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SLURRY REACTOR DESIGN STUDIES. SLURRY VS. FIXED-BED REACTORS FOR FISCHER-TROPSCH AND METHANOL: FINAL REPORT

BECHTEL GROUP, INC.
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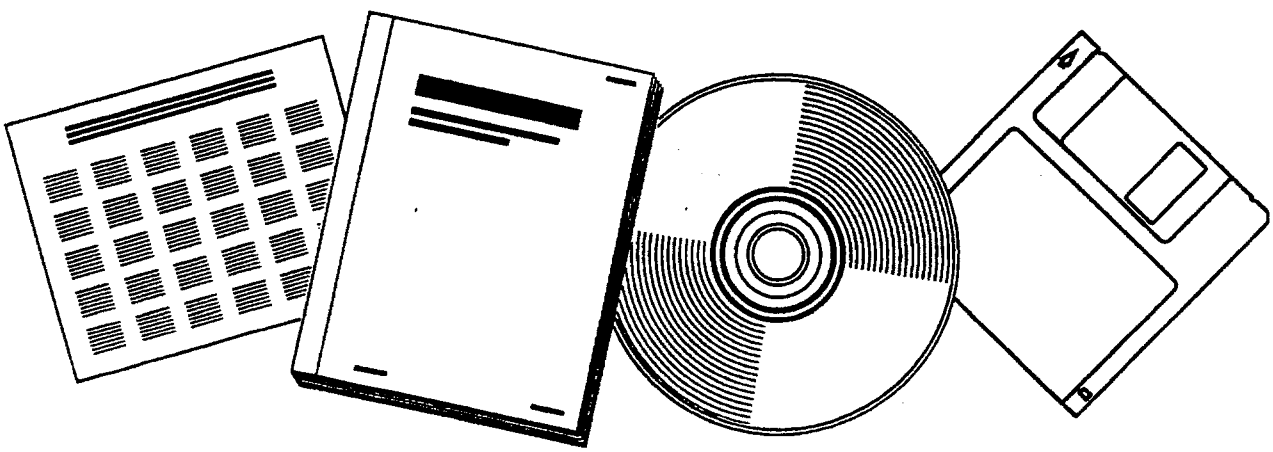

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Final Report

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For
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By
Bechtel Group, Inc.
San Francisco, California

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FINAL REPORT

SLURRY REACTOR DESIGN STUDIES

DOE Project No. DE-AC22-89PC89867
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SLURRY VS. FIXED-BED REACTORS FOR FISCHER-TROPSCH AND METHANOL

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JUNE 1990

EXECUTIVE SUMMARY

Background

The objective of this study was to set the groundwork for further development of indirect coal liquefaction technology via Fischer-Tropsch (F-T) or methanol synthesis. In particular, it was desired to know exactly how the slurry reactor concept could be used to best advantage. In the slurry reactor, developed in Germany in the 1950's, powdered catalyst is suspended in a heavy oil - the heavy end of Fischer-Tropsch product, for example - and the synthesis gas is bubbled through the mixture at reaction temperature and pressure. The reactants dissolve in the oil and react over the catalyst. Agitation from the gas flowing through the column - as in a bubble column reactor - provides for good mass transfer and heat transfer characteristics. The heat of reaction can be removed by cooling tubes inserted into the liquid.

The slurry reactor has been proposed for Fischer-Tropsch operations in the wax producing mode, which is preferred from a selectivity standpoint. Production of light ends is minimized and the heavy wax portion of the product can readily be upgraded to useful products. Fluidized-bed reactors cannot operate in this mode. Tubular-fixed-bed reactors have been in operation at Sasol for many years for wax production and will be used by Shell in their Middle Distillate Process being installed in Malaysia. A key question, therefore, is how does the slurry reactor stack up against the tubular-fixed-bed reactor?

This study indicates that a key advantage for the slurry F-T reactor is its ability to convert the low H_2/CO ratio synthesis gas (0.7 ratio or less) produced by coal gasifiers without ratio adjustment. An iron based Fischer-Tropsch catalyst can accept such a gas, converting it to liquid hydrocarbons with CO2 as the primary byproduct. The tubular-fixed-bed reactors cited above have been operated on 2.0 H_2/CO ratio gas and produce water as the byproduct. They would not be expected to be able to use a low ratio gas because of carbon formation. The main product from this study, therefore, is a cost comparison of slurry F-T reactors operating on low ratio gas versus fixed-bed F-T reactors operating on high ratio gas after composition adjustment. Designs have been prepared for those sections of a coal-based Fischer-Tropsch plant affected by reactor selection, equipment sized and costed with particular attention to the reactors themselves, and operating costs examined.

The slurry reactor has also been proposed for the synthesis of methanol and mixed alcohols. Chem Systems developed the original concept and Air Products has been piloting the so-called liquid phase methanol (LPMeOH™) process at LaPorte, TX, and now is designing a demonstration unit for Great Plains as part of Clean Coal 3. A specific niche has been identified for the slurry reactor in the coproduction of methanol and electric power in a combined cycle operation. A low conversion, once-through operation is used with low ratio gas as produced in the gasifier. Unconverted gas is directed to gas turbines for power generation. Methanol can be stored and used for supplemental firing of the gas turbines or sold.

Since the coproduction type of operation has been well studied and since it was not certain how to design a tubular-fixed-bed reactor for such an operation, attention was directed to ascertaining how well the slurry reactor would compete costwise with the fixed-bed reactor in a conventional, high yield methanol plant design with recycle. It was recognized that this would probably not be the optimum application for a slurry reactor, with its superficial velocity limitation, but it was felt that something could be learned about its preferred range of applicability. A brief look was also taken at mixed alcohols operation, using Lurgi's Octamix™ process as a model.

Reactor Design Criteria

Before attempting to perform an economic comparison, it was necessary to develop a rational basis for reactor comparison. Reaction kinetics, mass transfer, heat transfer and hydrodynamics were examined and effects of operating variables such as superficial velocity, slurry concentration, temperature and pressure were determined. It is necessary to allow for the effect of slurry concentration on mass transfer, for example, and this report provides a basis for doing so. A consistent process design basis was also developed based on the use of the Shell gasifier. This effort is the subject of Sections 2 through 5 of the report. This material should be of value to the DOE in setting the basis for the proposed baseline economic evaluation of advanced Fischer-Tropsch technology (RFP No. DE-RP22-90PC90027).

Scale-up of the slurry F-T reactor has been the subject of numerous technical articles. A high conversion per pass is preferred since recycle of unconverted syngas reduces the production from a slurry reactor, which has a superficial velocity limitation. Conversions of 90% or more have been demonstrated in high L/D pilot plant equipment but backmixing in a commercial reactor will limit the conversion which can be achieved. This study has taken a conservative approach by assuming that complete backmixing will occur and limits conversion per pass to 80%.

On the other hand, it has been assumed that superficial velocity and catalyst slurry concentration can be taken well beyond levels which have been demonstrated in F-T pilot plant operations to date. There is good reason for this since hydrodynamic studies sponsored by the DOE (e.g. Contract No. DE-AC22-86PC90012) have demonstrated reasonable gas holdup and gas dispersion under such conditions. Air Products' development work with the LPMeOH™ process in the LaPorte pilot plant is also considered very significant.

Conclusions

The primary conclusion from this study is that the slurry reactor has both advantages and disadvantages and that proper applications must be sought. Coal-based Fischer-Tropsch, as described above, appears to be such an application. It was found that, in a project producing 20,000 BPSD of Fischer-Tropsch products from 7500 TPD of moisture free Illinois No. 5 coal, plant investment can be reduced by $91 MM if the process scheme using slurry reactors is employed. This is a savings of about 8.5% on the total plant investment. The savings are roughly equally divided between the reactors themselves and the process simplifications resulting from the use of low H_2/CO ratio gas.

The fact that the slurry reactor can be operated continuously at the end of run temperature required for the fixed-bed reactor proved a significant advantage and permitted operation at roughly the same space velocity despite the fact that conversion was much higher (80% per pass versus 37% per pass). This enabled the use of 6 slurry reactors for the same capacity as 8 fixed-bed reactors, despite an intrinsically lower catalyst loading.

In order to gain these reactor savings it is necessary to design to an inlet superficial velocity of 0.14 to 0.15 m/s (0.46 to 0.49 ft/s) and a slurry concentration of 35 wt%. Typical pilot plant operations have been at about half these values. If the more conservative approach of using demonstrated pilot plant conditions is taken, the number of reactors increases from 6 to 11 and the cost of the reactors approaches that of the fixed-bed system. The net savings in investment reduces to $52 MM. There is thus a considerable incentive to demonstrate the higher levels of velocity and concentration.

There is even the possibility of going still further. It is understood that Air Products is designing the Great Plains demonstration unit for a superficial velocity of 0.24 m/s (0.8 ft/s). Slurry concentrations higher than 35 wt% are also possible. Again, the methanol system has been run up to 45 wt% slurry but under these conditions mass transfer resistance becomes a significant factor. This should not be true in the case of Fischer-Tropsch reactors which operate at only about one third the space velocity of a methanol reactor.

Operating cost was found not to vary greatly between the two reactors provided the slurry reactor catalyst is assumed to have an equivalent 60 day life (continuous replacement would be used) and the fixed-bed reactor catalyst, a life of one year. There is very little basis for either of these replacement rates and this is an item for further investigation. It was more difficult to balance energy requirements in the fixed-bed case leading to higher fuel gas requirement, but this is at least in part due to the use of the Shell gasifier. The Texaco or Dow type of gasifier would fit better into the fixed-bed processing scheme whereas the Shell gasifier appears a good choice for the slurry case..

For a conventional recycle methanol application the situation is reversed. The slurry methanol synthesis loop, at $41 MM for 1640 TPD of production, is almost twice as expensive as a tubular-fixed-bed system. The reasons are apparent when the design conditions are examined:

- In order to achieve design production from the slurry reactor, pressure is raised to 100 atmospheres. This reduces recycle requirement to a minimum and permits higher mass flows at a given superficial velocity.
- Using a stoichiometric feed gas it is possible to run the fixed-bed reactor at 55 atmospheres. Pressure drop is a limitation, but Lurgi assures that the design capacity can be produced.
- Space velocity is roughly the same for both reactors per unit weight of catalyst present. Because of the lower catalyst loading per unit of reaction volume, the slurry reactor is over twice the height of the fixed-bed reactor.
- The entire shell of the slurry reactor must be designed for reaction pressure of 100 atmospheres. With the fixed-bed reactor only the heads and tube sheets need be designed for reaction pressure. Thus even if the fixed-bed operating pressure were 100 atmospheres, shell weight would be less.
- The combined effect of the above is to negate the lesser tube weight of the slurry reactor and produce a more massive and costly vessel.
- Finally, feed gas compression is required, whereas the fixed-bed reactor can operate at the pressure level available from a Texaco gasifier.

No conclusions can be drawn from this study concerning once-through methanol operations. Without recycle, pressure can be reduced in half, essentially cutting the weight of the slurry reactor in half. The comparison would then depend on what design conditions can be developed for the fixed-bed reactor operation.

It was not possible to design a slurry reactor mixed alcohol plant without a better feel for what limits conversion. Lurgi requires 100 atmospheres for the fixed-bed operation with quite a low space velocity. Production is 460 TPD from the same size reactor used for 1640 TPD of methanol production. The high pressure was stated to be essential. This being the case, it does not appear that this is a good application for the slurry reactor in its present configuration

RECOMMENDATIONS

Further development of the slurry reactor for the Fischer-Tropsch application can be recommended without qualification. It appears to have intrinsic cost advantages over the fixed-bed reactor for this application and is more amenable to further improvement. The reactor is not easy to scale-up, however, and further experimental pilot plant work is recommended to demonstrate operation at the design conditions used in this study in a reactor of sufficient size that axial dispersion effects can be determined. Conversion of the LaPorte reactor to Fischer-Tropsch synthesis should be possible and is recommended.

A number of design needs are listed in Section 5 of this report. Foremost among these are the demonstration of backmixing effects, the possibility of reducing backmixing by use of baffles, possible advantages of higher conversion levels than were used in this study and better experimental definition of the effects of pressure, superficial velocity and catalyst concentration.

Reactor modifications that will limit backmixing and give higher conversion may be worth pursuing. A better feel for the economics could be gained by an analysis of the two extreme models, plug flow and complete backmixing. This would define the incentive for further development efforts. It is also noted that, when operating conditions are chosen to maximize capacity or new more active catalysts are developed, the number of cooling tubes increases to the point where alternate reactor designs with external cooling may once again become worthy of consideration.

Finally, it is felt that this study represents a good first step towards DOE's proposed baseline study on indirect liquefaction. It is recommended that the design assumptions used here be carefully reviewed and used for the definition of design conditions for that study.

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