



Short communication

Application of MnO_x/HNTs catalysts in low-temperature NO reduction with NH₃

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ABSTRACT

Halloysite (HNTs) was firstly adopted as a carrier to develop manganese based SCR catalysts. Supporting of manganese oxides (MnO_x) on halloysite was carried out via a simple in-situ reaction of Mn(AC)₂ and KMnO₄ on the surface of halloysite. The obtained MnO_x/HNTs catalyst was estimated for selected reduction of NO by NH₃. It was found that the SCR activity of the catalyst is perfectly high with a nearly 100% NO conversion at temperatures from 60 to 250 °C. Results showed that the adoption of Halloysite as carrier and the in-situ preparation method jointly play key function for the performance of the catalyst.

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1. Introduction

Selective catalytic reduction of NO_x with NH₃ had proved to be a cost-effective, reliable, and fuel-efficient way to control NO_x emission, especially for stationary sources such as power boilers and combustion furnaces [1,2]. V₂O₅-WO₃(MoO₃)/TiO₂ as a kind of commercial catalyst had high activity and selectivity in SCR of NO with NH₃ at 300–400 °C, but, there were some inevitable problem that remained in this catalyst, such as the toxicity of vanadium, SO₂ oxidation to SO₃, and the need to operate at high temperature [3,4]. Therefore, the development of novel catalysts for the low-temperature NH₃-SCR remained as an important challenging task for many researchers due to their potential application in downstream of the desulfurizer and electrostatic precipitator, where the flue gas temperature was usually below 200 °C or even lower [5–7]. In recent researches, Manganese based catalysts had been applied and exhibited excellent activity in low-temperature SCR reactions, such as MnO_x/Al₂O₃ [8,9], MnO_x/active carbon [10,11], but the actually low-temperature SCR activity (especially below 100 °C) for most catalysts remained unsatisfactory.

Recently, an accumulating number of researchers focused on the attempt of the nanocrystallization of catalyst carriers. Owing to the unique structure property and chemical stability, titania nanotubes and carbon nanotubes (CNTs) were widely applied as carrier materials to prepare catalysts such as MnO_x/TiO₂ [12–14], MnO_x/CNTs [15–17], Mn-CeO_x/TiO₂ [18,19], Mn-CeO_x/CNTs [20,21], which were found to be active for SCR at lower temperatures. Besides, in our previous report, we had successfully developed a manganese based SCR catalysts

adopting an fibrillar aluminosilicate caly material (palygorskite) as carrier to obtain an perfect lower temperature NO removal and indicated that the traditional aluminosilicate might be more suitable for catalyst carrier [22]. But, the use of HNTs (Al₂(OH)₄Si₂O₅·nH₂O) which is also characterized for a hollow nanotube structure similar with CNTs and titania nanotubes as the support for manganese oxides has not yet been investigated.

In this paper, a mild in-situ deposition method operated at room temperature was introduced to prepare a novel nano-sized MnO_x/HNTs catalysts. The catalysts thus obtained exhibited ultra-high low temperature catalytic activity for cleaning NO with NH₃ from 60 to 300 °C.

2. Experimental

2.1. Catalyst preparation

Halloysite (HNTs) from golden sun ceramics Co. Ltd. (Zhengzhou, China) was used as support of the catalysts in this work. Chemical composition and Textural property of HNTs were listed in Table 1.

MnO_x/HNTs catalysts were prepared by in-situ deposition method (SP) at room temperature as illustrated in Fig. 1. Catalyst with MnO_x as active component was prepared by impregnating HNTs with MnAC·4H₂O solution (1 mL/g HNTs), after 24 h of impregnation, the KMnO₄ solution was gradually added with thorough stirring. The mixtures were placed at room temperature for 8 h and then filtered and washed with de-ionized water 3–4 times. The resulting solid products were dried at 70 °C for 6 h and 110 °C for 12 h, respectively. For comparison, the palygorskite (PG) supported MnO_x catalysts were also prepared by in-situ deposition method. The reaction equations of KMnO₄

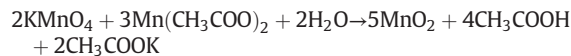
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Table 1
Physical properties of HNTs.

Chemical composition (wt%)						Textural property		
Na ₂ O	MgO	Al ₂ O ₃	SiO ₂	K ₂ O	CaO	Fe ₂ O ₃	S _{BET} (m ² /g)	V _{total} /(cm ³ /g)
0.8	0.2	36.47	42.92	0.91	0.61	1.24	86.362	0.291

and MnAC·4H₂O as follow:



For the comparison of preparation method of in-situ deposition and traditional wet-impregnation followed by calcination, MnO_x catalyst was also prepared by incipient-wetness impregnation method (DP). Carrier of HNTs was firstly impregnated with MnAC·4H₂O solution (1 mL/g HNTs), and then placed at room temperature for 8 h. The impregnated catalyst was dried at 70 °C for 6 h and 110 °C for 12 h, finally calcined in air flow at 300 °C for 2 h.

In this work, a series of MnO_x-supported catalysts based on those two support materials with 8 wt% MnO_x loading were prepared by two preparation routines. Catalysts prepared with different supports and methods were designated as MnO_x/HNTs-SP, MnO_x/PG-SP and MnO_x/HNTs-DP, and MnO_x/PG-DP as reported previously by Zhang [23]. All samples were crushed and sieved to a 20–40 mesh size for activity measurement.

2.2. Catalytic activity measurements

The SCR activity tests were conducted in a fixed-bed quartz reactor (i. d. 14 mm). The simulated flue gas consisted of 600 ppm NH₃, 600 ppm NO, 3 vol% O₂ and Ar gas as the balance. The total flow rate of 350 mL/min was maintained for all tests. Different gas hourly space velocities (GHSV) from 9000 h⁻¹ to 36,000 h⁻¹ were achieved by changing the volume of catalyst. The gas concentrations of NO were measured online by a flue gas analyser (Testo350-XL). All the dates were collected after 40 min when the SCR reaction reached a steady state.

2.3. Catalyst characterization

The chemical composition was analyzed by X-ray fluorescence (XRF-1800). Brunauer-Emmett-Teller (BET) analysis was conducted with a specific area and pore analyser (NOVA 2200e, Quantachrome, USA). Transmission electron microscopy (TEM) images of the samples were obtained by a JEM-2100F (Hitachi, Japan) analyser. Energy-

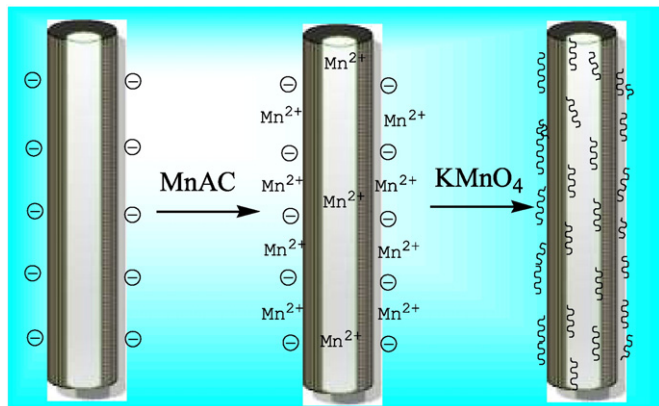


Fig. 1. Schematic illustration of the synthesis process of in-situ deposition method.

dispersive X-ray spectroscopy (EDX) was used to determine the element content on the sample surface. The powder X-ray diffraction (XRD) measurement was carried out on a Rigaku D/Max2500V system with Cu Kα radiation.

3. Results and discussion

3.1. Catalytic activity

In order to investigate the effect of different experiment method and carrier material on catalyst's SCR activities, the activities tests of four different catalysts with 8 wt% MnO_x loading were given at the same space

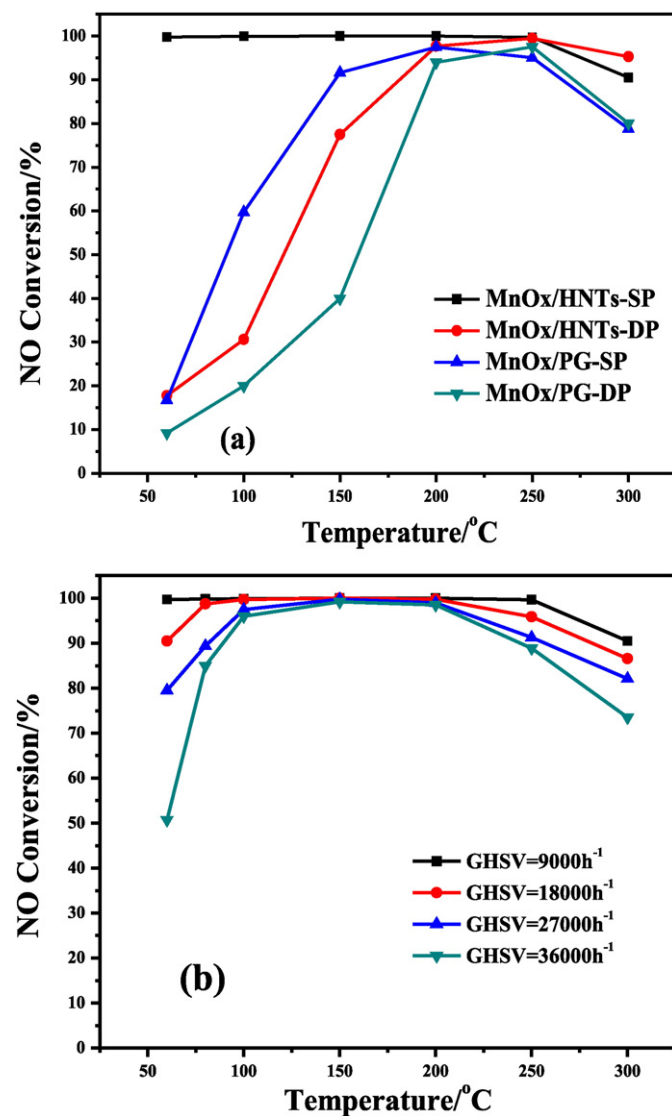


Fig. 2. (a) NO conversions for MnO_x catalysts prepared by different methods and carriers at the space velocity of 9000 h⁻¹ and (b) NO conversion over MnO_x/HNTs-SP with different GHSV. Reaction conditions: [NO] = [NH₃] = 600 ppm, [O₂] = 3 vol.%, N₂ as the balance gas, total flow rate = 350 mL/min.

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