

CAPTURE OF CO₂ BY VACUUM SWING ADSORPTION PROCESS USING ACTIVATED CARBON BEADS

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Summary

Pitch-based AC beads (particle size about 1mm) were synthesized from coal tar pitch through the emulsion method without using binder material in our laboratory. Adsorption equilibrium and adsorption kinetics of CO₂ and N₂ on the activated carbon beads were studied experimentally and theoretically. Based on the equilibrium and kinetics data, an improved vacuum-pressure swing adsorption (VPSA) process were designed for CO₂ capture, and simulations of the VPSA process were performed in gPROMS. Various operating conditions and different process steps were evaluated in order to obtain the high product purity and high CO₂ recovery at the expense of the reduced energy consumption.

Keywords: CO₂ capture, VPSA process, modeling, activated carbon beads

Introduction

Carbon dioxide from flue gas emitted by power plants, steel mills, and cement kilns is responsible for a considerable amount (90%) of the total anthropogenic CO₂ emissions throughout the world, which causes global climate change.¹ One of the alternatives to mitigate carbon dioxide emissions is to capture CO₂ from its original source, specially fossil-fuel power plants. The two general approaches to reducing carbon emissions from existing plants are post-combustion capture and oxy-combustion, while pre-combustion is mainly used at new power plants that employ integrated gasification combined cycle (IGCC) technology. Commercial CO₂ capture technologies that exist today, such as sequestration by direct injection into geologic, or monoethanolamine (MEA) chemical absorption, are energy intensive and expensive. Adsorption, becomes a viable alternative because of the reusable nature of the adsorbents used.² Particularly, pressure swing adsorption (PSA) processes for capturing CO₂ from fossil fuel combustion have been studied.^{3,4}

There exist many potential candidate adsorbent materials available to CO₂ capture, which include carbonaceous materials, zeolite molecular sieves, metal organic framework materials, amine supported mesoporous materials, hydrotalcite-like compounds, limestone, lithium zirconate, lithium silicate, and other metal oxides materials.⁵ Carbonaceous materials are the promising candidate adsorbents, since they have high BET surface area, better CO₂ adsorption capacity, water tolerant, novel morphologies (monolith, bead, fiber, granular, respectively). Activated carbon (AC) bead is

spherical; no binder material is used in producing AC bead. The spherical nature and hardness of AC bead minimizes dust formation and attrition losses during adsorption and regeneration processes. AC bead also exhibits excellent fluidization properties both in gas and liquid applications. These characteristics make AC bead the material of choice for higher performance in carbonaceous materials application. The AC beads used here are pitch-based synthesized by our laboratory.⁶ The objective of this work is to characterize this new adsorbent for carbon dioxide capture from flue gas in a way commonly.

Experimental and modeling study

Pitch-based AC beads was prepared from coal tar pitch through the emulsion method, followed by stabilization, carbonization and activation, and a detailed preparation procedure was given in a previous publication.⁶ Mercury porosimetry was employed to determine the porous structure of the adsorbent. The porosimetry showed some quite large pores (>100 μm) and macropores with average diameter of 0.4042 μm. Also, some mesopores (<0.01 μm) were observed. For a detailed determination of the micropore structure we had measured N₂ adsorption at 77 K. The BET surface area is 845.87m²/g.

Adsorption equilibrium of CO₂ and N₂ was gravimetrically measured at (303, 333, 363, 393 and 423 K) with pressures ranging (0-100) kPa on AC beads. The full set of data was fitted with both Virial adsorption

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equation and multi-site Langmuir model. The equilibrium isotherms were presented in Figure 1. All isotherms were completely reversible. Preferential adsorption of CO₂ on this AC beads can be seen from the equilibrium isotherms. The selectivity toward carbon dioxide decreases when increasing the pressure. Henry constant and isosteric heat of adsorption were obtained from the equilibrium data and compared with those on other activated carbons reported previously in literature⁷.

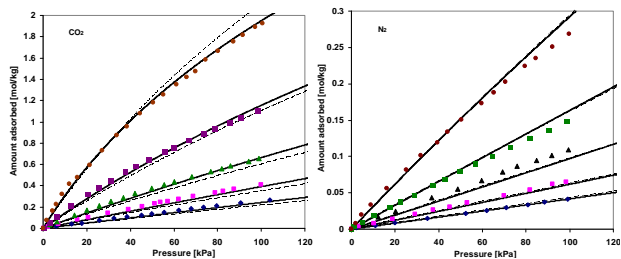


Figure 1. CO₂ and N₂ adsorption equilibrium on AC beads at the pressure range of (0-100) kPa. ●, T=303 K; ■, T=333 K; ▲, T=363 K; ◆, T=393 K; ◆, T= 423 K; Solid lines, Virial model; dotted lines, multisite Langmuir model.

The diffusion of single gases in the microporous structure of AC beads was studied by diluted breakthrough experiments performed over the same temperature range [(303 to 423) K]. A mathematical model was employed in the simulation of breakthrough curves. For both CO₂ and N₂ at all temperatures, micropore resistances control the diffusion. The micropore diffusivity constant (D_c / r_c^2) for CO₂ and N₂ obtained at 303 K is $1.058 \times 10^{-2} \text{ s}^{-1}$ and $7.185 \times 10^{-2} \text{ s}^{-1}$, respectively.

Based on the equilibrium and kinetics data, an improved vacuum-pressure swing adsorption (VPSA) process including feed, rinse with product, depressurization, counter-current blowdown and purge with product gas, were designed for CO₂ capture with the feed composition of (15-30)% CO₂ balanced with N₂ at (1.3-5) bar. Experiments of the cyclic adsorption/desorption were carried out. The diagram of the equipments set up for the VPSA process was shown in Figure 2. VPSA processes with and without product purge were studied. The effects of various operating conditions on the product gas CO₂ purity, CO₂ recovery and productivity were investigated in details.

Furthermore, a mathematical model taking into account mass balance, Ergun relation for pressure drop, energy balance, and Virial isotherm model was derived to describe the VPSA process with the product gas purge. Simulations of the mathematical model were performed in gPROMS (PSE Enterprise, UK) using the orthogonal collocation on finite elements (OCFEM). Different steps of the VPSA process and several parameters, such as the time of feed, feed temperature, feed flowrate, time of blowdown, the ultimate vacuum pressure were evaluated in order to obtain the high product purity and high CO₂

recovery at the expense of the reduced energy consumption.

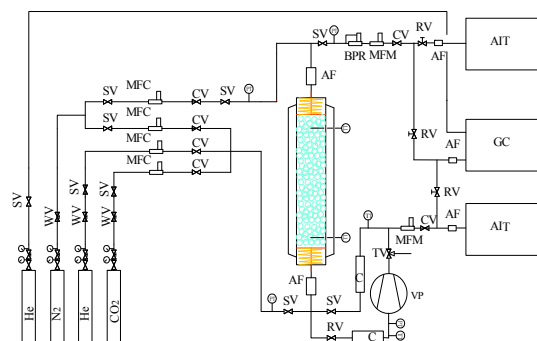


Figure 2. Schematic diagram of equipments set up for CO₂ capture by VPSA process. AF, air filter; BPR, back pressure regulator; C, condenser; CV, check valve; MFC, mass flowrate controller; PT, pressure transducer; RV, regulative valve; SV, ball valve; TV, three-way valve; TT, temperature transducer; VP, vacuum pump.

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