

## CHAPTER VIII

### CONCLUSIONS AND RECOMMENDATIONS

Silatrane and alumatrane were synthesized and used as precursors for synthesizing MFI zeolite. Aging time of the reaction mixture prior to heating plays a very important role in the formation of MFI. Complete conversion of SiTEA to MFI crystals was obtained with longer aging times. However, heating time also has an effect on SiTEA conversion, a longer heating time resulting in a better conversion. Higher temperatures can be applied to curtail aging and heating times. As for the effect of  $\text{OH}^-$ ,  $\text{Na}^+$ ,  $\text{H}_2\text{O}$  and template concentration, less perfect crystals of MFI are obtained when increasing  $\text{Na}^+$ , decreasing  $\text{OH}^-$ , or decreasing TBA concentration. The shape of MFI crystals appears to depend on the level of water present. Two types of template, TPA and TBA, were studied to compare their effect on the MFI morphology. The TPA shows better performance by decreasing the aging and heating times while showing no significant influence on the Si/Al ratio. Differences in the morphology of MFI were also observed between TPA and TBA, which appear related to differences in steric hindrance of the templates during crystal formation. The Si/Al ratio in MFI zeolite increases linearly relative to the loading composition. Increase in alumatrane loading causes more difficulty for MFI crystal formation, however, the tendency for MFI formation is improved by increasing the aging and heating times.

Similar to the study silicalite and ZSM-5 synthesis, VS-1 zeolite was synthesized using silatrane as precursor and the factors influencing the properties of the product were studied. Lower water content with higher reaction temperature is the preferable condition for VS-1 synthesis due to higher promotion of vanadium condensation into the zeolite structure. The amount of sodium and vanadium loading strongly influences the amount of intrinsic vanadium species within the VS-1 zeolite. Lower sodium hydroxide concentration gives higher vanadium in the structure, with being less leached out, when compared to VS-1 formed using higher sodium hydroxide concentration. Therefore, high sodium hydroxide increases extrinsic vanadium formation and does not promote incorporation of vanadium into the framework structure. Samples prepared with high sodium hydroxide concentration

are susceptible to destruction during calcination. ESR spectroscopy at room temperature shows that the vanadium species in all samples are immobilized and well dispersed in the MFI structure. Thus, the silatrane precursor provides good potential for preparing VS-1 zeolite with high vanadium loading via the hydrothermal synthesis method.

The influence of synthesis conditions on the catalytic properties in oxidative dehydrogenation of VS-1 zeolite, synthesized using the silatrane precursor, shows that propane conversion increases with increasing vanadium content in the zeolite structure while selectivity slightly decreases. However, the temperature has a more dramatic effect on both conversion and selectivity. Product selectivity decreases with increasing temperature due to the dominant radical mechanism or non-catalytic reaction at high temperature, causing more production of carbon monoxide. High sodium hydroxide concentration generates less vanadium in the VS-1 zeolite structure, but results in higher conversion and selectivity of carbon monoxide, probably owing to the reaction at defect sites. The nature of the active site of vanadium in VS-1 zeolite was studied by comparing the reaction of as-synthesized versus ammonium acetate treated VS-1. Our observations are consistent with the expectation that tetrahedral vanadium within zeolite is responsible for the selective oxidative dehydrogenation of propane, while extrinsic vanadium or polymeric species are responsible for over-oxidation to give higher carbon monoxide and carbon dioxide selectivity. A shorter contact time gives higher product yield and conversion though lower in selectivity.

Many evidences from this research work and references support the concept of well dispersion vanadium on support responsible for oxidative dehydrogenation. Various kinds of support has been studied to prepare well define vanadium structure and to understand the effect support on ODH reaction. It has several evidences that support can enhance reaction selectivity. Various kinds of alkali and alkali-earth were used for increase the selectivity. It is possible that selectivity of oxidative dehydrogenation on VS-1 catalyst can enhance by doping several kind of alkali and alkali-earth to modify electronic structure of vanadium for oxidative dehydrogenation reaction, as well.