



Moving bed adsorption process with internal heat integration for carbon dioxide capture



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ABSTRACT

A moving bed adsorption (MBA) process with heat integration is proposed as a potentially viable post-combustion process for the capture of CO₂ from large-scale CO₂-emitting plants. The proposed process consists of an adsorption bed and two desorption beds through which the adsorbent particles continuously circulate. The adsorbent particles and the flue gas flow in opposite directions in the adsorption bed, which is operated at low temperature and atmospheric pressure. The adsorbents discharged from the adsorption bed are transferred to a desorption bed operated at a high temperature and 1 atm, then transferred to another desorption bed operated at a high temperature and in a vacuum, and returned to the adsorption bed. A heat integration scheme in which the thermal energy removed from the adsorption bed is reused in the desorption beds was designed. After a numerical model of the proposed process was developed, the effects of the model parameters and operating conditions on the performance and behavior of the process were investigated. The proposed MBA process with the heat integration scheme was estimated to produce CO₂ with a purity of over 97% and a recovery of over 80% while the recovered energy from the adsorber can provide up to 54.4% of the regeneration energy.

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1. Introduction

The resolution of global warming is considered a compelling assignment to the human community to prevent the worsening of climate change. The Kyoto protocol was first adopted in 1997 and entered into force in 2005. As of 2011, more than 190 nations have signed and ratified the protocol, and have initiated specific actions to reduce greenhouse gas emissions. Among the four major greenhouse gases, which include CO₂, CH₄, NO_x, and SF₆, the primary concern has been CO₂ because this gas exhibits the largest amount of emission. Carbon capture and sequestration (CCS) has been considered the most promising technology that can immediately address the CO₂ problem. Presently, a major portion of the research on CCS is focused on CO₂ capture rather than on its transportation and storage because 70–80% of the total energy required for CCS is estimated to be used in the capture process (Haszeldine, 2009). The large energy requirement of the current capture process is the main obstacle to the early deployment of CCS. Thus, many research studies have focused on the development of low-energy-demand capture processes.

Post-combustion carbon capture processes are being researched using different approaches. The chemical absorption process using aqueous solvents, such as monoethanolamine (MEA), has been used for a long time in industries, and it is generally considered more appropriate to large CO₂ emission sources, such as power plants. However, this technology is still far from immediate deployment because of the large energy requirement in the CO₂ regeneration (Ramezan et al., 2007). The use of the current absorption-based CCS process in a power plant is expected to result in an energy penalty of 20–30%.

Processes using solid sorbents are also being actively researched in hopes of inventing less energy-intensive alternatives to the liquid absorption process (Wang et al., 2010; Shimizu et al., 1999; Yi et al., 2007; Drage et al., 2012; Veneman et al., 2012). The sorbent materials that are being investigated include alkali and alkali earth metals, such as K₂CO₃, Na₂CO₃, and CaO (Gupta and Fan, 2002; Abanades et al., 2004, 2005; Liang et al., 2004; Lee et al., 2008, 2009), amine-enriched porous sorbents (Gray et al., 2005; Monazam et al., 2013; Serna-Guerrero et al., 2010; Serna-Guerrero and Sayari, 2010), and metal organic frameworks (Millward and Yaghi, 2005; Sumida et al., 2012). Presently, circulating fluidized bed reactors are dominantly adopted for the absorber, and bubbling fluidized bed or moving beds are employed for the regenerator (Yi et al., 2007; Veneman et al., 2012; Abanades et al., 2004).

The amount of thermal energy liberated in the adsorber (or absorber) is roughly the same as that consumed in the regenerator. If the thermal energy liberated in the adsorber can be transferred

Abbreviations: ADB, adsorption bed; A-DEB, atmospheric desorption bed; V-DEB, vacuum desorption bed; CW, cooling water; DHI, degree of heat integration; HTW, heat transfer water.

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A_h	cross-sectional area of heating medium channel (m^2)
C	concentration in gas phase ($kmol/m^3$)
C_i	concentration of component i in gas phase ($kmol/m^3$)
D_{ax}	mass axial dispersion coefficient (m^2/s)
D_m	molecular diffusivity of gas (m^2/s)
ΔH_i	average isosteric heat of adsorption for component i ($kJ/kmol$)
L	bed height of packed region (m)
N_m	the number of heat exchanger plates
P	pressure (bar)
Pr	Prandtl number, $c\mu/k$
R	gas constant ($bar\ m^3/(kmol\ K)$)
Re	Reynolds number, $\rho\epsilon u_d/\mu$
T_h	heating medium temperature (K)
T_g	gas temperature (K)
T_s	adsorbent temperature (K)
T_s^{avg}	average adsorbent temperature in radial direction between plates (K)
T_w	wall temperature (K)
T_D	reference temperature (K)
U_{hs}	overall heat transfer coefficient between heating medium and solid phases ($J/(m^2\ s\ K)$)
W	width of heat exchanger plates (m)
a_{gs}	heat transfer area between the gas and solid phases per unit volume ($1/m$)
b	half distance between plates (m)
b_i	Langmuir constant of component i ($1/bar$)
c_g	specific heat of gas ($J/(kg\ K)$)
c_h	specific heat of heating medium ($J/(kg\ K)$)
c_s	specific heat of adsorbent ($J/(kg\ K)$)
d_p	particle diameter (m)
h_{gs}	heat transfer coefficient between gas and solid phases ($J/(m^2\ s\ K)$)
h_h	heat transfer coefficient of heating medium side ($J/(m^2\ s\ K)$)
h_s	heat transfer coefficient of solid side ($J/(m^2\ s\ K)$)
k_e	effective thermal conductivity ($J/(m\ s\ K)$)
k_g	thermal conductivity of gas ($J/(m\ s\ K)$)
k_s	thermal conductivity of solid ($J/(m\ s\ K)$)
k_i^L	coefficient of mass transfer model for component i ($1/s$)
k_i^Q	coefficient of mass transfer model for component i ($1/s$)
\dot{m}_{cw}	cooling water flow rate (kg/h)
\dot{m}_s	solid circulating flow rate (kg/h)
q_i	amount adsorbed of component i ($kmol/kg$ -ads.)
q_i^*	equilibrium adsorbed phase concentration ($kmol/kg$ -ads.)
$q_{m,i}$	maximum amount adsorbed of component i ($kmol/kg$ -ads.)
r_i	mass transfer rate of component i from bulk gas to adsorbed phase ($kmol/(kg$ -ads. s)
$r_{in,i}$	a positive mass transfer rate from the solid to the gas phase ($kmol/(kg$ -ads. s)
$r_{out,i}$	a positive mass transfer rate to the reverse direction ($kmol/(kg$ -ads. s)
u_{cw}	interstitial linear velocity of cooling water (m/s)
u_g	interstitial linear velocity of gas (m/s)
u_h	interstitial linear velocity of heating medium (m/s)
u_s	interstitial linear velocity of solid (m/s)

y_i	mole fraction of component i
y_i^{feed}	mole fraction of component i in feed stream

Greek letters

λ_{ax}	effective axial thermal conductivity of gas ($J/(m\ s\ K)$)
μ	gas viscosity (Pa s)
κ	CW purge ratio
ρ_g	gas density (kg/m^3)
ρ_h	density of heating medium (kg/m^3)
ρ_s	bulk adsorbent density (kg/m^3)
ϵ	bed porosity

to the regenerator, the thermal balance is satisfied within the process and the external energy supply can be minimized. The obstacle to the realization of this concept is the reversed temperature difference, which means that the adsorber is at a lower temperature than the regenerator. This difficulty can be overcome by upgrading the energy recovered at a low temperature to high-temperature energy through additional heating prior to being then transferred to the high-temperature desorber. To minimize the dependence on an external energy source, the recovered energy needs to be obtained at the highest temperature possible. However, the temperature inside a fluidized bed adsorber is rather uniform. In contrast, a temperature profile is formed inside the moving bed adsorber (MBA), and the design for the recovered energy discharged at a high temperature is possible. It seems that the MBA has not yet been considered in the study of the solid sorbent-based carbon capture process due to its potentially larger bed size compared with fluidized beds (Yang and Hoffman, 2008). However, the MBA process might offer a new opportunity to reduce the energy cost on a large scale in the CO₂ capture through intelligent heat integration, that is not easy to realize with circulating or bubbling fluidized bed processes.

The purpose of this paper is to propose a novel heat-integrated moving bed adsorber (MBA) process for CO₂ capture. The solid sorbent considered in this research is zeolite 13X, which is a representative adsorbent that has been employed for CO₂ separation using pressure swing adsorption (PSA). Other recently studied solid sorbents such as MOF (Metal Organic Framework) or alkali earth metals were not considered due to the lack of reliable information on mass transfer and reaction rates. As a continuous process, the proposed MBA process has several advantages over the conventional PSA process, which include the convenience to realize the temperature swing operation and heat integration. Compared with the fluidized bed processes that are employed with solid adsorbents, the realizability of efficient heat integration is also the most important advantage.

The proposed MBA process is composed of three beds: an atmospheric adsorption bed, an atmospheric desorption bed, and a vacuum desorption bed. Each bed was mathematically modeled based on first principles, and the individual bed models were integrated to construct a process simulator. Two different heat integration schemes were considered and incorporated into the simulator. The effects of various operating conditions on the performance of the proposed process, especially the purity and recovery of CO₂ and the degree of heat integration, were investigated.

2. Moving bed adsorption process for CO₂ capture

The basic configuration of the MBA process with heat integration is presented in Fig. 1. The process MBA consists of a low-temperature adsorption bed (ADB) operated at atmospheric pressure, and two

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