



Catalytic reduction of nitric oxide by carbon monoxide over coal gangue hollow ball



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ABSTRACT

The catalytic effect of coal gangue hollow ball with open pores on NO reduction by CO in fluidized bed was investigated, and the kinetics for the catalytic reduction of NO using CO was evaluated. In the absence of water vapor at 773–923 K, the reaction rate constant was

$$k = 5210 \exp(-113600/(RT)).$$

NO reduction ratio rapidly increased with reaction residence time at initial stage (<1 s) and $T > 873$ K; high NO reduction ratio was achieved by increasing reaction temperature when the initial concentration ratio of CO to NO was less than 4. In the presence of water, the catalytic effect of coal gangue hollow ball on NO reduction was weakened, and the reaction rate constant was

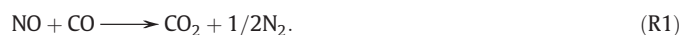
$$k = 0.31 \exp(-60750/(RT)).$$

NO reduction ratio is less than 0.2 when reaction residence time was 2.5 s at $T \leq 923$ K when the initial concentration ratio of CO to NO was 4, and the initial concentration ratio of CO to NO had negligible effect on NO reduction ratio at $T \leq 923$ K.

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1. Introduction

Emissions of nitrogen oxides (NO_x) are of great concern because of their impacts on the environment, including acid rain and photochemical smog formation. NO_x is the major combustion-generated pollutant from fossil fuel combustion [1]. The reduction of NO_x emissions has become one of the greatest challenges in environment protection. The most practical and convenient method for removing nitric oxide (NO) is to use unburned compounds such as carbon monoxide (CO) and hydrocarbons as reducing agent in flame [2]. The use of CO as reducing agent has advantages for NO_x reduction in practical application because of its presence in significant amounts in combustion process, CO oxidation is carried out by NO reduction, and NO removal is usually accomplished via reduction to form N_2 [3]:



The key steps in the reaction are the breaking of the NO molecule and a subsequent CO oxidation.

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The catalytic reduction of NO_x by CO has been one of the most important scientific challenges during the last few decades. For fluidized bed combustion of coal, several authors found that the rate of NO reduction increased significantly in the presence of CO, and solids presenting in a fluidized bed, such as calcined limestone (CaO), alumina (Al_2O_3) and sand (SiO_2), were found to catalyze NO reduction by CO [4–8]. It is well known that SiO_2 , Al_2O_3 and TiO_2 have interesting capacities as catalyst support for NO_x reduction, and base metal oxide catalysts are used to reduce NO by CO [9,10]. Tokio Iizuka found that the Fe_2O_3 supported on SiO_2 was the most active catalyst among a series of Fe_2O_3 catalysts for NO_x reduction [11]. Acke et al. [12] found that CaO was capable of catalyzing the NO reduction by CO and the effectiveness depended on its catalytic activity and surface. Water vapor is inevitable in the flue gas of hydrocarbon fuel combustion, and it influences NO catalytic reduction by CO [13].

Coal gangue is a main industrial solid waste, and the chemical compositions are SiO_2 , Al_2O_3 , Fe_2O_3 , CaO and some impurities, and these base metal oxides can be used as catalysts for NO reduction by CO. However, coal gangue has high sulfur low specific surface area, and the catalysis for NO reduction is not noticeable and results in high SO_x emission. If coal gangue is prepared to hollow ball and sintered under oxidizing condition, the catalysis for NO reduction is enhanced and no SO_x emissions.

The goal of the paper is to investigate the catalytic effect of coal gangue hollow ball with open pores on NO reduction by CO in fluidized bed and to evaluate the kinetics for the catalytic reduction of NO using CO.

2. Experimental

2.1. Structural properties of coal gangue hollow sphere with open pores

In the experiment, coal gangue hollow ball with open pores is used as catalyst for NO reduction by CO in fluidized bed reactor. The preparation process of coal gangue hollow ball with open pores includes the following steps: coal gangue is smashed into 0.1–10 μm ; the powders are mixed with water, Y_2O_3 sintering additive and carboxymethyl cellulose dispersing agent (CMC) to make slurry by mechanical stirring for 12 h, followed by the gradual addition of dissolved propyl gallate/TritonX-114 in the slurry, and the suspension foam slurry is prepared [14]; the stable foam slurry is rapidly atomized and dried, and then dry coal gangue hollow balls are prepared; the dry sphere is sintered at 1223 K under oxidizing condition for 2 h, and the microstructures are shown in Fig. 1. Field emission-scanning electron microscope (FE-SEM) images were recorded using a JSM-6701F instrument. Known from FE-SEM image of inner structure of hollow ball, there are many smaller hollow balls in the coal gangue hollow ball with open pores, thus the ball has high specific surface area (SSA). SSA (m^2/g) was calculated by the BET method from the N_2 adsorption/desorption isotherms obtained at 77 K [15]. The properties of hollow balls are shown in Table 1, and there are no C and S residuals due to the sintering oxidization of hollow balls at 1223 K for 2 h before experiments.

The chemical compositions of sintered coal gangue hollow ball were measured by Model 1800 X-ray fluorescence (XRF) analyzer, and the

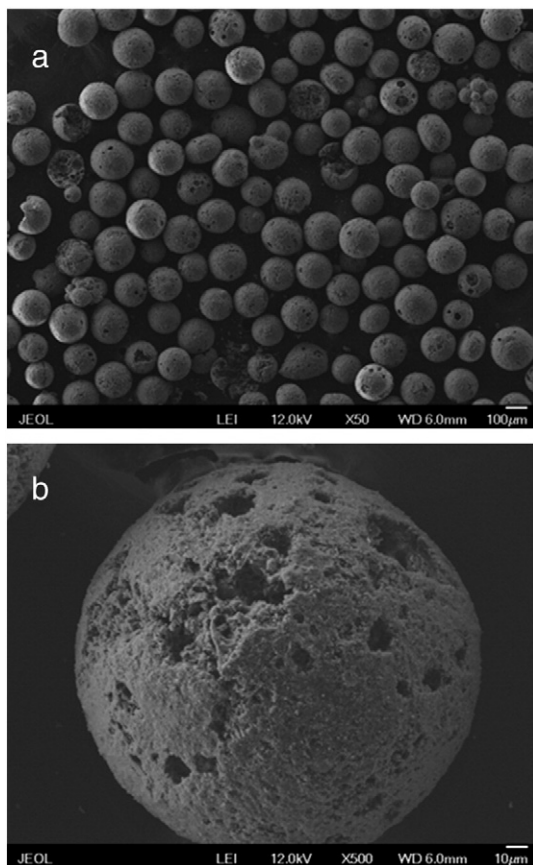


Fig. 1. FE-SEM images showing the microstructures of coal gangue hollow ball (a) and hollow ball with open pores (b).

Table 1

BET surface, average ball size and density of coal gangue hollow ball.

Specific surface area (cm^2/g)	Average ball size (μm)	Real density (kg/m^3)
253.9	210	2320

result is shown in Table 2. The coal gangue hollow ball consists of SiO_2 , Al_2O_3 , Fe_2O_3 , CaO , TiO_2 and K_2O .

2.2. Apparatus

Fig. 2 shows a schematic diagram of the experimental apparatus, and it consists of a gas assembly, reactor, gas analyzer and a temperature controller. The quartz reactor is placed inside of the electrically heating furnace, its inner diameter and height are respectively 25 mm and 600 mm, and the height of valid reaction zone is 300 mm. A gas distributor made of sintered quartz was mounted at the lower part of the reactor, which was located at 200 mm from the reactor inlet. The gas components were provided by high pressure cylinders of argon ($\text{Ar} \geq 99.99\%$), carbon monoxide ($\text{CO} \geq 99.99\%$), and standard NO prepared in advance (5% NO, Ar balance). The gas flow of every component gas was controlled precisely by mass flow meters, gas concentrations were adjusted by mass-flow controllers, and gases were mixed in a gas mixer. In order to investigate the effect of H_2O on catalytic reduction of NO, the gas mixture went through liquid H_2O contained in a bottle, and the relative humidity and temperature of gas mixture at the outlet were 84% and 305 K, respectively. The gas was analyzed with Flue Gas Analyzer (Ecom-J2KN) manufactured by Prism Gas Detection Pvt. Ltd. The maximum relative error for the measured species was 5%.

In the experiment, the initial concentration ratio of CO to NO is important for NO reduction, and the ratio (ε) is defined as

$$\varepsilon = \frac{[\text{CO}]_{\text{in}}}{[\text{NO}]_{\text{in}}} \quad (1)$$

where $[\text{CO}]_{\text{in}}$ and $[\text{NO}]_{\text{in}}$ are the CO and NO concentrations at the reactor inlet (ppm), respectively. $[\text{NO}]_{\text{in}}$ was kept constant (315 ppm), and ε ranged from 1 to 15.

In order to describe the extent of NO reduction, the conversion (x) is defined as

$$x = \frac{[\text{NO}]_{\text{in}} - [\text{NO}]_{\text{out}}}{[\text{NO}]_{\text{in}}} \quad (2)$$

where $[\text{NO}]_{\text{out}}$ is the NO concentration at the reactor outlet, ppm.

Gas reaction residence time (t) is defined as

$$t = \frac{V}{F} \times \frac{T_f}{273.15} \quad (3)$$

where F is the gas flow rate at 101.325 kPa and 273.15 K, and it is kept constant, $2 \times 10^{-5} \text{ m}^3/\text{s}$ in the experiment; V is the reactor volume of the valid reaction zone, m^3 ; T_f is the reaction temperature, K. Gas reaction residence time ranges from 1.5 to 2.6 s.

3. Results and discussion

3.1. Reaction scheme and rate measurement

Fig. 3 shows NO and CO concentration variations at reactor inlet and outlet, and the results indicate that the consumption of carbon monoxide coincides approximately with that of the nitric oxide:

$$[\text{NO}]_{\text{in}} - [\text{NO}]_{\text{out}} = [\text{CO}]_{\text{in}} - [\text{CO}]_{\text{out}} \quad (4)$$

The initial and final chemical compositions of sintered coal gangue hollow balls were not changed in the experiment of catalytic reduction

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