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Evaluating CO_2 desorption performance in CO_2-loaded aqueous tri-solvent blend amines with and without solid acid catalysts

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HIGHLIGHTS

- All the MEA-AMP-PZ blends showed better CO_2 desorption performance than MEA.
- The 3 M MEA-2.5 M AMP-0.5 M PZ with AMP/PZ molar ratio of 5 has the most bicarbonate and least carbamate.
- A possible catalytic CO_2 desorption pathway is proposed.
- The combination of solid acid catalyst with MEA-AMP-PZ can reduce 44.3–61.6% energy consumption for CO_2 capture.
- MSA × TAS of solid acid catalyst has the most important effect on CO_2 desorption.

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ABSTRACT

To enhance the energy efficiency of the CO_2 desorption process, the regeneration behaviors of the CO_2-loaded 6 M MEA-AMP-PZ (monoethanolamine, 2-amino-2-methyl-1-propanol and piperazine) tri-solvent blends with different AMP/PZ molar ratios with four different solid acid catalysts (H-ZSM-5, γ-Al_2O_3, SAPO-34 and SO_4^{2-}/TiO_2) and a blank at 96 °C were investigated in terms of CO_2 desorption rate, cyclic capacity and relative heat duty. For the no-catalytic runs, the results showed that all the tri-solvent blended amines greatly increased CO_2 desorption rate, cyclic capacity and decreased the relative heat duty in comparison with 5 M MEA. The ^{13}C NMR analysis indicated that the 3 M MEA-2.5 M AMP-0.5 M PZ blend with the highest AMP/PZ ratio produced the largest amount of bicarbonate and less carbamate, which resulted in the best desorption performance. With regard to catalyst, when the solid acid catalysts were introduced, the regeneration performance of the blend was further improved. The best of the blends along with H-ZSM-5 provided the combination with the best performance in CO_2 desorption, and reduced the relative heat duty by 61.6% as compared to 5 M MEA without catalyst (100%). Five relevant physicochemical properties of the catalyst were obtained and used to better understand the catalytic regeneration process. A possible catalytic CO_2 desorption mechanism was analyzed. The results revealed that the mesopore surface area coupled with total acid sites of the catalyst had the most positive influence on improving the CO_2 desorption performance. Findings from this work imply that the combination of solid acid catalyst with tri-solvent blended amines is a promising alternative method for further reduction of solvent regeneration energy requirement.

1. Introduction

Global warming caused by the emission and accumulation of greenhouse gases in the atmosphere has become the subject of public concern in past decades. Carbon dioxide (CO_2) is widely considered to be a predominant contributor of greenhouse gases, and the global annual CO_2 emissions have escalated by approximately 80% from 1970 to 2004 [1,2]. Given the continuing huge demand for fossil fuels and the resulting release of CO_2 to the atmosphere, it is thus an imperative to develop and improve methods to mitigate CO_2 emissions. According to Intergovernmental Panel on Climate Change (IPCC), atmospheric CO_2 concentration must be limited to 450 ppm by 2100 to avoid global

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Nomenclature

Abbreviations

IPCC	Intergovernmental Panel on Climate Change
CO_2	carbon dioxide
H_{des}	desorption heat
H_{vap}	vaporization heat
H_{sen}	sensible heat
MEA	monoethanolamine
AMP	2-amino-2-methyl-1-propanol
PZ	piperazine
DETA	diethylenetriamine
1-(2-HE)PRLD	1-(2-hydroxyethyl) pyrrolidine
1DEA2P	1-diethylamino-2-propanol
MDEA	methyldiethanolamine
EDA	ethylene diamine
BET	Brunauer-Emmett-Teller
B/L ratio	the ratio of Brϕnsted to Lewis acid sites
MSA	mesopore surface area (m^2/g)
TAS	total acid sites (μmol/g)
MSA × TAS	the product of MSA and TAS

Symbols

CC	cyclic capacity (mol)
α	CO_2 loading (mol CO_2/mol amine)
H	regeneration heat duty (kJ/mol)
RH	relative heat duty (%)
$[CO_2]_0$	the total amount of CO_2 in the amine solution (mol/L)
$C(H^+)$	hydrogen ion concentration (mol/L)
PZC	point of zero charge

temperatures rising by more than 2 °C [3–6].

Generally, there are three main process options for CO_2 capture: pre-combustion, oxy-fuel combustion and post-combustion [7–11]. Chemical solvent absorption, solid sorbent adsorption, cryogenic distillation and membrane separation are important techniques in post-combustion capture. Among all the capture technologies, chemical absorption technology using amine based solvent has attracted the most attention because of its cost-effectiveness, maturity and capacity to absorb large volumes of flue gas [12–16]. However, there are three major drawbacks of this method: amine solvent degradation, equipment corrosion, and high energy consumption for solvent regeneration. The large quantity of energy that is required during solvent regeneration for CO_2 stripping accounts for about 70–80% of the total cost in the CO_2 capture system, and is the one of biggest barriers for post-combustion CO_2 capture using chemical absorption [17,18].

According to the literature [13,19–21], the heat duty (H) for a CO_2-loaded amine solvent regeneration involves three parts: H_{des}, H_{vap} and H_{sen}, as follows.

$$H = H_{des} + H_{vap} + H_{sen}$$

where H_{des} is the desorption heat to destroy the chemical bonds between CO_2 and amine (or the absorption heat), H_{vap} is the vaporization heat to produce the water vapor for solvent regeneration, while H_{sen} is the sensible heat to raise the temperature of the rich amine solution. H_{des} primarily determined by the type of amine and the lean CO_2 loading of the regeneration process, which changes slightly under identical regeneration conditions. Oexmann et al. [22] specified that about half of the total regeneration heat duty comes from the H_{des} for 30 wt% monoethanolamine (MEA) solution. H_{des} is generally high for primary and secondary amines, while the tertiary amine solvents possess low H_{des}. The H_{vap} can be greatly affected by both the regeneration temperature and the amount of water vapor in the amine system. H_{sen} can be decreased by lowering the desorption temperature and stripping a larger amount of CO_2 (increasing the cyclic capacity).

In recent years, considerable research effort has been devoted to reducing the regeneration energy requirement. In general, there are several common methods, including process optimization, solvent innovation and other novel methods [13,23–25]. Khalipour et al. [26] decreased the energy consumption of CO_2 desorption by enhancing the efficiency of the Heat Exchanger Network (HEN). Other modified processes such as solvent split-flow, vacuum stripper, solvent inter-cooling and multi-pressure of the capture process were also proposed and studied [27,28]. On the other hand, the development of efficient solvents, including, (a) single amines (diethylenetriamine (DETA) [19], 1-(2-hydroxyethyl) pyrrolidine (1-(2-HE)PRLD) and 1-diethylamino-2-propanol (1DEA2P)) [29,30]; (b) hybrid solvents, such as MEA-methanol [31]; and (c) blends of amines have been studied for CO_2 capture to reduce the regeneration energy penalty. Chakravarty et al. [32] studied the heat duty of bi-solvent blends (binary mixtures) in the amine regeneration process. It was found that the blended amines perform better in terms of a faster desorption rate and a lower energy requirements for CO_2 desorption in comparison with a single amine solvent. Different bi-solvent blends (ternary mixtures) have also been investigated, such as MEA-MDEA (methyldiethanolamine) and MEA-AMP (2-amino-2-methyl-1-propanol) [33,34].

Recently, some researchers reported that tri-solvent blend amine solvents are an effective method for CO_2 capture, because they not only utilize the advantages of the individual aminea, but also address their individual disadvantages [35]. Nwaoha et al. [36,37] studied the CO_2 desorption performance of tri-solvent blends MEA-AMP-PZ (piperazine) and DETA-MDEA-AMP. Their results indicated that the tri-solvent amine blends had faster desorption rate and lower regeneration energy requirement in comparison with the single MEA solution. Liu et al. [38] investigated CO_2 regeneration performances of bi-solvent blends MEA-MDEA, and a tri-solvent blend MEA-MDEA-AMP. Based on the results of this work, the tri-solvent blended amines could be considered as potential novel absorbents for CO_2 capture in post-combustion processes in terms of heat duty. However, the heat duty of tri-solvent blends amines is still high, and needs further reduction.

Other novel methods also have been studied to reduce energy consumption for solvent regeneration. Several studies have focused on the addition of nanoparticles (SiO_2, Al_2O_3, TiO_2 and MgO) or soluble organic weak acid into the amine solvent to enhance liquid-side mass-transfer coefficient and improve desorption rate, thereby reducing the regeneration heat duty [39–42]. However, some of the additives cannot be separated completely from the amine system and may have negative influence on the CO_2 absorption rate. Also, these methods to reduce the energy demand for solvent regeneration still have not reached theoretical minimum regeneration heat energy value of 0.4356 MJ/kg CO_2 [43] and have not yet resulted in major decreases in operation costs. Thus, it remains a great challenge to develop highly efficient technologies to reduce the energy requirement for CO_2 desorption to an acceptable level.

Another promising innovative strategy to greatly reduce energy penalty has been proposed by Idem et al. [44,45]. They showed that the addition of solid acid catalysts such as H-ZSM-5 and γ-Al_2O_3 into a rich loaded amine solution for CO_2 stripping is an effective method to decrease the heat duty. The catalytic performance of solid acid catalysts in single solvent (MEA) and bi-solvent blend solvents (MEA-DEAB and MEA-MDEA) have been studied. Furthermore, Zhang et al. [46] revealed that the small-pore molecular sieve SAPO-34 and sulfated titanium dioxide (SO_4^{2-}/TiO_2) had good catalytic performance for the CO_2 loaded MEA solvent regeneration process.

Idem et al. [47] stated that the addition of solid acid catalysts to

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