

CHAPTER V

CONCLUSION

TS-1 catalysts were synthesized in basic condition using a gel composition of $\text{SiO}_2 : 0.025\text{TiO}_2 : 0.36 \text{TPA}^+ : 36.5\text{H}_2\text{O}$ by modifying the recipe reported by Thangaraj [7], using different types of template in pure TPAOH, pure TPABr, and mixture of TPABr and TPAOH, 1:1 mole ratio system were investigated. The gel was crystallization at 170°C in an oven designed to rotate the autoclaves at 60 rpm for 4 days. Upon the XRD measurement, the as-synthesized TS-1 prepared in the presence of all system, consists of only MFI phase. The particle size and morphology are affected by the type of template. The particle size increases when the amount of TPABr increases. Among these different templates the mixed template of TPAOH and TPABr provides the highest opportunity for titanium to locate at the tetrahedrally coordinating framework position of the MFI structure without formation of extraframework titanium species. The values of Si/Ti ratios were increases in the order of using TPAOH > TPABr > mixed templates. BET specific surface areas of TS-1 seems not different, but the trend can indicate the slight decrease in the order of using mixed templates > TPAOH > TPABr.

Ti-MWW catalysts were synthesized following the conventional procedure [6] using gel with a molar composition of $\text{SiO}_2 : 0.025\text{TiO}_2 : 0.67\text{B}_2\text{O}_3 : 1.4\text{HM} : 19\text{H}_2\text{O}$ and crystallization in an oven designed to rotate the autoclaves at 60 rpm. All Ti-MWW samples synthesized at different conditions for crystallization contained only the MWW phase as verified by XRD. The highest crystallinity was obtained by multistep crystallization at 130°C for 1 day, 150°C for 1 day and 170°C for 7 days. The SEM images of Ti-MWW revealed thin platelet crystals with a hexagonal morphology. The average size of the particles was 200-500 nm in length and 100 nm in thickness. The locations of titanium are in both tetrahedral and octahedral sites before acid treatment. After acid treatment of Ti-MWW, only tetrahedral site of titanium was observed along with the slight change of lamellar structure. Acid

treatment decreases the amount of Ti and B, but increases the BET and Langmuir types of surface area of catalyst.

The catalytic activities were studied in phenol hydroxylation using H_2O_2 as oxidant. The conversion of phenol is determined by the pore geometry, the external surface titanium sites, the particle size of the titanium-containing molecular sieve, the nature of the solvent, and reaction time. All TS-1 samples are active in the phenol oxidation. The conversion of phenol and the total product yield increase in the order of TS-1-Mix > TS-1-OH > TS-1-Br > Ti-MWW-acid > Ti-MWW. Ti-MWW led to a decreased conversion but an enhanced selectivity for p-benzoquinone. Water was the best solvent for all catalysts. The highest activity in water media and the use of H_2O_2 as oxidant make the process economic and friendly to the environment. In addition, the extension of reaction time from 1 h to 5 h increases the activity in both of the catalysts. The modification synthesized TS-1 mixture of TPABr and TPAOH, even it could reduce the production cost smaller than using only TPABr template, but the activity of catalytic properties of phenol hydroxylation was higher than the catalytic properties by using TPAOH in full level.

The suggestion for future work

- 1 Decrease amount of TPAOH to the lowest for synthesized TS-1 catalysts and test catalytic activity in phenol hydroxylation.
- 2 Delaminated Ti-MWW after acid treatment was producing the more external surface, which has higher active sites on the catalyst.
- 3 Co-solvents of organic solvent and water were interested to reaction of phenol hydroxylation.