

Evaluating the experimental CO₂ stripping performance of a new generation multicapillary Taylor flow reactor

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ABSTRACT

Among all physicochemical technologies commercially available to upgrade biogas to biomethane, chemical scrubbing with carbonated solutions stand as an easy-to-scale technology. However, the regeneration of the solvent via air-assisted CO₂ stripping is highly energy-intensive, representing >80 % of the total process costs. This study proposes the use of innovative Taylor flow reactors to enhance the liquid-gas mass transfer of CO₂ and therefore lower the cost of regeneration of carbonated solutions. In this study, a 200-glass capillary Taylor flow reactor supporting unprecedented mass transfer coefficients ($k_L a$) was tested for the first time for CO₂ stripping from carbonated solutions. The Taylor flow reactor achieved $k_L a$ coefficients higher than 100 h⁻¹ at an inorganic concentration (IC) of 1000 mgC L⁻¹, and $k_L a > 400$ h⁻¹ at an optimal gas superficial velocity of 0.30 m s⁻¹. Moreover, increasing IC concentrations to 2000 and 3000 mgC L⁻¹ resulted in a $k_L a$ increase of 250 % and 65 %, respectively, whilst no significant increase was recorded at an initial IC concentration of 4000 mgC L⁻¹. Indeed, multicapillary Taylor flow reactors demonstrated a superior and competitive performance during CO₂ stripping from carbonated solutions, representing a promising technology for solvent regeneration during biogas upgrading at industrial scale.

1. Introduction

Currently, biogas is facing an increased momentum mediated by the political incentives that encourage the production of clean energies to accelerate the independence from fossil fuels and decarbonize the economy. In 2023, biogas and biomethane global production accounted for 50 billion cubic meters, with Europe, China and The United States being the dominant regions and accounting for 90 % of the total worldwide production [1].

The energy content of biogas varies depending on its composition, which typically consists of 60–70 % methane (CH₄), 30–40 % carbon dioxide (CO₂) and other trace components in lower concentration, resulting in a heating value of 16–28 MJ m⁻³. To improve its energy content and achieve heating values similar to natural gas, biogas must be upgraded to biomethane by removing the CO₂ and trace components, especially if its final use is the injection in the natural gas grids or its use in the transportation sector [2].

Different physicochemical technologies including cryogenic

separation, membrane separation, pressure swing adsorption, solvent/chemical scrubbing and water scrubbing, have been established and are commercially available to upgrade biogas to biomethane. Currently, scrubbing techniques such as chemical absorption rank among the most popular techniques in Europe, due to its easy scalability and industrial commercialization [3]. Chemical absorption is based on the use of absorbents including alkanolamines (i.e. monoethanolamine or dimethylethanolamine) and alkali solutions (i.e. KOH, NaOH, Fe(OH)₃, FeCl₂ and carbonated solutions). In this context, carbonated solutions (i.e. K₂CO₃, NaCO₃) present several advantages over amine solutions due to their recyclability and non-corrosive nature [4]. Nonetheless, the use of carbonated solutions is limited by the high energy input required for solvent regeneration. Typically, the regeneration process consists of desorbing or stripping the captured CO₂ from the aqueous solution, which allows recovering the solvent/chemical solution for its reuse [5]. It is estimated that the CO₂ stripping process accounts for 80 % of the total cost of biogas upgrading during chemical absorption, since it requires high temperatures and pressures as a result of the poor CO₂

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diffusion from the liquid phase to the gas phase [6]. For instance, Abbasi et al. [7] demonstrated that carbonated solutions could remove up to 86 % of the CO₂ present in biogas in a spouted bed reactor operated at 55 °C, but solvent regeneration required temperatures ranging between 140 °C and 200 °C. Similarly, Ye et al. [8] conducted CO₂ stripping from carbonated solutions at temperatures of 140 °C and high pressures.

Different techniques have been implemented to reduce the energy input during CO₂ stripping, including the use of additives. Typically, CO₂ stripping is carried out in stripper columns or degassers, with limited mass transfer coefficients (k_La) ranging between 29 h⁻¹ and 72 h⁻¹ [9]. Higher k_La coefficients above 4000 h⁻¹ have been reported in microchannel reactors [10,11]. However, their application is limited to alkanolamines solutions. Thereby, intense engineering efforts are needed in reactor design to improve the liquid-gas CO₂ mass transfer, which would ultimately increase CO₂ removal efficiencies while reducing operational costs.

In this context, multicapillary Taylor flow reactors have recently demonstrated a superior performance compared to stirred tank and air-lift reactors, with up to 10-fold higher mass transfer coefficients. This could significantly increase the solubility of hydrophobic compounds [12]. For instance, Rocha-Ríos et al. [12] reported k_La coefficients > 3600 h⁻¹ in a capillary reactor devoted to CH₄ treatment. The CH₄ removal was mainly influenced by the gas flow, and increasing the gas flow from 20 mL min⁻¹ to 70 mL min⁻¹ resulted in a 19 % k_La increase. Similarly, Herrero-Lobo et al. [13] and Herrero-Lobo et al. [14] demonstrated the superior performance of a 25 capillaries Taylor flow reactor during CH₄ bioconversion to ectoine.

Hence, the aim of this study was to demonstrate the superior performance of a new generation of multicapillary Taylor flow reactors devoted to CO₂ stripping from carbonated solutions used in biogas upgrading units operated at room temperature. The improved design of the reactor consisted of 200-glass capillaries supporting high gas and liquid superficial velocities, which would ultimately entail a high CO₂ mass transfer in the gas-liquid interface. Different operational conditions were evaluated to improve the desorption of CO₂, including the superficial liquid and gas velocities (0.07 m s⁻¹ - 0.10 m s⁻¹ and 0.17 m s⁻¹ - 0.30 m s⁻¹, respectively) and the initial pH of carbonated solutions (8,9 and 10). Finally, the influence of inorganic carbon concentrations (1000 mg L⁻¹, 2000 mg L⁻¹, 3000 mg L⁻¹ and 4000 mg L⁻¹) on CO₂ stripping was also evaluated.

2. Materials and methods

2.1. Carbonated solution

Fresh carbonated solutions containing an inorganic carbon (IC) concentration of 1000 mg L⁻¹, 2000 mg L⁻¹, 3000 mg L⁻¹ and 4000 mg L⁻¹ were prepared with sodium bicarbonate (BICAR®FOOD, Spain) and sodium carbonate (PanReac AppliChem, Spain) at different initial pH values: 8.5, 9.0 and 10.0, depending on the studied test series. Antifoam 204 (Sigma-Aldrich, USA) was added to the solutions at a concentration of 0.3 mL_{antifoam}L_{carbonated solution}⁻¹ to prevent foam formation inside the reactor.

2.2. Experimental set-up

The multicapillary Taylor flow reactor was an improved version of the capillary bioreactor previously described in [15]. The innovative multicapillary Taylor flow reactor contained a working volume of 13.5 L and was composed of 200 glass capillaries of 0.3 cm of internal diameter and 150 cm length. The liquid inside the reactor was recirculated by a centrifugal pump (PRISMA 25-3 M, Spain), and a rotameter (Georg Fisher type 335, Germany) was used to measure the liquid recirculation flow rate. The bottom reservoir was filled with Kaldnes rings (6 mm, Kaldnes K1 rings, Evolution Aqua Ltd., UK) to improve the liquid-gas mixing and ensure an even distribution within the capillaries. The air

inlet was located at the bottom reservoir and it was controlled with an Aalborg rotameter S/N°512,416-2 (USA). A schematic representation of the capillary Taylor flow reactor system is shown in Fig. 1.

2.3. Experimental conditions

The stripping of CO₂ was conducted in the innovative multicapillary Taylor flow reactor by varying the liquid and gas superficial velocities, the initial pH and IC concentration of the carbonated solutions. Two different liquid superficial velocities (v_l), 0.017 m s⁻¹ and 0.10 m s⁻¹, were tested to recirculate the carbonated solution inside the reactor, depending on the test series. CO₂-free compressed air was injected at the bottom of the reactor under co-current flow operation at different gas superficial velocities (v_g) of 0.10 m s⁻¹ and 0.30 m s⁻¹. These gas and liquid superficial velocities were selected to ensure Taylor flow within the capillaries according to the flow-regime map described in [16], where similar system conditions were used, i.e. capillary diameter, air and water as fluids. However, it is important to address that the compact arrangement of the capillaries limited the visualization of the flow regime within the capillaries in our particular study. Three different initial pH were tested 8.5, 9.0 and 10.0, and four different initial IC concentrations were evaluated (1000 mg L⁻¹, 2000 mg L⁻¹, 3000 mg L⁻¹ and 4000 mg L⁻¹). These pH and IC concentrations were selected based on typical IC concentrations needed during biogas upgrading with carbonated solutions previously reported in [17,18]. pH was selected above the typical final pH achieved during biogas upgrading to prevent CO₂ stripping during solution preparation, and to test the mass transfer performance of the multicapillary reactor and high pH.

Three different test series were conducted to study the CO₂ stripping capacity of the multicapillary Taylor flow reactor. Briefly, test series I investigated the influence of v_l on the CO₂ stripping at different initial pHs (8.5, 9 and 10), while the v_g and the initial IC concentration remained constant at 0.10 m s⁻¹ and 1000 mgC L⁻¹, respectively. Test series II assessed the influence of v_g on the CO₂ stripping at different initial pHs (8.5, 9 and 10), while v_l and the initial IC concentration remained constant at 0.10 m s⁻¹ and 1000 mgC L⁻¹, respectively. Finally, test series III was devoted to study the influence of the initial IC concentration on the CO₂ stripping process at an initial pH of 9.0, v_l of 0.10 m s⁻¹ and v_g of 0.17 m s⁻¹. The experiments were conducted at 25 °C. The summary of the operational conditions evaluated in each test series is shown in Table 1. The CO₂ stripping process was monitored by periodically taking 10 mL of liquid samples to measure pH and IC

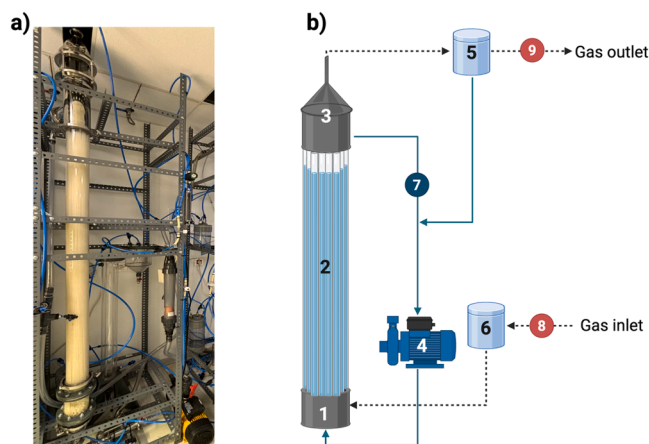


Fig. 1. a) Innovative multicapillary Taylor flow reactor; b) Schematic diagram of the multicapillary Taylor flow reactor system: 1) gas-liquid mixing bottom reservoir; 2) glass capillaries; 3) liquid-gas separation reservoir; 4) hydraulic pump; 5) liquid trap; 6) gas-mixing chamber; 7) liquid sample port; 8) gas inlet sample port; 9) gas outlet sample port. Dotted lines represent gas streams, continuous lines represent liquid streams.

Table 1
Operational parameters tested during the CO₂ stripping process.

Parameter	Test series		
	I	II	III
Initial IC (mg L ⁻¹)	1000	1000	1000,2000,3000,4000
Initial pH	8.5, 9.0, 10.0	8.5, 9.0, 10.0	9.0
Liquid superficial velocity (m s ⁻¹)	0.07, 0.10	0.10	0.10
Air superficial velocity (m s ⁻¹)	0.17	0.17, 0.30	0.30

concentration. Additionally, gas samples were taken at the reactor outlet to measure the CO₂ content in the gas outlet at 0 min, 10 min, 20 min, 30 min, 60 min, 90 min, 120 min, 150 min, and 180 min. Each experimental condition was run in duplicate.

2.4. CO₂ stripping, mass transfer and IC removal efficiency calculations

The carbonate buffer system was composed of four carbon species: dissolved CO₂, carbonic acid (H₂CO₃), bicarbonate (HCO₃⁻) and carbonate (CO₃²⁻). Dissolved CO₂ and H₂CO₃ are difficult to measure independently and are typically considered as one single species CO₂^{*}.



The equilibrium CO₂ concentration in an aqueous solution [CO₂]_{eq} is governed by the two following reactions, thus is highly dependent on the pH of the solution:



When CO₂ is stripped-out from the carbonated solution the pH increases and the equilibrium of Eqs. (2) and (3) is shifted to the left.

The CO₂ stripping process in the multicapillary Taylor flow reactor is characterized by gas bubbles that rise in the capillary ducts and spend within the ducts a time equal to the capillary length divided by their velocity. During the residence time of a single bubble (which is of the order of seconds) the concentration [CO₂]_{eq} is considered constant. The concentration of CO₂ in the gas bubbles can be described from the following mass balance:

$$-\frac{d[\text{CO}_2]_g}{dt} = \frac{k_L a}{\varepsilon_g} ([\text{CO}_2]_b - [\text{CO}_2]_{eq}) \quad (4)$$

The concentration of CO₂ in the gas is related to its molar fraction through the equation of the ideal gases:

$$[\text{CO}_2]_g = \frac{Py}{RT} \quad (5)$$

The concentration of CO₂ in the liquid phase that is in contact with the bubble is related to the molar fraction in the gas through the Henry constant K_H.

$$[\text{CO}_2]_b = K_H Py \quad (6)$$

The ratio $\frac{a}{\varepsilon_g}$ is just the surface volume ratio of the gas bubbles.

Eq. (4) can thus be rearranged as follows:

$$-\frac{dy}{dt} = \frac{k_L a}{\varepsilon_g} K_H RT \left(y - \frac{[\text{CO}_2]_{eq}}{K_H P} \right) \quad (7)$$

Integrating the previous equation and rearranging the terms we obtain:

$$y_f = \frac{[\text{CO}_2]_{eq}}{K_H P} \left(1 - \exp \left(-K_H RT \left(\frac{k_L a}{\varepsilon} \right) t_R \right) \right) + y_0 \exp \left(-K_H RT \left(\frac{k_L a}{\varepsilon} \right) t_R \right) \quad (8)$$

where: y_f is the molar fraction of CO₂ in the exiting gas; y_0 is the molar fraction of CO₂ in the inlet gas; $[\text{CO}_2]_{eq}$ is the concentration of dissolved CO₂ in equilibrium with the total inorganic carbon in mol L⁻¹; K_H is the Henry constant, at 25 °C, 0.0349 mol L⁻¹ atm⁻¹; R is the ideal gas constant 0.082 atm L mol⁻¹ K⁻¹; T is the temperature 298 K; t_R is the residence time of the bubbles in the column (length divided by velocity); P is the pressure in atmospheres; $\frac{k_L a}{\varepsilon}$ is the mass transfer coefficient divided by the volumetric fraction of gas in the column.

Previous studies have demonstrated that when the mass transfer at the gas-liquid interface is low, there is no significant difference in the calculated gas holdup values when using superficial velocities or slug length. Since CO₂ mass transfer is low, especially at high pH, where the concentration of the carbon specie that can be stripped, CO₂^{*} (CO₂ + H₂CO₃), is low and keeps decreasing with time, the gas hold-up in the system was calculated according to [19]:

$$\varepsilon_G = \frac{v_g}{v_g + v_l} \quad (9)$$

where v_g is the gas superficial velocity (m s⁻¹) and v_l is the liquid superficial velocity (m s⁻¹).

Then, the $k_L a$ can be estimated by linear regression.

Finally, the IC removal efficiency was calculated considering the operation time of the reactor as:

$$\text{IC}_{Rem. efficiency} (\%) = \frac{IC_{t0} - IC_t}{IC_{t0}} \times 100 \quad (10)$$

where IC_{t0} is the IC concentration (mg l⁻¹) at time t_0 ; and IC_t is the IC concentration (mg L⁻¹) in the solution at time t of operation. In this particular case, time t of operation was 180 min.

2.5. Analytical procedures

The CO₂ concentration in the gas stream was monitored by gas chromatography using a Varian CP-3800 GC-TCD (Palo Alto, USA) equipped with a CP-Molsieve 5A and a CP-Pora BOND Q columns according to [20]. The IC concentration in the liquid phase was determined in a Shimadzu TOC-VCSH analyzer (Japan). The pH was measured with a SensION™+ PH3 pHmeter (HACH, Spain). Alkalinity of the carbonated solutions was measured by titration following standard methods [21] whilst the kinematic viscosity was determined with a viscometer Anton Par SVM1001. Finally, superficial tension was measured with a tensiometer Kruss Easy Dyne (K20).

2.6. Statistical analysis

ANOVA analysis of variance considering a significant level $\alpha = 0.05$ was conducted to identify differences in the operational conditions tested during the CO₂ stripping experiments.

3. Results and discussion

3.1. Influence of the liquid superficial velocity and pH on CO₂ stripping

The initial pH (8.5, 9 and 10) significantly influenced IC removals regardless of the liquid superficial velocity set (Table 2). Higher IC removals (> 20 %) were obtained when the initial pH was 8.5 while the CO₂ dissolved concentration decreased up to 97 % at an initial pH 8.5. This high IC removal efficiency was related to the time course of the pH in the aqueous solution, since at higher pH the equilibrium of Eq. (2) is shifted towards HCO₃⁻. Additionally, it is important to highlight that the

Table 2

Influence of the liquid superficial velocity (v_l) and gas superficial velocity (v_g) on the gas holdup (ϵ_G), IC removal efficiency and CO₂ stripping efficiency.

(m s ⁻¹)	v_g (m s ⁻¹)	ϵ_G	G/L ratio	Initial pH	Alkalinity (mg CaCO ₃ L ⁻¹)	IC removal efficiency (%)
0.07	0.18	0.70	2.5	8.5	4640	28 ± 2
0.07	0.18	0.70	2.5	9.0	5010	16 ± 3±
0.07	0.18	0.70	2.5	10.0	6847	-
0.10	0.17	0.63	1.7	8.5	4640	22 ± 0.3
0.10	0.17	0.63	1.7	9.0	5010	20 ± 1
0.10	0.17	0.63	1.7	10.0	6847	2 ± 1
0.10	0.30	0.74	3	8.5	4640	21 ± 0.5
0.10	0.30	0.74	3	9.0	5010	23 ± 0.1
0.10	0.30	0.74	3	10.0	6847	-

alkalinity of the solutions was influenced by the initial pH, resulting in an increased buffer capacity when the initial pH was set to 10.0. Therefore, at an initial pH 10.0, the IC removal was significantly lower regardless of the v_l . In this context, during the first 30 min of the experiment the pH significantly increased, suggesting that during this period the largest amount of H₂CO₃ was stripped out (Fig. 2).

Moreover, when v_l was increased from 0.07 m s⁻¹ to 0.10 m s⁻¹, no significant differences were observed in the IC removal regardless of the initial pH ($p > 0.05$). Thus, the major mass transfer from the gas-liquid interface under Taylor flow regimes is typically carried out in the liquid film, where the mass transfer depends on the bubble length. In this context, air bubbles longer than 2 cm saturate the liquid film [22], while shorter air bubbles typically lead to a higher mass transfer. However, the reduced size of the air bubbles mediated by the increased v_l , did not significantly impact the k_{La} , as the air bubbles were saturated with CO₂, thus limiting the CO₂ stripping process (Fig. 3). Additionally, when v_l was increased from 0.07 m s⁻¹ to 0.10 m s⁻¹, the liquid residence time in the capillaries decreased, resulting in a reduced contact time between the gas and the liquid interphase [6].

The k_{La} coefficients exhibited the same tendency as the removal of IC was significantly influenced by the initial pH (Fig. 3a) regardless of the v_l tested ($p > 0.05$). When the v_l was increased to 0.10 m s⁻¹, the k_{La} at an initial pH of 8.5 was increased by 28 % ($p > 0.05$), whilst at an initial pH 9.0 the k_{La} did not change significantly. This result support the theory that the increased v_l reduced the contact time between the liquid-gas interfaces. It is important to address that at initial pH 10, the CO₂* concentration was lower and did not change significantly, particularly when the v_l was 0.07 m s⁻¹ where the CO₂* concentration remained at the initial value of 1.11×10^{-6} M. Hence, the combination of high alkalinity, low CO₂* concentration and high buffer capacity mediated by the chemical equilibrium shifted to the formation of HCO₃⁻ anions, resulted in no significant mass transfer at an initial pH 10.0. Therefore, the k_{La} coefficient was not calculated for none of the conditions at an initial pH 10.0. Finally, it is important to highlight the superior design of the 200-glass-capillary Taylor flow reactor herein used, as k_{La} coefficients ranging from 100 h⁻¹ to 270 h⁻¹ were recorded at G/L ratios of 1.7 - 2.4. The obtained k_{La} coefficients are in the range of the k_{La} coefficients of 462 h⁻¹ and 294 h⁻¹ previously reported by Kraakman et al. [23], at similar G/L ratios of 1.5 - 2.9 in a 25-capillary Taylor flow reactor [23]. Despite the fact that the k_{La} coefficients herein obtained were lower, the improved design of the multicapillary Taylor flow reactor, increased the packing density by almost an order of magnitude, enabling higher gas and liquid flow rates than the previously reported in multicapillary Taylor flow reactors [23]. Notably, these gas and liquid flow rates surpass the capacity of other reactor configurations, namely external loop airlift reactors by a factor of 2 to 5 times, resulting in k_{La} coefficients up to 33 % higher [24].

3.2. Influence of the gas superficial velocity on CO₂ stripping

During the second test series of the study, the influence of v_g was

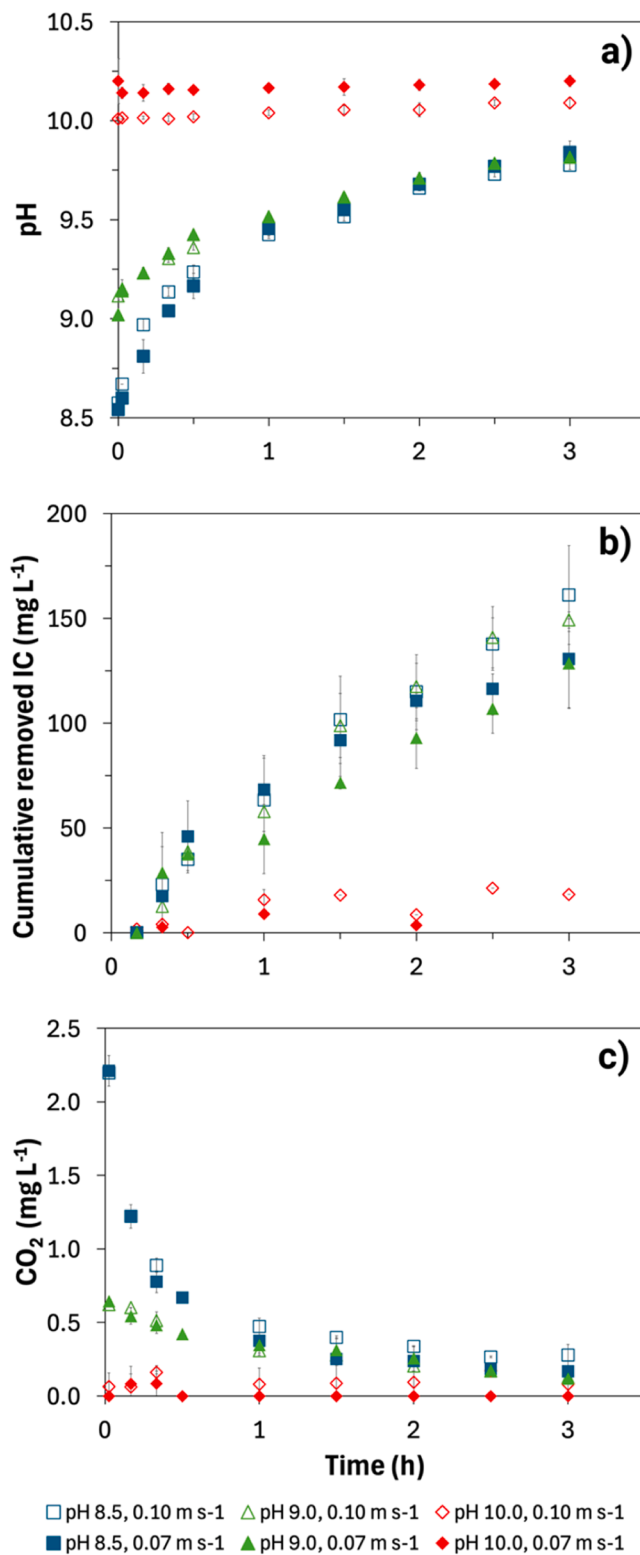


Fig. 2. Time course of a) pH; b) cumulative IC concentration removed (mg L⁻¹) in the liquid phase; c) gas CO₂ concentration (mg L⁻¹) in the effluent gas stream; at two different liquid superficial velocities (0.07 m s⁻¹-filled markers; and 0.10 m s⁻¹-empty markers).

evaluated by maintaining v_l at 0.10 m s⁻¹ and increasing v_g from 0.17 m s⁻¹ to 0.30 m s⁻¹, which resulted in a final G/L ratio of 3. Interestingly, the pH in the absorption broth increased faster when v_g was increased to 0.30 m s⁻¹, especially at an initial pH of 9.0 (Fig. 4a). This sharp increase

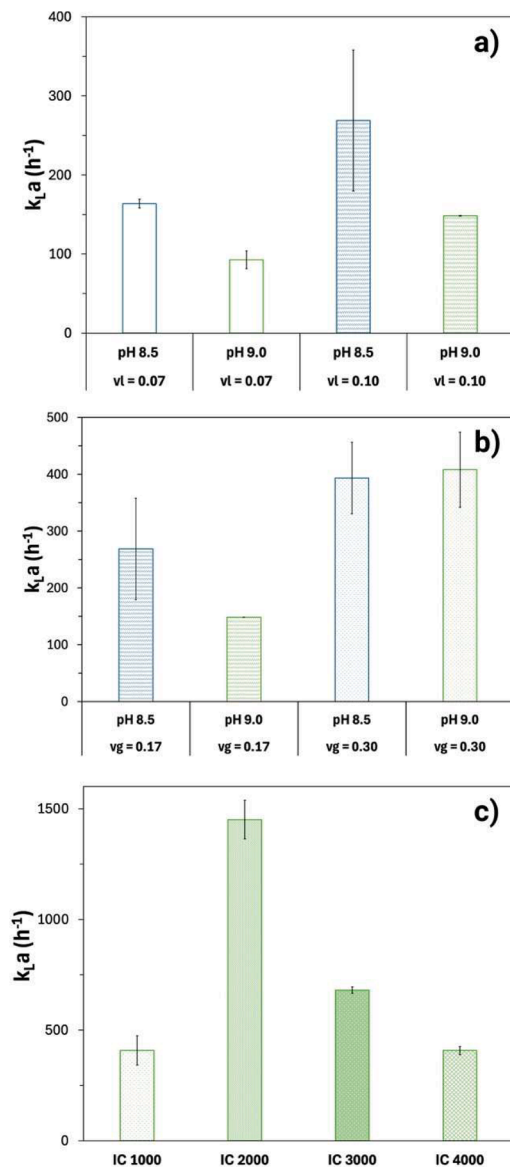


Fig. 3. Influence of the a) pH and liquid superficial velocity; b) pH and gas superficial velocity; and c) initial IC concentration on the mass transfer coefficients ($k_L a$) during CO_2 stripping.

in pH of the solution occurred mainly in the first 30 min of experiment, and was supported by the air-assisted CO_2 desorption (Fig. 4c). Similarly, the cumulative IC removed significantly increased at an initial pH of 9.0 when the v_g was 0.30 m s^{-1} (Fig. 4b) ($p < 0.05$), whilst no significant difference was observed at an initial pH of 8.5 ($p > 0.05$). This improved CO_2 stripping performance at an initial pH 9.0 was supported by the increased $k_L a$ coefficient (Fig. 3b).

In this context, the $k_L a$ coefficients increased by 51 % and 165 % at an initial pH of 8.5 and 9.0 respectively, when v_g was increased from 0.10 to 0.30 m s^{-1} ($p < 0.05$). Interestingly, no significant difference was observed between the $k_L a$ coefficients obtained at a v_g of 0.30 m s^{-1} regardless of the initial pH (Fig. 3b). Similar behaviors have been previously reported by [12] in a capillary bioreactor when v_g was increased from 0.12 m s^{-1} to 0.44 m s^{-1} , resulting in $k_L a$ increased of 38 %. Similarly, Bordel et al. [22] have theoretically demonstrated that the mass transfer in a multicapillary Taylor flow reactor increases with increasing bubble velocity. The increase in $k_L a$ coefficients can be attributed to the increased ϵ_G as the interfacial area between gas-liquid phases increases. For instance, it has been recently demonstrated that at an ϵ_G of 0.75 the

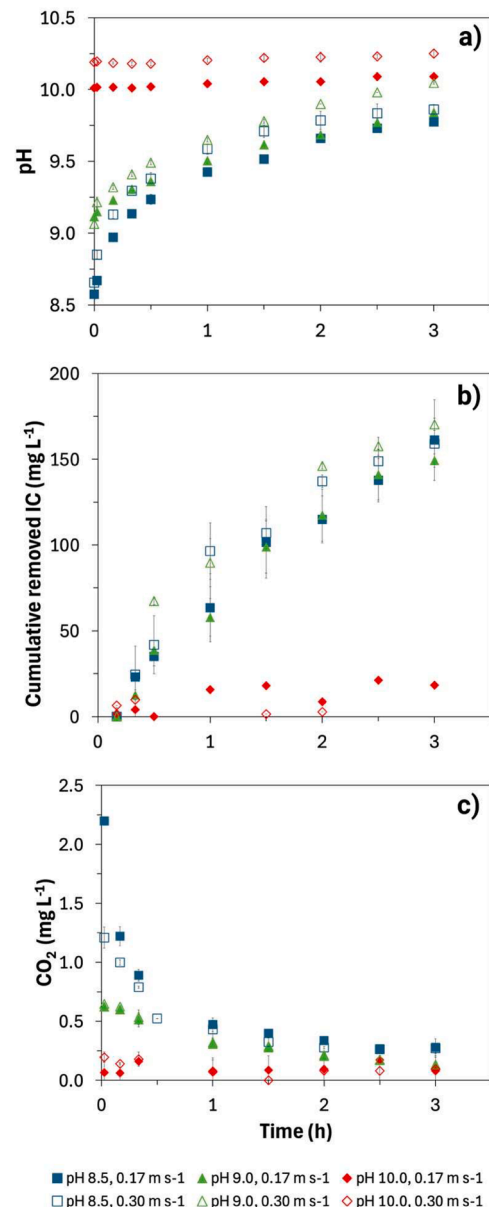


Fig. 4. Time course of a) pH; b) cumulative IC concentration removed (mg L^{-1}) in the liquid phase; c) stripped CO_2 concentration (mg L^{-1}) in the gas phase at different air superficial velocities of 0.17 m s^{-1} (filled markers) and 0.30 m s^{-1} (empty markers).

$k_L a$ coefficient is significantly increased, particularly when the unit length (bubble length plus slug length) is low [22]. Typically, ϵ_G increases when v_g is increased and v_l is either decreased or maintained [16], resulting in a higher volume of gas to stripped out the dissolved CO_2 . In this context, ϵ_G increased by 17 % when v_g was increased to 0.30 m s^{-1} , confirming the positive correlation between $k_L a$ and ϵ_G . Similarly, Abufalgha et al. [25], studied the effect of increasing gas superficial velocity on the $k_L a$ coefficient in a bubble column reactor, and observed that the $k_L a$ was correlated with the increased gas-liquid interfacial area. Hence, the $k_L a$ increased almost by 40 % (162 h^{-1} to 234 h^{-1}) when the v_g increased from 0.016 m s^{-1} to 0.031 m s^{-1} . In the same context, Mild et al. [26], observed an increase in the $k_L a$ coefficient of a bubble column from 90 h^{-1} to 360 h^{-1} when the v_g increased from 0.0012 m s^{-1} to 0.59 m s^{-1} [26]. These results support the superior performance of the 200-multicapillary Taylor reactor as $k_L a$ coefficients of around 400 h^{-1} were obtained at a v_g half of the one used by Mild et al. [26]. $k_L a$ coefficients

of 72 h^{-1} , 96 h^{-1} , 4968 h^{-1} and 9648 h^{-1} have been reported in literature for packed bed columns, cascade degassers, micro-packed bed reactors, and microchannel reactors, respectively during CO_2 stripping [6,9–11, 27]. Certainly, microchannels reactors exhibit higher k_{La} values, with one particular study reporting a k_{La} of 9648 h^{-1} [10], however their size could limit their industrial applications due to clogging mediated by carbonate precipitation. Moreover, it is important to highlight that the higher k_{La} 's, particularly the values of 4968 h^{-1} and 9648 h^{-1} , were obtained using amines-based solutions (i.e. MDEA) at high temperatures ($> 70^\circ \text{C}$), which significantly influence the absorption/desorption of CO_2 compared to the carbonated solutions herein used.

3.3. Influence of the initial IC concentration on CO_2 stripping

The effect of the initial IC concentration on CO_2 stripping is shown in Fig. 5. It can be observed that the increasing concentration of IC impacted the kinetics of increase of pH in the carbonated solution (Fig. 5a) and the final pH was significantly lower at higher IC concentrations. In contrast, the cumulative removed IC and the desorbed CO_2 significantly increased with increasing IC concentrations (Fig. 5b and c). Similarly, the IC removal efficiency was increased by 139 % when the IC increased to 4000 mg L^{-1} . This can be supported by the Fick's law, which states that molecules from the solute with higher concentration will diffuse to the zone of lower concentration [28]. In this context, it was expected that the increased concentration of CO_2 dissolved and H_2CO_3 concentrations in the liquid phase at higher IC concentrations would result in a higher CO_2 diffusion to the gas phase, thus increasing the k_{La} coefficient [6]. Indeed, the k_{La} coefficients significantly increased at increasing IC concentrations, but this increment was not linear (Fig. 3c). Similar trends were observed in the surface tension of the solutions, which did not exhibit a significant change and were in a range of 0.036 N m^{-1} and 0.041 N m^{-1} . In addition, the limited vision of the fluid within the capillaries due to the compact in configuration of the reactor, could not confirm the influence of the surface tension on the bubble patterns.

A 2.5 fold increment in the k_{La} was obtained in solutions when the initial IC concentration increased from 1000 mgC L^{-1} to 2000 mgC L^{-1} , while k_{La} was only increased by 67 % when the IC concentration was increased to 3000 mgC L^{-1} and no significant difference was observed at an initial IC of 4000 mgC L^{-1} ($p > 0.05$). This behavior in the k_{La} can be attributed to two main physico-chemical properties of the solutions (viscosity and alkalinity). The viscosity of a liquid is related to the mass transfer resistance, and as shown in Table 3, the kinematic viscosity of the solutions slightly increased with the increased IC concentration [6]. On the other hand, in our particular study, the IC concentration played a key role in CO_2 mass transfer as higher IC concentrations entailed a higher buffer capacity mediated by an increased alkalinity [17]. The alkalinity of the carbonated solutions was drastically increased at increasing IC concentrations (Table 3), fostering the buffer capacity of the system, which was reflected in the lower final pHs recorded with increasing IC concentrations. These findings are supported by the results previously reported by Summerfelt et al. [29], who observed that increasing alkalinities did not affect the CO_2 stripping efficiency of a degasser but led to a more stable pH through the time course of the experiment. Different studies have demonstrated that increasing solvent concentration influences the k_{La} coefficient. For instance, Abufalgha et al. [25] observed that when the alkane concentration increased from 2.5 % to 20 % v/v, the k_{La} of a bubble column reactor declined from 252 h^{-1} to 216 h^{-1} mainly due to the change in the viscosity of the fluid. In addition, Ruen-ngam et al. [30] studied the effect of salinity on the k_{La} coefficient of an airlift reactor and observed that increasing salinity from 0 ppt to 45 ppt did not influence the k_{La} at $v_g < 0.03 \text{ m s}^{-1}$. However, when the v_g was higher than 0.03 m s^{-1} , the increased salinity decreased the k_{La} , but this reduction was not linear [30]. Hence, in this study the viscosity of the liquid fluid did not significantly change with increasing IC concentrations, supporting the fact that the decrease in k_{La} was likely due to the increased alkalinity, which conferred an increased buffer

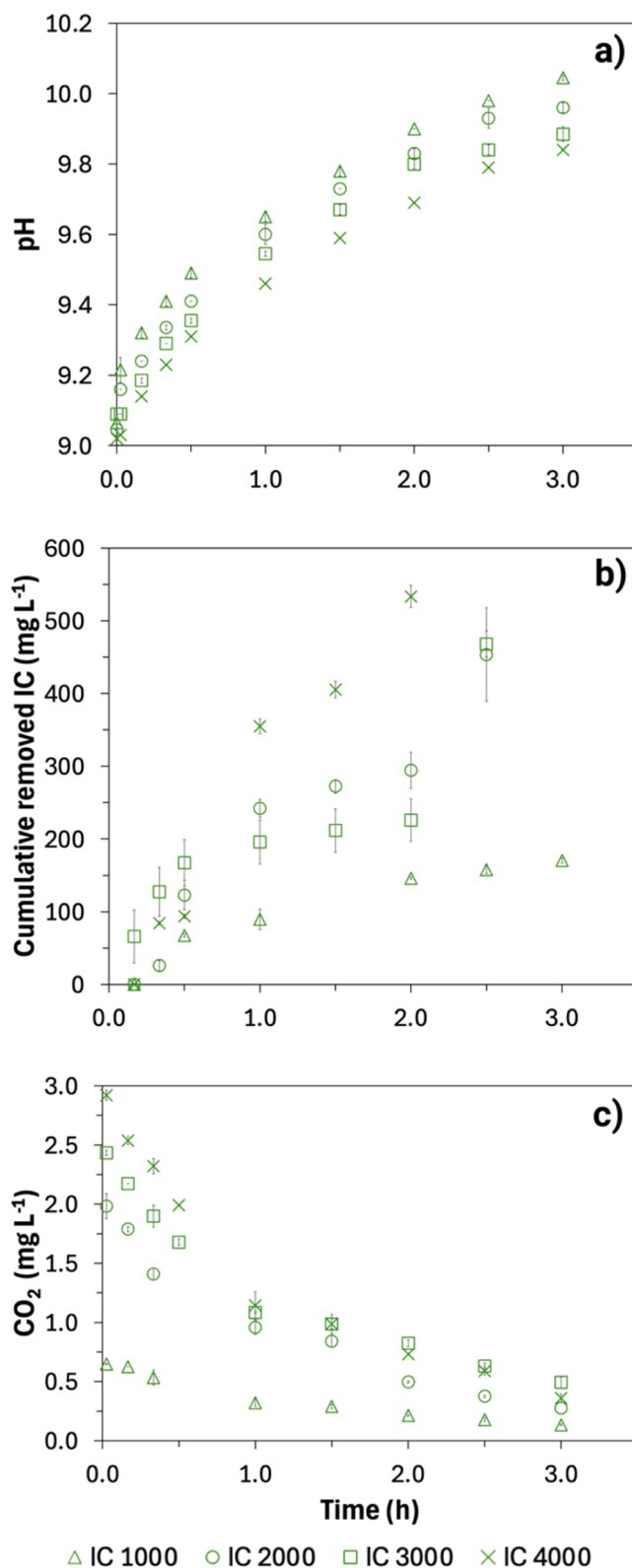


Fig. 5. Time course of the influence of different IC concentrations (1000, 2000, 3000 and 4000 mg L^{-1}) in CO_2 stripping process at a liquid superficial velocity of 0.10 m s^{-1} and gas superficial velocity 0.30 m s^{-1} at an initial pH of 9: a) pH; b) cumulative removed IC (mg L^{-1}) of the liquid phase; c) stripped CO_2 concentration in the gas outlet (mg L^{-1}).

Table 3

Influence of the initial inorganic carbon (IC) concentration on the kinematic viscosity, IC removal efficiency and CO₂ stripping efficiency during CO₂ stripping in a multicapillary Taylor flow reactor operated at a liquid superficial velocity of 0.10 m s⁻¹ and a gas superficial velocity 0.30 m s⁻¹.

Initial IC concentration (mg L ⁻¹)	Kinematic viscosity (mm ² s ⁻¹)	Alkalinity (mg CaCO ₃ L ⁻¹)	IC removal efficiency (%)
1000	0.923	5010	23.1 ± 0.1
2000	0.942	9754	24.9 ± 0.3
3000	0.958	14,270	21.8 ± 0.4
4000	0.972	23,325	32.3 ± 1.3

capacity to the system.

Despite the high alkalinity of the carbonated solutions, the multicapillary Taylor flow reactor still achieved superior IC removal efficiencies (> 20 %) to those previously reported in degassers, which typically range between 38 % at low alkalinities < 10 mg L⁻¹ CaCO₃ and < 10 % at relative high alkalinities > 100 mg CaCO₃L⁻¹.

4. Conclusions

This study aimed at demonstrating the superior performance of an innovative 200-capillary Taylor-flow reactor during the CO₂ stripping process from carbonated solutions devoted to biogas upgrading. The mass transfer of the dissolved CO₂ from the liquid phase to the gas phase was mainly influenced by the superficial gas velocity and inorganic carbon concentration. At an inorganic carbon concentration of 1000 mg L⁻¹, k_La coefficient of 407 h⁻¹ was obtained at optimal conditions. Increasing inorganic carbon concentration increased the alkalinity of solutions, fostering the buffer capacity of the system and significantly affecting the mass transfer of CO₂. However, the innovative Taylor flow reactor herein tested supported mass transfer coefficients of 1450 h⁻¹, 680 h⁻¹ and 407 h⁻¹ at inorganic carbon concentrations of 2000 mg L⁻¹, 3000 mg L⁻¹ and 4000 mg L⁻¹, respectively with IC removal efficiencies > 20 %. Indeed, the superior design of the Taylor flow reactor resulted in competitive k_La coefficients outperforming other reactor configurations, obtaining similar k_La coefficients than bubble columns at half gas superficial velocities. Moreover, IC removal efficiencies and mass transfer coefficients compared to conventional CO₂ stripping technologies were obtained without the need of an additional energy input or the intense use of chemicals i.e. MDEA. Therefore, multicapillary Taylor flow reactors stand as a promising low-cost technology for CO₂ stripping.

CRedit authorship contribution statement

Laura Vargas-Estrada: Writing – original draft, Methodology. **Sergio Bordel:** Validation, Methodology. **Raquel Lebrero:** Writing – review & editing, Funding acquisition. **Raúl Muñoz:** Writing – review & editing, Supervision, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Data availability

Data will be made available on request.

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