

Amine-impregnated silicic acid composite as an efficient adsorbent for CO₂ capture

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HIGHLIGHTS

- A new efficient adsorbent was developed for CO₂ capture.
- A maximum CO₂ sorption capacity of 4.54 mmol/g was obtained.
- Effects of amine loading, adsorption temperature, and H₂O addition on CO₂ capture.
- The sorbent is regenerable and thermally stable.

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ABSTRACT

A new high efficiency inorganic–organic composite solid sorbent was developed by the simple impregnation of aminoethylethanolamine (AEEA) on the nanoporous silicic acid (SiO(OH)₂) for CO₂ capture from flue gas. Experimental results revealed that amine loading amount, adsorption temperature, and moisture addition could greatly affect the CO₂ adsorption capacity. At the optimal AEEA loading of 55 wt%, the CO₂ sorption capacity reached a maximum value of 4.54 mmol/g at 25 °C under 10 vol% CO₂ and 10 vol% H₂O. To the best of our knowledge, this is the first time that an AEEA based adsorbent has been reported as an efficient adsorbent for CO₂ capture. The AEEA impregnated SiO(OH)₂ also had good stability and reusability during cyclic adsorption/desorption tests, and the sorption capacity loss may be recovered by mixing condensed AEEA with cyclic sorbents. The Avrami's fractional order kinetic model was applied for the kinetic analysis of CO₂ adsorption and desorption on the optimized sorbent. The obtained activation energy for CO₂ desorption was 33.5 kJ/mol. The estimated regeneration heat duty on the optimized AEEA sorbent was 53.29 kJ/mol CO₂, a great energy penalty reduction compared to that of a typical aqueous monoethanolamine system.

1. Introduction

One of the major concerns of our society today is the climate change associated with greenhouse gas emission. A huge amount of CO₂ emission resulting from the use of fossil fuels has been considered as one of the major contributors to global warming [1,2]. Hence, the development of cost-effective technologies for the reduction of CO₂ emission is essential. CO₂ capture, utilization, and storage is one of the potential technologies for reducing anthropogenic CO₂ emissions [3]. The captured CO₂ can be used for enhanced oil recovery [4,5], supercritical CO₂ solvents [6], synthesis of chemical intermediates and fuels [7], etc. Among various CO₂ capture technologies, the amine-based chemical absorption is one of the most widely studied technologies for

CO₂ capture [8,9]. Amine scrubbing process has been commercially used for separation of CO₂ in the natural gas industry. However, it is still challenging to use aqueous amine for separating CO₂ from flue gas due to the high energy consumption of liquid solvent regeneration [10].

To avoid the drawbacks of amine scrubbing process, the development of solid supported amine-based sorbents has received much attention [9–14]. Compared to the aqueous amine sorbent, the solid supported amine sorbents not only have the potential to reduce the energy consumption by avoiding the need to heat bulk water for the regeneration of sorbent, but also have high CO₂ capacity and the ability to eliminate the corrosion problems of high concentrated aqueous amine sorbent [9,15]. Grafting and impregnation are the two most common methods for the preparation of amine modified solid sorbents.

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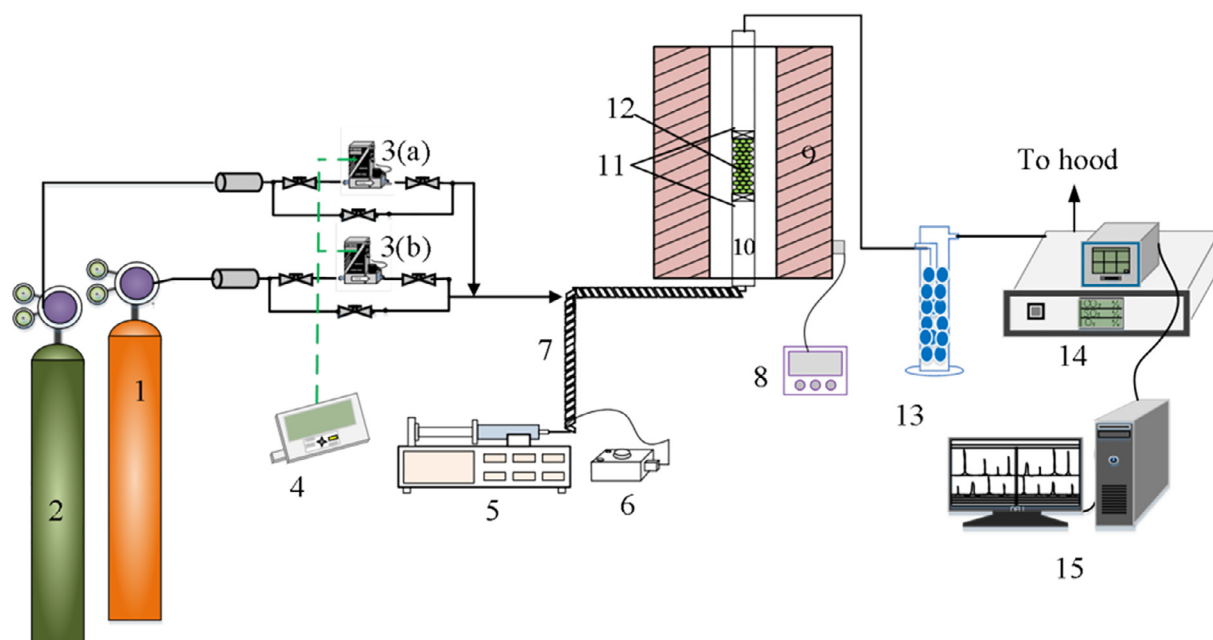


Fig. 1. Schematic diagram of CO₂ capture setup (1, 10 vol% CO₂ gas cylinder; 2, nitrogen cylinder; 3(a) and 3(b), mass flow controllers; 4, control module of mass flow controller; 5, syringe pump; 6, temperature controller of heating tape; 7, heating tape; 8 temperature controller of the tube furnace; 9, tube furnace; 10, quartz tube reactor; 11, quartz wool; 12, sorbent bed; 13, moisture removal unit; 14, gas analyzer; 15, data recording unit).

The sorbents prepared by chemically grafting amine to the support are normally more stable than those prepared by physical impregnation. However, in contrast to the grafting method using expensive chemicals, the impregnation method has several advantages, such as easier preparation, lower cost, and higher amine loading [16].

A variety of porous materials have been used as supports for different kinds of amine modified solid sorbents, such as mesoporous silica [13,17–22], porous carbon [23–28], aerogel [29,30], zeolite [31–33], mesoporous alumina [34,35], clay [36,37], porous titanium oxide, titanate [38–40], etc. Mesoporous materials with high surface area, high pore volume, and large pore size are most widely investigated among all porous materials. Some reports have shown that large pore size and pore volume are beneficial to achieving high CO₂ adsorption capacity. The high CO₂ adsorption capacity is important for making solid adsorbent more economic than a typical aqueous monoethanolamine (MEA) system [15]. However, even using mesoporous materials, most of the reported amine modified solid sorbents could not reach a CO₂ adsorption capacity of 4.0 mmol/g at 0.1 atm of CO₂ [9–11]. In addition, some porous materials are very expensive, which would limit their practical application. Therefore, seeking low-cost materials supported amine sorbent with a high CO₂ adsorption capacity is meaningful for industrial-scale practical application.

Wang et al. developed a solid sorbent with inexpensive clay and polyethylenimine (PEI) and achieved a capacity of 3.23 mmol/g at 0.15 atm of CO₂ [36]. Irani et al. reported an acid modified low-cost nanosepiolite supported tetraethylenepentamine (TEPA) for CO₂ capture [37]. The highest CO₂ sorption capacity reached by their optimal sorbent was 3.88 mmol/g at 0.01 atm of CO₂ [37]. Li et al. synthesized low-cost polyethylenimine–nano silica composites with a capacity of 4.23 mmol/g at 1.0 atm of CO₂ [41]. Martin et al. developed a commercial low cost silica supported diethylenetriamine adsorbent showing a capacity of 0.691 mmol/g at 1.0 atm of CO₂ [42]. However, there were still few reports on inexpensive supporting materials supported efficient adsorbent for CO₂ capture. Currently, most of the reported amine-based solid adsorbents prepared by the impregnation method use TEPA and PEI amines. Therefore, developing a new type of amine-based adsorbents for CO₂ capture that is inexpensive but has high CO₂ adsorption capacity is still imperative. To the best of our knowledge, the

aminoethylethanolamine-based solid sorbent has not been reported as an effective sorbent for CO₂ capture yet. In this work, a new sorbent was developed by using low-cost commercially available nanoporous silicic acid (SiO(OH)₂) powder and aminoethylethanolamine (AEEA) via a simple impregnation process without using any solvent. The impacts of major factors, such as AEEA loading, absorption temperature, moisture addition, and cyclic ability, on sorption capacity were investigated. The Avrami's fractional order kinetic model was also applied for the kinetic analysis of CO₂ adsorption and desorption on the sorbent. The energy consumption for adsorbent regeneration was also estimated.

2. Experiment

2.1. Preparation of sorbents

The SiO(OH)₂ powder and AEEA were purchased from Sigma Aldrich. Unlike the impregnation method applied for the preparation of adsorbents reported in literature [36,43], the silicic acid supported AEEA based sorbents in this work were prepared by physically mixing SiO(OH)₂ powder and AEEA together without any solvent. Certain amounts of AEEA and silicic acid were put in a glass beaker and then mixed well with a glass rod. AEEA/SiO(OH)₂ sorbents with AEEA loadings of 30, 40, 50, 55, 60, and 70 wt% were prepared with the same method in this work. These prepared sorbents were denoted as AEEA-x/SiO(OH)₂, where x represents the weight percentage of AEEA in the sorbent.

2.2. Sorbent characterization

The surface area and pore structure of the sorbent were measured by the N₂ physisorption at 77 K using a Quantachrome Quadrasorb SI. Powder X-ray diffraction (XRD) tests were conducted by a Rigaku Smart Lab diffraction system (Cu K α radiation, 40 kV and 40 mA). Thermogravimetric analysis (TGA) of samples was carried out by a TA Instruments SDT Q600 using a temperature rate of 10 °C/min. The morphologies of SiO(OH)₂ before and after loading of AEEA were characterized by Scanning electron microscopy (SEM) using a FEI

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