

## Enhancements in mass transfer for carbon capture solvents part II: Micron-sized solid particles



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### ABSTRACT

The mass transfer of CO<sub>2</sub> into amine-based carbon capture solvents was improved with the addition of heterogeneous micron- and nano-sized particle additives. Enhancement in CO<sub>2</sub> removal rate was determined in a stirred reactor with a flat steady gas-liquid interface and in a small packed-bed absorption column. The screening of several particle types in the stirred reactor indicated that carbon particles are best suited to enhance the rate of CO<sub>2</sub> diffusion in 30 wt.% monoethanolamine (MEA), and tests in the small packed-bed absorption column showed improved CO<sub>2</sub> mass transfer in three solvents, MEA, piperazine (PZ), and hexanediamine (HDA), upon addition of nano-sized activated carbon particles. The enhancement of CO<sub>2</sub> capture efficiency and  $K_G$  increases with increasing solvent kinetics, in order of methyldiethanolamine (MDEA) < MEA < PZ < HDA, confirming the particle additives have a larger impact on kinetically faster, liquid-side diffusion limited, solvents. The combined benefits of the particle additives were then demonstrated on an absorption-desorption integrated bench-scale CO<sub>2</sub> capture unit, in both MEA and the thermally stable advanced amine blend: 1-amino-2-propanol (A2P)/2-amino-2-methyl-1-propanol (AMP). The combined system translated to an overall 26% reduction in energy penalty for CO<sub>2</sub> capture on the CAER Bench-scale unit.

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### 1. Introduction

Climate change is of global concern, and many countries have agreed to reduce their CO<sub>2</sub> emissions in order to mitigate its impact (Rogelj et al., 2016; Paris Agreement, 2016). Reducing CO<sub>2</sub> emissions from stationary sources such as coal-fired or natural gas-fired power plants will be a major part of any CO<sub>2</sub> reduction strategy, and aqueous amine-based post-combustion CO<sub>2</sub> capture is the most studied due to its flexibility of heat integration, fast kinetics, and scale of demonstration (Yang et al., 2008; Rochelle, 2009; Wang et al., 2011; Boot-Handford et al., 2014). In amine-based capture processes, two equivalents of a primary or secondary amine, or one equivalent of a tertiary amine, react with CO<sub>2</sub> in an absorber column to form carbamate, or bicarbonate and protonated amine respectively (Scheme 1). The ion pair is then subjected to a stripping process to generate a concentrated stream of CO<sub>2</sub>, which is isolated

for storage or utilization, and the regenerated amine is recycled back to the absorber (Fig. 1).

Ethanolamine (MEA) is a primary amine that readily forms carbamate upon exposure to CO<sub>2</sub>, and its 30 wt.% (5 M) aqueous solution is the widely accepted benchmark CO<sub>2</sub> capture solvent (Tan et al., 2012). Many research efforts have focused on the development of advanced amine solvents with more desirable properties, such as enhanced CO<sub>2</sub> capture rates, loading capacity, increased thermal stability, or lower regeneration energy requirements relative to MEA (Liang et al., 2015; Lee et al., 2013; Puxty et al., 2009). Tertiary amines such as *N*-methyldiethanolamine (MDEA) and *N,N*-diethylethanolamine (DEEA) boast lower regeneration energy than MEA but are slow to react with CO<sub>2</sub> (Chowdhury et al., 2013). Secondary amines, piperazine in particular, have attractive CO<sub>2</sub> capture properties compared to MEA, such as faster absorption, lower regeneration energies, and higher capacities (Bishnoi and Rochelle, 2000). However, secondary amines readily react with flue gas contaminants (NO<sub>x</sub>) to form environmentally hazardous nitrosamines (Dai et al., 2012). Alkanolamine alternatives to MEA are also more expensive (Bara, 2012), leading some researchers to look at solvent blends containing MEA in combina-

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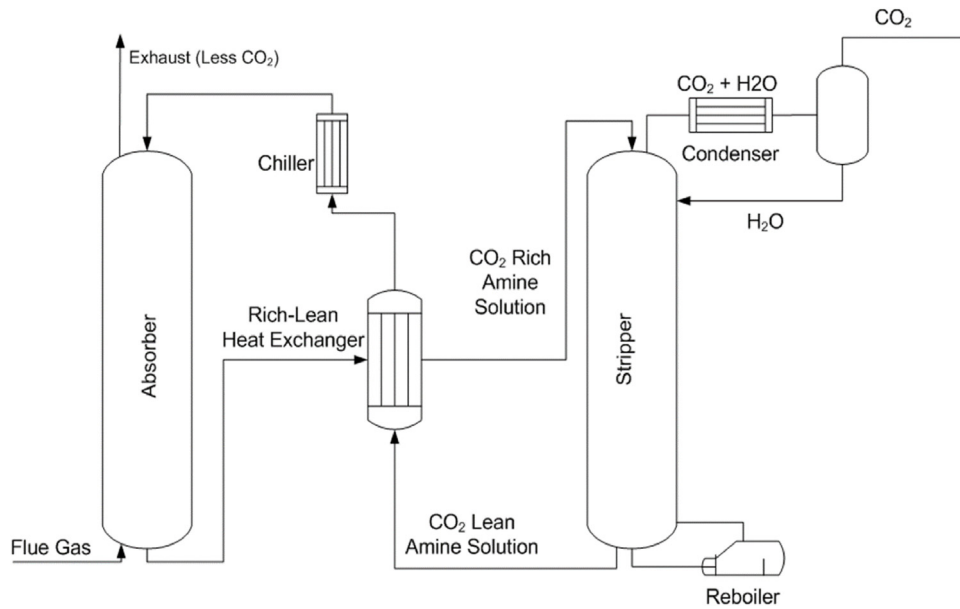
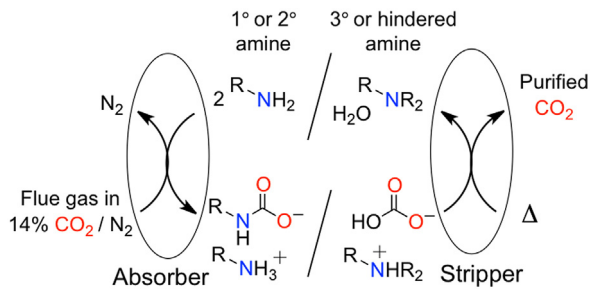


Fig. 1. Overall process flow diagram of the carbon capture process.



Scheme 1. Reversible reaction of MEA with CO<sub>2</sub> to form carbamate and protonated amine.

tion with other amines to obtain the desirable properties of both (Hagewiesche et al., 1995; Conway et al., 2015). This strategy comes with its own problems, e.g., higher amine degradation and (still) higher cost, among others (Idem et al., 2006). The difficulties in improving the chemical properties of CO<sub>2</sub> capture amines inspired us to consider improving the physical aspect of the process such as diffusivity, and thus the overall CO<sub>2</sub> flux into the capture solvent.

Early reports on the mass transfer of slurries show that the addition of particles enhances the volumetric mass transfer coefficient  $k_L a$  (Joosten et al., 1977; Alper et al., 1980; Quicker et al., 1984). This increase was determined to be an enhancement of the liquid-side mass transfer coefficient  $k_L$  and not the gas–liquid interfacial area  $a$  (Quicker et al., 1984, 1987). This mass transfer enhancement has also been tested and observed in CO<sub>2</sub> absorption into aqueous amines (Park et al., 2008; Jiang et al., 2014; Komati and Suresh, 2008). Herein we report on the ability of a series of solid-particle additives to improve overall mass transfer, as determined in lab-scale experiments. Activated carbon additives showed the greatest enhancement in initial screening, and was subsequently scaled to an absorption-desorption integrated bench-scale carbon capture unit using simulated flue gas to quantify overall possible energy savings.

### 1.1. Mass transfer fundamentals

The overall flux of CO<sub>2</sub> ( $N_{CO_2}$ ) into or out of the solvent is proportional to the CO<sub>2</sub> driving force, the difference between the gas

phase CO<sub>2</sub> partial pressure ( $P_{CO_2}$ ) and the liquid phase CO<sub>2</sub> partial pressure ( $P^*_{CO_2}$ ), as shown in Eq. (1).

$$N_{CO_2} = K_G (P_{CO_2} - P^*_{CO_2(l)}) \quad (1)$$

The overall mass transfer coefficient ( $K_G$ ) for a stable liquid–gas interface layer can be expressed in terms of the gas side ( $k_G$ ) and liquid side resistances ( $k'_G$ ). The liquid-side resistance can then be further expressed in terms of the reaction resistance and physical mass transfer resistance, as shown in Eq. (2):

$$\frac{1}{K_G} = \frac{1}{k_G} + \frac{1}{k'_G} = \frac{1}{k_G} + \frac{H}{\sqrt{Dk_2} [Am]} + \frac{\Delta P^*_{CO_2}}{k_{l,prod} \Delta \alpha [Am]} \quad (2)$$

Where  $D$  is the diffusivity of CO<sub>2</sub>,  $k_2$  is the reaction rate constant,  $[Am]$  is the concentration of amine,  $H$  is Henry's Law constant,  $\Delta P^*_{CO_2}$  is the log mean pressure difference of CO<sub>2</sub> between the liquid and gas sides during absorption process,  $\alpha$  is the C/N molar ratio, and  $k_{l,prod}$  is the physical liquid film mass transfer coefficient. For amine solvents with fast CO<sub>2</sub> absorption kinetics ( $k_2$ ), overall mass transfer is limited by the diffusion of CO<sub>2</sub> and carbon-associated molecules from the liquid film into the bulk liquid ( $k_{l,prod}^\circ$ ) and/or the unreacted amine from bulk liquid to interface liquid film when the film is in the static mode (Dugas and Rochelle, 2011). A reduction in the diffusion resistance will therefore increase the overall mass transfer that can be achieved by adding nano/micro-scale particles to the amine solution.

## 2. Materials and methods

### 2.1. Materials and solvent preparation

Carbon dioxide (>99.9%) and span gas (14 vol.%, CO<sub>2</sub>, in N<sub>2</sub>) were purchased from Purity Plus. Particles were purchased from the vendors as detailed in Table 1. Aqueous amine solvents were prepared from high purity monoethanolamine (MEA), 2-amino-2-methyl-1-propanol (AMP), methyldiethanolamine (MDEA), piperazine (PZ), 1,6-hexanediamine (HDA), and 1-amino-2-propanol (A2P) as summarized in Table 2, mixed with deionized water. To prepare solutions with particle additives, the desired amount of particles were suspended in the aqueous amine solutions and ultrasonicated for 1 min to disperse the particles evenly in solution.

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