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論文 / 著書情報
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（博士課程）
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論 文 要 旨
THESIS SUMMARY

系・コース： Department of, Graduate major in	応用化学 応用化学	系 コース	
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要旨（英文 800 語程度）
Thesis Summary（approx.800 English Words ）

ZSM-5 zeolite is a crystalline aluminosilicate material and has widely been used as a catalyst in chemical industries. Due to the chemical processes carried out in industry involving bulky-sized compounds, the original ZSM-5 often faces obstacles, such as the catalyst deactivation problem and undesired reactions. Moreover, the catalytic reactions are significantly influenced by the condition of acid sites as well, as the aluminum distribution in the framework. Therefore, modification of the framework structure and aluminum distribution on the hierarchical ZSM-5 intracrystalline becomes important. This summary highlights key findings from two chapters of a comprehensive study focused on the way to create a homogenous hierarchical structure of ZSM-5 zeolite with controlling Al distribution in the framework.

In Chapter 2, adjustment of aluminum distribution had been carried out on the hierarchical ZSM-5 intracrystalline. It was found that aluminum more located in the channel intersections than in the channels was produced by the hierarchical ZSM-5 with Na, which is opposite to the microporous ZSM-5's trend. The hierarchical ZSM-5 using a surfactant as mesoporogen gave rise to the occurrence of Al migration process from the micropores to mesopores and/or the external surface. Meanwhile, prolonging the hydrothermal time until 144 h resulted in not only a more well-structured hierarchical structure but also better chemical properties, which increased the hierarchy factor and strength of acid sites in the zeolite. Furthermore, it also influenced the rate of Al migration. The impact of the framework structure and the controlled aluminum was also seen in MTO and *n*-hexane cracking reactions. The hierarchical structure was predominant in producing lower olefins, especially in propene, compared to the micropore only, in which the hierarchical ZSM-5 without Na was superior to that with Na.

In Chapter 3, the effect of the synthesis condition and the composition of the starting materials on the aluminum distribution in the hierarchical ZSM-5 intracrystalline has been investigated. For the parameter of the crystallization temperature, the high crystallization temperature influenced not only aluminum distribution in the framework but also the hierarchical structure. Overheating crystallization temperature resulted in the enhancement of Al pairs distribution in the channel intersections, with a higher concentration on Hi_Z5_[TPA+Na]. However, the high crystallization temperature did not influence the ratio of Al distribution on the channels to the channel intersections, but it impacted on Hi_Z5_[TPA]. Moreover, a high crystallization temperature gave rise to an inhomogeneous structure between the microporous structure and mesoporous structure. In addition, it would lead to the formation of the microporous structure. Whereas for the parameter of the synthetic gel pH, the hierarchical ZSM-5 without pH adjustment (pH of ~14) possessed Al distribution was more concentrated in the channel intersection for Hi_Z5_[TPA+Na]_180_4.4_WApH, like the hierarchical ZSM-5 with pH adjustment which proved by ratio of T_{56}/T_{54} in the framework and ratio of C3=/C2= selectivity in the reaction of methanol conversion. Furthermore, the high pH of synthetic gel resulted in an increase of Al pairs distribution in the channel intersections. Meanwhile, the hierarchical structure produced by this treatment was not achieved. Even, it tends to the formation of the microporous structure only. On the other hand, Al distribution produced by the hierarchical ZSM-5 with timing mesoporogen addition into the synthetic gel switched to microporogen demonstrated the reversed trend with the hierarchical ZSM-5 synthesized using the original procedure. Its trend was the same as the original ZSM-5, in which the sample containing sodium (Hi_Z5_[TPA+Na]_180_4.4_Csw) had a higher ratio of T_{56}/T_{54} than the sample without

containing sodium (Hi_Z5_[TPA+Na]_180_4.4_Csw). This result was also proved by the ratio of C3=/C2= selectivity in the methanol conversion. Moreover, it resulted in a higher percentage of Al pairs distribution in the channel intersections compared to the control samples. Whereas the produced hierarchical structure, this treatment produced the mesopore inside the framework very little, which was shown by the appearance of a very low mesoporous step, indicating the microporous structure was more dominant than the mesoporous structure.

For the parameter of the composition of the starting materials, Al distribution on the hierarchical ZSM-5 with a lower concentration of mesoporogen (Hi_Z5_[TPA+Na]_180_2.2 and Hi_Z5_[TPA]_180_2.2) demonstrated the same trend as the hierarchical ZSM-5 with a higher concentration, in which Al distribution on Hi_Z5_[TPA+Na]_180_2.2 was more located in the channel intersections than Hi_Z5_[TPA]_180_2.2. This result was supported by the ratio of C3=/C2= selectivity in the methanol conversion reaction. Furthermore, the produced Al pairs distribution in the channel intersections was higher than the hierarchical ZSM-5 with a higher concentration, indicating the low concentration of mesoporogen would force the formation of Al pairs.

In conclusion, this work gives valuable insights into the zeolite field, especially the field of hierarchical zeolite, as a catalyst for increasing product selectivity in catalytic processes.

備考 ： 論文要旨は、和文 2000 字と英文 300 語を 1 部ずつ提出するか、もしくは英文 800 語を 1 部提出してください。

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