



Short communication

Lignin templated synthesis of porous carbon–CeO₂ composites and their application for the photocatalytic desulphuration



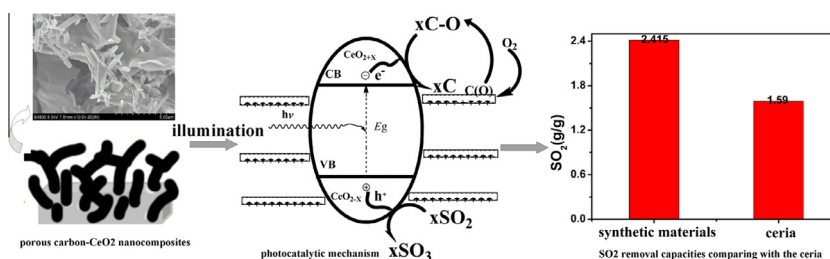
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HIGHLIGHTS

- Lignin was used as template for the synthesis of porous carbon–CeO₂ composites.
- The prepared porous carbon–CeO₂ composites were used for the photocatalytic removal of SO₂.
- The possible formation mechanism of carbon–CeO₂ was proposed.
- Desulfurization mechanism of porous carbon–CeO₂ composites was also proposed.

GRAPHICAL ABSTRACT



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ABSTRACT

In this work, lignin was used as template for the synthesis of porous carbon–CeO₂ composites via the calcination method. The structure and morphology of composites were investigated by X-ray powder diffractometer, scanning electron microscope and high-resolution transmission electron microscopy. The characterized results indicated that porous carbon was formed due to the decomposition of lignin, and CeO₂ nanorods are grown uniformly in the porous carbon to form porous carbon–CeO₂ composites. The prepared porous carbon–CeO₂ composites were used for the photocatalytic removal of SO₂ at room temperature under light irradiation. The photocatalytic activity of carbon–CeO₂ was greatly enhanced compared with CeO₂. The maximum of photocatalytic conversion of SO₂ can be about 51.89% much higher than that of CeO₂. The possible mechanism for the photocatalytic oxidation of SO₂ was proposed based on the experimental results, and the prepared composites may have potential application for the SO₂ removal in actual life.

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1. Introduction

SO₂ emissions have attracted much attention due to their detrimental effects on either human health or the environment. The release amount of SO₂ is huge every year. For example, only in 2010, over 100 Tg were released into the air, as reported in the literature [1]. Much effort has been developed to remove SO₂. Traditional methods for the removal of SO₂ are flue gas desulfurization [2,3], lime/limestone-based process [4,5], ionic liquid

absorption [6,7] and hydrotalcite-like material adsorption [8]. However, there remained several drawbacks in these techniques, such as high costs, complex equipment, large occupation of land area, large consumption of fresh water, and formation of secondary pollutants [9]. Recently, carbonaceous materials have been developed for physical adsorption SO₂, including activated carbon [10], coke [11], semicoke [12,13], and activated carbon fibers [14]. However, physical adsorption cannot completely eliminate SO₂ from the environment.

Combination of absorbing materials and catalytic oxidation materials has been reported to be an effective method for completely removing SO₂ from the environment. For example, Yan et al. synthesized nanosized CeO₂ particles loaded on activated

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carbon via a single-step hydrothermal process [15]. Their results demonstrated that adsorption and catalytic oxidation of SO_2 could be remarkably enhanced, indicating that carbon supported materials play an important role in the enhancement of SO_2 removal. Carbon-based materials have become important and promising materials due to their various advantages, such as cost-effective, low density, easy accessibility, high absorption capacity and high stability [13,16–18]. Carbon materials can be obtained by different raw materials. Lignin, one of the carbon raw materials, has aroused growing interest recently. However, Lignin is difficult to be exploited due to its complicated three-dimensional network [19], which usually acted as waste to discharge from paper factory, thus causing huge environmental pollution. Therefore, lignin as a raw material to prepare carbon materials would be an important way to utilize lignin.

In this work, to reduce its pollution to the environment and improve its utilization efficiency, lignin was used as template to in situ synthesize porous carbon– CeO_2 composites for SO_2 removal. The calcination temperature, calcination time and ratio of precursors are discussed in the experiment. Additionally, compared with CeO_2 , the catalytic and oxidizing ability of the composites for SO_2 was also discussed. The provided method not only utilizes lignin efficiently resulting in decreasing pollution of lignin to the environment, but also removes SO_2 from the environment, which may have potential applications for the environment pollution remedy.

2. Experimental section

2.1. Synthesis process

Carbon– CeO_2 composites were successfully prepared in different concentrations, temperature and time. During a typical procedure to obtain the carbon– CeO_2 structures, certain amounts of $\text{Ce}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ and sodium lignin sulfonate (reagents in Supporting Information) were weighed on the analytical balance, and then grinded and blended on the agate stone in dry condition. Herein, the mixture was dried using the lamp light in the whole grinding process. The mixture of processed was heated at a certain temperature for hours in the static air with a ramp rate of $5^\circ\text{C}/\text{min}$ in the tube furnace. The calcination temperature was $400\text{--}600^\circ\text{C}$, and the rate of sodium lignin sulfonate to cerium nitrate varied from 1:1 to 2:1 in this study (Table S1, the Supporting Information). In

addition, the samples were characterized (characterization in Supporting Information).

2.2. Catalytic activity measurement

Desulfurization capacity was detected by a simple reaction device at room temperature as shown in Fig. 1. A 0.1 g sample was dispersed in 20 mL deionized water in a suction filter bottle with SO_2 generating device, and the whole reaction was kept stirring. SO_2 was obtained by reacting dilute sulfuric acid with sodium sulfite, and was blown into the catalytic device to ensure saturation during the experimental process. Tail gas treating unit using sodium hydroxide as absorbent was added on the catalytic device. After the sample was added into the device, the device was immediately put under the sunlight. Then, 1 mL sample solution was taken out every 15 min once. The samples were centrifuged to obtain white precipitates. The efficiency of eliminating SO_2 is calculated from the quantity of oxidizing SO_2 to sulfate anions. The samples were further processed (the samples post-processing in Supporting Information).

3. Results and discussion

3.1. Characterizations of the products

The crystal structures and phase purities of the samples were investigated by X-ray powder diffraction. The XRD pattern (Fig. 2a) shows that all the marked peaks are CeO_2 peaks and the other peaks are carbon peaks, which correspond to the PDF cards (43-1002) and (46-0943), respectively. It can be seen that carbon– CeO_2 composites are uniformly formed under different calcination temperatures and calcination times from Fig. 2b. XRD results demonstrate that the prepared materials are carbon– CeO_2 composites.

SEM was conducted to observe the morphology of the carbon– CeO_2 structures. Fig. 2c and d shows typical SEM images of carbon– CeO_2 obtained at 400°C for 4 h. The carbon– CeO_2 architectures were constructed by porous carbon and intersectional CeO_2 nanorods. Obviously, the products present porous and rod-like mixed morphology. The porous structure was presented in Fig. 2e. The porous structure can be clearly observed, and CeO_2 nanorods can also be seen in the internal of the porous carbon. These results indicate that the intersectional CeO_2 nanorods grow on the porous carbon wall. To further investigate the morphology and crystallographic features of the carbon– CeO_2 composites, HRTEM image was recorded (Fig. 2f). We calculated a lattice spacing of 0.31 nm, corresponding to the [1 1 1] plane of the CeO_2 . The XRD pattern also showed the strong intensity of the [1 1 1] peak. These results demonstrate that each plate that forms the carbon– CeO_2 composites has a well-defined structure.

The laser-spectroscopy was conducted as shown in Fig. 3a. It was found that about 34% lies in 0–500 nm fraction, the relatively high ratio of size distribution can also be observed in 500–5800 nm fraction, which is consistent with what was observed in SEM image.

The N_2 -adsorption test was also conducted in our experiment. Nitrogen adsorption isotherm is a standard procedure for the determination of porosity of carbonaceous adsorbents. In Fig. 3b, it can be observed that the isotherm of the composite presents IV shape as defined by IUPAC classification, associated with a mesoporous structure. The surface area is $58.80\text{ m}^2\text{ g}^{-1}$, the pore volume is $0.1121\text{ m}^3\text{ g}^{-1}$, and the pore size is 3.77 nm. As a comparison, the carbon calcinated by lignin at the same conditions has the surface area of $3.139\text{ m}^2\text{ g}^{-1}$, pore volume of $0.008\text{ m}^3\text{ g}^{-1}$, average pore size of 2.637 nm. This clearly indicated that the carbon calcinated by lignin without adding $\text{Ce}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ has almost no porous

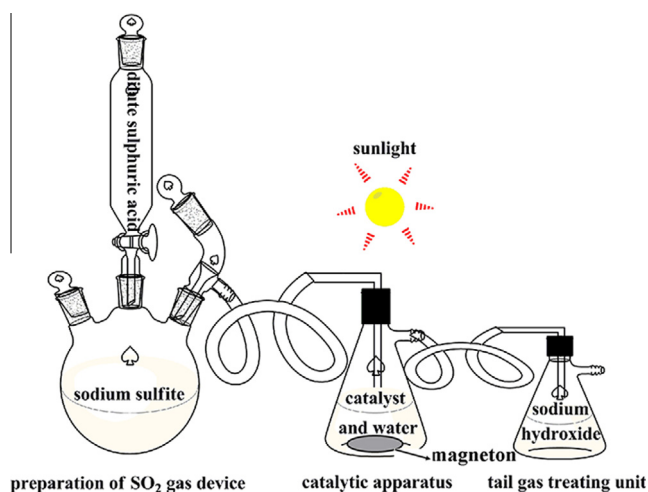


Fig. 1. Experimental setup.

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