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学 位 論 文 審 査 の 要 旨

博士の専攻分野の名称 博士（工学） 氏名 コウ ムブン

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学 位 論 文 題 名

Gallium-hydrides in Zeolites for Catalytic Dehydrogenative Transformation of Alkanes
(ゼオライト中のガリウム水素化物を用いたアルカン脱水素反応)

Metal-exchanged zeolite catalysts have great potential to form unique metal species for activation of small molecules such as light alkanes. Group 13 metal-loaded zeolites have attracted considerable attention for light alkane transformations, such as Cyclar process. Ga-loaded zeolites are one of the most widely studied zeolites for dehydrogenative transformations of light alkanes. The speciation of active Ga species in zeolites and their reaction mechanisms have been discussed in several studies, but the formation and catalysis of Ga-hydrides in various kinds of zeolites have rarely been reported. In this thesis, the author investigated Ga-hydrides in zeolites for dehydrogenative transformation of light-alkane.

Chapter 1 provides the general background of this paper. Chapter 2 shows the formation of Ga-hydrides in MFI zeolites during the catalyst preparation through reductive solid-state ion-exchange (RSSIE) and revealed their catalysis for non-oxidative ethane dehydrogenation (EDH). Their detailed structures and reaction mechanisms are discussed in combination with spectroscopic, and kinetic studies. The obtained results and insights are comprehensively discussed, including the relationship between the local structure of the active Ga species and reaction selectivity.

Chapter 3 demonstrates the formation and catalysis of isolated Ga-hydrides in various zeolites for EDH reaction. The effect of relative amount of Ga-hydride in Ga-loaded CHA, MOR, MFI on the EDH activity was studied. The catalytic activity of the relative amount of Ga-hydrides changed in the order of MFI > MOR >CHA. The result highlights the crucial role of zeolite frameworks in Ga-hydride formation and its impact on EDH.

Chapter 4 highlighted the speciation of Ga(I) cations and Ga hydrides in X-ray absorption fine structure (XANES) studies. The author developed Ga-exchanged zeolites after RSSIE for XANES study under operating conditions. The in situ XANES studies under various conditions, including CO, vacuum, and pressurized hydrogen, showed that the whiteline intensity in the spectra was responsible for the absence or presence of hydride sources in Ga-exchanged zeolites.

Chapter 5 investigated the potential of Ga-loaded zeolites in other dehydrogenative light alkane transformation reactions based on their high dehydrogenation activity. The author investigated the selective acetonitrile production from ethane and ammonia using Ga-loaded zeolite under anaerobic conditions. Various zeolite supports, including MFI, MOR, Y, BEA and FER, were compared for their catalytic activities, and the Ga-loaded FER zeolite exhibited the highest activity. The kinetic analysis suggests that acetonitrile is produced via a consecutive reaction pathway: the dehydrogenation of ethane to ethylene, which reacts with ammonia to yield acetonitrile.

Chapter 6 is the general summary. This study extensively explores the impact of Ga hydrides in zeolites on light alkane transformation reactions, particularly ethane dehydrogenation and acetonitrile synthesis. This not only enhances the understanding of Ga-zeolite catalysts but also provides crucial guidance for the development of novel catalysts for the synthesis of value-added chemicals from inexpensive feedstocks.

Thus, the author is qualified to receive the PhD degree in engineering, Hokkaido University.