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THESIS OUTLINE

Thesis Title: Study on the control of distribution of Al atoms in the framework of the MFI-type zeolite

Control of the Al atoms distribution in zeolite frameworks has attracted much attention. It has been reported that the Al atoms in ZSM-5 synthesized with only TPA cation in the absence of Na cation are selectively located at the intersections, and that the Al atoms in ZSM-5 synthesized with both TPA and Na cation are located not only at the intersections but also in narrow straight and/or sinusoidal channels. However, the preparation of the ZSM-5 zeolite with Al atoms preferentially located at straight and sinusoidal channels, not intersection, has not been attained to date.

Therefore, in this thesis, first I focused on the synthesis of ZSM-5 zeolites by using various organic molecules as OSDAs or pore-filling agents in the presence or absence of Na cations. The effect of the different organic molecules in the synthesis gel on the Al distribution was investigated. Then, I tackled the clarification of the influence of the distribution of Al atoms in ZSM-5 on catalytic properties by carrying out the MTO reaction and catalytic cracking of *n*-hexane. This thesis begins with general introduction (this chapter) and consists of six chapters.

In Chapter I

I reported on a comprehensive review on the introduction of zeolites, which included the synthesis, characterizations and catalytic applications of zeolite catalysts, especially the MFI-type (ZSM-5) zeolite. Recent development of the control of Al atoms distribution in zeolite framework was also included.

Chapter 2

I reported on the synthesis of the ZSM-5 zeolites with different Al distributions by using the organic molecules as structure-directing agents (OSDAs) and the obtained ZSM-5 zeolites were characterized by scanning election microscopy (SEM), X-ray diffraction (XRD), N_2 adsorption-desorption, NH_3-TPD, and elemental analysis. The Al distribution in the ZSM-5 zeolites was estimated by constrain index (CI).

Chapter 3

I reported on the synthesis of ZSM-5 zeolites with different Al distributions by using various kinds of alcohols as pore-filling agents in the presence of Na cations. The obtained ZSM-5 zeolites were characterized by SEM, XRD, N_2 adsorption-desorption, NH_3-TPD, and elemental analysis. The Al distribution in the ZSM-5 zeolites was estimated by constrain index (CI).

Chapter 4

I reported on a new class of ZSM-5 zeolite with Al atoms preferentially located at straight and sinusoidal channels. The strategy for achieving this is based on the use of a

bulky alcohol, Trimethylolethane (TME), in combination with Na cation; TME is a bulky molecule like TPA cation, and it works as a pore filling agent as well as organic-structure-directing agent. In this case, considering the size of TME molecule, it would occupy the channel intersections and Na^+ species are located at straight and sinusoidal channels. Meanwhile, TME molecules have no charge unlike TPA cation so that Al^{3+} species are located near Na^+ species, resulting in the unique distribution of Al atoms in the MFI framework; Al atoms are preferentially located at straight and sinusoidal channels, not the channel intersections. Thus prepared Al-site controlled ZSM-5 exhibited a longer catalytic life in the cracking of *n*-hexane and the methanol to olefins reaction than that with Al atoms located at intersection.

Chapter 5

I reported on the control of the particle size of the ZSM-5 catalysts with a unique Al distribution (described in Chapter 4). It was achieved by chaging the sizes or/and types (*i.e.*, calcined or as-synthesized) of the seeds (silicate-1) in the synthesis gel of ZSM-5. The produced samples were characterized and applied as catalysts for the MTO reaction.

Chapter 6

Summary: the aim of this study is to control the distribution of Al atoms in ZSM-5 zeolites and to investigate the Al atoms distribution on the catalytic performance. This thesis is composed of six chapters. Chapter I is a comprehensive review focusing on the introduction of zeolites. Chapters II and III focused on the synthesis of ZSM-5 zeolites by using various kinds of organic molecules as OSDA and/or pore-filling agents.

Chapter IV focuses on the effect of Al atoms distribution in ZSM-5 on the catalytic performance. Chapter V focuses on the control of crystal size of ZSM-5 with unique Al atoms distribution. Chapter VI is a general conclusion summarizing the significance of the research done in this dissertation.