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Results from a pilot plant using un-promoted potassium carbonate for carbon capture

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Abstract

A pilot plant facility has been designed and built to trial potassium carbonate solvent technology for carbon capture under a range of conditions. The rig is capable of capturing 4 - 10 kg/hr of CO_2 from 30 - 55 kg/hr of an air-CO_2 mixture, with different packings. A series of trials have been completed with a range of solvent concentrations from 20 wt% to 30 wt% potassium carbonate. The experimental holdup, solvent loading and absorber temperatures have been matched with rate-based simulations in Aspen Plus® software.

Keywords: solvent; potassium carbonate; simulation

1. Introduction

The removal and sequestration of carbon dioxide (CO_2) from post combustion flue gas streams by absorption processes is being actively investigated as a technology to help mitigate global warming due to human activities. The process of using potassium carbonate (K_2CO_3) for CO_2 removal from high pressure gases such as synthesis gas has been known for many years [1, 2] and follows the overall reaction (shown in ionic form):

$$CO_2 + CO_3^{2-} + H_2O \rightarrow 2HCO_3^- \tag{1}$$

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In recent years this use of potassium carbonate has reduced and aqueous alkanolamines such as monoethanolamine (MEA) or diethanolamine (DEA) have replaced it.

When compared to potassium carbonate for the application to post combustion flue gas streams, alkanolamines have significant limitations including: high levels of corrosion; high degradation rates due to high oxygen concentration; formation of heat stable salts due to side reactions with flue gas impurities; and high solvent losses through high vapor pressures. However, the traditional potassium carbonate system has limitations also, primarily a slow rate of reaction and high solvent circulation rates which lead to a prohibitively large solvent regeneration energy requirement.

The CO2CRC has patented a precipitating potassium carbonate process (termed UNO MK 3), which uses a highly concentrated solvent to improve reaction kinetics and reduce energy usage [3].

In order to design and implement efficient and cost effective carbon dioxide capture facilities based on precipitating solvent systems, such as the UNO MK 3 process, two key areas need to be addressed. They are:

- The development and / or validation of appropriate solvent handling equipment for bicarbonate slurries.
- The ability to adequately simulate system performance.

To test the performance of solvent handling equipment and models to predict the capture of carbon dioxide with a precipitating potassium carbonate solvent, a pilot scale experimental rig has been built at the University of Melbourne laboratories and in the first instance the performance evaluated using synthetic gas and aqueous potassium carbonate solvent.

2. Description of the Pilot Plant

A pilot plant facility to test a precipitating carbonate system was completed and commissioned in early 2012 at the Parkville campus of the University of Melbourne, Australia. The plant was designed to test the hydraulic aspects of carbon capture with a precipitating solvent, as well as to validate simulation models developed in Aspen Plus®. The plant consists of an absorber and stripper operating in counter-current mode. The stainless steel reboiler is attached directly to the base of the stripper and is heated by a low-density electrical two-stage heater made of Incoloy 800. Unlike in traditional amine plants, a water wash section was not included in the design as potassium carbonate is non-volatile. Figure 1 shows a process flow diagram for the plant.

The plant was initially trialed with solvent concentrations ranging from 20 to 30 wt%, sufficiently low such that precipitation was not observed. These tests enabled validation of the Aspen Plus® simulation results for absorber temperature profile, solvent loadings, and exit gas CO_2 concentration.

In a typical experimental run, compressed filtered air is mixed with pure CO_2 using Bronkhorst EL-FLOW mass flow controllers in parallel. The gas mixture is passed at 30 kg/hr through a heated water bath and enters the bottom of the absorber via a stainless steel spray gas distributor, which is centered in the absorber 0.2 m above the solvent exit. The absorber is made of borosilicate glass and is 4.25 m high and 0.1 m in diameter, with PTFE gaskets. It consists of three packed sections, each of 0.8 m in height. The packing utilised for this set of experiments is SS 304 Pall rings of 0.01 m diameter. The lean solvent from the regenerator column flows through a spiral heat exchanger and is pumped to the top of the absorber via a plate lean solvent cooler. The solvent/gas ratios were varied from 2 - 6 on a mass basis. When saturated

gas was used, hot water was sprayed into the gas line via a perforated stainless steel tube. The water flow rate was adjusted between 50 - 200 mL/min using a rotameter and then heated via a water bath at 80 °C. A humidity meter located near the gas entry into the absorber was used to record the gas temperature and humidity. The gas exiting the absorber was passed through a glass heat exchanger and a drum to drop out the majority of moisture in the gas. The gas then flows through a rotameter before going to exhaust. The exhaust line is sampled by a Horiba VA-3000 gas analyser via a sampling unit that cools the gas to 5 °C to drop out moisture before it contacts the IR cell. Horiba readings were taken every 15 minutes and solvent samples were taken every 30 minutes at the absorber inlet, outlet and between packed sections. The absorber temperatures were recorded every 10 s into an Excel file, using LabView software and Pt100 RTDs. The absorber top and bottom pressures were also recorded every 10 s from GE PTX1400 pressure transducers.

The solvent samples were titrated using a Metrohm 905 Titrando auto-titrator using 0.4 M H_2SO_4. Exactly 2 mL of the sample was pipetted into a beaker using an Eppendorf 5000 pipette, and diluted using 60 mL of RO water. This sample was then titrated to determine the two end points at pK_a values of 10.33 and 6.37, corresponding to the carbonate and bicarbonate species respectively. A fixed gas flow rate was chosen to satisfy a range of L/G ratios and to maximize the pressure drop through the column.
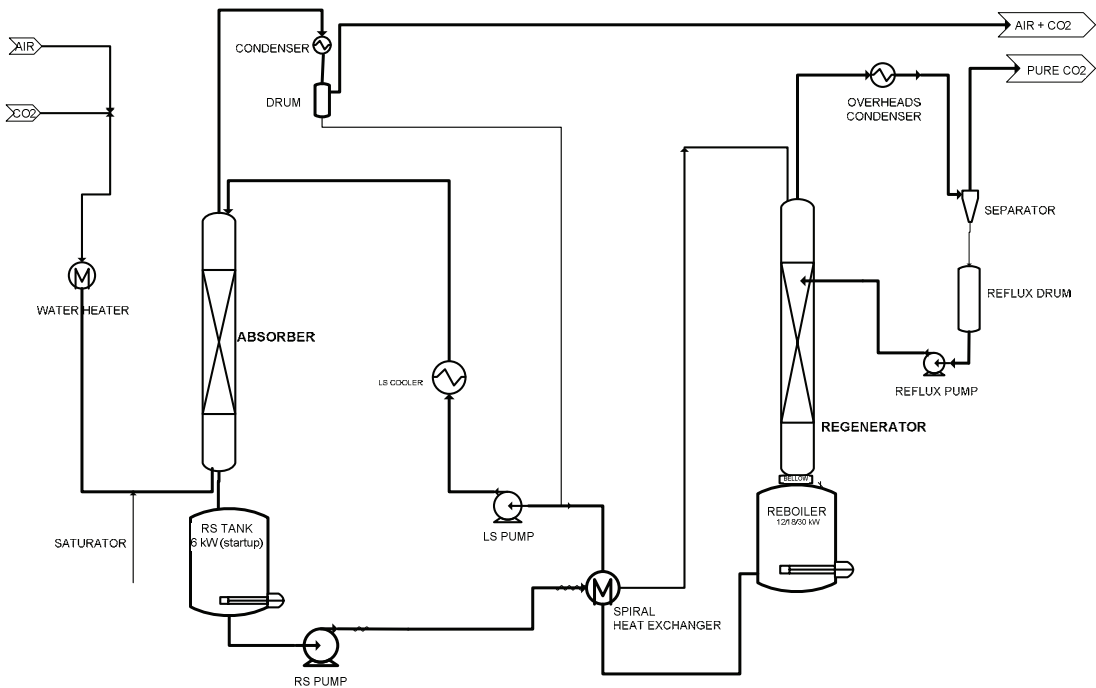

Figure 1: Process flow diagram for pilot plant

3. Description of ASPEN^{TM} simulation model

In this work a non-equilibrium rate-based model for CO_2 absorption into aqueous K_2CO_3 was developed using Aspen Plus$^{®}$ RateSep^{TM} to interpret the results from the pilot plant. The absorber model incorporates the VLE model, rate constants and physical properties correlations developed by the University of Melbourne [3].

4. Results and Discussion

4.1 Pressure drop

A comparison of theoretical and experimental pressure drops across the absorption column for varying air flow rates at a fixed water flow rate is presented in Figure 2. The theoretical pressure drops were estimated by interpolating between lines of constant pressure drop in the generalized pressure drop correlation (GPDC) developed by the Norton Company [4]. This figure shows that the theoretical prediction is lower than measured values at lower air flow rates, and the trend is reversed at higher air flow rates. This variability may be due to the limitations of the GPDC correlation for a non-industrial sized packing, as described by Kister *et. al.* [5]. The pressure drop for the rate-based model developed in this work was estimated using the correlation proposed by Stichlmair *et al.* [6]. The Stichlmair constants corresponding to 10 mm Pall ring packings were adjusted to match the measured pressure drop across the column.
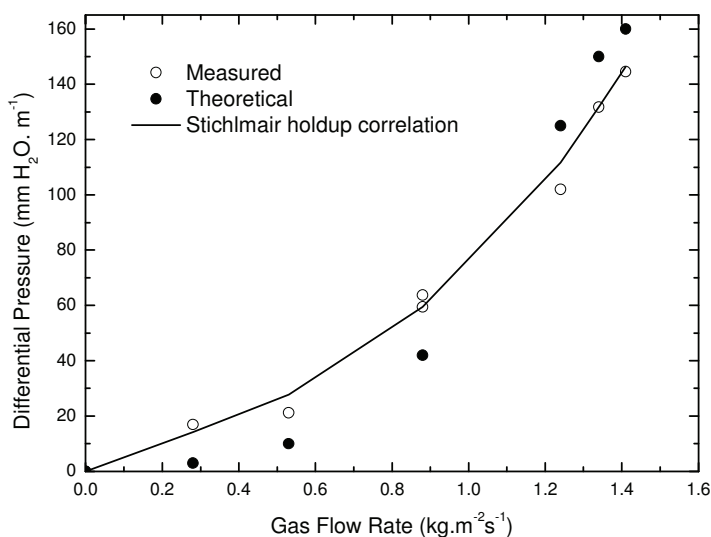

Figure 2: Comparison of experimental and theoretical pressure drops

4.2 Dynamic Holdup

The dynamic holdup (m^3/m^3) in the absorber column as a function of solvent flow rate, for different potassium carbonate concentrations is presented in Figure 3. The dynamic holdup was measured by collecting the volume of liquid that drained from the packing after simultaneously stopping solvent and gas flows. Prior experiments confirmed that the liquid holdup remained constant at constant liquid flow rate and varying gas flow rate, confirming that the column was operating below the loading point. The figure shows that the dynamic holdup increases linearly with liquid flow rate, for each set of experiments.
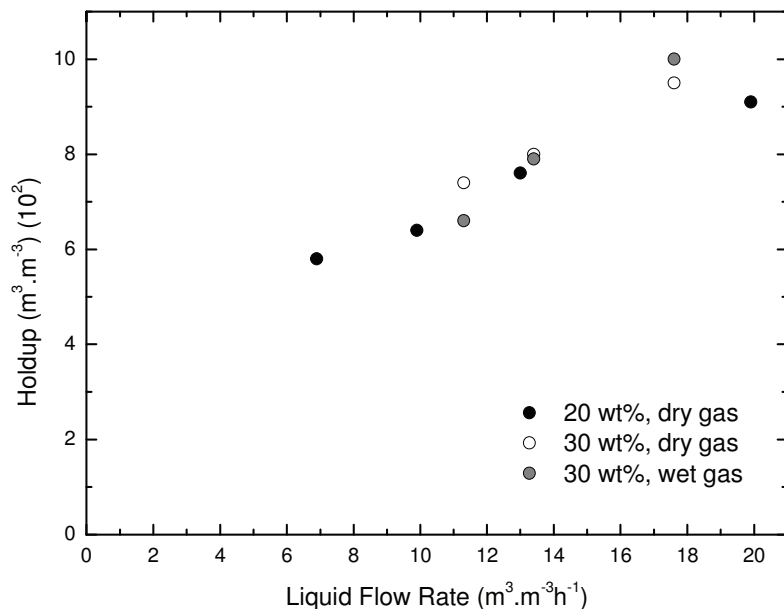

Figure 3: Dynamic liquid holdup as a function of solvent flow for 20 wt% and 30 wt% solutions

4.3 Simulation Results

A comparison of the results from the simulation model and pilot plant experimental data is presented in Table 1. The developed rate-based model was found to match the temperature and concentration profiles in terms of CO_2 volume % and rich solvent loading (mol_{CO2}/mol_{K2CO3}) of the pilot plant data well.

Table 1 Comparison of experimental results and the outputs from the rate-based model

Experiment number	K$_2$CO$_3$ (wt%)	L/G	CO$_2$ output (vol %)		ΔT (lean-rich)		Rich solvent loading (mol$_{CO2}$/mol$_{K2CO3}$)		Inlet gas conc (vol%)	Inlet solvent temp (°C)	Lean solvent loading
			Exp	Rate-based model	Exp	Rate-based model	Exp	Rate-based model			
20W3005	20	4	22.27	23.76	10.4	9.59	0.19	0.17	25	50	0.08
20W0506	20	6	21.48	23.91	8	10.32	0.18	0.17	25	50	0.09
20W0706	20	4	22.29	23.74	17.4	12.95	0.27	0.23	25	54.5	0.13
30W1406	30	4	8.86	9.35	19.9	15.9	0.10	0.11	10	57	0.08
30W1506	30	4	22.24	23.98	14.5	11.10	0.16	0.15	25	51.9	0.10
30W1906	30	4	21.82	23.98	17.5	13.10	0.20	0.19	25	54.8	0.14
30W0407	30	4	21.99	23.76	13.9	14.26	0.18	0.2	25	56.6	0.12
30W0907	30	4	22.03	23.94	13.6	12.78	0.16	0.16	25	54.5	0.11
30W1007	30	4	21.93	23.86	13.9	13.08	0.18	0.17	25	54.9	0.11
30W1707	30	4	22.04	23.81	9.8	8.63	0.22	0.2	25	54.8	0.13
30W2407	30	4	24.12	23.91	11	10.46	0.20	0.18	25	55	0.12
30W2707	30	4	9.44	9.52	12.3	11.59	0.15	0.14	10	54.7	0.12

5. Conclusions

A pilot scale rig for carbon dioxide capture from air has been trialed with aqueous potassium carbonate solvent at concentrations ranging from 20 wt% to 30 wt%. It was found that the measured air-water pressure drop for the Pall ring packing follows the trend predicted by the generalized pressure drop correlation. The operating hold-up did not vary significantly with gas water content, and varied linearly with liquid loading.

The results of the pilot scale rig have enabled verification of AspenPlus® simulation models developed for the process. These simulations will be a valuable tool to predict performance in future trials with precipitating potassium carbonate solvent.

Acknowledgements

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