I. CONTRACT OBJECTIVE

The objective of the contract was to consolidate the advances made during the previous contract in the conversion of syngas to motor fuels using Molecular Sieve containing catalysts and to demonstrate the practical utility and economic value of the new catalyst/process systems with appropriate laboratory studies.

II. SCHEDULE

The contract work was initiated on September 18, 1984 and was divided into six tasks.

Task 1 consisted of the preparation of a detailed, non-proprietary work plan covering the entire performance of the contract. This was completed in November, 1984.

Task 2 consisted of a preliminary techno-economic assessment of the UCC catalyst/process system. This assessment, based on a sensitivity analysis which MITRE conducted on an updated version of their previously completed economic evaluation of the Union Carbide Corporation (UCC) system, was completed in 1986.

Task 3 consisted of the optimization of the most promising catalysts developed under prior contract DE-AC22-81PC40077 toward goals defined by the MITRE and Task 2 studies. This work was completed in March of 1988.

Task 4 consisted of the optimization of the UCC catalyst system in a manner which would give it the longest possible service life. This was completed in March of 1988.

Task 5 consisted of the optimization of a UCC process/catalyst system based upon a tubular reactor with a recycle loop (i.e., the ARGE reactor) containing the most promising catalyst developed under the previous task studies. This optimal performance was estimated from a

mathematical model of the tubular reactor which incorporated reaction rate constants determined from completed Berty reactor runs. This work was completed in June of 1988.

Task 6 consisted of an economic evaluation of the optimal performance found under Task 5 for the UCC process/catalyst system. This effort was based upon the MITRE sensitivity analysis referred to in the description of Task 2, and was completed in the third quarter of 1988.

III. ORGANIZATION

This contract was carried out by the Catalyst Research and Development Group of the Molecular Sieve Technology Department, Catalysts and Services Division, Union Carbide Corporation, Tarrytown, New York. Since the initiation of the contract the Molecular Sieve Department has become a part of a new subsidiary of UCC and Allied Chemical known as UOP Inc.

The principal investigator was Dr. J. G. Miller.

The program manager was Dr. A. C. Frost.

The program director was Dr. Jule A. Rabo.

IV. SUMMARY OF PROGRESS

This is the Final Technical Report and summarizes all of the work performed at the Union Carbide laboratories in Tarrytown, New York during the period of the contract between September 18, 1984 and June 1989. All of the tasks were successfully completed.

The two major steps of indirect coal liquefaction are the gasification of the coal into syngas and the subsequent conversion of the syngas into liquid hydrocarbon fuels with Fischer-Tropsch (F-T) catalysts. A previous contract (DE-AC22-81PC40077) had explored the use of Molecular Sieve catalysts in conjunction with F-T metal components (MC) to provide improved performance. The results were very encouraging. Subsequently, the present contract was initiated to pursue the leads discovered.

The work was divided into six (6) tasks as outlined in the Organization section of this report. The results will be summarized below.

Task 1 was a detailed plan of the work planned for the other tasks in the contract.

Task 2, a preliminary techno-economic assessment of the UCC catalyst/process system, was based on a sensitivity analysis which MITRE conducted on an updated version of their previous evaluation of the UCC catalytic system.

This sensitivity study graphically showed the differential cost (around the base case cost), expressed as differential cents per gallon of motor fuels, for changes in each of the operating parameters of space velocity, catalyst life, methane make, product Schulz-Flory alpha values, overall conversion, feed H2:CO ratio, reactor temperature, and reactor pressure.

These differential cost-operating parameter curves showed that catalyst activity, methane make, and catalyst life were the major contributors to the cost of the process. These factors became the focus of the development program

Tasks 3 and 4. The evolution of the catalysts from those identified by the work of the previous contract to those developed during the course of this contract represents the major effort of the program. Catalysts which provided high levels of syngas conversion were ultimately achieved which yielded very desirable product properties. These catalysts were based upon cobalt as the F-T component and used in conjunction with Molecular Sieves and were modified with additives. Additional efforts resulted in a great improvement in catalyst life. Studies were completed on catalyst regeneration and/or rejuvenation to possibly improve the economics further. Finally, comparisons were made with commercial type F-T catalysts and components, incorporating

what was learned about catalyst preparation, activation, etc., to confirm the performance benefits that had been demonstrated.

Tasks 5 and 6. In the Fifteenth Quarterly Report the following items were reported; (a) the correlation of the process runs into rate and selectivity expressions for the Co/X11/X9/TC-123 catalyst (No. 55), (b) the incorporation of these equations into the FIXBD computer simulation of a commercial ARGE-type reactor, (c) checking the results of this program against experimental results, and (d) the final use of the program in the techno-economic evaluation.

This evaluation showed that the economically optimum set of process conditions was 250 C, 500 psig, 450 GHSV, and a 1.75 H2/CO feed ratio.

The resulting conversion per pass through the reactors at these conditions was 70%. This was the economic optimum balance between more (or larger) reactors for a higher conversion per pass (with less downstream equipment for a smaller recycle stream) and fewer (or smaller) reactors for a lower conversion per pass (with more downstream equipment for a larger recycle stream).

At these conditions, the cost was $1.88 per gallon of all liquid fuels with a one year life for the catalyst in 1983 dollars or $2.12 per gallon in 1988 dollars.

It should be noted that a fixed bed reactor, with a suitable external recycle stream, should be run at 70%

conversion to confirm the predicted results. This is necessary since it is not clear that a one-to-one correlation between the Berty reactor and a fixed bed reactor can be assumed due to the different partial pressures of components which will exist in the two systems throughout the length of the reactor. It is believed that the Berty reactor provides a conservative result. In addition, the Berty reactor, having proved itself to be an outstanding device for conducting studies on highly exothermic reactions and extremely responsive to process control requirements, is not recommended for long term life studies. The complexity of the equipment which provides the benefits mentioned above, becomes the Achilles' heel when long term (six to twelve months) stability studies are undertaken.

V. FUTURE WORK

This report represents the completion of all work on this contract.