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Optimization of Progressive Freeze Concentration on Apple Juice via Response Surface Methodology

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Abstract. In this work, a progressive freeze concentration (PFC) system was developed to concentrate apple juice and was optimized by response surface methodology (RSM). The effects of various operating conditions such as coolant temperature, circulation flowrate, circulation time and shaking speed to effective partition constant (K) were investigated. Five different level of central composite design (CCD) was employed to search for optimal concentration of concentrated apple juice. A full quadratic model for K was established by using method of least squares. A coefficient of determination (R^2) of this model was found to be 0.7792. The optimum conditions were found to be coolant temperature = -10.59 °C, circulation flowrate = 3030.23 mL/min, circulation time = 67.35 minutes and shaking speed = 30.96 ohm. A validation experiment was performed to evaluate the accuracy of the optimization procedure and the best K value of 0.17 was achieved under the optimized conditions.

1. Introduction

The selection of the technology used in fruit juices concentration process plays an important role in order to maintain flavour, colour and functional properties of concentrated fruit juices. The use of freeze concentration (FC) system is an excellent idea in the manufacturing of high quality fruit juices [1]. During FC, water is separated from a liquid solution by cooling or freezing it until ice crystals are formed [2]. A very pure ice crystal which contains only water is left behind a concentrated liquid product. Several FC methods have been conceived to improve the effectiveness of the concentration process such as suspension freeze concentration (SFC) and progressive freeze concentration (PFC). SFC system consists of crystallization, growth, and separation of ice crystals where it comprises a scraped surface heat exchanger (SSHE) and a recrystallizer [3]. SSHE is the most expensive processing unit in an FC plant where 30% of the total investment costs come from SSHE [4]. Although a high separation efficiency is achieved but the initial and operational costs are relatively high. Thus, the industrial future of FC is associated more with PFC because of the simpler separation step and low operational costs.

PFC is one of the other methods of FC where only a single ice crystal is formed on a cooling wall with a moving ice front [5]. In this study, a PFC system was proposed which is a spiral finned freeze concentrator [6]. This system has spiral fins as the main part, providing highest contact surface area and optimum flow characteristic. The productivity can be increased with the increase of contact surface area between the coolant and the solution through the cooling surface. In the present paper,



PFC has been applied to apple juice where the application of this PFC system on the real solution can be known. Optimum conditions for the apple juice concentration were determined using Response Surface Methodology (RSM).

2. Materials and Methods

2.1. Materials

Fuji apples were purchased from AEON Taman Universiti and washed with water to remove any adhering substances and sliced. Juice was extracted from the apples by homogenizing in a juicer. Then, the concentration of the apple juice was tested using a refractometer. The chosen concentration of apple juice was 0.1 g/mL according to its standard concentration.

2.2. Construction of Progressive Freeze Concentrator

The research activities start with the designing new crystallizer for PFC system, as depicted in Figure 1. After the crystallizer was successfully designed and fabricated, the new crystallizer was then tested in order to check whether it can produce a layer of ice crystal. The details of designing process has been explained in Samsuri et al. [6] where it also proves that the crystallizer is capable to produce the ice crystal from glucose solution. Then, the optimization process was performed to investigate the effect of four operating conditions on the response and optimizing these operating conditions of the apple juice concentration process. STATISTICA software Version 8.0 StatSoft Inc., USA was used to allow the optimization process to be effectively conducted. Experiments were conducted according to the experimental arrangements from the Design of Experiment (DOE) tool by the software.

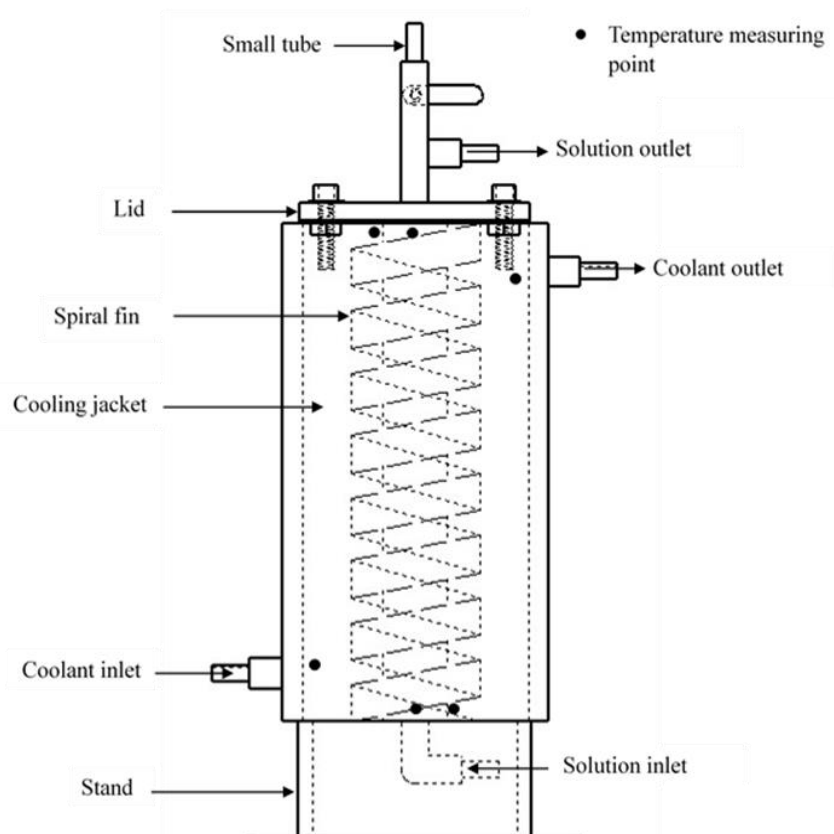


Figure 1 Schematic diagram of spiral finned crystallizer

2.3. Evaluation of system efficiency

The effective partition constant, K , is the most important parameter which portrays the effectiveness of the system. The ice purity increases when K decreases. Thus, the efficiency of the FC is high. The K between ice and liquid phase can be defined by Eq. (1):

$$K = 1 - \frac{\log \frac{C_o}{C_L}}{\log \frac{V_L}{V_o}} \quad (1)$$

where V_L and V_o are volume (mL) of concentrated and initial solution and C_L and C_o are concentration of apple juice (g/mL) in the concentrated and initial solution [7].

2.4. Experimental design

Based on the result of single factor at a time basis experiments, four variables including coolant temperature, circulation flowrate, circulation time and shaking speed were designed. The variables and their coded and uncoded values are presented in Table 1.

Table 1. Experimental range and coded value of variables.

Factors	Range and Levels				
	$-\alpha$	-1	0	+1	$+\alpha$
Circulation Flowrate, X_1 (mL/min)	2600	2800	3000	3200	3400
Circulation Time, X_2 (minutes)	20	40	60	80	100
Coolant Temperature, X_3 ($^{\circ}\text{C}$)	-16	-14	-12	-10	-8
Shaking Speed, X_4 (ohm)	20	30	40	50	60

3. Results and discussion

3.1. Apple juice concentration

The designing process of new apparatus to carry out PFC process with the goal of improving the concentration performance as well as better productivity has resulted in a new crystallizer as described before. During freezing, ice crystal appeared as a layer on the inner surface of the spiral finned crystallizer wall. The thickness of the ice crystal varies for all experiments according to the operating conditions.

3.2. Model fitting

All 26 of the designed experiments and results were depicted in Table 2.

Table 2. Response (K) for each run.

Run	Manipulated Variables				K
	X_1	X_2	X_3	X_4	
1	-14(-1)	40(-1)	2800(-1)	30(-1)	0.67
2	-14(-1)	40(-1)	2800(-1)	50(+1)	0.70
3	-14(-1)	40(-1)	3200(+1)	30(-1)	0.65
4	-14(-1)	40(-1)	3200(+1)	50(+1)	0.71
5	-14(-1)	80(+1)	2800(-1)	30(-1)	0.54
6	-14(-1)	80(+1)	2800(-1)	50(+1)	0.55
7	-14(-1)	80(+1)	3200(+1)	30(-1)	0.66
8	-14(-1)	80(+1)	3200(+1)	50(+1)	0.69
9	-10(+1)	40(-1)	2800(-1)	30(-1)	0.36
10	-10(+1)	40(-1)	2800(-1)	50(+1)	0.36
11	-10(+1)	40(-1)	3200(+1)	30(-1)	0.61
12	-10(+1)	40(-1)	3200(+1)	50(+1)	0.21
13	-10(+1)	80(+1)	2800(-1)	30(-1)	0.36
14	-10(+1)	80(+1)	2800(-1)	50(+1)	0.42
15	-10(+1)	80(+1)	3200(+1)	30(-1)	0.20
16	-10(+1)	80(+1)	3200(+1)	50(+1)	0.33
17	-16($-\alpha$)	60(0)	3000(0)	40(0)	0.73

18	-8(+ α)	60(0)	3000(0)	40(0)	0.32
19	-12(0)	20(- α)	3000(0)	40(0)	0.51
20	-12(0)	100(+ α)	3000(0)	40(0)	0.48
21	-12(0)	60(0)	2600(- α)	40(0)	0.36
22	-12(0)	60(0)	3400(+ α)	40(0)	0.26
23	-12(0)	60(0)	3000(0)	20(- α)	0.23
24	-12(0)	60(0)	3000(0)	60(+ α)	0.11
25 (C)	-12(0)	60(0)	3000(0)	40(0)	0.29
26 (C)	-12 (0)	60(0)	3000(0)	40(0)	0.29

The empirical mathematical model of the predicted K (Y) as a function of coolant temperature (X_1), circulation time (X_2), circulation flowrate (X_3) and shaking speed (X_4) and their interaction using linear and quadratic regression coefficient of main factors and linear-by-linear regression coefficients of interaction is represented in Eq. (2):

$$Y = 10.68565 + 0.63149 X_1 - 0.02411 X_2 - 0.00424 X_3 + 0.00922 X_4 + 0.02002 X_1^2 + 0.00018 X_2^2 - 0.0001 X_4^2 + 0.00009 X_1 X_2 - 0.00006 X_1 X_3 - 0.0011 X_1 X_4 + 0.00017 X_2 X_4 - 0.00001 X_3 X_4 \quad (2)$$

The fit of the model was evaluated by coefficient of determination (R^2) and analysis of variance (ANOVA).

3.3. ANOVA

The reliable way to evaluate the quality of the fitted model is by the application of ANOVA [8, 9]. The ANOVA in Table 3 indicates that the model for K gives good predictions. The F-value for K is 2.77, higher than the tabulated F-value ($F_{0.05, 14, 11} = 2.74$) at 0.05 significant level.

Table 3. ANOVA for quadratic model.

Source	Sum of Squares of Error (SS)	Degree of Freedom (DF)	Mean Squares (SS)	F-value
Regression (SSR)	0.690	14	0.049	2.77
Residual	0.196	11	0.018	
Total (SST)	0.886	25		
R^2	0.779			

After the validity of the regression model has been evaluated, it is also important to identify the variables that can influence the process significantly. The sorted multiple regression results are presented in Table 4, which would be used to evaluate the significance of regression coefficients of the model for response of K, where standard error estimates the common within-group standard deviation.

From Table 4, it can be seen that the mathematical model is statistically significant ($P < 0.001$) and this contributes largely towards the response variable. The quadratic term shows a substantial significant effect at 97% confidence level (p-value < 0.03). In general, the significant order of the independent variables on K from high to low based on the F- and p-values is: coolant temperature, circulation time, circulation flowrate and shaking speed. The R^2 value was found to be 0.7792 which is acceptable because it is larger than 0.75. R^2 should be at least 0.75 for the good fit of the model [10].

Table 4. Significance of regression coefficients of the model for response of K.

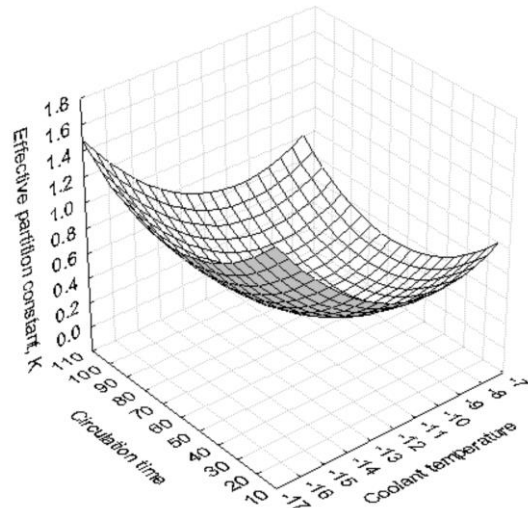
Factor	Coefficient Estimation	Standard Error	F	p
X_1	0.631	0.326	23.109	0.001
X_1^2	0.020	0.008	6.297	0.029
X_2^2	0.000	0.000	5.029	0.047
$X_2 X_4$	0.000	0.000	0.993	0.340

X_2	-0.024	0.029	0.788	0.394
X_3^2	0.000	0.000	0.675	0.429
X_1X_3	-0.000	0.000	0.505	0.492
X_1X_4	-0.001	0.002	0.434	0.524
X_3X_4	-0.000	0.000	0.260	0.620
X_4	0.009	0.061	0.2109	0.655
X_4^2	-0.000	0.000	0.102	0.756
X_2X_3	-0.000	0.000	0.028	0.870
X_3	-0.004	0.005	0.027	0.872
X_1X_2	0.000	0.001	0.011	0.920

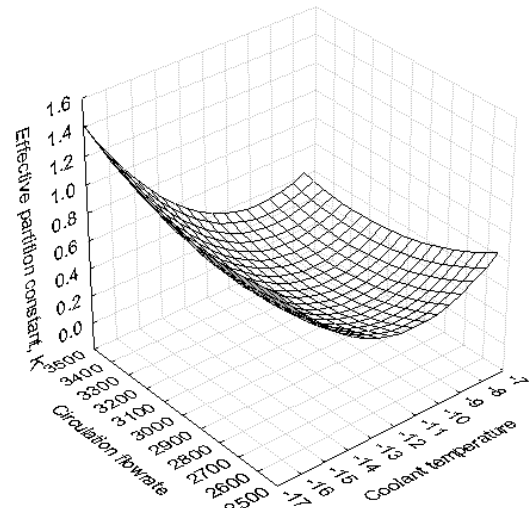
3.4. Effect of independent variables

The three dimensional (3D) response surfaces could also demonstrate the effect of four variables on the K. It could also visualize the significant ($p < 0.05$) interaction effects of factors on the PFC process. For the effects of coolant temperature and circulation time on the K, as depicted in Figure 2(a), the solution is saturated with solutes after 80 minutes, thus causing some inclusions of the solute into the ice formed. Longer circulation time than 80 minutes is not suitable for this PFC process. Anywhere outside of these range, the K-value is high as affected by decreasing coolant temperature and circulation time. Elliptical contour obtained from the interaction between coolant temperature and circulation flowrate in Figure 2(b) shows that there is a perfect interaction between the variables [11]. It could also be seen that K-value is low when the coolant temperature is intermediate while the circulation flowrate is high. The K-value decreases with the increasing circulation flowrate. The solutes were not easily trapped and were rapidly brought away from the surface of the stagnant ice layer, causing higher amount of solutes remaining in the concentrated solution and lower solute entrapment in the ice crystal. However, a decrease in the coolant temperature beyond the optimum region resulted in an increase in the K-value, as can be seen in Figure 2(c). During this time, ice crystal had grown faster which had overtaken the movement of solute particles in the direction of the solution flow. Thus, there is a high solute inclusion in the ice formed.

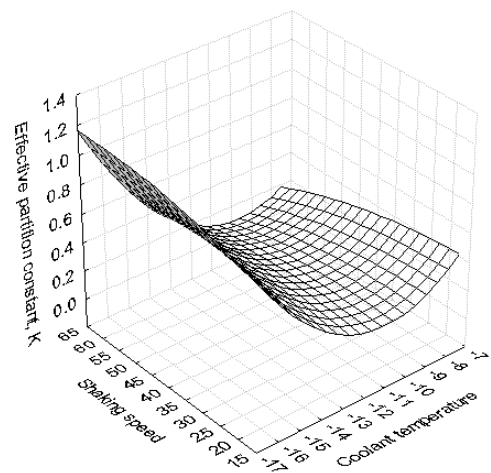
From Figure 2(d), both intermediate values of circulation time and circulation flowrate yielded the best value of K in these interactions. At circulation time of more than 70 minutes, the formed ice crystal has nearly filled the entire volume of the crystallizer. Thus, the diameter of the path for solution flow has been reduced. The solutes also became saturated in the concentrated solution and this led to the contamination of solutes in the ice layer. It can be seen from the interaction between circulation time and shaking speed in Figure 2(e) that intermediate circulation time is giving the best value of K although the shaking speed is changing. The result shows that circulation time has a greater effect on K than the shaking speed, especially at intermediate circulation time. Initially, ice growth rate is fast and decreases over time due to increased heat transfer resistance and the concentration of solute interface, which then lowers the freezing point, making it more difficult to freeze. The decrease of the ice growth rate making less inclusion of solute into the ice crystal. It can be seen in Figure 2(f) that the value of K decreases as the circulation flowrate and shaking speed increase from 3000 to 3400 mL/min and from 60 to 65 ohm, respectively. Besides, the value of K increases with further increase of shaking speed. Increase of shaking will erode not only the interface containing rejected solute, but also the pure ice crystal layer formed on the inner wall, thus making the solution less concentrated where much water returned back to the solution.



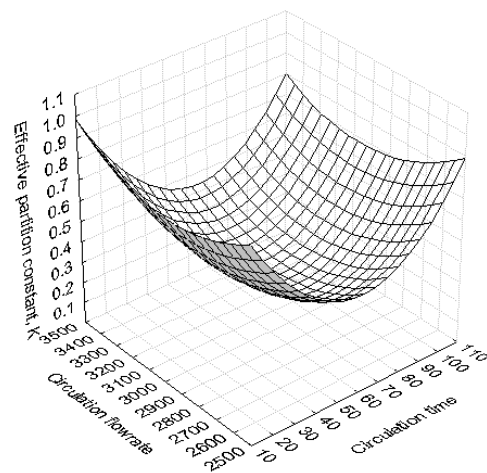
(a)



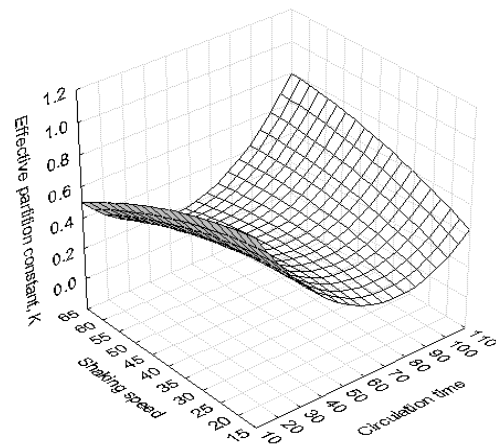
(b)



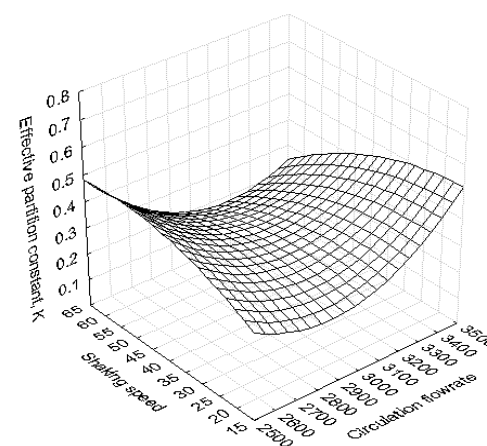
(c)



(d)



(e)



(f)

Figure 2. 3D surface plot of K as a function of (a) coolant temperature and circulation time; (b) coolant temperature and circulation flowrate; (c) coolant temperature and shaking speed; (d) circulation time and circulation flowrate; (e) circulation time and shaking speed; (f) circulation flowrate and shaking speed.

3.5. Optimization of variables and model verification

Additional experiment was carried out to validate the optimization result obtained by the RSM. Hence, Table 5 lists the optimum value for each parameter involved accordingly and the validation value is shown. K-value for validation experiment is better than prediction value and the values of prediction and validation are close enough to each other.

Table 5. Optimum conditions found by the model and verification of the model.

Response	X ₁ (°C)	X ₂ (min)	X ₃ (mL/min)	X ₄ (ohm)	Prediction	Validation
K	-10.59	67.35	3030.23	30.96	0.25	0.17

4. Conclusion

RSM was successfully used to optimize K for the PFC process on the apple juice concentration. All R² and F-value for K signify good accuracy of fitted model generated by the software. The experimental value is in agreement with the predicted value. Under the optimum conditions (-10.59 °C of coolant temperature, 67.35 minutes of circulation time, 3030.23 mL/min of circulation flowrate, 30.96 ohm of shaking speed), the K-value reaches 0.17 which represents the high efficiency of the PFC process. This study may provide useful tools to develop an efficient process for industrial concentration of apple juice.

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