Control of Si/Al in chabazite zeolite using Na/Si ratio during hydrothermal synthesis

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Copper ion exchanged chabazite (CHA) zeolite has been utilized extensively for selective catalytic reduction of NOx using ammonia.[1] The content of copper and the corresponding location affected the catalytic performance critically and also hydrothermal stability. The ion exchange level of Cu^{2+} in CHA can be determined with the Si/Al ratio which has been difficult to control because the direct conversion of Y zeolite resulted in the overall Si/Al in addition to the zeolite product yield.[2]

In this work, the Si/Al in the CHA zeolite was controlled readily to the range of 3.8-13.1 while the Na/Si ratio in the synthesis gel was varied. The CHA zeolite was synthesized employing trimethyl adamantylammonium ion as a structure directing agent at 413 K for 4 days under rotation at 40 rpm. The CHA zeolite of different Si/Al was also subjected to the Cu ion exchange for further study on the location of Cu, its hydrothermal stability and finally the catalytic performance in selective catalytic reduction of NO.

Fig. 1 shows the scanning electron micrographs of the obtained CHA with different Si/Al ratios. The particle shape and size were affected by the Na/Si ratio. Increasing Na/Si ratio resulted in the formation of the large agglomerate of CHA zeolite with low Si/Al ratios. The CHA zeolite with high Si/Al ratio, 11-13 was found to be 100-200 nm cube when the low Na/Si ratio was adopted in the synthesis gel. More Al incorporation in the framework of zeolite also increased the lattice parameter because of large Al-O distance compared to that of Si-O, which was referred from the shift of (101) peak to lower angle in X-ray diffraction pattern with the decrease of the Si/Al ratio.

Fig. 2 shows the Si/Al ratio of the obtained CHA zeolite against the Na/Si ratio in the synthesis gel. Increase of the Na/Si ratio was resulted in the formation of CHA zeolite with low Si/Al ratio. Therefore, the low Na/Si ratio should be employed in order to obtain high siliceous hydrothermally stable CHA zeolite. The effect of the Na/Si ratio also supports the role of Na to control the Al location and structured directing effect in addition to that of trimethyl adamantylammonium ion.

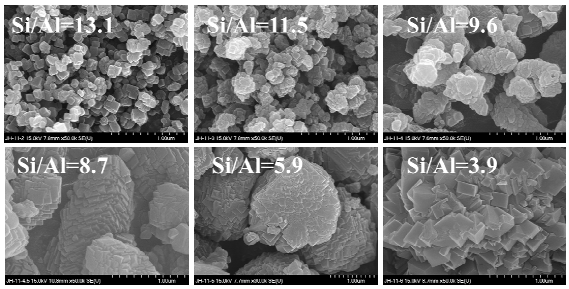

Fig.1 SEM images of the CHA zeolite.

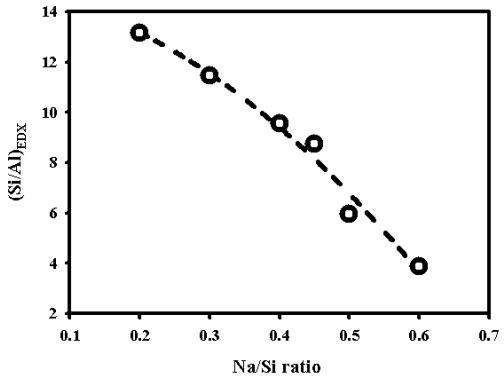

Fig. 2 The Si/Al ratio of CHA zeolite against the Na/Si ratio in the synthesis gel.

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