



## The removal and capture of CO<sub>2</sub> from biogas by vacuum pressure swing process using silica gel



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### ABSTRACT

In this study, silica gel was employed as adsorbent in a vacuum pressure swing adsorption process for the removal and capture of CO<sub>2</sub> from biogas. Adsorption isotherms of CH<sub>4</sub> and CO<sub>2</sub> on silica gel were measured experimentally, meanwhile a series of breakthrough experiments were also performed on a fixed bed packed with silica gel. In order to design experiments of VPSA process more reasonable, Central Composite Design Methodology was employed to implement the design of experiments, while Response Surface Methodology was used to analyze experimental results. Experimental results showed that the biogas simulated by 55% CH<sub>4</sub> balanced with CO<sub>2</sub> could be concentrated to an enriched CH<sub>4</sub> stream with CH<sub>4</sub> purity higher than 98%, after most of the CO<sub>2</sub> had been depleted from feed gas by VPSA process. Moreover, dynamic and transient behaviors, such as temperature profiles and concentration profiles in adsorption bed, were revealed by numerical modeling. A good consistency between experimental data and simulation results was observed. Furthermore, an industrial scale dual pressure swing adsorption unit was designed and evaluated by numerical simulation to achieve the goal of CH<sub>4</sub> enrichment and CO<sub>2</sub> capture simultaneously. Simulation results indicated that the simulated biogas could be separated to an enriched CH<sub>4</sub> stream at 98.01% CH<sub>4</sub> purity and 97.31% CH<sub>4</sub> recovery, as well as a concentrated CO<sub>2</sub> stream at 96.74% CO<sub>2</sub> purity and 97.58% CO<sub>2</sub> recovery.

### 1. Introduction

With the growing demand of energy and the exhausting of fossil fuel, the research and development of renewable energy have attracted more attention. Biogas generated by the biological degradation of organic compound, has been considered as a valuable energy carrier, and it is now playing a key role in emerging market of renewable energy [1,2]. In China, biogas engineering has been extensively promoted in urban and rural areas. According to the statistics, the total biogas production increased from  $3.6 \times 10^9$  cubic meter in 2001 to  $1.55 \times 10^{12}$  cubic meter in 2014 [3,4].

Raw biogas mainly consists of methane and carbon dioxide with traces of water, hydrogen sulfide, siloxanes, ammonia, oxygen, nitrogen and carbon monoxide [5]. It can be directly used for power generation and household cooking but the low fuel value of raw biogas caused by the presence of a large volume of CO<sub>2</sub> limits the economic feasibility of

its use. Therefore, upgrading biogas with the removal of CO<sub>2</sub> is an efficient way to improve the quality of biogas as well as to enhance the heating value of purified biogas [6]. Recently, vacuum pressure swing adsorption(VPSA) technology has drawn more attention for the removal of CO<sub>2</sub> from biogas because of its low capital investment and low energy consumption, as well as its stronger applicability in water-deficient area [7,8].

The primary task for VPSA process design is the choice of the adsorbent that should possess the quality of high capacity, high selectivity, as well as easy regeneration and low costs [9]. Currently, studies of adsorbents used in VPSA process for biogas upgrading and CO<sub>2</sub> depleting mainly focus on carbon molecular sieve(CMS), activated carbon, zeolite, and metal-organic framework(MOF) [8]. Yang's group firstly presented the study of VPSA process using carbon molecular sieve as adsorbent for CO<sub>2</sub>/CH<sub>4</sub> separation [10]. Experimental results demonstrated that an enriched methane product with a CH<sub>4</sub> purity

*Abbreviations:* ANOVA, analysis of variance; CCD, central composite design; CMS, carbon molecular sieve; CSS, cyclic steady state; LDF, linear driving force; MOF, metal-organic framework; RSM, response surface methodology; VPSA, vacuum pressure swing adsorption; PDEs, partial differential equations; UDS1, upwind differencing scheme 1

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| Nomenclature         |  |
|----------------------|--|
| $b_o$                | Langmuir isotherm parameter, ( $\text{bar}^{-1}$ )   |
| $T_{\text{amb}}$     | ambient temperature (K)  |
| $b_i$                | Langmuir isotherm parameter of component $i$ , ( $\text{bar}^{-1}$ )   |
| $T_g$                | gas phase temperature (K)  |
| $c_i$                | gas phase concentration of component $i$ ( $\text{mol} \cdot \text{m}^{-3}$ )                                      |
| $T_s$                | solid temperature (K)  |
| $C_{pa,i}$           | specific heat capacity of adsorbed phase of component $i$ ( $\text{J} \cdot \text{mol}^{-1} \cdot \text{K}^{-1}$ ) |
| $T_w$                | bed wall temperature (K)   |
| $C_{ps}$             | specific heat capacity of adsorbent ( $\text{J} \cdot \text{kg}^{-1} \cdot \text{K}^{-1}$ )                        |
| $v_g$                | gas phase superficial velocity ( $\text{m} \cdot \text{s}^{-1}$ )  |
| $C_{pw}$             | specific heat capacity of bed wall ( $\text{J} \cdot \text{kg}^{-1} \cdot \text{K}^{-1}$ )                         |
| $V$                  | standard volume flow rate ( $\text{m}^3 \cdot \text{h}^{-1}$ )   |
| $C_{vg}$             | specific gas phase heat capacity at constant volume ( $\text{J} \cdot \text{mol}^{-1} \cdot \text{K}^{-1}$ )       |
| $W_t$                | thickness of bed wall (m)  |
| $D$                  | mass diffusivity ( $\text{m}^2 \cdot \text{s}^{-1}$ )  |
| $y_i$                | Mole fraction of component $i$ (-)   |
| $D_{ax,i}$           | axial dispersion coefficient of component $i$ ( $\text{m}^2 \cdot \text{s}^{-1}$ )                                 |
| <b>Greek symbols</b> |  |
| $D_b$                | bed diameter (m)   |
| $\alpha_p$           | specific particle surface area per volume of bed ( $\text{m}^{-1}$ )   |
| $D_{e,i}$            | effective diffusion coefficient of component $i$ ( $\text{m}^2 \cdot \text{s}^{-1}$ )                              |
| $\rho_g$             | gas phase molar density ( $\text{mol} \cdot \text{m}^{-3}$ )   |
| $D_{k,i}$            | knudsen diffusion coefficient of component $i$ ( $\text{m}^2 \cdot \text{s}^{-1}$ )                                |
| $\rho_b$             | adsorbent density ( $\text{kg} \cdot \text{m}^{-3}$ )  |
| $D_{m,i}$            | molecular diffusion coefficient of component $i$ ( $\text{m}^2 \cdot \text{s}^{-1}$ )                              |
| $\rho_w$             | density of bed wall ( $\text{kg} \cdot \text{m}^{-3}$ )  |
| $F$                  | molar flow rate (SLPM)   |
| $\Delta H_i$         | isostatic heat of adsorption of component $i$ ( $\text{kJ} \cdot \text{mol}^{-1}$ )                                |
| $h_{\text{amb}}$     | wall-ambient heat transfer coefficient ( $\text{W} \cdot \text{m}^{-2} \cdot \text{K}^{-1}$ )                      |
| $\mu$                | bulk gas phase mixture viscosity ( $\text{kg} \cdot \text{m}^{-1} \cdot \text{s}^{-1}$ )                           |
| $h_f$                | gas-solid heat transfer coefficient ( $\text{W} \cdot \text{m}^{-2} \cdot \text{K}^{-1}$ )                         |
| $\psi$               | shape factor of adsorbent particle (dimensionless)   |
| $h_w$                | gas-wall heat transfer coefficient ( $\text{W} \cdot \text{m}^{-2} \cdot \text{K}^{-1}$ )                          |
| $\gamma$             | ratio of specific heats ( $C_p/C_v$ ) (dimensionless)  |
| $H_b$                | height of adsorbent layer (m)  |
| $\eta$               | compressor efficiency (dimensionless)  |
| $\varepsilon_b$      | bed porosity (dimensionless)   |
| $k_g$                | gas phase thermal conductivity ( $\text{W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$ )                              |
| $\varepsilon_p$      | particle porosity (dimensionless)  |
| $k_{LDF,i}$          | mass transfer coefficient for component $i$  |
| $\tau$               | pore tortuosity (dimensionless)  |
| $k_s$                | solid thermal conductivity ( $\text{W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$ )                                  |
| $k_w$                | thermal conductivity of bed wall ( $\text{W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$ )                            |
| $M_i$                | molar weight of component $i$ ( $\text{kg} \cdot \text{mol}^{-1}$ )  |
| $P$                  | pressure (kPa)   |
| $q_i$                | adsorbed phase concentration of component $i$ ( $\text{mol} \cdot \text{kg}^{-1}$ )                                |
| $q_i^*$              | adsorbed phase concentration in equilibrium with bulk component $i$ ( $\text{mol} \cdot \text{kg}^{-1}$ )          |
| $q_{m,i}$            | specific saturation adsorption capacity of component $i$ ( $\text{mol} \cdot \text{kg}^{-1}$ )                     |
| $r_p$                | adsorbent particle radius (m)  |
| $Re$                 | Reynolds number  |
| $R_g$                | ideal gas constant ( $\text{J} \cdot \text{mol}^{-1} \cdot \text{K}^{-1}$ )  |
| $Sc$                 | Schmidt number   |
| $t$                  | time (s)   |

higher than 90% could be achievable from a 50%/50% CO<sub>2</sub>/CH<sub>4</sub> mixture. Meanwhile the methane recovery was able to reach 90%. Subsequently, Rodrigues's group performed a systematic investigation related to the removal of CO<sub>2</sub> from biogas by VPSA process using CMS and Zeolite 13X, respectively [11–13]. Comparisons of experimental results showed that VPSA process with CMS adsorbent for biogas purification could achieve a better product recovery and productivity at a specific CH<sub>4</sub> purity, compared with that using zeolite 13X [12]. Besides, a new VPSA configuration with a lead-trim concept was proposed by this group and was evaluated through numerical simulation for further improving process performances [13]. A feed mixture containing 67% CH<sub>4</sub> and 33% CO<sub>2</sub> could be concentrated to a CH<sub>4</sub> product gas with 98% CH<sub>4</sub> purity and 88% CH<sub>4</sub> recovery by using this new VPSA configuration. Khunpolgrang et al. employed a new purge strategy to enhance CH<sub>4</sub> recovery in VPSA configuration for CH<sub>4</sub>/CO<sub>2</sub> separation [14]. High purity N<sub>2</sub> produced by another PSA unit was used as purge gas instead of a portion of high purity CH<sub>4</sub> product for adsorbent regeneration at the purge step. Results represented that the new VPSA configuration with zeolite 13X could produce an enriched methane stream with 99% CH<sub>4</sub> purity and 93% CH<sub>4</sub> recovery from biogas but it came at the cost of extra capital investment and energy consumption. Pevida et al. evaluated biogas upgrading by using biomass-based carbon that was prepared from cherry stones [15–17]. Results indicated that biomass-based activated carbons possessed higher adsorption capacity, faster kinetics, as well as good cyclability and renderability, compared with a commercial activated carbon Calgon BPL. Ferreira et al. demonstrated that a water stable porous aluminum terephthalate MIL-53(Al) was the promising adsorbents for CO<sub>2</sub>/CH<sub>4</sub> separation [18]. Their simulation results demonstrated that a four bed VPSA process with MIL-53(Al) as adsorbent could produce 99.4% CH<sub>4</sub> product with a recovery of 92.8% and 95.4% CO<sub>2</sub> product at a recovery of 99.6% from a biogas mixture consisting of 60%CH<sub>4</sub> and 40%CO<sub>2</sub>. Wu et al. compared the energy

consumption of VPSA process employed metal-organic framework 508b (MOF-508b), carbon molecular sieve 3K(CMS-3K) and zeolite 13X as adsorbents to reject CO<sub>2</sub> from biogas [19]. Simulation results revealed that energy consumption of VPSA process with MOF-508b and CMS-3K was 56% and 50% lower than that with zeolite 13X. Lately, Augelletti et al. investigated a two-stage VPSA process for biogas upgrading using zeolite 5A as adsorbent by numerical simulation [20]. The first stage unit was used to produce biomethane with a purity of 97% while the second stage unit was applied to produce an almost pure CO<sub>2</sub> stream by using residues gas of the first unit as feed gas. More importantly, the CO<sub>2</sub> depleted gas stream from the second unit was recycled to the first unit to enhance the overall methane recovery.

According to the above-mentioned studies, adsorbent materials can be used to separate CO<sub>2</sub> from biogas in practice mostly focused on zeolite and CMS. Recently, Ferella et al. investigated the separation of CO<sub>2</sub> from dry CO<sub>2</sub>/CH<sub>4</sub> mixtures by synthesized zeolites and available commercial adsorbents at a lower adsorption pressure [21]. Experimental results initially indicated that silica gel was the best adsorbent within commercial adsorbents for CO<sub>2</sub>/CH<sub>4</sub> separation but the detailed investigation of silica gel in VPSA process for methane enrichment was lacked. It is well known that silica gel considered as an inexpensive and robust adsorbent has been widely used in adsorption refrigeration and adsorption desalination because of its suitability for low grade heat source [22–24]. Besides, silica gel has also been extensively reported for CO<sub>2</sub> capture due to lower energy requirement for its regeneration as well as high selectivity of CO<sub>2</sub> with respect to weakly adsorbed component [25–27]. It can be assumed that it might possess the potential ability for the removal of CO<sub>2</sub> from biogas but there are few of papers related to biogas upgrading using silica gel as adsorbent. Hence, in this study, a four bed VPSA process was constructed for biogas purification with silica gel adsorbent. The Central Composite Design Methodology was applied to design experiments of VPSA process, while the Surface

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