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HIGH CONVERSION OF COAL TO TRANSPORTATION FUELS
FOR THE FUTURE WITH LOW HC GAS PRODUCTION

Progress Report No. 14
(Covering the period January 1 - March 31, 1996)

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PERSONNEL

In addition to the Principal Investigators, research personnel participating in this project during the reporting period are:

Belma Demirel, Graduate Student

OBJECTIVES AND SCOPE OF THE RESEARCH PROJECT

An announced objective of the Department of Energy in funding this work, and other current research in coal liquefaction, is to produce a synthetic crude from coal at a cost lower than \$30.00 per barrel (Task A). A second objective, reflecting a recent change in direction in the synthetic fuels effort of DOE, is to produce a fuel which is low in aromatics, yet of sufficiently high octane number for use in the gasoline-burning transportation vehicles of today. To meet this second objective, research was proposed, and funding awarded, for conversion of the highly-aromatic liquid product from coal conversion to a product high in isoparaffins, which compounds in the gasoline range exhibit a high octane number (Task B).

Experimental coal liquefaction studies conducted in a batch microreactor in our laboratory have demonstrated potential for high conversions of coal to liquids with low yields of hydrocarbon (HC) gases, hence small consumption of hydrogen in the primary liquefaction step. Ratios of liquids/HC gases as high as 30/1, at liquid yields as high as 82% of the coal by weight, have been achieved. The principal objective of this work is to examine how nearly we may approach these results in a

continuous-flow system, at a size sufficient to evaluate the process concept for production of transportation fuels from coal.

A continuous-flow reactor system (1/2 inch inside diameter) is to be designed, constructed and operated. The system is to be computer-operated for process control and data logging, and is to be fully instrumented. The primary liquid products will be characterized by GC, FTIR, and GC/MS, to determine the types and quantities of the principal components produced under conditions of high liquids production with high ratios of liquids/HC gases. From these analyses, together with GC analyses of the HC gases, hydrogen consumption for the conversion to primary liquids will be calculated. Conversion of the aromatics of this liquid product to isoparaffins will be investigated, to examine the potential for producing a transportation fuel from coal with satisfactory octane rating but low in aromatic content.

ACTIVITY FOR THE REPORTING PERIOD

A no-cost extension was granted, to extend this Grant to September 28, 1996. The principal purpose in seeking the extension was to allow for completion of the research being performed by the Graduate Student, Belma Demirel, and writing of her thesis for the Ph.D. Degree. A second purpose was to provide time and funding within the Grant for preparation of the Final Report, including payment from the Grant for the costs of printing the report, and for necessary time of the P.I. in the preparation. When the time extension commenced, the funding still available in the Contract would scarcely accomodate the objectives stated here, with no funds available to continue research with the main reactor system.

TASK A

In work performed in our laboratory under a previous DOE Contract, utilizing a batch microreactor, it was demonstrated that conversion in excess of 60% by weight of the coal to liquids can be achieved in a single pass, and as high as 82% with recycle of unreacted solids, with residence times as short as 3 seconds, accompanied by ratios of liquids/hydrocarbon (HC) gases as high as 30/1. The principal objective in Task A was to design, construct and operate a continuous-flow reactor system in which to investigate whether conversions and ratios of liquids/hydrocarbon (HC) gases could be achieved in a continuous system which would approach those obtained in the batch microreactor, while also providing reasonable assurance that such a continuous system can be scaled to a larger size, still achieving approximately the same conversions and high ratios of liquids/HC gases.

A continuous system has been constructed and operated, with a one-half inch inside diameter (ID) tube as the reaction vessel. After minor modifications, the system has operated as intended. By proper selection of parameters, it is possible in a continuous system to convert a high percentage of coal to liquids while producing much lower quantities of HC gases than are produced in other technologies, thereby conserving expensive hydrogen.

It had been previously demonstrated in our laboratory that if this is to be accomplished, residence time in the hot zone must be short, although it was not known at the outset how short this time may be, or in fact would need to be. A second cost benefit was anticipated by reason of the short time, hence higher throughput leading to smaller reactor size for a given throughput.

As the work in this project proceeded toward its conclusion, a third and unexpected benefit was discovered. As the residence times were decreased to values of 10 seconds or less, ratios of liquids/HC gases of 20/1 or higher were achieved. But very importantly, it was discovered that the chemical reactions which produce the primary liquids can be carried to high conversions at pressures much lower than reported, and indeed required, in the processes at longer times. Specifically, it was found that pressures as low as 500 psig could still give conversions to liquids of the order of 50 percent by weight of the coal in a single pass, and approaching 75 percent with recycle of unreacted coal, accompanied by low HC gas yields, with ratios of liquids/HC gases of 20/1 or greater. High yields of liquids were obtained in times observed to be as short as 3 seconds, while production of

significantly-large quantities of HC gases were not observed until times of 10 seconds or greater in the hot zone, an observation of great significance.

A very significant element of this latest observation involving pressures as low as 500 psig, or perhaps lower when parameters are optimized, is concerned with feeding the coal. In order to obtain high conversions in times as short as 3 seconds, the coal particles must be heated extremely rapidly. Other coal liquefaction processes begin by dissolving the coal in an appropriate solvent, in order to pump it into the pressurized system. However, when the coal particles are surrounded by a liquid, particle heat-up rates are slow, and times to achieve high conversions are measured in times of several minutes, generally 15 minutes or longer, and even 5 minutes in other so-called short-residence-time processes. A system for feeding dry solids to a system, at pressures of 2000 psig or greater typical of other technologies, has not been demonstrated. Lurgi coal gasification systems have demonstrated satisfactory reliability for feeding dry coal to the pressurized reactor, utilizing the lock hopper, at pressures up to 500 psig. Therefore, it can be concluded that a feed system for the present technology, operating at pressures as low as 500 psig, or lower, is available in the market place. This potential area of uncertainty has been argued by detractors of the very-short-time approach to coal liquefaction, but no longer is a valid argument.

Therefore, the principal objectives of high conversion to liquids with high ratios of liquids/HC gases, in a continuous-flow

system, has been demonstrated. Also, the potential for production of liquids from coal at a substantially-reduced cost, when compared with other, advanced technologies, has also been demonstrated. The reduced cost results from a combination of lower hydrogen consumption, smaller reactor per amount of coal throughput, and much less expensive reactor by reason of the low pressures. Two separate and independent evaluations have concluded that the cost of a primary liquid from coal by the technology under development in this work may be lower than the costs from other, advanced technologies by as much as 25 percent, a very significant advance.

In order to attract an industrial partner to invest in development of the technology in a major way, several of the parameters need to be optimized. Such optimization was not a part of the proposal which led to the present work. Indeed, it was not then known the roll to be played by each parameter, or the general range of values for each, such as pressure of the order of 500 psig. It is concluded that one additional round of research needs to be performed under government financing. A new proposal has been submitted to the University Coal Research program, to perform the optimization studies. Further, through these studies it will be shown just how high the conversion of coal to liquids may be in a single pass. The technology will be significantly simpler if recycle of unreacted solids is not required in order to obtain a sufficiently-high liquid yield. Conversions in the present work are approaching values high enough to indicate that, through optimization of parameters, a sufficiently-high conversion in a single pass is possible.

The general climate in the energy industry, with prices of crude oil on the world market remaining low, does not allow for this next stage of the work to be financed wholly by the private sector, particularly the optimization studies. It is hoped that funding can be made available through DOE.

TASK B

During this reporting period, the Graduate Student, Belma Demirel, has completed writing her thesis for the Ph.D. Degree in Fuels Engineering. She has successfully defended her thesis before her Committee, and completed all requirements for the degree. She has received a Post-doctoral research appointment at the University of Kentucky, under another DOE-sponsored project on coal.

The title of her thesis is "Production of High-Octane Gasoline Components by Hydroprocessing of Coal-derived Aromatic Hydrocarbons". As indicated in earlier Progress Reports, we have succeeded in demonstrating that a high-octane gasoline can be produced from aromatic compounds representative of the compounds found in coal-derived liquids, by straight-forward, practical chemistry, without including aromatic compounds in the gasoline.

As soon as a copy of the thesis in its final form is received, the Final Report will be prepared, to be submitted by the end of the Grant period as presently defined, namely September 28, 1996.

The support of DOE in my work, which over the years has led to the production of many Ph.D. degrees, and a lesser but significant number of Masters degrees, is gratefully acknowledged and appreciated.