

CHAPTER VIII

CONCLUSIONS AND RECOMMENDATIONS

8.1 Conclusions

Among all of the investigated oxide supports (Al_2O_3 .Acid, Al_2O_3 C, α - Al_2O_3 , Al_2O_3 .Neutral, Al_2O_3 .Fused, SiO_2 90, SiO_2 380, MOX 90, MOX 380, TiO_2 P25, nano SrTiO_3 , TiO_2 (sol-gel), SrTiO_3 (sol-gel), MgTiO_3 (sol-gel), CaTiO_3 (sol-gel), and BaTiO_3 (sol-gel)), SrTiO_3 was the most effective support for ethylene epoxidation, giving unusually high selectivity and yield of ethylene oxide. The sol-gel method was used to synthesize mesoporous-assembled titanate supports (TiO_2 , MgTiO_3 , CaTiO_3 , SrTiO_3 , and BaTiO_3). The support calcination temperature was found to have a significant effect on the physical and textural properties, affecting the catalytic activities towards ethylene epoxidation of all synthesized catalysts. The optimum calcination temperature of the support was 650 °C for all supports, except for TiO_2 which the optimum calcination temperature was found to be 600 °C. Among Ag catalysts, the 17.16 wt.% Ag/ SrTiO_3 catalyst gave the best catalytic activity to produce ethylene oxide with the highest EO yield of 4.7 % and the maximum EO selectivity of 99.12 % at a reaction temperature of 275 °C. The superior performance of this catalyst may be related to the moderate Ag particle sizes together with its high oxygen and ethylene adsorption ability as compared to the Ag catalysts loaded on the other supports. Moreover, Sr atoms that are incorporated in the SrTiO_3 support play an important role in making this catalyst more selective in EO formation.

The effect of loaded metals both monometallic (Ag and Au) and bimetallic (Au-Ag, Cu-Ag, Ba-Ag, Pd-Ag, and Sn-Ag) on the SrTiO_3 support were studied. The bimetallic Cu-Ag catalyst showed the significant improvement of the catalytic performance as compared with the traditional monometallic Ag catalyst. Furthermore, tin promoter was added in this catalyst to enhance the long-term stability. The 1.39 wt.% Cu-17.16 wt.% Ag/ SrTiO_3 with 0.32 wt.% Sn promoter was found to be the best catalyst for the ethylene epoxidation reaction, providing the highest EO yield of 5.2 % and the maximum EO selectivity of 95.1 % at 72 h of time on stream with the best long-term stability. This result might correspond to the high

oxygen and ethylene uptakes and the maximum fraction of metallic Ag. Moreover, addition of tin significantly reduced the amount of coke formation on the spent catalyst and lowered the catalyst sintering as well as less formation of Ag₂O and Cu₂O species, leading to the superior long-term stability.

The best reaction conditions for the epoxidation of ethylene, including diluent gas were also investigated. The optimum condition for the ethylene epoxidation reaction was using the bimetallic 1.39 wt.% Cu-17.16 wt.% Ag/SrTiO₃ catalyst with 0.32 wt.% Sn promoter, 6 % O₂ and 6 % C₂H₄ as a reactant feed in the 35 % CH₄ balanced with He as a diluent gas, a space velocity of 6000 h⁻¹, a pressure of 24.7 psia, and a reaction temperature of 275 °C. It should be noted that the CH₄ balanced with He as a diluent gas enhanced the ethylene conversion and less CO₂ formation (higher EO selectivity).

8.2 Recommendations

- To set up a high pressure operating system and compare the experimental results with this study.

- To modify the existing reaction line to have the recycling system of the reactant feed to obtain a higher ethylene conversion.

- To change O₂ in the reactant feed to N₂O to compare the catalytic performance in terms of EO selectivity and yield.

- To investigate the effect of other promoters i.e. Cs, Cl, Re, etc. on the improvement of the catalytic activity and long-term stability towards ethylene epoxidation.

- To further apply the 0.32 wt.% Sn-promoted on 1.39 wt.% Cu-Ag/SrTiO₃ catalyst to the epoxidation reaction of other olefins.

- To apply the optimum catalyst in the pilot scale test.

- To run the experiments for longer time on stream and regenerate the spent catalysts.