



# Detailed experimental study on the performance of Monoethanolamine, Diethanolamine, and Diethylenetriamine at absorption/regeneration conditions



Weidong Fan<sup>\*</sup>, Yacheng Liu, Kang Wang

School of Mechanical and Power Engineering, Shanghai Jiao Tong University, Shanghai 200240, People's Republic of China

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## ABSTRACT

Experiments were carried out in a lab-stripper to investigate the regeneration performance of CO<sub>2</sub>-loaded solutions of Monoethanolamine (MEA) under various operational conditions (energy input, rich solution flow rate, rich solution loading). To better understand the regeneration process of a rich MEA solution, appropriate evaluation of the absorption and regeneration capacities of an aqueous MEA solution were first determined. Thus, first of all, a bubbling reaction system was employed to assess the absorption and desorption capacities of MEA, and other amines, such as Diethanolamine (DEA) and Diethylenetriamine (DETA), were introduced to compare these capacities in terms of amino group. The influences of the solution temperature and the solution concentration on CO<sub>2</sub> absorption and desorption performances were investigated. The results show that these factors significantly affect CO<sub>2</sub> absorption and desorption. Moreover, experiments in the bubbling reaction system were conducted to compare the regeneration capacities of the three CO<sub>2</sub>-loaded solutions (MEA, DEA, DETA) with increasing concentrations under the condition of different sweep gases (100% N<sub>2</sub>, 14% CO<sub>2</sub> and 100% CO<sub>2</sub>). The data show that under the condition of each type of sweep gas, the regeneration capacity of the three types of amino groups followed the rule: DEA > DETA > MEA. On the basis of these data obtained from the reaction system, regeneration experiments with loaded 2 mol/L MEA solutions in a stripper column were conducted to analyze the relationships between the regeneration ratio and the stripper operating parameters.

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## 1. Introduction

Because global warming has caused worldwide concern and carbon dioxide (CO<sub>2</sub>) emissions are continually growing, reducing CO<sub>2</sub> emission has become an important task for the whole world (Mao et al., 2014; Kainiemi et al., 2015). The main source of CO<sub>2</sub> is from the production of electricity, the majority of which is derived from pulverized coal-fired boilers. Therefore, people have made a great effort to develop suitable technologies for capturing CO<sub>2</sub> from large-scale CO<sub>2</sub> emission sources in industry. Although numerous technologies are generally compatible with carbon capture and storage (CCS) activity and are applied in the chemical and petroleum industries, relatively few have gained any measure of acceptance for reducing the considerable CO<sub>2</sub> emissions from coal-fired

plants (Glier and Rubin, 2013). In general, for large point sources, especially coal-fired power plants, three acceptable technology options are suitable for commercial deployment; these are amine-based post-combustion CO<sub>2</sub> capture, oxy-fuel combustion, and calcium looping technologies (MacDowell et al., 2010). However, amine-scrubbing technology may be a more feasible technology for CO<sub>2</sub> capture from coal-fired power plants because of its retrofit approach for capturing CO<sub>2</sub> from existing power plants (Mazari et al., 2015). Moreover, it is well-suited to capturing CO<sub>2</sub> from dilute, low-pressure streams. The amine solvents absorption method as one main mode of post-combustion CO<sub>2</sub> capture has become a more mature CO<sub>2</sub> capture approach among CCS technologies applied in large coal-fired power plants for capturing CO<sub>2</sub> because of its higher reliability, greater suitability, and lower investment and running cost for decarbonization retrofit (Molina and Bouallou, 2015).

Monoethanolamine (MEA), as a primary amine, has been widely employed as the CO<sub>2</sub>-absorbent of the chemical absorption method

<sup>\*</sup> Corresponding author. Tel.: +86 21 34208287; fax: +86 21 34206115.  
E-mail address: [wdfan@sjtu.edu.cn](mailto:wdfan@sjtu.edu.cn) (W. Fan).

because of its high CO<sub>2</sub> reactivity. Therefore, researchers gave more attention to MEA-based processes, such as the development of novel technology using MEA, its model setup, its applications in pilot plants and energy consumption analysis (Kuntz and Aroonwilas, 2009; Faramarzi et al., 2010; Moser et al., 2011). Energy consumption in the regeneration process is higher for MEA, which is its main drawback compared to Diethanolamine (DEA) and Diethylenetriamine (DETA) (Zhang et al., 2014). DEA, as a secondary amine, is also suitable for the removal of CO<sub>2</sub> from flue gas but exhibits slow kinetics. DETA, as an alkyl amine, has also received great attention because it owns three amino groups in each single molecule, and it has been used as an activator to improve CO<sub>2</sub> absorption performance because of its fast reaction kinetics and high absorption capacity (Fu et al., 2012). DETA has three function groups, two primary amino groups and one secondary amino group, while MEA has one primary amino group and DEA has one secondary amino group. Most studies compared absorption/desorption capacities among different aqueous solvents in terms of the same mole amine concentration rather than the same amino group concentration (Rivera-Tinoco and Bouallou, 2010). The absorption capacity of DETA is higher than that of MEA and DEA because it has two primary amino groups and one secondary amino group per mole amine concentration (Liu et al., 2014). However, studies comparing CO<sub>2</sub> absorption and desorption capacity for various solvents in terms of amino group concentration may be more reasonable. Schäffer et al. (2012) did research on the comparison of MEA and Triethylenetetramine (TETA) under the same mole amino groups, but they analyzed the results from the aspect of amine molecule instead of amino group. In comparison with the major attention paid to improving the efficiency of the CO<sub>2</sub> absorption process, experimental research on the regeneration performance of aqueous MEA solution in a bubbling reaction system and a stripping column is less abundant. A similar experimental apparatus was employed in the literature reported by Galindo et al. (2012). They compared the regeneration characteristics between 20 wt. % MEA and 34 wt. % DEA for the same number of amino groups with the temperature ranging from 80 °C to 100 °C, and the results showed a higher cyclic capacity for DEA and lower dependence on CO<sub>2</sub> partial pressure for desorption compared to MEA. However, in their lab-scale stripper, the stripping steam was mainly produced in the reboiler.

The objective of this paper was to examine and compare the performances of three aqueous solutions of MEA, DEA, and DETA in terms of amino group under various operational conditions in the bubbling reaction system. Then, the regeneration experiments of CO<sub>2</sub>-loaded solutions of MEA were conducted under various operational conditions in a lab-stripper column. The main aim was to analyze the relationships between the regeneration ratio and the stripper operational parameters (energy input, rich solution flow rate, rich solution loading). First, the absorption capacity of the three CO<sub>2</sub>-loaded solutions at different amino group concentrations and temperatures were determined. The effects of the temperature and concentration with various sweep gases (100 vol. % N<sub>2</sub>, 14 vol. % CO<sub>2</sub> and 100 vol. % CO<sub>2</sub>) on the regeneration capacity were investigated. In light of these data obtained from the reaction system, the regeneration experiments on loaded 2 mol/L MEA solutions in a stripper column were conducted.

## 2. Theoretical background

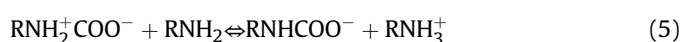
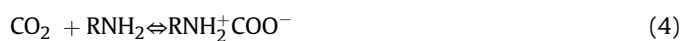
### 2.1. Absorption reaction mechanism

The reaction mechanism between CO<sub>2</sub> and MEA (DEA) can be described by the zwitterion mechanism, and both MEA as the primary amine and DEA as the secondary amine can react with CO<sub>2</sub> to

form carbamate and protonated amine molecules, limiting the theoretical loading to 0.5 mol CO<sub>2</sub>/mol amine by stoichiometry. However, the formation of bicarbonate or carbonate results in CO<sub>2</sub> loading over the theoretical value (Choi et al., 2014). Thus, overall reactions of CO<sub>2</sub> with MEA (RNH<sub>2</sub>) and DEA (RR'NH) can be represented as follows:



MEA as a primary amine can react with CO<sub>2</sub> faster and form more stable carbamates than DEA. The reaction of CO<sub>2</sub> with MEA consists of two steps: first, the formation of zwitterions, and then, deprotonation of the zwitterions (Choi et al., 2009).



As an alkyl amine, DETA has two primary amino groups and one secondary amino group. Hartono et al. (2007) performed the qualitative determination of the species present in the DETA–CO<sub>2</sub> system by using the nuclear magnetic resonance (NMR) technique; the results suggested that carbamate was the main species in the system for loading below 1.0 mol CO<sub>2</sub>/mol amine, and, while exceeding that value, dicarbamate was dominating and bicarbonate or carbonate were also formed.

### 2.2. Regeneration reaction mechanism

During the regeneration process of aqueous solutions of amine, the regenerable carbamate and bicarbonate or carbonate are thermally decomposed to liberate CO<sub>2</sub> from the rich solutions. Because of the unstable ions' rank (Choi et al., 2014): bicarbonate or carbonate > secondary carbamate > dicarbamate > primary carbamate, the regeneration of a rich MEA solution is more difficult than that of DEA or DETA. Thus, the overall regeneration process consists of the following reactions (6 and 7) and the reverse reactions (1, 2, and 3).



## 3. Experimental considerations

### 3.1. Experimental apparatus and procedure of the bubbling reaction system

The absorption and regeneration capacities of three aqueous solutions (MEA, DEA, DETA) were measured in the bubbling reaction apparatus illustrated in Fig. 1; the main experimental operational parameters are summarized in Table 1. The solution temperature was controlled by an electrical heating jacket with a deviation of ±1 K. A condenser circulating cooling water was used at the outlet of the three-neck flask to reduce the loss of water and solvent because of evaporation. The inlet gas was fed into the amine solution through the transfer switches and the mass flowmeter, and the outlet gas flow rate was monitored by another mass flowmeter. Prior to the absorption test, aqueous solutions with concentrations of from 1 to 4 mol amino group/L were prepared.

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