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Carbon dioxide capture by single droplet using Selexol, Rectisol and water as absorbents: A theoretical approach

Wei-Hsin Chen [a,*], Shu-Mi Chen [b], Chen-I Hung [b]

[a] Department of Aeronautics and Astronautics, National Cheng Kung University, Tainan 701, Taiwan, ROC
[b] Department of Mechanical Engineering, National Cheng Kung University, Tainan 701, Taiwan, ROC

HIGHLIGHTS

- A theoretical method for carbon dioxide capture is developed.
- Selexol, Rectisol, and water are taken into account as absorbents.
- The absorption time is more sensitive to the operating temperature.
- Rectisol has the highest capacity to capture CO_2 among the three absorbents.
- The absorption rate of Rectisol is larger than the others by an order of magnitude.

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ABSTRACT

A theoretical method is developed to analyze carbon dioxide capture by a stationary single droplet for evaluating the fundamental mass transfer behavior. In the method, the gas-phase diffusion is predicted using a similarity method and the technique of separation of variable is employed to approach the liquid-phase diffusion. At the interface, a finite difference method is applied to connect the CO_2 diffusion between the two phases. The individual capture processes of CO_2 by three different absorbents of Selexol, Rectisol and water, are taken into account. The operating pressure and temperature of Selexol and water are in the ranges of 30–60 atm and 303–333 K, respectively, and they are 30–60 atm and 240–270 K for Rectisol. The analysis indicates that an increase in temperature decreases the CO_2 capture amount and absorption time by Selexol and Rectisol droplets. The absorption time is more sensitive to the operating temperature than the capture amount. As a result, the CO_2 absorption rates by the droplets are increased when the temperature increases. Among the three absorbents, Rectisol has the highest capacity to capture CO_2 and its absorption time is in a comparable state to the other two absorbents. This results in that its absorption rate is larger than the others by an order of magnitude.

1. Introduction

Burning fossil fuels is the main source of carbon dioxide emissions from anthropogenic activities and it accounts for approximately 95% of the total global annual carbon dioxide emissions [1]. Currently carbon capture and storage (CCS) is considered to be technically feasible at commercial scale to reduce anthropogenic carbon dioxide emissions into the atmosphere [2–6]. A number of carbon dioxide separation methods, such as solution absorption, membrane separation, adsorption separation [7,8], and cryogenics [1,9], can be employed. When one is concerned with solution absorption, it can be classified into chemical absorption and physical absorption. Monoethanolamine (MEA), 2-amino-2-methyl-1-propanol (AMP), aqua ammonia, dual-alkali, and the proprietary solvent marketed by Mitsubishi Heavy Industries, KS-1, have been employed as chemical solvents [10] for a variety of applications in industries. With regard to physical absorption, the gas treating solvents of Selexol (dimethylether polyethylene glycol), Rectisol (chilled methanol), Fluor (propylene carbonate), and Purisol (N-methyl-2-pyrollidone) are becoming increasingly popular, especially in the application of coal gasification [11].

When physical absorption is practiced, CO_2 is absorbed into a absorbent obeying Henry's law, and the gas capture amount by the absorbent depends on the partial pressure of CO_2 and the temperature of absorbent [12]. In general, CO_2 removal using physical absorption allows consuming energy on a reasonable level for its application in large industrial scale. For example, physical absorption may be a suitable process to remove CO_2 from metallurgical fuel gases, such as blast furnace gases and Corex gases [13]. The solubility of CO_2 in an absorbent is higher at the higher partial

* Corresponding author. Tel: +886 6 2757575x63600; fax: +886 6 2389940.
E-mail address: weihsinchen@gmail.com (W.-H. Chen).

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Nomenclature

$\bar{c}$	average molar concentration (M)
D	diffusion coefficient ($m^2\ s^{-1}$)
D_m	mass diffusion number
H	nondimensional Henry's law constant
H'	Henry's law constant ($M\ atm^{-1}$)
m	nondimensional solute absorption amount
p	pressure (atm)
p_c	critical pressure (atm)
R	gas constant (=0.082 atm $M^{-1}\ K^{-1}$)
r	radial coordinate (m)
r_s	aerosol radius (m)
t	time (s)
T	temperature (K)
T_s	temperature at standard condition (=298.15 K)
T_c	critical temperature (K)
T_r	reduced temperature
u	nondimensional concentration
x	nondimensional radial coordinate
X	molar fraction
M	molecular weight ($g\ mol^{-1}$)
V_i	molar volume of CO_2 at its normal boiling temperature ($cm^3\ mol^{-1}$)
ΔH	enthalpy of solution ($atm\ K^{-1}$)

Greek letters

η	viscosity (cP)
θ	polar angle
Σ_v	diffusion volume ($m^3\ mol^{-1}$)
τ	nondimensional time
ϕ	association factor
ψ	azimuthal angle
ω	acentric factor

Subscript

air	air
c	diffusive characteristic time
g	gas phase or continuous phase
i	absorbent i
l	liquid phase or discrete phase
qss	quasi-saturated state
ss	saturated state
s	droplet surface or interface
SL	saturated liquid
t	total pressure
∞	infinity
vp	vapor phase

pressures of the gas and lower ambient temperatures. The required operating conditions of various absorbents are different. For example, the operating pressure and temperature of Selexol are over 30 atm and approximately at 313 K, respectively [14,15]. The operating pressure and temperature of Rectisol are in the ranges of 30–80 atm and 213–263 K, respectively [11,16,17]. These conditions reveal that the operating temperature of Rectisol is much lower than that of Selexol. When CO_2 captured by Rectisol (chilled methanol) and water are compared with each other, the former has a higher removal capability, especially at low temperatures. Specifically, the CO_2 solubility in methanol at a normal temperature is larger than in water by a factor of approximately 5, and 8–15 times when the temperature of methanol is below 273 K [16].

The integrated gasification combined cycle (IGCC) is a crucial system for the application of CCS [18–20] where the synthesis gas (or syngas) and power are generated. If the produced syngas undergoes water gas shift reactions in association with CCS, carbon free fuel can be produced. In this aspect, the research of CO_2 capture via absorption in IGCC systems has been reported in some studies. Strube and Manfrida [21] focused on two different CO_2 capture configurations using Selexol as a physical solvent. In the first configuration CO_2 and H_2S were individually captured, whereas they were simultaneously captured in the other configuration. They found that the efficiency of CO_2 capture was higher in the individual capture system. Chen and Rubin [22] developed a model to analyze the performances and costs of IGCC plants where a GE quench gasifier along with water gas shift reactors in the presence/absence of a Selexol system for CO_2 capture was employed. They pointed out that the CO_2 avoidance cost was lowest when the total CO_2 removal efficiency was approximately 90%, indicating that it was the optimal CO_2 capture efficiency for the designed plant.

In respect of the application of Rectisol, Li and Robin [23] used Rectisol as an absorbent to study CO_2 removal from syngas in a polygeneration system. Two configurations, including a single-stage wash process and a two-stage wash process, were analyzed and simulated in Aspen Plus for the process comparison. They reported that the two configurations could fulfill CO_2 separation, but they were different in the aspect of process power and energy demand. Another application of CO_2 removal using Rectisol is the indirect coal liquefaction (ICL). Zhou et al. [17] analyzed CO_2 removal in an ICL process utilizing three absorbents of dimethyl carbonate (DMC), MEA, and Rectisol. They discovered that Rectisol was the most economic option for capturing CO_2 in the ICL process.

Although many studies have been published on CO_2 capture by physical solvents, such as Selexol and Rectisol, the theoretical analyses of CO_2 capture by these absorbents are still absent so far. In industry, a variety of devices, such as Venturi scrubbers, spray towers, stirred tanks, packed columns, and bubble columns, have been adopted to remove CO_2 from flue gases [24–26]. When one is concerned with spray towers, the basic mass transfer unit is CO_2 uptake by single liquid droplets [27]. To evaluate the capture characteristics of CO_2 in spray towers through physical absorption, the mass transfer behavior of CO_2 by three different absorbents, Selexol, Rectisol, and water, in the form of droplet will be explored theoretically. The absorption processes at various operating temperatures and pressures will be examined. Finally, the difference in the capture phenomena of the three absorbents will be addressed.

2. Mathematical formulation

2.1. Physical description

This study focuses on CO_2 capture by a single droplet and three absorbents of Selexol, Rectisol, and water are considered. A theoretical method is developed and the following assumptions are adopted to simplify the physical problem: (1) the droplet is in a quiescent environment; (2) the droplet and its surrounding are isothermal so that no vaporization or condensation occurs; (3) the fluid properties such as the densities and diffusivities of the gas and liquid phases are constant; (4) phase equilibrium prevails at