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An experimental and theoretical investigation of pure carbon dioxide absorption in aqueous sodium hydroxide in glass millichannels

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ABSTRACT

In this work, chemisorption experiments of pure CO_2 (99% (v/v)) in different concentrations of sodium hydroxide solutions were carried out in a T-junction glass circular milli-channel in the slug flow regime. The absorption of the gas from the slug results in a shrinking of the slug as it moves along the channel. The rate of decrease of slug size and slug velocity are used to quantify the mass transfer rate. A lumped parameter model is developed to study the dissolution dynamics of CO_2 slugs in the aqueous solution. The experimental data in this controlled study and the model developed are used to predict the liquid side volumetric mass transfer coefficient. The enhancement of mass transfer due to chemical reaction in the liquid phase is also quantified. The effect of various parameters like gas and liquid flow rates, concentration of NaOH on system behaviour is examined. An empirical correlation for dimensionless mass transfer coefficient is obtained from the experimental data which can be used to design absorption columns.

1. Introduction

In the context of global warming, CO_2 capture from flue gases and its sequestration is gaining importance [1]. The post-combustion capture by chemical absorption using solvents with high CO_2 affinity is widely employed in industries. CO_2, thus captured is stripped from the solvent and stored in cylinders for further sequestration. This involves storage of concentrated CO_2 in old cold mines, in older oil fields for enhancing oil recovery or storing CO_2 under the ocean. In this present work, the focus is to use a solvent that chemisorbs concentrated CO_2 i.e. absorbs and reacts with CO_2 and yields products for commercial use. In this work NaOH in excess is used as the solvent and the final product obtained is sodium carbonate. It is a strong base, and used as a pH regulator. Being a very good conductor, it can also be used as an electrolyte in electrolysis. It is commercially produced from salt and limestone in Solvay process. In places where there is a localized need the approach proposed here using milli-fluidics can be adopted to generate sodium carbonate at the point of demand.

Gas–liquid contacting is usually done in packed towers to increase the surface area to volume ratio. Design of these units needs an accurate estimate of mass transfer coefficient. In order to determine these mass transfer coefficients, controlled experiments are carried out in micro or milli-channels [2–4]. Absorption in the slug flow regime in a milli channel has been recently studied [5]. Slugs are elongated gas bubbles with diameters almost equal to the channel diameter i.e. they occupy the entire cross-section of the channel [4].

Several researchers have studied the dissolution of gases with a low concentration of CO_2 in various solvents in different flow regimes [6,7,5]. Absorption in millichannels improves the efficiency of CO_2 capture from flue gases (mixture of other gases with CO_2). The dissolution of highly concentrated CO_2 slugs into MEA that is characterised by shrinking of CO_2 slugs has been recently studied [8]. To understand the physical dissolution dynamics, researchers have carried out numerical, CFD and experimental studies on CO_2 absorption [4,9,10]. These studies helped in developing correlations for the bubble velocity and interfacial mass transfer rate for a chain of CO_2 bubbles in water [11]. Using these correlations in the bubble flow and slug flow regimes, the physical dissolution dynamics of CO_2 were studied [12]. Several experimental investigations on physical absorption of methane in water, air in water in the slug flow regime have been carried out in capillaries of varying diameters [13]. These studies show that the channel diameter significantly influences the liquid side mass transfer coefficient. Several experimental studies have focussed on kinetics of CO_2 absorption in various solvents like ethanol [14], water, secondary amines in different types of rectangular micro-channels [15].

Mikaelian et al studied dissolution of a chain of gas bubbles in a non-volatile liquid. They focussed on the shrinking of slugs and suggested that the incomplete dissolution is influenced by physico-chemical properties of the solution and the operating conditions [11,16]. The incomplete dissolution in this case was attributed to the presence of

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Nomenclature

k_L	Liquid side mass transfer coefficient, m/s
R	Gas constant, m^3.Pa/K mol
T	Temperature, K
2r	Height of the gas slug, m
P_{CO_2}	Partial pressure of CO_2 in the slug, N/m^2
V_s	Velocity of the gas slug, m/s
r	Radius of the cap, m
$C_{CO_2}^{aqueous}$	Concentration of CO_2 in the aqueous phase, mol/l
H	Henry's Solubility constant, Dimensionless
H^{cp}	Henry's law constant, $H^{cp} = \frac{C_{CO_2}^{liquid}}{P_{CO_2}}$,mol/ L atm
$C_{CO_2}^{gas}$	Concentration of CO_2 in the gas, mol/L
j_G	Gas phase superficial velocity, m/s
j_L	Liquid phase superficial velocity, m/s
Re	Reynolds number, Dimensionless
Sc	Schmidt number,$Sc_L = \frac{\mu_L}{\rho_L D}$, Dimensionless

other gases [8]. There are several studies on chemical absorption of CO_2 in various solvents [17,13,18]. Most of these focussed on capturing CO_2 present at low concentrations in a mixture of gases. The liquid and overall mass transfer coefficient were determined by analysing the shrinking of slugs. Tan et al investigated the early stages of slug flow in the absorption of a mixture of CO_2-N_2 in NaOH in a rectangular microchannel [19,20]. They determined the mass transfer coefficient analysing the formation stage of slug flows. Ganapathy et al studied the absorption of CO_2 present in low concentrations in DEA in a mini-channel [21].

In the present work, the hydrodynamics and mass transfer of pure CO_2 (representing the gas stream available after sequestration) absorption in NaOH solution, is analysed. Here the mass transfer is rate limiting as the reaction rate in the liquid phase is very fast compared to the mass transfer rate. The shrinking of slugs induces a variation in the slug velocity along the channel length. A parametric study is performed by carrying out experiments for different gas and liquid flow rates and solvent concentrations. The experiments are carried out for a wide range of operating conditions and a lumped parameter model is used to determine the liquid side mass transfer coefficient. The controlled study reported here enables us develop a dimensionless correlations for the mass transfer coefficient in terms of Sherwood and Reynolds numbers.

2. Experimental apparatus and method

Gas, i.e. pure CO_2 99%(v/v) is sent from a cylinder to a moisture trap and then to a Mass Flow controller (MFC, Model-F-201CB-050-AGD-00-V-Bronhorst). Aqueous NaOH solution is prepared by dissolving sodium hydroxide pellets in Milli Q water. This solution is filled in a 60 ml Beckton and Dickinson Plastic syringe. It is pumped to the channel using a Harvard syringe pump (serial number-D-300150). The two phases are supplied to the two symmetric limbs of a T-shaped glass milli channel with a circular cross-section (Fig. 1).This channel has a diameter of 2 mm and the length of each of the inlet limbs is 10 cm. The two phases (CO_2 and NaOH) first come in contact at the junction of the channel. The two phase mixture thus formed, flows along the straight limb of the "T" which acts as the milli channel where gas-liquid contact and absorption occurs.

The experiments are conducted for two different concentrations of NaOH (0.5 M and 1 M). The volumetric flow rate of CO_2 is varied from 1.2 to 10 ml/min and that of NaOH is varied from 0.2 to 8 ml/min. All the experiments are conducted at 27 °C and at atmospheric pressure. The images of the slugs formed during the two phase flow through the milli channel are captured using a high-speed camera (Canon 1200D) at 10,000 frames per second. The two phase mixture leaving the channel flows through a gas-liquid separator. The liquid phase is collected at the bottom of this separator and it is titrated using Warder's method to determine the concentration of the aqueous phase. This allows us to estimate the amount of CO_2 absorbed in the liquid phase. The final product in the liquid phase is characterised using titration against hydrochloric acid. Two base indicators such as Phenolphthalein and Methyl orange, are used to identify the products sodium carbonate and sodium bicarbonate respectively. During the process of titration, the following reactions take place:

$$NaOH + HCl \rightarrow NaCl + H_2O \tag{a}$$

$$Na_2CO_3 + HCl \rightarrow NaHCO_3 + NaCl \tag{b}$$

$$NaHCO_3 + HCl \rightarrow NaCl + H_2O + CO_2 \tag{c}$$

The occurrence of reactions (a) and (b) is indicated by the indicator, phenolphthalein whereas that of reaction (c) is indicated by methyl orange. If the volume of hydrochloric acid consumed during the titration process in the presence of phenolphthalein and methyl orange is represented as V_{ph} and V_{mo} respectively, then the concentration of sodium hydroxide and sodium carbonate is determined from:

$$NaOH\ \% = \frac{[V_{Ph} - (V_{mo} - V_{ph})] \times N_{HCl} \times meq_{NaOH}}{V_{sample}} \times 100\,\%$$

$$Na_2CO_3\ \% = \frac{[2(V_{mo} - V_{ph})] \times N_{HCl} \times meq_{Na_2CO_3}}{V_{sample}} \times 100\,\%$$

These expressions are valid when $V_{mo} > V_{ph}$. Operating conditions were chosen such that the gas slug is dissolved completely in the liquid by the time it reached the exit. This helps in obtaining only the sodium carbonate as the end product. To ensure this, all experiments were conducted with NaOH in excess.

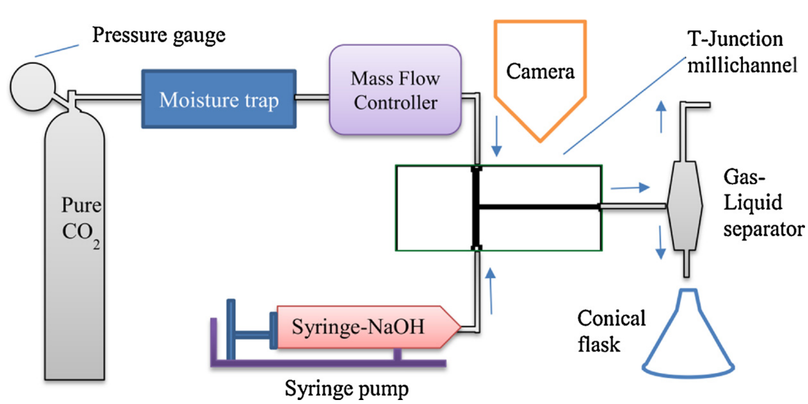

Fig. 1. Schematic diagram of setup used for CO_2 absorption in NaOH in milli channel.