



Experimental studies of regeneration heat duty for CO₂ desorption from diethylenetriamine (DETA) solution in a stripper column packed with Dixon ring random packing



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HIGHLIGHTS

- The regeneration heat duty of DETA was experimentally evaluated in a bench-scale stripper packed with Dixon ring.
- The regeneration heat duty of DETA was very sensitive to the operational parameters within the range presented in this work.
- The regeneration heat duty of DETA was lower than that of MEA for the same amount of CO₂ released.

ARTICLE INFO

Article history:

Received 8 April 2014

Received in revised form 3 July 2014

Accepted 9 July 2014

Available online 30 July 2014

Keywords:

Desorption

Carbon dioxide

Energy consumption

Diethylenetriamine

Packed column

ABSTRACT

The regeneration heat duty (Q_{reg} , kJ/kg CO₂) is a critical parameter in the post-combustion CO₂ capture process using a chemical solvent. In this study, the Q_{reg} of CO₂ desorption from CO₂ rich diethylenetriamine (DETA) solutions was experimentally evaluated in a bench-scale stripper column packed with Dixon ring random packing. The experiments were conducted to evaluate Q_{reg} over a solvent flow rate (L) range of 2.92–11.69 m³/m² h, amine concentration (C) range of 1.0–4.0 kmol/m³ and CO₂ cyclic capacity (Δa) range of 0.21–0.79 mol/mol. It was found that Q_{reg} was greatly influenced by all the three factors. In addition, a comparison of the regeneration performances between DETA and monoethanolamine (MEA) was performed to evaluate the potential for DETA's application in the CO₂ capture process. The results obtained in this work showed that for regeneration at the same absorption capacity, the heat duty of DETA was lower than that of MEA.

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1. Introduction

The implementation of post-combustion carbon dioxide (CO₂) capture from fossil fuel-fired power plants using reactive amine solvents is by far the most popular for reducing CO₂ emissions [1]. However, the main drawback of the amine-based CO₂ capture process is that the consumption of heat energy for solvent regeneration, referred to as regeneration heat duty (Q_{reg}), which accounts for about 70% of overall operating cost [2], is very high. Thus, the study of saving energy in the regeneration step is of vital importance.

In the industrial CO₂ capture process, Q_{reg} , provided by heat transfer from an external higher-temperature energy source such as low-pressure steam or hot oil in the reboiler, consists of three

parts: (1) absorption heat (q_{abs}) for breaking the chemical bond between CO₂ and amine-type solvent, (2) sensible heat (q_{sen}) for raising the temperature of the solution, and (3) vaporization heat (q_{vap}) for evaporating liquid water to vapor for CO₂ stripping. Consequently, Q_{reg} can be significantly affected by both the solvent type and operating conditions. Therefore, a comprehensive evaluation of Q_{reg} requirement for amine regeneration is crucial to providing accurate and reliable data for design as well as an economic evaluation of the amine-based CO₂ capture process. Up to date, there are three different important strategies for reducing Q_{reg} of the process, namely developing alternative energy efficient solvents, design of higher performance devices for mass and heat transfers, and optimization of the process configuration. Among these strategies, the development of new effective solvents is seen to be the biggest contributor in this area of research because it directly affects the capture performance and operating conditions. Also, it is easy to apply to existing CO₂ capture plants [3]. Presently,

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Nomenclature

a_{rich}	CO ₂ loading of the rich solutions, mol CO ₂ /mol amine	M_{CO_2}	molecular weight of CO ₂ , g/mol
α_{lean}	CO ₂ loading of the lean solutions, mol CO ₂ /mol amine	n_{amine}	molar flow rate of amine solution, kmol/h
Δa	difference in rich and lean CO ₂ loading, mol CO ₂ /mol amine	P_{CO_2}	partial pressure of CO ₂ , kPa
C	concentration, kmol/m ³	Q_{reg}	regeneration heat duty, kJ/kg
$C_{\text{oil},f}$	specific heat of heat transfer oil, kJ/(kg °C)	q_{abs}	absorption heat, kJ/kg
C_s	specific heat of the solution, J/(mol °C)	q_{sen}	sensible heat, kJ/kg
H_{reb}	reboiler heat duty, kJ/h	q_{vap}	vaporization heat, kJ/kg
H_{loss}	the system energy loss, kJ/h	R	universal gas constant, J/(mol °C)
L	solvent flow rate, m ³ /(m ² h)	T_{in}	temperature of the heat transfer oil inlet the reboiler, °C
m_{oil}	mass flow rate of heat transfer oil, kg/h	T_{out}	temperature of the heat transfer oil outlet the reboiler, °C
m_{CO_2}	mass flow rate of CO ₂ , kg/h	T_{reb}	temperature of the solution in reboiler, °C
m_s	mole flow rate of the solution, kmol/h	T_{feed}	temperature of the solution at the stripper inlet, °C

a variety of promising new amine-typed absorbents has been developed. These can be divided into three categories: (i) single amines such as diethylenetriamine (DETA) [4–7], 4-diethyl-amino-2-butanol (DEAB) [8,9], and 2-(1-piperazinyl)-ethyl-amine (PZEA) [10]; (ii) blended amines, such as monoethanolamine (MEA)–methyldiethanolamine (MDEA), 2-amino-2-methyl-1-propanol (AMP)–piperazine (PZ), and PZ–N, N-diethylethanolamine (DEEA) [11–14]; and (iii) hybrid solvents, such as MEA–Methanol, and MDEA–ionic liquids [15,16].

Diethylenetriamine (DETA), which has three amine functionalities, has been considered as a promising amine solvent for capturing CO₂ because of its characteristic higher performance than MEA (a benchmark solvent) in terms of absorption capacity, reaction kinetics and mass transfer rate. The work of Hartono et al. [7] shows that the kinetics rate constant of DETA is about 10 times higher than that of MEA at 298.1 K. Hartono et al. [6] also show that DETA had a higher solubility compared with other conventional amines. Fu et al. [4,5] reported that the mass-transfer performance in term of gas-phase volumetric overall mass transfer coefficient ($K_G a_V$) of CO₂ absorption into aqueous DETA in a packed column was better than that of MEA. However, Q_{reg} of DETA, a very important parameter, has not yet been reported, which makes the proper evaluation of DETA's potential for use as an absorbent for CO₂ capture to be incomplete. It is therefore essential to perform such studies using DETA. A packed column has been considered to be a practical equipment to use for investigating Q_{reg} in studies of amine regeneration. This is because a packed column can provide an operating system similar to the real CO₂ capture process, and has been extensively applied to investigating the regeneration performance of different amine solvents [17–21].

In the present work, Q_{reg} of CO₂ desorption from aqueous solutions of DETA was experimentally measured in a bench-scale stripper packed with Dixon ring random packing. The effects of key operating parameters, including solvent flow rate (L), amine concentration (C) and CO₂ cyclic capacity (Δa) on Q_{reg} were investigated. Furthermore, Q_{reg} as well as its components (i.e., q_{abs} , q_{sen} and q_{vap}) for DETA were compared with those of MEA. The results obtained are presented and discussed in this paper.

2. Regeneration heat duty

In this work, the reboiler heat duty (H_{reb} , kJ/h), which was supplied by heat transfer oil, was calculated by the following equation:

$$H_{\text{reb}} = m_{\text{oil}} C_{\text{oil},f} (T_{\text{in}} - T_{\text{out}}) \quad (1)$$

where m_{oil} and $C_{\text{oil},f}$ represent the mass flow rate (kg/h), specific heat (2.3 kJ/kg °C, as provided by the vendor) of the heat transfer

oil, respectively. Also, T_{in} and T_{out} are the inlet and outlet temperatures of the heat transfer oil from the reboiler (°C), respectively.

The Q_{reg} (kJ/kg) for solvent regeneration can be calculated from the ratio of the effective reboiler heat duty and the CO₂ mass flow rate as follows:

$$Q_{\text{reg}} = \frac{H_{\text{reb}} - H_{\text{loss}}}{m_{\text{CO}_2}} \quad (2)$$

$$m_{\text{CO}_2} = n_{\text{amine}} (\alpha_{\text{rich}} - \alpha_{\text{lean}}) M_{\text{CO}_2} \quad (3)$$

where H_{loss} is the system energy loss (kJ/h), m_{CO_2} is the CO₂ mass flow rate (kg/h), n_{amine} is the molar flow rate of amine solution (kmol/h), M_{CO_2} is the molecular weight of CO₂ (g/mol). α_{rich} and α_{lean} are the CO₂ loadings of the rich and lean solutions (mol/mol), respectively. It should be noted that H_{loss} inevitably exists in the experiment, and the approach to obtain H_{loss} in this work is given in Section 3.3.

As mentioned earlier, the regeneration heat duty, Q_{reg} , provided for the solvent regeneration generally includes three parts, namely q_{abs} , q_{sen} and q_{vap} (kJ/kg), as follows.

$$Q_{\text{reg}} = q_{\text{abs}} + q_{\text{sen}} + q_{\text{vap}} \quad (4)$$

where q_{abs} , q_{sen} , and q_{vap} can be calculated as in the following equations:

$$q_{\text{abs}} = R \frac{d(\ln P_{\text{CO}_2})}{d(1/T)} \quad (5)$$

$$q_{\text{sen}} = m_s C_s (T_{\text{reb}} - T_{\text{feed}}) / m_{\text{CO}_2} \quad (6)$$

$$q_{\text{vap}} = Q_{\text{reg}} - q_{\text{abs}} - q_{\text{sen}} \quad (7)$$

where R is the universal gas constant (J/(mol °C)), P_{CO_2} is the CO₂ partial pressure (kPa), T is the temperature (°C), m_s and C_s denote the molar flow rate (kmol/h) and the specific heat of the solution (J/(mol °C)), respectively. T_{reb} and T_{feed} are the temperatures of the solution in the reboiler and the stripper inlet (°C), respectively. The reaction heat and the specific heat of solution for DETA and MEA were obtained from the literature [22–24].

3. Experimental section

3.1. Chemicals

Reagent grade DETA and MEA were purchased from Tianjin Kermel Chemical Reagent Co. Ltd., China, each with purity of $\geq 98.0\%$. Commercial grade CO₂ was supplied by Changsha Rizhen Gas Co. Ltd., China, with a purity of $\geq 99.9\%$.

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