CHAPTER V
CONCLUSIONS AND RECOMMENDATIONS

Based upon previous works, low surface area Al_2O_3 is to be good for the ethylene epoxidation reaction. Several attempts have been done unsuccessfully to develope high surface area Al_2O_3 as support for Ag catalysts to produce ethylene oxide. In this research work, high surface area fumed Degussa C alumina as support for Ag catalysts was investigated because its non porous property is believed to give large particles and a good distribution of Ag particle sizes which are essential for the epoxidation reaction. This non porous support material provided large Ag particles, around 21 nm for metallic Ag and 30 nm for silver oxide which enhanced the ethylene epoxidation reaction. The formation of ethylene oxide was confirmed by DRIFTS experiment. Therefore, it can be stated that Ag on high surface area alumina can promote ethylene epoxidation provided that the Ag particle size is large. Moreover, high oxygen coverage on silver catalyst increases ethylene oxide activity.

Addition of gold as promoter on Ag/Al_2O_3 catalysts was found experimentally to enhance the ethylene oxide selectivity. It is believed that gold can create single silver sites that favor molecular oxygen adsorption. In other words, gold can reduce the dissociation of molecular oxygen on Ag sites, lowering the concentration of atomic oxygen. In addition, the interaction between Ag and Au causes a weakening of the adsorption bond strength between silver and oxygen. In this work, 0.54% Au on 13.18% Ag/Al_2O_3 gave the highest ethylene epoxidation activity.

It has been known that gold catalyst is good for low temperature CO oxidation, water-gas shift reaction, propylene epoxidation and photocatalytic reaction. Thus, Au/TiO_2 and Au/CeO_2 with different methods of preparation were studied. From the results, the gold particle size of Au/TiO_2 influenced the ethylene epoxidation and the suitable gold particle was found to be around 4 nm. It is known that oxygen species located at the perimeter interface are mostly molecular oxygen which favors to ethylene epoxidation. If the particle size of Au was less than 2 nm, the epoxidation reaction would not occur. The different methods of catalyst preparation gave different gold particle sizes. Au/TiO_2 with impregnation gave the

highest activity due to its optimum particle size. For Au/CeO_2, the results showed that it gave poor activity, both in terms of conversion and selectivity even though the gold particle size was large (6 nm). It may be due to the physical property of CeO_2 as oxygen storage material with high oxygen mobility.

The mechanism of adsorbed oxygen should be studied by using isotope O^{18} and O^{16}. Thus, the evidence of atomic and/or molecular oxygen adsorbed on this high surface area supports such as Al_2O_3, TiC_2 and CeO_2 can be verified. Moreover, the reaction mechanism of ethylene with the adsorbed oxygen species can be clarified.

Both Ag and Au particle sizes influence significantly on ethylene epoxidation. Therefore, catalysts with a wide range of particle sizes should be prepared by varying the calcination temperature. As known, a high temperature can induce the agglomeration of the metal particle to form a bigger size.