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FTIR STUDIES OF HYDROCARBON SYNTHESIS ON PD/ZSM5 CATALYSTS. QUARTERLY PROGRESS REPORT

NOTRE DAME UNIV., IN. DEPT. OF CHEMICAL ENGINEERING

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FTIR Studies of Hydrocarbon Synthesis on Pd/ZSM5 Catalysts

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BRIEF SUMMARY OF PROGRESS

The main objectives of the work performed during the last quarter were: 1) To prepare new catalysts and reproduce the high selectivity to oxygenates obtained with a Pd/ZSM5 catalyst existing in our laboratory.

 To study² the effect of catalyst pretreatment on the activity and selectivity.

SUMMARY OF PROGRESS

Studies summarized in our previous report indicate that the activity and selectivity of the Pd/ZMS5 catalyst depended on the cation exchanged on the ZMS5 support. Efforts to reproduce these results, which indicated that the Na exchanged ZSM5 was the only form of the support to produce dimethyl ether, were unsuccessful when using a low temperature pretreatment, with all catalysts producing mainly C_1-C_3 hydrocarbons.

The above results led us to examine carefully the pretreatment procedures used in the literature. It was concluded that the dispersion of Pd was a critical factor affecting selectivity. Consequently, a pretreatment procedure combining high temperature oxidation (600°C, 2 hrs.) and high temperature reduction (500°C, 2 hrs.) was adopted on the basis that it will increase the dispersion of Pd on the ZSM5 support.

The results shown in Fig. 1, indicate that, as previously reported, the initial rate of dimethyl ether (DME) production was an order of magnitude higher than the methanation activity. Fig. 1 also shows that the DME activity decreases steadily and in fact it is negligible after 60 hrs. of time on stream. The total activity (Fig. 2), and the methane activity, decreased by about 50% in the same period. The decrease in DME production was compensated by an increase in C_3^+ hydrocarbons.

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The high temperature pretreatment had an adverse effect on the Pd/La-ZSM5 catalyst. As shown in Fig. 2, the activity of this catalyst, previously found to be higher than the activity of Pd/Na-ZSM5, is now about an order of magnitude lower. However the activity of the Pd/La-ZSM5 increased slightly with time on stream. Work is underway to rationalize these findings.

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Figure 1 Yields vs. Time on Stream





Total Activity vs. Time on Stream

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