



Investigation of CO₂ adsorption performance and fluidization behavior of mesoporous silica supported polyethyleneimine



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ABSTRACT

A thorough and comprehensive study of different parameters affecting the sorbents prepared by impregnating polyethyleneimine (PEI) on silica for CO₂ capture has been conducted. The CO₂ capture performances of resulting sorbents were evaluated on both packed-bed and fluidized-bed reactors. Obtained results indicated that CO₂ adsorption-desorption performances of sorbents are greatly affected by the operation temperature in both simulated flue gas from coal fired power plant (SCF) and natural gas combined cycle power plant (NGCC). The optimal adsorption temperatures are from 50 to 80 °C for NGCC flue gas and from 70 to 90 °C for SCF flue gas, while the optimal regeneration temperatures are from 110 to 130 °C for both flue gas. Adsorbent containing 30 to 35 wt% PEI fluidized well with both simulated flue gas and steam. Results from this study revealed that PEI impregnated silica is a promising sorbents for CO₂ capture process, using a fluidized-bed reactor, from both coal-fired power and natural gas combined cycle power plants.

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1. Introduction

CO₂ capture technologies have undergone significant development in the last several years, and today large-scale Carbon Capture & Sequestration (CCS) demonstrations are underway. Several concepts for capturing CO₂ in industrial processes and power plants are being developed. Among these, amine scrubbing based on CO₂ absorption using ethanolamine (MEA) solutions [1,2] is considered as a current state-of-the-art industry process. However, the liquid phase amine scrubbing technology suffers from high regeneration cost, high degradation, and equipment corrosion [3–5]. As a result, there has been a strong interest to develop alternative more stable and less energy-intensive CO₂ removal technologies. Adsorption using solid sorbents [6–9], particularly amine-functionalized sorbents, is appealing due to their potentially lower energy requirements for sorbent regeneration [1,2,10–22]. By immobilizing amine functionalities on high-surface porous solid supports, the amino solid sorbents offer higher amine efficiency and thus superior sorbent performance [18,19,23,24]. Among various solid sorbent supports including zeolites [25], activated carbons [26–29], activated alumina [30–32], and membranes [33], polyethyleneimine (PEI) impregnation on mesoporous silicas have been attracting great attention [6–9,34–42], because of its simple preparation procedure which

can be easily translated to commercial scale as well as the potential to reduce the overall costs related to CO₂ capture by reducing the energy required to release the CO₂ during regeneration [10].

Previous studies indicated that the heat of CO₂ adsorption reduced considerably by immobilizing amine on solid substrate. For example, the heat of CO₂ adsorption of sorbent prepared by immobilizing PEI on CARIACT G10 silica is around 50.0 kJ/mol [43]. 3-Aminopropyltriethoxysilane functionalized mesoporous silica generates the heat of CO₂ adsorption about 60.0 kJ/mol [44]. Meanwhile, the heat of CO₂ adsorption of a conventional aqueous MEA solution is much higher; it is about 85 kJ/mol [10]. Generally, the heat of CO₂ adsorption is reduced due to the impregnation of amine solvents on a solid support. Because of low heat capacity, solid sorbent also requires less the sensible heat to raise temperature of adsorption bed to the regeneration temperature, however, the most significance of solid sorbent is the avoidance of solvent vaporization which could account for 30–50% of total regeneration energy [10,22,42].

Nevertheless, several factors may influence the amine-containing sorbents performance, in particular, adsorption capacity, kinetics, and the stability of the sorbents under different operating conditions such as adsorption and desorption temperature, pressure and number of cycles as well as their interactions with different species that may occur in industrial gases. Typical species include not only impurities such as water vapor, SO₂, H₂S, NO_x and O₂ [45–52], but also CO₂ itself, which deactivates amine-containing materials under dry conditions [34,51,53].

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The aim of the current study is to investigate the CO₂ adsorption performance and fluidization behavior of mesoporous silica-supported PEI under different operating conditions. Different types of silicas with various physical properties have been studied to evaluate their effect on the CO₂ capacity of the sorbent. In addition, different solvents and impregnating conditions were also thoroughly evaluated. The CO₂ adsorption performances of the resulting sorbents have been investigated in a packed-bed reactor (PBR) and their fluidization has been visually studied in fluidized-bed reactor (FBR). Finally, the influence of impurities on CO₂ adsorption performance during the long-term operation has been performed including humidity, oxygen and SO₂.

2. Experimental

2.1. Chemical reagents

Silicagels with different structural properties including 8 samples of irregular shape particles having different particles sizes and porosity, namely Silica from I to VIII and 1 spherical bead sample, namely Silica-IX were purchased from Sigma Aldrich. The irregular shape silica samples have surface area ranging from 155 to 353 m²/g, pore volume ranging from 1.1 to 1.2 cm³/g, and pore size ranging from 113 to 320 Å. All the silica substrates, used as support in this study, were dried at 130 °C for 2 h prior to impregnation with PEI. Branched polyethyleneimine (PEI, 99% and average Mw ≈ 600) was obtained from Alfa Easer (Massachusetts, USA). Methanol (MeOH, 99.9%), ethanol (EtOH, 99.9%), and isopropanol (iPrOH, 99.9%) were provided by Fisher Scientific (Massachusetts, USA). Deionized (DI) water was collected from a centralized water deionizing and filtering system at RTI International.

2.2. Sorbent preparation

Typical adsorbent preparation procedure involved a wet modification method [35,54]. First, a desired amount of PEI was dissolved in a solvent, which is MeOH, EtOH, iPrOH, or water, in a 1-L flask before adding a desired amount of silica under vigorous mixing in order to produce homogeneous slurry. The solvent was evaporated using a rotary evaporator (IKA RV 10 Rotovapor, USA). The resulting PEI-impregnated silica was dried in vacuum oven at 80 °C over night before used for testing.

2.3. Sorbent characterization

Characterization of the prepared adsorbents involved the methods of surface area measurement, pore volume measurement, pore size measurement, nitrogen content analysis, and Thermal Gravimetric Analysis (TGA). The surface area, pore volume, and pore sizes were measured by a nitrogen adsorption-desorption method using a Micromeritics Surface and Porosity Analyzer (ASAP 2020). First, sample was transferred into an analyzing tube with small bulb, whose mass was previously determined using analytical balance and degassed at 110 °C for 10 h. The mass of analyzing tube containing sample was determined again after degassing and the tube was mounted on Micromeritics Surface and Porosity Analyzer for analyzing. The mass of sample was determined by the mass difference between the tube with sample after degassing and empty tube. The pore volume and pore size were calculated by the Barrett-Joyner-Halenda (BJH) method using the desorption data. Nitrogen content within the sorbents was analyzed on a Thermo Scientific CNS elemental analyzer (Flash 2000). TGA was conducted on a thermogravimetric analyzer (SDT Q600), which involved heating sorbent samples from room temperature to 600 °C in nitrogen gas at a rate of 10 °C/min.

2.4. CO₂ adsorption-desorption on packed-bed reactor

The CO₂ adsorption-desorption performance of the adsorbent was investigated using packed-bed reactor system (PBR) designed and constructed by RTI International (RTI, North Carolina, USA) as shown in Fig. 1. The composition of the feed gas was adjusted by changing the flow rate of individual gases controlled by mass flow controllers. Water vapor was introduced to the gas stream by flowing the mixture of air and N₂ to the temperature controlled humidifier (A). The temperature of the humidifier can be adjusted to vary the moisture content in the simulated flue gas stream.

The CO₂ stream combined with the N₂/Air/H₂O mixed gas at the outlet of the humidifier to avoid the CO₂ from dissolving in the water in humidifier, which would result in the increase acidity of the water and inaccurate CO₂ content in the simulated flue gas stream. The effluent of the PBR (B) entered a condenser (C) and water collector (D) to remove water vapor prior to entering the CO₂ analyzer. The CO₂ concentration in the PBR effluent was analyzed using a Horiba CO₂ analyzer (VA-3000). PBR operated autonomously by National Instruments Lookout program coupled with customized PLC control developed by RTI's engineers. The software allows the PBR to be controlled according to preset test conditions (defined by operator) and to collect data points every second. The data including operating temperatures, the flow rate of input gases and CO₂ concentration in the output gas are recorded. The PBR is capable of performing multiple adsorptions-regeneration test cycles with varying test conditions without the need for operator attendance. In a typical experiment, approximately 2 g of adsorbent was mixed with silicon carbide beads, a filler and heat transfer material, then loaded into the PBR, which was made of stainless steel column with 1.27 cm inner diameter and 20 cm length.

A typical test undergoes 5 stages including 1) feed test, 2) purge, 3) adsorption, 4) regeneration, and 5) cooling. The feed test stage allows system to adjust the temperature of the process lines, humidifier, and reactor to defined values for the adsorption stage and to analyze and adjust the feed gas composition at the same flow rate being used in the adsorption stage. The feed gas can be tailored to various simulated flue gas conditions, in these tests in particular, flue gas from coal fired power plant (SCF) or natural gas combined cycle power plant (NGCC). The SCF flue gas has following composition: CO₂ = 15 vol%, O₂ = 4.5 vol%, and water vapor = 5.65 vol% in balance with N₂. The NGCC flue gas has following composition: CO₂ = 3 vol%, O₂ = 14 vol%, and water vapor = 5.65 vol% in balance with N₂. When the CO₂ concentration in the feed gas is stable, the system will enter the purge stage by flowing N₂ through the system until no CO₂ is detected in the outlet gas of the reactor. The adsorption stage is initiated by switching a valve to allow CO₂-laden feed gas to enter the reactor with a flow rate of 150 mL/min. The end of the adsorption process was determined by the breakthrough point for CO₂ which occurs when CO₂ concentration in outlet gas reached 99.95% of that in the feed gas. The end of the adsorption stage may also be determined by the minimum and maximum adsorption times, which was defined as 30 and 45 min, respectively, for the SCF flue gas and 60 and 90 min for NGCC flue gas. The regeneration stage follows the adsorption and is carried out by flowing humidified N₂ through the reactor at the flow rate of 150 mL/min at a desired temperature. The regeneration end point was defined by the regeneration off-gas (outlet gas) having concentration of CO₂ of <0.1%. Otherwise, it was determined by the minimum or maximum desorption times of 30 and 45 min, respectively. The cooling of the system is performed by flowing N₂ at the flow rate of 350 mL/min to reduce reactor temperature to either the temperature of the adsorption stage or to room temperature (if the tests are complete). The CO₂ loading (wt%) was calculated from regeneration data by dividing the total amount of CO₂ desorbed by the mass of the adsorbent and multiplying with 100. The CO₂ loading is defined as the amount of CO₂, which desorbs at a specific regeneration condition and does not include CO₂ lean loading. Each test consisted of

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