Effect of Cation in High Silica CHA on Hydrothermal Stability and Selective Catalytic Reduction of NOx

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Copper containing high siliceous small pore zeolite, chabazite has been applied successfully for NOx abatement for mobile source [1]. High catalytic activity of selective catalytic reduction of NOx using NH_3 over copper containing chabazite has been observed in the range of over 423 K and 823 K. However, the corresponding catalytic activity is significantly deteriorated when the catalyst is subject to hydrothermal aging at 1073 K or more under 10% water [2]. The hydrothermal stability of the catalyst can be affected the Si/Al ratio, cation presented in the zeolite, and crystal size. More siliceous zeolite is stable under hydrothermal aging but the copper content can be limited due to the Al content.

In this work, high siliceous chabazite of which the Si/Al_2 ratio was 24 has been prepared using benzyltrimethyl ammonium ion as organic structure directing agent through direct hydrothermal conversion of ultrastable Y zeolite. The obtained samples was calcined at 823 K and subsequently ion exchanged with copper salts. The copper containing chabazite (Cu/Z) was subjected to the catalytic reaction in which the NO and NH_3 were 350 ppm, respectively with 5% oxygen, 3% water and nitrogen balance.

Fig. 1 shows the scanning electron micrograph of chabazite prepared in this work. The particle shape was cube in which the length was ~ 500 nm. There was no observable amorphous phase other chabazite. The particle size was found to be controllable from 100 nm to 2 μm. After NH_4^+ and Na^+ ion exchange, the surface area of the sample were 614 and 581 $m^2\ g^{-1}$, respectively. Such textural properties were found to be deteriorated modestly 5% and 12%, respectively when the zeolite was subjected to the hydrothermal aging at 1173 K under 10% water for 24 h. The result suggested that the presence of Na^+ enhances the transformation of chabazite to quartz, which is further accelerated for the chabazite with low Si/Al_2, 12. The loading of copper into chabazite further aggravated the textural properties. H form chabazite after copper loading was found to be resistant to the sintering. Also, the catalytic activity of Cu/Z of H form showed superior performance at high temperature as shown in Fig. 2.

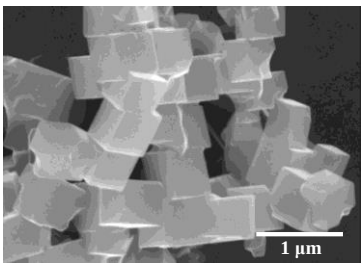

Fig.1 SEM micrograph of zeolite (Si/Al_2=24).

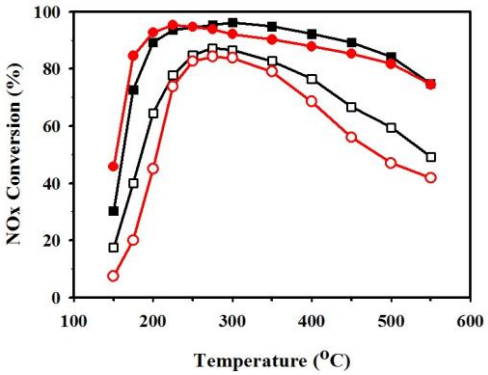

Fig. 2 Catalytic conversions of NOx over Cu/H-Z (■), Cu/H-Z-HTA (□), Cu/Na-Z (●), and Cu/Na-Z-HTA (○).

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