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Catalytic effect of boric acid in CO_2 absorption

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Abstract

As a consequence of the accelerated industrial growth, the problem involved in the treatment of gases emitted into the atmosphere by the industrial sector, especially carbon dioxide, takes on greater relevance, therefore, the development and exploration of new technologies for the capture of CO_2 must always be under investigation. This study employed a pilot-scale absorption unit to investigate the capacity of the absorption of carbon dioxide using an aqueous solution of sodium hydroxide and boric acid as catalyst. The evaluated parameters included two different concentration of absorbent, operating temperature and the kinetic effect due the presence of boric acid. The results showed that when the concentration of the absorbent was increased, the removal of CO_2 is highly favored, also the kinetic effect was favored when the operating temperature was stepped up and the presence of boric acid improves the CO_2 kinetic rate.

Keywords: Carbon dioxide, absorption, aqueous sodium hydroxide, boric acid.

1. Introduction

As a consequence of the accelerated industrial growth, the problem involved in the treatment of gases emitted into the atmosphere by the industrial sector, especially carbon dioxide, takes on greater relevance. In connection with both environment and economic, this sector is directly responsible for 30% of the global emission of carbon dioxide (Fischedick et al., 2014), product of the combustion of fossil fuels (coal, oil and natural gas), and the main factor causing the intensification of the greenhouse effect and climate change (Siriwardane et al., 2007; Ye et al., 2013). The concentration of carbon dioxide in the atmosphere has increased considerably, going from 280 ppm (before the industrial revolution) to 315 ppm in 1958, then to 377 ppm in 2004 (Canadell et al., 2007; Yang et al., 2008) and reaching currently to exceed 400 ppm (NOAA, 2014).

Therefore, in recent years there has been increasing concern among the scientific and industrial community for the development and exploration of new technologies for the capture of carbon dioxide. These technologies include chemical, physical absorption (Endo et al., 2011), adsorption, separation by membranes (C. Zhao et al., 2013) and cryogenic methods (Kordylewski, Sawicka, & Falkowski, 2013; Peng, Zhao, & Li, 2012). Chemical absorption has been used for various industrial applications but especially for the capture of carbon dioxide application, it is in a state of development more advanced than other technologies. The main advantages of chemical absorption over other processes are high capture capacity and simple operational management (Peng et al., 2012). In addition, the desorption process that follows the absorption, needs less thermal energy compared to the other alternatives.

Although the process of chemical absorption is the method of carbon dioxide capture most used in the industrial sector, its performance depends largely on the cost and characteristic of the absorbent (composition, corrosivity, volatility, degradability and ease of recovery). Therefore, many solvents have been investigated for the absorption of carbon dioxide, including primary and secondary amines, such as monoethanolamine (MEA) and diethanolamine (DEA) (Han et al., 2011), respectively, potassium carbonate and aqueous solutions of sodium hydroxide (Chiang, Lee, & Liu, 2017; Han et al., 2011). Of the primary amines, MEA is considered the most economical of the commercial amines and has significant advantages such as high rate of absorption and the facility to absorb carbon dioxide from low concentration combustion gases. In addition, the MEA has the highest absorption capacity for acid gases, which means less requirement of absorbent (Ghosh, Kentish, & Stevens, 2009; Peng et al., 2012). However, its performance is limited by several factors that include, the high consumption of thermal energy in the regeneration of the absorbent, the degradation at high temperatures in the presence of oxygen, the formation of corrosive and toxic organic products, the high volatility (Du, Yuan, & Rochelle, 2017; Wang et al., 2011; B. Zhao et al., 2011) and the requirement of pretreatment processes for the elimination of sulfur dioxide (Thee et al., 2012).

Industrially, aqueous solutions of potassium carbonate have been used to absorb carbon dioxide. These solutions are the basis of the industrial process known as the Benfield process that is for the elimination of carbon dioxide in the synthesis of ammonia (Benson and Field, 1954; Benson et al., 1959). Aqueous solutions of potassium carbonate require less energy for regeneration. In addition, they present less corrosivity, less degradation and have a low environmental impact when compared with traditional amine-based absorbers (Anderson et al., 2013; Ghosh et al., 2009). However, the absorption rate of carbon dioxide in potassium carbonate is slow and it is necessary to add a catalyst that decreases the reaction time for this process. In this

sense, additions of boric acid have been studied as a catalyst during the process (Smith et al., 2012; Thee et al., 2012). Boric acid is environmentally benign, economically affordable and tolerant to oxidative and thermal conditions. It also has no significant influence on the vapor liquid equilibria (VLE) of carbon dioxide at low concentrations and no interaction with other minor components such as sulphuroxide in the flue gas (Endo et al., 2011; Ghosh et al., 2009; Hu et al., 2016; Thee et al., 2012).

In this work, the performance of the mixture sodium hydroxide and boric acid for the capture of carbon dioxide by chemical absorption was evaluated. The effect of sodium hydroxide and boric acid concentrations, and the temperature of operation on absorption process was determined. Additionally, a kinetic expression was developed for the absorption rate of carbon dioxide in the solution of sodium hydroxide and boric acid. Finally, a costs analysis of the process was carried out.

2. Materials and methods

2.1 Materials

The purity and suppliers of all chemicals used for the experimental work are given in Table 1. All chemicals were used as provided by the manufacturer without further purification or correction.

Table 1
Purity and suppliers of chemicals used for experimental work.

Chemical name	**Purity**	**Supplier**
*Sodium hydroxide (NaOH)	> 98.0 wt.%, Na_2CO_3 <0.9 as impurity	Dequímicos Ltda
*Boric acid (H_3BO_3)	> 98.0 wt.%	Dequímicos Ltda
Carbon dioxide gas (CO_2)	≥ 99.9 mol%	Cryogas
Air		
Water (H_2O)		

*Commercial grade reagents
Source: Data provide by manufacturing

2.2 Apparatus

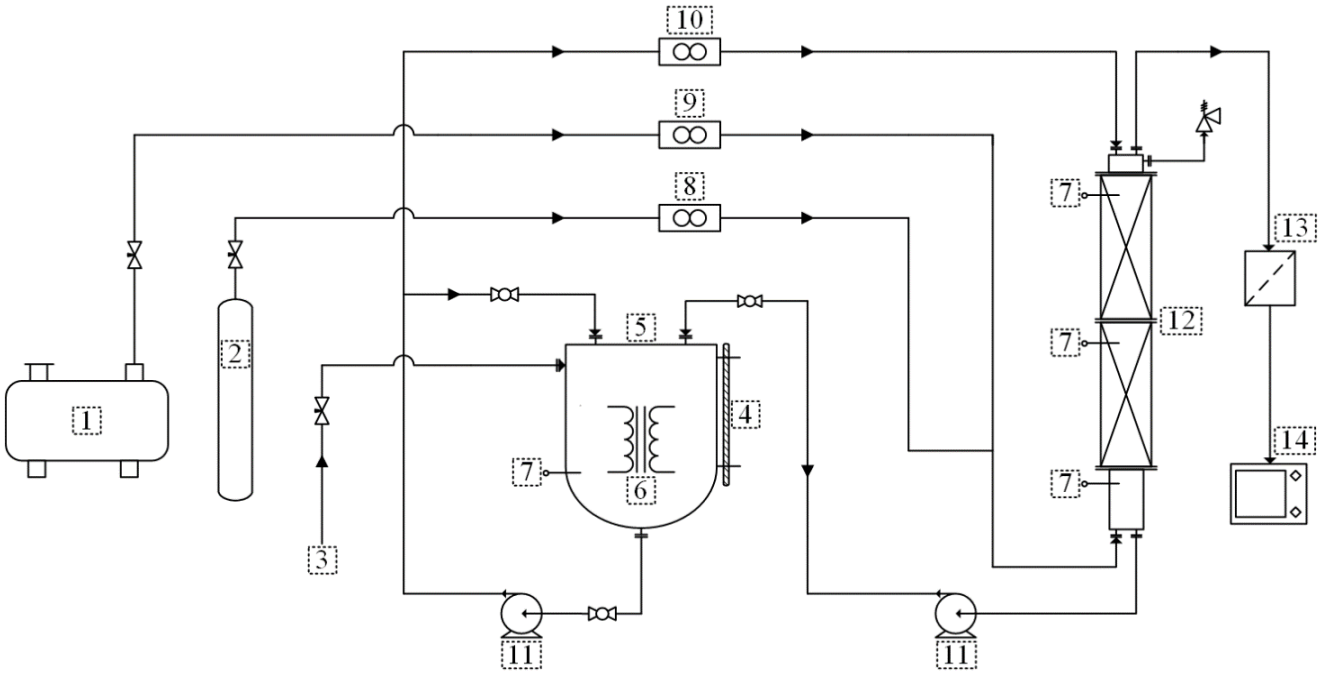

Figure 1. Diagram of the pilot absorption unit: (1) Air compressor, (2) CO_2 cylinder, (3) Absorbent inlet, (4) Level indicator, (5) Storage tank, (6) Electrical resistance, (7) Thermocouple, (8) CO_2 flow meter (9) Air flow meter, (10) Liquid flow meter, (11) Centrifugal pump, (12) Absorption column, (13) Maintenance unit, (14) Gas analyzer.
Source: The authors

Fig. 1 shows a diagram of the pilot absorption unit that was designed and built by the company Didacontrol S.A. The pilot absorption unit consists of a Pyrex glass column of 0.1 m inner diameter and 1.25 m height, randomly packed with Rasching Miniving rings (1/2 in) that have a high specific surface, which facilitates the interaction between a liquid phase and a gaseous mobile phase. The column was equipped with a gas inlet and a distribution space in the lower part; a liquid inlet and a distributor at the top; outlets for gas and liquid at the top and bottom, respectively. Furthermore, three thermocouples were located at the top, in the middle section and at the lower part of the column.

A maintenance unit (STNC) (located at the top outflow of the column), consisting of a regulating filter (STNC, model TW4000-04) and an air lubricator (STNC, model TL4000-04), was used to remove all the impurities (metal particles, dirt and condensed water) and to keep the operating pressure at a constant value. Then, a non-dispersive infrared-based gas analyzer (OUTEST, model HT-2000, range 0~9999 ppm) with a ± 50.0 ppm precision was also located at the top of the column to measure the CO_2 composition in the outlet gas from the absorber.

The other equipments in the pilot absorption unit include a storage tank, flow meters, an air compressor and centrifugal pumps. The storage tank consists of a cylindrical tube, which was made of stainless steel with 0.5 m inner diameter, 1.0 m height and with a storage capacity of 40 000 cm^3. The reservoir was equipped with an electrical resistance to adjust the temperature of the liquid solutions that enter the column. A thermocouple and a level indicator were located at the bottom of the tank to keep a constant control of the temperature and the level of the solution, respectively. The volumetric flow meters ALICANT SCIENTIFIC (model MCR-250LPM-D-DB15/5M and model MC-100LPM-D-DB15) and IFM ELECTRONICS (model SM6001) were installed in the air streams, carbon dioxide and liquid that feed the absorber, respectively. Centrifugal pumps (PEDROLLO, ALL-RED 135) were installed in the liquid streams that enter and leave the absorption column to pump the absorbent solution to the top of the column and to recycle the solution to the storage tank.

2.3 Experimental Conditions

The chemical absorption process was carried out at three temperature conditions (25°C, 35°C and 45°C), two concentrations of sodium hydroxide (5.0wt.% and 8.0wt.%) and one concentration of boric acid (1.0wt.%). The inlet concentration of CO_2 in the gas stream was 1.0% by volume for all the experimental runs. The concentration of CO_2 at the inlet was determined by the maximum concentration value that can be registered by non-dispersive infrared-based gas analyzer (9999 ppm). By several pre-tests, the liquid and inlet gas flow rates were determined to be 2.0 L/min and 40.6 L/min, respectively. The liquid/gas ratio was controlled at 0.05 to prevent flooding in the absorption column.

2.4 Experimental setup

For experimental runs, an absorbent solution was prepared in the feed tank at a given concentration. The absorbent solution was also heated to the desired operating temperature by electric heaters before being fed

into the column. The experiment began by introducing a gas mixture of clean air from compressor air, and CO_2 from a cylinder to the bottom of absorption column at a desired flow rate. After the operating temperature of the absorbent solution and the concentration of inlet CO_2 reach the steady state, the prepared liquid solution was pumped to the top of the column to create counter-current contact between gas and liquid. At the same time, the CO_2 analyzer are turned on. Gas and liquid stream properties (including pressure, temperature and flow) were continuously monitored via an online data logging system. The CO_2 composition in the outlet gas from the absorber was measured every 5 s and was also recorded automatically by a computer. The contact time of CO_2 with absorbent is about 20 s. The operating time of each experiment was 30 min, which was determined by several pre-tests, and guarantees that the system reaches the reaction equilibrium.

2.5 Kinetic rate equation

Based on differential analysis, a kinetic rate equation was considered for CO_2 capturing in a batch reactor and the kinetic parameter alpha (α) and K of Eq. (4) (shown later), were calculated by fitting experimental data to a kinetic model by using the Matlab® fsolve function.

3. Results and discussion

When the gaseous mixture formed by air and CO_2 was brought into contact with the caustic soda solution, the solubilization of the carbon dioxide occurred instantaneously due to the physical absorption process. (Darmana, Deen, & Kuipers, 2005). Subsequently, the chemical absorption took place when the aqueous CO_2 reacted with the NaOH present in the absorbent solution, the mechanism of reaction was explained by Fleischer et al., (Fleischer, Becker, & Eigenberger, 1996).

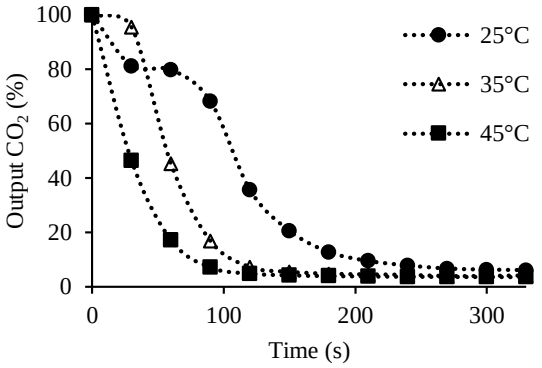

Figure 2. CO_2 absorption kinetics (NaOH 5%, without catalyst).

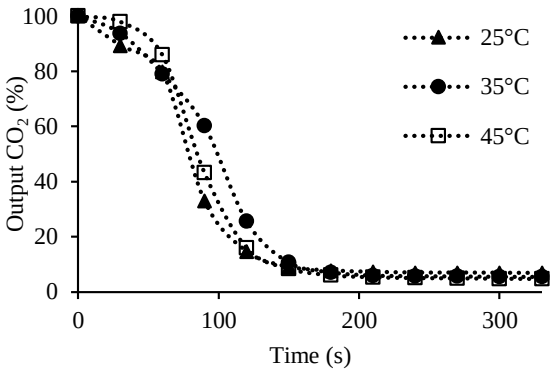

Figure 3. CO_2 absorption kinetics (NaOH 8%, without catalyst).

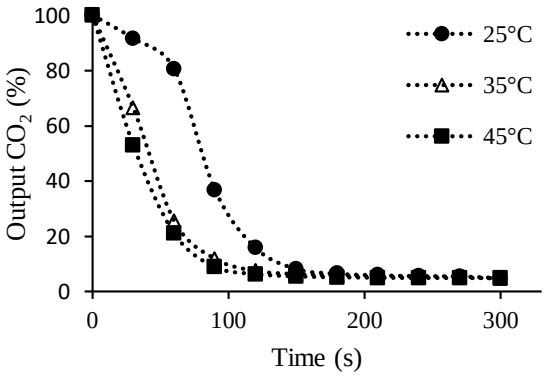

Figure 4. CO_2 absorption kinetics (NaOH 5%, with catalyst)

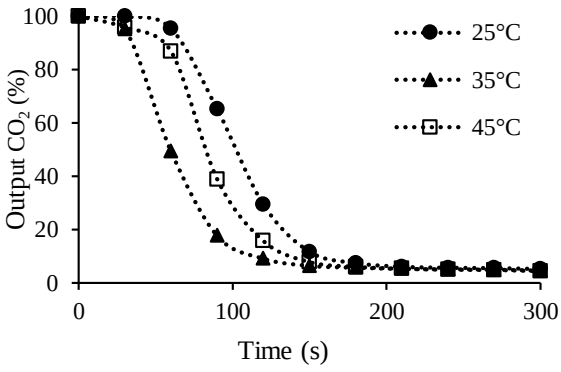

Figure 5. CO_2 absorption kinetics (NaOH 8%, with catalyst).

The decrease in the output composition of gaseous CO_2 was rapid for all the temperatures worked, obtaining the lowest values at 90, 120, 210 seconds for temperatures 45, 35, and 25°C, respectively (see Fig. 2). After these times, all the output concentrations tended towards a constant value, after 250 seconds, the output concentration of CO_2 coincided for temperatures of 35 and 45°C and was very close but not equal when the temperature was 25°C. This behavior can be attributed to the fact that when the operating temperature is close to 25°C the reaction kinetics becomes slower, making the time it takes to reach the final concentration obtained for the other two temperatures be greater.

For the three temperatures mentioned above, the reaction kinetics improved as the temperature of the absorbent solution was increased, indicating that the chemical absorption process was governed by the kinetic behavior; which is favored by a gradual increase in the temperature of the absorbent.

Fig. 3 shows the behavior of the CO_2 output composition in the absorption tower when the sodium hydroxide solution was in an 8% concentration (%wt.). This time, the lowest values of CO_2 output concentration were obtained around 150 seconds for the three temperatures. Contrary to what is observed in Fig. 2, in this situation it was not dominated by the kinetic behavior because there was no relationship between the temperature and the time it takes to obtain a certain concentration of CO_2. In this case, the values of concentration in the stability are quite close for the three temperatures, however, the final concentration values increase when the temperature of the solution decreases with sodium hydroxide, habitual behavior for an absorption. This indicates that by increasing the concentration of sodium hydroxide from 5%wt. to 8%wt. in the absorbent solution, the excess of OH- ions favors the aqueous CO_2 production causing an increase in the production of sodium carbonate (Yoo et al., 2013). Due to the high alkalinity of the aqueous solution only the sodium carbonate formation was considered in this study. (Hecht, 2013).

The final concentrations of CO_2 decreased, in all cases, when the concentration of sodium hydroxide was increased from 5% to 8%. This result supports the results obtained by G. Yincheng et al. (2011), who worked with aqueous solutions of sodium hydroxide with concentrations between 2%wt. and 10%wt., obtaining that at high absorber values, the removal of CO_2 is significantly favored. The explanation for this behavior is that the increasing concentration of absorbent produces a greater amount of available NaOH to be diffused towards the gas-liquid interface to react with CO_2 (Yincheng, Zhenqi, & Wenyi, 2011).

When 1% boric acid (%wt.) is added to the absorbent solution as a catalyst (Fig. 4), maintaining the conditions described in Fig. 2, the lowest values of carbon dioxide concentration began to be obtained at the 90, 95 and 120 seconds for the temperatures of 45, 35 and 25°C, respectively. As in Fig. 2, when the process was carried out with an absorbent solution at 5% sodium hydroxide, the chemical absorption is governed by the reaction kinetics, where an increase in the temperature leads to an increase in the reaction rate.

Analogously to Fig. 3, when the concentration of sodium hydroxide in the absorbent solution increased by three percentage points (up to 8%), the reaction kinetics ceases to be the phenomenon that governs the chemical absorption process (see Fig. 5).

It is verified that for both cases (Fig. 4 and 5), the boric acid accelerates the reaction rate fulfilling its work as a catalyst, because, the formation of bicarbonate is increased, which leads to an increase in the rate of CO_2 consumption. The support of this result is shown below.

The mechanism through which the boric acid accelerate the rate of reaction of the CO_2 is similar to the mechanism of the carbonic anhydrase to hydrate the CO_2 in the human body (Guo et al., 2011). The B(OH) reacts with the CO_2 to increases the formation of bicarbonate, as listed in the next reaction mechanism:

$$B(OH)_3 + H_2O \leftrightarrows B(OH)_4^- + H^+ \quad (1)$$

$$B(OH)_4^- + CO_2 \leftrightarrows B(OH)_4CO_2^- \quad (2)$$

$$B(OH)_4CO_2^- + H_2O \leftrightarrows B(OH)_3.H_2O + HCO_3^- \quad (3)$$

3.1 Kinetic model of the absorption of CO2 in a solution of Sodium Hydroxide

The reaction where the CO_2 is physically absorbed is a first order reaction. The next equation shows the rate absorption:

$$r_B = KC_B^{\propto} \quad (4)$$

Where: r_B is the rate of absorption, C_B is the molar concentrations of CO_2, K is the rate constant, and α the order of the reaction.

As noted in the 2.5 section, the rate constant K was calculated by fitting experimental data to a kinetic model, one rate constant was obtained for each temperature for each of the experiments (Fig. (2)-(5)). The algebraic equation that relates the rate of disappearance of a reagent with the concentrations of the species is called kinetic expression or rate law. The reaction rate constant K^*, was assumed to depend only on the temperature for the present study. The Arrhenius equation shows this relationship (Dormillore, 1996)

$$K^*(T) = A * e^{-\frac{E_a}{RT}} \quad (5)$$

Where: A pre-exponential factor, Ea is the activation energy (J/mol), R is the universal gas constant (8.314 J/mol.K) and T is absolute temperature (K)

Eq. (5) represents the kinetic expression obtained for this work, and the Table 2 summarizes the values of the parameters found for each of the experiments represented in fig. 2 to 5.

Table 2.
Values of the pre-exponential factor and activation energy of the study.

Figure	A	Ea (J/mol)
2	5.5085	14900
3	$2.72x10^{-03}$	-5176.06
4	$1.03x10^{-07}$	-32151.4
5	8.3028	15231.8

As mentioned above, when the concentration of the absorber was 5%, kinetics was the dominant phenomenon, obtaining values for the activation energy of 14900.05 J/mol and 32151.4 J/mol for runs with and without the presence of acid boric, respectively. The higher the activation energy, the more sensitive to temperature is the rate of disappearance, which explains why the curves present in Fig. 2 are much more distant from each other, when compared with the curves from Fig. 4. When a greater activation energy was obtained in the run performed without catalyst, the output composition decreased faster as the temperature was increased, compared to the run with the presence of a catalyst.

Although the simulation must be done with the concentration of aqueous CO_2 which can be calculated using Henry's law, in this work it was determined with the concentration of gaseous CO_2. Therefore, temperature effect by Henry is intrinsic in the constant K^* obtained by Arrhenius.

3.2 Preliminary cost estimation

The parameter that was calculated for this evaluation was the cost of the energy needed to absorb a certain amount of CO_2 / liter of absorbent solution. To carry out this analysis, initially, the amount of carbon dioxide absorbed during the experimental run was determined at a temperature of 45 ° C, an operating time of 150 s

and a load of sodium hydroxide and boric acid at 8% wt. and 1% wt, respectively. Subsequently, the hydraulic power of the pump that moves the solution towards the top of the absorber was calculated, taking into account that the volumetric flow of the absorber solution fed to the column was 2 L / min for all the runs. The hydraulic power was calculated by Eq. (6)

$$bhp = \frac{Q * \triangle P}{1714} \quad (6)$$

Where, bhp is expressed in hp, Q is the volumetric flow driven by the pump (gpm) and ∆ P are the load losses in the pipeline (psia).

Considering the previously mentioned operating conditions, it was found that 15.35g of CO_2 / L of absorbent solution was captured during the process. This result also indicates the maximum absorption capacity of that solution, because it was calculated at 150 s of operation (time in which it stabilized the concentration of carbon dioxide). In addition, it was determined that the hydraulic power required to drive 2 L / min of the aqueous solution of sodium hydroxide and boric acid was 0.00115 kW. It is worth mentioning that the energy consumption found is independent of the size and characteristics of the pump. According to the value of hydraulic power found and the cost of energy per kW.h, it was determined that the operating cost to capture 15g of CO_2 / L of solution was approximately 0.01953 COP.

4. Conclusions

The boric acid, used as catalyzer, enhanced in the rate of CO_2 capturing in the absorption process under the operation condition evaluated in this study. The removal of CO_2 was strongly dependent on the NaOH concentration, when the concentration of the absorbent was increased, the removal of CO_2 is highly favored, due the excess of OH^- ions, causing an increase in the production of sodium carbonate. When the NaOH concentration was 5%wt., the chemical absorption process was governed by the kinetic behavior, which is favored by a gradual increase in the temperature of the absorbent.

The liquid and inlet gas flow rates were determined to be 2.0 L/min and 40.6 L/min, respectively. The liquid/gas ratio was controlled at 0.05 to prevent weeping in the absorption column.

This study sets a precedent to continue investigating the use of chemical substances as catalysts in the process of absorbing carbon dioxide. The effect of the variation of boric acid concentration should be analyzed in future investigations.

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