Optimization of Boric Acid Extraction From Colemanite With An Environmentally Friendly Process By Using The Taguchi Method

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Summary: Boric acid, one of the boron products, is obtained from colemanite sulfuric acid process in some countries. In this process, sulfur dioxide was used instead of sulfuric acid to obtain H_3BO_3. The purpose of this research is to produce boric acid from a cheaper and more environmentally friendly process. In this research where H_3BO_3 was obtained, optimization of the dissolution of the original colemanite containing clay in SO_2-saturated water was investigated using the "Taguchi Method". In experiments, solid-liquid ratio, mixing speed, reaction temperature, reaction time and particle size were selected as parameters. Optimization studies were carried out in two stages. According to the results obtained in the first stage, the best value for the mixing speed was found to be 300 rpm and the mixing speed is found to have little effect. Therefore, the mixing speed was kept constant in the second stage. According to the second design, the optimum conditions were found at 50°C for reaction temperature, -212+150 µm for particle size, 0.25 g/mL for solid liquid ratio, and the B_2O_3 passing through the solution under these conditions was 98.5%.

Keywords: Colemanite; Boric Acid; SO_2; Dissolution; Taguchi Method

Introduction

Turkey has the biggest boron reserves in the world. 72% of world reserves are in Turkey. The commercial boron reserves in Turkey are uleksite, colemanite and tincal. These ores are enriched by being subjected to a concentration treatment after being extracted as raw. In some countries such as Turkey, H_3BO_3 is produced by sulfuric acid process from colemanite. The boron content of colemanite with chemical composition $2CaO.3B_2O_3.5H_2O$ is 15.7% and the content of B_2O_3 is 50.8%.

Among the contaminants in the gas phase, sulfur dioxide is the most common primary air pollutant. Acid rain caused by SO_2 causes corrosion on earth rocks, constructions and metal surfaces and disrupts the natural balance in various ecosystems. SO_2 also forms a fog layer over industrial areas and large cities.

In the chemical industry, one of the most important sectors in environmental pollution, one of the alternatives to prevent environmental pollution, perhaps the most important, is to develop environmentally friendly technologies. It is more common in the industry to obtain boric acid from colemanite or ulexite by reaction with sulfuric acid. In the applied technology, gypsum is formed as waste along with boric acid and this waste is pumped to the environment. This may cause major problems in the future. In addition, the raw boric acid produced contains various impurities, mainly sulphate and calcium, and obtaining a refined product requires additional processing, which increases the cost. In this respect, it is important to develop environmentally friendly new processes and technologies to obtain cheaper, purer boric acid and to protect the environment.

The aim of this work is to obtain cheaper and more pure boric acid than the sulfuric acid process with an environmentally friendly process without waste or waste of process can be evaluated in another way. In sulfuric acid process, boric acid is obtained by dissolving concentrated colemanite in sulfuric acid solutions at 90-92 ºC, after filtering the mixture in pressure filters and then crystallizing in vacuum crystallizers. The by-product at the reaction is gypsum and it is disposed as waste after being filtered. Although some studies have been carried out in recent years on the evaluation of this waste, no action is taken on the assessment of this waste. In this process, in the crystal boric acid which obtained has sulphate, calcium and magnesium pollution, which decreases the market value of boric acid produced. Crystal boric acid is washed with water to make up for it and to get rid of these pollutants. This causes to both boric acid losses and to more water entering to the process and adversely affects to cost. In addition to this, due to excessive water penetration to the process, water containing boric acid is sent to the

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waste dam, resulting in a loss of boric acid and a negative situation in terms of environmental pollution. Therefore, it is important to develop a process that is both cheaper and more environmentally friendly than the existing process.

For this purpose, optimum conditions for dissolving colemanite mineral in water saturated with SO_2 at high solid/liquid ratios were investigated and solid-liquid ratio, mixing speed, reaction temperature, reaction time and particle size were used as parameters for this research. The Taguchi experimental design method, one of the optimization methods, has used to determination of the optimum dissolution conditions.

Taguchi's Orthogonal Array analysis is used for optimal design processes where the best parameters are obtained with minimal experimentation. In recent years, Taguchi method has been applied extensively in the finding of optimum working conditions due to many advantages. In Taguchi method, the parameters affecting an experiment are examined in two groups, one being controllable and the other being uncontrollable, and enabling a large number of parameters to be examined at two or more levels [1].

There are many studies on dissolution and optimization of colemanite in different solution. Imamutdinova, studying the dissolution of colemanite in various mineral acids, found that the solubility in H_2SO_4, H_3PO_4, HNO_3 and HCl solutions were the same, that there was a maximum solubility value for each acid, and researcher expressed that these maximum values varied depending on the acid type and temperature [2]. The dissolution of colemanite in CO_2-saturated water was investigated by Gülensoy and Kocakerim [3]. The researchers found that the original and calcined colemanite samples were dissolved in CO_2–saturated water and that the dissolution rate of the calcined ore was the highest at 380°C. Alkan et al. [4] studied the dissolution kinetics of colemanite in CO_2-saturated water. The researchers found that the dissolution process was chemically controlled, the dissolution rate was increased by increasing the calcination and reaction temperatures, decreasing the particle size. The activation energy for the calcined samples at 400°C was found as 57.7 $kJ.mol^{-1}$. Özmetin et al. [5] studied the on dissolution kinetics of colemanite in CH_3COOH solutions and proposed a kinetic model. The investigator determined that the dissolution rate increased by increasing the acid concentration up to the concentration of 3,365 M acetic acid and decreased after this concentration, by decreasing the particle size and solid/liquid ratio, and the activation energy for this process being 51.49 $kJ.mol^{-1}$. Yartaşı *et al.* studied the kinetic and dissolution mechanism of colemanite in H_3BO_3 solutions. As a result, the researchers determined that the dissolution process was be diffusion controlled from the ash film and that occured the various calcium borates [6]. Ceyhun et al. [7] examined the dissolution kinetics of colemanite in chlorine-saturated water. They were found that the rate of dissolution increased with decreasing particle size and solid / liquid ratio and with increasing the temperature. It was found that he reaction rate for this process was chemically controlled and the activation energy is 35.56 $kJ.mol^{-1}$. Temur *et. al.* investigated the dissolution kinetics of colemanite in phosphoric acid solutions. It was identified the rate of dissolution increased up to an acid concentration of 19.52% and then decreased [8]. In a study for the production of boric acid, Tunç et. al. [9] studied the solubility of colemanite in $(NH_4)_2SO_4$ solutions. In this study, it was found that the dissolution rate increased with decreasing particle size and solid/liquid ratio and with increasing $(NH_4)_2SO_4$ concentration and temperature. Gür and Alkan [10] studied the leaching kinetics of colemanite in perchloric acid solutions. Künkül et. al. [11] They have investigated the production of boric acid from the reaction of $(NH_4)_2CO_3$ solutions with calcined colemanite. Küçük et. al. [12] examined the dissolution of the original Kestelek colemanite in saturated water with sulphur dioxide. Researchers have expressed that this dissolution process is similar to the dissolution of pure colemanite in the water saturated with sulfur dioxide [13] and is chemically controlled, have calculated the activation energy to be 39.53 $kJ.mol^{-1}$. The authors attributed this behavior to impurities in the ore. Davies et al. [14] studied the formation of boric acid by dissolving colemanite samples that were subjected to flash calcination in water saturated with CO_2 and found that the calcined samples showed the fastest dissolution at 600°C. Ata et al. [15] examined the optimization of the dissolution of arsenic colemanite in water satureted with carbon dioxide. At the optimum conditions determined as 70°C reaction temperature, 0.1 solid/liquid ratio, 2.7 bar gas pressure and 120 minutes reaction time, almost all of the colemanite ore was converted to boric acid. Yeşilyurt [16] determined the optimum conditions for boric acid extraction from colemanite ore in HNO_3 solutions using the Taguchi method. Also in a study used Taguchi method [17], the optimization of the dissolution of ulexite in SO_2-saturated water has been investigated. Doğan and Yartaşı have used Taguchi

Method in determine of optimum conditions for the dissolution of ulexite in phosphate acid solutions [18]. They found that the optimum conditions were found to be 60°C, 0.15g.mL^{-1}, 30 minutes, 200 rpm, (-850+452) µm and 0.70M. In other two research, it was studied application of Taguchi method in optimization of dissolution of ulexite in $(NH_4)_2SO_4$ and NH_4Cl solutions [19,20].

Experimental

Materials Preparation

The colemanite mineral used in the experiments was obtained from the Bandirma Boric Acid plant of Etibank and was taken directly from the mineral deposit without being cleaned from ore impurities and broken with a laboratory size crusher and ground with a laboratory type crusher. The ground ore were divided into particle sizes of -1700+1180, -1180+850, -850+600, -600+355, -355+212 µm with ASTM standard sieves. The ore sample was chemically analyzed and found to contain 46.68% B_2O_3, 27.27% CaO, 0.02% Na_2O, 20.00% H_2O (Depending on the structure), and 6.75% other impurities. X-ray diffractogram of original sample obtained by Rigaku DMAX 2000 Series X-ray diffractometer is given in Fig. 1.

Dissolution operation was carried out in a 250 mL double-wall glass reactor and atmospheric pressure. A mechanical stirrer with a tachometer was used for mixing and a constant temperature circulator was used to keep the reaction temperature constant. In the dissolving process, 100 mL of purified water was added to the reaction vessel at each time, the reaction vessel and its content were brought to reaction temperature, and pure water was saturated with SO_2 gas at the reaction temperature, and experiments were carried out using other parameters and their levels specified in the designs. In order not to reduce the SO_2 concentration and to saturate the solution, the SO_2 gas was continuously fed to the reactor throughout the dissolution reaction. At the end of the specified times the stirring was stopped and after the reaction vessel was filtered, B_2O_3 analysis was made. Sulfur dioxide gas was passed through a scrubbing flask filled with sulfuric acid for drying before being fed to the system and then passed from a drying tower filled with coke for the trapping of drifting H_2SO_4. The gas was passed through a flow meter which was used to adjust the flow rate, and then it was givenm the reactor. The gas flow in all studies rate is constant. The experimental setup used is shown in Fig. 2.

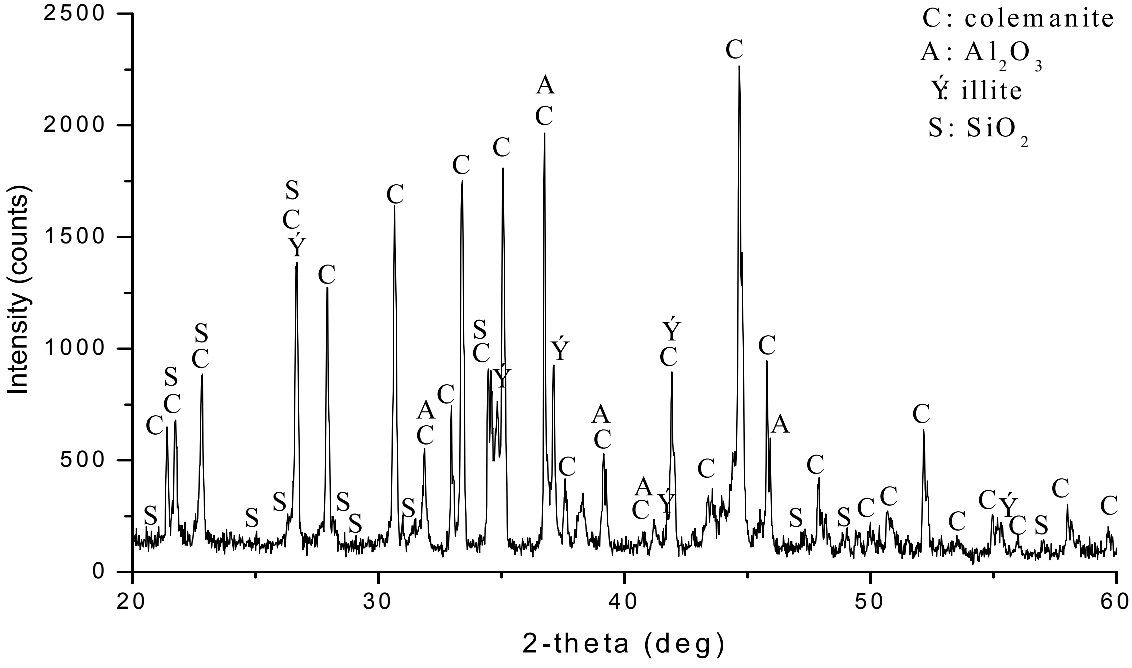

Fig. 1: X-ray diffractogram of original sample.

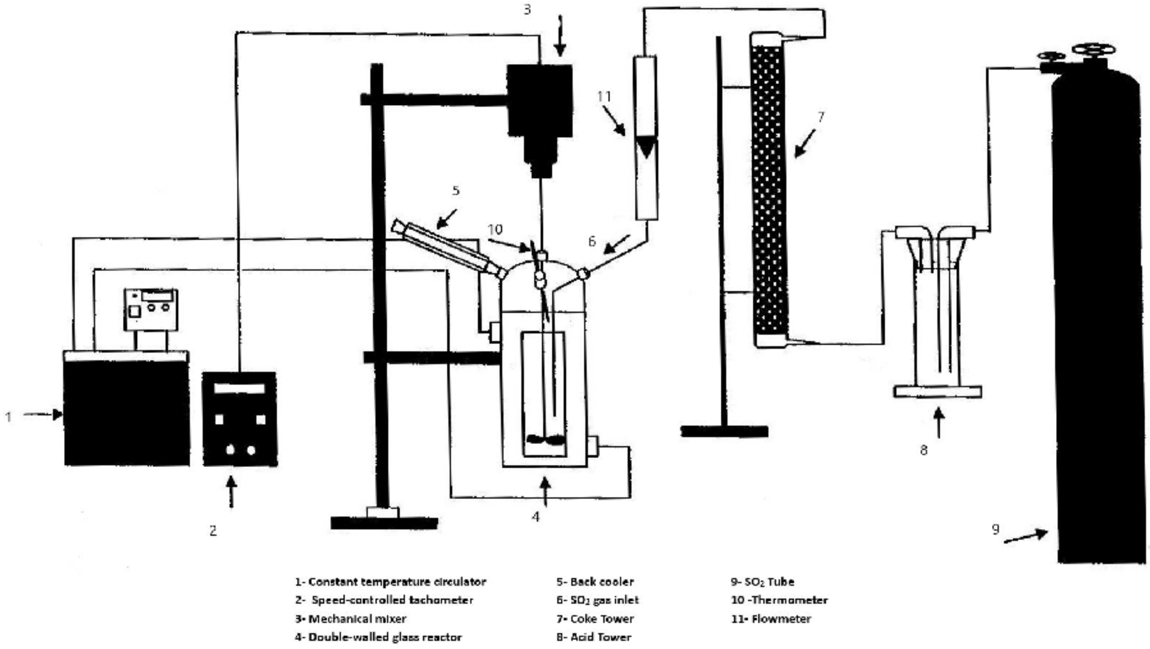

Fig. 2: Test setup used in the dissolution process.

Taguchi Method

One method based on the principle of least experimentation to keep costs at a minimum is "Taguchi Methodi" developed by Japanese scientist Genichi Taguchi [21]. This method differs from other statistical experiment design methods; The parameters affecting an experiment are examined in two groups, one being controllable and the other being uncontrollable, and enabling a large number of parameters to be examined at two or more levels. Generally, the performance characteristic of each of the product or process must have a nominal value or target value. The goal is to reduce the variability around this target value. The optimum operating conditions to be determined at the end of the experimental study should always be the same or very close to each other in different operating environments or at different times. The optimization criterion to be used for this should be able to control the variability around the performance value to a minimum. According to Taguchi, such an optimization criterion is performance statistics. The following performance statistical formulas are used to determine the optimum levels of the parameters [22].

For Larger-the-better; $SN_L = -10Log\left(\frac{1}{n}\sum_{n=1}^{n}\frac{1}{y^2}\right)$ (1)

For smaller-the-better; $SN_S = -10Log\left(\frac{1}{n}\sum_{n=1}^{n}y^2\right)$ (2)

If the goal is to achieve the maximum value, the parameter levels that make the maximize SNL is optimum If the goal is to reach the minimum, the parameter levels that make the SNs maximum are optimum [23]. Performance value corresponding to optimum operating conditions determined by Taguchi method can be estimated with the help of equation (3).

$$Y_{it}= \mu +X_i + e_i \qquad (3)$$

If the test results are given as a percentage, before the Y value is calculated, the percent values converted to decibel value with the help of equation (4).

$$\Omega_i=-10Log(y_i/(1-y_i)) \qquad (4)$$

Because Eq. (3) is a point estimation, which is calculated by using experimental data in order to determine whether the additive model is adequate or not, the confidence limits for the prediction error must be evaluated [22]. The prediction error is the difference between the observed Yi and the predicted Y_i. The confidence limits for the prediction error, Se, is

$$\mathrm{Se} = \pm 2\sqrt{\left[\frac{1}{\mathrm{n}_0}\right]\sigma_e^2 + \left[\frac{1}{n_r}\right]\sigma_e^2} \qquad (5)$$

$$\sigma_e^2 = \frac{\text{sum of squares due to error}}{\text{degrees of freedom for error}} \quad (6)$$

$$\frac{1}{n_o} = \frac{1}{n} + \left[\frac{1}{n_{A_i}} - \frac{1}{n}\right] + \left[\frac{1}{n_{B_i}} - \frac{1}{n}\right] + \left[\frac{1}{n_{C_i}} - \frac{1}{n}\right]. \quad (7)$$

If the prediction error is outside these limits, the model is not adequate. Otherwise, the model can be considered to be adequate. Verification experiment is a powerful tool for detecting the presence of interactions among the control parameters. If the predicted value under the optimum conditions does not match the observed value, the interactions are important. If the predicted response matches the observed response, then the internal interactions are probably not important and the model is a good approximation [21].

In this study, two different Taguchi factorial fractional experiment design plans were made and the L_{25} (5^5) Taguchi factorial fractional experiment design was planned by considering 5 which was the number of parameters determined for the initial design and, 5 different levels for each parameter. The parameters and their levels of the first design are given in Table-1, and the orthogonal experimental design plan and results are given in Table-2.

When the results of the experiments in the first design were examined, it was found that the effect of the stirring speed was almost non-existent so that the stirring speed was fixed at 300 rpm in all the experiments for the Second Design. By considering the number of parameters determined for the second study to be 4 and 3 different levels for each parameter, L_9 (4^3) Taguchi factorial fractional experiment design was planned. Parameters and levels are given in Table-3, and experimental design and results are given in Table-4.

Table-1: Parameters and their levels for first design.

	Parameters	Levels				
		1	2	3	4	5
A	Reaction Temperature (°C)	30	40	60	75	90
B	Particle Size (µm)	-1700+1180	-1180+850	-850+600	-600+355	-355+212
C	Solid-to-Liquid Ratio (g/mL)	0.05	0.1	0.15	0.2	0.25
D	Stirring Speed (rpm)	300	400	500	600	700
E	Reaction Time (minute)	15	30	45	60	90

Table-2: Experiment plan and its results for first design.

Deney No	A	B	C	D	E	%B_2O_3 Experiment 1	%B_2O_3 Experiment 2
1	1	1	1	1	1	96.4	99.9
2	1	2	2	2	2	83.09	82.63
3	1	3	3	3	3	71.06	73.85
4	1	4	4	4	4	76.32	76.73
5	1	5	5	5	5	82.39	83.99
6	2	1	2	3	4	99.99	99.98
7	2	2	3	4	5	77.11	79.83
8	2	3	4	5	1	71.45	72.72
9	2	4	5	1	2	69.04	69.9
10	2	5	1	2	3	99.99	99.98
11	3	1	3	5	2	86.92	86.12
12	3	2	4	1	3	74.04	79.06
13	3	3	5	2	4	75.77	77.19
14	3	4	1	3	5	99.99	99.98
15	3	5	2	4	1	95.44	98.87
16	4	1	4	2	5	78.86	76.61
17	4	2	5	3	1	61.91	63.25
18	4	3	1	4	2	99.99	99.98
19	4	4	2	5	3	96.87	98.8
20	4	5	3	1	4	97.67	94.6
21	5	1	5	4	3	39.72	38.86
22	5	2	1	5	4	91.02	93.42
23	5	3	2	1	5	87.10	84.88
24	5	4	3	2	1	71.62	70.42
25	5	5	4	3	2	76.44	74.47

Table-3: Parameters and their levels for second design.

	Parameters	Levels		
		1	2	3
A	Reaction Temperature (°C)	50	60	70
B	Solid-to-Liquid Ratio (g/mL)	0.15	0.2	0.25
C	Particle Size (µm)	-355+212	-212+150	-150 +125
D	Reaction Time (minute)	30	45	60

Table-4: Experiment plan and its results for second design.

Experiment Number	A	B	C	D	%B_2O_3 Experiment 1	%B_2O_3 Experiment 2
1	1	1	1	1	95.27	97.83
2	1	2	2	2	99.90	99.98
3	1	3	3	3	95.64	96.96
4	2	1	2	3	99.96	99.90
5	2	2	3	1	99.99	98.07
6	2	3	1	2	85.70	91.20
7	3	1	3	2	98.65	99.90
8	3	2	1	3	95.74	98.10
9	3	3	2	1	98.29	99.99

Result and Discussion

Dissolution Reactions

Reactions occurring during the dissolution process are

$$4SO_2 + 4H_2O \rightarrow 4H_2SO_3 \qquad (8)$$

$$4H_2SO_3 + 4H_2O \rightarrow 4H_3O^{+} + 4HSO_3^{-} \qquad (9)$$

$$2CaO.3B_2O_3.5H_2O + 4H_3O \rightarrow 2Ca^{+2} + 2H_2O + 6H_3BO_3 \qquad (10)$$

The total reaction is as follows

$$2CaO.3B_2O_3.5H_2O + 6H_2O + 4SO_2 \rightarrow 2Ca^{+2} + 4HSO_3^{-} + 6H_3BO_3 \qquad (11)$$

Statistical Analysis

The collected data were analyzed with the MINITAB software. For determination effective parameters on the dissolution process and their confidence levels was found with an analysis of variance. The F-Test is a tool used to see which process parameters have a significant effect on the dissolution value. Generally, the larger the F-value, the larger the effect of the parametres on the dissolution process. The optimal combination of process parameters with the performance characteristics and ANOVA analyses can be predicted [1]. The results of variance analysis for first stage and second stage experiments are given in Table-5 and Table-6.

SN_L equations were used to determine the parameter levels that maximize the B_2O_3 value passing through the solution from the colemanite ore. To determine the parameter levels that maximize the dissolution of colemanite with SO_2-saturated aqueous media, SN_L values were found from equation 1 by using dissolution fractions. The results obtained from the first design by calculating the SN_{ort} marginal average performance statistical values for the parameter levels with these SN values are shown graphically in Fig. 3 and in Fig. 4.

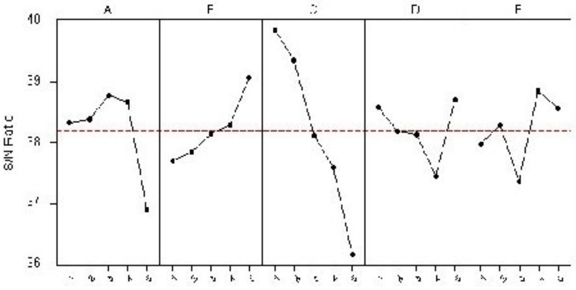

Fig. 3: SN_{ort} values according to parameter levels for the first design.

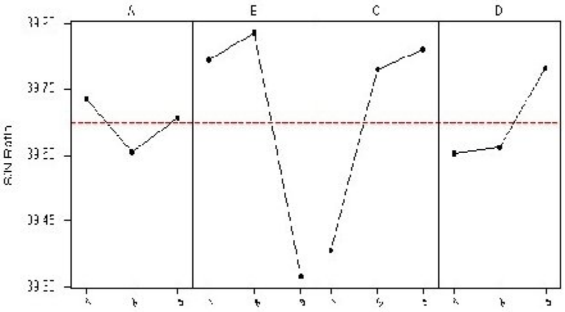

Fig. 4: SN_{ort} values according to parameter levels for the second design.

Table-5: Analysis of variance for the first stage optimization experiment.

	Parametreler	Degrees of Freedom	Sum of Squares	Mean of Squares	F	Efficiency
A	Reaction Temperature (^oC)	4	1419.24	354.81	20.74	Effective
B	Particle Size (μm)	4	817.25	204.31	11.94	Effective
C	Solid-to-Liquid Ratio ($g.mL^{-1}$)	4	6790.35	1697.59	99.23	Effective
D	Stirring Speed (rpm)	4	457.29	114.32	6.68	Effective
E	Reaction Time (minute)	4	669.80	167.45	9.79	Effective
	Error	29	496.12	17.11		
	Total	49	10650.05			

Table-6: Analysis of variance for the second stage optimization experiment.

	Parametreler	Degrees of Freedom	Sum of Squares	Mean of Squares	F	Efficiency
A	Reaction Temperature (^{o}C)	2	22.985	11.448	3.27	Not Effective
B	Solid-to-Liquid Ratio ($g.mL^{-1}$)	2	58.568	29.284	8.36	Effective
C	Particle Size (µm)	2	122.192	61.096	17.43	Effective
D	Reaction Time (minute)	2	26.543	13.271	3.79	Not Effective
	Error	9	31.545	3.505		
	Total	17				

Fig. 2A shows the variation of SN_L with reaction temperature. The optimum condition for the first data point is level 3 which is 60°C for A parameter. The experiments that has 3. level for A parameter (from Table-2) are experiments no 11, 12, 13, 14 and 15. The value of the third data point is thus the average of results obtained from experiments 11, 12, 13, 14 and 15. All the points in 2A, 2B, 2C, 2D and 2E graphs and other 3A, 3B, 3C and 3D graphs are established by the same way. For each plot, the numeric value of the maximum point refers to the best value for that parameter. When we look at Fig. 3 for the first stage experiments, These values is seen to be A3(60 ◦C), B5 (-355+212 µm), C1 (0,05 g/mL), D5 (700 rpm) and E4 (60 min.) Therefore, for first stage experiments A3, B5, C1, D5 and E4 conditions were determined as optimum dissolution conditions and the dissolution fraction under these conditions was found to be 99.36%. If the experimental plan given in Table-1 is examined carefully, it can be seen that experiments in optimum conditions have not been carried out during the experimental work. The results of this optimization experiments and statistical analysis (Table-5) showed that solid-to-liquid ratio and reaction temperature were the most effective parameters. Although this conditions is the optimum, We was aimed to reach 100% dissolution at high solid / liquid ratios by keeping the mixing speed at 300 rpm and changing other parameters, since working with high solids-liquid ratios is a preferred factor in industrial scale. A second design that is given parameter ve their level in Table-3 and experimental results in Table-4 has been prepared for this purpose and the figure drawn from the results obtained from this design is given in Fig. 4.

When Fig. 4 is examined, it is seen that the parameter levels that make the SN_{ort} value maximum are A1, B2, C3, D3. Therefore, parameter values that maximize the dissolution rate of colemanite will be A1, B2, C3 and D3. Optimum conditions was found as reaction temperature(50°C), the solid / liquid ratio(0.2 $g.mL^{-1}$), the particle size(-150 µm+125 µm) and the reaction time(60 minutes). Under optimal conditions, the estimated B_2O_3 passing through the solution is 100% and the experimental value is 99.8%.

To work by using a high solids-to-liquid ratio and large particle size and by shortening the reaction time is the reason for preference in industrial scale processes. Therefore, parameter levels was selected as 50°C for the temperature, 0.25 g / ml for the solid liquid ratio, 30 minutes for the reaction time and -212 µm +150 µm for the particle size and 300 rpm for stirring speed. In the experiment at these condition we did, we found that the estimated B_2O_3 as 98.9% and the experimental value as 98.5%. In addition to a reduction of about 1%, A1, B3, C2, D1 were taken as optimal values considering that higher the solid / liquid ratio, larger particle size (lower the grinding costs) and shorter reaction time. Thus, it should be noted that values in Table-7 and 8 are predicted results by using Equation (2) and the observed results for the same conditions. Also, the results in Table 7 and 8 are confidence limits of predictions for $\%B_2O_3$. In order to confirmation the predicted results, confirmation experiments were carried out twice at the same working conditions. The percentages of dissolution obtained from the validation experiments are within the confidence interval calculated from Equations (3-7) (see Tables 7 and 8) showed that the test results are within ± 5% error range [21].

Conclusion

Solid-to-liquid ratio, particle size, reaction temperature, stirring speed and reaction time were used as parameters in dissolution experiments. The reaction is shown below.

$$2CaO.3B_2O_3.5H_20 + 6H_2O + 4SO_2 \rightarrow 2Ca^{+2} + 4HSO_3^{-} + 6H_3BO_3 \quad (11)$$

On the other hand, by-product in this process is $Ca(HSO_3)_2$ and $CaSO_3.xH_2O$. When the solution is heated, $Ca(HSO_3)_2$ decomposes and

$$Ca(HSO_3)_{2(s)}+(x-1)H_2O \rightarrow CaSO_3.xH_2O_{(k)}+SO_{2(g)} \quad (12)$$

According to the reaction (12) calcium sulphide precipitate. It would be possible to obtain the product boric acid in high purity because the by-product formed in the reaction (The calcium sulfide solubility is considered to be 0.0043 g in 100 mL of water at 18°C) will remain in solid waste. SO_2 formed in this reaction can be used again. If necessary, SO_2 can be reused by treating $CaSO_3.xH_2O$ with cheap acids or by roasting of $CaSO_3.xH_2O$. The XRD graph of the residue remaining after the reaction is shown in Fig. 5.

In the case of acid use,

$$CaSO_3.xH_2O + 2H^{+} \rightarrow Ca^{+2} + (x+1)H_2O + SO_2 \quad (13)$$

In the case of roasting;

$$CaSO_3.xH_2O \rightarrow CaO + SO_2 + xH_2O \quad (14)$$

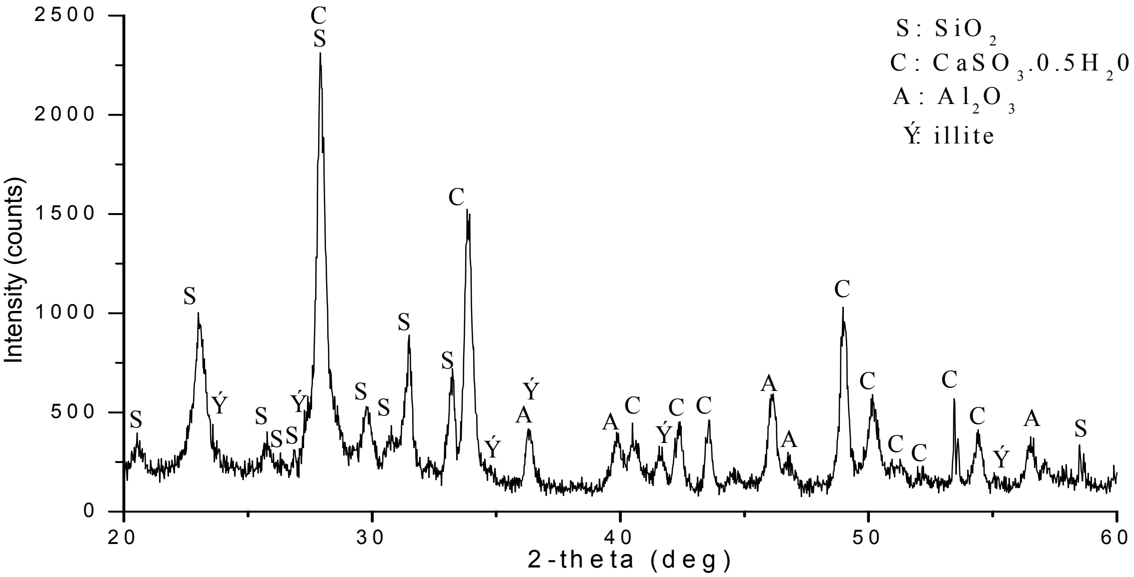

Fig. 5: The XRD graph of the residue remaining after the reaction.

Table-7: Optimum operating conditions in the first stage experiments, observed and predicted dissolved quantitiy for B_2O_3

	Parameters	Case 1*		Case 2**		Case3***	
		Level	Value	Level	Level	Value	Level
A	**Temperature (°C)**	**60**	**3**	**60**	**3**	**60**	**3**
B	**Particle Size (µm)**	**283.5**	**5**	**283.5**	**5**	**283.5**	**5**
C	**Solid-to-Liquid Ratio (g.mL^{-1})**	**0.05**	**1**	**0.2**	**4**	**0.25**	**5**
D	**Stirring Speed (rpm)**	**300**	**1**	**300**	**1**	**300**	**1**
E	**Reaction Time (minute)**	**60**	**4**	**60**	**4**	**60**	**4**
	Observed value for B_2O_3 (%)	**100**		**95**		**93**	
	Predicted value for B_2O_3 (%)	**100**		**97**		**89**	
	Confidence limits of prediction for B_2O_3 (%)	**89.32-100.00**		**86.32-00.00**		**78.32-99.68**	

*The parameter levels for maximum dissolution of B_2O_3
** Alternative optimum conditions
*** Optimum conditions selected

Table-8: Optimum operating conditions in the second stage experiments, observed and predicted dissolved quantitiy for B_2O_3

	Parameters	Case 1*		Case 2**		Case3***	
		Value	Level	Value	Level	Value	Level
A	**Temperature (°C)**	**50**	**1**	**50**	**1**	**50**	**1**
B	**Solid/Liquid Ratio (g.mL^{-1})**	**0.20**	**2**	**0.2**	**2**	**0.25**	**3**
C	**Particle Size (µm)**	**137.5**	**3**	**181**	**2**	**181**	**2**
D	**Stirring Speed (rpm)**	**300**	**--**	**300**	**--**	**300**	**--**
E	**Reaction Time (minute)**	**60**	**3**	**30**	**1**	**30**	**1**
	Observed value for B_2O_3 (%)	**100**		**100**		**98.5**	
	Predicted value for B_2O_3 (%)	**100**		**100**		**98.9**	
	Confidence limits of prediction for B_2O_3 (%)	**95.16-100.00**		**95.16-100.00**		**94.06-100.00**	

*The parameter levels for maximum dissolution of B_2O_3
** Alternative optimum conditions
*** Optimum conditions selected

It is thought that sulfur dioxide can be formed according to the above reactions and can be used again in the process.

On the other hand, the boric acid obtained as a result of the reaction of colemanite with SO_2 will give clearer boric acid crystals than the sulfuric acid process, since it is not sulphate in the environment and the solubility of calcium sulphite is about 100 times less than that of calcium sulphate. In this respect, at centrifuges washing of crystal boric acid will not be required to, or less washing will be sufficient. The suspension obtained by dissolution of crude colemanite in water saturated with SO_2 was easily filtered and boric acid-containing solutions were obtained. Obtained solution can be more easily filtered than solution containing clay and $CaSO_4$ solids.

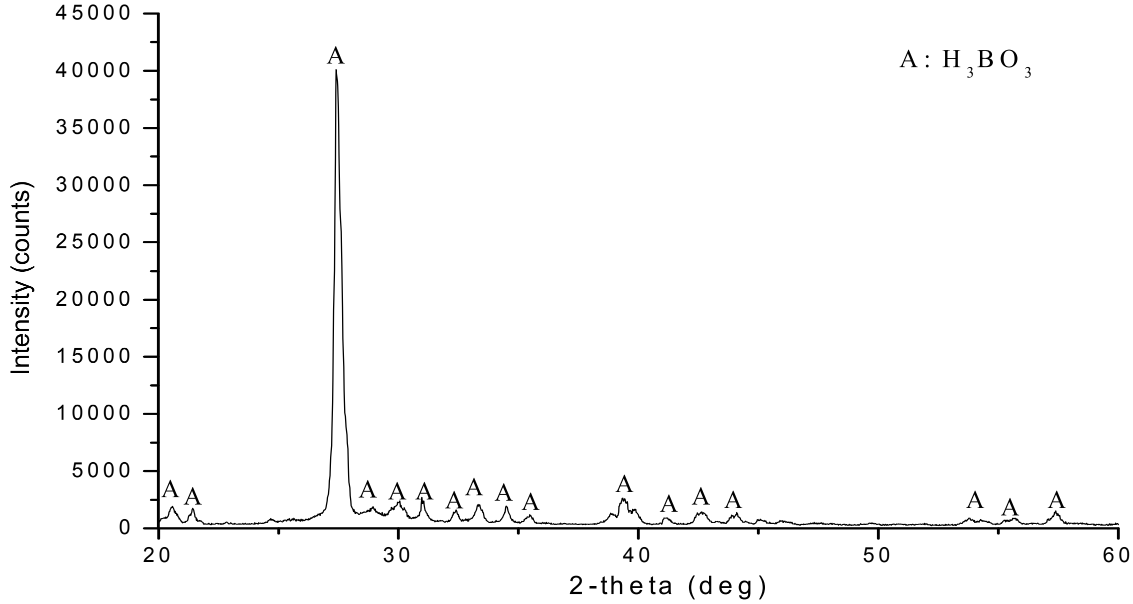

Fig. 6: XRD Graph of the boric acid obtained under the optimum conditions.

As a result, the optimal conditions for the parameters and levels used was obtained a reaction temperature of 50°C, a solid-to-liquid ratio of 0.25 g / mL, a reaction time of 30 minutes, a particle size of -212 µm+150 µm and a stirring speed of 300 rpm. The boric acid purity obtained under the optimum conditions was found to be 96.4% and XRD graph of product are also given in Fig. 6. Findings of this work can be very useful for industrial scale enterprises since optimum conditions obtained by Taguchi method in laboratories can also be produced in real production environments.

Nomenclature

SN_L	performance characteristics for Larger-the-better
SN_S	performance characteristics for Smaller-the-better
Y_i	performance value of ith experiment
µ	the overall mean of performance value
X	the fixed effect of the parameter level combination used in ith experiment
e_i	the random error in ith experiment
	Ω(db) the decibel value of percentage value subject to omega transformation
P	the percentage of the product obtained experimentally
S	the two-standard-deviation confidence limit
n	the number of rows in the matrix experiment,
n	the number of repetition for confirmation experiment or experimental combination
	n_{Ai}, n_{Bi}, n_{Ci}, the replication number for parameter level Ai, Bi, Ci, …

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