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Fluid flow and mass transfer characteristics of enhanced CO_2 capture in a minichannel reactor

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HIGHLIGHTS

- Enhanced CO_2 capture using aqueous DEA in a microreactor.
- Observed flow patterns and calculated interfacial area by high-speed visualization.
- Achieved close to 100% absorption efficiency under certain operating conditions.
- Compared pressure drop and Sherwood number with predictions from model.
- Mass transfer coefficient 2–4 orders of magnitude higher than conventional reactors.

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ABSTRACT

CO_2 absorption using amine solvents can be significantly enhanced by the use of microscale reactors having high surface area to volume ratio. The present paper reports an experimental investigation of the fluid flow and mass transfer characteristics during reactive gas–liquid absorption in a minichannel reactor. Absorption of CO_2 mixed with N_2 into aqueous diethanolamine was studied in a channel having hydraulic diameter of 762 μm and a circular cross-sectional geometry. High-speed imaging of the two-phase flow was conducted to visualize the flow regimes. Image-processing analysis of the acquired flow patterns was performed to determine the interfacial area. The performance of the reactor was studied with respect to the absorption efficiency, pressure drop, mass transfer coefficient, interfacial area, enhancement factor, and Sherwood number. Parametric studies investigating the effects of phase superficial velocity, liquid reactant concentration, and CO_2 concentration in the gas phase were performed and are discussed. High levels of absorption efficiency, close to 100%, were observed under certain operating conditions. An empirical model for the Sherwood number was developed and compared against experimental data. The mass transfer coefficient was found to be higher at reduced channel lengths, which was attributed to improved utilization of the absorption capacity of the amine solution for a given reactor volume. The presently achieved values of mass transfer coefficient and specific interfacial area are between 1 and 4 orders of magnitude higher than those reported for most conventional absorption systems.

1. Introduction

In May 2013, one of the world's oldest observatories in Mauna Loa, Hawaii, involved in measuring atmospheric CO_2 concentration levels in real time since the year 1958 reported a daily average value that crossed a symbolic, but nonetheless significant threshold of 400 ppm for the first time [1]. To reduce these anthropogenic greenhouse gas emissions that may lead to adverse climate change, new technologies are rapidly emerging in the energy industry [2]. Several investigations have shown that cost-effective mitigation of CO_2 emissions from power plants can be accomplished by carbon capture and storage (CCS) [3–9]. CO_2 can be separated from a gas stream by different approaches, including physical/chemical absorption [10–13], adsorption [14–18], membrane separation [19–24], and cryogenic distillation [25–29].

Three major techniques for CCS are post-combustion, pre-combustion, and oxy-combustion (or oxyfuel). The present study focuses on post-combustion CO_2 capture using aqueous amine solvents. The concentration of CO_2 in the post-combustion flue gas stream varies depending upon various factors including the operating conditions and the nature of the fuel being used. Concentrations have been estimated to range between 12% and 14% for coal-fired boilers and integrated gasification combined cycles

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Nomenclature

a	specific interfacial area (m^2/m^3)
b	empirical constants in *Sh* number correlation (–)
Bo	Bond number (–)
c	Chisholm parameter (–)
C_A	concentration of specie A (mol/m^3)
C_A^*	interfacial concentration of specie A (mol/m^3)
d	diffusivity (m^2/s)
D	hydraulic diameter (m)
E	enhancement factor (–)
f	friction factor (–)
G	mass flux (kg/m^2 s)
H	Henry's constant (Pa m^3/mol)
k_{-1}	backward first-order reaction rate constant (1/s)
k_2	forward second-order reaction rate constant (m^3/mol s)
k_B	forward second-order reaction rate constant for base B (m^3/mol s)
k_L	liquid-side mass transfer coefficient (m/s)
k_La	liquid-side volumetric mass transfer coefficient (1/s)
L	length (m)
MAE	mean absolute error (%)
N_A	absorption flux of specie A (mol/m^2 s)
n_A	molar flow rate of specie A (mol/s)
p	partial pressure (Pa)
P	pressure (Pa)
Q	volumetric flow rate (m^3/s)
R	reaction rate (mol/m^3 s)
Re	Reynolds number (–)
Sc	Schmidt number (–)
Sh	Sherwood number (–)
U	velocity (m/s)
V	volume (m^3)
x	gas mass fraction (–)
X	Martinelli parameter (–)

Greek symbols

α	volume fraction (–)
β	volumetric flow ratio (–)
ϕ	two-phase friction multiplier (–)
η_{abs}	absorption efficiency (%)
μ	dynamic viscosity (Pa s)
ρ	density (kg/m^3)
σ	surface tension (N/m)
τ	gas–liquid contact time (s)

Subscripts and superscripts

A	acceleration
F	friction
G	gas
in	channel inlet
L	liquid
m	log-mean
out	channel outlet
TP	two-phase

(IGCC), 11–13% for oil-fired boilers, 3–4% for gas turbines, and 7–10% for natural gas fired boilers [2]. The underlying gas–liquid absorption phenomenon during carbon capture can also be applied to the natural gas sweetening process. The yield from sour gas fields can typically contain as much as 8% CO_2 and 17% H_2S in addition to the major component of natural gas which is methane [30]. Certain ultra-sour fields such as the Shah Gas Field in the United Arab Emirates involve streams containing up to 23% H_2S and 10% CO_2 [31]. Both of these acid gases warrant removal prior to subsequent processing, transport or liquefaction [32].

To improve the efficiency of gas–liquid absorption, microscale technologies are a promising area of research that could yield both environmental and economic benefits. Microscale devices are quickly penetrating new application areas in diverse engineering applications due to their ability to intensify processes, improve process control, increase safety, and reduce the overall size [33,34]. When optimally designed, the inherently higher surface area to volume ratio of these systems substantially enhances heat and mass transfer performance, while keeping the pressure drop at moderate levels [35]. Subsequent discussions in the present study follow the widely accepted classification system defining minichannels as having hydraulic diameters between 3 mm and 200 μm, and microchannels as those having hydraulic diameters between 200 μm and 10 μm.

Yue et al. [36] studied the reactive mass transfer characteristics between pure CO_2 and an aqueous solution of $NaHCO_3/Na_2CO_3$ in a 667 μm minichannel. The experimental two-phase frictional pressure drop was well predicted by the Lockhart–Martinelli approach [37] with a modified expression for the Chisholm parameter, c, that was obtained by regression. The liquid-side mass transfer coefficient and specific interfacial area were estimated and shown to be at least one to two orders of magnitude higher than those observed in conventional gas–liquid absorption systems.

Niu et al. [38] studied the absorption of CO_2 mixed with N_2 into piperazine-activated methyldiethanolamine (MDEA) solution in minichannels (0.5–2 mm). They reported an improvement in the absorption efficiency with decreasing channel diameter. However, the reactor performance was not characterized in terms of mass transfer coefficient.

Shao et al. [39,40] studied the absorption of CO_2 mixed with N_2 into aqueous NaOH in minichannels (345–816 μm). Different channel lengths were studied, which demonstrated that longer channels yield better absorption efficiencies. However, the effect of channel length on pressure drop and mass transfer coefficient was not discussed. Further, their work was limited to studying only the Taylor flow regime. Similar studies focusing on this regime were reported by Kundu et al. [41] and Tan et al. [42,43].

Su et al. [44] studied the absorption of H_2S mixed with N_2 in aqueous MDEA in a minichannel (1 mm). High values of interfacial area and gas-side mass transfer coefficient were achieved, which were reported to be at least 1–2 orders of magnitude higher than those of conventional absorption technologies. Empirical correlations for the Sherwood number and the interfacial area, obtained by regression, were proposed.

Ye et al. [45] tested the absorption of CO_2 mixed with N_2 in methylethanolamine (MEA) in a minichannel (408 μm). The system performance was analyzed while operating at both ambient pressure and an elevated pressure of 3 MPa, and enhanced transport phenomena were reported with the latter case.

Shooshtari et al. [46] studied the absorption of pure CO_2 in aqueous diethanolamine (DEA) in a minichannel (750 μm). Different channel lengths were tested, and the reactor performance was reported in terms of absorption efficiency, pressure drop and acid gas loading ratio. However, their study considered the absorption of pure CO_2 into the amine solution, while most practical applications involve relatively dilute mixtures, wherein the gas to be absorbed is usually present as a small fraction along with a dominant non-absorbable fraction.

While the above-reviewed studies (summarized in Table 1) focus largely on laboratory-scale single channel reactors, scaled-up