Effect of starting materials on Al distribution of the CHA-type aluminosilicate zeolites

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INTRODUCTION

Recently, the CHA-type zeolite has attracted much attention in small pore zeolites and has been extensively studied by many groups. Iorio and coworkers reported that the amount of bivalence cations in the CHA-type zeolite synthesized from using amorphous aluminosilicate was dependent on the ratio of organic structure-directing agent (OSDA) and Na^+ cation [1]. On the other hand, for the CHA-type zeolite synthesized from the FAU-type zeolite as a starting material, the introduction of heteroatoms is governed by the composition of the FAU-type zeolite. [2] In this work, the effects of the synthesis method on the acidic and catalytic properties were investigated in detail.

EXPERMENTAL

CHA type zeolites were synthesized by using amorphous aluminosilicate and FAU type zeolite as raw materials. The synthesis was carried out by changing the proportion of FAU type zeolite and amorphous aluminosilicate with the SiO_2/ Al_2O_3 ratio in the gel of 20 kept at the crystallization temperature of 150 or 170 ºC. When the proportions of FAU in the raw material were 10 and 100 %, the products were designated as "CHA-F-0.10" and "CHA-F-1.0", respectively.

RESULTS AND DISCUSSION

The XRD patterns of the products were shown in Fig. 1. The CHA-type zeolites were obtained regardless of the amount of FAU as a raw material at the crystallization temperature of 170 ºC. At the crystallization temperature of 170 ºC, a mixture of CHA and FAU zeolites was formed when FAU was only used as a raw material. These results suggest that the dissolution rate of FAU influences the formation of the CHA-type zeolite.

Fig. 2 shows the results of ^{29}Si MAS NMR measurement of CHA-F-0.10 and CHA-F-1.0 synthesized at 170 ºC. It was found that CHA-F-0.10 had a higher $Si(OSi)_3(OAl)_1$ spices (Si(1Al)) peak. Further **Si**$(OSi)_2(OAl)_2$ spices (Si(2Al)) peak was lower than CHA -F-1.0. Thus observed differences imply that the Al distribution of the products would be influenced by the proportion of FAU (Si / Al = 2.8) in the raw material gel.

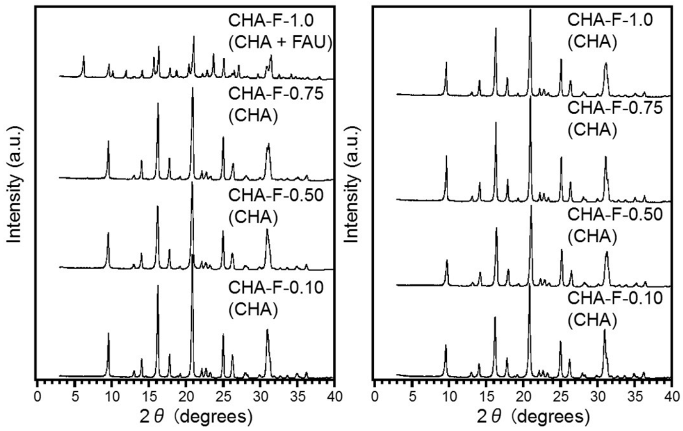

Fig.1 XRD pattern of the products at crystallization temperatures of 150ºC (Left) and 170ºC (right).

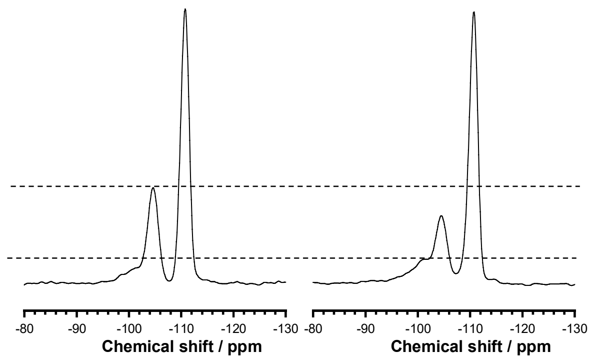

Fig.2 ^{29}Si MAS NMR spectra of CHA-F-0.1 (left) and CHA-F-1.0 (right).

The catalytic properties of the CHA-F-0.10 and CHA-F-1.0 samples synthesized at 170 ºC for the MTO reaction at 350 ºC were representatively evaluated. There was no marked difference in the products distributions, while CHA-F-0.10 exhibited a much longer catalytic life than CHA-F-1.0. This would be caused by the difference in the Si(1Al)/Si(2Al) ratio, *i.e.*, Al distribution in the framework.

Thus, we have successfully found that the proportion of FAU in the raw material strongly affect the distribution of Al in the CHA framework, leading to the difference in the catalytic properties.

REFERENCES

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