478b Synthesis of Dimethyl Carbonate, Dimethoxy Methane, and Methyl Formate on Cu-Y and Cu-Zsm-5

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Copper-exchanged zeolites have been reported to be active catalysts for the oxidative coarbonylation of methanol to dimethyl carbonate (DMC). Depending on zeolite structure and level of exchange, dimethoxy methane (DMM) and methyl formate (MF) are often observed as by products. In the present study, Cu-Y and Cu-ZSM-5 were prepared by "dry" exchange using CuCl vapor. The as-prepared catalysts were characterized by XRD, TPR, and Al and Cu XANES and EXAFS. The results of these analyses show Cu to be present exclusively as Cu+ cations. At high space velocity and methanol conversions approaching zero, the only products formed are DMC and water. As the conversion of methanol is increased, the yields of DMM and CO2 increase, while the yield of DMC increases. A small increase in the yield of MF is also observed. These results are attributed to the formation of H2 via the water-gas-shift reaction and subsequent hydrogenation and hydrogenolysis of DMC to form DMM and MF respectively. Differences in the activity and selectivity of Cu-Y and Cu-ZSM-5 will be discussed.