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Optimization of three-bed VPSA system for biogas upgrading

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HIGHLIGHTS

- A three-bed 12-step VPSA process applied to upgrade biogas.
- The r-SQP algorithm used to optimize the decision parameters of this process.
- Improving the CH_4 recovery and the unit productivity of CH_4 while reducing the unit energy consumption.
- An important improvement in the performance of the three-bed 12-step VPSA process.

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ABSTRACT

An optimization for biogas upgrading process by three-bed 12-step vacuum pressure swing adsorption (VPSA) was performed in this work. The raw gas composed of 60% CH_4 and 40% CO_2 was used at 313.15 K with the pressure of 101.325 kPa. Co-current replacement with heavy component step and counter-current pressurization with replaced exhaust step were applied in the VPSA system. The results indicated that the adoption of these two steps actually helped enhance the CH_4 recovery and the specific productivity of CH_4. The objective pursued in this work was evaluated by the combination of the specific energy consumption, the CH_4 recovery and the specific productivity of CH_4. The effects of these decision variables on the objective were evaluated by the r-SQP algorithm. According to the optimization performed for a stream of biogas of 37 N m^3/h, it was possible to obtain bio-methane with a purity of 98.00%, a recovery of 93.23% with a specific productivity of 0.1331 N m^3/kg/h and a specific energy consumption of 0.3697 kWh/kg of produced methane.

1. Introduction

Sejian et al. pointed that the greenhouse warming potential of methane is 25 times higher than that of conventional well-known greenhouse gas CO_2 (Sejian et al., 2011). Consequently, controlling the emission of CH_4 has significant contribution to alleviate global warming. Meanwhile, the recovery of CH_4, a kind of clean and renewable fuel or energy (Chynoweth et al., 2001), is helpful to the remission in current tense situation of energy demand, generating significant environmental and economic benefits (Börjesson and Berglund, 2007; Karapidakis et al., 2010).

Biogas produced in anaerobic digestion units of livestock manure is one of the sources of CH_4 emission in many developing countries, whose CH_4 content ranges from 40% to 70%, the rest being CO_2 and trace of NH_3, H_2S and H_2 (Tauseef et al., 2013). As the major contaminant, carbon dioxide's content in biogas is usually higher than 25%, hence its removal is the most critical step for the entire biogas upgrading process. After the removal of minor contaminants, CH_4 and CO_2 can be then separated by physical or chemical absorption (Rasi et al., 2008; Tippayawong and Thanompongchart, 2010), membrane-based separation (Harasimowicz et al., 2007; Makaruk et al., 2010), cryogenic separation (Tuinier and Annaland, 2012), pressure swing adsorption (PSA) or vacuum pressure swing adsorption (VPSA) (Knaebel and Reinhold, 2003; Cavenati et al., 2005; Grande and Rodrigues, 2007a, 2007b; Santos et al., 2011). Within the above feasible methods, VPSA is an attractive option for biogas upgrading, due to its advantage (Sircar, 2002) in low cost as well as highly automatic operations.

Different adsorption affinity of gas components to adsorbent under various pressures is the principle of gas separation by PSA or VPSA (Ruthven et al., 1994; Waldron and Sircar, 2000). For biogas upgrading process by VPSA, adsorption process is performed beyond atmospheric pressure, and large amount of CO_2 is enriched in bed. Desorption process is executed below atmospheric pressure, and adsorbent achieves regeneration. It is crucial for VPSA to choose an appropriate adsorbent. Cavenati et al. studied the performance of

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carbon molecular sieve 3K in biogas upgrading by Skarstrom cycle (Cavenati et al., 2005; Skarstrom, 1960). Grande et al. researched and compared the performance of zeolite 13X and carbon molecular sieve 3K in biogas upgrading process, and they reported that carbon molecular sieve 3K may result in higher specific productivity of CH_4 than zeolite 13X, as a results of its higher CH_4 recovery (Grande and Rodrigues, 2007a). In their subsequent publications (Grande and Rodrigues, 2007b; Santos et al., 2011), the performance of zeolite 13X was significantly improved by enhancing the temperature of raw gas and by adding an equalization pressurization step. Delgado et al. reported that silicalite had good performance on the separation of CH_4 and CO_2 (Delgado et al., 2006). In despite of lower CH_4 recovery than the value reported by the reference mentioned above, the specific productivity of CH_4 was relatively higher. It indicates the prospect application of silicalite for upgrading biogas process.

In the past 10 years, the optimization of PSA has been widely studied, and some significant achievements have been made. Smith et al. presented a mixed-integer nonlinear program (MINLP) model to achieve the minimum annual cost by optimizing certain design conditions, such as the number of beds (Smith and Westerberg, 1991). Ko et al. used an approach based on r-SQP to optimize the CO_2 capture by PSA and fractionated VPSA (Ko et al., 2003, 2005). Jiang et al. applied direct sensitivity, a simultaneous tailored approach and r-SQP optimization strategy to optimize a PSA process for air separation (Jiang et al., 2003). In order to improve the computational efficiency, Jiang et al. adopted parallelize sensitivity calculation, and achieve a close-to-linear speed up rate (Jiang et al., 2005). Cruz et al. adopted an adaptive multi-resolution method and a successive quadratic-programming algorithm to simulate and to optimize the general cyclic adsorption separation processes (Cruz et al., 2005). Agarwal et al. proposed a novel PSA superstructure, and applied it to design optimal PSA cycle configurations for pre-combustion and post-combustion CO_2 capture (Agarwal et al., 2010a, 2010b). Khajuria et al. addressed an optimization strategy for simultaneously incorporating PSA design, and operational, and control aspects under process uncertainty (Khajuria and Pistikopoulos, 2013).

In this article, it was assumed that methane's content in biogas was 60%, and the balance component was carbon dioxide. A three-bed 12-step VPSA system was adopted to separate CH_4 from biogas based on the fundamental research work in our laboratory (Zhang Zhengwang et al., 2014). The adsorbent used in the work is silicalite (Li and Tezel, 2007). In order to enhance the CH_4 recovery and the specific productivity of CH_4, a co-current replacement with heavy component step was adopted in this system, and the replaced exhaust was used as final rising gas for another bed. The designed three-bed 12-step VPSA process is a typical process which can incorporate all the existing cycle steps such as equalization, replacement etc. when it is compared with traditional two-bed PSA process. And it is suitable for laboratory-scale research purposes compared to multi-beds PSA process. The separated performance of this process can be estimated in terms of the CH_4 recovery, the specific productivity of CH_4, and the specific energy consumption. The three together made up the objective function of this work, the effects of these decision variables on the objective were studied by the r-SQP algorithm. Furthermore, an optimization was performed to intensify the process, in the prerequisite of meeting some necessary constraints.

2. VPSA system model

VPSA model mainly includes adsorption bed, valve (uni-direction or bi-direction), vacuum pump, buffer tank and compressor, and their detailed mathematical models had been well established. As the core part of the entire VPSA model, mathematical model of the adsorption bed was described as follows, and some assumptions were made for it (Jiang et al., 2003):

1. All the gases obey the ideal gas law;
2. There are no gradients of gas concentration, temperature and pressure in the radial direction;
3. The gas phase and the solid phase are in thermal equilibrium, and the bulk density of the solid phase keeps constant;

Table 1
Mathematical model for adsorption bed (da Silva et al., 1999; da Silva and Rodrigues, 2001a, 2001b).

Parameter	Expression
The component *i* mass balance	$-\varepsilon_b D_{ax}\frac{\partial^2 y_i}{\partial z^2}+\frac{\partial(v_g c_i)}{\partial z}+(\varepsilon_b+(1-\varepsilon_b)\varepsilon_p)\frac{\partial c_i}{\partial t}+\rho_p(1-\varepsilon_b)\frac{\partial q_i}{\partial t}=0$
The energy balance	$((\varepsilon_b+(1-\varepsilon_b)\varepsilon_p)\sum_{i=1}^{N}c_i(C_{pg,i}M_i-R)+(1-\varepsilon_b)\rho_s C_{ps}+(1-\varepsilon_b)\rho_s\sum_{i=1}^{N}q_i(C_{pg,i}M_i-R))\frac{\partial T}{\partial t}$ $+v_g\rho_g\sum_{i=1}^{N}C_{pg,i}M_i\frac{\partial T}{\partial z}+\rho_s(1-\varepsilon_b)\sum_{i=1}^{N}\frac{\partial q_i}{\partial t}\Delta H_i+2h\frac{T-T_w}{R_b}-(\varepsilon_b+\varepsilon_p(1-\varepsilon_b))\frac{\partial P}{\partial t}-k_g\frac{\partial^2 T}{\partial z^2}=0$
The momentum balance	$-\frac{\partial P}{\partial z}=\frac{150\mu(1-\varepsilon_b)^2}{(2R_p)^2{\varepsilon_b}^3}v_g+1.75\frac{(1-\varepsilon_b)\rho_g}{2R_p{\varepsilon_b}^3}\lvert v_g\rvert v_g$
Extended Langmuir equation	$q_i^*=\frac{q_{m,i}b_iP_i}{1+\sum_{i=1}^{n}b_iP_i},\quad b_i=b_0\exp\left\{-\frac{\Delta H_i}{R}\left(\frac{1}{T}-\frac{1}{313.15}\right)\right\}$
LDF model	$\frac{\partial q_i}{\partial t}=15\frac{D_{c,i}}{{R_p}^2}(q_i^*-q_i)$
Dispersion coefficient	$D_{ax}=0.73D_m+\frac{v_gR_p}{\varepsilon_b\left(1+9.49\frac{\varepsilon_bD_m}{2v_gR_p}\right)},\ D_{c,i}=\frac{\varepsilon_pD_{k,i}D_m}{\tau(D_{k,i}+D_m)}$ $D_{k,i}=48.5D_p\sqrt{\left(\frac{T}{M_i}\right)},\ D_m=\frac{0.01013T^{1.75}\sqrt{\left(\frac{1}{M_A}+\frac{1}{M_B}\right)}}{P\left({D_{v,A}}^{1/3}+{D_{v,B}}^{1/3}\right)^2}$
Boundary conditions	$z=0:\quad v_{g,z}>0,\ T_z=inlet.T,\ y_{i,z}=inlet\cdot y_i$ $v_{g,z}\le 0,\ \frac{\partial T_z}{\partial z}=0,\ \frac{\partial y_{i,z}}{\partial z}=0$ $z=H_b: v_{g,z}>0,\ \frac{\partial T_z}{\partial z}=0,\ \frac{\partial y_{i,z}}{\partial z}=0$ $v_{g,z}=<0,\ T_z=outlet\cdot T,\ y_{i,z}=outlet\cdot y_i$