Optimization of Carbon Dioxide Capture Process Parameters in Sodium Metaborate Solution

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ABSTRACT: *In this study, parameters affecting the carbonation reaction of carbon dioxide with sodium metaborate solutions were determined and optimized for a variety of process conditions. These parameters include reaction temperature, sodium metaborate concentration, carbon dioxide flow rate, and sodium metaborate/sodium hydroxide molar ratio. Two experimental designs were created for the carbonation reaction with different parametric ranges based on the solubility of the reactants. One of the designs contains high solubility of carbon dioxide and the other has high solubility of sodium metaborate. The modeling results exhibit a good agreement with the experimental values for the low-temperature design. The modeled conditions exhibit an optimal reaction temperature of 24.0±1.0 °C, a carbon dioxide flow rate of 300±10 mL/min, and a molar ratio of 1.23±0.03 mol $NaBO_2$/mol NaOH. The design conditions show that the rate of carbon dioxide consumption is 0.80 mol CO_2/min at optimum, which is coherent with the experimental mean value of 0.77 mol CO_2/min.*

KEYWORDS: *Carbonation reaction; Carbon dioxide capture; Experimental design; Sodium borohydride hydrolysis; Sodium metaborate recovery.*

INTRODUCTION

Over the past century, some sectors such as production industries of electricity, (petro)chemicals, food, fertilizer, transportation, and mining have been developed [1]. Although technological developments provide prosperity, they also force the reduction of sources to satisfy the demand. There are many approaches and studies to fill the gaps between the production yields in any of the sectors which are mentioned above and consumption of sources. In order to accelerate the production with minimum consumption rates of sources, energy has to be used efficiently, therefore, from past to present energy has been the most important phenomenon related to the needs of humans [2]. Because of the depletion of fossil fuels, alternative fuels which are renewable and environmentally friendly have attracted attention. Solar, wind, geothermal energies and biofuels are used to fulfill the energy demand [3]. Hydrogen (H_2) can also contribute to the alternative energy source phenomena. Hydrogen is not an energy source but is a good energy carrier [4,5]. In the last decade, sodium borohydride ($NaBH_4$) was found to be an efficient hydrogen carrier. Hydrogen gain of 10.6 wt.% has been reported for sodium borohydride hydrolysis [6]. Sodium metaborate ($NaBO_2$) is a waste by-product of sodium borohydride hydrolysis reaction [7,8] and is shown in Eq.(1).

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$$NaBH_4 + (2 + x_h)H_2O \rightarrow NaBO_2.x_hH_2O + 4H_2 + Q \quad (1)$$

This study envisages using the waste sodium metaborate for carbon dioxide storage. Carbon dioxide (CO_2) is generally the direct or indirect output of many production processes, burning fossil fuels and transportation. International Energy Agency (IEA) and Environmental Protection Agency (EPA) declare that global greenhouse gas emissions primarily consist of 76% of CO_2 by volume. Hence, it is a key player in associated global warming and climate change. Scientists have suggested that capture and storage of CO_2 is a mandatory step to save the environment [9].

It is emphasized in the literature that recycling processes have to be a part of energy-saving systems [10]. Carbonated substances can be produced by gas-solid reactions (dry carbonation) and gas-liquid (wet carbonation) reactions [11]. CaO, MgO, $Ca(OH)_2$, NaOH, $NaBO_2$ are some examples of solid chemical adsorbents [12–15]. The gas-liquid reaction system can be summarized as a gas reactant that partially or fully dissolves in a liquid phase and the active anions react with the cations in the solution [16–19]. Therefore, the yield of the carbonation reaction depends on the physical and chemical properties of solvent, solute, and gas. Carbonation reaction or solution temperature, (partial) pressure of the gas, concentration of solution and gas, type and acidity of solvent affect the physical and chemical properties of the reactants [20]. Experimental design serves to optimize the variables, therefore, it is a well-established tool for the determination of optimum conditions in multi-variable systems [21-23].

In order to determine the variables of carbonation reaction, the hydrolysis reaction of sodium borohydride also has to be considered with the initial conditions of hydrolysis such as $NaBH_4$ concentration, NaOH concentration, and amount of water that the hydrolysis takes place in [24]. $NaBH_4$ tends to hydrolyze rapidly when it contacts water. Therefore, in order to stabilize and control the rate of hydrolysis, alkaline media is preferred. NaOH is generally used for the alkalinity of the $NaBH_4$ solution. Theoretically, NaOH should be in the hydrolysis reaction media without any change. According to the hydrolysis reaction equimolar $NaBO_2$ also should be in the reaction media after the hydrolysis is completed. In literature different $NaBH_4$ concentration ranges are given for hydrolysis such as 1.38 M-8.77 M [25] and 0.26 M-2.63 M [26] and also the solubility of $NaBH_4$ at room temperature is given as 4.26 M (55 g$NaBH_4$/100 gH_2O) [27]. It is obvious that the concentration of $NaBO_2.x_hH_2O$ can differ according to the concentrations of $NaBH_4$ solutions. Also, another important parameter is the amount of water that is used for hydrolysis reaction as shown in Eq. (1).

The by-product may contain structural crystalline water (x_h) with the completion of hydrolysis. The hydration factor of $NaBO_2$ changes and stable forms of $NaBO_2.x_hH_2O$ can be given for x_h=4, 2, and 0 with regard to the amount of water [28]. Carbonation reaction temperature should also be considered according to the solubilities of the reactants because the carbonation reaction proceeds on the ions from $NaBO_2$ and CO_2 [29] in an aqueous media. The solubility curves of the reactants ($NaBO_2$ and CO_2) are given in Online Resource 2. The solubility of $NaBO_2$ is calculated according to Eq. (2) [30] and the solubility of CO_2 is sketched with the data from [31].

$$Solubility\ of\ NaBO_2(g/100g\ H_2O) = 0.915xT(K) - 245 \quad (1)$$

The solubilities of the reactants are strictly dependent on the temperature. The solubility of CO_2 exhibits a parabolic change between 0-30 ^oC temperature range and above 30 ^oC it decreases almost linearly. On the contrary, the solubility of $NaBO_2$ linearly increases with the increasing temperature. The low temperature will enhance the CO_2 capture due to the solubility of CO_2 but depending on the number of cations from the solids in solution, it will probably be low to trap the anions.

Especially the concentrations of sodium borohydride and sodium hydroxide directly affect the concentration profile of the residual solution. Another effect is the hydration factor of sodium metaborate which is related to the amount of water used for hydrolysis [32]. Subsequently focusing on the carbonation reaction, the solubilities of the reactants are bottlenecks due to their response to the temperature.

This work which was performed in Kocaeli University – Catalyst Research and Development Laboratory (2017/2018) – presents an extensive study of multivariable optimization for carbon dioxide capture in sodium metaborate solution. In virtue of preliminary experiments, the number of variables was reduced according to the

hydrolysis reaction conditions and physical properties of the reactants, the carbonation reaction variables were determined as carbonation reaction temperature, the concentration of sodium metaborate, the flow rate of carbon dioxide, and the molar ratio of sodium metaborate per sodium hydroxide. The experimental designs were configured with different boundary conditions of the variables due to the solubilities of the reactants according to the central composite design. The study emphasizes that the residual sodium borohydride hydrolysis reaction can be used as a reactant for the carbon dioxide capture process and this process can be modeled. In this study, in addition to the improvements on the capture process made, sodium metaborate and carbon dioxide which are described as wastes are also evaluated to produce value-added chemicals.

EXPERIMENTAL SECTION

Calcination

Three types of $NaBO_2$ were used as reactants for the carbonation reaction. The first one is commercial $NaBO_2.4H_2O$ (S0251, Sigma-Aldrich) and the others were obtained by calcination of commercial $NaBO_2.4H_2O$ at 400°C and 600 °C in the air environment. 0.4200±0.0005 g of commercial $NaBO_2.4H_2O$ with a calcination height of 1.0 cm was placed between two quartz wool plates to avoid the heterogeneous heat and mass transfer, due to volume expansion [28]. Then it was put in the middle of a quartz reactor (ID=1.0 cm) and the reactor was placed in a tubular furnace (Protherm PTF 12/20/250). For the calcination, pure air was passed through the commercial $NaBO_2.4H_2O$ for 2.5 h at a flow rate of F_{gas}= 300 mL/min.

Reaction tests

Solutions of calcined and commercial samples were reacted with CO_2 (99.99 %) in a semi-batch bubble reactor. The volume of the reaction medium was kept constant at 100 mL. The gas mixture flow rate of F_{total}=600 mL/min (F_{N2}=500 mL/min and F_{CO2}=100 mL/min) at varying temperatures. The experimental setup is given in Online Resource 1.

Carbonation conversion was calculated by using Equation (3) at the end of 150 min for the reaction time for all fresh sample and the difference of CO_2 amounts before and after the reaction were obtained by online data collection from CO_2 analyzer (TRL-CO_2, 2290, USA). The analyzer output shows the number of CO_2 molecules that pass across the infrared detector and were assigned an arbitrary unit (a.u.), therefore the output number of molecules and molar flow rates were correlated for further mathematical calculations.

$$\text{Carbonation conversion}(x) = \left[\frac{\int_{t=0}^{t=t} f\left(n_{CO_{2,in}}\right) - \int_{t=0}^{t=t} f\left(n_{CO_{2,out}}\right)}{\int_{t=0}^{t=t} f\left(n_{CO_{2,in}}\right)}\right] \quad (3)$$

n : moles of CO_2

The amounts of CO_2 in the feed and reaction exit streams were translated into mathematical functions which were related to the flow rates of the streams by regression. Molar flow rates were calculated by using the trapezoidal rule. At least three repeats were performed for the conversion values, and maximum or minimum data were discarded according to the statistical error analysis.

Characterization

The solutions were evaporated at 70 °C to avoid the decomposition and the remaining solid was characterized on X-Ray Diffractometer (XRD) (Rigakku, Miniflex 2, Japan). 2θ values were scanned from 10° to 80° with a step size of 0.02°/min using CuKα radiation (λ = 0.15418 nm) at 45kV/40mA.

Experimental Design

Face Centered Composite (FCC) design was used to determine the optimal conditions and to study the effects of four variables; reaction temperature (°C), the concentration of $NaBO_2$ (mol/L), the flow rate of CO_2 (mL/min), and the molar ratio of $NaBO_2$/NaOH (mol/mol)) on the response of carbonation conversion. Two different experimental design patterns (ED1 and ED2) were constructed with respect to the solubility data of the reactants. From the preliminary experimental conditions (data not shown, [33]), process variables and the ranges were determined and the summary of actual values were given in Table 1. The statistical analysis was performed using Design Expert Statistical Software package 9.0.6.2 (Stat Ease Inc., Minneapolis, USA).

Table 1: Experimental design summary of ED1 and ED2.

Design Summary (ED1)					
Study Type	Response Surface		Runs	30	
Design Type	Central Composite				
Design Model	Quadratic				
Factor	Name	Units	Type	Minimum	Maximum
A	Temperature	^{o}C	Numeric	0	30
B	Concentration	M	Numeric	0.1	0.3
C	CO_2 flow rate	mL/min	Numeric	100	500
D	$NaBO_2$/NaOH	Molar ratio	Numeric	0.2	1.8
Design Summary (ED2)					
Study Type	Response Surface		Runs	30	
Design Type	Central Composite				
Design Model	Quadratic				
Factor	Name	Units	Type	Minimum	Maximum
A	Temperature	^{o}C	Numeric	25	75
B	Concentration	M	Numeric	0.4	2.0
C	CO_2 flow rate	mL/min	Numeric	100	500
D	$NaBO_2$/NaOH	Molar ratio	Numeric	0.2	1.8

RESULTS AND DISCUSSION

Determination of process variables

The following experiments were performed to determine the effects of possible variables (concentration of $NaBO_2$, calcination temperature (effect of hydration factor), carbonation reaction temperature, the concentration of NaOH, and interaction of OH^- ion). In Figure 1, the output of the analyzer for CO_2 capture is given for the observation of the effect of hydration factor and NaOH.

The carbonation reaction temperature was kept constant at 25 ^{o}C and the total flow rate was 600 mL/min (16.67 % vol.CO_2/vol.N_2). It is clear that total CO_2 capture is affected by the amount of $NaBO_2$ and NaOH ions. The output curves overlap for the carbonation reactions of hydrated and dehydrated $NaBO_2$ solutions because at the reaction conditions numbers of total ions are equal. This result indicates that the volume from the hydration factor is negligible and therefore the hydrated $NaBO_2$ was considered for further discussion. The dilute solution easily reaches completion or an equilibrium almost in 100 min with the minimum CO_2 removal. From Fig. 1, it can be speculated that the carbonation reaction exhibits a diffusion-controlled reaction mechanism. This result also supports the importance of the carbonation reaction temperature related to the solubility of the reactants. The repeatability of an experiment is one of the most important requirements for stability. Fig. 2 shows the repeatability of the carbonation reaction.

As can be seen in Fig. 2, the maximum CO_2 consumption was achieved with 0.04 moles of $NaBO_2$ at 25 ^{o}C, and higher concentrations of $NaBO_2$ have not increased CO_2 consumption. In literature, the solubilities of $NaHCO_3$, Na_2CO_3, and NaOH are given as 10g/100g H_2O, 23g/100g H_2O, and 100g/100g H_2O, respectively at 25 ^{o}C. During the experiments with 0.05 moles and 0.075 moles of $NaBO_2$, nucleation and crystallization in the solution were observed with the naked eye, probably as a result of the formation of less soluble product(s). As the next step of characterization, the effect of carbonation reaction temperature was investigated for 0.02 moles of $NaBO_2$ and the result is given in Fig. 3.

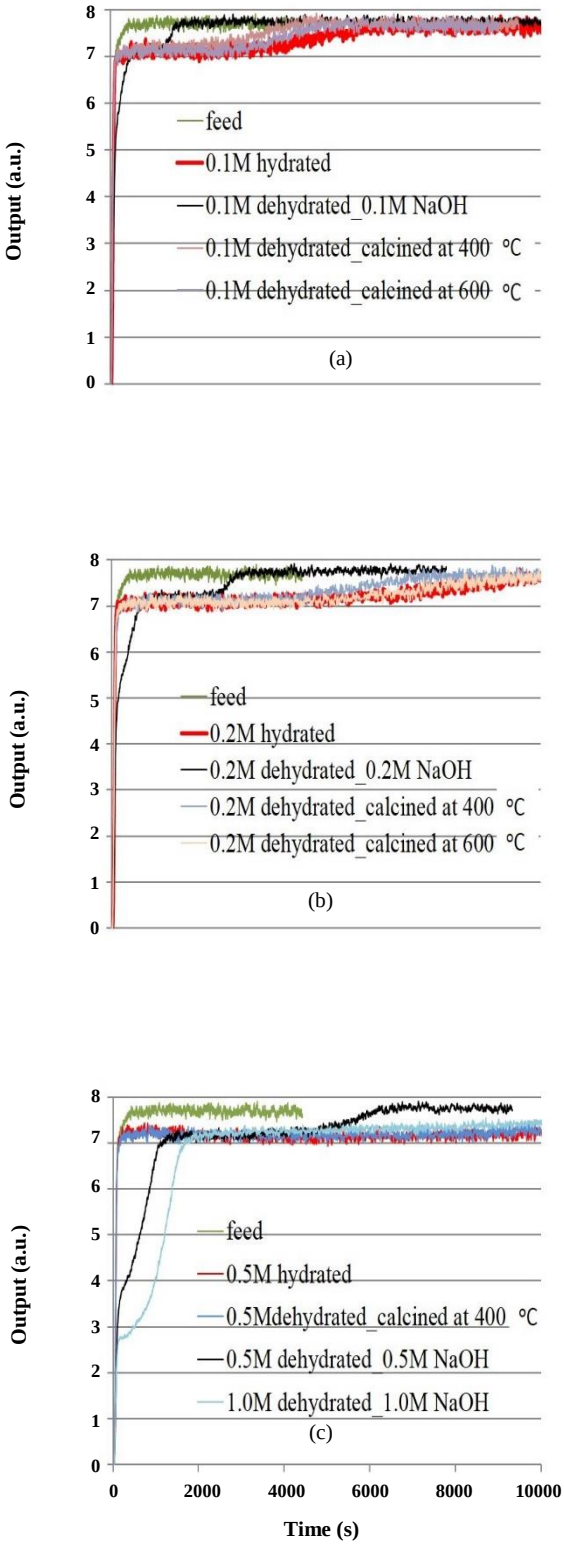

Fig. 1: Time on stream of carbonation reaction a) 0.1 M b) 0.2 M c) 0.5 M.

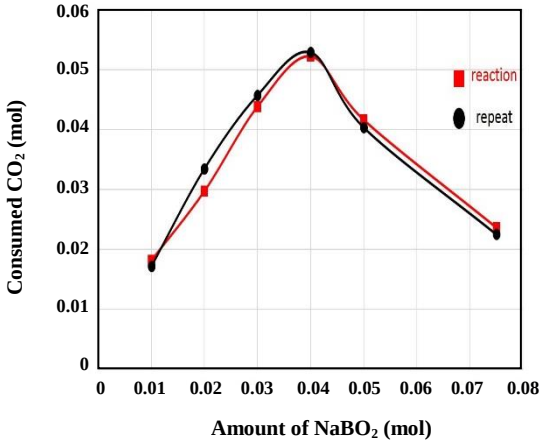

Fig. 2: Repeatability test of carbonation reaction.

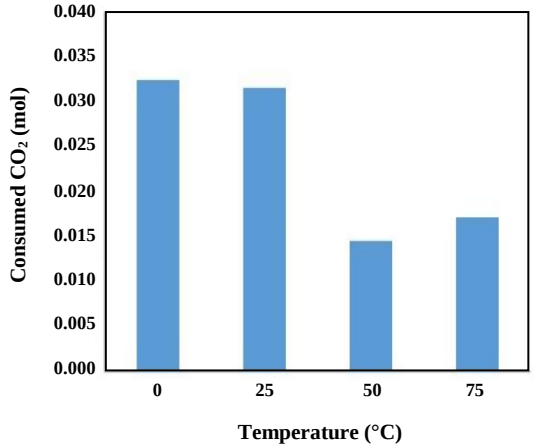

Fig. 3: Temperature dependency of CO_2 consumption.

Despite the low solubility of $NaBO_2$ at low temperatures, CO_2 capture was obtained more efficiently at low temperatures (0-25 °C) due to the solubility of the gas. This result suggests that the concentration of $NaBO_2$ can also be a dominant parameter especially at high temperatures, so it is concluded that $NaBO_2$ and CO_2 concentrations can affect the consumption yield of CO_2 simultaneously.

As it is reported in the literature and mentioned above, NaOH is used to suppress the hydrolysis reaction of $NaBH_4$. In this situation, additional ions such as Na^+ and OH^- are present in the solution and these ions can also react with the available reactants. The possible reactions of the reactants can be given by the following Equations (4-10);

$$NaBO_2 \rightarrow Na^+_{(aq)} + BO^-_{2(aq)} \quad (2)$$

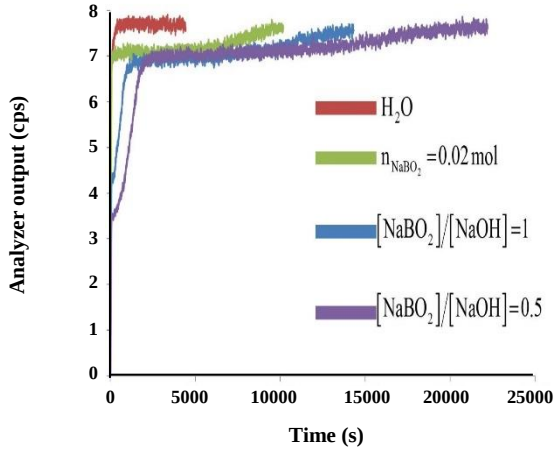

Fig. 4: Effect of NaOH concentration on carbonation reaction.

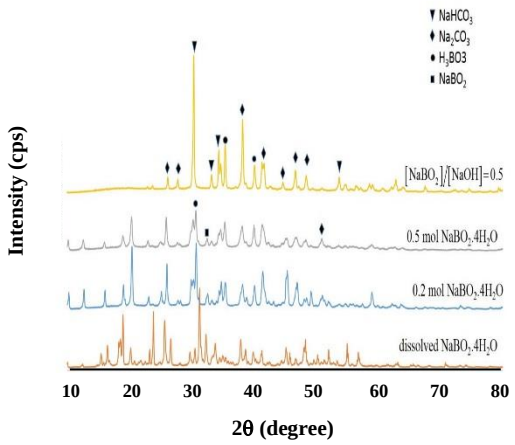

Fig. 5: XRD spectrum of the solids from carbonation reaction.

$$2H_2O \rightarrow H_3O^+_{(aq)} + OH^-_{(aq)} \quad (5)$$

$$CO_{2(g)} + H_2O \leftrightarrow H_2CO_3 \leftrightarrow \quad (3)$$

$$H^+_{(aq)} + HCO^-_{3(aq)} \leftrightarrow 2H^+_{(aq)} + CO^-_{3(aq)}$$

$$CO_{2(g)} + H_2O \leftrightarrow H_2CO_3 \leftrightarrow 2H^+_{(aq)} + CO^-_{3(aq)} \quad (7)$$

$$Na^+_{(aq)} + HCO^-_{3(aq)} \rightarrow NaHCO_{3(k)} \quad (4)$$

$$2Na^+_{(aq)} + CO^-_{3(aq)} \rightarrow Na_2CO_{3(k)} \quad (5)$$

$$H_3O^+_{(aq)} + BO^-_{2(aq)} \rightarrow H_3BO_{3(k)} \quad (6)$$

In Figure 4, the behavior of CO_2 (time on stream data) is given related to additional Na+ ion where the carbonation reaction is performed at 25 ºC with constant flow rate (F_{total}=600 mL/min) by defining the ion concentration in terms of molar ratio of $NaBO_2$ to NaOH as $NaBO_2$/NaOH.

It is clear that the increase in the amount of NaOH enhances the carbonation yield but the reaction ends in a longer time period. To associate the Equations (4-10) and Fig. 4, it is concluded that the presence of additional ions in the solution affects the carbonation yield. Therefore another parameter defined as the molar ratio of $NaBO_2$ to NaOH should be investigated.

The solutions are dried at 70 ºC to avoid the decomposition of the products at the end of carbonation reactions and XRD spectrum of selected precipitates is given in Fig. 5. For comparison, commercial $NaBO_2.4H_2O$ was also dissolved in distilled water and the same drying procedure was followed.

$NaHCO_3$ (PDF No. 99-100-8785), Na_2CO_3 (PDF No. 99-101-1517), and H_3BO_3 (PDF No. 99-100-9191) are identified from the spectrum. The following Equation (11), can be proposed with its stoichiometric constants with respect to the products of the carbonation reaction in an aqueous media.

$$3NaBO_2 + 2CO_2 + 5H_2O \leftrightarrow \quad (11)$$
$$Na_2CO_3 + NaHCO_3 + 3H_3BO_3$$

It is obvious that the carbonation reaction of $NaBO_2$ exhibits a complex system depending on the process variables. The process variables, in other words, reaction parameters, such as temperature, amount of CO_2, $NaBO_2$, and NaOH affect each other and also the CO_2 consumption. Experimental design is a useful tool to optimize the process variables. In this situation, $NaBO_2$ concentration, carbonation reaction temperature, CO_2 flow rate, and $NaBO_2$/NaOH molar ratio were selected as variables and CO_2 consumption was the response of the designs.

Experimental Design

Two series of experimental designs were configured according to the solubility data and the temperature effects. Basically, experimental design 1 (ED1) consists of low-temperature effects and experimental design 2 (ED2) contains high-temperature effects on the carbonation reaction of $NaBO_2$. One of the independent variables is the temperature (denoted as A) and the range is determined as 0-30 ºC for

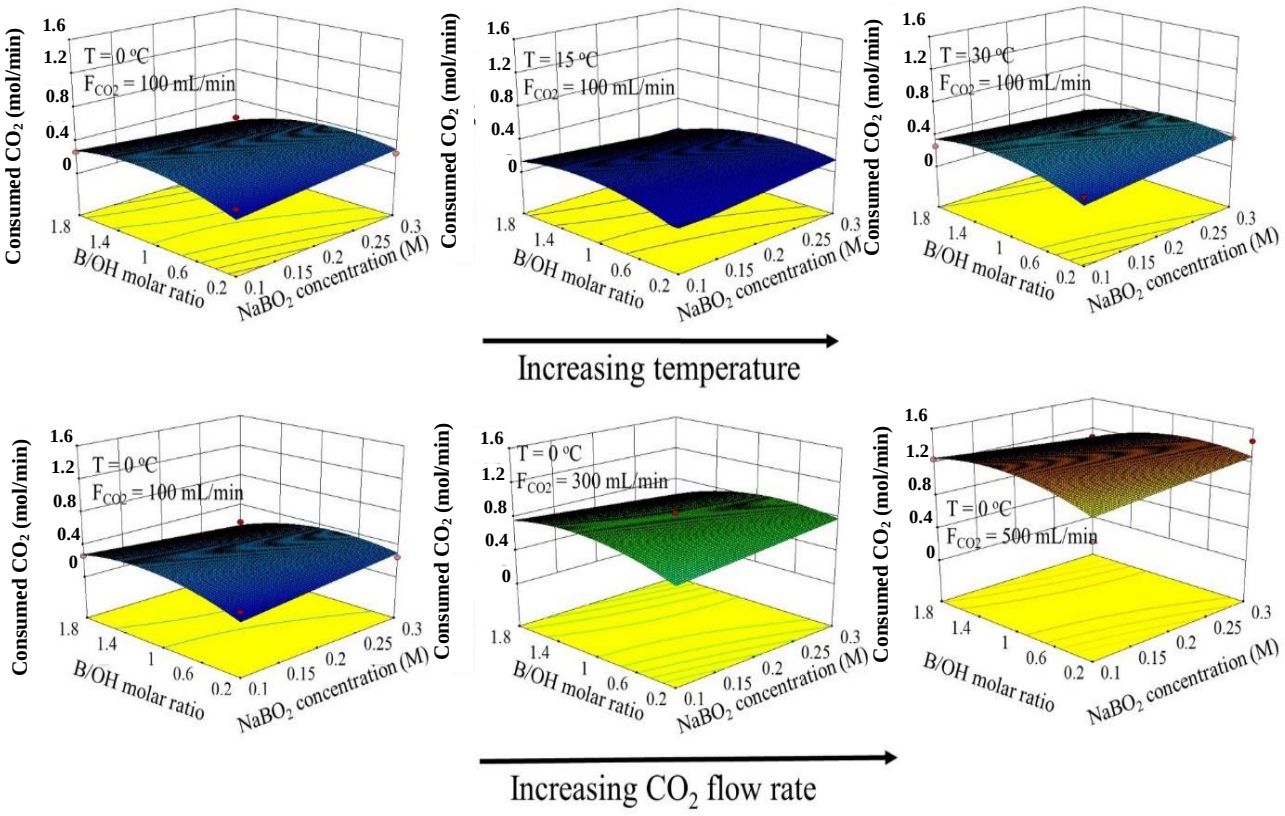

Fig. 6: Response surface plots and their contour plots for CO_2 consumption versus individual effects of the variables of ED1.

ED1 where the solubility of CO_2 exhibits a parabolic decrease with the increasing temperature (Figure 2). The temperature range is defined as 25-75 °C for ED2 where the solubility of CO_2 exhibits a linear change. Related to the solubility curve of $NaBO_2$ (denoted as B) as presented in the same Figure, the ranges are defined as 0.1-0.3 M for ED1 and 0.4-2.0 M for ED2. The variables, CO_2 flow rate (denoted as C) and $NaBO_2$/NaOH molar ratio (denoted as D), are defined in the same boundaries for both ED1 and ED2, 100-500 mL/min and 0.2-1.8, respectively. The molar ratio values demonstrate the real conditions of the hydrolysis reaction of $NaBH_4$. A central composite design is used with the controlled variables, giving 30 cases shown in Online Resource 3 with the response of CO_2 consumption (denoted as Y) of ED1.

The relationship between the independent variables was analyzed by analysis of variance (ANOVA) and is given in Online Resource 4. The p-value of a variable indicates that the variable in the model is significant. In this case, the terms; Y; A, C, BD, A^2, and D^2 were significant. The predicted polynomial model with R^2; 0.9359 obtained for Y is expressed according to the following Equation (12) in terms of actual factors:

$$\text{Consumed } CO_2 \text{ (mol/mL)} = 0.69 + 0.033A + 0.49C - 0.087BD + 0.18A^2 - 0.152D^2 \quad (12)$$

The individual effects of $NaBO_2$ concentration and molar ratio variables which are in first-order were determined as insignificant terms. However, interestingly the product of the variables (B*D) term is a significant term. This can be expressed by the interaction of these variables since the ions from NaOH are also active for CO_2 consumption. In Figure 6 the effects of process variables are given for three levels where the response is the consumption rate of CO_2.

It is concluded that the temperature had a relatively low effect on the response within the selected range but the effect of the flow rate on the CO_2 consumption was extreme at a constant temperature. The design of ED2 is given in Online Resource 5.

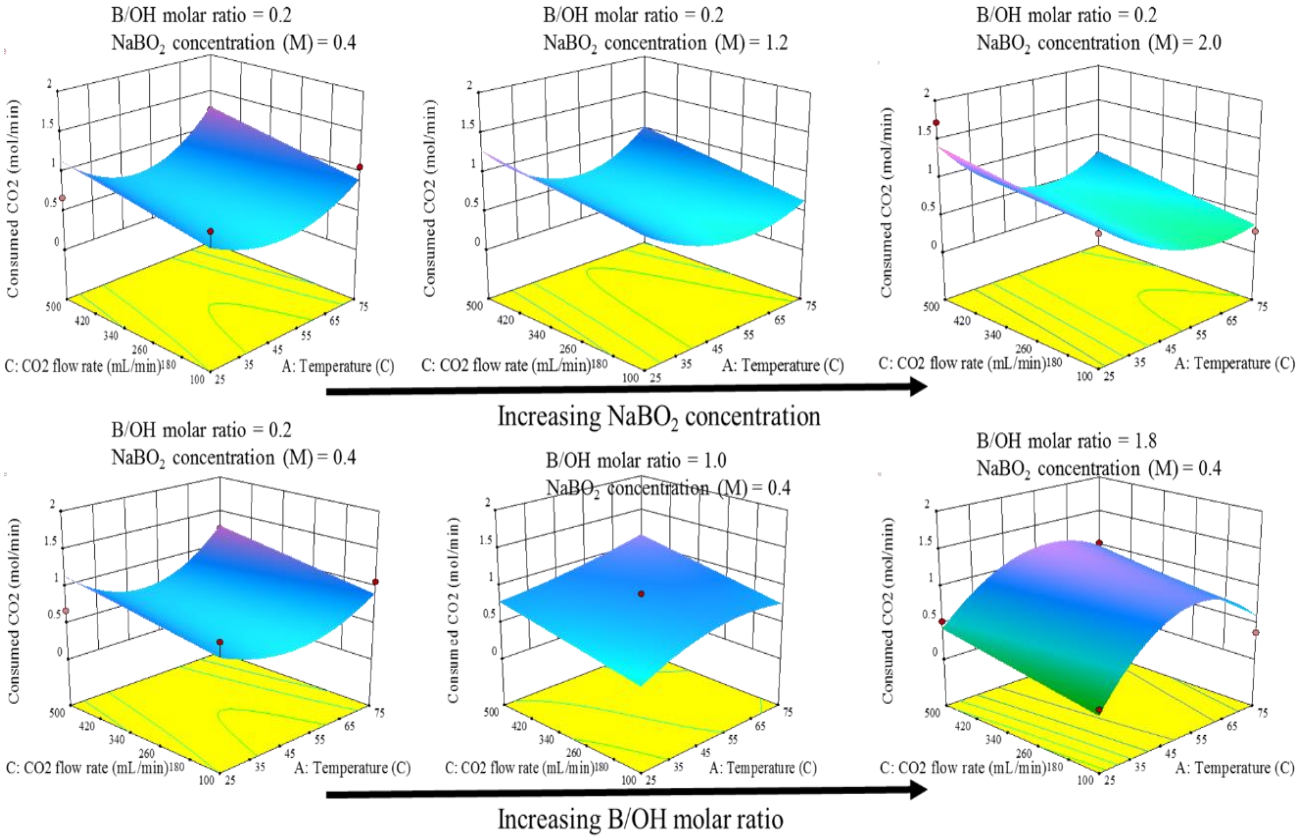

Fig. 7: Response surface plots and their contour plots for CO_2 consumption versus individual effects of the variables of ED2.

The same procedure of ED1 was applied to the statistical determination of ED2 and a significant model was derived with a p-value of 0.0034 and R^2 value of 0.6901. The ANOVA of ED2 is given in Online Resource 6. The response, Y, is expressed with the following Equation (13) in terms of actual factors:

$$\text{Consumed } CO_2 \text{ (mol/mL)} = \quad (13)$$
$$0.79 - 0.031A - 0.064B + 0.072C$$
$$+0.24D - 0.20AB + 9.2\times10^{-3}AC + 0.11AD$$
$$-0.073A^2 + 0.11A^2C - 0.50A^2D$$

According to Equation (13) it can be speculated that the higher consumption rates can be achieved at high temperatures only if the flow rate is high. However, it is known physically that the variables temperature and flow rate significantly affect the consumption rate due to the solubility of CO_2. Based on the graphical optimization it is also concluded that high CO_2 consumption was achieved when the temperature and flow rate were low i.e. 30 °C and 315 mL/min. When the temperature was kept constant the optimization did not exhibit a reasonable response above 315 mL/min due to the interaction between the variables. By comparing the results, the carbonation reaction was found to proceed more effectively at the low-temperature range (25-50 °C) and low flow rate range (100-250 mL/min).

In Fig. 7 the effects of process variables are given for three levels where the response is the consumption rate of CO_2.

As it can be seen in Figure 7, increasing $NaBO_2$ concentration at a constant molar ratio of B/OH=0.2 causes a sharp decrease at high-temperature levels due to the low solubility of carbon dioxide at high temperatures. An increase in molar ratio exhibits a gradually increasing trend for CO_2 consumption at mild carbonation reaction temperatures, T=50-55 °C. A drastically reduced carbon dioxide consumption was observed at the edge points of temperature and flow rate. As expected, when the carbonation reaction temperature has increased the consumption of carbon dioxide was also increased due to the solubility of $NaBO_2$. In this case the low solubility of

Table 2: Comparison of theoritical and experimental results of optimized conditions of carbonation reaction

Variable				Response		Deviation
A	B	C	D	$Y_{theoritical}$	$Y_{experimental}$	
Temperature	$NaBO_2$ concentration	CO_2 flow rate	$NaBO_2$/NaOH	Consumed CO_2	Consumed CO_2	
(^oC)	(M)	(mL/min)	(mol/mol)	(mol/min)	(mol/min)	(%)
25	0.2	300	1.26	0.80	0.77	3.8
40	0.9	300	1.20	0.82	0.73	10.9

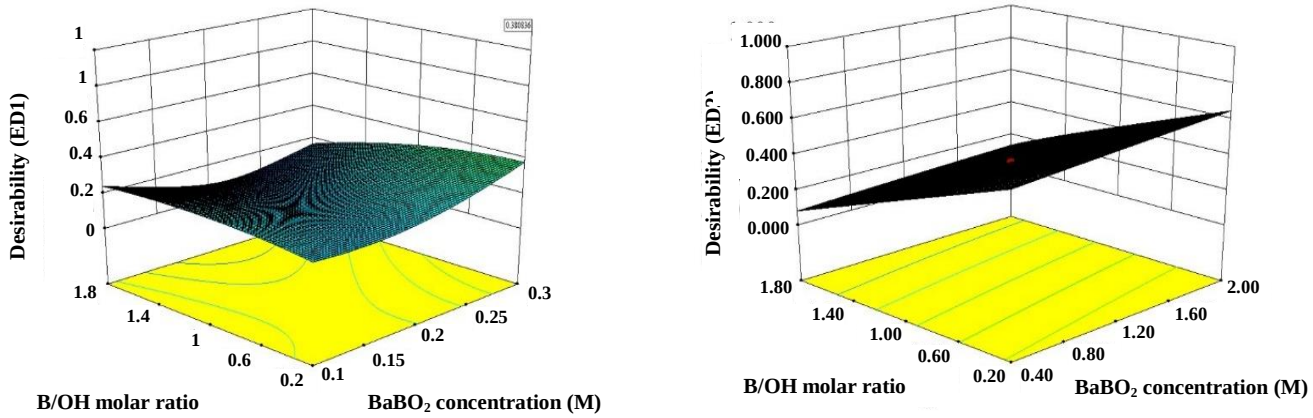

Fig. 8: Desirability plot of optimized process variables (T=25 ^oC, F_{CO2}=300 mL/min).

carbon dioxide at high temperatures also affects the carbonation yield. In order to compare and optimize the multivariable system, the graphs of desirability functions are given in Fig. 8.

In order to obtain a maximum CO_2 consumption at high-temperature levels, the flow rate should be high for ED1, whereas at low temperatures, $NaBO_2$ concentration should be at the maximum solubility level. It seems that at high temperatures the concentration does not affect the desirability. The desirability of ED2 shows that at low carbonation reaction temperatures and low flow rates higher carbonation yields can be obtained. In both cases (ED1 and ED2), the molar ratio of $NaBO_2$ and NaOH has to be balanced and this result can lead to optimizing the NaOH solution which is used for suppressing the hydrolysis reaction of $NaBH_4$. High flow rates of CO_2 can also be evaluated for both cases. Carbonation yield can be affected by the selected variables therefore boundary conditions should be determined carefully. In order to validate the results, two different temperatures were selected for the carbonation reaction, 40^{o}C, where the hydrolysis reaction performs, and 25^{o}C, where the maximum consumption rate can be obtained. Theoretical and experimental results are given in Table 2. Experimental results are the means of three experiments without any elimination of data.

As mentioned before, ED1 is a more robust design than ED2, thus the deviation from the desired value is lower than the one obtained via ED2.

CONCLUSIONS

In the present work, the carbonation reaction of sodium metaborate solution was studied. The main intention was to determine and optimize the carbonation reaction parameters and conditions. Four different parameters such as reaction temperature, sodium metaborate concentration, CO_2 flow rate, and $NaBO_2$/NaOH molar ratio were determined. The central composite design was used to optimize the process variables. Related to the solubility data of the reactants two different designs were applied to the process by changing the boundary conditions. The results showed that the process variables have significant influences on the carbonation yield. The results are summarized as follows;

● The hydration factor of sodium metaborate does not make any difference in the carbonation reaction pathway or yield. The important parameter is the number of reactive ions.

● Increasing concentrations of $NaBO_2$ and NaOH enhances the carbonation yield, but the reaction ends in a longer time period, therefore the molar ratio of $NaBO_2$ to NaOH was optimized and determined to be about 1.23±0.03 (mol/mol).

● Two different ranges of temperature were investigated due to the solubilities of the reactants. The process was more robust in lower temperatures than the higher temperatures and the optimum temperature was determined as 24.0±1.0 ºC for carbonation reaction. The flow rate of CO_2 was also 300±10 mL/min for all conditions.

● All parameters of the carbonation process interact, thus a small change can differ the yield significantly. Therefore, an optimum condition needed to be selected. Robust design can be achieved in a low-temperature range with a considerable deviation from the carbonation yield mean value.

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