

A new ECO-friendly catalytic system for removing NOx from automotive engine by urea/SCR technology

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Introduction

Selective Catalytic Reduction (SCR) of NOx by urea is one of the most reliable and unique technology to meet the future NOx emission regulations including EURO V and SULEV for world-wide automotive industry [1]. CuZSM5 and V₂O₅/TiO₂ catalysts have been commonly recognized as commercial catalyst for urea/SCR technology. However, these catalysts still suffer from their low temperature activity and the public perception on the use of copper and vanadium as a catalyst composition, mainly due to their toxicity and low melting point [2,3].

The MnOx-based catalyst has been proposed and reported as an excellent low temperature SCR catalyst [4]. Recently, Mn-Fe/TiO₂ catalyst prepared by sol-gel method as an Eco-friendly SCR catalyst has been reported [5]. To overcome the drawbacks of Mn-Fe/TiO₂ catalyst including low temperature activity, N₂ selectivity and narrow operating temperature window, a new Eco-friendly Mn based ZSM5 catalytic system has been suggested to remove NOx from next generation vehicle including diesel engine in the present study. Note that Mn compounds are less toxic than the heavy metals including as iron, nickel, copper and vanadium. It is also an essential trace nutrient for human and animal lives [6].

Materials and Methods

The Mn based ZSM5 catalysts have been prepared by the wet-impregnation method with respect to the Mn contents. Their deNOx activities were examined over a packed-bed down flow reactor system at the reactor space velocity of 100,000 hr⁻¹ under the reaction condition including 500 ppm NO, 500 ppm NH₃, 5% O₂, 10% H₂O and N₂ balance [7]. To confirm the deNOx activity and N₂ selectivity of the catalysts prepared, the on-line chemiluminescence NO/NO₂ analyzer and FT-IR analyzer with gas cell have been employed in the present study.

Results and Discussion

The physicochemical properties of the catalysts prepared are listed in Table 1. The metal contents of Mn based ZSM5 catalysts were optimized for their high deNOx performance. The deNOx performance of Mn-Fe/ZSM5 proposed as an Eco-friendly catalyst is compared to those of the CuZSM5 and Mn-Fe/TiO₂ catalysts reported. As shown in Fig. 1, the Mn-Fe/ZSM5 catalyst containing 20 wt. % of Mn and 10 wt. % of Fe reveals the highest deNOx activity and N₂ selectivity in the temperature range upto 350 °C. However, the NOx conversion of the Mn-Fe/ZSM5 catalyst rapidly decreases by the further increase of the reaction temperature. It is mainly due to the oxidation reaction of NH₃ by MnO₂. Although the NOx conversion of the Mn-Fe/ZSM5 catalyst is relatively low in the high temperature

region compared to that of CuZSM catalyst, the catalytic system still contains excellent low temperature activity, appropriate operating temperature window and competitive N₂ selectivity to remove NOx from diesel engine. It should be noted that the highest temperatures of exhaust stream from light- and heavy-duty diesel engines are typically 250 and 350 °C. In addition, Mn-Fe/ZSM5 catalyst has been characterized by XRD, XPS, TPD and in-situ FT-IR to elucidate its high deNOx performance and the active reaction site on the catalyst surface.

Table 1. The physicochemical properties of the catalysts prepared

Catalyst	Metal Contents (wt.%)			BET surface area (m ² /g)	Calcination condition ^a
	Mn	Fe	Cu		
Mn/ZSM5	20	-	-	250	Oven with air flow
Mn-Fe/ZSM5	10	10	-	252	
	20	10	-	220	
	30	10	-	184	
Mn-Fe/TiO ₂	28	14	-	148	Oven with static air
CuZSM5	-	-	3.1	330	

^a: 500 °C for 5hr

Significance

A new ECO-friendly Mn based ZSM5 catalytic system revealing high deNOx performance has been proposed to effectively remove NOx from automotive engine by urea/SCR technology.

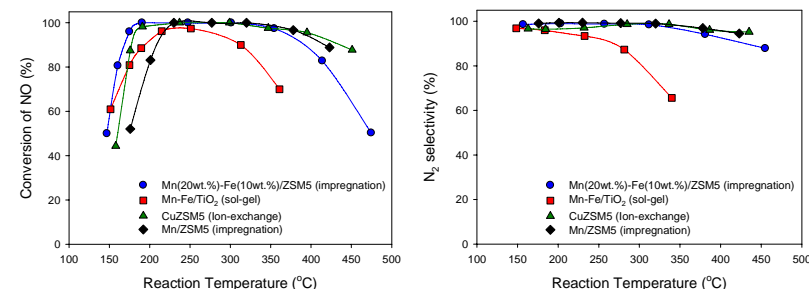


Figure 1. High deNOx performance of Mn-Fe/ZSM5, CuZSM5 and Mn-Fe/TiO₂ catalysts.

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