Improved high temperature activity of NH_3-SCR over Cu/SUZ-4

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NO_x emissions are major pollutant that causes photochemical smog, acid rain and respiratory problems. SCR (Selective Catalytic Reduction) is a reaction to convert nitrogen oxide with the aid of catalyst into nitrogen using ammonia/urea as gaseous reductant[1]. Copper ion exchanged zeolite is the one candidate among several zeolite-based catalysts for SCR. Copper supported on ZSM-5 has been known to have good SCR activity at low temperature. In this communication, we report NH_3-SCR activity of Cu/SUZ-4.

SUZ-4 (Si/Al=6.57) synthesized by hydrothermal method using the synthesis gel having the composition of 7.35 K_2O: Al_2O_3: 3.10 TeA_2O: 33.3 SiO_2: 781 H_2O. ZSM-5 (Si/Al=20) was obtained from TOSOH corporation. Cu/H-zeolite catalysts were prepared by an ion exchanged using 0.015 M $Cu(CH_3COO)_2$ method at 80°C for 1 h. Standard NH_3-SCR were performed by using reaction mixtures containing 240 ppm NH_3, 200 ppm NO, 7 vol% O_2, 5 vol% H_2O at a GHSV = 80,000 h^{-1}. The catalyst sample (500 mg, 30-60 mesh) was loaded in a quartz tube and placed inside an electric furnace. Hydrothermal aging treatment was carried out for 3 h at 750°C.

Table 1 gives copper amounts loaded on zeolites. Figure 1 shows the NO conversion over Cu/zeolite catalysts in presence of oxygen. Cu/H-SUZ-4 showed higher activity than Cu/H-ZSM-5 at higher temperature. Though the NH_3-SCR activity of Cu/H-ZSM-5 decreased rapidly after 400°C due to NH_3 oxidation, Cu/H-SUZ-4 kept high activity even at above 600°C. Even after the hydrothermal treat, the levels of NO conversion were similar to those of fresh catalyst. The XRD measurements were performed for both catalysts to examine possible structural changes. The XRD patterns indicated that the intensities of reflection peaks decreased but the structure remains the same.

Figure 2 shows the NH_3 desorption profiles. Cu/H-SUZ-4 gave NH_3 desorption at higher temperatures. The acidity generally influences the activation of NH_3 and provides NH_4^+ adsorbed at high temperature and the overall acidity of framework zeolite is affected by Si/Al ratio[2]. This would be consistent with NH_3-SCR results observed, strong acid probably contributed to the appearance of high SCR activity of Cu/SUZ-4 at high temperatures. UV-Vis results suggested that copper states on zeolite, would be mixture of Cu^{2+} species interacting with oxygen of zeolite structure, copper dimer and Cu^{2+} cations in CuO particles. We expect that improvement of catalyst preparation to control Cu states is expected to lead to further improvement of SCR activity.

Table 1 Loaded amounts of Cu

Catalyst	Cu loading /wt%	Cu/Al
Cu/H-ZSM-5	1.68	0.872
Cu/H-SUZ-4	1.49	0.277

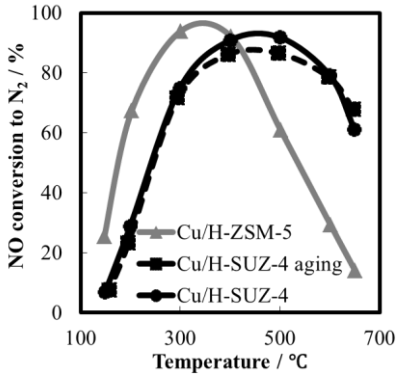

Fig. 1 NO conversion over Cu/zeolites.

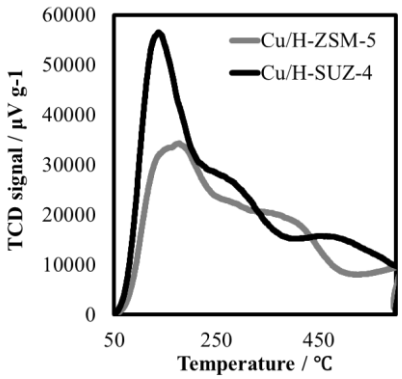

Fig. 2 NH_3 desorption profiles.

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