Experimental study of carbon dioxide solubility in aqueous solutions of citric acid at CO₂ low partial pressures

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In this work, citric acid as a new absorbent for carbon dioxide was investigated. The chemical solubility of CO_2 expressed as loading capacity (mol CO_2 /mol citric acid) was measured experimentally in an equilibrium cell in aqueous 0.1, 1, 2.5 and 3M citric acid solutions over the partial pressure of carbon dioxide ranging from 10 to 50kPa at temperatures 298.15 and 303.15K. The results of gas solubility are presented as loading capacity versus corresponding partial pressure of CO_2 and time for all experimental runs. It was observed that CO_2 solubility increases with increase in concentration of citric acid solution and with decrease in temperature. It also increases with an increase in partial pressure of CO_2 for the pressure range studied.

1. Introduction

Removal of carbon dioxide (CO₂) from gas streams is an important operation in many industrial processes, for a number of technical and economical reasons. Carbon dioxide present in natural gas reduces the heating value of the gas and can cause corrosion in pipes and process equipment. In the ammonia synthesis process, CO₂ would poison the catalyst, whereas in LNG (liquefied natural gas) plants, for prevent freezing in the lowtemperature chillers, CO₂ should be removed (Derks et al. 2005). Finally, CO₂ is regarded as a major greenhouse gas which has the greatest adverse impact on the greenhouse effect causing approximately 55% of the observed global warming (Yeh and Bai, 1999).

There are several technologies available for capturing CO_2 from gaseous mixtures. These include chemical and physical absorption, physical adsorption, cryogenic separation, membrane separation and biological fixation. Among these techniques, chemical absorption by alkanolamines is recognized as an effective, reliable and relatively low cost method for removing CO_2 from processes gas. Alkanolamines more often uses for CO_2 removal are monoethanolamine (MEA), diethanolamine (DEA), diisopropanolamine (DIPA), N-methyldiethanolamine (MDEA), and 2-amino-2-methyl-1-propanol (AMP). Among them, aqueous MEA solution is the most suitable chemical solvent which has been widely used as industrially absorbent because of high absorption rate, low solvent cost, high resistance to thermal degradation, low molecular weight and low solubility for hydrocarbons (Lee et al. 2008). However, it has some disadvantages

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include low loading capacity, high energy requirements for regeneration, degradation through oxidation of the amine, vaporization losses because of high vapour pressure, high viscosity, and also it can causes operational problems such as corrosion, foaming and fouling of the process equipment (Ma'mun et al. 2007). To avoid such problems, various absorbents have been widely investigated.

This work focuses on selecting new absorbent for CO_2 capture from gas streams. After pre-screening, we have selected citric acid as a new chemical absorbent because of low vapour pressure, low viscosity, commercially available and relatively low cost. The solubility of CO_2 in aqueous citric acid solutions measured at atmospheric pressure in different concentrations over the partial pressure of CO_2 from 10 to 50 kPa at temperatures 298.15 and 303.15 K.

2. Experimental

1.1 2.1 Material

MEA (purity > 99.5 %) and citric acid (purity > 99.5 %) were obtained from Merck chemical company. carbon dioxide with high purity (> 99.95 %) were supplied from commercial cylinders.

1.2 2.2 Apparatus and Procedure

The apparatus used to measure the solubility of CO_2 is a modification of that reported by Rinaldi (1997). The batch reactor (round-bottomed flask) with an internal volume 2000ml was made of Pyrex. The temperature is controlled by means of thermostatic water bath with temperature precision $\pm 0.1^{\circ}$ C. In a typical experiment, known volume of absorbing solution was introduced into the reactor. After that, a certain amount of CO_2 by using a syringe gas with constant rate (from SGE Co.) injected into the vessel. The solution was continuously stirred with the magnetic stirrer to increase the mass transfer rate and achieve equilibrium quickly. Then, the contents of the reactor were allowed to reach equilibrium at the desired temperature. As the carbon dioxide is absorbed, total pressure decreases. During the experiment, pressure of the system was measured by a pressure transmitter sensor (from GEFRAN) with an accuracy ± 0.15 % of the gauge range. The measuring data from pressure sensor are sent to computer via a data logger and record by Lab VIEW software. When the total pressure of the cell does not change (or remained constant), the equilibrium was assumed to be reached. The partial pressure of CO_2 was calculated from the difference between initial and final pressure in the absorbent vessel and the CO₂ loading of solution was determined by ideal gas law.

3. Result and discussion

The experimental setup and procedure were tested by measuring the solubility of CO_2 in an aqueous 2.5M of MEA solution at 298.15K (Muhlbauer and Monaghan, 1957 and Lee et al. 1976), as shown in Figure 1. We are unaware of any published data on CO_2 solubility in citric acid aqueous solution which to compare our results.



Figure 1: Solubility of CO₂ in aqueous 2.5 M MEA solution at 298.15 K

The experimental data of the loading capacity against corresponding CO_2 partial pressure in aqueous 0.1, 1, 2.5 and 3M citric acid solutions at temperatures 298.15 and 303.15 K are shown graphically in Figures 2 and 3. It was observed that loading capacity increases with an increase in applied pressure of CO_2 and decreases with an increase in temperature. As CO_2 absorption is exothermic in nature and an increase in temperature should decrease the extent of chemical absorption in accordance with Le Chatelier's principle. Also, it is clear that loading capacity increases with increase in concentration of citric acid solution.



Figure 2: Solubility of CO₂ in aqueous citric acid solutions at 298.15 K



Figure 3: Solubility of CO₂ in aqueous citric acid solutions at temperature 303.15 K

The loading capacities of citric acid solutions versus time in different partial pressures of CO_2 at 298.15 K are illustrated in Figures 4a-d. It was clear that, it increases with time.



Figure 4a: Loading capacity versus time for aqueous 3M citric acid solution at temperature 298.15 K



Figure 4b: Loading capacity versus time for aqueous 2.5M citric acid solution at temperature 298.15 K



Figure 4c: Loading capacity versus time for aqueous 1M citric acid solution at temperature 298.15 K



Figure 4d:Lloading capacity versus time for aqueous 0.1M citric acid solution at temperature 298.15 K

4. Conclusions

In this article, after investigation on different chemical solvent, citric acid selected as a new absorbent for CO_2 removal because of low vapour pressure, low viscosity, commercially available and relatively low cost. The solubility of CO_2 in aqueous solutions of citric acid was measured in different concentrations at temperatures 298.15 and 303.15 K. The experimental result showed that citric acid seems to be a potentially good absorbent for capturing CO_2 according to the above criteria.

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