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NOVEL REACTOR CONFIGURATION FOR SYNTHESIS GAS
CONVERSION TO ALCOHOLS

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QUARTERLY REPORT

July 1, 1991 - September 30, 1991

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I. Objectives for the Fourt Quarter, Year 2:

Our objectives during this quarter were to complete construction and shake down runs on our new reactor assemblies, obtain activity data on the catalyst employing the slurry reactor, incorporate pressure drop and change in number of moles options into our reactor simulation code and start experiments in the trickle bed reactor.

II. Accomplishments, Fourth Quarter, Year 2:

We have completed shake down runs in our 100 cc slurry reactor and completed two long period on line studies using different batches of catalyst. The results in terms of methanol productivity versus time on stream is given in Figure 1 for both runs. In the first run, 359 hours on stream, the oil in the slurry was not circulated and additional oil was added to the reactor periodically in order to compensate for oil lost from the system. In the second run, 200 hours, the oil in the reactor was circulated continuously. Although there were large differences in the initial activity of the catalyst for both runs, which may be due to slight differences in the reduction procedure of the catalyst, after about 50 h on stream the activity for both catalysts are about the same within experimental error. The activity of the catalyst, when normalized to the initial activity, is presented in figure 2. The activity function derived is used to correlate the data and to correct the data on variable space velocities for slight activity changes due to time on stream.

As mentioned in the previous Quarterly Report, we started Task V which involves simulation of the design equations presented in the Quarterly Report for 1/90 - 3/90 (Task IIIA). By quasilinearization combined with orthogonal collocation, the model equation set is transformed into a set of linear

algebraic equations and solved. In the previous Quarterly report we have indicated that gas-liquid and liquid-solid mass transfer coefficients do not have any effect on the reactor performance and that low effective diffusivities will reduce the productivity and conversion. Most significant parameter was the particle size, which affected both the productivity and conversion.

We have also studied the effect of partial wetting of the catalyst particle on reactor productivity and conversion. As shown in Figure 3, the wetting effectiveness is not important at low space velocities in both small and large particles indicating that effective diffusion into the catalyst pores will control the mass transport phenomena. Figure 4 shows that at high space velocities external wetting has an effect which is more pronounced for small diameter particles. This is consistent with our simulations presented in the previous Quarterly Report. Particle diffusion effects are more pronounced for larger particles due to the length of the diffusion path, hence it is the controlling mass transfer mechanism whereas for small particles effectiveness factor is closer to unity and for complete wetting external mass transfer may have a slight effect.

The review panel that reviewed our project suggested that we incorporate pressure drop calculations and how these affect the performance of the reactor especially for methanol synthesis since there will be a net change in the number of moles in the gas phase. We have incorporated that in our simulation code using the Leva Equation and performed simulations for two cases, a 2 inch diameter (~ 5 cm) reactor of varying length from 25 cm to 100 cm and a hypothetical 1 m diameter reactor with length 2.5 m to 10 m.

Figure 5 shows our simulations for the 2 inch reactor. The results

are obtained two ways, by neglecting the pressure drop and by considering the pressure drop. In this size reactor, the pressure drop does not affect the reactor performance. The productivity simulations at lengths 50 cm to 100 cm are very close to each other indicating that axial dispersion is not very important. The decrease in productivity for reactor of 25 cm length is due to the backmixing effect. In these simulations particle diameter was set at 3 mm. Figure 6, on the other hand, indicates that the pressure drop would have an effect at high space velocities and long reactors if the diameter is 1 m. The dashed line in the figure is for the case where pressure drop is neglected and as expected the same values are obtained at all space velocities for any reactor length, i.e the conversion and the productivity are functions of the space velocity only. For a reactor of 2.5 m, when the pressure drop is included in the simulation, we do not see any effect of the pressure drop. In fact even for a reactor of 5 m length the effect is seen at space velocities in excess of 10,000. However, longer reactors will be affected by the pressure drop resulting in a decrease in productivity. The effect is more pronounced at high space velocities.

III. Problems Encountered, Fourth Quarter, Year 2:

We originally planned to immerse our trickle bed reactor in a heating oil which is electrically heated. The oil was supposed to serve as a heat source and sink at the same time enabling us to maintain constant temperature in the reactor and also due to high natural convection in the liquid phase (the heating oil) we were expecting to avoid temperature fluctuations in the reactor due to heater control. However, the oil we used caught fire twice and fire control personnel had to be called to put out the fire. Although there

was no damage to property or personnel, due to unacceptable safety of the design, we modified the heating section of the assembly. We now immerse the reactor in a volume filled with aluminum shots that has air circulation and is heated electrically. We finished our long duration no reaction hot flow studies and were able to maintain the reactor at constant temperature, $250 \pm 0.5^{\circ}\text{C}$, for a long period on stream using the mineral oil that will be used in the reactor together with nitrogen. We hope to start our trickle bed experiments very shortly.

Methanol Productivity versus Time on Stream

FIGURE 1:

Relative Activity versus Time on Stream

FIGURE 2:

FIGURE 3

CH₃OH Productivity and H₂ Conversion versus Wetting Coefficient

MD2: WHSV= 6,000 l/hr.kg-cat

FIGURE 4

CH₃OH Productivity and H₂ Conversion versus Wetting Coefficient

MD2: WHSV= 20,000 l/hr.kg-cat

Figure 5

**Figure 6 Methanol Productivity
for $dr=1m$ and $dp=3mm$**

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