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Taguchi application on sodium bicarbonate production using modified Solvay process

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Abstract. Sodium bicarbonate ($NaHCO_3$), known as baking soda, can be used as a baking powder, disinfectant, fire extinguisher, mild disinfectant, antiseptic to help prevent infections, acid neutralizer, cleaning agent, odor control, and antacid to treat acid indigestion and heartburn in medical and health use. Sodium bicarbonate is commonly produced using the Solvay process that uses brine (seawater), ammonia (NH_3) gas, and carbon dioxide. To reduce the costs of sodium bicarbonate production, this study used ammonium hydroxide (NH_4OH) solution, a by-product of sodium cyclamate factory, to replace the NH_3 gas. Hence, this modified Solvay process is more environmentally friendly. Ammonium hydroxide is a weak base suitable to produce food-grade sodium bicarbonate because it does not contain heavy metal impurities. The following parameters were studied and optimized namely reaction time, NaCl concentration, and CO_2 flow rate. The appearance of the microstructure, particle size, percentage of elements, and phase composition of the material was examined X-Ray Diffraction (XRD) and Field Emission Scanning Electron Microscopy with Energy Dispersive X-Ray Spectroscopy (FESEM-EDX).

Keywords: ammonium hydroxide; food-grade baking soda; modified Solvay; sodium bicarbonate; Taguchi;

1. Introduction

Sodium bicarbonate ($NaHCO_3$) is generally produced through the Solvay process, which consists of reactions with brine (sodium chloride), ammonia (NH_3), and carbon dioxide [1]. The production of sodium bicarbonate by the Solvay process gives the best results in terms of technological development and economic costs [2]. The Solvay process was originally developed for sodium bicarbonate production, wherein a concentrated brine solution is reacted with NH_3 gas and carbon dioxide to form soluble ammonium bicarbonate, which reacts with sodium chloride to form soluble ammonium chloride sodium bicarbonate precipitate as shown in equations (1) to (4) [3].

$$NaCl + H_2O + NH_3 \leftrightarrow NaCl + NH_4OH \quad (1)$$

$$2\ NH_4OH + CO_2 \leftrightarrow (NH_4)_2CO_3 + H_2O \quad (2)$$

$$(NH_4)_2CO_3 + CO_2 + H_2O \leftrightarrow 2\ NH_4HCO_3 \quad (3)$$

$$2\ NH_4HCO_3 + 2\ NaCl \leftrightarrow 2\ NaHCO_3 + 2\ NH_4Cl \quad (4)$$

Despite its wide use, NH_3 gas production is expensive, hence will increase the production cost of sodium bicarbonate. In this study, NH_3 was replaced with ammonium hydroxide (NH_4OH), a by-product of the sodium cyclamate (artificial sweetener) industry. The NH_4OH is cheaper than NH_3. Ammonium hydroxide is a weak base that usually does not contain heavy metals impurities, therefore this process is more environmentally friendly and suitable to produce food-grade sodium bicarbonate.

The chemical reaction to produce sodium bicarbonate from NH_4OH is shown in equation (5).

$$NH_4OH_{(aq)} + CO_{2\,(g)} + NaCl_{(s)} \leftrightarrow NaHCO_{3\,(s)} + NH_4Cl_{(aq)} \quad (5)$$

The Solvay process is usually carried out in a bubble column reactor using two stages of gas input (CO_2 and NH_3) [4]. This bubble column reactor can produce high-purity sodium bicarbonate [5]. In this study, 20% NH_4OH solution was used instead, therefore an NH_3 column reactor was not required. The NaCl powder was diluted in NH_4OH solution, then being transferred to the carbonization column. Hence, the process of sodium bicarbonate production using NH_4OH was simpler. The Taguchi method of experimental design was used to study and optimize the influence of process parameters on sodium bicarbonate production [6][7]. The parameters studied and optimized in this study were reaction time, NaCl concentration, and CO_2 flow rate.

2. Experimental Evaluation

In this study, the materials used were NaCl (sodium chloride), NH_4OH (ammonium hydroxide) derived from a by-product of sodium cyclamate (artificial sweetener) factory at Golden Sari Co. Ltd. (Lampung, Indonesia), and CO_2 (carbon dioxide) gas from Aneka Gas Co. Ltd. (Indonesia). The characteristics of the NaCl used are presented in Table 1.

Tabel 1. Characteristics of NaCl.

Element	Na	Mg	Al	P	S	Cl	K	Ca
Concentration (%)	15.727	0.482	0.253	0.217	0.527	81.467	0.184	0.925

For sodium bicarbonate production, the Taguchi experimental design was applied to study the following parameters: reaction time, NaCl concentration, and CO_2 flow rate. Each parameter had four levels as shown in Table 2. The experimental design in the Taguchi analysis used the L16 (4^3) orthogonal array matrix which was generated using the Minitab software. The results were analyzed with the Taguchi method using signal-to-noise (S/N) ratio of 'larger is better", which was calculated with equation (6):

$$S/N = -10\ log\ log\ \left(\frac{1}{n}\sum_{i=1}^{n} \frac{1}{{y_i}^2}\right) \quad (6)$$

where *Yi* is the responses for the given factor level combination and *n* is several responses in the factor level combination.

The first stage in this work was dissolving NaCl into 2 L of 20% NH_4OH solution. The solution was stirred using an overhead stirrer (IKA EUROSTAR 60 digital overhead stirrer, IKA Corp., Germany) at a speed of 1500 rpm until NaCl was completely dissolved. The mixed solution was then filtered using polypropylene filter paper. The filtered solution was transferred into a reactor with an inner diameter of 20 cm and a height of 20 cm (Figure 1). The CO_2 gas was flowed into the reactor using a hose with an inner diameter of ¼ inch near the bottom of the reactor. During the reaction, the solution was stirred at a speed of 1500 rpm using a 4-bladed propeller stirrer (IKA R1342, IKA Corp., Germany).

Tabel 2. Taguchi experimental design and results.

Run	Experimental design			Results
	Operation time (min)	**CO_2 flow rate (L/min)**	**NaCl (g)**	**Sodium bicarbonate (g)**
1	10	2.5	300	105.3984
2	10	5	400	129.6772
3	10	7.5	500	0.0001
4	10	10	550	0.0001
5	15	2.5	400	70.0452
6	15	5	300	237.9858
7	15	7.5	550	190.0982
8	15	10	500	47.6508
9	20	2.5	500	182.6441
10	20	5	550	153.5589
11	20	7.5	300	0.0001
12	20	10	400	45.5836
13	25	2.5	550	0.0001
14	25	5	500	0.0001
15	25	7.5	400	2.8428
16	25	10	300	40.8152

After the reaction, sodium bicarbonate was separated from the NH_4Cl solution using filter paper. The solid residue (sodium bicarbonate) was dried at 90 °C. The sodium bicarbonate product was characterized using an analytical mass balance, X-ray Diffraction analysis (X'Pert3 Powder XRD, PANalytical Corp., the Netherlands), and Field Emission Scanning Electron Microscope−Energy Dispersive X-ray Spectroscopy (FESEM Quanta, ThermoFisher Scientific Corp., USA). Data of XRD results were processed using High Score Plus Version (3.0e) 3.0.5 with ICDD (International Centre for Diffraction Data) database.

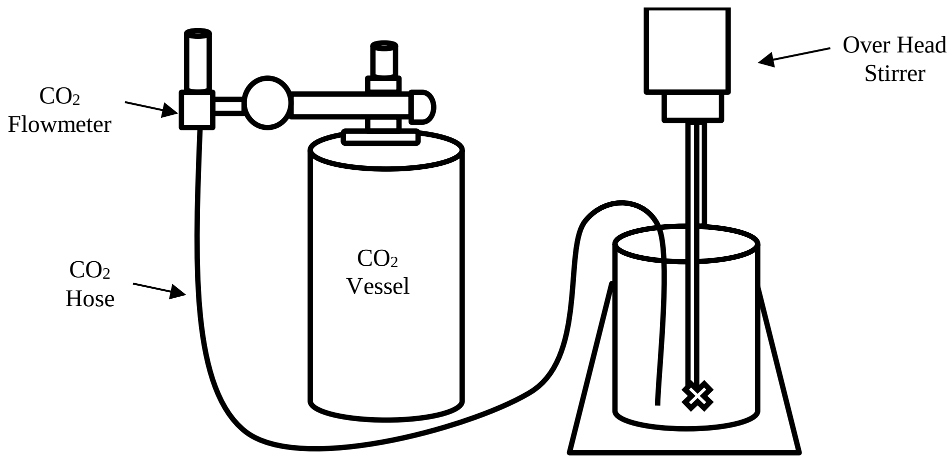


Figure 1. Experimental set-up for sodium bicarbonate production.

3. Results and Discussion

3.1. Taguchi analysis
The results of this study were analyzed using the "larger is better" signal-to-noise (S/N) ratio of the Taguchi method as presented in Table 3. The table shows that the highest delta value was the reaction time parameter. The rank indicates the order of influence or contribution of a parameter on product quality [8]. Reaction time was found to be the most effective operating condition or had the largest influence on the yield of sodium bicarbonate.

Tabel 3. The results of the S/N ratio analysis of the Taguchi method "large is better".

Level	Reaction time	NaCl concentration	CO_2 flow rate
1	-19.322	10.051	10.649
2	40.895	30.354	13.378
3	10.533	-20.302	-26.336
4	-29.677	-17.674	4.739
Delta	70.572	50.656	39.715
Rank	1	2	3

3.1.1. Influence of reaction time on sodium bicarbonate production
The results of Taguchi analysis in Figure 2 show that the optimum reaction time for sodium bicarbonate production was 15 min. The reaction time obtained was the effective time for NaCl, NH_4OH, and CO_2 to react completely to form sodium bicarbonate. A longer reaction time would increase side reactions from impurities that were present in the solution. The longer reaction time also was not effective to increase yield due to NH_4OH and NaCl limitation, while CO_2 consumption still increased and added the production costs [9].

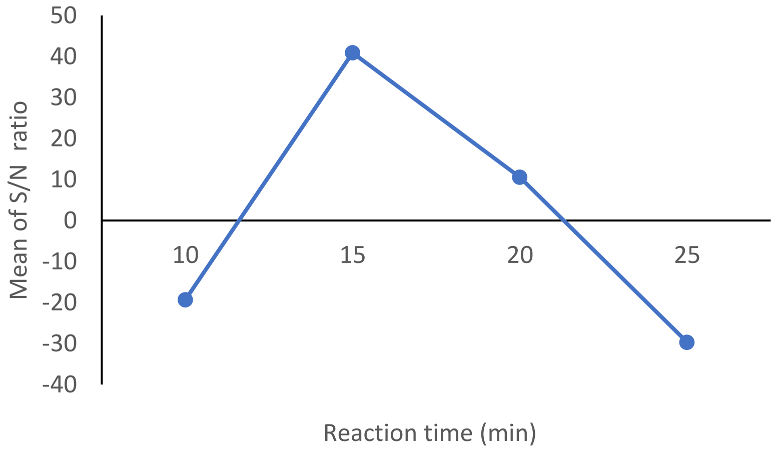


Figure 2. Means of reaction time S/N ratio.

3.1.2. Influence of NaCl concentration on sodium bicarbonate production
The results of Taguchi analysis in Figure 3 show that the optimum NaCl concentration for sodium bicarbonate production was 400 g. The optimum suggests that at concentrations higher or lower than 400 g, the results were influenced by NaCl that was not completely dissolved in NH_4OH solution, thereby reducing the purity of sodium bicarbonate product. At too high a concentration, NaCl could not dissolve completely.

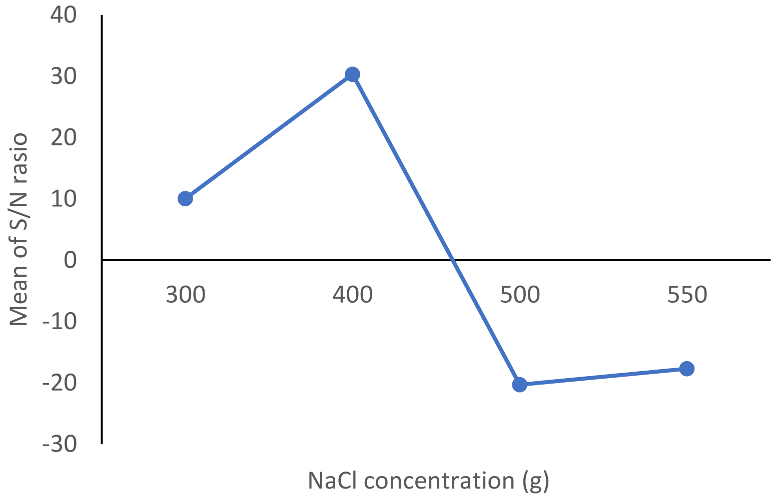


Figure 3. Means of NaCl concentration S/N ratio.

3.1.3. Effect of CO2 flow rate on sodium bicarbonate production

The results of Taguchi analysis in Figure 4 show that the optimum CO_2 flow rate for sodium bicarbonate production was 5 L/min. At higher flow rates, many large bubbles would be generated so it was unstable and CO_2 was difficult to react with NH_4OH and NaCl in the solution. The higher CO_2 flow rate also had no significant effect on sodium carbonate yield, since most of the NaCl and NH_4OH in the solutions had reacted to become sodium carbonate. Moreover, with increasing gas flow rate, the absorption capacity of NH_4OH and NaCl would decrease [10]. In this study, the gas was supplied directly from a small pipe and did not use a gas sparger to break big bubbles into small bubbles. The use of a gas sparger would generate small bubbles and the gas would be evenly distributed and react more effectively.

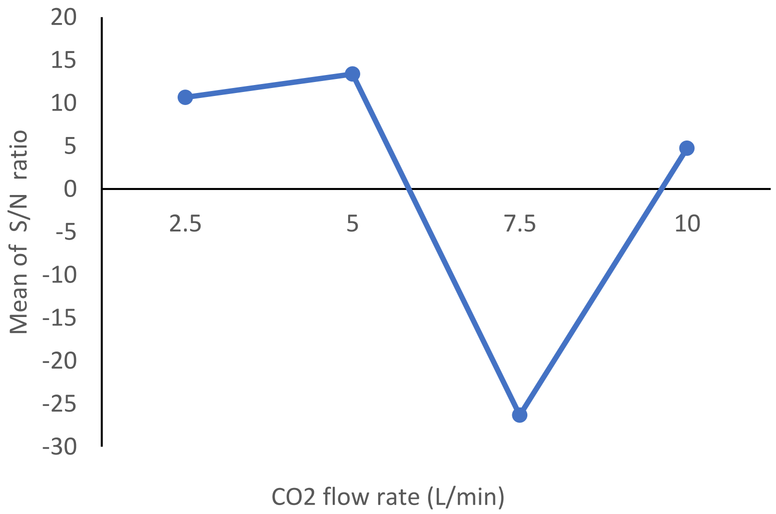


Figure 4. Mean of CO_2 flow rate S/N ratio.

3.2. XRD and FESEM-EDX characterization

XRD characterization was carried out to see the percentage of the phases contained in the sodium bicarbonate product. The XRD characteristics of the sodium bicarbonate powder produced at optimum conditions are presented in Figure 5. The figure shows a diffractogram pattern that is dominated by peaks of leucite, chlorocalcite, larnite, lomonosovite, and halite phases. The dominant phase in the sodium bicarbonate was the lomonosovite ($Na_5Ti_2(Si_2O_7)(PO_4)O_2$) phase at 47%, shown as number 4 with ref. ICDD code 01-076-0485 and the highest peak located at 2θ = 33.6106°. The second most dominant phase was the chlorocalcite ($KCaCl_3$) phase at 19%, shown as number 2 with ref. ICDD code 00-021-1170 and the highest peak located at 2θ = 31.7312°. Phase number 3 was larnite (Ca_2SiO_4) at 14%, with ref. ICDD code 01-070-0388 and the highest peak located at 2θ = 36.7067°. Phase number 5 was halite (NaCl) at 14%, with ref. ICDD code 00-005-0628 and the highest peak located at 2θ = 45.4785°. The phase that had the smallest percentage (6%) of total sodium bicarbonate was the leucite ($Na_{.15}K_{.85}AlSi_2O_6$) phase, with ref. ICDD code 01-074-2128 and the highest peak located at 2θ = 37.9268°.

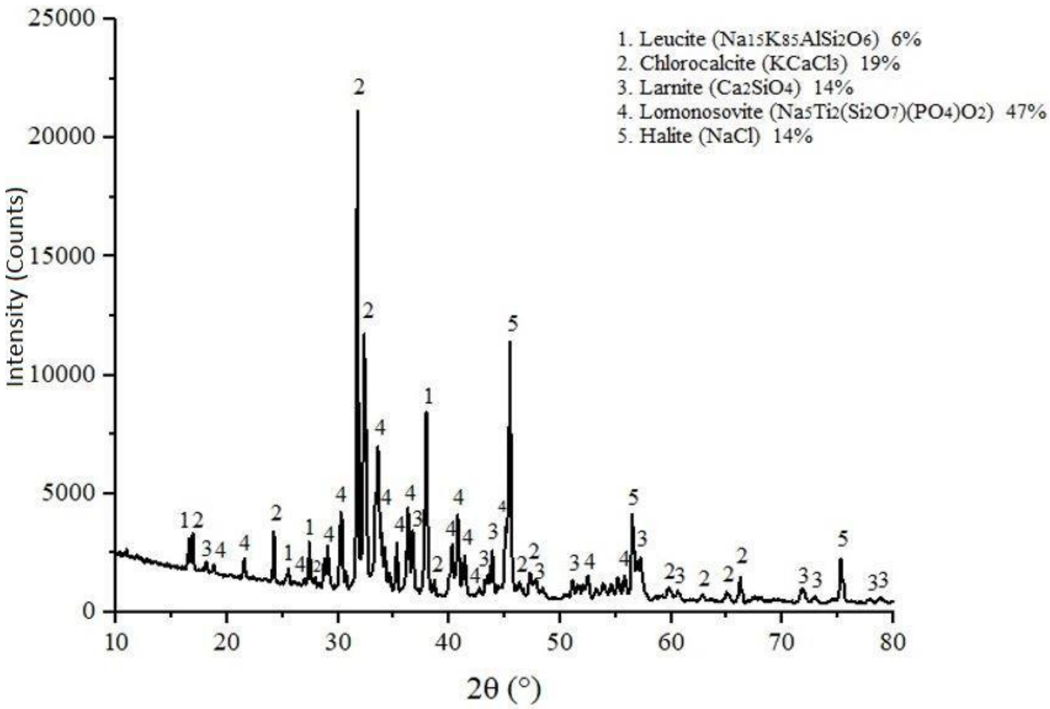
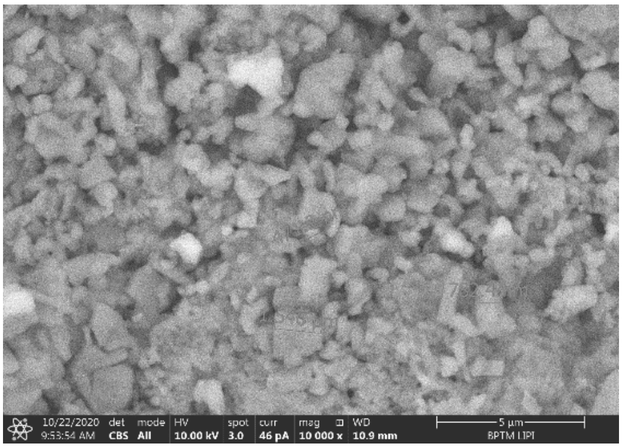


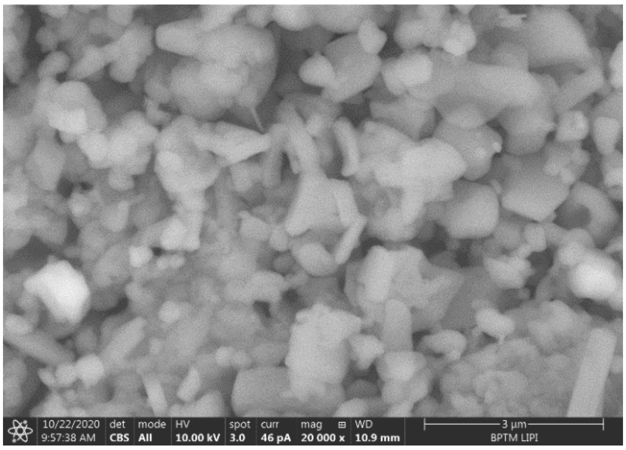
Figure 5. The optimum conditions of sodium bicarbonate XRD diffractogram pattern.

Field Emission Scanning Electron Microscope−Energy Dispersive X-ray Spectroscopy (FESEM-EDX) characterization was carried out to determine the surface morphological structure of the sodium bicarbonate product and to determine the composition of the elements contained in the sodium bicarbonate. Figures 6a and 6b show images of the morphological structure of sodium bicarbonate taken at 10000× and 20000× magnification, respectively.

The SEM results show that the morphological structure of sodium bicarbonate at optimum conditions did not have a uniform particle size, resembling irregular spherical shapes like lumps that had varying sizes. The fraction of solid particles in the product would depend on the method of sodium bicarbonate production. Sodium bicarbonate has particles that appear to be generally spherical (although not necessarily spherical in the true geometric sense) and a perforated structure [11]. Based on the image processing analysis at 10000× magnification (Figure 6a), it can be seen that the particle size of sodium bicarbonate product was 515 μm in diameter.



(a)



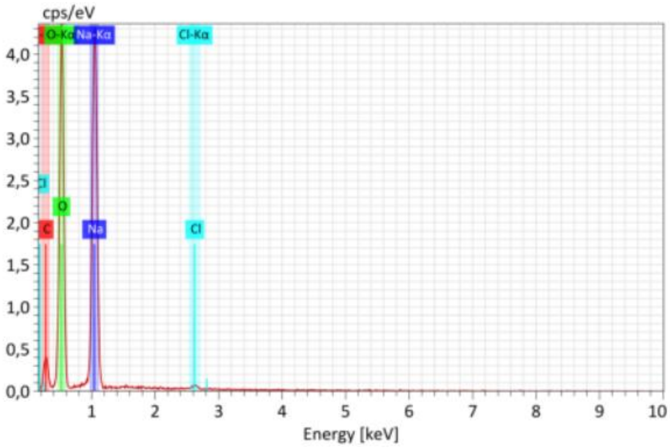
(b)

Figure 6. Particle structure of the SEM sodium bicarbonate magnification of (a) 10000× (b) 20000×.

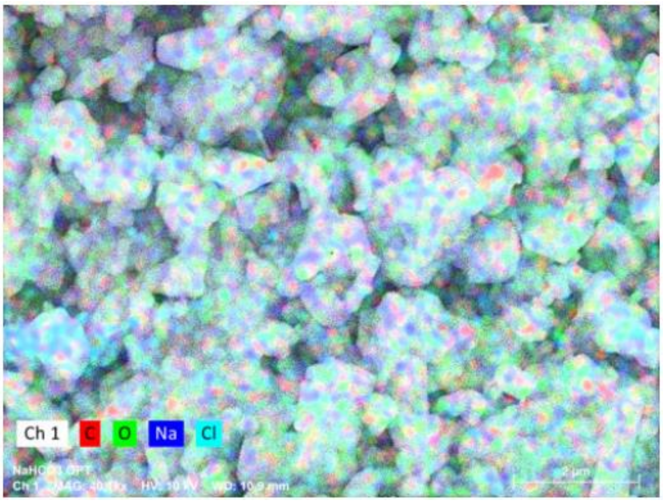
Figure 7a shows the spectrum of sodium bicarbonate characterization using FESEM-EDX. Figure 7b shows the mapping distribution of the elements carbon, oxygen, sodium, and chloride with different colors (mineralography). Table 4 shows that oxygen (O) in sodium bicarbonate had the largest mass percentage of 40.61%, followed by sodium (Na, 29.92%), carbon (C, 7.25%), and chloride (Cl, 0.78%). This result was comparable with commercial (high purity) sodium bicarbonate containing approximately 27.3% sodium [12]. The presence of Cl was due to the presence of residual NaCl that was not completely filtered, therefore still present in the sodium bicarbonate product.

Table 4. The elemental composition of sodium bicarbonate is based on the EDX results.

Element	Mass (%)
Carbon	7.25
Oxygen	40.61
Sodium	29.92
Chloride	0.78



(a)



(b)

Figure 7. (a) The spectrum of sodium bicarbonate characterization using EDX, (b) The mapping of elements in sodium bicarbonate.

4. Conclusions
Production of sodium bicarbonate using NH_4OH was successfully conducted and optimized using the Taguchi experimental design. The Taguchi analysis shows that reaction time is the parameter that influences sodium bicarbonate production the most. The optimal conditions for sodium bicarbonate production are 15 min of reaction time, 400 g of NaCl, and 5 L/min of CO_2 flow rate. The FESEM-EDX results show that the sodium bicarbonate product consists of 40.61% oxygen, 7.24% carbon, 29.92% sodium, and 0.74% of chloride. The presence of a small amount of chloride is due to the unreacted NaCl, which becomes impurities in the sodium bicarbonate product. The EDX results also show that the sodium bicarbonate product does not have any heavy metal impurities, hence it can be used for food application.

Acknowledgments
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