CHAPTER I

INTRODUCTION

Beta zeolite is potentially an important catalyst and wide applications in petrochemical and chemical industries. It has been interesting potential applications in acid catalyzed reactions because of high thermal, hydrothermal and acid treatment stability, high strength acid sites and hydrophobicity [1].

Beta zeolite is a high-silica, large-pore zeolite with a three-dimensional channel system and 12–membered ring pore openings. It was first synthesized in 1967 by Wadlinger et al. [2] from alkaline aluminosilicate gels in the presence of sodium and tetraethylammonium cations.

Zeolites are mostly synthesized by hydrothermal crystallization under an autogeneous pressure and in the presence of a large excess of water. Numerous types of aluminosilicate, metallosilicate and aluminophosphate molecular sieves have been prepared by this method. Each zeolite has a definite structure and a unique pore size and most of them have limitations on their chemical compositions [3]. In 1990, Xu et al. [4] introduced a new technique. They converted a dry aluminosilicate gel in contact with steam and vapors of volatile amines into ZSM-5. Matsulata et al. developed a new crystallization method and named dry gel conversion (DGC) [5]. In this method, dried aluminosilicate gel powder containing organic template is crystallized to zeolites in the presence of steam. Hari et al. have shown that a new crystallization method, the dry gel conversion technique, is useful for the synthesis of beta zeolite with a wide range of SiO_2/Al_2O_3 ratio. This method has the following advantages over the hydrothermal crystallization method, which possible development of new structures, reduction of limitations on chemical composition, minimizing the waste, reduction of the consumption of expensive organic template and rapid crystallization [6]. Further studies on the crystallization of several types molecular sieves such as [Al]EU-1, [Ga]EU-1 [7], EMT [8], ZSM-5 [4], ZSM-22 [9], ZSM-48 [10].

Recently, Mintova et al. [11] studied the synthesis of nanosized ZSM-5 from amorphous silica grains by dry gel conversion. They have shown that the crystallinity, the average size of crystallites and mechanical properties of the nanozeolite depend strongly on the synthesis procedure. Arnold et al. [7] prepared EU-1 zeolite synthesized by dry gel conversion. They were found that the formation of EU-1 zeolite without impurities was obtained only optimal parameters of the dry gel synthesis of EU-1 zeolite that amount of water content in the autoclave during the crystallization process, the content of sodium cations and template molecule in the dry gel.

However, the study on beta zeolie was insufficient, because different kind species of beta zeolite were not available so far, and our study was limited to commercially available ones.

In this study, we have investigated the synthesized of beta zeolites by dry gel conversion with various water content as a source of steam, M_{gel}/M_{water} ratio and SiO_2/Al_2O_3 ratio. We shall show that effect of various their parameters on formation and characterization of beta zeolites. The resultant beta zeolites were characterized by X-ray diffraction (XRD), X-ray fluorescene (XRF), scanning electron microscopy (SEM), BET surface area (BET), nuclear magnetic resonance (NMR). Its catalytic performance for methanol conversion was also studies. The objectives and scope of this study will be described as follows:

1.1 The objective of this work

To study the synthesis of beta zeolite by dry gel conversion and its catalytic performance

1.2 The scope of this study

1.2.1 To study the synthesis of beta zeolites using dry gel conversion method with various parameter as follows:

1.2.1.1 Water content 5, 10, 20, 30, 50 ml

1.2.1.2 M_{gel}/M_{water} ratio 0.8, 1.5, 3.0

1.2.1.3 SiO_2/Al_2O_3 ratio 30, 60, 100, 200

1.2.2 Charcterzation 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 Fluorscence (XRF).

(c) Specific surface area by N_2 adsorption based on BET method (BET).

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

(e) Quantitative analysis of tetrahedral aluminium in samples by ^{27}Al nuclear magnetic resonance (^{27}Al NMR).

1.2.3 To investigate the performance of the prepared catalyst on methanol conversion by using N_2 for carrier gas under the following conditions:

1.2.4.1 Atmospheric pressure

1.2.4.2 SiO_2/Al_2O_3 ration in gel of 30-200

1.2.4.3 Reaction temperature 450°C

1.2.4.4 Space velocity 4000 h^{-1}

The reaction products were analyzed by gas chromatographs.

The present thesis is arranged as follows:

Chapter II presents the literature reviews of investigation, synthesis, properties, and reaction of beta zeolite.

Chapter III presents the theoretical consideration on beta 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 overall conclusion emerged from this work. Finally the calculation of beta zeolite preparation, calculation of percent relative crystallinity and calculation of BET surface area are included in appendices at the end of this thesis.