68a Olefin Epoxidation on Silver: from Mechanism to Catalyst Design

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Recent work from this laboratory has shed light on the mechanism of olefin epoxidation. From ultra-high vacuum (UHV) experiments and density functional theory calculations we have isolated the key oxametallacycle intermediate responsible for the formation of ethylene oxide. Also identified were the two transition states that govern the selectivity of the process. DFT calculations showed that the transition state for ethylene oxide was more favorable on a copper-silver alloy than on silver, relative to the transition state leading to acetaldehyde and further combustion. The results of the computational studies were validated by reactor studies. Experiments were conducted using a continuous flow reactor system, and employed ceramic foam monoliths as catalyst supports. The reactor data clearly showed that supported Cu-Ag bimetallic catalysts are more selective to EO than supported silver catalysts over a wide of ethylene and oxygen feed pressures and stoichiometries at similar ethylene conversions. The addition of copper to the catalyst surface was also found have an additive effect with traditional promoters such as cesium and chlorine with respect to enhanced EO selectivity. This example shows how fundamental studies can be extremely valuable in the design of catalytic materials.