JOURNAL OF SCIENCE
SAKARYA UNIVERSITY

Sakarya University Journal of Science

ISSN 1301-4048 | e-ISSN 2147-835X | Period Bimonthly | Founded: 1997 | Publisher Sakarya University | http://www.saujs.sakarya.edu.tr/

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Recieved: 2019-03-19 16:18:44

Accepted: 2019-05-07 13:35:45

Article Type: Research Article
Volume: 23
Issue: 5
Month: October
Year: 2019
Pages: 916-923

Sakarya University Journal of Science 23(5), 916-923, 2019

An Experimental Study on Extraction of Sugar from Carob Using by Taguchi Method

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Abstract

The carob, a perennial plant, is grown in regions where the Mediterranean climate is dominant. Because of its properties, it is preferred in many sectors especially in food and health sectors. Carob is mostly used as sugar and molasses. Moreover, seeds of carob are also used as thickening material in the food sector and its residue is utilized as animal feeding stuff as well. Molasses are mostly produced from carob, grape, mulberry, apple and pear for many years in Turkey. Molasses production is different from each other with regard to the structure of each fruit. Molasses are produced by pressing in the event of soft. The molasses of hard dried fruits such as carobs which cannot be pressed are obtained by extraction method using water. The parameters which are important for the quality of product such as extraction temperature and duration are not taken into account in the carob molasses obtained by traditional methods. Increasing the amount of water in extraction decreases production efficiency and quality while increasing energy consumption. In this study, the effects of parameters such as water quantity, extraction temperature, duration and batch number that affect quality of molasses were experimentally investigated by using Taguchi method.

Keywords: sugar extraction, mass transfer, batch system, taguchi method.

1. INTRODUCTION

The Latin name of Ceratonia silique L. is hard carob. The carob has been known for more than 5000 years. Before the production of white sugar, the carob was mostly used in the pastries and desserts. It is grown in regions where the Mediterranean climate dominates such as İzmir, Antalya, Cyprus and Mersin. There are long, straight and curved types of carob. The carob has 10 to 30 cm longness, 1.5 to 3.5 cm wideness and about 1 cm thickness. Since the carob is consumed as dried, after harvesting, it is desiccated one to two months. The carob is usually consumed as carob molasses in the food industry. The core of carob is also used in ice cream production and in fruit concentrates since it increases its consistency. The carob gum is used in textile field in order to provide an equal

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permeability in cotton fabrics. It is a very effective fruit for allergic asthma. It has many properties such as antioxidant, anti-carcinogenic, antiseptic, and antiallergic. Due to the chemical and pharmacological properties of carob, it has been known the beneficial and medical properties especially in the digestive system [1]. It is frequently preferred in industry because of its remarkable properties. For this reason, it is aimed to obtain sugar from the carob fruit without waste of energy and resources. There are many studies in the literature on HMF formation, extraction and liquid sugar production.

N. Basak Ozhan [2] examined the kinetics of non-enzymatic browning reactions of carob molasses during storage. She investigated the browning reaction of carob molasses for 4 different grades. As a result of storage in the research, no change was observed in water activity and brix values, whereas the acidity values decreased at 25ºC, 35ºC and 45ºC. It is indicated that the reaction rates of HMF and browning reactions increase with the temperature. S. Khatib and J. Vaya [3] examined the effects of the carob on human health. The carob pulp has been determined that it is rich in soluble fibers and it has reduction ability of total and LDL cholesterol on the hypercholesterolemia patients. A. Mulet et al. [4] investigated the importance of grain size in the extraction of the carob. It was observed that the effect of time and temperature was higher in the use of small grain size in carob extraction. I. Turhan et al. [5] determined soluble quantitative of the total mass transfer coefficient and total phenolic components with carob extraction. The efficiency of the process temperature was determined using the Arrhenius equation. The activation energies were calculated as 5.84 and 0.072 $x10^{-3}$ kJ/mol for the diffusion of soluble mass and total phenolic components. Higher concentrations were obtained at higher temperatures. Although it has high temperature, it is stated that it does not cause HMF formation. F. Senay [6] studied the optimum extraction value and the clarification conditions for the production of liquid sugar from the carob. The effects of the amount of water, the extraction temperature, the particle size, the mixing and pressing on the efficiency were investigated. The titration acidity, pH value, total dry matter and the amount of moisture were also examined. The highest efficiency was obtained at 90 ºC, in the rate of 1: 4 water, in the small particle size and at 3 hours. It was applied mixing and pressing in the extraction process. O. Demirtas [7] studied on the production of carob gamut from the carob beans and the physical and on chemical properties of these gums. For the production of carob gamut, it was kept in various solutions at 75 °C for an hour and was washed, then it was dried at 30 °C for a day and it was grained in a mill. In order to remove the shell of the carob kernel were used various compounds such as HCl, H_2SO_4 and NaOH. As a result of this, the moisture, protein, ash, insoluble matter in the acid and viscosity values of the carob gamut were compared. The highest viscosity was obtained from HCl, while the content of the insoluble material in the acid was highest in NaOH. The negative effects of these chemicals are also compared. M. Yalcin Yilmaz [8] tried to obtain clear sugar from the carob. The seeds of the carob were removed and carob pods were cut in different sizes. The clarification process was carried out at different water rates and temperatures. Kizelsoldan, bentonite, and gelatin were used in the clarification process. After the removal of color and turbidity, it was concentrated in vacuum evaporation and its rheological properties were determined.

U. Yurdagel and I.Teke [9] examined the roasting trials in different sizes, different temperatures and different times. The carob bean pieces of 3 to 6 mm size were found to be suitable for roasting. After roasting at 160 ºC and for 30 minutes, the darkest carob powder was obtained according to Hunterlab measurements. I. Turhan [10] kept constant the carob amount and the solvent flow rate, then extraction of the carob was done depending on temperature. The experiments were carried out at 20 ºC, 50 ºC, 85 ºC with continuous extraction. The increase of soluble dry matter, phenolic compound diffusion and 5-HMF formation were determined in extraction. The titration acidity, crude fiber content, pH value and total amount of nitrogenous substances were analyzed. B. K. Tiwari [11] investigated the ultrasonic extraction method which is used with the aim of extraction and harmless to the earth. It was indicated that it increases the extraction efficiency and the aqueous extraction process without the use of solvents. For the carob fruit, the conditions of the ultrasound-assisted extraction method are stated as 40 kHz frequency, 45-297 W power, at 30 to 50 ºC, 30 to 120 minutes. N. Tetik and E. Yuksel [12] studied on the optimization of extraction conditions of D-pinitol compound by using carob ultrasound and four independent variables. It was stated that these variables are temperature, ultrasonic power, dilution rate and time. As a result of the experiments, the highest D-pinitol concentration was obtained at 50 ºC temperature, 207 W ultrasonic power, at 1: 4 dilution rate and at 120 min extraction time. It was stated that ultrasound assisted extraction can be used as an alternative to conventional hot water because of increasing the concentration of D-pinitol by ultrasonic action during extraction. B. A. Sarvin et al. [13] studied on the ultrasound-assisted extraction method for isolation of 17 sugars and sugar alcohols from conifers. The methanol concentration in the extraction solution, the extraction time, the type of plant sample and the extraction temperature was investigated as extraction parameters using Taguchi method. The optimum extraction conditions were obtained as 30% MeOH concentration, 30 minutes, plant sample type-II and 60 ºC. The proposed optimum parameters can be used for profiling of sugars and sugar alcohols in a wide range of plant species. A. W. Go et al. [14] studied on Taguchi method to improve the production of sugar which is rich in hydrolysate from non-dilapidated spent coffee grounds. A sugar recovery of %81 to %98 of the available sugars was achieved using 4% v/v sulfuric acid at an SSR of 8 mL/g for a hydrolysis time of 3 to 4 hours at 95 ºC. They suggest that their approach allows a potential energy savings as much as 48% according to conventional approaches. There are more studies about the extractions, Taguchi methods, and the heat and mass transfers in the literature [12], [15]–[18].

In this study, it is aimed to obtain maximum sugar from carob by using Taguchi method with optimum level of parameters which are water quantity, temperature, duration, and batch number. Three different levels are determined for each parameters. At 30 ºC, 35 ºC, 40 ºC, and at 100 (g), 200 (g), 250 (g) water, 30, 60, 90 minutes were studied on the time periods. Batch number is added as a parameter to determine the optimum and sufficient amount. The effects of parameters on the efficiency and the rate of S/N are shown in Fig. 3. At the end of the experiments, the optimum conditions which should be applied according to the efficiency of the sugar amount were determined.

2. MATERIALS AND METHODS

Taguchi method is used to minimize the number of experiments. Thus, the best test results can be obtained without having to do all the experiments. L9 orthogonal array and four parameters were used. Each parameter has three levels. The orthogonal array and the numerical value of each level are shown on Table 1.

Table 1. Controllable factors and their levels

Run	Water(g)	Temp.(°C)	Duration(min.)	Batch
T1	Level 1 100	Level 1 30	Level 1 30	Level 1 4
T2	Level 1 100	Level 2 35	Level 2 60	Level 2 5
T3	Level 1 100	Level 3 40	Level 3 90	Level 3 6
T4	Level 2 200	Level 1 30	Level 2 60	Level 3 6
T5	Level 2 200	Level 2 35	Level 3 90	Level 1 4
T6	Level 2 200	Level 3 40	Level 1 30	Level 2 5
T7	Level 3 250	Level 1 30	Level 3 90	Level 2 5
T8	Level 3 250	Level 2 35	Level 1 30	Level 3 6
T9	Level 3 250	Level 3 40	Level 2 60	Level 1 4

Four batch experiment scheme is shown in Fig. 1 for better understanding of batch system.

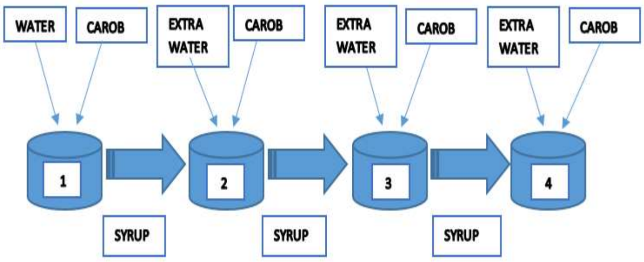
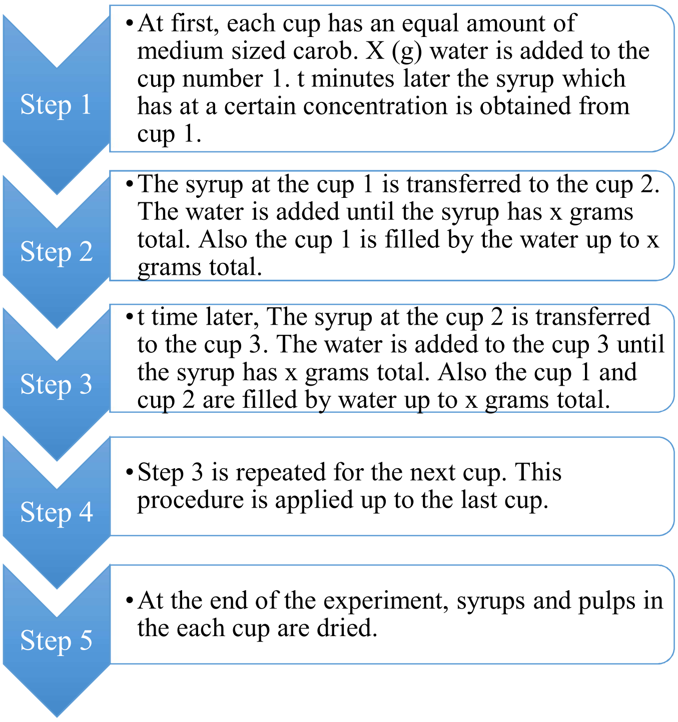


Figure 1. Four batch experiment scheme.

A step-by-step description of a four-batch experiment is shown below. Here, X (water quantity) and t (time) represent any level of the parameters.



During the experiments, thermos cups were held in the styrofoam box to the temperature. At the end of the runs, water of the syrup of each cup was evaporated by drying at 105 ºC. The dried syrups for six batches are shown in Fig. 2.

Figure 2. The picture of medium sized carob and the dried syrups for six batches

Errors in temperature measurement: Error due to the structure of the glass thermometer: ±0.25 ºC. Error in measuring the temperature of the test environment: ±0.25 ºC. Error in temperature measurement in drying oven: ±0.5ºC. Error in the mass measurement of the scales: ± 1 g.

According to these error values, uncertainty analysis can be obtained with the following equation [19].

$w_1, w_2, w_3, \ldots, w_n$ are uncertainties.

The result R is function of the independent variables ($x_1, x_2, x_3, \ldots, x_n$).

$R = R(x_1, x_2, x_3, \ldots, x_n)$

$$W_R = \left[\left(\frac{\partial R}{\partial x_1} w_1\right)^2 + \left(\frac{\partial R}{\partial x_2} w_2\right)^2 + \cdots + \left(\frac{\partial R}{\partial x_n} w_n\right)^2\right]^{1/2} \quad (1)$$

It can be calculated with the equation presented by Kline and Mc. Clintock. Errors according to the total temperature measurement:

$$W_R = [(0.25)^2 + (0.25)^2 + 0.5^2]^{1/2} \quad (2)$$

The total error according to the temperature measurement: ± 0.612 ºC

3. RESULTS AND DISCUSSION

As a result of the experiments, the total amount of sugar in each experiment was obtained as follows as in Table 2.

Table 2. The results of experiments

Run	1st Exp.	2nd Exp.	3rd Exp.
1	34	35	30
2	51	52	53
3	63	70	71
4	91	90	89
5	58	60	59
6	65	70	72
7	83	82	81
8	89	91	90
9	58	61	64

According to the Taguchi method, it is necessary to calculate the S / N ratio of each experiment to reach the results. There are 3 equations developed for the S / N ratio.

First equation is ‘Larger is better’.

$$S/_N = -10 \times \log\left(\frac{1}{n}\sum_{i=1}^{n}\frac{1}{y_i^2}\right) \qquad (3)$$

‘y_i’ is the yield of sugar extraction.

Second equation is ‘Smaller is better’.

$$S/_N = -10 \times \log\left(\frac{\sum_{i=1}^{n} y_i^2}{n}\right) \qquad (4)$$

Third equation is ‘Nominal is best’.

$$S/_N = 10 \times \log\left(y^{-2}/_{s^2}\right) \qquad (5)$$

The S/N values of the results are calculated from Eq. 3. The calculated S/N ratios for each experiment is shown in Table 3.

Table 3. Yield of the experiments and S / N ratio

Run	1st Exp. (%)	2nd Exp. (%)	3rd Exp. (%)	S/N
1	25.00	25.73	22.05	27.64
2	30.00	30.58	31.17	29.70
3	30.88	34.31	34.80	30.42
4	44.60	44.11	43.62	32.89
5	42.64	44.11	43.38	32.74
6	38.23	41.17	42.35	32.14
7	48.82	48.23	47.64	33.66
8	43.62	44.60	44.11	32.89
9	42.64	44.85	47.05	33.01

Table 4 shows the effects of each parameter on the yield of extraction.

Table 4. Response Table for S/N

Level	Water	Temperature	Duration	Batch
1	29.25	31.40	30.89	31.13
2	32.59	31.78	31.87	31.84
3	33.19	31.86	32.28	32.07
Delta	3.93	0.46	1.39	0.93
Rank	1	4	2	3

According to Table 4, the parameters influenced the sugar extraction, respectively, are water quantity, duration, batch, temperature. If the first level of water quantity is increased to second level (200 g), S/N ratio increases from 29.25 to 32.59. S/N ratio is also achieved 33.19 on third level. In other words, it is clearly that the water quantity is the most effective parameter for the sugar extraction. If the S / N ratios put in equation 3, the effect of parameters on yield can be calculated. For the water quantity; the yield of extraction is determined about % 29 on the first level (100 g). The yields of extraction on the second and third levels, respectively are determined % 42.60 and % 45.65. The yield of extraction for the other parameters can be seen at the Table 5.

Table 5. The effects of the parameters as yield of extraction

Level	Water %	Temperature %	Duration %	Batch %
1	29	37.15	35.03	36.01
2	42.60	38.81	39.21	39.08
3	45.65	39.17	41.11	40.13

Analysis of variance is shown in Table 6.

Table 6. Analysis of variance

Source	DF	Adj SS	Adj MS	F-Value	P-Value
Regression	4	30.4980	7.6245	23.33	0.005
Water	1	25.9905	25.9905	79.52	0.001
Temperature	1	0.3183	0.3183	0.97	0.380
Duration	1	2.8789	2.8789	8.81	0.041
Batch	1	1.3102	1.3102	4.01	0.116
Error	4	1.3074	0.3268		
Total	8	31.8054			

Fig. 3 that shows the effect of the parameters on the extraction was obtained by using Minitab program. Here, the inclinations of each parameter in the graphs show the effect of the level differences of the parameters. According to the Fig. 3, the highest S/N ratio was obtained in the combination of 3.3.3.3 as an expected result because 'larger is better' function was chosen. However, since the inclinations for levels of the parameters in sugar extraction are more important, it is suggested that the optimal combination is 3.1.3.2. The combination of 3.1.3.2 is included in the experiments. Therefore, there is no need to clarification test.

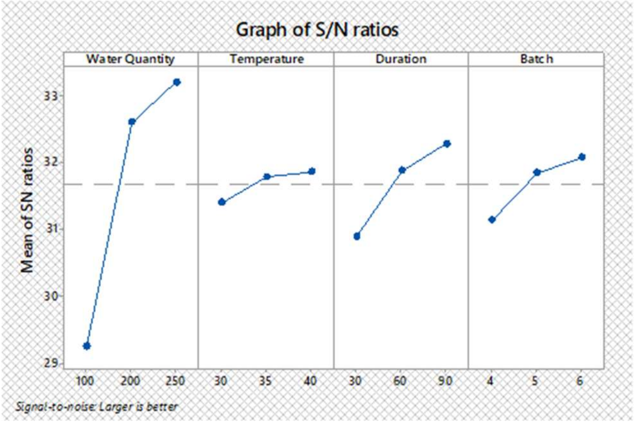


Figure 3. Graph of S/N ratios

The yield and the amount of obtained sugar can be calculated by the below equation which is depending on levels of parameters.

$$F = 3.26786 + 0.11236Water + 0.07200Temperature + 0.08880Duration + 1.51111Batch \quad (6)$$

Here, F is proposed equation. The yield of extraction can be calculated as a percentage by using this equation.

4. CONCLUSION

In this study, the sugar was recovered by extraction method from carob using Taguchi method. It is aimed to determine the optimum extraction conditions in order to consume less energy and resources. The yield of sugar is clearly depending on the extraction conditions. The effects of four parameters which are the water quantity, the temperature, the duration and the batch number were examined as the extraction conditions. It is proposed an equation depending on the parameters to obtain the yield of extraction. According to these results, it was found that the most effective parameter was the water quantity. The duration was found as an important parameter as the water quantity in the experiments. It is seen that the water quantity and duration were more effective than the other two parameters. The batch number was found to be more effective than the temperature. The effects of the temperature and the number of batch also increased the yield of extraction. However, the caramelization of sugar at high temperatures should be taken into consideration. Maximum efficiency of 49.67%

was obtained for the conditions of 250 (g) water, at 40 ºC, 90 minutes, 6 batch. On the other hand, of 48.23% efficiency was obtained for the conditions of 250 (g) water, at 30 ºC, 90 minutes, 5 batch. Since the difference is very small, the conditions that of 48.23% yield is achieved can be considered as the optimum condition.

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