

High Catalytic Activity of Platinum-ZSM-5 Zeolite below 500 K
in Water Vapor for Reduction of Nitrogen Monoxide

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Platinum ion-exchanged ZSM-5 zeolite efficiently reduced nitrogen monoxide to nitrogen by ethene in an oxidizing atmosphere below 500 K and the catalytic activity was not affected by the presence of 8.6 vol% water vapor.

The selective reduction of nitrogen monoxide (NO) by hydrocarbons in an oxidizing atmosphere has been reported to be catalyzed over various catalysts.¹⁻⁸⁾ Although iron or copper ion-exchanged zeolite is active below 573 K,⁸⁾ the catalytic activity is still low for the exhaust gases from diesel and lean-burn gasoline engines in a state of idling and is greatly decreased by the presence of water vapor.⁹⁾ Here we wish to report a pronounced catalytic activity of platinum ion-exchanged ZSM-5 zeolite below 500 K for the selective reduction of NO by ethene even in the presence of water vapor.

Parent zeolites, ZSM-5 (silica/alumina = 23.3) and mordenite (18.9) were supplied by Tosoh Corporation and they were abbreviated as MFI and MOR, respectively. Platinum, copper, and iron ion-exchanged zeolites were prepared as described previously.^{1,9,10)} The exchange levels of the metal ions were estimated on the assumption that the oxidation numbers of Pt, Fe, and Cu ions are +2. Hereafter the sample was abbreviated as Pt-MFI-97 (cation - zeolite structure - degree of exchange). The catalytic reduction of NO was carried out with a fixed bed flow reactor (inner diameter, 7.5 mm) in 1000 ppm of NO, 1000 ppm of C₂H₄, and 2% of O₂ system (balance: He) in the presence or in the absence of 8.6 vol% water vapor. Total flow rate was 150 cm³·min⁻¹. The catalyst of 0.0625 g (particle size, 250 - 425 μm; volume, 0.125 cm³; GHSV = 72000 h⁻¹) was used without any diluents. The catalytic activity for NO removal was evaluated by the conversion into N₂.

The catalytic activity of Pt-MFI-97, Fe-MOR-71, or Cu-MFI-105 at 473 - 523 K was somewhat changed with reaction time and approximately steady formation of N₂ was attained after 15, 6, or 3 h on each catalyst. Figure 1 shows the temperature dependences of their catalytic activities in the absence of water vapor. The most active temperature of Pt-MFI-97 was 485 K and the conversions into N₂ and N₂O were 18% and 36%, respectively, at 485 K and GHSV of

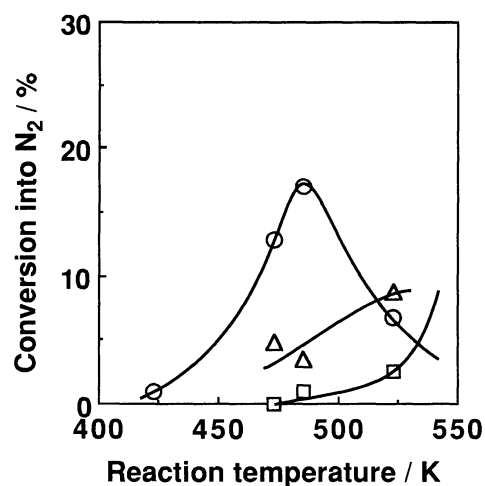


Fig. 1. Temperature dependences of the catalytic activities of (○) Pt-MFI-97, (Δ) Fe-MOR-71, and (□) Cu-MFI-105 in the absence of water vapor.

72000 h⁻¹. Although Cu-MFI and Fe-MOR catalysts have been reported to be the most active in the range 473 - 573 K,^{1,9)} the results of Fig. 1 show that Pt-MFI-97 is more active than these catalysts below 500 K. The conversions of C₂H₄ at 485 K on Pt-MFI-97, Fe-MOR-71, and Cu-MFI-105 were 100, 4, and 1%, respectively. The higher ability for the oxidation of C₂H₄ is probably the reason for the high ability to reduce NO into N₂.

The effects of water vapor on the catalytic activities of Pt-MFI-97, Fe-MOR-71, and Cu-MFI-105 are depicted in Fig. 2. The catalytic activity of Pt-MFI-97 was not decreased by the introduction of 8.6% of water vapor. On the other hand, the conversions into N₂ on Fe-MOR-71 and Cu-MFI-105 were reduced nearly to 0% by the addition of water vapor though the activities were approximately restored by the interrupt of water addition.

It follows that Pt-MFI-97 has the bright prospect of effective reduction of NO into N₂ in the presence of oxygen and water vapor below 500 K.

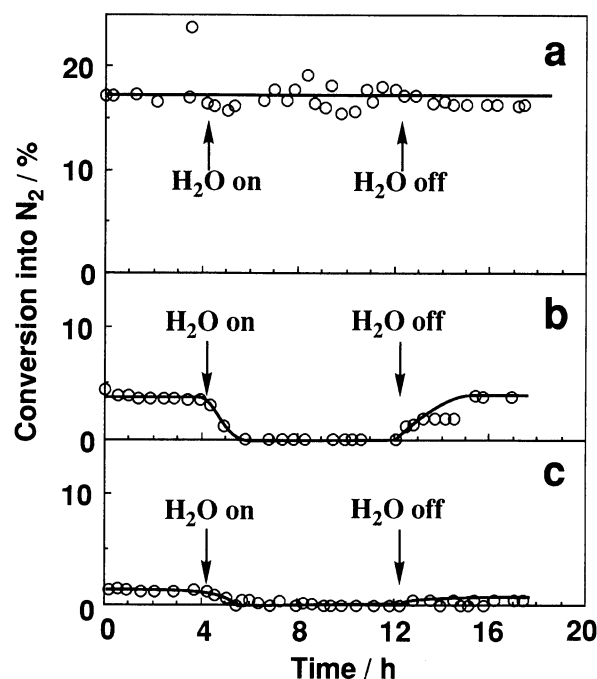


Fig. 2. Effect of water vapor (8.6 vol%) on the catalytic activities of (a) Pt-MFI-97, (b) Fe-MOR-71, and (c) Cu-MFI-105 for the selective reduction of NO. Reaction temp., 485 K.

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