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論文審査の要旨（2000 字程度）

The aim of this study is to control the distribution of Al atoms in ZSM-5 zeolite frameworks 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.

In Chapter I, there was 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.

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

In Chapter III, ZSM-5 zeolites with different Al distributions were synthesized by using various kinds of alcohols as pore-filling agents in the presence of Na cations. The obtained samples were characterized by SEM, XRD, NH_3-TPD, and elemental analysis. The Al atoms distribution in the ZSM-5 zeolites was estimated by constrain index (CI).

In Chapter IV, a new class of ZSM-5 zeolites with Al atoms preferentially located at straight and sinusoidal channels were synthesized. It was achieved by using bulky alcohols, such as Trimethylolethane (TME), as pore-filling agents, in combination with Na cation. 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 molecule has 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 (MTO) reaction than that with Al atoms located at the intersections. The part of this work has been published in Journal of Catalysis.

In Chapter V, the crystal size of ZSM-5 zeolites with a unique Al atoms distribution (described in Chapter 4) was controlled. It was achieved by changing the crystal size, type (i.e., as-made, Na-type, NH_3-type, and H-type), and amount of the seeds in the synthesis gel. The produced samples were characterized and applied as catalysts for the MTO reaction. The part of this work is now in preparation for submission.

In Chapter VI, a general summary and future prospect of this study are presented.

In short, the findings in this thesis will contribute to the development of a new class of zeolite catalysts with the Al atoms in the pores properly controlled. Furthermore, the synthesized zeolite with efficient reaction space and high diffusion pathways was a great contribution to the engineering science. Therefore, it is recognized that this paper is sufficient worth as a doctor (engineering) thesis.