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Spray absorption of CO₂ into monoethanolamine: Mass transfer coefficients, droplet size, and planar surface area

Y. Tamhankar^a, B. King^a, J. Whiteley^a, T. Cai^b, K. McCarley^b, M. Resetarits^c, C. Aichele^{a,*}

^a School of Chemical Engineering, Oklahoma State University, Stillwater, OK 74078, USA

^b Fractionation Research Incorporated, Stillwater, OK 74074, USA

^c Independent Separations Consultant, Lakewood Ranch, FL 34202, USA

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ABSTRACT

Spray columns show potential to absorb CO₂ into aqueous amine solvents such as monoethanolamine (MEA). Low gas phase pressure drop, less susceptibility to corrosion due to the absence of internals, and greater tolerance to use of precipitating solvents, are some of the favorable attributes associated with spray columns. At present, there is a need for robust spray absorption data with the CO₂-MEA system. Experimental droplet size measurements with MEA as the test solvent have never been reported. Measurement of droplet size distributions can help ascertain the surface area available for absorption. In this spray study, absorption rates in terms of overall mass transfer coefficients, drop size data, and planar surface area for the CO₂-MEA system, quantified in a 0.2 m ID, lab-scale column are presented. The droplet size measurements are made with a Phase Doppler Interferometry (PDI) system. The effects of liquid rate, inlet loading, MEA concentration, and gas-liquid contact height on the performance of a full-cone nozzle are elucidated. Results from the study show an increase in mass transfer coefficients with MEA concentration and liquid rate. The mass transfer data, droplet size data and surface area quantification indicate that the free MEA content dominates spray absorption. The data also indicate the occurrence of a great degree of mass transfer in the region immediate downstream of the nozzle tip. The study provides fundamental insight into spray absorption phenomena and can aid in more robust, application specific design of spray columns.

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1. Introduction

Aqueous amine absorption and stripping of CO₂ from flue gas, shown in Fig. 1, is the most well established technology for post combustion Carbon Capture and Storage (CCS) (Kuntz, 2006; Oexmann and Kather, 2010; Rochelle et al., 2011). At present, there is a need to reduce operating and capital costs of the process to make it commercially viable (Rochelle, 2009a). High pressure drops associated with column internals

(packings or trays), and large reboiler duties (steam driven) required to regenerate the solvent are two significant sources which contribute to the high capital and operating costs (Oyeneke and Rochelle, 2009; Tsai, 2010). The costs for the process can be reduced by employing more efficient gas-liquid contactors, and or better solvents.

30 wt% Monoethanolamine (MEA), is widely used in natural gas processing for CO₂ removal, and is considered as the standard against all other solvents which are benchmarked.

* Corresponding author. Tel.: +1 405 744 5280; fax: +1 405 744 6338.

E-mail address: clint.aichele@okstate.edu (C. Aichele).

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Nomenclature

A	area of concentric circular zone, (m ²)
A _C	column cross sectional area, (m ²)
A _{plume}	cross sectional area of spray plume, (m ²)
C	correction factor for drop count
C _{MEA}	concentration of MEA, (kmol/m ³)
d	drop diameter, (μm)
D _{CO₂,1}	diffusivity of CO ₂ in MEA, (m ² /s)
d _o	orifice diameter or free passage of nozzle, (m)
D ₃₂	Sauter mean diameter, (μm)
E	enhancement factor
F _A	fractional surface area
G	total gas rate, (m ³)
G ₁	inert gas rate, (kmol/min)
H _{CO₂}	Henry's constant for CO ₂ in MEA, (m ³ atm/kmol)
k _g	local gas side mass transfer coefficient, (kmol/m ² s atm)
K _G	overall gas side mass transfer coefficient, (kmol/m ² s atm)
k _{g'}	local liquid side mass transfer coefficient in gas units, (kmol/m ² s atm)
K _{Ga_e}	overall volumetric mass transfer coefficients, (kmol/m ³ min atm)
k ₁ ⁰	physical local liquid side mass transfer coefficient, (m/s)
k ₂	rate constant, (m ³ /kmol s)
L	total liquid rate, (Lit or m ³)
L _{MEA}	MEA liquid rate, (kmol/min)
m _L	mass of liquid or solvent, (kg)
N _{CO₂}	CO ₂ flux, (kmol/min)
n	drop count
p _{CO₂}	CO ₂ partial pressure, (atm)
P _L	liquid pressure, (Pa or atm)
P _{SA}	planar surface area, (m ²)
R	gas constant, (m ³ atm/kmol K)
S	surface area of all drops through concentric circular zone, (m ²)
T	temperature, (K)
V	volume of solvent, (m ³)
V _{spray}	volume of solvent sprayed, (m ³)
Y _{CO₂,in}	CO ₂ mole ratio in inlet gas
Z	column height, (m)
Subscript	
i	bin in dropsize measurement
in	gas inlet
j	1, 2, 3... = concentric circular zone number
lm	logarithmic
out	gas outlet
Superscript	
*	equilibrium
Greek	
α _{lean}	lean loading, (mol CO ₂ /mol MEA)
α _{rich}	rich loading, (mol CO ₂ /mol MEA)
ρ _L	density of MEA solution, (kg/m ³)
μ _L	dynamic viscosity, (kg/m s)
σ	surface tension, (kg/s ²)
Δ	difference
∞	infinite

Other

[]	free or active
'	rate, (s ⁻¹)

Fast mass transfer rates, high cyclic capacity, high heat of absorption, and low relative cost are favorable attributes of MEA (Maddox, 1974; Kohl and Nielsen, 1997; Clossmann, 2011). The concentration of MEA is limited to 30 wt% in order to prevent corrosion of column internals (Kohl and Nielsen, 1997). For a fixed CO₂ removal duty, the MEA concentration governs the recirculation rate and the regeneration duty.

Spray columns seem to be well suited for absorbing CO₂ into MEA. The low gas phase pressure drop associated with spray columns can significantly reduce blower costs (Mehta and Sharma, 1970). Additionally, the absence of internals permits the use of higher MEA concentrations since the corrosion of column internals is avoided. A 12% reduction in reboiler duty by use of 40 wt% MEA instead of 30 wt% MEA has been reported (Abu-Zahra et al., 2007). Further, sprays can offer greater tolerance to solvents which tend to precipitate out from the solution. Such solvents will likely choke packings. Thus, spray columns show sufficient promise in reducing the cost of the absorption-stripping CCS process to warrant investigation.

Design of spray columns is highly empirical and application specific. Spray absorption rate data quantified in terms of the overall volumetric mass transfer coefficients is required for robust design of spray columns (Javed et al., 2010). At present, reliable spray absorption data for the CO₂-MEA system is not readily available. Experimental dropsize measurements inside MEA sprays are non-existent. Further, there is a need to ascertain the effect of solvent concentration on the spray absorption rates, dropsize distribution, and available surface area. Increasing MEA concentration results in greater free MEA content on one hand, resulting in greater absorption rates. On the other hand, the increased diffusive resistance due to increased viscosity results in the lowering of absorption rates. The present study is a step towards addressing this gap.

2. Background

Investigation of CO₂ absorption into liquid sprays has been moderately studied in the literature. However, most of the previous studies involved absorption of CO₂ into NaOH (Mehta and Sharma, 1970; Taniguchi et al., 1997; Taniguchi et al., 1999; Dimiccoli et al., 2000; Turpin et al., 2008; Javed et al., 2010; Bandyopadhyay and Biswas, 2012). Spray absorption of CO₂ in a Monoethanolamine (MEA) spray has scarcely been investigated (Kuntz and Aroonwilas, 2008; Koller et al., 2011).

Kuntz and Aroonwilas (2008) were the first to study absorption of CO₂ into an MEA spray. Mass transfer measurements were made inside a small, lab scale spray column. The effect of inlet gas CO₂ composition, inlet loading, liquid rate, gas rate, MEA concentration and nozzle size on the mass transfer coefficients in the spray zone (no wall flow) was elucidated. The performance of the spray column was compared to another lab-scale packed column. Further, an attempt was made to ascertain the interfacial area and the gas side local film mass transfer coefficient.

Koller et al. (2011) measured cyclic absorption rates of CO₂ into MEA inside a pilot plant facility with 30 wt% MEA as the

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