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Effect of Ultrasonic Treatment on the Solubility of Sodium Sulphate Using Response Surface Methodology

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The effect of ultrasonic treatment on the solubility of sodium sulphate was investigated in this study. The influence of several single-factor experiments including temperature, ultrasonic time, ultrasonic power and rotation rate on the solubility of sodium sulphate was carried out. Response surface methodology (RSM), based on a three level, three variable central composite design (BBD), was employed to obtain the best possible combination of temperature (X_1), ultrasonic time (X_2) and ultrasonic power (X_3) for maximum solubility according to the results from single-factor experiment. The 3-D response surface plot and the contour plot was derived from the mathematical models. The results also showed that the best conditions to attain the maximum solubility are met when temperature was 35 ºC, time was 20 min, power was 140 w. Under this condition, the experimental yield was 52.6321 g, which was well matched with the predictive yield.

Key Words: Sodium sulphate, Crystallization, Ultrasonic wave, Response surface methodology, Solubility.

INTRODUCTION

As is well known, crystallization is an attractive separation technique for manufacturing because of the advantages of low energy consumption, high efficiency and low pollution[1]. Crystallization out of solutions is a major separation and purification unit operation, which is applied in many fields of chemical and pharmaceutical industries. Crystallization in general can be divided into nucleation and growth. Nucleation may be compared to birth. However, before nucleus formation, the mother phase must be supersaturated or supercooled. This is the thermodynamic driving force for crystallization[2]. Sonocrystallization[3-6] is the use of power ultrasound to control the course of a crystallization process. Recently, sonocrystallization has received much attention. Several research groups used ultrasound to control solubility. Wang *et al.*[7] have used ultrasound to improve the solubility of Wheat Gluten, followed in turn by pulse "on" and "off" time cycle. Laugier *et al.*[8] have investigated the effect of ultrasound on solubility in gas-liquid systems. Jambrak *et al.*[9] also observed the effect of ultrasound and sonication on whey proteins in order to improve their function properties. Since Na^+ (aq) and SO_4^{2-} (aq) are very common components of natural fluids, their thermodynamic properties in aqueous solution is of crucial importance in understanding various industrial and geochemical process such as crystallization[10]. Thus sodium sulphate was used to be the principal raw material.

Response surface methodology (RSM), which combines statistical and mathematical techniques is useful for developing, improving and optimizing processes[11-15]. The main advantage of response surface methodology is the reduced number of experimental trials needed to evaluate multiple parameters and their interactions. Therefore, it is less laborious and time-consuming than other approaches required to optimize a process.

Sodium sulphate was used to observe the effect of ultrasound on solubility. The objective of this study is to improve the solubility of sodium sulphate by ultrasonic using response surface methodology.

EXPERIMENTAL

Single-factor experiment: Balance method was applied to get the solubility of sodium sulphate. The solubility of sodium sulphate as target and selecting temperature (20, 30, 40, 50, 60, 70, 80 and 90 ºC), time (15, 20, 25, 30, 35, 40 and 45 min), power (80, 100, 120, 140, 160, 180 and 200 w) and rotational velocity (200, 300, 400, 500, 600 and 700 r/min) as single-factor.

Design of statistical experiments: After determining the preliminary range of solubility through single-factor test, a three-level-three-factor, Box-Behnken factorial design (BBD)[16] was adopted in this optimization study. Temperature (X_1), time (X_2), power (X_3) were the independent variables selected to be optimized for the solubility of sodium sulphate. The range of independent variables and their levels were presented in Table-1. Solubility (s) was taken as the response for the combination of the independent variables given in Table-2. Analysis of the experiment design and calculation of predicted data were carried out by using Design-expert Software 7.1.3 to estimate the response of the independent variables.

TABLE-1
INDEPENDENT VARIABLES AND THEIR LEVELS USED IN THE RESPONSE SURFACE DESIGN

Independent variables	Levels		
	-1	0	1
Temperature (X_1) (ºC)	20	30	40
Time (X_2) (min)	15	20	25
Power (X_3) (w)	100	120	140

TABLE-2
BOX-BEHNKEN EXPERIMENTAL DESIGN AND RESULTS FOR SOLUBILITY

No.	X_1 (temperature)	X_2 (time)	X_3 (w)	Solubility (g)
1	1	0	1	49.6587
2	1	-1	0	49.4631
3	-1	1	0	25.1603
4	0	0	0	50.0395
5	-1	-1	0	25.5767
6	0	-1	-1	46.7411
7	0	0	0	50.2680
8	0	0	0	49.4450
9	0	-1	1	49.1482
10	0	1	1	50.7356
11	-1	0	1	26.6351
12	0	1	-1	50.5951
13	0	0	0	49.6269
14	0	0	0	50.5756
15	-1	0	-1	30.0374
16	1	1	0	48.4399
17	1	0	-1	49.1540

RESULTS AND DISCUSSION

Effect of temperature on the solubility: Temperature is an important factor that could influence the solubility of sodium sulphate. Firstly, solubility curve was obtained without ultrasound. Then the effect of ultrasonic temperature on solubility was investigated when time is 20 min, power 120 w, rotational velocity is 300 r/min. From Fig. 1, it is found that the solubility increased as temperature ascended from 20-40 ºC, highest at *ca.* 35 ºC and no longer increased when the temperature exceeded 40 ºC. On the other hand, obviously ultrasound can increase solubility, but can not change the change trend. Ultrasound did improve the solubility and play an important role in crystallization process[17,18].

Effect of time on the solubility: Different ultrasound time from 15-45 min can also affect the solubility of sodium sulphate. Solubility increased along with the extension of time. When time was over 40 min, solubility would not have change obviously. The results were listed in Fig. 2.

Effect of power on the solubility: A higher power also presents a positive effect on the solubility of sodium sulphate. The solubility of sodium sulphate affected by different power is shown in Fig. 3. When the other three factors (temperature, time, rotational velocity) were 35 ºC, 20 min, 300 rpm, respectively. It showed that the solubility increased as the power ascended from 80-140 w. After this point, the solubility started to maintain a dynamic equilibrium with the increasing of power and no longer increased when power exceeded 140 w.

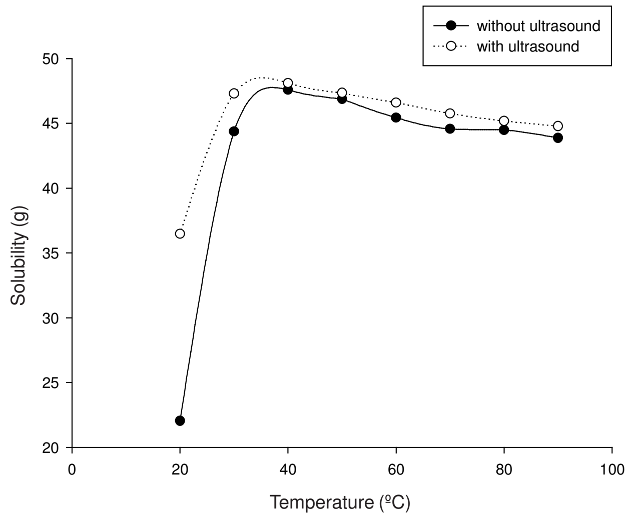


Fig. 1. Effect of temperature on the solubility with ultrasound and without ultrasound

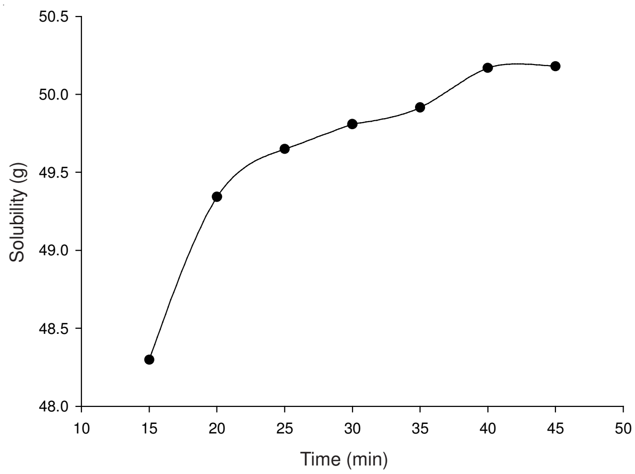


Fig. 2. Effect of time on the solubility

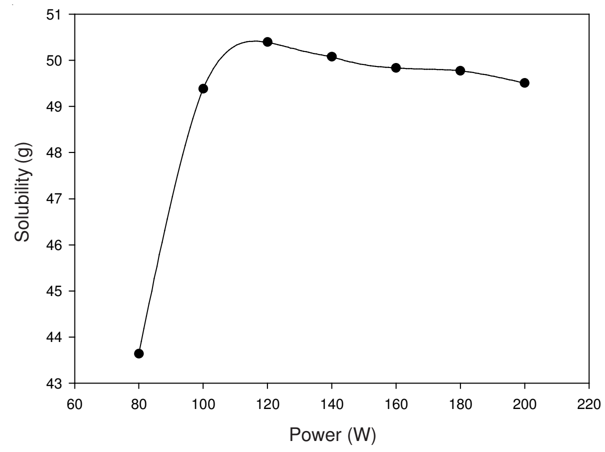


Fig. 3. Effect of power on the solubility

Effect of rotational velocity on the solubility: Fig. 4 shows a wavy line that the effect of rotational velocity on the solubility has no significant change. Therefore it need not consider this factor when solubility is studied in the future.

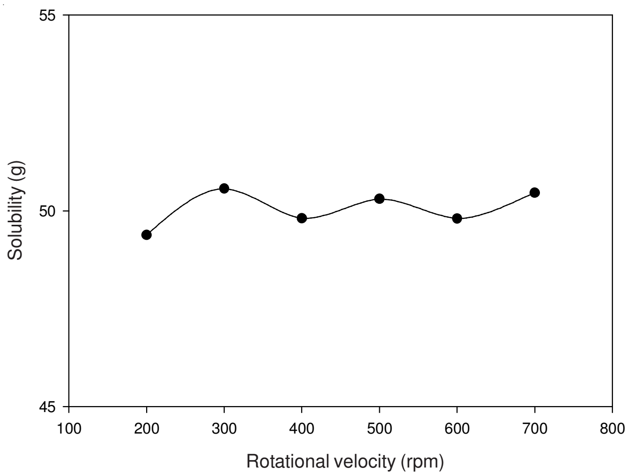


Fig. 4. Effect of rotational velocity on the solubility

Statistical analysis and model fitting: A multiple regression analysis of data was carried out to calculate the coefficients of the second order polynomial equation[19] proposed to correlate the response to the three parameters:

$$S = k_0 + \sum_{i=1}^{3} k_i \times X_i + \sum_{i=1}^{3} k_{ii} \times X_i X_i + \sum_{i<j}^{3} k_{ij} \times X_i x_j \quad (1)$$

By applying multiple regression analysis on the experimental datas, the following second order polynomial equation was found:

$$S = +49.99100 + 11.16327X_1 + 0.50023X_2 - 0.043750X3 - 0.15170X_1X_2 + 0.97675X_1X_3 - 0.56665X_2X_3 - 11.63235X_1^2 - 1.19865X_2^2 + 0.51265X_3^2 \quad (2)$$

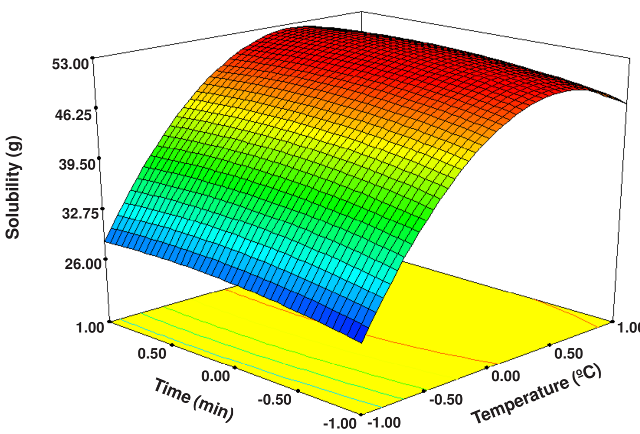
where S is the predicted response; X_1, X_2, X_3 are coded values of temperature, time and power, respectively.

The ANOVA data were analyzed to evaluate the significance of the different models equations associated with models parameters established by regression calculations to fit all of the polynomial models to the selected response. Table-3 sums up the test of significance for regression coefficient. The model F-value of 90.59 implies the model is significant. Values of "Prob > F" less than 0.0500 indicate model terms are significant. The "Lack of Fit F-value" of 20.01 implies the lack of fit is significant. The temperature is significant. No interaction among the parameters is significant. The order of influencing solubility that can be saw from Table-3 is: temperature > time > power. One top of that, the R^2 values were 99.15 %, confirming the excellent accuracy of the model in predicting the solubility.

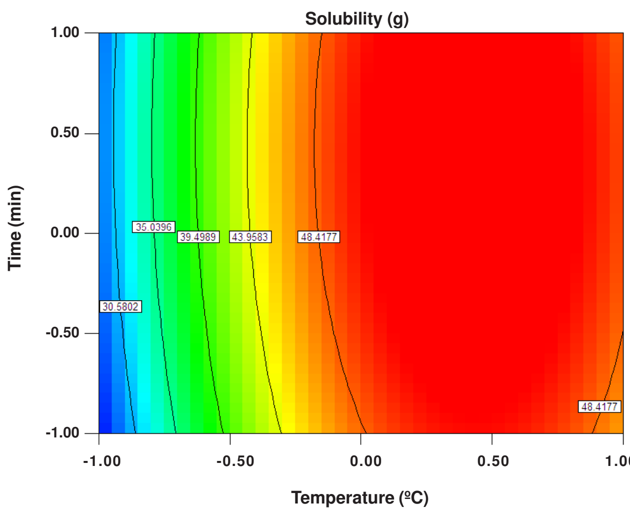
Optimization of solubility of sodium sulphate: Response surface optimization is more advantageous than the traditional single parameter optimization in that it saves time space and raw material. The 3D response surface plots described by the regression model were drawn to illustrate the effects of the independent variables and the interactive effects of each independent variable on the response variables. The shape of the corresponding contour plots indicates whether the mutual interactions between the independent variables are significant or not[20,21]. Fig. 5a-b shows the 3D plot and contour plot of the effect of the independent variables temperature and time on solubility. Fig. 6a-b depicts the 3D plot and contour plot, showing the effects of temperature and power on the solubility. Fig. 7a-b shows the 3D plot and contour plot of the effect of the independent variables time and power on solubility.

TABLE-3
REGRESSION COEFFICIENTS OF THE PREDICTED SOLUBILITY

Source	Sum of squares	Mean square	F-Value	P-Value
Modle	1587.91	176.43	90.59	<0.0001
X_1	996.95	996.95	511.87	<0.0001
X_2	2.00	2.00	1.03	0.3444
X_3	0.015	0.015	7.862E-003	0.9318
X_1X_2	0.092	0.092	0.047	0.8341
X_1X_3	3.82	3.82	1.96	0.2043
X_2X_3	1.28	1.28	0.66	0.4435
X_1^2	569.73	569.73	292.52	<0.0001
X_2^2	6.05	6.05	3.11	0.1214
X_3^2	1.11	1.11	0.57	0.4756



(a)



(b)

Fig. 5. 3D plot and contour plot of the effect of the independent variables temperature and time on solubility

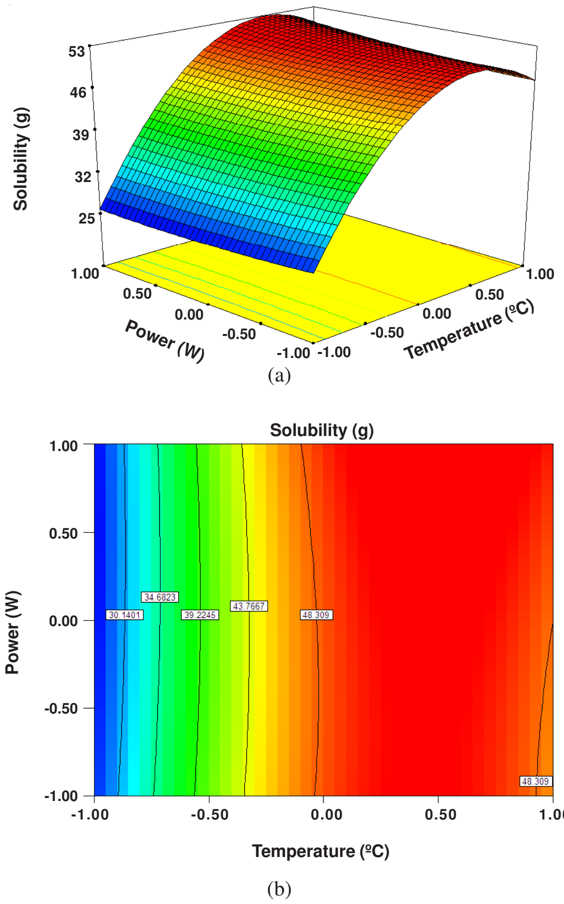


Fig. 6. 3D plot and contour plot of the effect of the independent variables temperature and power on solubility

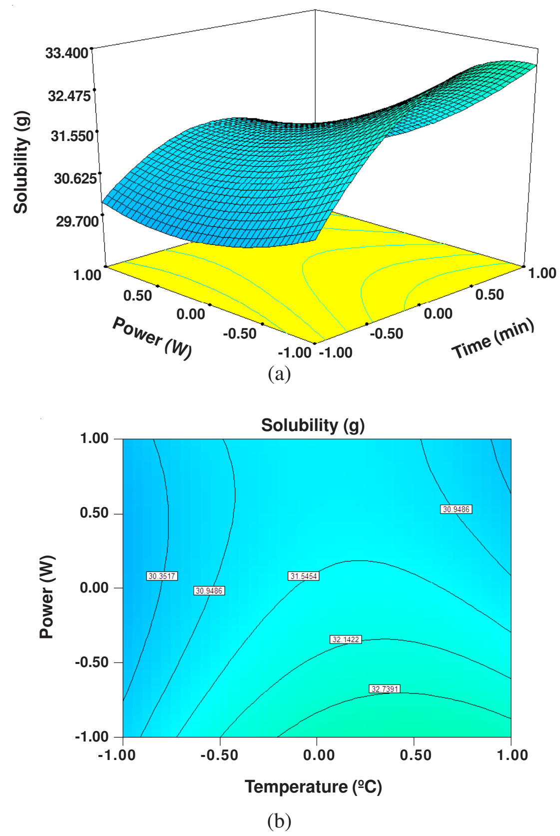


Fig. 7. 3D plot and contour plot of the effect of the independent variables time and power on solubility

Fig. 5a-b shows that the solubility yield was maximal when temperature was 34.3 to 36 ºC during from 18.4 to 20 min. By using eqn. 2, the maximum solubility was obtained at 35.2 ºC and with the time of 19.7 min. This evidences that the solubility increased at a minimum time when raised the temperature. Fig. 6a-b shows the solubility yield was maximal that when temperature was 34.3 to 36 ºC during from 120 to 140 w. The maximum solubility was obtained at 35.2 ºC and 140 w. We can also obtain the maximum solubility.

The response surface methodology allowed us to simulate the effect of temperature, time and power on solubility. The following operating conditions: 19.7 min, at 35.2 ºC with a power of 140 w were selected in order to combine optimized solubility, and solubility is 53.6318 g.

Verification of the models: The suitability of the model equation for predicting the optimum response values was tested by using the selected optimal conditions. The maximum predicted yield and experimental yield of solubility were given in Table-4[22]. Additional experiment by using the predicted optimum conditions for solubility was carried out: temperature of 35.2 ºC, time of 19.7 min, power of 140 w. To ensure the predicted result was not biased toward the practical value, experiment rechecking was performed by using these modified optimal conditions: temperature of 35 ºC, time of 20 min, power of 140 w. The solubility of 53.5926 g was obtained, demonstrated the validation of the RSM model. The results of analysis confirmed that the response model was adequate for reflecting the expected optimization (Table-4) and the model of eqn. 2 was satisfactory and accurate.

TABLE-4
PREDICTED AND EXPERIMENTAL VALUES OF THE RESPONSE AT OPTIMUM AND MODIFIED CONDITIONS

	Temp. (ºC)	Time (min)	Power (w)	Yield of solubility (g)
Optimum conditions	35.2	19.7	140	53.6318
Modified conditions	35	20	140	52.6321

Conclusion

The effect of ultrasonic irradiation on the solubility of sodium sulphate was studied. Ultrasound can increase solubility, but can not change the trend. Ultrasound did improve the solubility and play an important role in crystallization process. Based on the single-factor experimental , RSM was used to estimate and optimize the experimental variables: temperature (ºC), time (min) and power (w). Temperature has highly significant effects on the response values. The optimal conditions for solubility was as follows: temperature of 35.2 ºC, time of 19.7 min and power of 140 w. Under these conditions, the experimental yield of solubility was 52.6321 g, which was close with the predicted yield value.

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