Investigation of a lab-scale physicochemical CO_2 capture system

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RHODES2024
11th International Conference on Sustainable Solid Waste Management

Introduction

Human activities have led to a 400% increase in global CO_2 emissions since 1950, with the 2015 Paris Agreement aiming to limit Earth's temperature rise to below 1.5°C. To achieve this, hundreds of tons of CO_2 must be captured and stored annually until 2030, using techniques like adsorption, absorption, cryogenic process, and membrane separation. This study aims to investigate chemical absorption as a carbon dioxide capture method in a modified lab-scale scrubber system by testing different absorbent solutions, gas inlet flows and solution volumes.

Methodology

The **mechanism** of CO_2 capture in aqueous media includes the dissolution of the CO_2 gas molecules in water (equation 1) according to Henry's equilibrium, the reversible conversion by deprotonation of the neutral $CO_{2(aq)}$ species (hydration) to form anionic bicarbonate species HCO_3^- (equation 2) according to a chemical equilibrium which is pH dependent, and the transport of both the neutral and anionic aqueous CO_2 species, from the CO_2 capture side towards the CO_2 release side, by molecular diffusion inside the aqueous medium and/or by forced fluid circulation and the reverse process.

$$CO_2(g) + H_2O \leftrightarrow CO_2(aq) \quad (1)$$

$$CO_2(aq) + H_2O \leftrightarrow H^+ + HCO_3^- \quad (2)$$

The equilibrium constant of equation 2 is $K_a=10^{-6.35}$ at 25ºC. Therefore, if the pH can be maintained above the pK_a value of 6.35 using a buffer, the formation of ionic HCO_3^- species is favoured, while the concentration of neutral $CO_{2(aq)}$ species remains fixed at the gas-liquid interface according to Henry's law. The solubility of HCO_3^- anions in water is much higher than that of the neutral $CO_{2(aq)}$ species so a larger total CO_2 concentration can be dissolved in an aqueous solution.

The **experimental apparatus** of the modified lab-scale scrubber includes inlet and outlet flowmeters for the measurement and control of the gas flow. Moreover, a pump is used in order to achieve recirculation of the solution and an outlet pipe is used for liquid sampling after the gas dissolution. The gas input is supplied from a commercial cylinder of a mixture of 80% atmospheric air (O_2, N_2) and 20% CO_2.

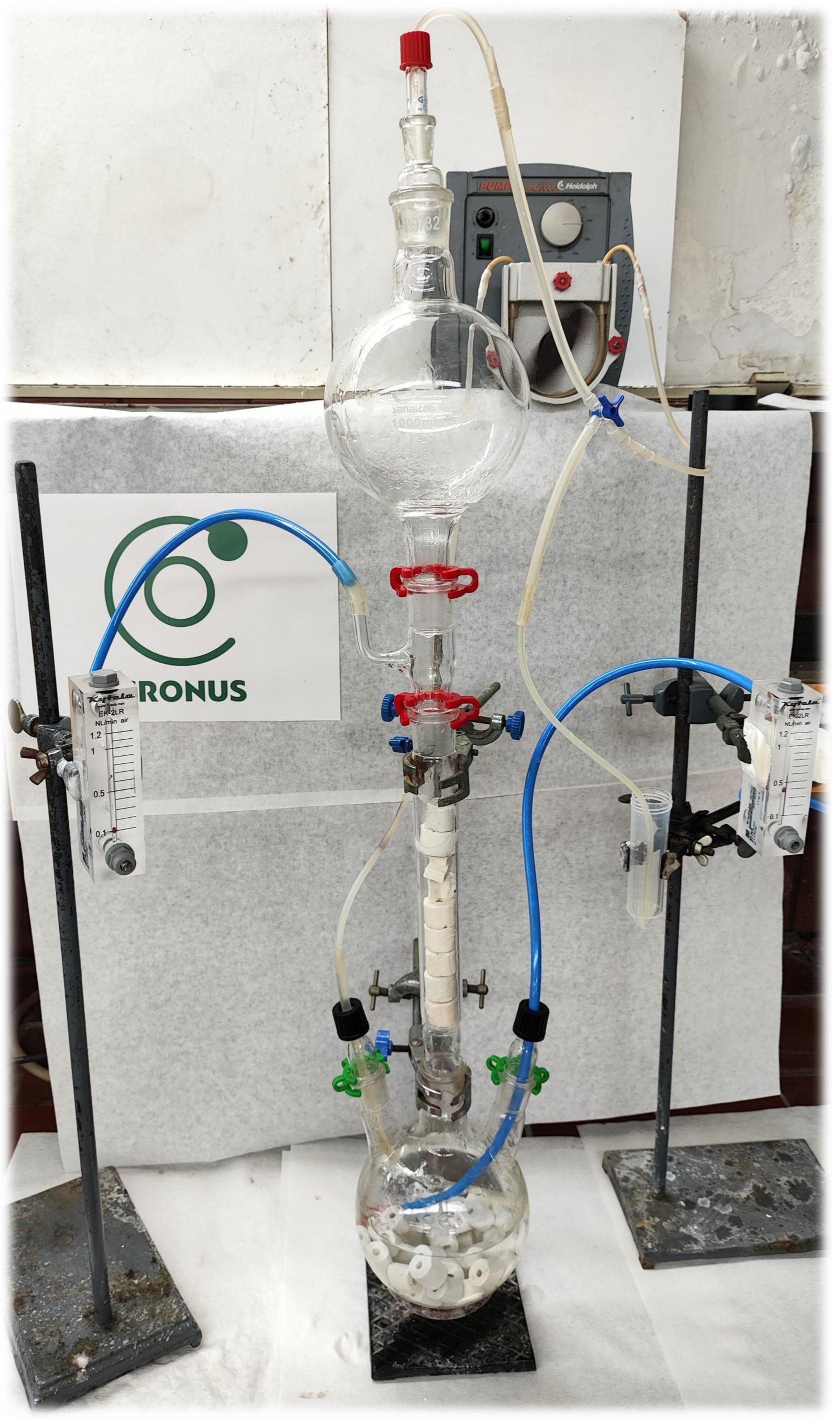


Figure 1: Set-up of the CO_2 capture system

Samples are taken at regular time intervals and the pH, dissolved oxygen, inorganic carbon (IC), organic carbon, and total nitrogen concentrations are measured in each sample until carbon concentration reaches a plateau.

Table 1 sums up the **parameters** investigated and the CO_2 capture percentage is determined based on the IC measurements in the liquid and the inlet CO_2 flow in the scrubbing system as explained (equation 3).

$$CO_2\ capture\ efficiency\ (\%) = \frac{total\ CO_2\ dissolved}{total\ CO_2\ input} 100\% \quad (3)$$

Table 1: Parameters investigated in the lab-scale scrubber

Absorbent solution	pH	Volume (mL)	Inlet Gas flow (L/min)
Na_2HPO_4 / NaH_2PO_4 (Henderson-Hasselbalch buffer)	7	600	0.1
NH_3 / NH_4Cl (Henderson-Hasselbalch buffer)	8.2		
	9		
	10		
Na_2HPO_4 / NaOH (Henderson-Hasselbalch buffer)	11		
	12		
NH_3 / NH_4Cl (Henderson-Hasselbalch buffer)	10	400	
		700	
		800	0.3
			0.5
			1.0
Na_2CO_3 (Carbonate)	11.7	700	0.1

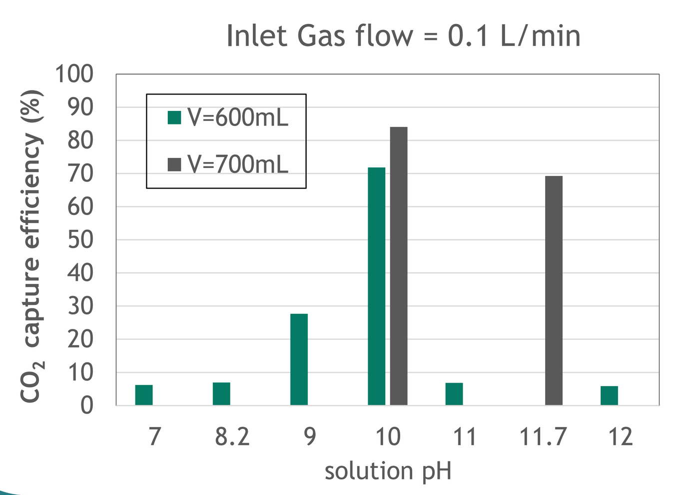
Gas Retention Time (GRT), CO_2 loading, and liquid-to-gas (L/G) ratio are important operating parameters that affect the scrubbing process. GRT and CO_2 loading depend on the inlet gas flow as explained in Table 2.

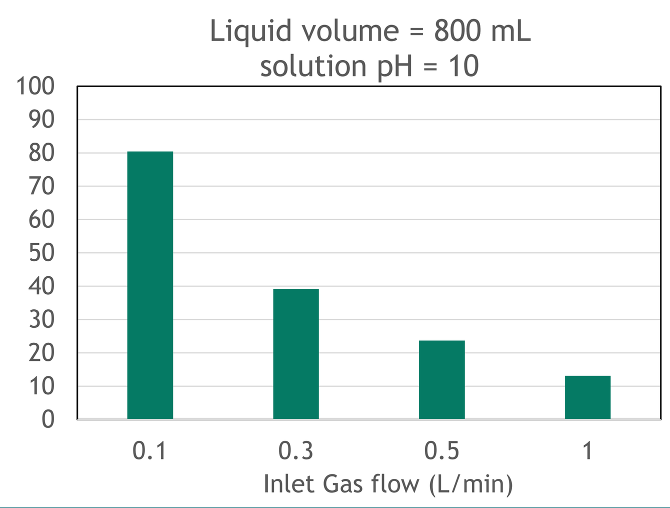
Table 2: GRT and CO_2 loading values based on inlet gas flow

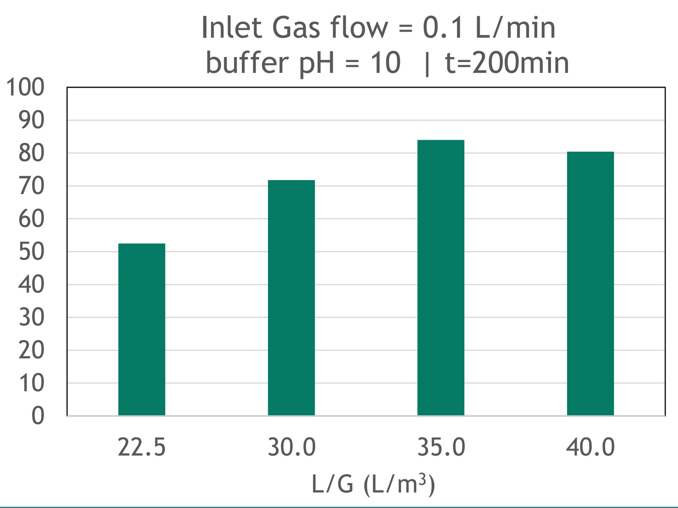
Inlet Gas Flow (L/min)	GRT (s)	CO_2 Loading (g/L/min)
0.1	930	0.0253
0.3	310	0.0759
0.5	186	0.1265
1.0	93	0.2529

Results and Discussion

The summarised results based on the parameters (Table 1&2) and equation (3) are shown below.







- The best buffering capacity and the highest capture efficiency were observed with the NH_3 / NH_4Cl buffer with pH 10.
- CO_2 capture yield decreases gradually as the inlet gas flow rises and the L/G ratio reduces.
- Higher GRT and thus lower CO_2 loading leads to higher rates of CO_2 capture.
- Carbonates and ammonia-based solutions can both provide high CO_2 capture efficiency but carbonates have lower toxicity, volatility, corrosiveness, and degradation. However, ammonia can escape and produce secondary pollution.

Conclusions

The chemical absorption of CO_2 was studied in a lab-scale scrubber system and it is concluded that the CO_2 capture efficiency maximizes on pH 10 and it rises when inlet gas flow decreases and L/G ratio increases.

Next steps

Addition of activators such as Carbonic Anhydrase enzyme to improve the low reaction rate and process efficiency of CO_2 absorption with carbonates.

Scale-up of the scrubbing system in pilot scale.

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Acknowledgements

The authors of this work acknowledge that this project has received funding from the European Union's Horizon 2022 research and innovation programme under grant agreement No 101084405 (CRONUS).

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