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Hydrodynamics and mass transfer performance of a microreactor for enhanced gas separation processes

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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

Chemical absorption processes for gas separation applications can be substantially enhanced using microreactors that utilize micro-structured surfaces/channels and fluid feed systems. The present paper focuses on the absorption of CO_2 using aqueous diethanolamine solvent in microreactors having hydraulic diameters ranging from 254 to 762 μm. 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 were conducted varying the channel hydraulic diameter, liquid solvent concentration, and CO_2 concentration in the gas phase. The two-phase pressure drop was compared against an available empirical model and a reasonably good agreement was obtained for the present range of channel diameters. An empirical model for the Sherwood number was developed and compared against experimental data. Absorption efficiencies close to 100% were observed under certain operating conditions. Liquid-side volumetric mass transfer coefficients close to 400 s^{-1} were achieved, which is between 2 and 4 orders of magnitude higher than those reported for most conventional gas–liquid absorption systems. Such high levels of process intensification were attributed to the increased area per unit volume offered by microchannels, thus increased gas–liquid specific interfacial area at reduced channel diameters. Interfacial areas close to 15,000 m^2/m^3 were achieved which is between 1 and 2 orders of magnitude higher than those of 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]. Increasing anthropogenic greenhouse gas emissions have the potential to cause global warming and consequently leading to adverse climatic changes. This threat has spurred the development of new technologies targeted at combating this issue [2]. Fossil fuel-driven power plants are currently the largest emitters of CO_2. The concentration of CO_2 in the post-combustion flue gas stream varies depending upon several factors, including the operating conditions and the nature of the fuel being used. Typical concentration levels range between 12% and 14% for coal-fired boilers and integrated gasification combined cycles (IGCC), 11–13% for oil-fired boilers, 3–4% for gas turbines, and 7–10% for natural gas fired boilers [2]. It has been widely shown that cost-effective mitigation of CO_2 emissions from these electricity-generating sources can be accomplished by carbon capture and storage (CCS) [3–10].

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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	acceleration due to gravity (m/s^2)
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)
K_c	contraction coefficient (–)
K_e	expansion coefficient (–)
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
exp	experimental
F	friction
G	gas
in	channel inlet
L	liquid
m	log-mean
out	channel outlet
SP	single-phase
th	theoretical
TP	two-phase

The separation of CO_2 from post-combustion flue gas can be conducted by various approaches, including physical/chemical absorption [11–21], adsorption [22–31], membrane separation [32–44], and cryogenic distillation [45–49]. The present study focuses on post-combustion CO_2 capture by chemical absorption using aqueous amine solvents. It is worth noting that a relatively similar process termed gas sweetening has been conducted by the natural gas industry for several decades. While the major component of natural gas is methane, the yield from sour gas fields can typically contain up to 8% CO_2 and 17% H_2S, both of which are corrosive in nature and can adversely affect pipelines and operating equipment [50]. Furthermore, H_2S is highly toxic when present even in small concentrations and can be lethal. Hence these acid/sour gases warrant removal prior to transportation, processing, or liquefaction of natural gas. Furthermore, in recent years there has been an increased interest in the development of certain extreme-sour fields such as the Shah and Bab gas fields that involve streams containing up to 23% H_2S and 10% CO_2, and 33% H_2S and 10% CO_2, respectively [51,52]. Hence, there are numerous economic, safety, and environmental factors that strongly incentivize the development of novel technologies for efficient gas separation processes.

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 [53–56]. 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 [57,58]. Microreactors, if successfully applied to gas–liquid absorption processes, can offer significant reductions in the cost and footprint of carbon capture systems. 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 and 10 μm.

Yue et al. [59] studied the absorption of pure CO_2 into aqueous $NaHCO_3/Na_2CO_3$ in a 667 μm channel and reported results for the two-phase pressure drop, mass transfer coefficient and interfacial area. Likewise, Shooshtari et al. [60] investigated the absorption of pure CO_2 into aqueous diethanolamine (DEA) in a 750 μm channel and presented results for absorption efficiency, pressure drop and reactant utilization efficiency. It is worth noting that while both these studies utilized pure CO_2 as the gaseous phase, in most practical applications, CO_2 is present as a small fraction along with a dominant non-absorbable fraction.

Su et al. [61] investigated the absorption of H_2S mixed with N_2 into aqueous methyldiethanolamine (MDEA) in a 1 mm channel. Ganapathy et al. [62] studied the absorption of CO_2 mixed with N_2 into aqueous DEA in a 762 μm channel. Both works reported various performance parameters including the absorption efficiency, pressure drop, mass transfer coefficient, interfacial area, and Sherwood number. Further, Ganapathy et al. [62] studied the effect of channel length and proposed a two-phase frictional pressure drop model for amine-based working fluids. The effect of system pressure on mass transfer was investigated by Ye et al. [63] for the absorption of CO_2 mixed with N_2 in methylethanolamine (MEA) in a 408 μm minichannel. Atmospheric and elevated pressure levels were tested and the latter was reported to yield better mass transfer performance.

All of the above-reviewed works remarked on the enhanced mass transfer characteristics achieved by using microreactors.