Electronic Supplementary Information

Low-temperature NO reduction with NH₃ over Mn-CeO_x/CNTs catalysts prepared by a liquid-phase method

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Experimental section

1. Catalyst preparation

The raw multiwall CNTs with an outer diameter of 60-100 nm were firstly refluxed and oxidized with concentrated HNO₃ under stirring for 4 h in order to remove impurities and introduce oxygenated surface groups for further deposition of the active phase. Then certain amount of acid-treated CNTs and cerium nitrate were mixed in 50 ml deionized water and stirred for 6 h. After that, 40 ml of potassium permanganate solution was added into above mentioned mixture with vigorous stirring at room temperature for 12 h. Finally, the solid was filtered and washed with deionized water for 3-4 times, followed by desiccation in a vacuum drier at 120 °C for 12 h. The obtained catalyst by this method is denoted as Mn(y)-CeO_x/CNTs, where y represents the molar ratio of Mn/(Mn+Ce) used in the preparation process.

2. Catalytic activity tests

The evaluation of the catalyst activity was performed in a fixed-bed quartz reactor. Prior to activity tests, samples were pretreated in air at 200 °C for 0.5 h to completely remove the residual water, which was combined with metal oxides during the preparation process. 180 mg sample (ca. 1.4 ml) was used in each test. The gas composition was 500 ppm NO, 500 ppm NH₃, 5% O_2 balanced by N₂. The total flow rate was 700 ml/min, which corresponded to a gas hourly space velocity (GHSV) of 30,000 h⁻¹. The gas composition (NO, NO₂ and O₂) was monitored by a flue gas analyzer (Kane International Limited, KM940). All the data were collected after 30 min when the SCR reaction reached a steady state.

3. Characterization techniques

X-ray diffraction (XRD) patterns were recorded on an X'Pert Pro MPD X-ray diffractometer using Cu K α radiation (λ =0.15406 nm). The data were collected for scattering angles (2 θ) ranging between 10 and 80° with a 0.02° step size. Transmission electron microscopy (TEM) images were obtained using a JEOL model JEM 2010 EX instrument. X-ray photoelectron spectroscopy (XPS) measurement was performed on a Thermo Scientific ESCA Lab250 spectrometer. Temperature programmed reduction by hydrogen (H₂-TPR) was carried out on a custom-made TCD setup using 50 mg catalysts. Prior to H₂-TPR experiments, samples were pretreated in N₂ at 200 °C for 1 h. H₂-TPR runs were carried out with the linear heating rate (10 °C/min) in pure N₂ containing 6 % H₂ at a flow rate of 30 ml/min. The thermal stability was determined by thermogravimetic analysis (TGA: SDT-Q600) from room temperature to 800 °C at a heating rate of 10 °C min⁻¹ in air.

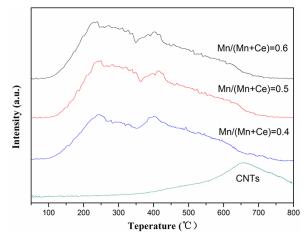


Fig. S1 H₂-TPR profiles of the acid-treated CNTs and Mn-CeO_x/CNTs catalysts prepared by the liquid-phase method with constant (Mn+Ce)/C molar ratio (4 %) but different Mn/(Mn+Ce) molar ratios.

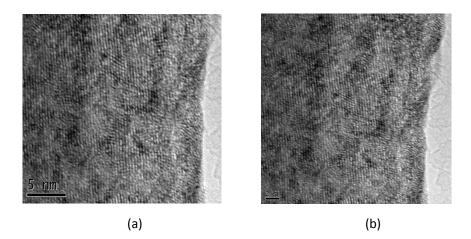


Fig. S2 HRTEM images with different magnification of the Mn(0.5)-CeO_x/CNTs catalyst.

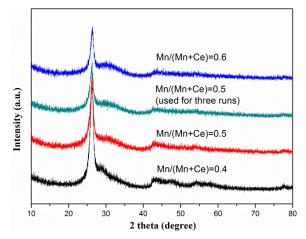


Fig. S3 XRD spectra of Mn-CeO_x/CNTs catalysts (Ce/C =2 %).

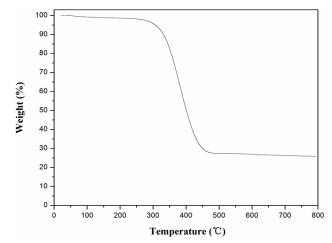


Fig. S4 TGA curve of Mn(0.5)-CeO_x/CNTs catalyst showing weight loss at a heating rate of 10 °C min⁻¹ in air.

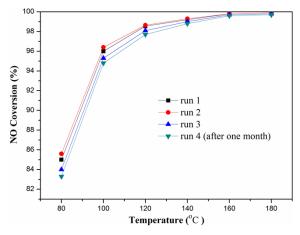


Fig. S5 Cyclic activities of the Mn(0.5)-CeO_x/CNTs catalyst for SCR of NO with NH_3