

CHAPTER I

INTRODUCTION

According to IUPAC, three types of pores can be present in a solid depending on d , the pore diameter, as follows: micropores have diameter less than 2 nm, mesopores have diameter between 2 and 50 nm and macropores have diameters greater than 50 nm (IUPAC, 1972).

Mobil's researchers successfully synthesized a new family of mesoporous silicate and aluminosilicate molecular sieves, designated as M41S. M41S including MCM-41 (hexagonal), MCM-48 (cubic) and MCM-50 (laminar) synthesized with liquid crystal templates possesses a well-order array of structure and a uniform pore size distribution engineered in the range from 15 to 100 Å. MCM-41, possessing a hexagonal arrangement of channels, is one of the most important member of the M41S family (S.H. Liu, H. Paul Wang, 2002). MCM-41 has attracted the attention of scientists due to its elevated specific surface area, high thermal and hydrothermal stability, possibility of controlling its pore size and its hydrophobicity and acidity. These characteristics have made MCM-41 a promising material as catalyst and/or support and to be used in industrial processes of adsorption, ion exchange and environmental control, hosts for the inclusion of compounds and nanosize clusters, and molecular sieves for large molecules (Ryong Ryoo, 1997). The wide and uniform uni - dimensional channels of MCM-41 allow a faster diffusion of larger organic molecules than the microporous zeolite and aluminophosphate - based materials with channels and cavities only up to 15 Å in diameters.

MCM-41 can be synthesized following a wide variety of preparation procedures. However, there is one thing that all these procedures have in common next to the obvious presence of a silica source, *viz.* a templating agent. Actually, there are 2 types of surfactants, supramolecular templating and liquid - crystal templating (LCT). Many research groups exploited this technique of supramolecular templating to produce not only mesoporous silicate and aluminosilicate materials, but also mesostructured metal oxides.

The extension of this liquid - crystal templating (LCT) mechanism into the field of transition metal oxide synthesis represented a major step forward towards tailoring catalytic, electronic and magnetic properties of these redox - active materials. (S. Sathayanon, 2003)

However, the catalytic applications of pure silica MCM-41 are limited. It is known that incorporation of transition-metal ions in the silica framework may help to improve the catalytic performance of MCM-41. One of the interesting metals is iron due to the various applications such as epoxidation, polymerization, oligomerization.

There are two ways in which the iron can modify the silica structure. The iron may well be incorporated by substitution into the silica framework, and thus be attached by *covalent* bonds. Iron lies in a tetragonal geometry and is referred to as *framework* iron. In the second way, the metal may simply be in contact with the surface of the pore walls, and upon calcination, nanoclusters of octagonal crystalline iron oxide (Fe_2O_3) are formed. This *extra-framework* iron is retained to the support only by *weak* Van der Waals bonds. The major problem with iron atoms is the formation of extraframework iron species during template removal by calcinations (N. Crowther, 2003).

The major problem with iron atoms is the formation of extraframework iron species during template removal by calcinations (A. Tuel, 1999). Thus, the highest conversion of styrene over the Fe-MCM-41 via direct hydrothermal technique showed at 14% conversion over the catalyst of 1.1 wt% of Fe (Y. Wang, 2001).

Sol-gel technology is the preparation of ceramic, glass, or composite materials by the preparation of a sol, gelation of the sol, and removal of the solvent. In general, the sol-gel process is the synthesis of an inorganic network at low temperature by a chemical reaction in solution. This technique involves the transition characterized by a relatively rapid change from a liquid (solution colloidal solution) into a solid (gel-like state). Generally, the precursor is dissolved in a suitable organic solvent in order to obtain a solution. The solvent must be carefully selected so that a solution with high concentration of the required component is obtained. Sol-gel process involves the following steps: hydrolysis of precursor, polycondensation, gel formation, organic pyrolysis by heat treatment (N. Thanabodeekij, 2002).

Iron is known to be the active center in monooxygenase enzymes such as cytochrome P-450 and methane monooxygenase, which catalyze the selective oxidation of methane or several other hydrocarbons. Moreover, many iron-containing compounds and solid catalysts have been reported to catalyze the selective oxidation of benzene or alkanes. In this work, the epoxidation of styrene is used to study the efficiency of the catalyst. Styrene oxide and benzaldehyde were two main products in the epoxidation of styrene with H_2O_2 over Fe-MCM-41. Styrene glycol and benzoic acid were also formed with low selectivity (Q. Zhang, 2001). Many factors were studied to find the optimal condition of this catalyst such as temperature, reaction time, amount of catalyst and amount of loaded Fe (B. Su, 2001).

Thus, the objective of this research is synthesis via sol – gel process and study of the optimal condition for the synthesis of the iron loaded MCM-41, including the characterizations of the product. The catalytic activity of styrene epoxidation was studied.