CHAPTER VII
CONCLUSIONS AND RECOMMENDATIONS

7.1 Conclusions

The catalysts prepared from sol-gel derived technique show the unique activity and selectivity over those prepared from the conventional methods. Gold catalysts show high activity for NO_x reduction by propene. They gave almost 100% NO_x conversion. However, their activities depend on the preparation methods. Among different catalysts derived from sol-gel technique, the one prepared from single step sol-gel method gave the highest activity while the others prepared from impregnation and deposition-precipitation method gave lower activity. However, the impregnated sol-gel alumina was the best catalyst in terms of overall performance which the formation of N_2 was concerned as well. It shows that catalyst preparation method has strong effects on the catalytic morphology resulting in different catalytic behaviors.

Generally, gold catalyst reached the maximum conversion around 400 ^{o}C. Its activity was also influenced by oxygen and water. Water considerably enhanced the catalytic activity. However, it depressed the selectivity towards N_2. Nitric acid, surface nitrates or organonitrosyl compound are possible by-products. These catalysts worked well in the large excess oxygen even to 14% which is equivalent to lean-burn condition in the exhaust stream. The higher amount of the oxygen improved the catalytic activity and enlarged the activity window. Gold catalyst also showed its stability for prolonged time on stream in both dry and humid conditions.

In the selective catalytic reduction of NO_x with urea, gold catalysts gave relatively fair activities over the experimental temperature range compared to their acitivities in NO_x reduction by hydrocarbon. The preparation method also affected to the activities due to the difference of their structures. Platinum catalyst also gave an average NO_x conversion. It reached the maximum conversion around 250-300 ^{o}C which was lower than that of gold catalyst. Oxygen was found to enhance NO_x conversion over both catalysts as well. In addition, poisoning condition with SO_2 had slight effect on both catalysts. The platinum catalyst was further studied with urea

delivered from aqueous solution. Under oxidizing condition, nitrogen-containing compounds derived from urea were oxidized to form NO, NO_2 and N_2O during the NO_x reduction. This results in negative NO_x conversion at high temperatures.

In summary, noble metal catalysts from gold and platinum give the promising performance in the selective NO_x reduction by both hydrocarbon and urea. This shows the possibility to develop these catalysts for NO_x removal in lean-burn condition which can be applied for both automotive and stationary exhaust clean-up.

7.2 Recommendations

This study shows that the preparation strongly influences on the activity of the catalysts as it affects their structures. Since sol-gel technique can give high surface area and is able to control other characteristics such as pore size, pore volume, surface acidity, etc. Therefore, the chemisty of sol-gel is an attractive issue for catalytic properties modification. Different precursors and procedures in sol-gel synthesis are suggested to employ in the catalyst preparation. Further investigation of catalytic behavior in order to understand the correlation between the catalytic activity and its physical and chemical properties will be beneficial in developing a novel catalyst.

In addition, studying in the selective NO_x reduction by urea should be extended as its chemistry over catalyst is still less literated. In fact, the NO_x reduction by urea is more complex than that by propene because it is indirect source of secondary reducing agent, ammonia. Urea itself originates several parallel reactions through its decomposition and polymerization. Moreover, its derivative products such as NH_3 and HNCO also undergo various reactions such as oxidation, hydrolysis and polymerization. These circumstances result in difficulty to understand the actual pathways in catalytic NO_x reduction. Therefore, urea decomposition and hydrolysis including other parallel reactions over gold and platinum catalysts are recommended to further investigate.