

## CHAPTER V

### CONCLUSIONS AND RECOMMENDATIONS

#### 5.1 Conclusions

The methylation of toluene with methanol over synthesized HZSM-5 with SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> ratio of ca. 200 and various modified methods of HZSM-5 catalysts were demonstrated in this work. The HZSM-5 catalyst has been successfully synthesized by hydrothermal condition based on TPABr chelating agent. The morphologies of those were irregular hexagonal prism with rectangular insertion and provide SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> molar ratios of synthesized HZSM-5 catalyst of 214. Chemical liquid deposition (CLD), dealumination (DeAl), and combination of CLD and DeAl modification methods were applied. For the silylated HZSM-5 modified by CLD method, its SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> ratio is considerably increased and significantly decreases of BET surface area and Brønsted acid site. For the dealuminated HZSM-5, its SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> ratio is increased and considerably increased the Lewis acid site due to extra-framework aluminum species, thus creation the mesopores. Furthermore, for the HZSM-5 modified by combined silylation and dealumination, their SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> ratios are increased. The micropore volume, total pore volume, and the Lewis acid site of CLD-DeAl-HZ5 are higher than DeAl-CLD-HZ5 but Brønsted acid site is lower. For catalytic activity, the silylated HZSM-5 with 1 ml/g cat (CLD(1.0)-HZ5) and that with the silylation-dealumination sequence (DeAl-CLD-HZ5) gave a comparable *p*-xylene selectivity of ca. 75 % and toluene conversion of ca. 8 % at temperature 400 °C, T/M molar ratio of 4:1, and WHSV 24 h<sup>-1</sup>. The silylation-dealumination sequence (DeAl-CLD-HZ5) catalyst was further studied to improve its *p*-xylene selectivity by various parameters. The results indicated that the DeAl-CLD-HZ5 provided the highest *p*-xylene selectivity around 80 % and decreasing of toluene conversion about ca. 4 % at 400 °C, a WHSV 40 h<sup>-1</sup>, and T/M molar ratio of 8:1.

## 5.2 Recommendations

In order to improve the catalytic activity of synthesized HZSM-5 for this reaction, methods of catalyst modification catalysts are important. Acid property modification such as the dealumination method should be used for the a lower  $\text{SiO}_2/\text{Al}_2\text{O}_3$  molar ratio than 200. The variations of temperature on reaction should be further considered.