A Taguchi Optimization Study about the Dissolution of Colemanite in Ammonium Bisulfate (NH_4HSO_4) Solution

Mumcu Şımşek, Havva*+
Osmaniye Korkut Ata University, Department of Chemical Engineering, 80000, Osmaniye, TURKEY

Gulıyev, Rövşen
Ardahan University, Department of Environmental Engineering, 75002, Ardahan, TURKEY

ABSTRACT: *In this study, the optimum conditions of the dissolution of colemanite ($2CaO.3B_2O_3.5H_2O$) ore in ammonium bisulfate (NH_4HSO_4) solution were examined by using Taguchi fractional design methods, and an alternative reactant for boric acid extraction process from colemanite ore was specified. The studied parameters and optimum conditions for the dissolution process were the following; reaction temperature: 50°C, solid/liquid ratio: 0.1 g/mL, particle size: -80 mesh, mixing speed: 600 rpm, and reaction time: 25 minutes. Particle size, mixing speed, and reaction time had the most significant effect on dissolution when compared to the others. Accordingly, the dissolution efficiency of colemanite was found to be 99.54% under optimum conditions, and using NH_4HSO_4 as a solvent enabled the selective separation of boron from the colemanite ore. The ammonium sulfate formed as a by-product was converted into NH_4HSO_4 by the addition of the appropriate stoichiometric amount of sulfuric acid and fed back into the dissolution vessel.*

KEYWORDS: *Colemanite; Ammonium bisulfate; Dissolution; Optimization; Taguchi method.*

INTRODUCTION

Boron ore and its compounds, which constitute one of the most important underground treasures of Turkey, are strategically important substances with several application areas. Related compounds with the highest industrial value are colemanite, ulexite, and tincal. 72% of the world's boron reserves are found in Turkey [1-3], and the usage areas of boron minerals and products have been expanding in Turkey recently. Approximately 10% of those are solely used as minerals, and the remaining part is evaluated as raw material for the production of boric acid (H_3BO_3), which is utilized in many industries [4]. Generally, H_3BO_3 that is the starting material in the preparation of organic boron salts, is synthesized from colemanite [5,6].

In Turkey, H_3BO_3 is procured according to the following reaction of colemanite with sulfuric acid [7];

$$Ca_2B_6O_{11}.5H_2O_{(s)} + 2H_2SO_{4(aq)} + 6H_2O \quad 6H_3BO_{3(aq)} + 2CaSO_4.2H_2O_{(s)} \tag{1}$$

Borogypsum ($CaSO_4.2H_2O$) is formed as a by-product after the completion of the aforementioned reaction (Eq. (1), and is separated by filtration. But it is important to point out that, the remaining H_3BO_3 in the medium subsequent

** To whom correspondence should be addressed.*
+ E-mail: rovsenguliyev@ardahan.edu.tr
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to filtration should have high purity. Because sulfuric acid is a very strong acid and has the capability of dissolving gangue minerals in the boron ore. The main problem in the current technology of H_3BO_3 is that the gangue minerals in the colemanite ore are degraded in the reaction environment and hence, some problems arise during the process, and product quality decreases. Ammonium bisulfate (NH_4HSO_4) solution can be added for determining the optimum dissolution conditions of colemanite in order to overcome these complications.

The dissolution of colemanite with NH_4HSO_4 solution causes the formation of calcium sulfate crystals which have larger sizes and uniform geometric shapes in the reaction medium, accelerates the leaching process, and reduces the amount of B_2O_3 remaining in the solid waste of boro gypsum. Considering these findings, the major target of this paper is focused on developing a new technology process in H_3BO_3 production.

Regarding previous research, determining the optimal conditions for the production of H_3BO_3 from colemanite depends on a properly designed process. Traditional process optimization which involves studying one variable at a time requires a series of trial combinations such as labor, cost, and time [8]. Taguchi fractional experiment design procedure was applied in this study for saving time and cost within this context. The differences between Taguchi from other statistical techniques are to allow to provide the opportunity for the investigation of a large number of parameters at more than two levels, as well as analyzing the influencing factors in the experiment by dividing them into two parts controllable and uncontrollable. Furthermore, this method minimizes the variability around the target by adjusting the desired performance value, and hence the results obtained under trial conditions and the actual production have considerably similar values [9].

There are several studies in the literature about Taguchi experimental design and the dissolution of boron minerals [10-12]. *Küçük* (2006) [13] determined the optimum levels of parameters for the dissolution of colemanite ore in potassium bisulfate ($KHSO_4$) solution by using the Taguchi approach and the following results were obtained; solid/liquid ratio 1:10 g/mL, reaction temperature 50˚C, particle size -80 mesh, reaction time 15 minutes, and mixing speed 600 rpm. *Yeşilyurt*, *Çolak*, & *Çalban* (2004) [14] investigated the dissolution kinetics of colemanite ore in potassium dihydrogen phosphate (KH_2PO_4) solution and the reaction temperature, KH_2PO_4 concentration, mixing speed, solid/liquid ratio, and particle size were chosen as parameters affecting the dissolution rate. Statistical software was used to evaluate the experimental findings. The dissolution rate of colemanite increased when both reaction temperature and KH_2PO_4 concentration enhanced, but the changes in particle size and solid/liquid ratio did not cause such an influence on the related response. The value of activation energy (41.88 kJ/mol) also, confirmed this outcome. *Bilga* (2016) [15] benefitted from the Taguchi method in order to specify the optimum dissolution conditions of metals. *Dogan* (2013) [16] dissolved colemanite ore which was obtained from Bandırma boric acid plants in phosphoric acid solution and stated the optimum conditions by Taguchi fractional design methods as follows; reaction temperature 60°C, solid/liquid ratio 0.15 g/mL, mixing speed 200 rpm and reaction time 30 minutes. *Logothetis* (1992) [17] applied Taguchi method for dissolving ulexite in NH_4Cl solution and under optimum conditions, the dissolution percentage was indicated as 98.37. *Greenberg*, *Trussell*, & *Clesceri* (1985) [18] calcined colemanite ore in a free-falling flash calcination furnace and investigated the effects of calcination parameters on solubility. Optimum calcination conditions were identified according to Taguchi method and they emphasized flash calcination furnace temperature as 700°C, average ore particle size as 0.232 mm, heated flash calcination furnace size as 125 cm, and ore feed rate as 2 g/min for an efficient H_3BO_3 extraction. H_3BO_3 recovery from colemanite was 99.66%. On the other hand, *Ekince* (2020) [19] and *Copur* (2015) [20] remarked similar results regarding the dissolution of colemanite. To the best of our knowledge, there is not enough information concerning the dissolution of colemanite in NH_4HSO_4 solution. Therefore, the aims of this article are (i) to determine the optimum conditions using Taguchi method, and (ii) to develop a new technology process in H_3BO_3 production.

EXPERIMENTAL SECTION

Material

The colemanite ore was obtained from Emet district of Kütahya province in Turkey and crushed by a lab-size crusher device and ground by a grinder (ISOLAB, 038). Then, the material was screened by A.S.T.M. standard

Table 1: The chemical composition of colemanite.

Compound (%)	CaO	B_2O_3	H_2O	SiO_2 and the others
	24.42	43.52	18.90	13.16

Table 2: Factors and actual levels studied in trials.

Codes	Factors	Units	Levels			
			1	2	3	4
A	Temperature	°C	50	60	70	80
B	Solid/liquid ratio	g/mL	0.2	0.1	0.07	0.05
C	Mixing speed	rpm	300	400	500	600
D	Time	minutes	5	15	25	35
E	Particle size	mesh	-20	-40	-60	-80

sieves and divided into particle sizes of -20, -40, -60, -80 mesh. The chemical composition of colemanite ore was given in Table 1. The NH_4HSO_4 solution was procured from MERCK (Germany).

Setup and Design of the Experiment (DOE)

The experiments were accomplished in a 250 mL double-necked glass reactor equipped with a condenser in order to prevent solution loss by evaporation, a temperature circulator for controlling the reaction temperature, and a mechanical stirrer. 100 mL of 1.5 M NH_4HSO_4 solution was put into the reactor, and it was heated up to a certain reaction temperature. Then, the ore was added to the solution and the reaction was started. At the end of the specific reaction period, the content was filtered and the amount of boron was determined by a titrimetric and potentiometric method (1mL samples of the reacted solution were taken for the assay of B_2O_3 and analyzed. It was calculated as in equation 2) according to [21,26]. The experimental factors and studied levels were demonstrated in Table 2.

$$X\,(\%) = \frac{\text{amount of dissolved } B_2O_3 \text{ in the solution}}{\text{amount of dissolved } B_2O_3 \text{ in the original sample}} \times 100 \quad (2)$$

Optimization of the dissolution of colemanite in NH_4HSO_4 solution was carried out by Taguchi fractional design method starting with the choice of the orthogonal array. The minimum number of experiments in the orthogonal array is given by [22];

$$N_{min} = (l-1)k + 1 \quad (3)$$

k: Number of parameters

l: Number of levels

Reaction temperature (A), solid/liquid ratio (B), mixing speed (C), time (D), and particle size (E) were the considered process parameters during trials. Since 4 different levels were selected for each factor, the L_{16} (5^4) sequence was employed as Taguchi factorial fractional orthogonal index (Table 3) [23]. The orthogonal index plan with responses was depicted in Table 3.

Taguchi loss function, in other words, signal-to-noise (S/N) ratio is a multi-objective function [21] that is estimated by averaging the measurement values. Thanks to S/N ratio, the responses are able to be evaluated as behaving linearly [24]. Equations (4), (5), and (6) are valid for "larger the better", "smaller the better" and "nominal the better" conditions of purpose, respectively.

For larger the better;

$$SN_L = -10\log\left(\frac{1}{n}\sum_{i=1}^{n}\frac{1}{y_i^2}\right) \quad (4)$$

For smaller the better;

$$SN_S = -10\log\left(\frac{1}{n}\sum_{i=1}^{n}{y_i}^2\right) \quad (5)$$

For the nominal the better;

$$SN = -\log\left(\frac{\tilde{y}^2}{S^2}\right) \quad (6)$$

Table 3: L_{16} (4^5) standard experimental matrix of orthogonal design.

Run Order	A	B	C	D	E	B_2O_3 (%)	S/N
1	1	1	1	1	1	56.15 ± 1.94	38.24
2	1	2	2	2	2	87.01 ± 0.05	39.71
3	1	3	3	3	3	92.80 ± 0.58	39.60
4	1	4	4	4	4	98.11 ± 0.79	39.39
5	2	1	2	3	4	96.88 ± 0.76	38.68
6	2	2	1	4	3	90.22 ± 0.53	39.44
7	2	3	4	1	2	100.00 ± 0.00	39.22
8	2	4	3	2	1	100.00 ± 0.00	39.60
9	3	1	3	4	2	100.00 ± 0.00	38.07
10	3	2	4	3	1	100.00 ± 0.00	39.11
11	3	3	1	2	4	89.15 ± 0.49	39.81
12	3	4	2	1	3	93.31 ± 1.61	39.96
13	4	1	4	2	3	100.00 ± 0.00	38.38
14	4	2	3	1	4	98.53 ± 0.66	38.39
15	4	3	2	4	1	84.46 ± 2.18	39.46
16	4	4	1	3	2	91.07 ± 3.92	39.61

$$\tilde{y} = \left(\frac{1}{n}\sum_{i=1}^{n} y_i\right) \quad (7)$$

$$S^2 = \left(\frac{1}{n-1}\sum_{n=1}^{n} (yi - \tilde{y})^2\right) \quad (8)$$

y_i: Observation value of i^{th} performance response,

n: Number of tests,

$\tilde{y}$: Mean of observation values,

S^2: Variance of observation values

If the target is to reach the minimum, the parameter levels that make the SN_s minimum, are optimum. On the contrary, the maximization of SN_L gives optimum for achieving a maximum goal. Performance value, which is suitable for optimum experimental conditions designated by the Taguchi method could be predicted with the help of Eq. (9).

$$Y_i = \mu + X_i + e_i \quad (9)$$

Y_i: Estimated performance value of the ith experiment,

μ: General average of performance value,

X_i: Measure the effectiveness of the ith experiment,

e_i: Experimental error

Conversion of the test results given as a percentage to decimal values is able to be done with the help of Eq. (10), before the calculation of the Y_i value.

$$\Omega = -\log 10\left(\frac{1}{x} - 1\right) \quad (10)$$

Ω: decibel value,

x: dissolution fraction of the ith experiment

The confidence limits for the prediction error, S_e is

$$S_e = \pm 2\sqrt{\left[\frac{1}{n_0}\right]\sigma_e^2 + \left[\frac{1}{n_r}\right]\sigma_e^2} \quad (11)$$

$$\frac{1}{n_0} = \frac{1}{n} + \left[\frac{1}{n_{Ai}} - \frac{1}{n}\right] + \left[\frac{1}{n_{Bi}} - \frac{1}{n}\right] + \left[\frac{1}{n_{Ci}} - \frac{1}{n}\right] + \cdots \quad (12)$$

σ^2: Sum of squares of the error/degrees of freedom of the error

n: the number of rows in the matrix experiment

After defining the optimum conditions, a confirmation analysis was performed. When the factor-level combination found at the end of the experiment reached the best characteristic value, it was accepted that the desired situation was actualized and the aim of the experiment was achieved [25]. All statistical evaluation was performed by MINITAB package program (version 19, USA).

RESULTS AND DISCUSSION

Dissolution reactions

Dissolution processes were carried out using NH_4HSO_4 solutions, and the reactions occurring in the medium were as follows [26];

$$4NH_4HSO_{4\ (aq)} \rightarrow 4NH_4^+{}_{(aq)} + 4HSO_4^{-1}{}_{(aq)} \quad (13)$$

$$4HSO_{4(aq)}^{-1} + 4H_2O_{(aq)} \leftrightarrow 4H_3O_{(aq)}^+ + 4SO_4^{-2}{}_{(aq)} \quad (14)$$

Colemanite reacted with NH_4HSO_4 solution as in Eqs (15) and (16);

$$2CaO.3B_2O_3.5H_2O_{(s)} + 4H_3O_{(aq)}^+ \rightarrow 2Ca_{(aq)}^+ + 6H_3BO_{3(aq)} + 2H_2O_{(l)} \quad (15)$$

$$2Ca_{(aq)}^{+2} + 2SO_{4(aq)}^{-2} \rightarrow 2(CaSO_4.2H_2O)_{(s)} \quad (16)$$

The total reaction was;

$$2CaO.3B_2O_3.5H_2O_{(s)} + 4NH_4HSO_{4(aq)} + 6H_2O_{(aq)} \rightarrow 2(CaSO_4.2H_2O)_{(s)} + 6H_3BO_{3(aq)} + 2(NH_4)_2SO_{4(aq)} \quad (17)$$

As a result of the total reaction, boro gypsum ($CaSO_4.2H_2O$), boric acid (H_3BO_3), and as a by-product ammonium sulfate ($(NH_4)_2SO_4$) were obtained. An appropriate stoichiometric ratio of sulfuric acid was added to the available $(NH_4)_2SO_4$ solution, and the released NH_4HSO_4 was fed back to the reactor [$(NH_4)_2SO_4+H_2SO_4 \rightarrow 2(NH_4HSO_4)$]. Thus, the amount of reactant added at the beginning was reduced, and the system was economically beneficial.

Statistical evaluation

Performance statistics are selected in accordance with the properties of more than one performance characteristic for the solution of the problem concerned in Taguchi method. The analysis of the data is conducted according to this selected way, so the correct determination is very significant. The most common performance statistic in Taguchi designs are the S/N ratio, which measures the performance of the robust design.

The "larger the better" analysis (Eq. (4)) was used because the objective of this research was to procure the highest dissolution rates. For this purpose, the parameter levels that make SN_L maximum were determined as optimum values. In assessing the factor stages which maximized the dissolution of H_3BO_3 in colemanite, SN_L performance statistics data that was calculated from related mathematical expressions were utilized, and S/N values were found for each experiment (Table 3).

The F test is a useful tool for observing the influences of the analyzed parameters on dissolution rate [14]. Variance analysis was conducted for benefitting from the dissolution fractions in order to determine the efficiency of factors, and F efficiency values were calculated. F efficiency tables were used to find out whether the parameters were effective or not. By comparing the experimental value of F to the F value included in the table, the efficiency of a factor on the dissolution was cleared (Table 4). If $F_{experimental}$ is greater than F_{table}, the relevant factor is effective, and vice versa [27-28]. $F_{(1.16),\ 0.95} = 4.49$, $F_{(1.16),\ 0.99} = 8.53$ in the table, and hence, all parameters were effective. *Ekinci*, *Kurdal*, & *Kizilca Coruh*'s papers were currently in line with this study [29]. Reaction temperature, solid/liquid ratio, SO_2/CO_2 ratio, particle size, stirring speed, and time (all parameters) were effective in the dissolution of colemanite in SO_2 and CO_2-water systems. *Küçük*, and *Korucu* (2018) [30] extracted H_3BO_3 from colemanite by Taguchi technique in an environmentally friendly way and stated that only solid/liquid ratio and particle size were effective at the second stage of the optimization test.

Test of each parameter in the dissolution process was performed by using S/N graphs (Fig. 1). It could be clearly understood that, each factor in the dissolution of colemanite in NH_4HSO_4 solution was significant. Optimum conditions were also defined with regard to the same illustration by Taguchi techniques[31-33]. The high level of S/N ratio indicated optimum among all levels of every factor. Accordingly, optimum temperature, solid/liquid ratio, mixing speed, time, and particle size were A1 (50˚C), B2 (0.1 g/mL), C4 (600 rpm), D3 (25 min.) and E4 (-80 mesh), respectively (Table 5). The results of the dissolution experiments were found to be consistent with the observed and predicted results. As the particle size decreases, the dissolution rate increases. Reducing the size increases the number of particles per unit weight. Thus, as the contact surface between the fluid and the solid particle will increase, it is possible result that the dissolution rate will increase. If the transfer rate of the products to the main

Table 4: Analysis of variance (ANOVA) outcomes of optimization experiments.

Codes	Factors	SDi	SSi	MSi	F	Efficiency
A	Temperature (˚C)	3	876.91	292.304	161.25	Effective
B	Solid/liquid ratio (g/mL)	3	244.40	81.468	44.94	Effective
C	Mixing speed (rpm)	3	1598.39	532.796	293.92	Effective
D	Time (minutes)	3	322.36	107.454	59.28	Effective
E	Particle size (mesh)	3	564.35	188.116	103.77	Effective
	Pure Error	16	29.00	1.813		
	Cor. Total	31	3635.42			

Table 5: Optimum conditions, predicted and experimental results.

Codes	Factors	Value	Level
A	Temperature (˚C)	50	1
B	Solid/liquid ratio (g/mL)	1	2
C	Mixing speed (rpm)	600	4
D	Time (minutes)	25	3
E	Particle size (mesh)	80	4
Experimental		99.54 ± 0.65	
Predicted		100.00	
Confidence interval		100 - 97.43	

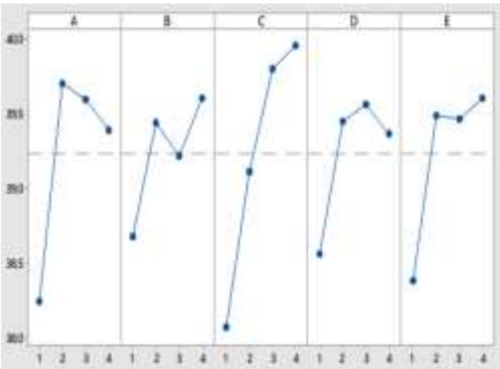

Fig. 1: The effect of factors on performance statistics in the dissolving process.

the solution is small, the products form a film around the colemanite particles. Over time, this film thickens and the transformation range of colemanite decreases. The increase in temperature caused an increase in the collision rate per unit times, which increases the reaction rate [28]. The optimum conditions of the experiments were found with the MINITAB package program according to the experiment plan given in Table 3. The dissolution data in Table 5 are the results were calculated, estimated, and observed from the equations in equations (9)-(10). For confirmation of findings, validation tests including three repetitions were conducted and the results were shown in Table 5 as well. It was found that the estimated amount of B_2O_3 was 100.00%, while the observed dissolution rate of B_2O_3 was 99.54% in solution under these conditions. Bingöl, & Çopur (2019) [31] declared the calculated and observed dissolution rate of B_2O_3 as 96.95% and 90.22% in the high-pressure reactor by using CO_2. The obtained dissolution percentage data are in the confidence interval calculated from Eqs (10) and (11). It has been seen that the Taguchi model is compatible with the experimental results were found to explain the relationship between the dissolution process and the parameters.

CONCLUSIONS

• In this study, NH_4HSO_4 solution was used instead of sulfuric acid in the production of H_3BO_3, and NH_4HSO_4

was a successful solution for high dissolution rates of H_3BO_3.

- Since sulfuric acid which is utilized in the production of H_3BO_3 in Turkey possesses a strong acid ability, undesirable dissolution of several minerals, as well as boron arises. Unfortunately, this situation leads to the contamination of the final product. When NH_4HSO_4 solution reacted instead of sulfuric acid, the other minerals were less dissolved, thus the contamination ratio was decreased and the dissolution efficiency was increased.
- Sulfuric acid formed by the result of the reaction, was added to $(NH_4)_2SO_4$ solution and NH_4HSO_4 which was fed back to the system solution, was produced. Therefore, the consumption of the reactant was reduced.
- The usage of NH_4HSO_4 as the solvent reactant was also suitable in order to prevent the corrosion of the device.
- The experimental data were evaluated statistically with the aid of one-way analysis of variance (ANOVA). When the results were examined, it could be inferred that all parameters were effective for the dissolution of colemanite. Optimum conditions were; temperature 50°C, solid: liquid ratio 0.1 g/mL, stirring speed 600 rpm, particle size -80 mesh, and reaction time 25 minutes.
- Considering the performance criteria and ANOVA table, it was seen that, the effective parameters for obtaining H_3BO_3 were mixing speed, particle size, solid/liquid ratio, temperature, and time.
- By Taguchi method, the impact of dissolution was determined in a short time, and the product quality was developed by reducing time and cost. The optimum process conditions suggested by Taguchi technique could be a reference in both pilot and industrial scale process design, during size enlargement.

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