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Ultrasound intensified CO_2 desorption from pressurized loaded monoethanolamine solutions. I. parameters investigation and modelling

Jiru Ying [a, *], Dag A. Eimer [a, b], Frode Brakstad [a], Hans Aksel Haugen [a]

[a] SINTEF Industry, Porsgrunn 3918, Norway
[b] University of South-Eastern Norway, Porsgrunn 3901, Norway

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ABSTRACT

CO_2 stripping from loaded monoethanolamine (MEA) aqueous solutions intensified by means of ultrasound was investigated in a lab-scale kettle reboiler with both gas and liquid continuous operations. The reboiler operating conditions were similar to those of a typical industrial reboiler with a pressure of 1 barg, and where the CO_2 loading is less than 0.25 mol CO_2/mol MEA. Intermittent ultrasound application was tested to find the effects of variables for CO_2 stripping from the CO_2 loadings 0.20–0.39 mol/mol at pressures up to 1.5 barg. Multi-variate data analysis was employed, and a model was built to explain and find the effects of six variables on CO_2 stripping by ultrasound. The six variables include pressure, liquid flow rate, CO_2 loading, intensity, frequency and on-stream time of ultrasound. The variable analysis results manifest that the CO_2 loading is the significant positive effect variable, pressure is negative on energy saving and CO_2 stripping rate and ultrasound parameters have varied effects. Experimental results show that the CO_2 stripping rate assisted by ultrasound is 4 times than by heat only when CO_2 loading is high, and the best result of specific energy consumption was 2.3 MJ/kg CO_2 in the present test conditions.

1. Introduction

CO_2 capture and utilization/storage (CCS or CCUS) attract more and more attentions due to the problem of global warming. At present, the most mature technology for CO_2 capture is an MEA-based capture technology, which has a high cost, partly due to the energy demand related to the desorption process.

Ultrasound enhanced CO_2 stripping could be one method to unlocking CCS cost savings. Ultrasonic waves have frequencies above 20 kHz making them inaudible. Ultrasound is a mature technology and is widely used in various fields. With respect to the frequency, ultrasound is divided into three categories: power ultrasound (20–100 kHz), which is used in chemically important systems; high frequency ultrasound (100 kHz–1 MHz), which is used for animal navigation and communication, detection of cracks or flaws in solids, and under water echo location; and diagnostic ultrasound (1–500 MHz), used as a diagnostic in the medical field [1–3]. In sonochemistry, ultrasound with a frequency ranging from 20 to 100 kHz is used to increase the reactivity through cavitation [2–4]. Cavitation bubbles in the liquid may grow larger than 100 μm through a series of compression and expansion cycles caused by the acoustic waves. Cavitation bubbles can be unstable and implode, producing high-speed liquid micro-jets, localized intense heating and high-pressure shock waves. The expelling liquid jet can move up to speeds of 400 km/h, the hot spots in the cavity can reach thousands of degrees Celsius and the shock waves have been shown sufficient to cleave polymers by mechanical breakage of the chains [5]. Thereby, the effects by cavitation can produce sufficient intensity to enhance chemical reactions and associated mass transfer. Ultrasound is also employed for degassing: removing air from distilled water in laboratory and or hydrogen from aluminum alloy process [6].

In recent years, ultrasound started to be introduced in gas purification: for stripping acid gas from loaded solutions, for absorbent regeneration and reducing degradation of absorbents. The major mechanisms for improving the mass transfer are the millions of cavitation bubbles that increase the interfacial area between gas and liquid. The micro-liquid jet and vortex produced by ultrasound

* Corresponding author.
E-mail address: jiru.ying@sintef.no (J. Ying).

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can increase mass transport as well. For degassing purposes, the frequency of ultrasound is often relatively low (e.g. 20–60 kHz) to avoid degradation of the absorbents. Gantert et al. [7] introduced ultrasound to degas CO_2 from amine solutions between 60 and 80 °C under ambient pressure. They found that CO_2 can be degassed at temperatures lower than 80 °C using ultrasound and appropriate absorbents, and there is no significant difference between desorption at 37.5 kHz and 25 kHz. Xue et al. [8] studied the desorption of sulfur dioxide from citrate solution using ultrasound with frequencies of 20, 40 and 60 kHz. They found that the use of ultrasound can improve the desorption efficiency of sulfur dioxide. Zhang et al. [9] reported a positive influence of ultrasound on the CO_2 desorption rate from a thermomorphic biphasic solvent (TBS). The frequency was fixed at 37 kHz in their investigation. Tanaka et al. [10] investigated the use of ultrasound to remove CO_2 from monoethanolamine (MEA) aqueous solutions in an ultrasound bath with a frequency of 28 kHz at 25 °C. In the above works [7–10], researchers attempted to enhance CO_2 desorption at lower temperature and tried to look for chemical effects introduced by ultrasound. In our previous work [11], we introduced ultrasound to strip CO_2 from amine solutions with high CO_2 loadings at various temperatures under ambient pressure, and found that most ultrasound energy was used for heating but not for stripping when the desorption temperature was low, because ultrasound did not decompose the carbamate by taking on a catalyst role. We also found that the fractional improvement in the CO_2 desorption rate by using ultrasound at lower temperature, is much slower than at high temperature, resulting in a higher energy consumption per unit of CO_2 desorbed at low temperature. Stripping CO_2 by means of ultrasound at low temperature does not seem to be feasible for industrial applications.

However, all these studies were performed at ambient pressure. In a typical MEA-based CO_2 capture plant, the pressure in the reboiler is about 1.0 barg. Ultrasound and its effect on desorption of CO_2 at pressurized conditions has not previously been researched in depth, and a full theoretical model is not available. Previously [12], preliminary work has been done to investigate the influence of pressure on CO_2 stripping at a pressure range 0–1.5 barg with two different pressure control methods. In this work, the ultrasound intensification effects of using ultrasound intermittently were investigated at varying pressures up to 1.5 barg at the boiling point of lean MEA aqueous solutions with CO_2 loadings from 0.20 to 0.39 mol/mol. The conditions were chosen to be representative of an industrial process. Steam heating was employed in the reboiler to simulate industrial conditions. The parameters studied were pressure/temperature, liquid flow rate, intensity of ultrasound, frequency of ultrasound, percentage on-stream time of ultrasound, and CO_2 loading. When in the reboiler, temperature and pressure are thermodynamically connected, and it was elected to use the temperature as the variable when analyzing the data produced.

2. Theoretical background

The interfacial area between gas and liquid, here manifested by the amount and size of bubbles, plays a key role in any process that involves mass transfer between gas and liquid. Under desorption conditions bubbles are particularly important in that they provide a mass transfer surface for gas that is still physically dissolved in the liquid but lacks the additional energy to form a bubble. This is because the inwards acting surface force and surrounding pressure are very large for a very small bubble [2,13], and an incipient bubble is liable to collapse unless it increases to a critical size. In equilibrium, the pressure balance of a bubble and the surrounding liquid is:

$$P_{liq} + P_s = P_{CO2} + P_{vap} \quad (1)$$

here, P_{liq} is the liquid pressure at the depth of the site, P_s is the pressure caused by surface tension, P_{CO2} is the partial pressure of CO_2 in the bubble and P_{vap} is the solvent vapor pressure in the bubble.

When P_{liq} decreases, the volume of the bubble will increase because the pressure inside the bubble is higher than on the outside. On expansion, both P_{CO2} and P_{vap} inside the bubbles become lower, and the driving force of CO_2 diffusing into the bubble becomes higher. Similarly, when the pressure increases continuously bubbles will reduce in size and could finally collapse.

A larger bubble is easier to inflate than a smaller one as evident from equation (2). The additional pressure due to the pressure caused by surface tension is inversely proportional to bubble size. For a spherical bubble with radius R, this Laplace pressure is given by [2,13].

$$P_s = \frac{2\sigma}{R} \quad (2)$$

where σ is the surface tension of the liquid.

Once a bubble begins to grow, the pressure due to the surface tension reduces, so the bubble expands rapidly. Ultrasound produces many cavitation bubbles large enough to grow compared to the very small sized heterogeneous/homogeneous bubbles formed when applying heat only.

Ultrasound enhances the mass transfer of gas from liquid phase to gas phase by providing cavities that can accommodate gas molecules such that more gas can be desorbed without being retained in a saturated, or even supersaturated, liquid. According to Schueller & Yang [14], ultrasound makes bubbles form more easily, and the activation energy for surface diffusion decreases. This results in a lower energy consumption for CO_2 stripping. Ultrasound also produces micro-streams and vortices that can intensify the mass transfer in the liquid.

The chemical reaction of CO_2 desorbed from MEA aqueous solution by heating can be described by the following chemical equations [15,16].

Carbamate converts to carbon dioxide:

$$RNHCOO^- + RNH_3^+ \leftrightarrow 2RNH_2 + CO_2(aq.) \quad (3)$$

Ionization of water:

$$2H_2O \leftrightarrow H_3O^+ + OH^- \quad (4)$$

Bicarbonate reversion to carbon dioxide:

$$HCO_3^- + H_3O^+ \leftrightarrow 2H_2O + CO_2(aq.) \quad (5)$$

Furthermore, to the extent that carbonate (CO_3^{2-}) is present, it will react with H_2O according to equation (6) to form bicarbonate (HCO_3^-). Carbamate may also be converted to bicarbonate (hydrolysis reaction) through equation (7):

$$CO_3^{2-} + H_3O^+ \leftrightarrow HCO_3^- + H_2O \quad (6)$$

$$RNHCOO^- + H_2O \leftrightarrow RNH_2 + HCO_3^- \quad (7)$$

The CO_2 produced from the chemical reactions will accumulate in the solution if it is not transferred to the gas phase according to: