

CO₂ capture performance of a novel synthetic CaO/sepiolite sorbent at calcium looping conditions



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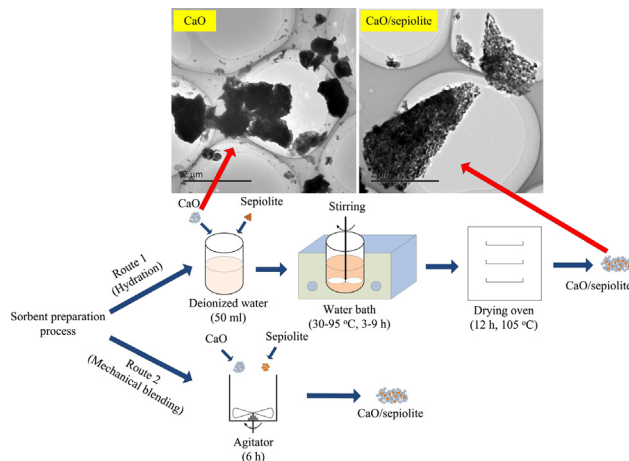
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HIGHLIGHTS

- A novel synthetic sorbent was fabricated from CaO and sepiolite by hydration.
- Hydration temperature has an important effect on CO₂ capture by CaO/sepiolite.
- MgO and Ca₂SiO₄ are formed in calcined CaO/sepiolite for hydration at 95 °C.
- MgO and Ca₂SiO₄ improve sintering resistance of CaO/sepiolite during cycles.

GRAPHICAL ABSTRACT



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ABSTRACT

A novel synthetic sorbent was fabricated from CaO and sepiolite by the hydration and its CO₂ capture performance was investigated during the calcium looping cycles. The effects of the sorbent preparation conditions including preparation method, hydration temperature, hydration duration and sepiolite content on CO₂ capture by synthetic CaO/sepiolite sorbent were examined in a dual fixed-bed reactor. The results showed that CaO/sepiolite possesses higher CO₂ capture capacity than original CaO. CO₂ capture capacity of CaO/sepiolite after 10 cycles is 39% and 56% higher than those of hydrated CaO and original CaO, respectively. The hydration temperature has an important effect on CO₂ capture by CaO/sepiolite during the preparation. When the hydration temperature is 95 °C, the obtained CaO/sepiolite exhibits the highest cyclic CO₂ capture capacity, because the good supports such as MgO and Ca₂SiO₄ are formed in the calcined CaO/sepiolite at 95 °C, which can improve the sintering resistance of CaO during the cycles. However, these supports are not found in the calcined CaO/sepiolite at the other hydration temperatures. In addition, CaO/sepiolite possesses more porous structure, larger surface area and pore volume, compared to hydrated CaO and original CaO. After the 1st calcination, the volume of pores in 10–100 nm in diameter of CaO/sepiolite is much higher than those of hydrated CaO and original CaO, respectively, which facilitates CO₂ capture of the sorbent. CaO/sepiolite appears promising as an effective and low-cost CO₂ sorbent at calcium looping conditions.

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1. Introduction

Worldwide, 30 billion tons of CO₂ are emitted into the atmosphere annually, with 40% from the coal-fired power plants [1]. Currently, as one of the most effective ways to solve the issue of lots of CO₂ emission from the coal-fired power plant, carbon capture and storage (CCS) has been adopted by the Intergovernmental Panel on Climate Change of the United Nation (IPCC) as the most crucial technological exploration to realize the goal of reducing the emission of greenhouse gases by 2050 [2–6]. Calcium looping, that is, the calcination/carbonation looping of calcium-based sorbent is considered to be one of the most potential CO₂ capture technologies for the industrial application [7–9]. It is reported that CO₂ capture capacities of calcium-based sorbents decrease gradually with the number of calcination/carbonation cycles due to the sintering of the sorbents [10–12]. Thus, the recent research has been laid on how to improve cyclic CO₂ capture performances of the calcium-based sorbents and enhance its sintering resistance [13–16].

Some methods have been used to improve CO₂ capture capacities of calcium-based sorbents, such as use of organic acids for the modification of sorbent [17,18], use of inert materials with high sintering resistance as supports of CaO [19–22], use of additives [14,23–25], pelletization [26], thermal pretreatment [27], discontinuously adding HCl in the carbonation step [28], prolonged carbonation [29] and hydration treatment [30,31], etc. Among these methods, adding the inert material as supports in calcium-based sorbent is an efficient method, producing the high-active synthetic sorbents with large surface area, high porosity and high sintering resistance. Al₂O₃ [32], ZrO₂ [19], CeO₂ [33], MgO [20,34–36], Y₂O₃ [37], MnO₂ [35,38], TiO₂ [39], CuO [40] and Nd₂O₃ [41] can be used as good supports. It should be noted that the preparation of the synthetic sorbents increases the cost. A compromise should be made between improving the sorbent performance and increasing its cost [8]. Chen et al. [42] used attapulgite as a support for calcined limestone, the carbonation conversion of the obtained synthetic sorbent contained 15 wt.% attapulgite after 20 cycles was 128% higher than that of the calcined limestone. Wang et al. [43] used the acid-leaching of metakaolin as a support to prepare the synthetic CaO/Ca₁₂Al₁₄O₃₃ sorbent and they found that under the severe calcination conditions (calcination at 950 °C under 100% CO₂), the carbonation conversion of the synthetic sorbent was 4 times as high as that of untreated CaO after 25 cycles. Li et al. [44] fabricated the synthetic sorbent with rice husk ash and CaO by hydration during the specific preparation process (hydration duration of 8 h, hydration temperature of 75 °C and SiO₂/CaO molar ratio of 1.0) and they found the maximum CO₂ capture capacity was 0.45 g/g after 20 cycles, which was twice as high as that of original CaO. Manovic et al. [45] prepared CaO/Ca₁₂Al₁₄O₃₃ sorbent by the calcium aluminate cement and CaO under 900 °C and the formation of Ca₁₂Al₁₄O₃₃ improved the sintering resistance the sorbent during long-term cycles. Chen et al. [46] produced a novel synthetic sorbent using fly ash and calcined limestone by hydration method, which exhibited higher CO₂ capture capacity than that of calcined limestone due to the formation of supports such as CaSiO₃ and Ca₁₂Al₁₄O₃₃ in the sorbent. Ma et al. [47] prepared the synthetic sorbent from carbide slag, aluminate cement and by-product of biodiesel by combustion method and they reported that the sorbent could achieve a CO₂ capture capacity of 0.62 g/g after 10 cycles, which was 2.2 times as high as that of carbide slag. Sanchez-Jimenez et al. [48] reported a synthetic sorbent fabricated from calcium nitrate and nanostructured silica by impregnation, which retained high CO₂ capture capacity during 50 cycles. Tian et al. [49] used steel slag and CaO by sol-gel method to prepare a synthetic sorbent and its CO₂ capture capacity was twice as high as that of CaO. Wang et al. [50] added 5 wt.%

diatomite into limestone to produce a synthetic sorbent by mechanical mixing and they pointed out that after calcination under an air atmosphere at 1000 °C, Ca₂SiO₄ was generated in the synthetic sorbent, which improved CO₂ capture capacity and sintering resistance of the sorbent. Valverde et al. [51] and Wang et al. [34] both thought that MgO in the calcined dolomite provided the inert skeleton, which was a good support to improve the sintering resistance and maintain cyclic stability of the sorbent.

Sepiolite is a natural mineral clay, the price is about \$43–110/t according to its quality [52], which has been widely applied in the building materials, cosmetics, mineral feed, catalysts, detergents and paper, etc. At present, the total reserves of sepiolite amounted to more than 8000 million tons in the world, which is widely distributed in Spain, China, United States, France, and so on [53–56]. As a clay mineral with a unit cell formula Si₁₂Mg₈O₃₀(OH)₄(OH₂)₄8H₂O [57,58], sepiolite contains a continuous two-dimensional tetrahedral sheet, however, it differs from other layer silicates in that they lack continuous octahedral sheets. Due to the structural morphology, the considerable attentions have been paid to sepiolite which is used as sorbent for organics, as support for catalysts, and so on [59,60]. For example, Liu et al. [61] used TiO₂ and CaO to prepare a new type of TiO₂/sepiolite composite to enhance the ability of photocatalytic elimination of formaldehyde by using the structural characteristics of sepiolite nanofibers, such as strong adsorption and high porosity. However, sepiolite as a support of CaO has never been reported in the calcium looping technology. Therefore, it is promising to use the structural morphology of sepiolite as a support to improve CO₂ capture performance.

In this work, a novel synthetic CO₂ sorbent was proposed, which was prepared from CaO and sepiolite by the hydration. CO₂ capture performance of the synthetic sorbent, i.e. CaO/sepiolite, was investigated at the calcium looping conditions. The effects of preparation method, hydration temperature, hydration duration and sepiolite content in CaO/sepiolite on the CO₂ capture by CaO/sepiolite were examined in a dual fixed-bed reactor. Also, the microstructures of the synthetic sorbent during the cycles were detected for better understanding its CO₂ capture performance.

2. Experimental

2.1. Preparation of sample

Calcium oxide (CaO ≥ 99.0% purity, size < 0.125 mm, Tianjin Kermel Chemical Reagent Co., Ltd., China) and sepiolite (sampled from Xiangtan in Hunan Province, China and sieved to size < 0.125 mm) were used to prepare the synthetic sorbent. The chemical components of the sepiolite detected by X-ray fluorescence (XRF) are shown in Table 1.

To study the influence of different production methods on the CO₂ capture performance of the synthetic CaO/sepiolite sorbent, two preparation routes were adopted: the hydration and the mechanical blending, as presented in Fig. 1. The hydration route is shown as follows: a certain amount of sepiolite and 1 g CaO were added to a beaker with 50 ml deionized water. The mixture was put into a thermostatic magnetic stirrer to stir for the different durations (1, 3, 6 and 9 h) under different hydration temperatures (30, 65 and 95 °C). The various sepiolite contents in CaO/sepiolite were specified as 5 wt.%, 8 wt.%, 10 wt.%, 12.5 wt.%, 15 wt.% and 20 wt.%, respectively. Then the beaker was placed in a drying oven for 12 h at 105 °C, and then the CaO/sepiolite sorbent by hydration was obtained. To examine the influence of sepiolite and hydration of CaO on the CO₂ capture behavior of the synthetic sorbent, the cyclic reaction performance of the hydrated CaO was also studied. The hydrated CaO was prepared according to route 1 as shown in Fig. 1, except for the addition of sepiolite.

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