



MgO-based adsorbent achieved from magnesite for CO₂ capture in simulate wet flue gas

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ARTICLE INFO

Article history:

Received 15 August 2017

Revised 11 January 2018

Accepted 6 February 2018

Available online 9 March 2018

Keywords:

Magnesite

Calcination

CO₂ adsorption performance

Wet flue gas

ABSTRACT

MgO based adsorbent for CO₂ capture can be achieved quite easily from the abundant, low cost and non-toxic magnesite (main ingredient MgCO₃). Water vapor was also added to simulate real flue gas. By investigation, the CO₂ capacity of magnesite calcined at 550 °C for 4 h can reach 1.82 mmol/g under the adsorption condition of 60 °C, 0.4 Mpa, 100 mL/min 10% CO₂ flow rate in the present of water vapor. And the cycling test results indicated that the adsorption capacity kept still 0.96 mmol/g after eighth cycles, showing the adsorbent had preferably cycle stability. XRD shows that the magnesite nearly don't decompose at 450 °C, higher temperature is needed to make magnesite decompose completely. The BET results verify that the magnesite calcined at 450 °C nearly has no pore structure. Under 500 °C, the BET surface is 122.8 m²/g. At 550 °C, the BET has a slightly decline, 118.6 m²/g, which indicates that the sintering occurred. The CO₂ adsorption capacity not only relates to the BET, but also the effect of steam condensation. The results of FT-IR and CO₂-TPD indicate that the CO₂ can be adsorbed on calcined magnesite successfully mainly by chemisorption, but the physical adsorption also exists.

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1. Introduction

Global warming induced by greenhouse gas CO₂ has caused public attention widely in recent years [1]. As of April 2016, the CO₂ concentration in the atmosphere has reached to its historically highest level of 405 ppm, which is 27% higher than that in 1960. With this rate, CO₂ concentration will be more than 700 ppm by the end of this century, which is very serious [2]. In order to solve the greenhouse phenomenon, extensive managements have been proposed to reduce CO₂ emission. CO₂ capture and storage (CCS) is regarded as the most practical approach for meeting the challenging goal of mitigating 20% of world emission from energy by 2020, while the key for this technology is to search a green adsorbent with high performance [3]. With the development of adsorbent, many of the new type adsorbents appeared, such as ionic liquids [4], MOFs [5–7], alkali metal oxide [8], activated carbon [9], porous materials [10,11] etc. [12–15]. Among them, alkali metal and alkaline oxide, especially MgO, CaO based sorbents, are studied as CO₂ adsorbents mostly due to abundant sources, low cost, as well as non-toxicity [16]. Compared with CaO adsorbents, MgO has advantages as it requires low regeneration temperature to regenerate MgCO₃ to MgO [16]. Until now, many types of MgO have been re-

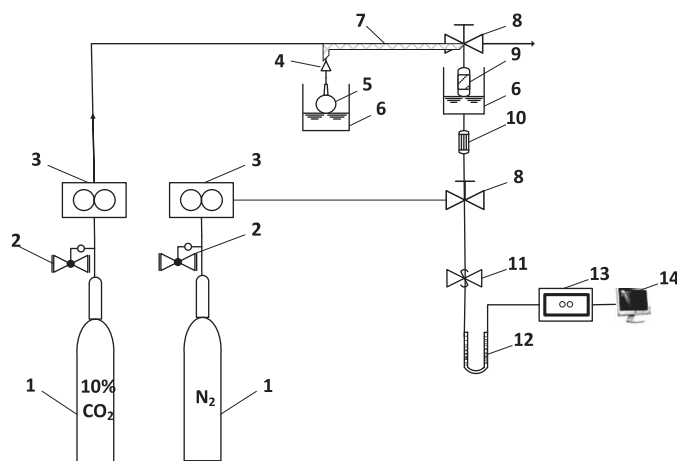
ported, e.g. MgO/Al₂O₃ [17], mesoporous MgO [18], MgO-TiO₂ [19], MgO-SiO₂ [20], MgO-C [21], MgO-ZrO₂ [22], and so on. Besides, in order to implement the circulation of the sorbents based on MgO in the industry application, the cyclic CO₂ removal process has been indicated.

Many investigations have been carried out using nesquehonite [23], forsterite [24], dolomite [25] and other minerals [26] to capture CO₂ internationally. Magnesite is also a rich mineral resource in China. A lot of magnesite production processing belongs to primary products and resource consumption, which mainly consume resources to gain economic benefits [27]. The MgO based adsorbent can be easily achieved by calcining abundant, low cost and non-toxic magnesite. For the CO₂ adsorbents, except for the capture capacity, another important issue is the cost. The simple preparation process of the MgO-based adsorbent derived from magnesite can not only reduce the cost, but also reduce the energy consumption, which is available and important to real industrial application. Besides, to simulate the real flue gas, the CO₂ capture capacities of calcined magnesite were tested systematically in the present of water vapor. Finally, the MgO-based adsorbent derived from magnesite also has the benefits of solving the problem of recycling difficulty and inconvenience for industrial application of liquid adsorbent. All of above are the benefits of the research for industrial applications.

The CO₂ capture capacity of magnesite on different adsorption conditions and the CO₂ adsorption and desorption cycling

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1-Gas cylinder; 2-Pressure regulator; 3-Gas mass flow controller; 4-One-way valve; 5-Flask;

6-Water bath pot; 7-Heat reserving belt; 8-Three-way valve; 9-Adsorber; 10-Filter;

11-Counterbalance valve; 12-Dryer; 13-Infrared gas analyzer; 14-Data collection.

Fig. 1. CO₂ adsorption experiment apparatus.

performance were evaluated in detail. Finally, the samples were thoroughly characterized using TG-DSC, FT-IR, XRD, CO₂-TPD and BET etc. to examine the structure.

2. Materials and methods

2.1. Materials and devices

The magnesite was obtained from Shan Dong Lai Zhou Magnesite mine. 10% CO₂(N₂ as balance gas) and 99.999% N₂ was supplied by Dalian Gas Co. Ltd. Gas mass flow controllers, D08-1D/ZM for N₂ and D08-1F for CO₂, were supplied by Beijing Sevenstar electronics Co. Ltd. Electric heating (DF-101S) was produced by Henan Yuhua Instrument Co. Ltd. Infrared gas analyzer (ANTARIS IGS Analyzer), was purchased from Thermo Scientific Instrument Co. Ltd. Circulating water bath pot (CS601) was produced by Shanghai Boxun Instrument Co. Ltd. Muffle furnace was produced by BangXi Technology Instrument Co. Ltd. Counterbalance valve, three-way valve and one-way valve were produced by Beijing Jiafa International Co. Ltd. Powder compressing machine (769YP-15A), were supplied by Tianjin Keqi Instrument Co. Ltd. Mortar, sieve, flask and adsorber were supplied by Yunnan Taifei Instrument Co. Ltd.

2.2. CO₂ adsorption and desorption test

The magnesite was calcined in muffle furnace. The resulting solid was pressed in the form of a pellet by powder compressing machine and further crushed into particles using a mortar. Screening of 0.2–0.4 mm diameter particle by sieve, the adsorbent was obtained.

CO₂ adsorption experiment apparatus is shown in Fig. 1. Firstly, N₂ with a flow rate 100 mL/min was used to purge the infrared gas analyzer for 15 min. 10% CO₂ with a flow rate 100 mL/min, which was used to simulate the real flue gas from industry, and steam from heating the water in the flask at 100 °C, were mixed to replace the gas in the path through the empty branch. When the flow was steady, by the control of the three-way valve, 10% CO₂ in the present of water vapor went through the adsorber with

3 g adsorbent, and the infrared gas analyzer was started to detect. The temperature and pressure were controlled by circulating water bath pot and counterbalance valve. The pipe between flask and adsorber should be as shorter as possible, and do heat preservation by insulation belt, to prevent the water vapor condensation. CO₂ desorption was carried out at 550 °C for 4 h with pure N₂.

2.3. Characterization instruments

Thermo-gravimetry and differential scanning calorimetry (TG-DSC) analyses were carried out on a SDT Q600 simultaneous thermal analyzer (TA Corp., America) under a flow of N₂ from room temperature to 800 °C at a heating rate of 10 °C/min.

X-ray fluorescence (XRF) was determined to analyze the composition of the adsorbent by AXIOS wavelength dispersive X-ray fluorescence spectrometer, (PANalytical B.V. Corp., Netherlands).

X-ray diffraction (XRD) was carried out on D/MAX-rA instrument (Rigaku Corp., Japan), with a Cu-target ka-ray. Nickel filter and scanning angle between 20 and 90° of 2θ at a scanning rate of 28 per minute were used.

Fourier transform infrared spectroscopy (FT-IR) spectra were recorded on an ALPHA-T Fourier Transform Infrared Spectrometer (BRUKER Corp., Germany). IR spectrometer in the wave number range between 4000 and 500 cm⁻¹ with a 2 cm⁻¹ resolution, and samples were diluted into KBr and pressed in the form of a pellet (diameter 13 mm).

The BET surface areas, and porosity analysis were achieved by N₂ adsorption/desorption using Tristar 3020 physisorption apparatus (MICROMERITICS Corp., America). BET specific surface area was calculated according to the Brunauer-Emmett-Teller (BET) model. Pore size distribution was determined from the adsorption branch of the isotherm using the Barrett-Joyner-Halenda (BJH) method. Total pore volume was calculated according to the N₂ amount adsorbed at a relative pressure of 0.99.

The basicity of the adsorbent was measured by CO₂ temperature-programmed desorption (CO₂-TPD) using Belcat II apparatus (Micortrac Bel Corp., America). Prior to the adsorption of CO₂, the adsorbent were heated at 550 °C for 4 h for cleaning the surface and the decomposition of magnesite. After cooling to

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