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KINETICS AND MECHANISMS OF CATALYTIC HYDROLIQUEFACTION AND HYDROGASIFICATION OF LIGNITE

Quarterly Report for the Period

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ABSTRACT

Delay of six months in the delivery of a new slurry pump has prevented extension of the range of available residence times in the continuous stirred tank reactor.

A quick-feed mechanism installed on the batch autoclave permits more accurate determination of running times at constant temperature.

A detailed study of decomposition of carrier oil (anthracene oil) showed that gasification of the oil was considerably greater at high temperatures than previously expected. Future runs will be limited to conditions under which the carrier oil loss is a relatively small fraction of the oil generated from lignite.

OBJECTIVE AND SCOPE OF WORK

The research project is aimed at investigation of the basic chemical kinetics and mechanisms in catalytic hydrogenation of lignite suspended in a carrier oil in a continuous reactor operating at steady state. Specifically, the research is designed to study the overall conversion rate and the elemental transformation from the raw materials into products under surface reaction controlling conditions and to postulate the reaction mechanisms of hydroliquefaction and hydrogasification of lignite based upon the results obtained.

From the results a better understanding of continuous lignite and coal conversion processes may be obtained. This may lead to an improved ability to predict the behavior of pilot or demonstration plants under altered operation conditions and to possible directions for process improvements.

SUMMARY OF PROGRESS TO DATE

DETAILED DESCRIPTION OF TECHNICAL PROGRESS

TASK 1

The objective of this task is to install a high-pressure continuous hydrogenation unit and to develop a safe, efficient procedure for operating all equipment.

Continuous Pumping

The Hills McCanna slurry pump which is expected to broaden the range of flow rates and residence times had not been delivered by June 30 though it was first promised for delivery in January, then later in April. Cancellation of the order was considered, but this would result in a long delay if the contract is extended as proposed and the pump is then reordered.

Batch Autoclave

The quick-feed mechanism described in the previous quarterly report was designed and built with a teflon piston to assure nearly complete transfer of slurry feed. Because of mechanical problems causing jamming, the piston was removed and the device was found to function better with direct gas pressure on the slurry interface. Runs made with this feed mechanism allow much clearer definition of starting time by nearly complete elimination of the heat-up period.

TASK 2

The objective of this task is to investigate the effect of process variables on overall rate of conversion and on yield of products in the catalytic hydrogenation of lignite.

Gasification of Anthracene Carrier Oil

Principal attention during this period was devoted to a more accurate determination of the decomposition of carrier oil under process conditions. Because the loss of carrier oil is much greater than was originally anticipated, considerable error is introduced in calculation of net oil yield from the lignite slurry. Consequently a series of runs was made on anthracene oil in the CSTR in the presence of catalyst and hydrogen, at reaction times and temperatures used in lignite liquefaction.

The results are shown in the following figure. At 440°C and reaction time of 50 minutes, the loss of oil is about 12%. This is just about equal to the oil produced from the lignite at the usual slurry concentration. For meaningful results then, the carrier oil decomposition would have to be known to a high degree of accuracy.

ANTHRACENE CARRIER OIL GASIFICATION

Unfortunately one cannot be certain that carrier oil losses in the presence of lignite are the same as for carrier oil alone. Oil fragments which would be expected to be lost as gases may combine with fragments from the lignite and be retained.

Future experimental conditions will be chosen so that expected carrier oil losses are held to a small fraction of the oil produced from lignite. For example, at 420°C and a contact time of 23.5 minutes, the carrier oil loss is only 2%. For typical conditions this correction represents about one quarter of the oil yield, which is much more acceptable.

During the first run on lignite slurry after the series on oil gasification, the hydrogen supply failed and the reactor filled with a mass of coke. During its removal the agitator and cooling coil were ruined. Delay in obtaining replacement parts has set back the schedule. As a result an extension of the time of the contract will be sought without additional funds.

TASK 3

The objective of this task is to characterize the products obtained in the runs of Task 2.

No significant progress on this task has been made since the last quarterly report. All results including those obtained during the remainder of the grant period will be incorporated in the final report.

TASK 4

The objective of this task is to analyze and interpret the data of Tasks 2 and 3 in terms of the kinetics and mechanisms of hydroliquefaction and hydrogasification of lignite and to compare the results with those obtained in the literature on related systems.

A computer program has been obtained which will be of considerable assistance in modeling and interpreting reaction network. Extensive modification and testing of the program will be necessary to adapt it to our needs. This phase of the work is being carried out under the direction of Dr. Guniz Guruz, who is visiting Worcester Polytechnic Institute as a Fulbright Fellow.

CONCLUSIONS

By limiting the reaction temperature and time it is possible to operate under conditions in which correction of lignite oil yield based on gasification of carrier oil can be made with confidence. Future runs will be made under such conditions.

Use of a quick-feed mechanism to deliver lignite slurry to the preheated batch autoclave will permit close determination of reaction time at fixed temperature.

Determination of the reaction network and the rate constants in that network will be facilitated by extensive modification and use of an available computer program.