CHAPTER V

CONCLUSION

To study the effect of supporting materials on catalytic activities of tungsten oxide on metathesis reaction of 1-hexene, various supported tungsten oxide catalysts were synthesized by incipient wetness impregnation method. Using conventional amorphous supports such as alumina and silica-alumina were compared to the high surface area materials like HMS, Al-HMS and ZSM-5.

Al-HMS mesoporous support was successfully synthesized in neutral condition by modifying the method reported by Tuel *et al.*¹ and Rao *et al.*² The gel molar composition of $1.0 \text{SiO}_2 : 0.0125 \text{Al}_2 \text{O}_3 : 0.25 \text{HDA} : 8.30 \text{EtOH} : 100 \text{ H}_2 \text{O}$ was aged with stirring at room temperature for 20 h. This procedure can be applied for pure silica HMS without addition of aluminum source added in the gel. The zeolite type ZSM-5 support can be synthesized in basic condition by following the recipe reported by Böhlmann³ using a gel composition of $1.0 \text{SiO}_2 : 0.0125 \text{Al}_2 \text{O}_3 : 0.27 \text{TPABr} : 4.67 \text{NH}_3 : 26 \text{H}_2 \text{O}$ and crystallization at $180 \,^{\circ}\text{C}$ for 7 days. The WO₃/support catalysts were prepared by incipient wetness impregnation method and subsequent calcinations at the temperature of $500 \,^{\circ}\text{C}$ to convert the tungstate ion to supported tungsten oxide.

From characterization of supported tungsten catalysts, XRD patterns of bared HMS and ZSM-5 supports exhibited the typical structure of distorted hexagonal and MFI, respectively. ²⁷Al-NMR spectra indicated occupancy of both tetrahedral and octahedral positions by aluminum in case of the Al-HMS support. The Si/Al ratio in Al-HMS and ZSM-5 was determined by ICP-AES. The Si/Al ratio of 18 and 38 are found Al-HMS and ZSM-5, respectively. The crystallinity of bared HMS and Al-HMS supports is an inverse

function of tungsten loading, whereas for the MFI structure of ZSM-5 is not affected. No tungsten oxide crystallite is observed on HMS and Al-HMS even at high loading of tungsten. In addition, BET specific surface area of each support is reduced at high tungsten loading. DR-UV spectra indicate the presence monomeric and polymeric tungstate species on the catalysts. The phase of tungsten oxide on catalysts at high tungsten loading is tungsten oxide monohydrate as confirmed by Raman scattering spectra.

Metathesis of 1-hexene was investigated using various supported tungsten catalysts. Type of supports plays a significant role on the catalyst activity and the selectivity to gas products. At high temperature using, cracking reaction over acidic silica-alumina, Al-HMS and ZSM-5 becomes competitive to metathesis. It can be concluded that the 1-hexene conversion depends on temperature. However, at low temperature HMS which is very weak acidic promotes metathesis reaction. In addition, the tungsten content has significantly effect on the yield of gas products.

The suggestion for future work

- 1) To avoid the effect of acidity of support on the catalytic activities. The support with the same Si/Al mole ratio should be utilized.
- 3) Cross metathesis of butene and ethylene to produce propylene should be attempted for the industrial application.