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Effects of operating parameters on physicochemical properties of red plum juice and permeate flux during membrane clarification

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ABSTRACT

Red plum juice was clarified by membrane processing by analyzing several parameters, including pore size, transmembrane pressure (TMP), feed flow rate, temperature, and membrane type, to optimize the performance of the membrane permeate flux and the quality of the clarified juice. Membrane clarification did not have a significant effect on pH, while total phenol content, anthocyanin, antioxidant activity, acidity, total soluble solid content, and turbidity decreased remarkably (about 99% rejection). The analytical results demonstrated that there was no significant difference by examining the permeate flux obtained using various pore sizes. Also, the highest steady-state flux (3.4 kg m^{-2} s^{-1}) occurred when the process was operated at 1.3 bar TMP. Moreover, increasing the velocity from 0.2 to 0.5 m s^{-1} and temperature from 20 to 40°C decreased the total fouling resistance by about 45 and 50%, respectively. Finally, based on the results, the mixed cellulose ester membrane with a pore size of 0.1 μm, a TMP of 1.3 bar, a velocity of 0.5 m s^{-1}, and a temperature of 40°C was selected as the most suitable membrane and operating conditions for clarification of red plum juice.

Keywords: Red plum juice; Membrane filtration; Fouling; Physicochemical properties

1. Introduction

Red plum juice is popular worldwide because of its attractive color, aroma and flavor, and its nutritional value. The nutritional importance of red plum is mainly due to its phenolic compounds, such as flavonoids and phenolic acid, which reduces the risk of oxidative damage and counteracts different types of cancer [1–3]. In some countries, such as Iran, plum juice, which is consumed at home or at local health food stores, is only available in specific seasons, because the turbidity of the juice has an unfavorable effect on its color and shelf-life (as it causes post-bottling haze formation). However, filtration removes the haze-causing components and produce plum juice that can be consumed all through the year [4]. Conventional clarification methods have been essentially replaced with membrane clarification; specially, microfiltration (MF) and ultrafiltration (UF) [5]. Membrane processing is one of the most important operations in industrial fruit juice processing. In comparison with conventional juice-processing methods, its advantages are low cost, mild required temperatures, ease of scale-up and simplicity of operation,

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which involves no phase change or chemical agents [6,7].

As reported by many studies, membrane fouling is a major obstacle in the economical and industrial application of membrane processing. It can occur by accumulation of compounds with high molecular weights, such as pectin, micro-organisms, and proteins, over the membrane surface. This blocks the membrane's pores with either a cake layer or other types of pore blockages [8], which may lead to reduction of product rate and juice quality [9]. Several efforts have been made to control or eliminate membrane fouling, including the use of shear-enhanced processing, fabrication of anti-fouling membranes, and pretreatment of feed juice [10,11]. Among these approaches, filtration at the best operating conditions, which minimized membrane fouling and maximized permeate flux, is technically attractive. Fouling phenomena can exhibit total resistance (R_t), cake resistance (R_c), reversible fouling resistance (R_{rev}), irreversible fouling resistance (R_{irr}), and membrane resistance (R_m) [12]. Several studies have been performed to study the clarification of fruit juices such as pomegranate [13], blood orange [14], pineapple [7], kiwifruit [15], and apple [16], using UF and MF. However, no studies have examined red plum juice.

In this study, the effect of different operating conditions, including pore size (0.22, 0.1, and 0.025 μm), transmembrane pressure (TMP) (0.5, 1.3, 2.1, and 2.9 bar), feed flow rate (0.2, 0.5, and 0.8 m s^{-1}), temperature (20, 30, and 40°C), membrane type (mixed cellulose ester (MCE), and polyvinylidene fluoride (PVDF)), on the performance of permeate flux was evaluated to identify the optimum clarification treatments (Table 1). The effects of operating conditions on fouling resistances were evaluated as well as their effects on physicochemical properties.

2. Materials and methods

2.1. Juice extraction

Red plum fruit (*Prunus domestica*) *vt*. Vampire was purchased from a local market (Karaj, Iran). Juice was manually extracted from mature and fresh fruits, and large particles such as peel were removed using a mesh filter (pore diameter = 2 mm).

2.2. Membrane setup

A cross-flow membrane unit with a flat sheet module in batch mode was used at laboratory scale (Fig. 1). A hydrophilic PVDF flat membrane with a pore size of 0.22 μm, and MCE flat membranes with pore sizes of 0.22, 0.1, and 0.025 μm and a total effective filtration area of 0.0209 m^2, were used in this study (Millipore, Billerica, MA, USA). Feed temperature was controlled using a two-layer jacket tank and a water bath. An inverter (LS, Model sv015ic5-1f, Cheongju, South Korea) coupled with a transmitter (WIKA, type ECO-1, Klingenberg, Germany) was used to hold the pressure of the rotary van pump (PROCON, Series 2, Milano, Italy) at the required level for different flow rates. Two separate pressure meters (WIKA, model 2 13.53.06 3, Klingenberg, Germany) were used to measure pressure in the feed and retentate sides. Permeate was collected in a permeate tank and was weighed to measure permeate flux. Retentate was recycled to the feed tank.

Table 1
Design of operating condition in experiments

Membrane type	Pore size (m)	Velocity (m s^{-1})	Temperature (°C)	Pressure (bar)
MCE	0.025	0.2	20	0.5
	0.1	0.2	20	0.5
	0.22	0.03	20	2.9
				2.1
				1.3
				0.5
		0.2	20	0.5
			30	
			40	
		0.5	20	0.5
		0.8	20	0.5
PVDF	0.22	0.2	20	0.5

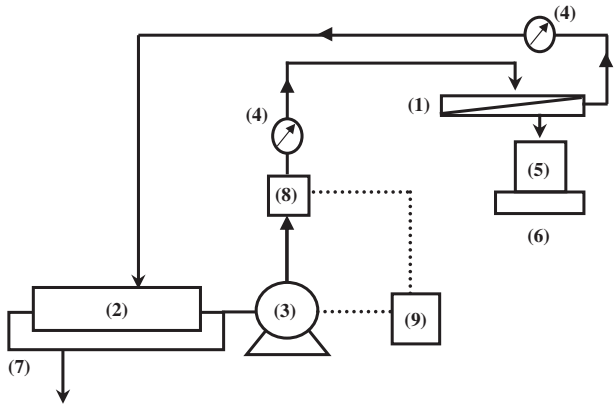

Fig. 1. Plate and frame membrane unit: (1) membrane module, (2) feed tank, (3) pump, (4) pressure meter, (5) permeate tank, (6) balance, (7) water bath, (8) transmitter, (9) inverter, (10) valve.

2.3. Red plum juice analysis

The soluble solid content of samples was measured by a hand refractometer (ATAGO, HSR-500, Tokyo, Japan). Juice turbidity changes were measured by a portable turbidimeter (WTW, 350 IR, Weilheim, Germany), and pH was determined by a digital pH meter at 25°C (Metrohm, Herisau, Switzerland). The titrable acidity was analyzed using the AOAC method and expressed as g malic acid in 100 g juice [17]. Color differences were measured by a colorimeter (Hunter lab, Model 45, USA) and expressed as L^*, a^*, and b^* values.

Total anthocyanin content (TAC) was measured using the pH-differential method by UV–visible spectrophotometer (CECIL, Model 2502, Cambridge, UK). Two solutions were prepared: a potassium chloride buffer (0.025 M), pH 1.0, and a sodium acetate buffer (0.4 M), pH 4.5. After that, 0.4 mL of diluted juice (1:10) was added to 3.6 mL of the prepared solutions. The absorbance of each dilution was determined at $\lambda_{vis\text{-}max}$ (510 for cyanidin 3-rutinoside as the dominant anthocyanin in red plum juice [18]) and 700 nm (to correct for haze) against a blank cell filled with distilled water [19]. Finally, TAC was calculated using the Eq. (1):

$$\mathrm{TAC}\left(\frac{\mathrm{mg}}{\mathrm{L}}\right) = \frac{A \times \mathrm{MW} \times \mathrm{DF} \times 100}{\mathrm{MA}} \quad (1)$$

where A is absorbance of diluted samples and calculated by the Eq. (2):

$$A = (A_{510} - A_{700})_{\mathrm{pH}\,1.0} - (A_{S10} - A_{700})_{\mathrm{pH}\,4.5} \quad (2)$$

MW is the molecular weight (595 for the cyanidin 3-rutinoside), DF is the dilution factor, and MA is the molar absorptivity of dominant anthocyanin (7,000 for the cyanidin 3-rutinoside).

Antioxidant activity (AA) of red plum juice was determined by the 2, 2 diphenyl-1-picrylhydrazyl (DPPH) method, based on the evolution of free radical-scavenging activity, as described by Sanchez-Moreno et al. [20] with some modifications. In this method, 0.1 mL of juice diluted at the ratio of 1:10 with distilled water was added to 2.46 mL of the DPPH solution (0.025 g L^{-1} in methanol) and mixed vigorously by vortex. After standing at room temperature for 30 min in darkness, the absorbance of the samples and control (0.1 mL methanol instead of plum juice) was measured at 517 nm by UV–visible spectrophotometer (CECIL, Model 2502, Cambridge, UK). Finally, the AA was calculated using the Eq. (3) [21]:

$$\text{Antioxidant Activity (AA)} = \left(1 - \frac{A_{\text{sample (517 nm)}}}{A_{\text{control (517 nm)}}}\right) \times 100 \quad (3)$$

The total phenolic content of samples was determined using the Folin and Ciocalteu method described by Singleton and Rossi [22]. In this method, 1 mL diluted sample or standard solutions of galic acid was added to a 25 mL balloon containing 9 mL distilled water. After that, 1 mL Folin and Ciocalteu's phenol reagent was added to samples and mixed. After 5 min, 10 mL Na_2CO_3 (7%) solution was added with mixing. The solution was then immediately diluted to a volume of 25 mL with distilled water and mixed thoroughly. The blank solution was prepared using the same recipe but with water instead of juice. After incubation at room temperature for 90 min, the absorbance of the samples vs. the prepared blank was measured by UV–visible spectrophotometer (CECIL, Model 2502, Cambridge, UK) at 750 nm. Finally, total phenolic content was reported as g GAE L^{-1} plum juice [3].

The varieties between parameters values in feeds do not permit us to compare changes in each characteristic at different conditions. To solve this problem, the rejection factor (R) was calculated according to the Eq. (4):

$$R = \frac{\varphi_F - \varphi_P}{\varphi_F} \quad (4)$$

where R is the rejection factor for parameter φ, and F and P are the amounts of parameter φ in feed and permeate, respectively.

2.4. Theory

The permeate flux of treated solutions was determined according to the Eq. (5):

$$J = \frac{\Delta m}{A \times t} \tag{5}$$

where Δm is the permeate weight (kg), A is the effective membrane surface (m^2), and t is time (h).

TMP was measured using the Eq. (6):

$$\text{TMP} = \frac{p_a + p_b}{2} - p_c \tag{6}$$

where p_a, p_b, and p_c are the pressures on the feed, retentate, and permeate side, respectively. p_c is approximately zero.

After juice processing, membrane was washed with distilled water for 30 min at minimum pressure and maximum flow rate. The cleaning process was followed by circulation of NaOH (0.5% w/w) for 30 min and then, circulation of HCl (pH 1) for 30 min at same pressure and flow rate. If the cleaning process has been effective and foulants have been removed from the membrane, the membrane system will provide the same performance again in the next process cycle [23].

Total resistance, including membrane and fouling resistance, was computed using the equation [12]:

$$R_T = R_m + R_f \tag{7}$$

where R_m is the intrinsic membrane resistance caused just by membrane, and R_f is fouling resistance including cake (R_c), reversible (R_{rev}), and irreversible (R_{irr}) resistance. They can be calculated according to the Eqs. ((8) and (9)):

$$R_m = \frac{1}{\mu_W L_p^0}, \quad L_p^0 = \frac{J_W}{\Delta P} \tag{8}$$

$$R_f = \frac{\Delta P}{\mu_p J_p} - R_m \tag{9}$$

where μ_w is water viscosity (Pa s), L_p^0 is the hydraulic permeability of new membrane ($m\,Pa^{-1}\,s^{-1}$) ΔP is TMP (Pa), J_w is the permeate flux of the membrane before experiment ($kg\,m^{-2}\,s^{-1}$), and μ_p and J_p are red plum permeate viscosity and flux, respectively. Reversible and irreversible fouling resistances were measured by the Eqs. ((11) and (12)):

$$R_{irr} = \frac{1}{\mu_W L_p^1} - R_m, \quad L_p^1 = \frac{J_W^1}{\Delta P} \tag{10}$$

$$R_{rev} = \frac{1}{\mu_p L_p^2} - R_m - R_{irr}, \quad L_p^2 = \frac{J_W^2}{\Delta P} \tag{11}$$

In these equations, L_p^1 and L_p^2 are the membrane hydraulic permeabilities after washing by water, alkaline and acid detergents, and after washing with water, respectively, at maximum velocity and minimum pressure for 30 min. J_W^1 and J_W^2 are water fluxes after each cleaning.

Finally, cake resistance (R_c) was calculated by the Eq. (13):

$$R_c = R_t - R_m - R_{irr} - R_{rev} \tag{12}$$

2.5. Statistical analysis

Results were expressed as the mean of triplicate determinations. Statistical analysis of data was performed using one-way analysis of variance. The mean comparisons were carried out using Duncan's multiple range tests by Minitab 15 software.

3. Results and discussion

3.1. Effect of pore size on permeate flux and physicochemical properties

Processing was performed at different pore sizes in selected operating conditions (TMP = 0.5 bar; velocity = 0.2 m s^{-1}; and temperature = 20 °C) to evaluate the effect of pore size on membrane processing. In general, at the initial stage of the process permeate flux declined rapidly (Fig. 2). The reduction rate became slower until it reached a steady state; this was related to the clogging of pores and formation of a cake layer [18]. Foulants likely included juice compounds such as cell wall, pectin, cellulose, lignin, and hemicellulose [24]. The permeate fluxes obtained from the membranes with pore sizes of 0.22, 0.1, and 0.025 μm were 11.42, 11, and 9.82 $kg\,m^{-2}\,s^{-1}$ at the beginning of the process, respectively. They reached an equal value at the end of the process, despite increases in the pore size of between 2 and 10 times. Since the effect of membrane pore size is a marginal, the filtration is cake layer controlling. Hence, flux may not necessarily improve with increasing pore size. This can be due to the fact that membranes with greater pore size are more prone to fouling, since more severe pore blocking, as well as

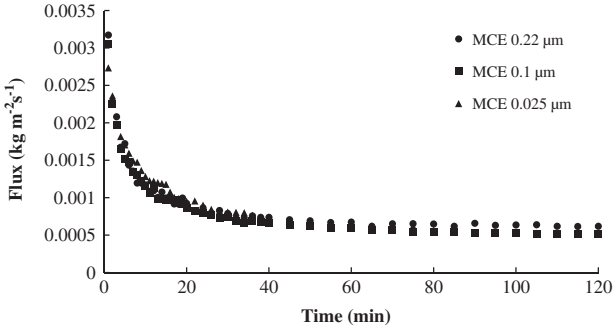

Fig. 2. Time course of permeate flux during filtration of red plum juice with various membrane pore sizes (cross-flow velocity 0.2 m s^{-1}, TMP 0.5 bar, and temperature 20°C).

accumulation of more particles, can occur in larger pores [25]. These results were in agreement with Girard and Fukumoto [26] and Laorko et al. [7]. Consideration of fouling phenomena showed that cake resistance in the PVDF membrane was 46% more than in the MCE membrane (Fig. 3). The hydrophobic characteristics of PVDF membrane prevent fluid movement into the membrane pores and compress particles on the membrane surface as a cake layer. The total resistance of the 0.025 µm membrane was noticeably higher than those of the 0.22 and 0.1 µm membranes. Cake resistance made up the major part of this additional resistance. Similarly, this was observed by Razi et al. [9] in their experiments on clarification of tomato juice with membrane filtration. The main cause of this phenomenon was attributed to the membrane's smaller pore size compared to the size of juice particles. Furthermore, the R_{rf} increased with smaller pore size, inversely to R_{if}. This could be because the smaller pore size led to a higher concentration polarization; this sort of

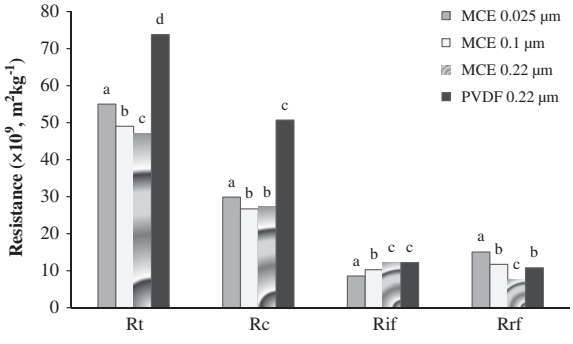

Fig. 3. The effect of membrane type and pore size on resistance (same letters in each resistance shows no significant difference between values).

blockage is removable. On the other hand, the larger pore membrane had more opportunities for internal pore blocking, which is not removable by washing. In this study, R_m of the 0.22, 0.1, and 0.025 µm membranes was 0.10, 0.339, and 1.50 (×10^9 m^2 kg^{-1}), respectively, which is negligible compared to the fouling resistance.

Chemical and physical properties of juices before and after treatment with various membrane pore sizes are shown in Table 2. The results showed that remarkable removal had been achieved in all studied parameters, except pH. Moreover, all membranes effectively removed juice turbidity by 99% after removal of suspended solid components. The reduction in density and soluble solid content confirmed this. These observations corroborate the results obtained by Cassano et al. [6] and Mirsaeedghazi et al. [13]. The brightness of the clarified juice, indicated by L*, was lighter and more clear than that of fresh juice for all treatments. Table 2 shows that the acidity value of clarified juice decreased with membrane processing. This decrease can be attributed to the rejection and accumulation in the cake layer of organic acid components affecting acidity. This was also reported by Mirsaeedghazi et al. [13] and Loarko et al. [7] in the cross-flow filtration of pomegranate and pineapple juice, respectively. Table 2 also shows the effect of membrane filtration on the total phenolic content, anthocyanin, and AA of red plum juice. Reduction of phenolic components such as anthocyanin was associated with the formation of a cake layer that affected membrane selectivity; therefore, they were not easily able to pass from the membrane. This directly decreased the AA.

Fig. 4 show the rejection parameter for various membrane types and pore sizes regarding the physicochemical properties of red plum juice. A comparison of UF and MF membranes was helpful in finally choosing the most suitable membrane with the highest physicochemical recovery. In an overall comparison between MCE and PVDF membranes as different types, it could be found that better recovery was made by MCE; for example, PVDF and MCE showed recoveries of 57 and 63%, respectively, regarding AA. Since PVDF membrane is less hydrophilic than MCE, rejection of this type is higher. As discussed, a remarkable reduction in turbidity was observed in the permeate fraction of all membranes (over 99% for the UF and MF processes). Rejections of 44, 31, and 17.9% were observed for the 0.22, 0.1 (as MF), and 0.025 µm (as UF) membranes, respectively, regarding total phenol content. For other nutritional compounds, such as anthocyanin, reductions of 32, 27, and 30% were noted in permeates for the 0.22, 0.1, and 0.025 µm membrane pores, respectively. In addition, with 20% rejection,

Table 2
Physicochemical properties of red plum juice before and after filtration at various membrane types and pore sizes

	MCE 0.22 m		MCE 0.1 m		MCE 0.0.25 m		PVDF 0.22 m	
	Feed	Permeate	Feed	Permeate	Feed	Permeate	Feed	Permeate
TSS (°Brix)	13.5[a]	10.2[b]	13.6[a]	11.65[b]	13.35[a]	11.45[b]	13.5[a]	10.2[b]
Acidity (% w/w malic acid)	1.69[a]	1.42[b]	1.68[a]	1.46[b]	1.87[a]	1.63[b]	1.72[a]	1.4[b]
Turbidity (NTU)	3,750[a]	19[b]	2,900[a]	10.5[b]	2,977[a]	9.7[b]	4,053[a]	36[b]
pH	3.25a	3.26[a]	3.27[a]	3.30[a]	3.29[a]	3.29[a]	3.27[a]	3.29[a]
Density (kg m^{-3})	1,061[a]	1,053[b]	1,053[a]	1,045[b]	1,055[a]	1,035[b]	1,060[a]	1,055[b]
Color								
L^*	19[a]	30[b]	15.17[a]	25.14[b]	17.24[a]	25.13[b]	21.32[a]	24.48[b]
a^*	22.50[a]	39.60[b]	22.67[a]	44.10[b]	22.08[a]	46.22[b]	22.4[a]	37.99[b]
b^*	9.5[a]	30.56[b]	8.1[a]	36.67[b]	8.6[a]	39.36[b]	12[a]	32.44[b]
Total phenol (g GAE L^{-1})	7.2[a]	4.0[b]	10.47[a]	7.15[b]	8.3[a]	6.82[b]	10.6[a]	5.40[b]
Anthocyanin (mg L^{-1})	51.6[a]	35.25[b]	72.67[a]	53[b]	64.6[a]	45.48[b]	62.4[a]	41[b]
Antioxidant activity (%)	62.6[a]	40.05[b]	78.5[a]	60[b]	68.6[a]	55[b]	77.43[a]	44.3[b]

Notes: MCE, mixed cellulose ester.
PVDF, polyvinylidene fluoride.
Same letters in each row of one membrane present no significant difference based on Duncan's multiple range tests at $p < 0.05$.

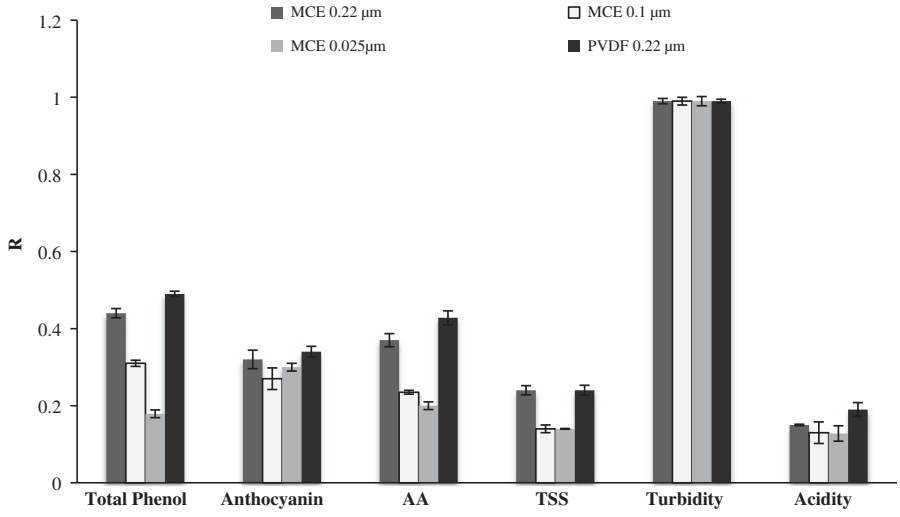

Fig. 4. Rejection factor of some chemical properties of red plum juice after clarification with different membrane types and pore sizes.

the UF membrane gave the highest recovery of AA in permeate. Significant recovery difference between 0.1 and 0.025 µm membranes was not observed for either acidity or total soluble solids (TSS), while the 0.22 µm membrane had more rejection.

Finally, the 0.1 and 0.025 µm membranes showed essentially equal recovery of valuable components, but the total resistance of the 0.1 µm membrane was less than that of the 0.025 µm; therefore, the MCE membrane with pore size of 0.1 µm was selected as the best membrane for achieving optimal permeate during clarification of red plum juice. Due to its higher rejection of nutritional material, the 0.22 µm membrane had no chance to be chosen for this purpose.

3.2. Effect of TMP on membrane clarification of red plum juice

To consider the effects of TMP on permeate flux and physicochemical properties, the juice was treated using the 0.22 µm MCE membrane at velocity of 0.03 m s^{-1} and temperature of 21 °C. The permeate flux decreased over time (Fig. 5). Also, the higher the TMP that was applied, the more permeate flux was

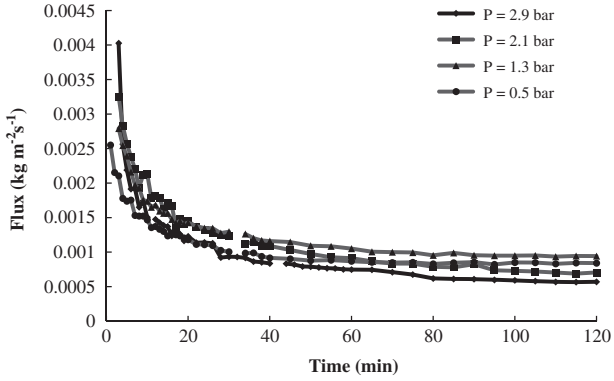

Fig. 5. Permeate flux over time during membrane filtration of red plum juice at various TMPs (MCE 0.22 μm, cross-flow velocity 0.03 m s^{-1}, and temperature 20 ℃).

obtained at the initial stage, since these membrane processes (MF and UF) are pressure-driven. Moreover, over time, the differences among the permeate fluxes decreased, and the cake layer that had formed acted as a selective layer. Fig. 5 show that the flux reduction rate was intensified by increasing the TMP. The permeate flux value obtained from a 1.3 bar TMP was 3.4 kg m^{-2} s^{-1} at the end of process; this was the highest flux of all TMPs tested. In this condition, as reported by Cassano et al. [6], flux becomes independent of pressure and any further increase in pressure has no considerable effect. This is due to increased precipitation and movement of the colloidal particles toward the membrane, which builds up a denser cake layer on the membrane surface at higher pressures [12,16]. This can also be confirmed by analysis of membrane resistance at different pressures (Fig. 6). R_T during membrane filtration was 29.5, 72.4, 132.4, and 208.9 ($\times 10^9$ m^2 kg^{-1}) for 0.5, 1.3, 2.1, and 2.9 bar TMPs, respectively. The higher permeate flux obtained at higher pressures (at the initial stage) can move more particles toward the membrane surface, which can increase membrane fouling [27]. Hojjatpanah et al. [5] presented similar results for the membrane clarification of black mulberry juice. In addition, there was a noticeable increase among irreversible resistances: 9.54, 21.3, 30.8, and 40.4 ($\times 10^9$ m^2 kg^{-1}) were obtained for 0.5, 1.3, 2.1, and 2.9 bar, respectively. Increasing pressure results in the further penetration of particles into the membrane pores.

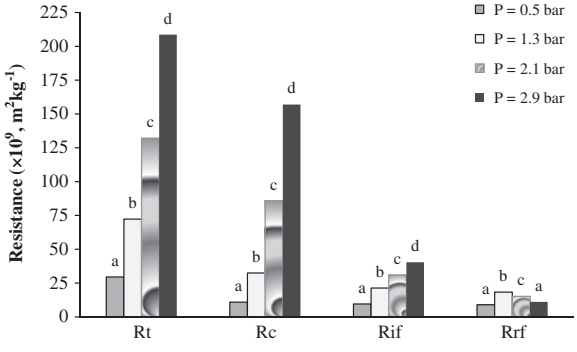

Fig. 6. The effect of different pressures on resistance (same letters in each resistance shows no significant difference between values).

Table 3 and Fig. 7 shows the analytical measurements and rejection factors for physicochemical properties carried out on samples from the feed and permeate of the processes at the different pressures. Better recovery of important components was accomplished at higher filtration pressures. Rejections of 32, 31, 22, and 19% were observed for TMPs of 0.5, 1.3, 2.1, and 2.9 bar, respectively, towards polyphenols. More force on the particles and components passing through the membrane is applied by intensifying pressure; this raises the amount of macromolecules in the permeate. This enhanced the permeate turbidity at pressures of 2.1 and 2.9 bar, achieving rejections of 97 and 93%, respectively. The brightness of the permeate (L^*) at a pressure of 2.9 bar was not improved by clarification. Moreover, since a reduction in acidity was ascribed to the cake layer, its further rejection at the higher pressures (2.1 and 2.9 bar) was justifiable.

However, any increase in operating pressure elevates energy cost, and fouling becomes a main problem. With regard to proper flux behavior (maximum steady-state permeate flux), good recovery of physicochemical properties and appropriate turbidity rejection, a TMP of 1.3 bar was preferred as the best operating pressure for optimal permeate flux.

3.3. Flux behavior and physicochemical changes during testing with different cross-flow velocities

Red plum juice was subjected to three velocities at temperature 20 ℃ and pressure 0.5 bar with a 0.22 μm membrane to study the effect of feed velocity on flux behavior and physicochemical changes. It was observed that the permeate flux was enhanced when the feed velocity was increased (Fig. 8). At the higher velocities, more wall shear stress was created on the membrane surface, since precipitation of colloidal particles diminished; consequently, concentration polarization and reversible fouling decreased on the membrane surface [12,28]. However, increasing the feed velocity from 0.5 to 0.8 m s^{-1} had no significant

Table 3
Physicochemical properties of red plum juice submitted to the membrane treatment at various pressures

	0.5 bar		1.3 bar		2.1 bar		2.9 bar	
	Feed	Permeate	Feed	Permeate	Feed	Permeate	Feed	Permeate
TSS (°Brix)	13[a]	11[b]	13[a]	11.35[b]	13.45[a]	11.85[b]	12.8[a]	11.35[b]
Acidity (% w/w malic acid)	1.7[a]	1.45[b]	1.78[a]	1.56[b]	1.82[a]	1.45[b]	1.8[a]	1.5[b]
Turbidity (NTU)	3,650[a]	18[b]	3,750	24[b]	1,965[a]	48[b]	4,050[a]	245[b]
pH	3.13[a]	3.15[a]	3.16[a]	3.16[a]	3.21[a]	3.23[a]	3.29[a]	3.30[a]
Density (kg m^{-3})	1,061[a]	1,053[b]	1,070[a]	1,055[b]	1,065[a]	1,055[b]	1,075[a]	16.34[a]
Color								
L^*	17.5[a]	27.84[b]	17.50[a]	28.14[b]	15.11[a]	23.30	16.50[a]	16.34[a]
a^*	20.7[a]	35.1[b]	20.70[a]	38.42[b]	22.58[a]	42[b]	22.50[a]	33.50[b]
b^*	8.4[a]	26.80[b]	8.40[a]	25.24[b]	8.33[a]	31.10[b]	8[a]	21.10[b]
Total phenol (g GAE L^{-1})	10.30[a]	7.03[b]	9.33[a]	6.4[b]	8.96[a]	6.95[b]	9.89[a]	8.03[b]
Anthocyanin (mg L^{-1})	67.50[a]	48.02[b]	63.3[a]	51.8[b]	66.3[a]	54.45[b]	66.3[a]	56.3[b]
Antioxidant activity (%)	76.7[a]	60[b]	70[a]	55[b]	71[a]	60[b]	74[a]	65[b]

Note: Same letters in each row of one pressure present no significant difference based on Duncan's multiple range tests at $p < 0.05$.

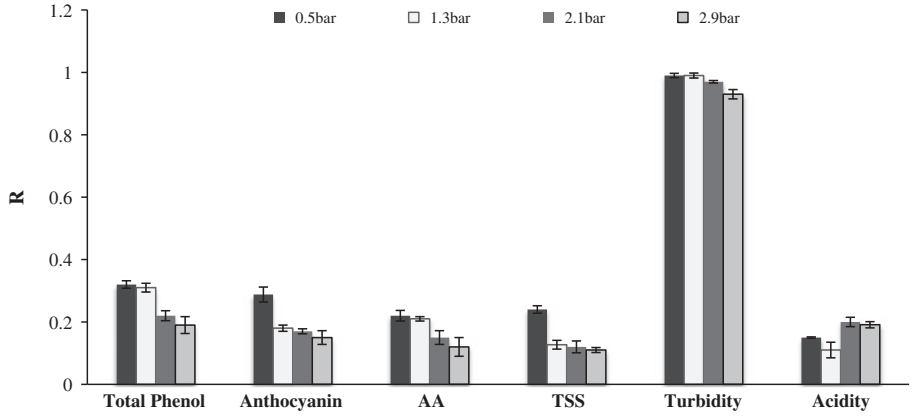

Fig. 7. Rejection factor of some chemical properties of clarified red plum juice at different TMPs.

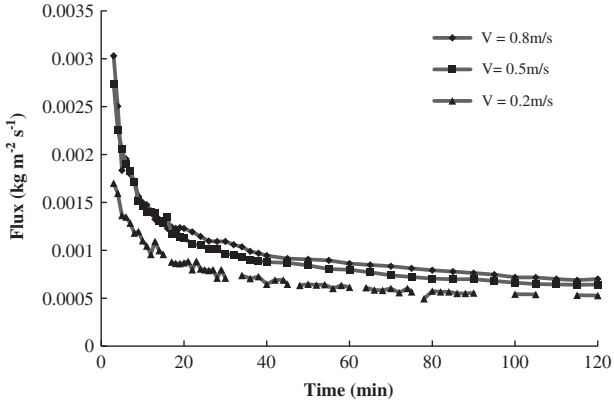

Fig. 8. Permeate flux of red plum juice vs. time during membrane filtration at various velocities (MCE 0.22 μm, TMP 0.5 bar, and temperature 20°C).

influence on the permeate flux (Fig. 8). Fig. 9 illustrate the effect of feed velocity on the fouling resistances during clarification of red plum juice. In general, resistance did not change with an increase in velocity from 0.5 to 0.8 m s^{-1}. Nevertheless, total resistance decreased by about 45% when the feed flow rate was increased from 0.2 to 0.5 m s^{-1}. Cake resistance contributed the most to the reduction in total fouling resistance, falling by about 60% due to sweeping by tangential forces of the cake deposited on the membrane surface.

Physicochemical characteristics and mean rejection factor of red plum juice submitted to membrane treatment at various feed velocities are shown in Table 4 and Fig. 10. Consideration of the turbidity and color parameters of the permeate in all tests showed that the permeate juice had appropriate clarity and appearance.

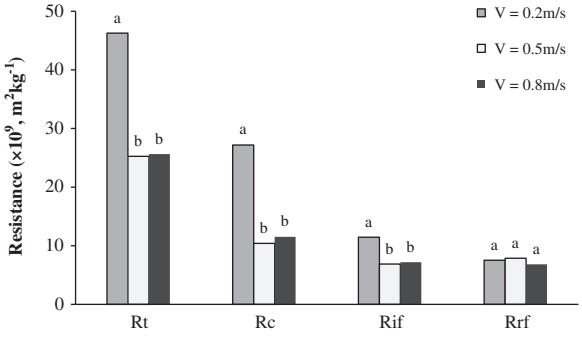

Fig. 9. The effect of various velocities on resistance (same letters in each resistance shows no significant difference between values).

With increased feed velocity, rejection of total phenol content, anthocyanin, AA, and TSSs decreased, since at the higher velocities, total resistance and cake layer fouling also decreased. However, a comparison between velocities of 0.5 and 0.8 m s^{-1} showed that lower velocity had better recovery rather than another. As discussed previously, raising velocity from 0.5 to 0.8 m s^{-1} did not greatly decrease cake layer resistances; also, components had less opportunity to pass from the membrane at the higher velocity. Recoveries of 74, 75, and 83% were observed for 0.5 m s^{-1} velocity regarding total phenol, anthocyanin, and AA, respectively. Finally, with regard to these results, a velocity of 0.5 m s^{-1} was preferred as the best operating cross-flow velocity for clarification of red plum juice.

Table 4
Physical and chemical characteristics of fresh and clarified red plum juice at various velocities

	0.2 m s^{-1}		0.5 m s^{-1}		0.8 m s^{-1}	
	Feed	Permeate	Feed	Permeate	Feed	Permeate
TSS (°Brix)	13.5^a	10.2^b	13.65^a	11.45^b	13^a	11^b
Acidity (% w/w malic acid)	1.69^a	1.42^b	1.83^a	1.43^b	1.77^a	1.61^b
Turbidity (NTU)	3750^a	19^b	2905^a	13^b	2982^a	11^b
pH	3.25^a	3.26^a	3.25^a	3.29^a	3.21^a	3.21^a
Density (kg m^{-3})	1,061^a	1,053^b	1,056^a	1,047^b	1,056^a	1,049^b
Color						
L*	19^a	30^b	18^a	22.84^b	18^a	21.71^b
a*	22.50^a	39.60^b	23.3^a	42^b	23.3^a	44.41^b
b*	9.5^a	30.56^b	10.4^a	30.66^b	10.4	31.8^b
Total phenol (g GAE L^{-1})	7.2^a	4.0^b	9.82^a	7.25^b	9.76^a	6.6^b
Anthocyanin (mg L^{-1})	51.6^a	35.25^b	65.7^a	48.9^b	66.7^a	50.7^b
Antioxidant activity (%)	62.6^a	40.05^b	73.21^a	60.65^b	73^a	56^b

Note: Same letters in each row of one velocity present no significant difference based on Duncan's multiple range tests at $p < 0.05$.

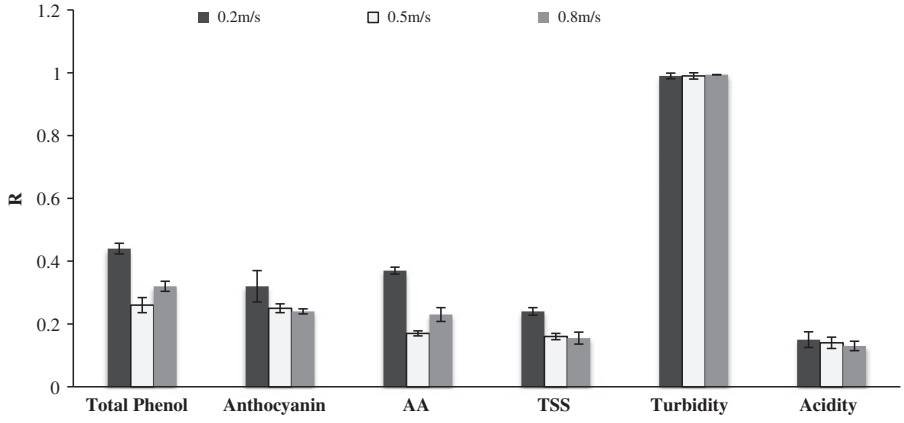

Fig. 10. Rejection factor of some chemical properties of red plum juice after clarification at different velocities.

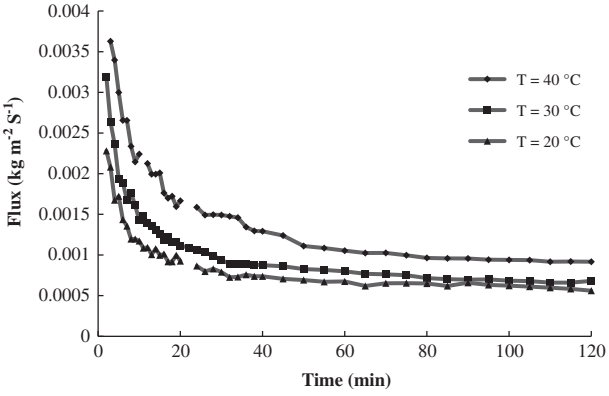

Fig. 11. Permeate flux during membrane filtration of red plum juice at different temperatures (MCE 0.22 μm, cross-flow velocity 0.2 m s^{-1}, and TMP 0.5 bar).

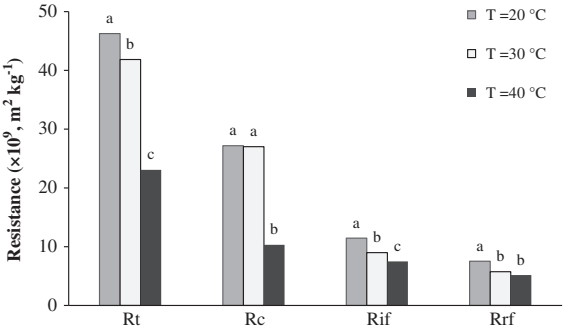

Fig. 12. The effect of different temperatures on resistance (same letters in each resistance shows no significant difference between values).

3.4. Influence of feed temperature on permeate flux and physicochemical characteristic

Several parameters, including TMP, feed flow, and temperature, should be studied for the optimization of membrane filtration performance [29]. For this purpose, the effect of temperatures of 20, 30, and 40˚C on permeate flux and physicochemical characteristics at a TMP of 0.5 bar and a cross-flow velocity of 0.2 m s^{-1} with a 0.22 μm MCE membrane was examined. Fig. 11 show the plot of permeate flux vs. time. The permeate value improved with increased operating temperature. The increase of temperature reduces feed viscosity and intensifies the diffusion coefficient of macromolecules, consequently enhancing mass transfer and permeation rate. This behavior was similar to that reported in other studies [29–31]. Meanwhile, the results showed that the permeate flux decreased gradually with the operating time due to concentration polarization and gel formation. Moreover, there was a remarkable discrepancy between permeate flux obtained from elevating temperature from 20 to 30˚C and from 30 to 40˚C: an efficiency effect on permeate flux was not achieved with a 10˚C increase in temperature (from 20 to 30˚C). Studying the effect of feed temperature on the total fouling resistance showed that increasing the feed temperature from 20 to 30˚C could decrease the total fouling resistance by about 9% (Fig. 12). In contrast, the total fouling resistance fell by about 50% when heat treatment was enhanced from 20 to 40˚C, and cake resistance and irreversible and reversible fouling resistances decreased by 62, 34, and 32%, respectively. This is due to the greater molecular diffusivity and mobility of particles.

Table 5
Physical and chemical characteristics of red plum juice submitted to membrane treatment at different temperatures

	20˚C		30˚C		40˚C	
	Feed	Permeate	Feed	Permeate	Feed	Permeate
TSS (˚Brix)	13.5^a	10.2^b	13.5^a	11.45^b	13^a	11^b
Acidity (% w/w malic acid)	1.69^a	1.42^b	1.93^a	1.6^b	2.12^a	1.81^b
Turbidity (NTU)	3750^a	19^b	2985^a	13.5^b	3555^a	13.5^b
pH	3.25^a	3.26^a	3.10^a	3.12^a	3.09^a	3.11^a
Density (kg m^{-3})	$1{,}061^a$	$1{,}053^b$	$1{,}060^a$	$1{,}048^b$	$1{,}063^a$	$1{,}054^b$
Color						
L^*	19^a	30^b	18.3^a	23.63^b	17.55^a	19.44^b
a^*	22.50^a	39.60^b	23.55^a	43.2^b	24.38^a	42.15^b
b^*	9.5^a	30.56^b	10.75^a	31.26^b	9.85^a	30.44^b
Total phenol (g GAE L^{-1})	7.2^a	4.0^b	10.56^a	7.81^b	9.86^a	7.85^b
Anthocyanin (mg L^{-1})	51.6^a	35.25^b	64.31^a	45.9^b	62.05^a	49.3^b
Antioxidant activity (%)	62.6^a	40.05^b	78^a	62.1^b	72.7^a	64.4^b

Note: Same letters in each row of one temperature present no significant difference based on Duncan's multiple range tests at $p < 0.05$.

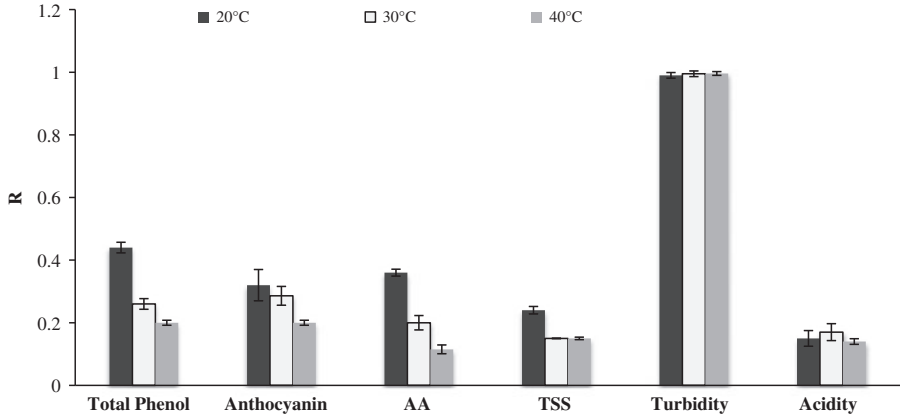

Fig. 13. Rejection factor of some chemical properties of clarified red plum juice at different temperatures.

Changes in different parameters at various temperatures are shown in Table 5 and Fig. 13. The mean rejection factor of prominent components decreased with increases in feed temperature. For example, approximate anthocyanin losses of 32, 28, and 20% and total phenol losses of 44, 26, and 20% were observed at 20, 30, and 40°C, respectively, due to the increased mass transfer coefficient of particles. Study of L^* and a^* values demonstrated that red plum juice was clarified and became light red. Also, the turbidity of permeate samples improved.

Finally, due to the highest permeate flux achieved and very fine recovery of important components, a 40°C value was selected as the best operating temperature for the membrane clarification of red plum juice.

4. Conclusions

The effect of membrane properties and operating parameters on the permeate flux, quality of clarified juice and membrane fouling were considered during filtration of red plum juice. Results showed that in all cases a totally clarified juice was obtained. Apart of pH, all other quality parameters were significantly different before and after clarification process. The cross-flow velocity and temperature showed positive effect on the permeate flux. As a final result, with regard to maximization of permeate output and quality along with minimization of energy consumption, the MCE membrane with pore size of 0.1 μm, TMP of 1.3 bar, velocity of 0.5 m s^{-1}, and temperature of 40°C was selected as the best membrane and operating conditions for the membrane clarification of red plum juice in point of view of the technological aspect. Filtration in these conditions enables to produce a clarified juice with physicochemical and nutritional properties similar to those of fresh juice.

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