



## Adsorptive removal of carbon dioxide using polyethyleneimine loaded glass fiber in a fixed bed



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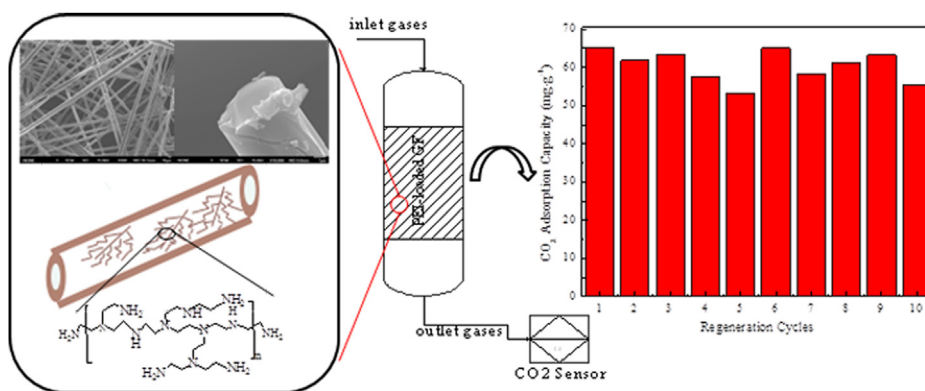
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### HIGHLIGHTS

- Polyethyleneimine (PEI)-loaded Glass Fiber was successfully prepared.
- PEI-loaded GF with core-shell structure performed well on CO<sub>2</sub> adsorption.
- The optimum condition for CO<sub>2</sub> adsorption in a fixed PEI-loaded GF bed was obtained.
- The regeneration temperature should be around 100 °C with RH lower than 30%.
- PEI-loaded GF showed good cyclic CO<sub>2</sub> adsorption-desorption performance.

### GRAPHICAL ABSTRACT



### ARTICLE INFO

#### Article history:

Received 6 February 2015

Received in revised form 29 April 2015

Accepted 3 May 2015

Available online 8 May 2015

#### Keywords:

CO<sub>2</sub> capture

Glass fiber

Polyethyleneimine

Dynamic adsorption

### ABSTRACT

Low-cost glass fiber (GF) was modified by loading polyethyleneimine (PEI) using a physical impregnation method and applied as an adsorbent for CO<sub>2</sub> capture. The structure and morphology of the PEI-loaded GF were characterized by thermogravimetric analyzer (TGA), fourier transform infrared spectroscopy (FTIR) and scanning electron microscope (SEM). The PEI-loaded GF possessed a core-shell structure. The CO<sub>2</sub> dynamic adsorption-desorption performances were investigated in a PEI-loaded GF fixed bed. The effects of PEI loading ratio, adsorption temperature, relative humidity (RH), flow rate and bed depth ( $H/D$ ) ratio were examined, and the maximal dynamic CO<sub>2</sub> adsorption capacity was 60.04 mg g<sup>-1</sup> with a PEI/GF ratio of 0.70 g g<sup>-1</sup>. In addition, the PEI-loaded GF was regenerated with only a slight decrease in the adsorption capacity after multiple regenerations using an optimum regeneration temperature of 100 °C. The results suggest that the PEI-loaded GF can be used as an economical adsorbent system for CO<sub>2</sub> capture.

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## 1. Introduction

Anthropogenic CO<sub>2</sub> emission from the burning of fossil fuels is causing general public concern because it is speculated to be the primary contributor to global warming and climate change [1–3]. According to NOAA, the current atmospheric concentration of CO<sub>2</sub> has reached 400 ppm compared to a level of 300 ppm in the pre-industrial period. The CO<sub>2</sub> emissions related to the energy sector will further increase by 130% by 2050 due to increasing fossil fuel usage [4,5]. Currently, chemical absorption [6–8], membrane [9,10], ionic liquids [11,12] and cryogenic techniques have been investigated for CO<sub>2</sub> capture. Among these technologies, aqueous amine chemical absorption systems including monoethanolamine and diethanolamine [13,14] have been commercially utilized for CO<sub>2</sub> capture from power plant effluents. However, this system suffers from several drawbacks including severe equipment corrosion and amine degradation as well as the high-energy consumption for the regeneration of aqueous solutions during stripping CO<sub>2</sub> from the spent amine solution [15]. Therefore, advanced CO<sub>2</sub> capture and storage technologies that are energy efficient and environmentally friendly are important.

As a promising technology, solid-form amine adsorbents have attracted much attention due to its less energy intensive regeneration process. The solid supports are normally porous materials with a high surface area and large pore volume (e.g., porous silica [16,17], activated carbon [18,19], zeolites [20,21], metal-organic frameworks (MOFs) [22,23] and some other materials, such as bentonite [24] and activated alumina [25]).

As the substrate for the amine, porous carbon has relatively weak retention, which results in poorer selectivity to CO<sub>2</sub> in the range of 50–120 °C. Due to its hydrophilic surface characteristics, zeolite supported amines are extremely sensitive to moisture, and their performance is substantially reduced in high humidity environments during CO<sub>2</sub> adsorption. MOFs exhibit exceptional CO<sub>2</sub> adsorption capacity with pure CO<sub>2</sub> at high pressures but its performance was dramatically reduced when exposed to gas mixtures. Silica-supported amine materials suffer from slow kinetics due to poor transport of CO<sub>2</sub> molecules from the surface into the loaded amine bulk due to the compact entity formed by the loaded amine [26].

A glass fiber, which has a small diameter and is an inexpensive and mechanically and thermally stable substrate, has been successfully applied as a substrate for the synthesis of novel carbon fibers [27–29] and other functional materials [30,31]. Polymeric-amine coatings have been widely studied due to their high selectivity towards CO<sub>2</sub> and high thermal stability [32,33]. Among all of the amines, polyethyleneimine (PEI) is a widely available and economical amine molecule [34–36]. To develop an economical and viable CO<sub>2</sub> adsorption system, a glass fiber that was 7 μm in diameter was introduced as a supporting substrate for polyethyleneimine. The performance of this material for CO<sub>2</sub> capture was investigated under various humidity, temperature, flow rate, bed depth and regeneration conditions.

## 2. Experimental

### 2.1. Materials

The glass fiber substrate, which is a nonwoven glass fiber mat with 7 wt% PVA binder (Craneglass 230), was provided by Crane & Co. Inc (Dalton, MA). Polyethyleneimine (PEI,  $W_n \sim 600$ ), epichlorohydrin (ECH) and methanol (99.8%) were purchased from Sigma–Aldrich and used as received. The CO<sub>2</sub> and N<sub>2</sub> gases were lab grade and obtained from S.J. Smith Welding, Inc.

### 2.2. Preparation of PEI-loaded GF

PEI and ECH were dissolved in methanol to yield a homogeneous solution. Glass mats were dip-coated with the mixture and dried in air at room temperature overnight followed by curing in a convection oven at 70 °C for a pre-set time.

### 2.3. Characterization

The morphology of the sorbents was observed using a Hitachi S-4700 scanning electron microscope (SEM). Prior to each characterization, the samples were dried overnight at approximately 70 °C under vacuum followed by conductive coating using a gold-palladium plasma spray for 20 s.

The coating content of PEI on the glass fiber was measured using a Hi-Res TA Instruments 2950 Thermogravimetric Analyzer (TGA) by burning off the coating in air at 800 °C. The surface chemistry functional groups were characterized by Fourier transform infrared spectroscopy (FTIR Nicolet Nexus 670) in diffuse reflectance mode (DRIFTS). The surface charge of the sorbents was determined through zeta potential measurements using a Zetasizer (Malvern Zetasizer Nano ZS).

### 2.4. CO<sub>2</sub> adsorption–desorption performance

CO<sub>2</sub> adsorption–desorption breakthrough cycles were conducted using the lab-scale setup shown in Fig. 1. The sorbent was loaded into the adsorption column with a fixed bed depth. Prior to each run, the column was purged with N<sub>2</sub> at a humidity level equivalent to the adsorption process. After 15 min of purging, the CO<sub>2</sub> and N<sub>2</sub> mixture gases were introduced until complete breakthrough was achieved. Then, pure N<sub>2</sub> was used to purge the adsorption column followed by online regeneration. Prior to each run, a blank adsorption experiment was carried without sorbents in the system under the same adsorption conditions. An electrochemical CO<sub>2</sub> sensor (CO<sub>2</sub> Meter, Inc., range of 0–30 vol% CO<sub>2</sub>, accuracy of ±0.2 vol% CO<sub>2</sub> ± 3 vol% of measured value) was employed to monitor the outlet CO<sub>2</sub> concentration.

Both the adsorption capacity and the desorption rate were used to evaluate the adsorbent. The CO<sub>2</sub> adsorption capacity was calculated using Eq. (1)

$$Q_{\text{CO}_2} = \frac{\int_0^t [(c_0 - c_t) \times V \times \rho_{\text{CO}_2}]}{m} \quad (1)$$

where  $Q_{\text{CO}_2}$  is the CO<sub>2</sub> adsorption capacity (mg g<sup>-1</sup>),  $c_0$  and  $c_t$  are the CO<sub>2</sub> concentration in the feed flow and at time  $t$ , respectively, (%),  $V$  is the flow rate, L min<sup>-1</sup>,  $\rho_{\text{CO}_2}$  is the CO<sub>2</sub> density (g L<sup>-1</sup>), and  $m$  is the weight of sorbent (g).

The desorption rate was calculated using Eq. (2)

$$k_{\text{Desorption}} = \frac{\int_0^t [(c_0 - c_t) \times V \times \rho_{\text{CO}_2}]}{Q_{\text{CO}_2} \times m} \quad (2)$$

where  $Q_{\text{CO}_2}$  is the CO<sub>2</sub> adsorption capacity calculated in the adsorption process (mg g<sup>-1</sup>) and  $m$  is the weight of the sorbent (g).

The amine utilization efficiency, which is defined as the moles of CO<sub>2</sub> molecules adsorbed for each mole atom in the amine functional group, was calculated using Eq. (3)

$$E_{\text{Amine}} = \frac{Q_{\text{CO}_2}/M_{\text{WCO}_2}}{r/(1+r)/M_{\text{WN}} \times (M_{\text{WN}}/M_{\text{WC}_2\text{H}_5\text{N}})} \times 100\% \quad (3)$$

where  $Q_{\text{CO}_2}$  is the CO<sub>2</sub> adsorption capacity calculated in the adsorption process (mg g<sup>-1</sup>) and  $r$  is the PEI coating ratio.

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