

CHAPTER I

INTRODUCTION

The classical definition of a zeolite is a crystalline, porous aluminosilicate. However, some relatively recent discoveries of materials virtually identical to the classical zeolite, but consisting of oxide structures with elements other than silicon and aluminium have stretched the definition. Most researches include all types of porous oxide structures having well-defined pore structures due to a high degree of crystallinity in their definition of a zeolite, the metal atoms (classically, silicon or aluminum) are surrounded by four oxygen anions to form an approximate tetrahedral consisting of a metal cation at the center and oxygen anion at the four apexes. The tetrahedral metals are called T-atoms, and these tetrahedra then stack in beautiful, regular arrays such that channels form. The zeolite channels (or pore) are microscopically small, and in fact, have molecular size and shape of the channels have extraordinary effects on the properties of these materials for adsorption processes, and this property leads to their use in separation process. Since silicon typically exists in a 4^+ oxidation state, the silicon-oxygen tetrahedra are electrically neutral. However, in zeolite, aluminum typically exists in the 3^+ oxidation state so that aluminum-oxygen tetrahedra form centers that are electrically deficient one electron. Thus, zeolite frameworks are typically anionic, and charge compensating cations populate the pores to maintain electrical neutrality. These cations are participated in ion-exchange processes. When the zeolite cations are protons, the zeolite becomes a strong solid acid. Such solid acids form the foundations of zeolite catalysism applications including the important fluidized bed cat-cracking refinery process. Other types of reactive metal cations can also populate the pores to form catalytic materials with unique properties. Thus, zeolites are also commonly used in catalytic operations and catalysis with zeolites is often called “shape-selective catalysis”.

Most zeolites used come from either nature or synthesis. Synthetic zeolites are obtained via the sol-gel process which amorphous gel is produced from an interaction between aluminate and silicate or silica sol. The sol-gel process involves two main reactions, hydrolysis and condensation reactions of precursor, such as

silicic acid, fumed silica, tetraethoxysiloxane (TEOS), sodium aluminate, etc. A three-dimensional gel network comes from the condensation of partially hydrolyzed species in which the degree of gelation impacts on the properties of the product. At the hydrolysis rate faster than the condensation rate, resulting in the gel structure is weakly branched, while in the opposite direction, the results obtained give highly branched and contained colloidal aggregates. To obtain the crystalline phase, further hydrothermal treatment is required. It can be processed by either conventional or microwave heating. In both techniques, the amorphous gel undergoes continual dissolution and reconstruction and crystalline phase grows under hydro-pressure. However, the nucleation rate in both techniques is different. In case of conventional heating, transferring heat from the generation source to media or materials deals with heat transfer coefficient of each material, causing convection current of heat or non-homogeneous heat distribution. As a result, the nucleation rate is very fast at the area of high temperature and very low at low temperature region. For microwave heating was considered as an alternative method to generate heat. It attributes fast homogeneous nucleation and easy dissolution of the gel due to the fast homogeneous heating and the formation of active water molecules. The microwave system shows many advantages over the conventional heat method, such as giving homogeneous heat for short period of time, rising temperature at very high rate even in large amount of samples. It is a fast and energy efficient technique, which prevents other side reactions owing to their nature in interaction caused from dielectric property of each material. Therefore, microwave technique is used to promote the reaction to go faster at required shorter reaction time. Our goaled zeolite, the MFI zeolite, can be successfully synthesized by microwave technique within few hours of heating.

The synthesis of MFI zeolite has become one of the increasing interests due to a variety of important applications. In particular, these zeolite can function either as a catalyst by itself or as a catalyst support. The latter can be done by addition of other metals, e.g. Fe, Ti, V, etc., as a catalyst into the zeolite structure, for example VS-1 containing V atom in the framework of MFI for oxidation reaction. The iron, our interest metal, has gained great interest in view of their excellent catalytic performance in various environmental applications and in selective oxidation for chemical production (Pérez-Ramírez and Gallardo-Llamas, 2004), however, gives

less surface area, resulting in less active site to interact with the reactant. The higher reactivity and more selectivity of product are obtained by the combination of high surface area together with regularity of the channel in the structure of zeolite.

Reduction of NO_x is of great of importance and interest in air pollution control especially in the high temperature combustion processes in automobile engines. Generally, NO is the major emission gas in these processes. However, CO is also found in ineffective combustion processes. The catalytic reduction of NO in the presence of CO has been widely studied, and it can be catalyzed by precious metals dispersed on various supports such as Pd on Al_2O_3 (Fernández-García *et al.*, 2004). However, due to the scarcity and high cost of these precious metals, Fe-MFI zeolite catalysts are of increasing importance in catalytic reduction of NO.

In the study, iron loaded-MFI zeolite was synthesized using synthesized silatrane as zeolite synthesis precursors and alkaline base as a hydrolysis agent and source of metal ions via the sol-gel process followed by microwave technique. Influence of the preparation method on the catalytic activity in the reduction of NO using CO as a reductant gas was investigated.