



Short communication

Novel promotional effect of yttrium on Cu–SAPO-34 monolith catalyst for selective catalytic reduction of NO_x by NH₃ (NH₃-SCR)

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ABSTRACT

Cu–SAPO-34 and CuY–SAPO-34 catalysts for NH₃-SCR were prepared by the wet-impregnation method. XRD, UV–vis DRS, ESR and NH₃-TPD results showed that the introduction of Y effectively improved the dispersion of copper species, increased the amount of isolated copper ions and enhanced the acid density. In addition, the activity test, NH₃-TPD and TGA results reflected that the CuY–SAPO-34 catalyst showed better C₃H₆ oxidation activity, lower dropping degree of acid sites after C₃H₆/O₂ treatment and less adsorption of C₃H₆/O₂ than Cu–SAPO-34 catalyst. Therefore, the addition of Y promoted the NH₃-SCR performance and the hydrocarbon (HC) resistance of Cu–SAPO-34 catalyst.

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1. Introduction

NO_x emitted from diesel vehicle would cause serious environmental issues, such as acid rain, photochemical smog, ozone depletion and haze pollution events. NH₃-SCR was recognized as one of the most efficient technologies to eliminate NO_x [1]. Recently, copper ion modified zeolite with CHA structure NH₃-SCR catalysts have attracted much attention owing to their excellent activity, N₂ selectivity and hydrothermal stability [2,3].

However, NH₃-SCR performance of Cu–SAPO-34 would be inhibited by HCs which were presented in the exhaust during the cold start period or in the case that upstream diesel oxidation catalysts were deactivated [4–7]. Compared with other types of HCs (paraffins, aromatics and long-term olefins), C₃H₆ had severer inhibition effect [5,8]. Thus, C₃H₆ was applied to determine the HC resistance of Cu–SAPO-34 catalyst. The poisoning effect of C₃H₆ on the Cu–zeolite catalyst was attributed to the formation of surface acrolein-like and/or coke species which would block the channels of the zeolite and cover the active sites [4,9], and the competitive adsorption between C₃H₆ and NH₃/NO_x which would inhibit the adsorption of NH₃/NO_x [4,10].

The aim of this study is to improve the HC resistance of Cu–SAPO-34 catalyst. Sultana et al. [6] confirmed that the addition of Na⁺ promoted the HC resistance of Cu–ZSM-5 by retarding the formation of coke. Meanwhile, Seo et al. [5] reported that the introduction of ZrO₂ improved the HC resistance of Cu–ZSM-5 by adjusting its acidity. Furthermore, Kim et al. [11] attributed the high HC resistance of MnFe–ZSM-5 catalyst to its excellent C₃H₆ oxidation activity. As declared in our previous study, the addition of yttrium could improve the C₃H₆ oxidation activity of Pt–TiO₂ catalyst [12]. Herein, the effect of Y on the HC resistance of Cu–SAPO-34 was investigated by N₂ adsorption, XRD, UV–vis-DRS, NH₃-TPD and TGA measurements.

2. Experimental

2.1. Catalyst preparation

CuY–SAPO-34 (CuY) catalyst was prepared by a simple wet impregnation method. 1.1325 g of Cu(NO₃)₂·3H₂O (99%, Kelong) and 0.6025 g of Y(NO₃)₃·6H₂O (99%, Jinshan) were dissolved into 5.1 ml of deionized water, then 11.5965 g of commercial NH₄-SAPO-34 (Nankai, Tianjin, mole ratio: Si / (Al + P + Si) = 0.1) was poured into the mixed solution to obtain a paste, afterwards the paste was stirred at 19 °C for 3 h followed by drying at 100 °C for 6 h and calcining at 550 °C for 3 h. Cu–SAPO-34 (Cu) catalyst was prepared by the similar method above. Finally the prepared catalysts were coated on honeycomb cordierites (2.5 ml, 400 cpi, Jiangsu Yixing) as described in our previous study [12], and

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Table 1
The chemical composition, surface areas, pore volumes and amount of acid sites of the catalysts.

Sample	Chemical composition of the catalyst ^a (wt.%)					Surface areas (m ² /g)	Pore volumes (ml/g)	Acid sites (mmol/g)	Acid density (μmol/m ²)
	Cu	Y	Si	Al	P				
Cu	3.1	–	2.1	19.6	18.2	616	0.24	0.81	1.31
CuY	3.0	1.3	2.6	20.1	17.2	580	0.23	0.79	1.36
Cu–HC ^b	–	–	–	–	–	556	0.22	0.60	1.08
CuY–HC ^b	–	–	–	–	–	570	0.23	0.67	1.18

^a Analyzed by ICP-AES.

^b Treated in the simulated gas with C₃H₆ at 300 °C for 50 min.

the loading of each monolith catalyst was ca. 130 g/L. The chemical component of each catalyst was determined by ICP-AES (Table 1).

2.2. Activity measurement

The catalytic activity measurement was conducted in a fixed bed quartz flow reactor. And the concentrations of the simulated gases were as follows: 350 ppm NO, 350 ppm NH₃, 8% O₂, 5 vol% H₂O, 700 ppm C₃H₆ (when added) and balanced with N₂. The gas hourly space velocity (GHSV) was 30,000 h⁻¹. The concentrations of NO_x, N₂O and C₃H₆ were continually analyzed by an FT-IR (Antaris IGS, Thermo Fisher Scientific).

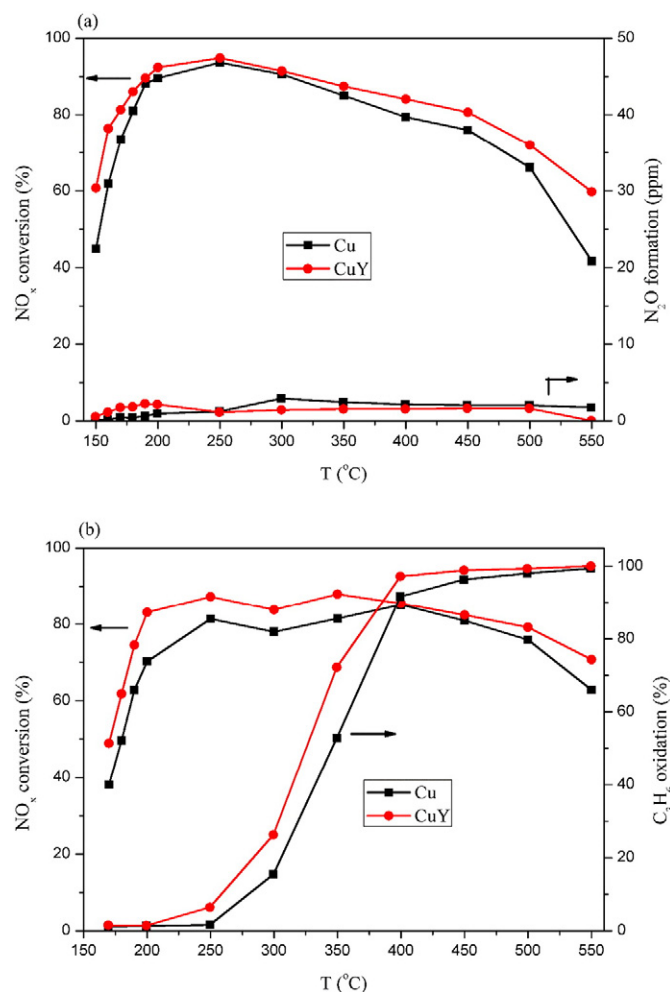


Fig. 1. The NO_x conversion and N₂O generation on Cu and CuY catalysts (a) and the C₃H₆ oxidation activity and NO_x conversion on Cu and CuY catalysts in the presence of C₃H₆ (b). Feed condition: 350 ppm NH₃, 350 ppm NO, 8% of O₂, 5 vol% of H₂O, 700 ppm C₃H₆ (when used) and N₂ balance, GHSV: 30,000 h⁻¹.

2.3. Catalyst characterization

N₂ adsorption was determined by a QUADRASORB SI automatic surface analyzer (Quantachrome Corporation). The structure of the samples was collected by XRD (Rigaku D/MAX-rA). UV-vis-DRS was taken on a “Shimadzu” UV-3600 PC spectrophotometer. ESR spectra were recorded at 103 K on the X-band using an ESR spectrometer (JES-FA200, JEOL). Prior to the ESR analysis, 30 mg of sample was evacuated (5 Pa) and heated from RT to 150 °C, then the sample was treated at 150 °C in vacuum for 3 h. NH₃-TPD was carried out on a commercial instrument (Xianquan, TP5076) with a TCD detector at a linear heating rate of 10 °C/min from 120 to 650 °C under He (30 ml/min) flow. TGA (Hengjiu) was conducted to determine the adsorption of propene on catalyst in mixture gas (35 ml/min) containing 700 ppm C₃H₆, 8% O₂ and N₂ as balance gas.

3. Results and discussion

3.1. Activity test

NH₃-SCR performance and N₂O formation of Cu and CuY catalysts were presented in Fig. 1a. CuY catalyst performed better NO_x conversions than Cu catalyst in the whole temperature range. In addition, the temperature range of NO_x conversion which was higher than 80% of CuY catalyst located between 167 and 454 °C, while that of Cu catalyst was between 179 and 393 °C. Meanwhile, less than 5 ppm of N₂O was formed, suggesting that a high selectivity of NO to N₂ was achieved. Furthermore, the effect of Y on the HC resistance and propene oxidation activity of the catalyst was displayed in Fig. 1b, C₃H₆ oxidation activity of CuY catalyst was better than that of Cu catalyst, and the inhibition effect of C₃H₆ on Cu catalyst was severer than that on CuY catalyst, indicating that the introduction of Y improved the HC resistance of Cu catalyst.

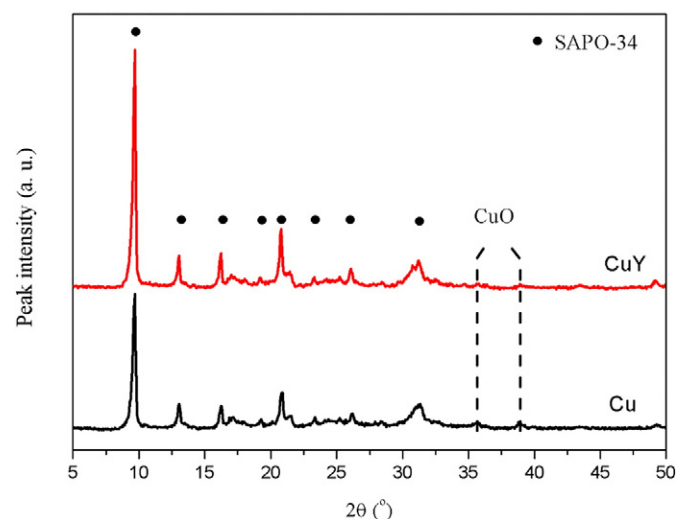


Fig. 2. XRD patterns of the Cu and CuY catalysts.

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