Clarification of sour orange juice by ultrafiltration: Optimization of permeate flux and fouling resistances using response surface methodology

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Abstract

The turbidity of sour orange juice after juice extraction affects on quality, shelf-life and concentration of juice. Therefore, juice clarification is an important operation in the fruit processing industry. The goal of this study was evaluating the effect of membrane operation parameters including pressure (120-220 kPa) and temperature (25-35 °C) on the permeate flux and hydraulic resistance of sour orange juice during membrane clarification. Response surface methodology (RSM) was used to optimizing the operating parameters. Results of the experiments showed that the permeate flux was raised with increasing of temperature, but total hydraulic resistance (R_T), concentration polarization resistance (R_c) and gel layer resistance (R_g) was decreased in mentioned condition. The permeate flux, membrane resistance (R_m), R_T , R_{cp} and fouling index was raised with increasing in pressure. The R_m and fouling index are showed different behavior depending on temperatures level. Results of process optimization indicated that the best conditions to maximize of permeate flux, and to minimize of fouling index and R_T achieved at 35 °C and 120 kPa for a maximum desirability of 0.761.

Keywords: Sour orange, Clarification, Response Surface Methodology, Ultrafiltration.

Introduction

The main applications of membrane operations in the food industry are in the beverage industry (wine, beer, fruit juices, etc.) and the dairy industry. In fruit juice processing, membrane technology is mainly used for several proposes: clarification of the juice by ultrafiltration (UF) and microfiltration deacidification by (MF), electrodialysis, recovery of aroma compounds by pervaporation and membrane distillation, and concentration or preconcentration of the juice by means of nanofiltration, reverse osmosis (RO), membrane distillation, or direct osmosis (Falguera & Ibarz, 2014). The sour oranges (Citrus aurantium L.) constitute a separate species of citrus fruit but are closely related to the sweet orange species. It is grown primarily for its peel, which is used in the manufacture marmalades of (Barrett, Somogyi, & Ramaswamy, 2005). The sour orange is

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popular for its characteristics, such as being a source of vitamin C, organic acids, mineral, soluble and insoluble fibers and also its antioxidant capacity. The sour orange juice is used as food additive, ingredient in salad dressing and also as a popular drink because of its rich flavor and aroma (Koshani, Ziaee, Niakousari, & Golmakani, 2014).

Thermal treatments are used in the preservation of fruit derivatives and in manufacturing operations. The negative effects of these treatments include non-enzymatic browning, nutrient loss and the formation of undesirable by-products such as 5-hydroxy methylfurfural (HMF) (Ibarz, Pagan, & Garza, 1999). Therefore, operation at low temperatures is of interest.

In pulpy juices, the raw juice obtained after pressing is very turbid, viscous, and of a dark color, and contains a lot of colloidal compounds that are stabilized in suspension by polysaccharides such as pectin, starch, cellulose, and gums. Therefore, many juices are clarified prior to their concentration.

Clarification is performed to remove the juice components that cause cloudiness, mainly pectin (Falguera & Ibarz, 2014). In the traditional clarification process, crude filtra-

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tion was performed directly after crushing the fruit. Pectinase was added to hydrolyze pectin, which reduced the viscosity of the juice before it was passed through a series of decantation and diatomaceous filtration steps to yield clear juice with a typical yield of about 90%. By replacing these final filtration steps with ultrafiltration, a very good-quality, almoststerile product can be produced with a yield of almost 97% (Jansen, Feron, Hanemaaijer, & Huisjes, 2002; Prasad, Runkle, & Shuey, 1994).

UF is very promising alternatives to conventional clarification processes. The operational costs of using membrane processes are considerably lower than those of more traditional processes (Yazdanshenas, Tabatabaee -Nezhad, Soltanieh, Roostaazad, & Khoshfetrat, 2010). Moreover, UF membranes is able to retain microorganisms, avoiding the need for thermal pasteurization processes, and UF is able to remove polyphenol oxidases, which is effective in stabilizing the colors of fruit juices (Falguera & Ibarz, 2014).

Ultrafiltration had been investigated for the clarification of Apple (Onsekizoglu, Bahceci, & Acar, 2010), pear (Alis Cassano, Conidi, Timpone, D'avella, & Drioli, 2007), orange (Galaverna *et al.*, 2008), lemon (Chornomaz, Ochoa, Pagliero, & Marchese, 2011), kiwifruit (Tasselli, Cassano, & Drioli, 2007), chicory (Zhu *et al.*, 2013), Black Currant (Pap et al., 2012) and pineapple juice (Laorko, Li, Tongchitpakdee, Chantachum, & Youravong, 2010).

One of the main drawbacks of membrane technology is the fouling of the membrane exhibited with total resistance, which is caused by the accumulation of solute molecules on the membrane surface or inside the pores. Membrane fouling causes a reduction of the permeate flux and also causes changes in selectivity and decreases the overall process productivity. The permeate flux can be restored by means of cleaning procedures, but the process must be stopped and large amounts of chemicals, energy, water, and time are consumed. Moreover, successive membrane cleaning operations can reduce the life of the membrane (Falguera & Ibarz, 2014).

RSM is a collection of statistical and mathematical techniques useful for developing, improving, and optimizing processes in which a response of interest is influenced by several variables and the objective is to optimize this response. RSM has important applications in the design, development and formulation of new products, as well as in the improvement of existing product design. It defines the effect of the independent variables. alone or in combination, on the process. In addition, in order to analyze the effects of the independent variables, this experimental methodology generates а mathematical model which describes the chemical or biochemical (Bas & Boyacı, processes 2007; Ruby Figueroa, Cassano, & Drioli, 2011).

Ruby Figueroa et al. (2011) evaluated the effect of process parameters including transmembrane pressure (TMP), temperature, and feed flow-rate on fouling resistances during ultrafiltration of orange press liquor and optimized Operation parameters. A strong interaction effect of temperature and feed flow-rate was observed on the permeate flux while interactions TMP-temperature and TMPfeed flow-rate were found to be less significant. In the case of fouling index, interactions TMP-temperature and TMP-feed flow-rate produced a significant effect (Ruby Figueroa et al., 2011). Baklouti et al. (2013) evaluated the resistance-in-series model to analyze flux behavior, which involved the resistances of membrane itself, the fouling and polarization. solute concentration Thev concluded that the resistance due to solute concentration polarization (R_{cp}) dominated the flux decline (40–74%).

The fouling resistance (R_f) varied from 12 to 46%. The selected UF conditions of the compromise were as follows: three bars, 0.95 L min⁻¹ and 30°C. Optimal values of R_f , R_{cp} and permeate limit flux were equal to 18%, 72% and 19 L h⁻¹ m⁻², respectively (Baklouti, Kamoun, Ellouze-Ghorbel, & Chaabouni, 2013). Nourbakhsh *et al.* (2014) evaluated the resistances (total, reversible, irreversible & cake) during microfiltration of watermelon juice and red plum juice. Results showed that the total resistance decreased by about 45% when the feed velocity was increased during clarification of red plum juice due to change in cake resistance. MCE membrane had a lower cake resistance compared to PVDF membrane.

Examination of the microfiltration of watermelon juice showed that Rt decreased by about 54% when the feed temperature was increased from 20 to 50°C, partially due to the reduction of reversible fouling resistance by 78% (Nourbakhsh, Alemi, Emam-Djomeh, & Mirsaeedghazi, 2014).

The aim of this work was to evaluate the potentiality of Polyvinylidene fluoride UF membrane in the clarification of sour orange juice. As well as the effects of Operation parameters including pressure (120-220 kPa) and temperature (25-35 °C) on the permeate flux and resistances investigated. Resistance in series model was applied to identify flux behavior in the UF process.

Materials and methods Chemicals Material

Nitric acid (HNO₃) and sodium hydroxide (NaOH) were purchased from Merck Company. Sodium sulfite (Na₂So₃) was obtained from Sigma–Aldrich Company.

Preparation of sour orange Juices

Sour Orange fruit were purchased from a local market in Gorgan (Iran) and washed with tap water in order to remove foreign material from the skin and drained. Then, the juice was extracted by FMC juice extractors with a 2-mm-diameter perforated plate and placed in a tank. Extracted juices were 130 L from 600 kg sour orange fruit. 4 gr kg⁻¹ Na₂So₃ was added to single strength juice to avoid browning reactions. The juice was stored at -18° C and was defrosted to room temperature before use. Preparation of fruits juices is shown in Figure. 1.

Ultrafiltration unit and procedures

UF experiments were performed by using a pilot plant unit equipped with a tubular

Polyvinylidene fluoride membrane module¹ with a nominal $MWCO^2$ of 200 kDa. The characteristics of UF membrane are summarized in Table 1.



Fig. 1. Preparation of sour orange juice.

Table 1. Characteristics of UF membrane and module used in this study

moune used in this study				
Membrane type	F01740			
Material	PVDF			
Effective area	0.1 m^2			
Length	120 cm			
Range of pH tolerance	1.5-10.5			
Maximum temperature	60 °C			
Maximum pressure	1 MPa			
Module	tubular			

^{1.} ITT PCI Membranes Technology Co., Ltd (Hampshire, United Kingdom)

^{2.} Molecular Weight Cut-Off

The juice was clarified according to the batch concentration procedure in which the permeate is collected separately and the retentate is recycled to the feed tank. The operating pressure was in the range of 120–220 kPa and the temperature varied from 25°C to 35 °C. The permeate flux was measured every one min for 45 min. In Figure. 2 a schematic diagram of the UF experimental setup is illustrated.



The membrane module was rinsed with distilled water for 20 min after the treatment of the juice; then it was submitted to a cleaning process with a NaOH solution (pH = 10, T= 50° C, pressure = 120 kPa, operating time = 0.5 h) followed by a cleaning with the HNO₃ (pH = 2, T= 50° C, pressure = 120 kPa, operating time = 0.5 h) and Backwashing with distilled water for 1 hour. A final rinse of the system with distilled water for at least 20 min was carried out. After each cleaning procedure the distilled water flux of the membrane module in fixed conditions (T= 35° C; pressure = 120 kPa) was measured.

Calculated parameters

The operating pressure calculated according to the following relationship:

DrivingForce =
$$TMP = \frac{P_i + P_o}{2} - P_P$$
 (1)

Where, TMP is the transmembrane pressure (Pa), P_i and P_o are inlet and outlet pressures, respectively, and P_p is permeate pressure. Since P_p is negligible, is not considered.

The permeate flux can be obtained from Darcy's law with assumption of resistance in series model as follows:

$$J_{p} = \frac{TMP}{\mu_{p}R_{T}} = \frac{TMP}{\mu_{p}(R_{m} + R_{g} + R_{cp})}$$
(2)

Where J_p is the permeation flux (kg/m².h), R_T is the total resistance (m⁻¹), μ_p is the dynamic viscosity of permeate (N.S/m²), R_m is the Intrinsic or hydraulic membrane resistance, R_{cp} is the Concentration polarization resistance (m⁻¹) and R_g represents the Gel layer resistance due to (i) the internal fouling of the membrane by adsorption of macromolecules on the internal walls of membrane pores and (ii) a thin layer that blocks the membrane pores on the surface which is created by adhering the particles to the membrane surface (Kazemi, Soltanieh, & Yazdanshenas, 2013).

 R_m was calculated by measuring the distilled water flux of clean membrane as follows:

$$R_m = \frac{TMP}{\mu_w J_w} (3)$$

Where μ_w and J_w represents the viscosity and flux of distilled water, respectively. For this purpose, water flux in the temperature and pressure range used in this study was obtained and then R_m was calculated using equation (3). At the end of each stage filtration, water flux was measured to calculate R_g on the surface of membrane according to Equation (4).

$$R_g = \frac{TMP}{\mu_w J_{wf}} - R_m \quad (4)$$

Where, J_{wf} represents the distilled water flux at the end of filtration process (blocked membrane). R_{cp} Calculated as follows:

$$R_{cn} = R_T - (R_o + R_m)$$
 (5)

The fouling index was calculated according to Equation (6):

FoulingIndex (%) = $\left(\frac{J_w - J_{wf}}{J_w}\right) \times 100$ (6)

The hydraulic permeability of the membrane was determined as follows:

 $L_{i}^{p}=J_{w}/\Delta P(7)$

Where, L_1^p , L_2^p and L_3^p are hydraulic permeability; after cleaning with distilled water, after cleaning with NaOH solution and after cleaning with HNO₃ solution, respectively.

The Viscosity was measured using a rotational type Brookfield LVDV-II digital viscometer (USA) at 25 °C at 80 RPM. A sample of sour orange juice was loaded into cylindrical sample chamber (ULA-31Y) of 16 mL capacity for all experiments and was allowed to equilibrate at 25 °C using a circulating water jacket (Model ULA-40Y Brookfield Engineering Laboratories). (Bodbodak, Kashaninejad, Hesari, & Razavi, 2013).

Experimental design and statistical analysis

The software Design-Expert (trial version 9.0.0.7, Stat-Ease Inc., Minneapolis, USA) was used for experimental design, analysis of data and plotting of graph. Response surface methodology was used to establish the relationships between operating parameters including pressure (120-220 kPa) and temperature (25-35 °C) and ultrafiltration (UF) efficiency and thus to determine optimal conditions. Thirteen treatments Consisted of 5 replications at the central point were conducted base on the rotatable central composite design (table 2).

Response functions of measurement parameters were examined by fitting experimental data on linear (Y_1) , 2FI (Y_2) and Quadratic models (Y_3) as follows:

$$Y_{1} = b_{0} + b_{1}x_{1} + b_{2}x_{2} (8)$$

$$Y_{2} = b_{0} + b_{1}x_{1} + b_{2}x_{2} + b_{12}x_{1}x_{2} (9)$$

$$Y_{3} = b_{0} + b_{1}x_{1} + b_{2}x_{2} + b_{11}x_{1}^{2} + b_{22}x_{2}^{2} + b_{12}x_{1}x_{2} (10)$$

Where, b_0 ; constant term, b_1 and b_2 ; linear effects, b_{11} & b_{22} ; quadratic effects and b_{12} ; interaction effects. The models were compared

based on the coefficient of determination (R^2) , adjusted coefficient of determination (R^2-adj) and predicted coefficient of determination $(R^2$ pred). The model with the highest values of R^2 -adj & R^2 -pred, was selected as the accurate model (Yolmeh, Habibi Najafi, & Farhoosh, 2014). Analysis of variance (ANOVA) was performed to assess the significant effects of process variables on each response.

 Table 2. Experimental design results of central composite design (CCD)

Run	Temperature (°C)	Pressure (kPa)	Average permeate flux (kg/m ² ,h)				
1	25	120	15.97				
2	35	120	20.47				
3	25	220	22.62				
4	35	220	28.40				
5	22.93	170	10 70				
6	37.07	170	27.00				
7	30	99 29	19.20				
8	30	240 71	62.20				
9	30	170	22.70				
10	30	170	24 00				
11	30	170	23.60				
12	30	170	23.20				
13	30	170	24.10				

Result and discussion Flux behavior

Permeate flux behavior during ultrafiltration of sour orange juice was shown in Figure. 3 The curve represented the evolution of permeate flux decline with time due to concentration polarization and gel formation. Permeate flux curve could be divided in three regions. An initial region in which a rapid decrease of permeate flux occurs; a second region, corresponding to a smaller decrease of permeate flux; a third region characterized by a small decrease of permeate flux up to a steady-state (A Cassano, Marchio, & Drioli, 2007).

hydraulic permeability

Hydraulic permeability is used for calculating of membrane fouling that is estimated with measuring the distillated water flux before and after the membrane filtration and after cleaning treatment. Figure. 4 represents the distillated water permeate flux of the membrane before and after cleaning treatments. According to figure. 4, after the cleaning with water & NaOH, the hydraulic permeability of the membrane is 63 & 43% lower than the initial value (0.06 kg/m².h.kPa), respectively. Hydraulic permeability of the primary causes of acid wash with distilled water recovery percentage was 9/97. The cleaning with a HNO₃ solution permitted the recovery of about 97.9% of the initial distillated water permeability of the membrane.

➡ 120 KPa 25 C → 120 KPa 35 C



Fig .3. Time course of permeate flux during ultrafiltration of sour orange juice.





Fig .4. Hydraulic permeability of the UF membrane before and after cleaning procedures (T = 35°C; pressure =120 kPa).

Permeate flux

The results of ANOVA for permeate flux, showed that the effects of various operating parameters (temperature and pressure) on flux was significant. Highest and lowest fluxes were related to treatment of 30 °C; 240.71 kPa and 25 °C; 120 kPa, respectively. The maximum values of R^2 , R^2 -adj and R^2 -pred obtained for linear model that revealed the adequate of this model for Prediction. The values of R^2 , R^2 -adj and R^2 -pred for the permeate flux was 90.42, 88.51 and 82.24, respectively. The model as follows:

Flux=-1.86056+(0.47974×temperature)+(6.11987×pressure) (11)

From equation (11), we found that the linear effect of pressure is the most effective factor in increasing of permeate flux. Ruby Figueroa et al (2011) during ultrafiltration of orange press liqueur reported that the linear coefficients of pressure were found to be the most significant effect to increase the permeate followed by linear coefficient flux. of temperature (Ruby Figueroa et al., 2011). Figure.5 represents effect of temperature and pressure on permeate flux. It seems that at such higher pressures, increasing the driving force led to increase in the flux, whereas, increasing the molecular diffusion and decreasing the viscosity led to flux improvement at such higher temperatures (Salehi & Razavi, 2012). The permeate flux was found to be pressure-depended in the pressure range studied.

An increase of temperature, enhanced permeate flux due to an increase of masstransfer coefficient according to the film model (Vladisavljević, Vukosavljević, & Bukvić, 2003).

Fouling index (FI)

The results of ANOVA for FI, showed that the effects of various operating parameters on FI was significant. Highest and lowest FI were related to treatment of 30 °C; 240.71 kPa and 37 °C; 170 kPa, respectively. The maximum values of R^2 , R^2 -adj and R^2 -pred obtained for quadratic model that revealed the adequate of this model for Prediction. The values of R^2 , R^2 -adj and R^2 -pred for the FI was 91.46, 85.35 and 67.74, respectively. From equation (12), it was found that the linear effect of temperature is the most effective factor in increasing of FI. The effect of temperature and pressure on the FI is shown in Fig. 6. The fouling index decreased with increasing temperature only at pressures higher than central point; at lower level the fouling index raised by increasing the temperature. The FI increased greatly with increasing pressure. In membrane processing which pressure is the driving force; also permeate flux can be raised with increasing in pressure; but, pressure is intensifies the membrane fouling index (Pabby, Rizvi, & Requena, 2009)

Resistance

Flux decline occurs during membrane filtration, because of several reasons including the gel layer formation and blocking the pores and the concentration polarization layer formation. This agents led to increase in the resistance of membrane against material passing. R^2 -adj and R^2 -pred obtained for linear model that revealed the adequate of this model for Prediction. The values of R^2 , R^2 -adj and R^2 pred for the R_T was 92.94, 91.53 and 86.95, respectively. From equation (13), we found that the linear effect of pressure is the most effective factor in increasing of R_T . The R_T increased with increasing in pressure. In addition, the flux is raised with increasing in pressure. This suggests that pressure have more effect on flux compared to the R_{T} . Figure. 7 represent effect of temperature and pressure on R_T. At high pressure, the rate of deposition would be high and the high pressure would compress the rejected solutes into a thicker and denser fouling layer with an R_T (De Bruijn, Venegas, Martinez, & Bórquez, 2003). This result was also observed by Nourbakhsh et al. (2014) working with red plum and watermelon juices (Nourbakhsh et al., 2014). Increasing in temperature, reduced viscosity and enhanced the diffusion coefficient of material from cake layer to feed, thus the RT decreased



Fig .5. 3D response surface plot for the effect of pressure and temperature on permeate flux.

Total hydraulic resistance (R_T)

The results of ANOVA of total resistance showed that the effect of various treatment of operating parameters on R_T was significant. Highest and lowest R_T were related to treatment of 25 °C; 220 kPa and 30 °C; 99.29 kPa, respectively. The maximum values of R^2 ,



Fig .7. 3D response surface plot for the effect of pressure and temperature on $R_{\rm T}.$

Membrane resistance (Rm)

The results of ANOVA of membrane resistance showed that the effect of various treatment of operating parameters on R_m was significant. The maximum values of R^2 , R^2 -adj and R^2 -pred obtained for quadratic model that revealed the adequate of this model for Prediction. The values of R^2 , R^2 -adj and R^2 -

pred for the R_m were found to be 89.92, 82.72 and 59.32, respectively. From equation (14), it was found that the linear effect of pressure on enhanced of R_m is more than the linear effect of temperature on reduced of this factor. The R_m changes during filtration by membrane fouling, which is due to either solute adsorption onto the membrane surface and membrane plugging (de Oliveira, Docê, & de Barros, 2012). The effect of temperature and pressure on the R_m is shown in Fig. 8. With increasing temperature and pressure, R_m was decreased and increased, respectively



Fig .8. 3D response surface plot for the effect of pressure and temperature on R_m .

Gel layer resistance (R_g)

The results of ANOVA of R_g showed that the effect of various treatment of operating parameters on R_g was significant. In this study, the largest proportion of the total resistance (47.8%) were related to Rg. Baklouti et al. (2012) applied the Hermia model to describe the fouling during the filtration of enzymatic treated of pomegranate juice and reported that the gel layer formation was the major cause of fouling (Baklouti, Ellouze-Ghorbel, & Mokni, 2012). Domingues et al. (2014) reported that gel formation was the major fouling factor during the microfiltration of centrifuged and enzyme treated passion fruit juice with polieterimide hollow fibre Membrane (Domingues, Ramos, Cardoso, & Reis, 2014).

The maximum values of R^2 , R^2 -adj and R^2 pred obtained for linear model that revealed the adequate of this model for Prediction. The values of R^2 , R^2 -adj and R^2 -pred for the R_g was 95.35, 94.41 and 90.4, respectively. From equation (15), we found that the linear effect of pressure on enhanced of R_g is more than the linear effect of temperature on reduced of this factor. Fig. 9 represents effect of temperature and pressure on R_g . increasing in pressure due to enhance of driving force and adsorption of particles on the membrane surface leads to increasing of R_g (Rai, Majumdar, Das Gupta, & De, 2007). According to the fig. 9, at higher temperatures, Rg slowly declined



Fig .9. 3D response surface plot for the effect of pressure and temperature on $R_{\rm g}.$

Concentration polarization resistance (R_{cp})

The results of ANOVA of R_{cp} showed that the effect of various treatment of operating parameters on R_{cp} was significant. In this study, the lowest proportion of the total resistance (9.39%) were related to R_{cp} . The maximum values of R^2 , R^2 -adj and R^2 -pred obtained for linear model that revealed the adequate of this model for Prediction. The values of R^2 , R^2 -adj and R^2 -pred for the R_{cp} was 79.25, 75.09 and 61.2, respectively. From equation (16), it was found that the linear effect of pressure on enhanced of R_{cp} is more than the linear effect of temperature on reduced of this factor. The effect of temperature and pressure on the R_{cp} is shown in Fig. 10. Concentration polarization occurs

in the all membrane processes which the driving force is the pressure. The accumulation of particles on the membrane surface may increase with increase of pressure. It enhances the membrane fouling via concentration polarization.

Fouling Index =
$$-11.89560 + (0.99212 \times temperature) - (0.83645 \times pressure) -$$

(0.00669349 × temperature × pressure) - (0.016689 × temperature²) + (0.69063 × pressure²)
 $R_T = 1.55245 \times 10^{10} - (2.24801 \times 10^8 \times temperature) + (6.56529 \times 10^9 \times pressure)$ (13)

$$R_{M} = 3.07333 \times 10^{10} - (1.18969 \times 10^{9} \times temperature) - (4.72727 \times 10^{9} \times pressure) - (2.71050 \times 10^{7} \times temperature \times pressure) + (1.18759 \times 10^{7} \times temperature^{2}) + (1.88512 \times 10^{9} \times pressure^{2})$$
(14)

$$R_{g} = 3.39272 \times 10^{9} - (9.12388 \times 10^{7} \times temperature) + (5.21875 \times 10^{9} \times pressure)$$
(15)
$$R_{cn} = 1.65555 \times 10^{9} - (1.98456 \times 10^{7} \times temperature) + (4.77544 \times 10^{8} \times pressure)$$
(16)

An increase in temperature enhanced back diffusion of solutes into the bulk solution, reducing consequently the thickness of the polarized layer: therefore R_{cp} decreased with increasing in temperature (A Cassano. Mecchia, & Drioli, 2008). He et al. (2007) during clarification of apple juice over 20 hours, reported that membrane fouling to the filtration performance can be neglected; and the major factors influencing permeate flux were the reversible concentration polarization (in contrast to our study) related to feed and concentration viscosity. not the irreversible fouling such as internal plugging, silting, etc (He, Ji, & Li, 2007)

Optimizing

In order to maximize the permeate flux and minimize the fouling index and total resistance, the desirability function approach was applied to analyze the regression model equations. The optimized operating variables were found to be 120 kPa, 35°C for an overall desirability of 0.761. In optimal condition, the permeate flux, fouling index and total resistance was 22.27 (kg/m².h), 2.09 % and 1.55×10^{10} (m⁻¹), respectively.

Optimization results for the UF of sour orange juice are summarized in table 3



Fig .10. 3D response surface plot for the effect of pressure and temperature on R_{cp} .

Table 5. Fredicted parameters and responses from KSWI for optimized condition						
			goal	The degree of importance		
dicted meters/ ponses	TMP	120 kPa	In range	-		
	Temperature	35 °C	In range	-		
	permeate flux	22.27	maximizing	+++++		
rra rra esl	fouling index	2.09 %	minimizing	++++		
H pa	total resistance	1.55×10^{10}	minimizing	++++		
	overall desirabilit	ty	0.761			

Table 3. Predicted parameters and responses from RSM for optimized condition

Conclusions

Sour orange is a source of vitamin C that is cultivated in the north and central regions of Iran. The effect of operating parameters including of pressure and temperature on the performance of a UF membrane in the clarification of sour orange juice was studied with the response surface methodology. A central composite design was used for regression modeling and optimizing the UF operating parameters. Results of the experiments showed that the permeate flux, was raised with increasing in temperature, but fouling index and all of resistances (R_T , R_M , R_{cp} & R_g), were decreased in this condition. Increasing in pressure, enhanced of permeate flux and all of the resistances. The permeate flux also decreased over the time. The best conditions to maximize of permeate flux, and to minimize of fouling index and R_T achieved at 35 °C and 120 kPa.

References

- Baklouti, S., Ellouze-Ghorbel, R., & Mokni, A. (2012). Clarification of pomegranate juice by ultrafiltration: study of juice quality and of the fouling mechanism. *Fruits*, 67(03), 215-225.
- Baklouti, S., Kamoun, A., Ellouze Ghorbel, R., & Chaabouni, S. (2013). Optimising operating conditions in ultrafiltration fouling of pomegranate juice by response surface methodology. *International Journal of Food Science & Technology*, 48(7), 1519-1525.
- Barrett, D. M., Somogyi, L. P., & Ramaswamy, H. S. (2005). Processing fruits: science and technology: *CRC Press Boca Raton*, FL.
- Baş, D., & Boyacı, İ. H. (2007). Modeling and optimization I: Usability of response surface methodology. *Journal of food engineering*, 78(3), 836-845.
- Bodbodak, S., Kashaninejad, M., Hesari, J., & Razavi, S. (2013). Modeling of Rheological Characteristics of "MalasYazdi"(Punicia granatum L.) Pomegranate Juice. *Journal of Agricultural Science and Technology*, 15(5), 961-971.
- Cassano, A., Conidi, C., Timpone, R., D'avella, M., & Drioli, E. (2007). A membrane-based process for the clarification and the concentration of the cactus pear juice. *Journal of food engineering*, 80(3), 914-921.
- Cassano, A., Marchio, M., & Drioli, E. (2007). Clarification of blood orange juice by ultrafiltration: analyses of operating parameters, membrane fouling and juice quality. *Desalination*, 212(1), 15-27.
- Cassano, A., Mecchia, A., & Drioli, E. (2008). Analyses of hydrodynamic resistances and operating parameters in the ultrafiltration of grape must. *Journal of food engineering*, 89(2), 171-177.
- Chornomaz, P. M., Ochoa, N. A., Pagliero, C., & Marchese, J. (2011). Synthesis, characterization and performance of membranes for clarification of lemon juice. Desalination and Water Treatment, 27(1-3), 294-298.
- De Bruijn, J., Venegas, A., Martinez, J., & Bórquez, R. (2003). Ultrafiltration performance of Carbosep membranes for the clarification of apple juice. *LWT-Food Science and Technology*, 36(4), 397-406.
- de Oliveira, R. C., Docê, R. C., & de Barros, S. T. D. (2012). Clarification of passion fruit juice by microfiltration: Analyses of operating parameters, study of membrane fouling and juice quality. *Journal of food engineering*, 111(2), 432-439.
- Domingues, R. C. C., Ramos, A. A., Cardoso, V. L., & Reis, M. H. M. (2014). Microfiltration of passion fruit juice using hollow fibre membranes and evaluation of fouling mechanisms. *Journal of food engineering*, 121, 73-79.
- Falguera, V., & Ibarz, A. (2014). Juice Processing: Quality, Safety and Value-Added Opportunities: CRC

Press.

- Galaverna, G., Di Silvestro, G., Cassano, A., Sforza, S., Dossena, A., Drioli, E., & Marchelli, R. (2008). A new integrated membrane process for the production of concentrated blood orange juice: Effect on bioactive compounds and antioxidant activity. *Food chemistry*, 106(3), 1021-1030.
- He, Y., Ji, Z., & Li, S. (2007). Effective clarification of apple juice using membrane filtration without enzyme and pasteurization pretreatment. *Separation and Purification Technology*, 57(2), 366-373.
- Ibarz, A., Pagan, J., & Garza, S. (1999). Kinetic models for colour changes in pear puree during heating at relatively high temperatures. *Journal of food engineering*, 39(4), 415-422.
- Jansen, A. E., Feron, P. H. M., Hanemaaijer, J. H., & Huisjes, P. (2002). Apparatus and method for performing membrane gas/liquid absorption at elevated pressure: *Google Patents*.
- Kazemi, M. A., Soltanieh, M., & Yazdanshenas, M. (2013). Mathematical modeling of crossflow microfiltration of diluted malt extract suspension by tubular ceramic membranes. *Journal of food engineering*, 116(4), 926-933.
- Koshani, R., Ziaee, E., Niakousari, M., & Golmakani, M. T. (2014). Optimization of Thermal and Thermosonication Treatments on Pectin Methyl Esterase Inactivation of Sour Orange Juice (Citrus aurantium). *Journal of Food Processing and Preservation*.
- Laorko, A., Li, Z., Tongchitpakdee, S., Chantachum, S., & Youravong, W. (2010). Effect of membrane property and operating conditions on phytochemical properties and permeate flux during clarification of pineapple juice. *Journal of food engineering*, 100(3), 514-521.
- Nourbakhsh, H., Alemi, A., Emam-Djomeh, Z., & Mirsaeedghazi, H. (2014). Effect of processing parameters on fouling resistances during microfiltration of red plum and watermelon juices: a comparative study. *Journal of food science and technology*, 51(1), 168-172.
- Onsekizoglu, P., Bahceci, K. S., & Acar, M. J. (2010). Clarification and the concentration of apple juice using membrane processes: A comparative quality assessment. *Journal of Membrane Science*, 352(1), 160-165.
- Pabby, A. K., Rizvi, S. S., & Requena, A. M. S. (2009). Handbook of membrane separations: chemical, pharmaceutical, food, and biotechnological applications: *CRC press*.
- Pap, N., Mahosenaho, M., Pongrácz, E., Mikkonen, H., Jaakkola, M., Virtanen, V., Vatai, G. (2012). Effect of ultrafiltration on anthocyanin and flavonol content of black currant juice (Ribes nigrum L.). *Food and Bioprocess Technology*, 5(3), 921-928.
- Prasad, R., Runkle, C. J., & Shuey, H. F. (1994). Spiral-wound hollow fiber membrane fabric cartridges and modules having flow-directing baffles: *Google Patents*.
- Rai, P., Majumdar, G., Das Gupta, S., & De, S. (2007). Effect of various pretreatment methods on permeate flux and quality during ultrafiltration of mosambi juice. *Journal of food engineering*, 78(2), 561-568.
- Ruby Figueroa, R. A., Cassano, A., & Drioli, E. (2011). Ultrafiltration of orange press liquor: Optimization for permeate flux and fouling index by response surface methodology. *Separation and Purification Technology*, 80(1), 1-10.
- Salehi, F., & Razavi, S. M. (2012). Dynamic modeling of flux and total hydraulic resistance in nanofiltration treatment of regeneration waste brine using artificial neural networks. Desalination and Water Treatment, 41(1-3), 95-104.
- Tasselli, F., Cassano, A., & Drioli, E. (2007). Ultrafiltration of kiwifruit juice using modified poly (ether ether ketone) hollow fibre membranes. *Separation and Purification Technology*, 57(1), 94-102.
- Vladisavljević, G., Vukosavljević, P., & Bukvić, B. (2003). Permeate flux and fouling resistance in ultrafiltration of depectinized apple juice using ceramic membranes. *Journal of food engineering*, 60(3), 241-247.
- Yazdanshenas, M., Tabatabaee-Nezhad, S. A. R., Soltanieh, M., Roostaazad, R., & Khoshfetrat, A. B. (2010). Contribution of fouling and gel polarization during ultrafiltration of raw apple juice at industrial scale. *Desalination*, 258(1), 194-200.
- Yolmeh, M., Habibi Najafi, M. B., & Farhoosh, R. (2014). Optimisation of ultrasound-assisted extraction of natural pigment from annatto seeds by response surface methodology (RSM). *Food chemistry*, 155, 319-324.
- Zhu, Z., Luo, J., Ding, L., Bals, O., Jaffrin, M. Y., & Vorobiev, E. (2013). Chicory juice clarification by membrane filtration using rotating disk module. *Journal of food engineering*, 115(2), 264-271.

شفافسازی آب نارنج با استفاده از اولترافیلتراسیون: بهینهسازی شار تراوه و مقاومت گرفتگی با

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چکیدہ

کدورت آب نارنج بعد از آبگیری بر روی کیفیت، ماندگاری و تغلیظ آبمیوه موثر است. بنابراین شفافسازی آبمیوه یک فرآیند مهم در صنعت فرآوری میوه است. هدف از این پژوهش بررسی پارامترهای عملیاتی غشاء شامل فشار (۲۲۰–۲۲۰ کیلوپاسکال) و دما (۳۵–۲۵ درجه سانتیگراد) بر شار تراوه و مقاومت هیدرولیک در حین شفافسازی غشایی آب نارنج بود. روش سطح پاسخ برای بهینهسازی پارامترهای عملیاتی استفاده شد. نتایج آزمایشات نشان داد با افزایش دما شار تراوه افزایش یافت اما مقاومت هیدرولیک کل (R_T)، مقاومت پلاریزاسیون تغلیظ (R_{cp}) و مقاومت لایه ژل (R_g) در ایـن شـرایط کاهش یافت. شار تراوه افزایش یافت اما مقاومت هیدرولیک کل (R_T)، مقاومت پلاریزاسیون تغلیظ (R_{cp}) و مقاومت لایه ژل (R_g) در ایـن شـرایط کاهش یافت. شار تراوه، مقاومت غشاء (R_m) مقاومت هیدرولیک کل (R_T)، مقاومت پلاریزاسیون تغلیظ (R_{cp}) و مقاومت لایه ژل (R_g) در ایـن شـرایط کاهش یافت. شار تراوه، مقاومت غشاء (R_m) مقاومت هیدرولیک کل (R_T)، مقاومت پلاریزاسیون تغلیظ (R_{cp}) و مقاومت لایـه ژل (R_g) در ایـن شـرایط توجه به تغییرات دما نشان دادند. نتایج بهینه سازی فرآیند نشان داد بهترین شرایط برای به حداکثر رساندن شـار تراوه و بـه حـداقل رساندن شـاخص گرفتگی و T_T در دمای ۳۵ درجه سانتی گراد و ۱۲۰ کیلوپاسکال با حداکثر مطلوبیت ۱۷۶۰ بدست میآید.

واژههای کلیدی: نارنج، شفافسازی، اولترافیلتراسیون، روش سطح پاسخ

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