

CHAPTER VI

CONCLUSIONS AND RECOMMENDATIONS

In this thesis, the zeolite catalysts were investigated for the selective reduction of nitric oxide with hydrocarbon and excess oxygen. The brief conclusions of this study are as follows;

1. Butane was the most active reductant for NO reduction with the saturated hydrocarbons on Cu/Na-ZSM-5 catalyst in the presence of excess oxygen compared with propane, ethane, and methane. For hydrocarbon saturation studies, ethylene was a more effective reductant than ethane but it is in contrast for the case of propylene and propane. The low effectiveness of propylene was due to the formation of carbonaceous deposits on catalyst surface.

2. For the oxidation of various hydrocarbons, methane was found to be the most difficult hydrocarbon to oxidize and propylene was the easiest. In general, an increase in carbon number resulted in a decrease in the required temperature for a given conversion. For a given carbon number, the required temperature for a given conversion decreased with degree of saturation.

3. The Al-free catalysts as Cu-incorporated and Cu-exchanged zeolites showed nearly the same catalytic activity whereas 0.2%Cu/Na-ZSM-5 showed higher activity.

4. The large decrement of Si/Al ratio in H-ZSM-5 increased the NO and C₃H₈ conversions for the reduction of NO with C₃H₈ under excess oxygen

condition whereas the NO and C₃H₈ conversions were not affected by a little different in Si/Al ratio.

5. The higher copper content on Cu/Na-ZSM-5 catalyst showed the higher NO reduction and C₃H₈ combustion while Na-Cu-Al-silicate showed the opposite results that the higher copper content on the catalyst exhibited the lower activity. However, Na-Cu-Al-silicate with a small amount of copper showed higher activity compared with Na-Al-silicate.

6. H-form of Cu-exchanged ZSM-5 and Cu-Al-silicate showed the similar results of copper content for NO reduction. The catalysts without copper performed higher activity than that with copper. For copper catalysts, the catalytic activity and thermal stability increased with increasing copper content on the catalysts.

7. The catalyst with copper ion-exchange and incorporate, Cu/Na-Cu-Al-silicate, showed the both advantages of Cu-exchanged and Cu-incorporated catalysts those were the high activity and broad temperature window.

8. Metal ion-exchanged zeolite and metallosilicate catalysts prepared by the rapid crystallization method also have the pentasil pore opening structure. The morphology of the catalysts were approximately spherical shape. In addition, pore size distribution of the catalyst was affected by the metal loading on the catalyst while the total surface area and the most pore size of the catalyst were not.

9. The results of pore size distribution, BET surface area, and total pore volume of spent and fresh catalysts indicate that the catalyst as 35%Cu/Na-ZSM-5 which formed coke on its surface during the reaction showed the higher results compared with fresh catalyst and coke had its own porous structure like the porous system of the catalyst. For Na-Cu-silicate, the catalyst which

performed a little activity showed the decrement of pore size distribution, BET surface area, and total pore volume because the pore structure collapsed by thermal decomposition of the catalyst.

10. From the result of CO₂ TPD studies, it was found that only Cu-exchanged ZSM-5 zeolite showed a sharp peak of CO₂. The increment of copper content in the catalyst increased the CO₂ desorption peak. It can be concluded that copper ion site can adsorb CO₂ and the correlation between cumulative CO₂ desorption and the content of copper ion-exchange (wt.%) can be considered as non-linear correlation.

From this research, the following recommendations for further study are as follows;

1. Develop the copper ion-exchanged copper aluminosilicate catalyst for NO reduction with propane and excess oxygen.
2. Study the coke formation on the zeolite catalysts during the selective reduction of NO by hydrocarbon in oxidizing atmosphere.
3. Determine the phenomenon of temperature programmed desorption of carbon dioxide of the catalysts.
4. Investigate the effect of copper content on the catalytic activity for NO reduction over Cu-exchanged and Cu-incorporated aluminosilicate catalysts in more detail.