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CO_2 capture from natural gas power plants by aqueous PZ/DETA in rotating packed bed

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ABSTRACT

An rotating packed bed (RPB) was applied to capture CO_2 from the flue gases of natural gas power plants containing 4 vol% of CO_2 using an aqueous solution with mixed absorbents piperazine (PZ) and diethylenetriamine (DETA) in this study. The experimental results showed that PZ/DETA with 4.0 m/4.0 m, as the most promising formulation, not only exhibited higher CO_2 capture efficiency and capacity but also showed lower regenerated energy in an RPB with 54.8% lower than the most common used absorbent of 7.0 m monoethanolamine (MEA). The effects of absorbent concentration, lean loading and regeneration in an RPB were all investigated. Based on the Aspen Plus simulation to 7.0 m MEA with various loadings, a packed bed (PB) required 4.6 times the volume of an RPB to achieve the same capture efficiency at the same operating conditions; the CO_2 capture efficiency and amount using an RPB were 63% and 4.6 times higher, respectively, than those for the same volume PB, indicating the superiority of an RPB to treat low CO_2 concentration flue gases.

1. Introduction

Since the Industrial Revolution, the greenhouse effect has significantly affected the environment of the whole world. The major portion of the greenhouse gases is CO_2 which is responsible for more than 60% of the global warming [1] and is now at a concentration higher than 400 ppm, significantly higher than the pre-industrial level of approximately 280 ppm. To satisfy the long-term goal of limiting global warming as to maintain the temperature well below 2 °C, the International Energy Agency (IEA) indicated that 57 billion tons of CO_2 emissions should be reduced to 14 billion tons by 2050 and that CO_2 capture and storage (CCS) technology contributes to approximately 14% of the reduction [1]. Therefore, development of CCS technology is essential.

Though various technologies have been proposed to capture CO_2 [2,3], chemical absorption is likely to be the most applicable technology to achieve the CO_2 capture purpose from the gases of fossil fuel power plants before 2030 [4]. However, major concerns for this technology including the requirement of a large equipment volume, solvent degradation, and high energy consumption are needed to be solved [5–7]. Lin et al. [8] has indicated that to employ an rotating packed bed (RPB) instead of a packed bed can overcome the above mentioned concerns. With high gravity in RPBs, liquid is cut into numerous small droplets and thin films when it passes through the packing, and the gas-liquid contact area increases substantially so that the mass transfer rate can significantly be increased.

Because of relatively short retention time in an RPB, reaction rate between absorbent and CO_2 is therefore the key factor. Freeman et al. [9] observed an aqueous solution containing piperazine (PZ) as a promising absorbent because of its high reaction rate, low corrosion to an apparatus and low volatility. However, the limitation of PZ solubility in water makes its inconvenient when a high PZ concentration solution is loaded under cold environment. Hartono et al. [10] presented the results on the use of diethylenetriamine (DETA) as the absorbent. The reaction rate of DETA is not as fast as that of PZ, but it is faster than that of the traditional alkanolamine absorbents, such as MEA and aminoethylethanolamine (AEEA). Moreover, DETA is in liquid form at room temperature, thus there will be no precipitation problem during the CO_2 capture process. Yu et al. [6] proposed a mixed alkanolamine absorbent as 2.5 m PZ/2.1 m DETA to capture CO_2 from the flue gases containing CO_2 concentration higher than 10 vol% that possessed the advantages of the two types of alkanolamines. PZ/DETA has the characteristics of not only a high reaction rate but also a lower precipitation during operation. The two alkanolamine mixed absorbent is therefore another practical formulation.

In recent decades, shale gas has become one of the most important energy resources in the US. According to the expectation of US

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Nomenclature

A	CO_2
a	interfacial area in packing, m^2/m^3
a_t	total surface area of packing, m^2/m^3
B	monoethanolamine (MEA)
C_A	CO_2 concentration in gas phase, vol%
C_{AL}	CO_2 concentration in liquid phase, vol%
C.A.	capture amount, L/min
C.E.	capture efficiency, %
D_{AB}	binary diffusivity for system A-B, m^2/s
D_G	diffusivity of CO_2 in liquid, m^2/s
D_L	diffusivity of alkanolamine in liquid, m^2/s
d_p	nominal size of packing = effective diameter of packing = $6\,(1-\varepsilon)/a_t$, m
G	superficial mass velocity of gas phase, $kg/s/m^2$
H	Henry's constant
h	height of rotating packed bed, m
I	enhancement factor
k_2	second order reaction rate constant, $m^3/kmol/s$
k_G	gas-phase mass transfer coefficient, m/s
k_L	liquid-phase mass transfer coefficient, m/s
K_Ga	overall volumetric mass transfer coefficient, 1/s
L	superficial mass velocity of liquid phase, $kg/s/m^2$
Q_G	gas flow rate, m^3/s
Q_L	liquid flow rate, m^3/s
r	radius, m
r_A	mass rate of production of species A by homogeneous chemical reaction, $kg/m^3/s$
r_i	inner radius of the packing, m
r_o	outer radius of the packing, m
v_r	velocity of species in r direction, m/s
v_θ	velocity of species in θ direction, m/s
v_z	velocity of species in z direction, m/s
Z	stoichiometric coefficient

Greek letters

α	CO_2 loading, mol of CO_2/mol of amine
μ_G	viscosity of gas, kg/m/h
μ_L	viscosity of liquid, kg/m/h
ρ	density, kg/m^3
ρ_G	density of gas, kg/m^3
ρ_L	density of liquid, kg/m^3
σ_c	critical surface tension, kg/s^2
σ_L	surface tension of liquid, kg/s^2
ν_G	dynamic gas viscosity, m^2/s
ν_L	dynamic liquid viscosity, m^2/s
ω_A	mass fraction of species A, dimensionless

Dimensionless groups

Fr_L	Froude number (L^2a_t/g)
Gr_L	liquid Grashof number ($d_p^2a_c/\nu_L^2$)
Ha	Hatta number
Re_G	gas Reynolds number ($G/a_t\nu_G$)
Re_L	liquid Reynolds number ($L/a_t\nu_L$)
Sc_G	gas Schmidt number (ν_G/D_G)
Sc_L	liquid Schmidt number (ν_L/D_L)
We_L	Weber number ($L^2\rho_L/a_t\sigma$)

Energy Information Administration [11], up to 2035, the shale gas production in the US will increase significantly and occupy 49% of the production of natural gas [12]. Natural gas can be applied extensively and is able to supply most of the needs of industrial energy and power generation. Thus, the development of the CCS process in natural gas power plants is required. However, due to the CO_2 concentration of natural gas power plants being lower than that of coal-fired power plants, in a range of 3–5 vol% [12,13], the conventional CO_2 capture processes applicable to high CO_2 concentration flue gases may not be conventional to the flue gases of natural gas power plants.

In addition to absorption, RPB has also been found to be applicable for stripping and adsorbent regeneration due to the existence of high mass and heat transfer areas as well as high mass and heat transfer rates [14–17]. Cheng et al. [18] indicated that the same regeneration efficiency could be achieved using an RPB whose volume was only one-tenth of the volume of the conventional stripper when using 7.0 m MEA as the absorbent. This approach thus is recommended to regenerate absorbent because less energy needs to supply during regeneration.

In this study, the mixed absorbent PZ/DETA with concentrations varying from 2.5 m/2.1 m to 4.0 m/8.0 m were applied to explore the feasibility to capture CO_2 using an RPB from the flue gases containing 4 vol% CO_2 and the absorbent regeneration in an RPB. As pointed out by Abu-Zahra et al. [19] that absorbent lean loading, concentration of absorbent and operating pressure in the stripping would affect the cost of the CO_2 capture process using a PB, those effects and the effects of the operation variables in an RPB were systematically examined in this study. Moreover, the CO_2 capture efficiency in a conventional PB was simulated using Aspen Plus as the comparison baseline to investigate the potential of volume reduction of an RPB in CO_2 capture process. With the study results, a more competitive process with a suitable absorbent formulation and an intensified apparatus were presented. In addition, the required energy in absorbent regeneration, the CO_2 absorption rate, and the absorber volume can be improved by applying the proposed approach to the low concentration CO_2 capture process of natural gas power plants.

2. Materials and methods

2.1. Materials

MEA, DETA, and PZ with a purity of 99% were purchased from Tedia, Aldrich, and Seedchem, respectively. N_2 and O_2 with a purity of 99.9% and CO_2 with a purity of 99.5% were purchased from Boclh Industrial Gases Co. (Taiwan). All the chemicals and gases were used as received. Aqueous alkanolamine solutions were prepared by adding a predetermined weight of individual or blended alkanolamines to deionized water. The CO_2 lean loading solutions were prepared by bubbling with pure CO_2 at room temperature. The CO_2 loading of each aqueous alkanolamine solution was measured by titration using an autotitrator (888 Titrando, Metrohm AG, Switzerland).

2.2. Apparatus

The details of the RPB apparatus and the operation used in this study can be found elsewhere [18,20]. For the RPB used to capture CO_2, the inner diameter, outer diameter, and height of the packing were 2.5, 12.5, and 2.3 cm, respectively. The total volume of the packing in the RPB was 270.9 cm^3. Stainless wire mesh was used as the packing material, with a surface area of 887.6 m^2/m^3 and a void fraction of 0.96. For the RPB used to regenerate the absorbents, the inner diameter, outer diameter, and height of the packing were 7.6, 16.0, and 2.0 cm, respectively. The total volume of the packing in the RPB was 311.4 cm^3. Stainless wire mesh was