CHAPTER I

INTRODUCTION

Incorporation of metals into catalysts has been used for many years for production of catalysts .at the present , Ag containing with several supports has been used for many catalytic process such as Ag/ Al_2O_3 ,Ag –ZSM-5 ect.

Several researches reported that Ag clusters were important active species for selective catalytic reduction of NO by hydrocarbon. Ag clusters increased NO conversion and increased the hydrocarbon combustion.[1,2,3,4]

Berthke *et al.*[1] reported that Ag/ Al_2O_3 catalyst has improved an activity for NO conversion in selective catalytic reaction with C_3H_6. But they found that when increasing Ag content about 2 wt% to 6 wt% .NO conversion decreased, on the other hand, C_3H_6 combustion increased .They concluded that the structure of Ag in 2 wt%Ag/ Al_2O_3 was Ag^{+1} while Ag in 6 wt%Ag/ Al_2O_3 were Ag^0 clusters. The presence of Ag^0 clusters increased C_3H_6 combustion while Ag^{+1} increased NO conversion.

Zeolites which contain clusters of different metals, is widely used as important industrial catalysts.

Zhijang Li *et al.*[2] has improved an activity of Ag-ZSM5 by adding cerium for selective catalytic reduction (SCR) of NO with methane in the presence of an excess of oxygen. They concluded that silver existing mainly as a dispersed Ag^+ ion in a low Ag-content and conversion of NO to N_2 was much higher than high Ag-content, while nanoparticles of silver of ~ 10nm size were found on the surface of high Ag-content. The dispersed Ag^+ state was more active for SCR reaction while silver particles effectuate more methane combustion reactions.

Shibata *et al.*[3] examined Ag clusters on MFI for selective catalytic reduction with propane in the presence of H_2. They reported that Ag-MFI after the

SCR reaction with propane by adding H_2. Ag^+ ions in Ag-MFI was converted to $Ag_n^{\delta+}$ cluster ($2 \leq n \leq 4$) and metallic Ag_m cluster ($3 \leq n \leq 5$). The formation of this Ag cluster increased NO conversion and increased C_3H_8 combustion.

Therefore, it is important to study the interplay of zeolite matrix and Ag clusters stabilizing in it and to find the parameters governing the Ag cluster formation during catalytic preparation. The numbers of parameters such as metal concentration, preparation conditions have been found to govern the physical and chemical of cluster embedded in zeolite. However the Si/Al ratio was not considered as a parameter for governing Ag clusters formation in zeolites. A few researches include preparation of clusters in zeolites with different SiO2/AlO3 ratio in Ag-mordenite. Bogdanchikova *et al.*[4] reported that the variation of SiO2/AlO3 molar ratio strongly changes a contribution of Ag clusters. However, the effect of Si/Al ratio of Ag-ZSM-5 catalyst on the silver cluster formation has not been studied.

In this study, we aim to examine the effect of Si/Al ratio of Ag-ZSM-5 catalyst on the silver cluster formation. The preparation of catalyst was characterized by X-ray diffraction (XRD), X-ray fluorescence (XRF), scanning electron microscopy (SEM), UV-Vis spectroscopy. The objectives and scope of this study will be described as follow.

1.1 The objective of this work

Studying the effect of silicon to aluminium ratio of Ag-ZSM-5 catalyst on the silver cluster formation

1.2 The scope of this study

1.2.1 Preparing ZSM-5 zeolite catalyst at different ratio of silica to aluminium Si/Al ratios of 20,22,28,32,60,110 and 150

Preparing Ag-ZSM-5 by ion exchange with $AgNO_3$

Preparing Ag clusters on Ag-ZSM-5 by reduced Ag-ZSM-5 in flow of H_2 at 300 °C

Studying the effect of silicon to aluminium ratio from (20-150) of Ag-ZSM-5 zeolite catalyst on metal cluster formation

1.2.2 Charcterization of prepared catalyst by the following methods

(a) Structure and crystallinity of samples by X-ray diffractrometer (XRD).

(b) Determination of chemical composition of catalysts by X-ray Fluorescence (XRF).

(c) Morphology of sample by Scanning Electron Microscopy (SEM).

(d)The acidity of catalysts was analyzed by Temperature programmed desorption of ammonia (NH_3-TPD)

(e) Characterization of Ag clusters in Ag-ZSM-5 by UV-Vis spectroscopy

The present thesis is arranged as follows:

Chapter II presents the literature reviews of investigation, synthesis, properties, and reaction of ZSM-5 zeolite.

Chapter III presents the theoretical consideration on ZSM-5 zeolite.

Chapter IV presents the experimental systems and operation procedures. The experimental results obtained from the laboratory scale and standard measurements are reported and discussed in chapter V.

The last chapter gives an overall conclusion emerging from this work. Finally, the calculation of ZSM-5 zeolite preparation.