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Research article

Simultaneous SO_2/NO removal performance of carbide slag pellets by bagasse templating in a bubbling fluidized bed reactor

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ABSTRACT

Carbide slag pellets fabricated by bagasse templating using extrusion-spheronization were proposed to simultaneously remove SO_2/NO from flue gas. The effects of the reaction temperature, O_2 concentration, mass ratio of bagasse to carbide slag and particle size on the simultaneous SO_2/NO removal performance of carbide slag pellets were investigated in a bubbling fluidized bed reactor. CaO in the carbide slag pellets has a significant promotion effect on NO reduction by CO. SO_2 and NO are completely removed by the carbide slag pellets at 825–875 °C. The presence of O_2 in the reaction gas results in an adverse effect on NO removal by carbide slag pellets, while it leads to a positive effect on SO_2 removal. With an increase in the mass of bagasse in the pellets, the duration for the complete NO removal becomes longer and the porosity of the pellets appears higher. Carbide slag pellets with a smaller particle size possess a higher NO removal capacity. The higher mass ratio of bagasse to carbide slag leads to a larger surface area and pore volume of the pellets during SO_2/NO removal process but results in a slight decrease in the attrition resistance. Carbide slag pellets by bagasse templating for simultaneous SO_2/NO removal seems promising in the industrial application.

1. Introduction

Currently, the excessive emission of sulfur dioxide (SO₂) and nitrogen oxides (NO_x) from coal-fired power plants have produced serious negative effects on the environment and human health, especially in China, where approximately 45% of the coal is used for electric power generation [1]. In 2015, the Chinese government issued comprehensive implementation of ultra-low emissions and energy-saving schemes for coal-fired power plants. The concentrations of SO₂ and NO_x should be controlled below 35 and 50 mg/Nm³, respectively. Currently, the mature technologies for SO₂ and NO removal in coal-fired power plants mainly include wet flue gas desulfurization (WFGD) [2-4], selective catalytic reduction (SCR) [5-7], selective noncatalytic reduction (SNCR) [8–10] and circulating fluidized bed (CFB) technology [11,12]. The system of SCR-WFGD is a conventional post-combustion control strategy. However, it has some serious potential problems such as separate removal of SO₂ and NO_x, a complicated system for SO₂ and NO_x, a costly SCR catalyst, easy poisoning of the SCR catalyst, and so on [13,14]. Therefore, it is worth developing an effective method for efficient and simultaneous removal of SO2 and NO with a low-cost and

simplified system.

Carbide slag (CS) is an industrial solid waste that is mainly composed of Ca(OH)₂. It is a by-product of acetylene production by the hydration of calcium carbide [15]. More than 40 million tons of CS is produced in the world every year [16]. CS was investigated as an SO₂ sorbent in the desulfurization process of flue gas. Wu et al. [17] studied cyclic SO₂ and CO₂ adsorption by CS and limestone in a dual fixed-bed reactor and found that CS possessed higher SO2 adsorption capacity than limestone at 700 °C. Cheng et al. [18] investigated the desulfurization properties of three calcium and sodium industrial wastes (white lime mud, CS and brine sludge) via a fixed-bed reactor, and they found that CaO derived from CS possessed the most SO₂ diffusion channels and the highest specific surface area among the three industrial wastes, which was beneficial for SO₂ removal. Thus, CS may be a promising SO₂ sorbent owing to its low price and high efficiency of desulfurization performance. Furthermore, CS has also been investigated as a CO₂ sorbent during the calcium looping process for CO₂ capture [19–21].

The CFB flue gas desulfurization technology is a promising dry flue gas desulfurization process with the competitive advantages of lower investment, simple operation and high removal efficiency in

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comparison with wet flue gas desulfurization processes [22]. However, raw CS particles are fine powders. Most CS particles are less than 100 μ m, which are not suitable for CFB flue gas desulfurization technology because the fine powder is easily elutriated from CFB [23]. Scala et al. [24–26] pointed out that the attrition of calcium-based sorbent resulted in an important change in its particle size distribution in a fluidized bed reactor for SO₂ capture, which caused the direct elutriation of the sorbent from the reactor. Thus, CS should be granulated to form pellets with a suitable particle size and good mechanical strength for application in a fluidized bed reactor.

Many granulation methods for fine particles have been previously reported. The conventional methods include extrusion [27-30], sprav water pelletization [31–33] and extrusion-spheronization [34–37]. CS pellets fabricated by the extrusion-spheronization granulation technique are characterized by high mechanical strength and uniform particle size distribution as a CO₂ sorbent in the calcium looping process [37]. However, this technique may also lead to the densification of the pellets and cause a loss in the surface area of CS pellets, which is not beneficial for CO_2 capture [35]. Hence, the pellets fabricated by the extrusion-spheronization granulation technique need pore-forming materials to act as templates to enhance their porosity. Sun et al. [38] chose five biomass materials (i.e., microcrystalline cellulose, corn starch, rice husk, sesbania powder and lycopodium powder) as poreforming materials to fabricate calcium-based pellets via an extrusionspheronization method to obtain calcium-based pellets with good porosity for CO₂ capture in the calcium looping process. Xu et al. [37] prepared porous spherical calcium-based sorbents by templating with bamboo for CO₂ capture and found that the extrusion-spheronization process with 2 wt% bamboo templating was promising for the fabrication of CO₂ sorbents with good mechanical strength.

Bagasse, a waste from the sugar industry, is expected to have an output of 20 million tons in China every year. In this work, we propose bagasse as a template in the preparation of CS pellets fabricated by the extrusion-spheronization method, which are then used as a SO₂ sorbent in a fluidized bed reactor. Bagasse is chosen as a pore-forming material to enhance the porosity of the CS pellets for high SO₂ removal efficiency. The thermal degradation of bagasse at a high temperature can release pyrolysis gas, which may contribute to the formation of a porous structure in CS pellets. Moreover, the pyrolysis products derived from bagasse including char and reducing gas (CO, C_xH_y, etc.), may also have the function of reducing NO_x, which involves the heterogeneous reaction between the char and NO and the homogeneous reaction between the pyrolysis gas and NO. Previous reports pointed out that CaO had a catalytic effect on NO reduction by char and CO [39,40]. Zhong et al. [39] illustrated that the reaction activation energy of NO reduction by coal char was significantly decreased with the addition of CaO. Thus, CS pellets produced by bagasse templating may have a dual function, coupling SO₂ removal and NO_x reduction.

In this study, CS pellets prepared by bagasse templating using the extrusion-spheronization method were proposed to simultaneously remove SO₂ and NO in a bubbling fluidized bed reactor. The effects of the reaction temperature, O₂ concentration, and mass ratio of bagasse to carbide slag and particle size on the simultaneous SO₂/NO removal performance of CS pellets were investigated. The microstructural characteristics of the CS pellets were also studied.

2. Experimental

2.1. Material

Carbide slag was sampled from a chlor-alkali plant in Shandong Province, China. Table 1 shows the chemical components of CS analyzed by X-ray fluorescence (XRF). For the XRF analysis, the chemical components of the materials are measured in the form of oxides. In fact, the main composition of CS is $Ca(OH)_2$ rather than CaO. The CS samples were sieved to a particle size below 0.125 mm. Bagasse was obtained

Table 1			
Chemical	components	of CS	(wt%).

CaO	SiO_2	Fe_2O_3	Al_2O_3	K ₂ O	Cl	Others	Loss on ignition
66.98	2.83	0.23	1.11	0.18	1.43	0.05	27.19

from a cane-sugar plant in Guangxi Province, China. Bagasse was dried for 12 h at 100 °C and then sieved into a size fraction of 0.125–0.18 mm. Table 2 shows the proximate and the ultimate analysis of the bagasse.

2.2. Preparation of CS pellets by bagasse templating

CS pellets by bagasse templating were prepared by the extrusionspheronization method. The preparation process is presented in Fig. 1. First, some bagasse and 30 g of CS were well-mixed in a beaker. The mass ratios of bagasse to CS were specified as 0:100, 10:100, 20:100 and 30:100. Then, the well-blended mixture was stirred constantly as approximately 10-20 mL of deionized water was added. Second, the well-mixed CS and bagasse was sent to an extruder, where the rodshaped samples with a diameter of 1 mm were produced. Third, the rodshaped samples were placed in a spheronizator to form the spherical CS pellets using the bagasse templating. Finally, the obtained CS pellets were dried for 12 h at 100 °C. The spherical CS pellets with a particle size of 1.0-1.4 mm were thus obtained. To reveal the role of CS in the process of the simultaneous removal of SO₂ and NO, bagasse pellets without the addition CS were fabricated with the particle size of 1.0-1.4 mm by the extrusion-spheronization method according to the above-described procedure. To investigate the effect of the particle size on SO₂/NO removal by the CS pellets, CS pellets with a larger particle size (1.4-2.3 mm) were also prepared at a mass ratio of bagasse to CS of 10:100. CS pellets with a particle size in the range of 1.0-1.4 mm were used in the following research, except for the research on the effect of particle size. CS pellets by bagasse templating are denoted as CS-BX, where X denotes that the mass ratio of bagasse to CS is X:100. For instance, CS-B10 denotes CS pellets with a mass ratio of bagasse to CS of 10:100.

2.3. SO₂/NO removal by CS pellets

The SO₂/NO removal performance of CS pellets was investigated in a bubbling fluidized bed reactor (BFBR) operated at 775-925 °C, as shown in Fig. 2. The BFBR consists of a quartz tube with an inside diameter of 30 mm inserted into an electric heating furnace. Four thermocouples were used to detect the temperature of the BFBR and to control the temperature of the electric heating furnace. The reaction gases included 15% CO₂, 300 ppm SO₂, 500 ppm NO, O₂ and N₂. The total flow rate of the reaction gas was 2 L/min, which was controlled by mass flowmeters. During each test, the BFBR was first heated to a specified temperature and the reaction gases from different gas cylinders were sent to the BFBR after preheating. After the concentrations of the reaction gases in BFBR were adjusted to the specified values for 2 min, 4 g of CS pellets were then added to the BFBR through a hopper on the top of the reactor. Meanwhile, the concentrations of the reaction gases were detected and recorded by a Testo 350 flue gas analyzer. SO $_2$ removal efficiency (η_{SO2}) and NO removal efficiency (η_{NO}) were calculated according to the concentration changes of SO₂ and NO as follows:

$$\eta_{\rm NO} = \left(1 - \frac{C_{\rm NO}(t)}{C_{\rm NO}(0)}\right) \times 100\%$$
(1)
$$\eta_{\rm SO_2} = \left(1 - \frac{C_{\rm SO_2}(t)}{C_{\rm SO_2}(0)}\right) \times 100\%$$
(2)

where *t* denotes the reaction time (*s*); C_{NO} (*t*) is the NO concentration at time *t* in the exhaust gas from the BFBR (ppm); C_{NO} (0) refers to the

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