B
SAMPLE	III-1	III-1	III-2	III-2
OILS	66.1	65.3	66.3	64.9
ASPHALTENES	11.0	11.4	11.5	12.8
PA	11.8	11.9	11.8	12.3
RESIDUE	11.0	11.4	10.4	10.0

**TABLE 10
UNIT REPRODUCIBILITY
YIELD DISTRIBUTIONS**

OPERATING CONDITIONS:

TEMPERATURE, °C	449/449
PRESSURE, PASCALS	1.39 × 10 ⁷
TIME, SECONDS	768/846

	III-1	III-2
HYDROCARBON GASES	8.6	7.0
OTHER GASES	2.6	2.5
H ₂ O	1.7	3.7
OIL	17.3	16.0
ASPHALTENES	21.5	24.0
PREASPHALTENES	31.3	31.4
IOM	18.4	17.0

**TABLE 11
EFFECT OF STAGED DISSOLVERS**

PRESSURE - 1.39 × 10⁷ PASCALS
H₂ FEED RATE - .87 m³/Kg COAL

PART NUMBER	I	II	III	IV	V	VI
TEMPERATURE, °C	415	432	449	415/415	432/432	449/449
RESIDENCE TIME, SECONDS	1716	1740	1686	726/882	768/870	768/846
YIELD, % maf COAL						
HYDROCARBONS	2.2	4.0	6.9	2.9	5.6	7.0
OTHER GASES	2.8	2.1	2.2	1.6	2.2	2.5
OILS	13.5	13.7	17.5	11.9	17.6	17.2
ASPHALTENES	22.2	25.7	23.7	30.6	26.6	24.0
PREASPHALTENES	46.7	40.6	32.9	42.3	35.3	30.2
RESIDUE	11.9	13.6	16.8	10.0	11.1	16.7
WATER	1.9	1.7	2.0	1.6	2.5	3.7
H ₂ CONSUMPTION, TOTAL	0.82	1.1	1.48	0.89	1.39	1.55
FROM GAS	0.67	0.64	0.84	0.55	0.92	0.92
FROM SOLVENT	0.15	0.46	0.64	0.34	0.47	0.63
S IN SRC	1.28	1.00	0.88	1.0	0.90	0.90
SOLVENT HYDROGEN, WT%	8.21	8.02	7.92	8.10	8.02	7.92

**TABLE 12
EFFECT OF STAGED TEMPERATURES**

PRESSURE - 1.39 × 10⁷ PASCALS
H₂ FEED RATE - 0.87 m³/Kg COAL

PART NUMBER	VII	VIII	VI	IX
TEMPERATURES, °C	415/449	432/449	449/449	449/432
TIME, SECONDS	798/906	726/822	768/846	750/852
YIELDS, maf COAL				
HYDROCARBONS	5.0	5.6	7.0	5.4
OTHER GASES	1.7	2.2	2.5	2.0
OILS	12.1	22.7	17.2	19.6
ASPHALTENES	23.3	23.0	24.0	22.3
PREASPHALTENES	38.9	31.6	30.2	33.5
IOM	13.5	12.4	16.7	14.8
H ₂ CONSUMPTION	1.42	1.74	1.55	1.37

FIGURE 1
SRC-I PROCESS
COAL REACTION EQUIPMENT

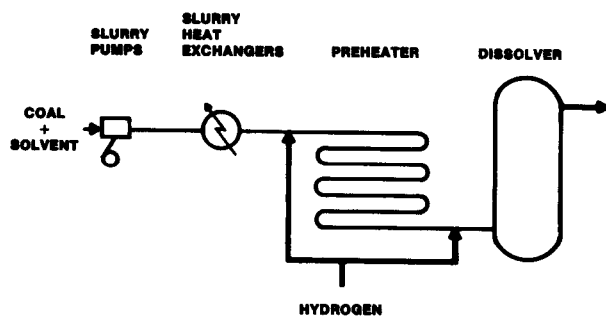


FIGURE 2
OIL YIELD FOR PYRO COAL, PREDICTED
FROM SEQUENTIAL KINETIC MODEL

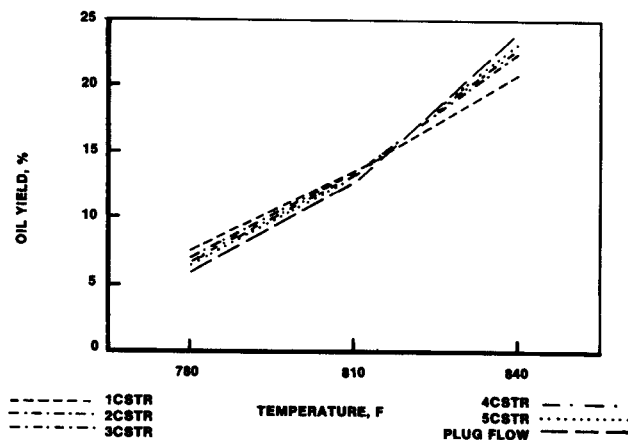


FIGURE 3
PREASPHALTENE YIELD FOR PYRO COAL,
PREDICTED FROM SEQUENTIAL KINETIC MODEL

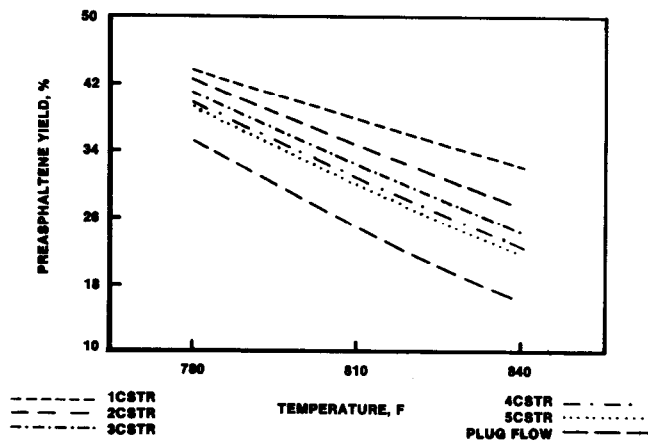
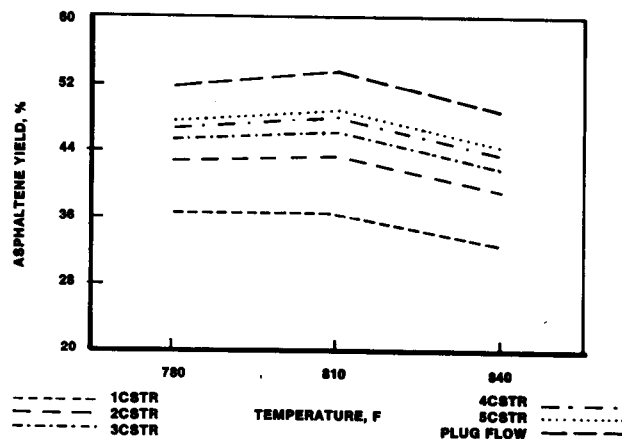


FIGURE 4
ASPHALTENE YIELD FOR PYRO COAL,
PREDICTED FROM SEQUENTIAL KINETIC MODEL



DIRECT SNG PRODUCTION BY THE CS/R
HYDROGASIFICATION PROCESS

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ABSTRACT

The CS/R Hydrogasification Process utilizes a short-residence-time entrained-flow reactor, derived from aerospace rocket reactor technology, for quickly reacting pulverized coal with hot hydrogen to directly produce substitute natural gas (SNG). Development testing has indicated the feasibility of two primary process options: (1) the production of SNG as the sole major product or (2) the coproduction of SNG and chemical-grade benzene. Recent emphasis has focused on process design, optimization, and economics. Preliminary design studies of commercial-scale (250×10^9 Btu/day) grassroots SNG plants have been completed for two widely different types of feedstock: Kentucky No. 9 hvAb coal and Minnesota peat.

This paper summarizes the pertinent experimental data and analytical modeling studies of flash hydrolysis used as a basic input to the process design effort. The commercial process flowsheets for each feedstock are described, and the resultant capital and operating costs are discussed. Sensitivity analyses are presented relating the cost of gas to the major process operating variables and economic parameters.

INTRODUCTION

Rockwell International has been developing technology for the flash hydrolysis (FHP) of coal and other solid carbonaceous feedstocks under DOE contract since 1975. Cities Service Company has been a major subcontractor in this work, and Rockwell and Cities Service are engaged in the joint development and commercialization of the resultant technology, under the general heading of "CS/R Hydrolysis."

CS/R Hydrolysis is carried out in entrained-flow reactors to accomplish controlled reactions between an entrained, pulverized-solid feedstock and a hot, hydrogen-rich carrier gas stream. Residence times in the reactors are relatively short, and a large fraction of the feedstock's carbon is converted directly to a selected slate of gaseous and liquid hydrocarbon products. The makeup of the product slate can be varied over a broad range — from predominantly a syncrude oil with SNG byproduct, to SNG with relatively pure benzene byproduct, to SNG alone

as the sole hydrocarbon product — by varying the severity of the hydrolysis reactor's operating conditions. (Severity is increased by raising the reactor's exit temperature, the reactor residence time, or both.)

As part of its contractual work for DOE, Rockwell International had the CE-Lummus Company conduct two preliminary conceptual design studies of 250×10^9 Btu/day commercial SNG plants based on CS/R Hydrogasification. Both studies considered grassroots, mine-mouth SNG plants, one fed Eastern U.S. bituminous coal, the other fed Minnesota peat. The objectives in each case were to assemble an approximately optimum preliminary process flowsheet for the entire plant; to calculate material and energy balances for the process; and to estimate overall economics, including total installed cost, total capital requirements, annual operating costs, and average cost of gas.

ROCKWELL INTERNATIONAL HYDROGASIFIER CONCEPT

The basic reactor concept underlying the CS/R Process is the Rockwell Hydrogasifier illustrated in Figure 1. The reactor design is derived from aerospace rocket reactor technology. Impinging streams of dense-phase-fed pulverized coal and hot hydrogen are injected into the reactor. Complete and uniform mixing and heat transfer are accomplished in several milliseconds. All the reactor heat is input with

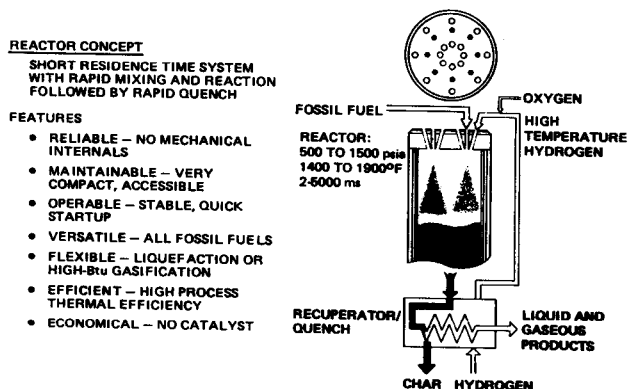


Fig. 1. Schematic of the Rockwell Flash Hydrolysis Reactor

the hot hydrogen, with the exception of the exothermic heat of methane formation. Typically, the initial theoretical mix temperature is about 1500°F, which will increase to about 1800°F as the result of methane formation.

The dense-phase transport technique minimizes the amount of carrier gas fed with the coal, thereby minimizing the heat required from the injected hot hydrogen stream. In dense-phase feeding, only the interstitial gas surrounding the coal particles at rest (in a high-pressure feeder vessel) is injected with the coal.

Because of the extremely high coal heatup rates (~500,000°F/s) in this type of entrained flow reactor, the coal particles are very rapidly devolatilized, thereby precluding any possibility of agglomeration when caking coals are used. Thus, the reactor has successfully handled a wide range of coal types, including peat, subbituminous coal, and highly caking bituminous coals.

Following conventional aerospace rocket reactor practice, reactor scaling is accomplished by "multiplicity." First a single reactor injection element is characterized. A typical hydrogasifier element consists of a central coal feed stream with several surrounding hot hydrogen jets impinging on it. Once the single-element geometry has been established, several of these identical elements can be appropriately clustered onto the injector face of the scaled up reactor.

CURRENT STATUS OF ROCKWELL HYDROGASIFIER DEVELOPMENT

Rockwell has conducted extensive hydrogasifier development testing in two different reactor test facilities: a 1/4-ton/h coal feed rate unit capable of short duration (approximately 10 min) screening tests and a 3/4-ton/h engineering-scale test unit (ETU) that could run for up to 30 min with better material balance accounting. Several gasification tests were also made in a 1-ton/h hydroliquefaction PDU at 1/4-ton/h and durations up to 75 min.

Highly and mildly caking Eastern bituminous coals (Western Kentucky No. 9 hvAb and Illinois No. 6 hvCb), nonagglomerating Western subbituminous coal (Montana Rosebud), and Minnesota peat have been used as feedstocks in Rockwell's hydrogasification testing. Substantial data bases have been accumulated on overall conversions and conversions to specific products over rather wide ranges of key reactor operating variables (i.e., temperature, residence time, hydrogen/solid feed rate ratio, and pressure). More detailed summaries of test facility design and operation and of experimental results have been given in contractual reports for DOE (1-4) and discussed in several technical papers (5-7).

The practical, engineering scale of Rockwell's extensive experimental test program has clearly established the viability of entrained-flow hydrogasifier operations. Elemental overall conversions and conversions to the major products

have been determined quantitatively with sufficient accuracy for preliminary design and economic assessments of commercial applications. For those feedstocks having the largest data bases (Kentucky No. 9 hvAb, Montana Rosebud subbituminous, and Minnesota peat), the conversion data have been correlated against a mathematical model of FHP kinetics, and this has improved confidence in their validity.

DOE and Rockwell have planned for some time to build and operate a 3/4-ton/h integrated process development unit (IPDU) capable of 30-day-long runs as the next step in developing CS/R Hydrogasification technology. However, with IPDU design and construction approximately 60% complete, the project was placed on an indefinite hold because of reductions in the DOE budget for fossil energy programs.

REACTOR PERFORMANCE MODELING

Rockwell has developed a flash hydropyrolysis reaction model that very successfully correlates and predicts experimental FHP reactor performance (8). The model expands on the formulation of Anthony and Howard (9) in which the devolatilization kinetics are modeled in terms of multiple parallel reactions having a Gaussian distribution of activation energies. Volatiles and rapid-rate carbon are produced, which in turn can react either with hydrogen, producing hydrocarbon gases plus char, or can simply cross-link and form char.

When extremely complicated chemical reactions are modeled by a set of reactions that are essentially phenomenological, the modeling scheme should be kept as simple as possible. Following this philosophy, the generalized scheme shown in Figure 2 was evolved to model total carbon conversion, conversion to liquids, and conversion to BTX in an FHP reactor. Coal is assumed to devolatilize according to multiple parallel reactions to light gases, vapors of nonreactive liquid species, vapors of reactive liquid intermediates, and activated char. The liquid intermediates may be fractionated and/or hydrogenated to more stable liquids or polymerized to solid carbon or

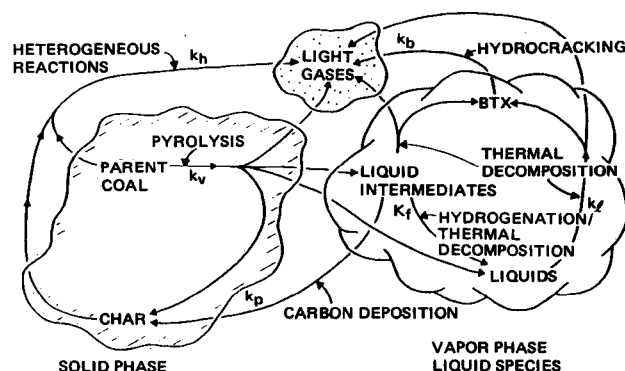


Fig. 2. Reaction Model for Flash Hydropyrolysis of Coals

char. The reactive char can be hydrogenated heterogeneously to gases or stabilized to non-activated char. The nonreactive liquids are subject to thermal decomposition, forming light gases, BTX, and liquids. The non-BTX liquids decompose to gases and BTX, and the BTX decomposes further to gases.

Experimental FHP conversion data for bituminous and subbituminous coals and peat have been correlated with the Rockwell model. The resultant correlation models can, in turn, be used to generate specific reactor performance plots, thereby providing extremely valuable design tools for selecting reactor operating conditions having potentially attractive commercial-scale economics.

Two examples of model-calculated conversion charts are given in Figures 3 and 4. Figure 3 is a plot of Kentucky No. 9 hvAb coal carbon conversions at a hydrogen partial pressure of 1000 psia and three different reactor temperatures as functions of reactor residence time. Three curves are shown for each temperature — overall carbon conversion, carbon conversion to liquids, and carbon conversion to BTX liquids. Performance aspects that are evident in Figure 3 are the increase in gas production with increasing reactor temperature and the fall-off in total conversion as residence time is increased above about 0.4 s at the lower temperatures. The latter effect is attributed to the back-deposition of carbon on the char particles as the pyrolysis vapors are hydrocracked to form BTX and light gases.

Figure 4 is a similar model-calculated conversion plot for peat. The increase in gas production and decrease in non-BTX liquid production with increasing residence time and increasing temperature are evident. BTX production passes

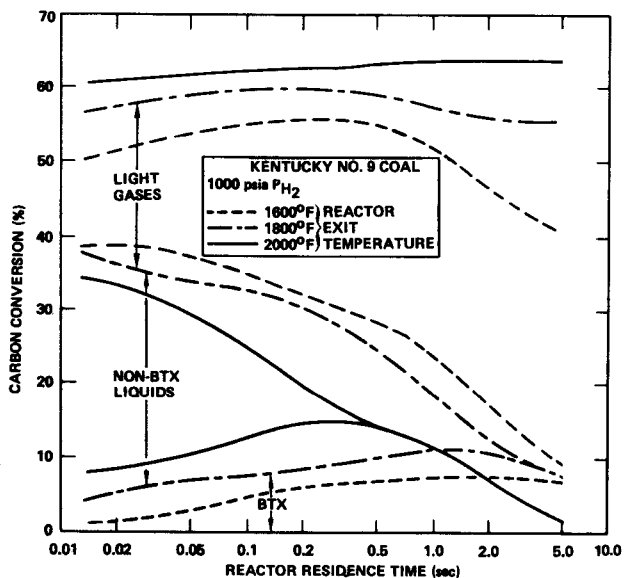


Fig. 3. Model-Calculated Carbon Conversions for Hydrogasification of a Bituminous Coal

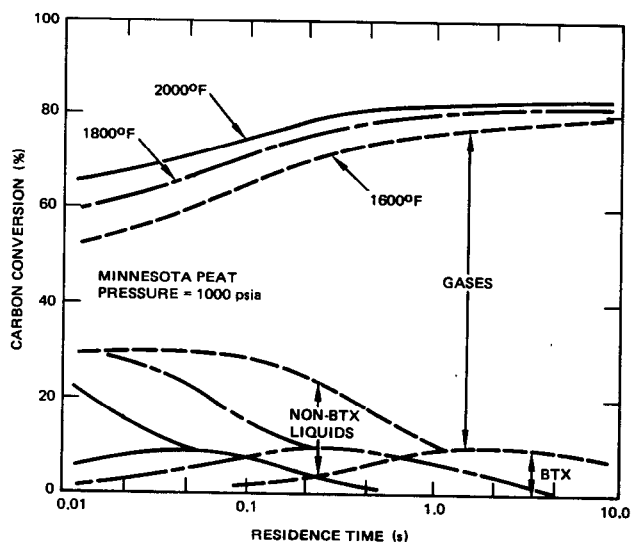


Fig. 4. Model-Calculated Carbon Conversions for Hydrogasification of Peat

through a maximum at residence times of about 2.0, 0.3, and 0.06 s at temperatures of 1600, 1800, and 2000°F, respectively. At these temperatures, the yield of non-BTX liquids becomes essentially nil at residence times of about 1.0, 0.2, and 0.05 s, respectively.

Similar curves were generated at additional reactor pressures and temperatures to cover the entire range of interest for process design applications. Lummus used these plots to optimize reactor conditions for the commercial plant studies described in the next section.

PROCESS DESIGN AND ECONOMICS

Coal Hydrogasification

As an integral part of the Coal Hydrogasification Process Development Program sponsored by the U.S. Department of Energy, the CE-Lummus Company prepared under subcontract to Rockwell a preliminary design of a complete 250×10^9 Btu/day commercial-scale SNG-from-coal plant using the CS/R Hydrogasification Process. The prime objective of the study was to integrate the Rockwell single-stage coal hydrogasifier with an appropriate combination of plant subsystems to yield the most effective technical and economic result. The study was conducted over an 18-month period, culminating in the issuance of a report submitted to Rockwell in May 1982 (10). The process flowsheet developed by Lummus is conservative, providing a baseline framework for future process optimization. Lummus identified the key process steps which, during normal process development, will be refined and substantially improved.

A simplified block flow diagram of the CS/R hydrogasification plant configuration is shown in Figure 5. Run-of-mine Kentucky No. 9 hvAb coal

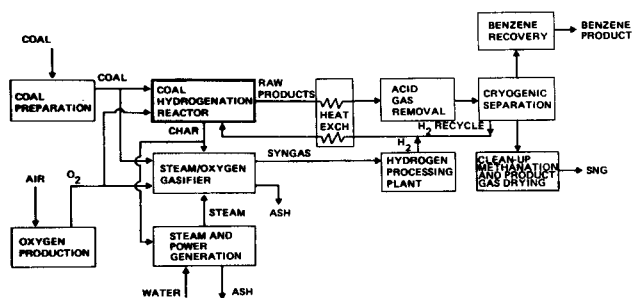


Fig. 5. Simplified Flow Schematic of the CS/R Coal Hydrogasification Process

is ground to 70% through 200 mesh and dried to a moisture content of 2% in a conventional flash-drying system. Part of the coal is pressurized in lock hoppers and hydrogasified in the Rockwell short-residence-time entrained-flow gasifier, using hydrogen as the hydrogasifying medium. The effluent gas is cooled by preheating the hydrogen feed and raising high-pressure steam, then separated from the char, and passed through an MEA acid gas removal system to remove hydrogen sulfide and carbon dioxide. The methane and residual carbon monoxide contained in the desulfurized gas are cryogenically separated from unreacted hydrogen, vaporized, compressed, and trim methanated in conventional catalytic methanators to convert the residual carbon oxides to methane. This gas, after being dried in glycol units, is delivered to a gas pipeline at 1000 psig and ambient temperature. The hydrogen stream from the cryogenic unit is compressed, along with makeup hydrogen, and recycled to the hydrogasifiers.

The char removed in the particulate cleanup section of the hydrogasifier trains is sent to a dry-fed oxygasification system. The raw gas produced in the oxygasifiers is divided into two parts. The first part is treated in a Selexol unit to remove hydrogen sulfide and then is sent to a combined-cycle plant to generate electric power. The second part is used to produce the makeup hydrogen required in the hydrogasifiers. In addition, fresh coal is gasified in the oxygasifiers to generate the balance of makeup required. Raw gas generated in the oxygasifiers contains primarily hydrogen, methane, carbon monoxide, carbon dioxide, and hydrogen sulfide. This gas is sent through a shift conversion unit, to convert carbon monoxide to hydrogen, and subsequently is passed through an acid gas removal unit before being compressed and mixed with the recycle hydrogen stream. Westinghouse oxygasification technology was chosen for use in this study.

The process condensate from both the hydrogasification and the Westinghouse coal/char gasification trains is treated for recovery of ammonia and aromatics. The aromatics recovered from the process condensate and the cryogenic hydrogen recovery plant are upgraded in stripping

columns followed by the Houdry-Litol process for producing high-purity, chemical-grade benzene. Besides ammonia and aromatics, sulfur in the form of hydrogen sulfide is also recovered from the process condensate. This hydrogen sulfide and the sulfur-containing streams from the acid gas removal units in the hydrogen plant, the power generation plant, and the main hydrogasification plant are treated in a Claus/Scot plant to recover elemental sulfur.

The preliminary results obtained by Lummus showed excellent economics and confirmed the predictions of earlier studies. The cost of pipeline gas (SNG) calculated by the utility financing method (75/25 debt/equity ratio) is \$3.68/10⁶ Btu on a mid-1979 basis. The economic guidelines used in computing these cost estimates were obtained directly from Ref. 11.

The capital and operating costs determined by Lummus have been escalated by Rockwell to an August 1982 economic basis using the CE Plant Cost Index and updated operating costs and credits. The resultant cost of SNG on this basis is plotted in Figure 6 as a function of coal

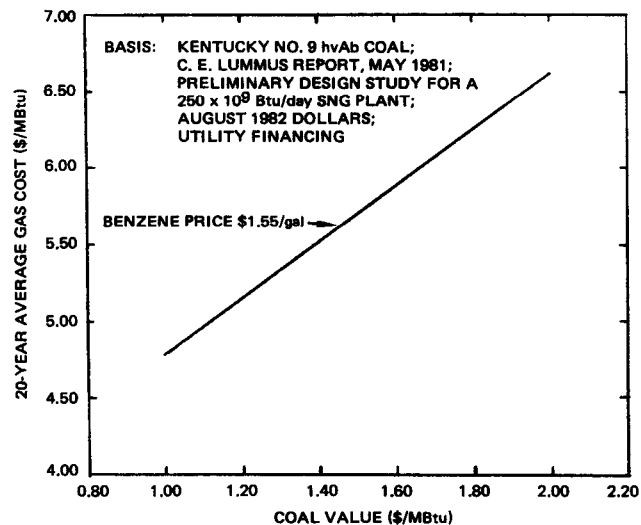


Fig. 6. CS/R Coal Hydrogasification Process - Effect of Coal Value on Gas Cost

value. The total plant investment, including contingency, is estimated to be \$1.76 billion in August 1982 dollars. The total capital requirement, which includes allowance for funds during construction, startup costs, working capital, etc., is \$2.19 billion. Annual byproduct credit, primarily for chemical-grade benzene (valued at \$1.55/gal), amounts to \$134 million and reduces the cost of gas by approximately \$1.60/10⁶ Btu. The specific effect of benzene price on SNG cost for an assumed value of coal at \$1.50/10⁶ Btu is illustrated in Figure 7.