International Food Research Journal 27(6): 1156 - 1166 (December 2020)
Journal homepage: *http://www.ifrj.upm.edu.my*

Vibration-assisted forward osmosis process for Mao (*Antidesma bunius* L. Spreng) juice concentration: water flux enhancement and preservation of phytochemicals

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Article history

Received: 19 February 2020
Received in revised form: 13 June 2020
Accepted: 2 September 2020

Keywords

forward osmosis, Mao juice, membrane vibration, flux enhancement

Abstract

The experimental water flux of the forward osmosis (FO) process is much lower than the theoretical flux due to the existence of the internal concentration polarisation (ICP), external concentration polarisation (ECP), and membrane fouling. In the present work, vibration was integrated with the FO process to enhance water flux in water and Mao (*Antidesma bunius* L. Spreng) juice concentration. In addition, the capability of the FO process in preserving phytochemicals was studied. The use of the vibration assisted technique could enhance the water flux up to 23% during the FO process of distilled water due to the reduction of ICP, and a much higher water flux enhancement (up to 70%) was attained during the FO of Mao juice due to the reduction of ICP, ECP, and fouling. Phytochemicals including total phenolic compounds, anthocyanin, and ascorbic acid were preserved up to 82.7, 72.6, and 95.9%, respectively. These results suggest that membrane vibration is a promising technique for the enhancement of the FO process performance.

Introduction

Consumers nowadays are more concerned about the nutrition and safety of food products. Therefore, foods that go through minimal processing and contain no preservatives or additives are in high demand. Fruit juices are of high nutritive value which is beneficial to human health. "Mao" (aka "Mao-Luang"; *Antidesma bunius* L. Spreng) is a tropical fruit-like-berry in the Euphobiaceae family. It is widely planted in the north-eastern part of Thailand. The ripe fruit contains high levels of bioactive compounds such as anthocyanin and ascorbic acid (Sripakdee *et al.,* 2015). These compounds are beneficial to human health due to their antioxidant properties (Jorjong *et al.,* 2015). In fruit juice processing, the concentration process is widely employed to enhance microbiological stability and to reduce packaging, storage, and transportation costs (Babu *et al.,* 2006). Conventionally, fruit juices are subjected to thermal processing to evaporate water, and consequently, to obtain the concentrated product. However, thermal processing requires a high amount of energy to heat and evaporate water, and has a severe impact on heat-sensitive compounds and organoleptic attributes (Sant'Anna *et al.,* 2012). For instance, anthocyanin is a heat-sensitive compound, and susceptible to degradation under heat treatments above 70°C (Sarkis *et al.,* 2013). Ascorbic acid on the other hand, tends to degrade rapidly at heating temperatures above 60°C (Vieira *et al.,* 2016). Because of the presence of heat-sensitive compounds in fruit juice, technologists have made attempts to develop novel technology that can avoid or minimise the use of heating process. Over the past decade, forward osmosis (FO) has been introduced to separate water from solution, allowing water to be obtained as well as concentrated solution. It has been widely studied for water desalination (Akther *et al.,* 2015) and liquid food processing (Chen *et al.,* 2019; Pei *et al.,* 2020). Studies indicate that FO is a promising process for the concentration of fruit juice and for bioactive compound solutions at ambient temperatures, allowing the minimisation of their deterioration (Nayak and Rasogi, 2010; Coday *et al.,* 2014; Sant'Anna *et al.,* 2016). In addition, the FO process requires less energy as compared to evaporation and reverse osmosis (RO; Sant'Anna *et al.,* 2012). FO also has low fouling tendency as compared to RO (Lee *et al.,* 2010). Besides, since it is operated at low temperatures and pressures,

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FO is less complex as compared to RO and evaporation. Therefore, FO likely requires less investment as well as operational costs.

FO is a membrane separation process, governed by the osmotic pressure difference of the feed and draw solutions, arising across a selective layer membrane. This osmotic pressure difference acts as a driving force to draw water from the feed solution (low concentration e.g., fruit juice) to the draw solution (high concentration e.g., NaCl solution). As a result, the feed concentration is increased while the draw solution concentration is decreased. The diluted draw solution is usually re-concentrated with suitable processes. For instance, it can be re-concentrated by evaporation using industrial waste heat (Chekli *et al.,* 2012). Additionally, the draw solution can be prepared from liquid waste discharged from food processing plant e.g., waste brine of cheese processing (Chen *et al.,* 2019). The water permeate rate or water flux is usually used to indicate FO process performance. It is known that the experimental FO water flux is much lower than the theoretical estimation, owing to the occurrence of internal concentration polarisation (ICP) and external concentration polarisation (ECP), as well as membrane fouling. The degree of ICP, ECP, and fouling in FO varies depending on membrane characteristic, operating condition, and nature of the feed and draw solutions. Membrane scientists have employed different techniques to control concentration polarisation and fouling in FO. Chanukya and Rastogi (2017) for instance, utilised ultra-sonication to improve water flux during the FO of sucrose solution. They found that ultrasound at 50 kHz could significantly reduce the ECP on the feed side and dilutive ICP on the draw side. However, the ultrasound could not mitigate the ECP and gel formation during FO of the feed containing pectin. Membrane vibration is another technique that has been employed to reduce concentration polarisation and fouling in pressure driven membrane processes such as ultrafiltration and RO (Sriniworn *et al.,* 2016; Su *et al.,* 2018). In FO, Yuan and Zhang (2013) studied the effect of membrane vibration and magnetic stirrer on the water flux under the PRO mode (feed face to support layer) of operation. The results indicated that the use of membrane vibration could remarkably enhance the water flux due to the reduction of ECP on the feed side. The water flux improvement using membrane vibration was much higher as compared to using a magnetic stirrer. They also reported that the enhancement of water flux was dependent on the membrane vibration frequency. They also suggested that the liquid film at the feed side which was compressed under vibration led to reduced film thickness and consequently increased the water flux. In fruit juice concentration, however, FO (active layer face to feed solution and support layer face to draw solution) was more suitable as compared to the PRO mode owing to the complexity of the juice containing wide range of components with low and high molecular weight components (Chanukya and Rastogi, 2017). In addition to the existing ICP of draw solution on the support layer, it is possible that components found in fruit juice would accumulate on the active layer surface, and consequently generating ECP as well as fouling. Therefore, it is possible that operating FO under vibrating conditions may lead to reduced membrane fouling, ICP, and ECP during FO of Mao juice.

In view of this, the main objectives of the present work were to study the potential use of membrane vibration technique to improve the water flux during the FO of Mao juice, under the FO mode of operation. The study focused on the effect of membrane vibration amplitude and concentration of the juice. In addition, selective-phytochemicals preservation during FO and the concentrated juice obtained were also studied.

Materials and methods

Chemicals

Sodium chloride (NaCl, molecular weight: 58.44 g/mol, Merck), Folin-Ciocalteu's phenol reagent (Merck), sodium carbonate ($NaHCO_3$, molecular weight: 84.01 g/mol, LOBA), gallic acid ($C_7H_6O_5$, molecular weight: 170.12 g/mol, Merck), potassium chloride (KCl, molecular weight: 74.56 g/mol, Merck), sodium acetate ($CH_3COONa.3H_2O$, molecular weight: 136.08 g/mol, Ajax), and 2,6 dichlorophenol-indophenol ($C_{12}H_7NCl_2O_2$, molecular weight: 268.1 g/mol, LOBA) were of analytical grade, and used as received.

Mao juice preparation

Mao juice was extracted by cold pressing followed by filtration using a filter cloth. The extraction and filtration were carried out in the Sakon-Nakhon Province, Thailand, and stored at -20°C before transporting to the laboratory. The Mao juice continued to be stored at -20°C until ready to be used.

Forward osmosis experiment

Forward osmosis membrane and system

Self-synthesised TFC membranes were used in all experiments. These membranes consisted of polyamide selective layer and polysulfone support

layer, and the properties of the membranes have been reported in our previous work (Sirinupong *et al.,* 2018). All FO experiments were carried out using a crossflow permeation cell with the total effective membrane area of 29.75 cm^2. NaCl solution (4 M) was used as draw solution, where feed was either distilled water or Mao juice. During FO experiment, the concentration of draw solution was maintained constantly by dosing of NaCl solution (6 M). The dosing process was properly carried out by monitoring the conductivity of the draw solution. The feed and draw solution were circulated in counter-current mode using two peristaltic pumps (Masterflex L/S) at a constant crossflow velocity (0.025 m/s) and temperature (25 ± 2°C). The FO membrane cell was vibrated by placing the cell between the plates of vibration generator (Endecotts, C.V.S.1., England). The upper chamber was filled with the feed (distilled water or Mao juice), while the lower chamber was filled with the draw solution.

Effect of membrane vibration amplitude on water flux and reverse solute flux during forward osmosis of distilled water

The experiments were carried out at a constant vibration frequency (3000 min^{-1}) but at different vibration amplitudes, i.e., 0 (control), 0.6, 1.2, and 1.8 mm. Each experiment was performed for 30 min with triplication to yield an average result. The water flux was determined through the changes in the weight of the draw solution. The draw solution tank was placed on a digital weight balance, and weight change was automatically logged onto a computer. The data was then used to calculate water flux (J_w) using Eq. 1. To analyse the reverse salt flux (J_s), feed solution conductivity was monitored as a function of time using a conductivity meter (Mettler-Toledo). The conductivity was then converted to solution concentration using conductivity-concentration calibration curve. J_s was then calculated using Eq. 2 (Emadzadeh *et al.,* 2016):

$$J_w = \frac{\Delta m/\rho}{A_m \Delta t} \quad \text{(Eq. 1)}$$

$$J_s = \frac{\Delta(C_t V_t)}{A_m \Delta t} \quad \text{(Eq. 2)}$$

where, Δm = weight change of draw solution (kg), ρ = density of the feed solution ($kg.m^{-3}$), A_m = effective membrane area (m^2), C_t and V_t = salt concentrations ($g.L^{-1}$) and the feed solution volume (L) at the end of experiment, respectively, and Δt = measured time period (h).

The water flux improvement was indicated by the ratio of water flux at different amplitudes of membrane vibration (J_{wi}) to control treatment (J_{w0}), and calculated using Eq. 3:

$$\text{Water Flux Ratio} = \frac{J_{wi}}{J_{w0}} \quad \text{(Eq. 3)}$$

The theoretical or ideal water flux (J_{wid}) across membrane in the FO mode of operation can be described using Eq. 4:

$$J_{wid} = A(\pi_d - \pi_f) \quad \text{(Eq. 4)}$$

where, π_d and π_f = osmotic pressures (Pa) of the draw and feed solutions, respectively, A = water permeability coefficient of the selective membrane ($kg.m^{-2}.h^{-1}.Pa^{-1}$). The theoretical water flux using this equation does not take into account the effects of ICP, ECP, and fouling that usually occur during FO. The water permeability of the self-synthesised membrane used was 1.12×10^{-9} $kg.m^{-2}.h^{-1}.Pa^{-1}$ (Sirinupong *et al.,* 2018). The osmotic pressure of the draw solution (NaCl 4 M) was estimated according to Geraldes *et al.* (2001). The flux reduction (FR), which is the ratio of water flux difference of the theoretical water flux and the water flux of feed solution (distilled water and Mao juice) to the theoretical water flux, can be used to indicate the significant effects of ICP and ECP plus fouling. The FR due to ICP was obtained during the FO of distilled water, while flux reduction due to ICP, ECP, and fouling was obtained during the FO of Mao juice. Then, the FR obtained during FO of distilled water was used to separate the significant on FR between ICP and combination of ECP and fouling.

Concentration of Mao juice by FO

The experimental procedure of Mao juice concentration using FO is described as follows: Firstly, distilled water (40 ± 2°C) was circulated on both sides of the TFC membrane for 30 min to clean the system. Prior to concentration, the water flux of clean membrane using distilled water as feed solution and NaCl as draw solution was determined (J_{w1}). After that, distilled water was replaced by Mao juice. Two stages of the FO process were employed to increase the concentration of Mao juice with the initial total soluble solid (TSS) content of 12 and 20 °Brix for the first and second stage, respectively. The feed solution in the second stage was the pre-mixed Mao juice concentrated obtained from the first stage to standardise the juice concentration. At the end of each concentration stage, the membrane was cleaned using a simple hydraulic washing method. Distilled water was introduced to the system to replace the Mao juice, and draw solution was circulated for 30 min at a constant crossflow velocity of 0.025 m/s (Lv *et al.,*2017). Following

hydraulic washing, distilled water and NaCl were used as the feed and draw solution respectively, to determine water flux (J_{w2}). The water flux recovery was demonstrated as the ratio of the water flux of fouled membrane to water flux of clean membrane, and calculated using Eq. 5:

$$\text{Water flux recovery (\%)} = \frac{J_{w2}}{J_{w1}} \times 100 \quad \text{(Eq. 5)}$$

In addition, Mao juice samples were taken during FO to determine the TSS content using a hand refractometer (ATAGO MASTER-53M 0-58 °Brix), and the other properties of samples taken (12, 15, 20, 25, 30, and 35 °Brix) were further analysed

Analysis of Mao juice properties

The properties of concentrated juice produced by FO were analysed and compared with the properties of fresh Mao juice. All juice samples were analysed at 25 ± 2°C with respect to their properties. Water activity (a_w) and pH were measured using a_w analyser (PreAQUA LAB) and pH meter (Hanna, edge pH), respectively. A transmission colorimeter (HunterLab, Vista) was used to perform colour analysis. It is classified by CIE (Comission Inter-148 nationale l'Eclairage) into three dimensions; L* (brightness), a* (reddish to greenish), and b* (yellowish to blueish). The total colour difference (ΔE) and chroma were analysed from L*, a*, and b* values using Eqs. 6 and 7 (Diamante *et al.,* 2010):

$$\Delta E = [(L_i^* - L_f^*)^2 + (a_i^* - a_f^*)^2 + (b_i^* - b_f^*)^2]^{1/2} \quad \text{(Eq. 6)}$$

where, L_i^*, a_i^* and b_i^*= initial color values of fresh juice, and L_f^*, a_f^* and b_f^*= final color of concentrated juice.

$$\text{Chroma} = (a^{*2} + b^{*2})^{1/2} \quad \text{(Eq. 7)}$$

where, a^* and b^* = colour values of juice.

In addition, Mao juice samples were collected at different concentrations (12, 15, 20, 25, 30, and 35 °Brix) during FO for the analysis of phytochemical content. Total anthocyanin content was measured by the pH-different method as described by Wang and Xu (2007). Potassium chloride (0.025 M, pH 1) and sodium acetate (0.4 M, pH 4.5) were utilised as buffer in the system. Mao juice was brought into buffer at pH 1.0 and pH 4.5, and was allowed to equilibrate for 30 min. The absorbance of each equilibrated solution was measured at a wavelength of 514 nm (λ_{max}) and 700 nm by spectrophotometer (GENESYS 10S UV-VIS) at room temperature (25 ± 2°C). The total monomeric anthocyanin was then calculated as cyanidin-3-glucoside using Eq. 8:

$$\text{Anthocyanin (mg/l)} = A \times MW \times DF \times 1000/(\varepsilon \times L) \quad \text{(Eq. 8)}$$

where, $A = (A_{514} - A_{700})pH_{1.0} - (A_{514} - A_{700})pH_{4.5}$, MW = molecular weight of cyanidin-3-glucoside (449.2 $g.mol^{-1}$), DF = dilution factor, L = pathlength of absorbance detection in cm, ε = molar extinction coefficient of cyanidin-3-glucoside (26,900 $L.mol^{-1}.cm^{-1}$), and 1000 = conversion factor from g to mg.

The total phenolic and ascorbic acid were determined using the same approach as reported in the work of Sripakdee *et al.* (2015). The total phenolic content was determined using the Folin-Ciocalteu reagent. The diluted phenolic (0.5 mL) was transferred into a test tube, and mixed with 2.5 mL of 10% ($v.v^{-1}$) Folin-Ciocalteu reagent. After mixing for 3 min, 0.5 mL of saturated sodium carbonate (7%, w/v) was added, and the sample was allowed to equilibrate for 120 min. Then, the absorbance was measured at 765 nm using a spectrophotometer (GENESYS 10S UV-VIS). The measurement was compared with a standard curve which was prepared by gallic acid solution. The total phenolic content was then expressed as micrograms of gallic acid equivalent (GAE) per mL of juice.

The total ascorbic acid content was determined using 2,6-dichlorophenolindophenol reagent. Mao juice was diluted by distilled water and transferred into a test tube followed by a mixing with 9 mL of 2,6-dichlorophenolindophenol. The absorbance of mixed solution was measured at 518 nm using a spectrophotometer (GENESYS 10S UV-VIS) within 30 s. The measurement was then compared with the L-ascorbic standard curve, and expressed as mg of L-ascorbic per mL of juice.

Statistical analysis

All measurements were performed in triplicate. The data were then subjected to analysis of variance, and appropriate mean separation was conducted using Duncan's multiple-range test. The data are presented as mean ± standard deviation (SD) of three observations.

Results and discussion

Effect of membrane vibration on water flux and reverse salt flux during FO of water

In the present work, FO was operated under the FO mode (feed solution face to the selective layer and draw face to support layer). It is worthy to note

that since distilled water was used as the feed solution, the ECP on the feed side can be neglected. In this case, we can assume that the changes in water flux due to vibration were solely due to ICP occurrences in the support layer structure. Figure 1A illustrates the water flux, and Figure 1B the reverse solute flux of FO operated at various vibrating amplitudes. The results demonstrated that the water flux was dependent on vibrating amplitudes. The highest water flux ratio of 1.24 corresponding to the improvement of 23% achieved at a vibrating amplitude of 0.6 mm. However, further increase in membrane vibration amplitude (1.2 and 1.8 mm) did not increase the water flux. The vibration or motion of membrane at certain amplitudes (at 0.6 mm in this case) possibly led to an increase in homogenisation of draw solution and reduced the impact of dilutive ICP adjacent to the selective layer, thus resulting in a decrease in ICP. Vibrating membrane at an amplitude higher than 0.6 mm possibly led to affecting the membrane support layer structure, thus leading to an increase in solute diffusion resistance. However, intensive investigation for future work is required. Higher water flux improvement using membrane vibration during FO of water when draw solution facing to active layer and feed solution facing to supported layer (PRO mode) was reported elsewhere (Yuan and Zhang, 2013). These authors concluded that the improvement was due to the effect of the dilutive ECP. The water flux tended to increase with increasing vibration frequency. However, the effectiveness of vibration frequency on water flux enhancement reduced when higher frequencies (> 40 Hz) were applied. The authors also suggested that the thickness of the liquid layer at the membrane surface resulted in a lower water flux. It is important to note that vibration amplitude as found in the present work as well as applied frequency as reported by Yuan and Zhang (2013) play an important role in controlling either ICP or ECP, depending on mode of operation (FO or PRO). The present work also showed that the use of membrane vibration led to an increase in reverse solute flux as compared to un-vibrating condition as shown in Figure 1B. This was due to an increase in solute concentration at the selective layer facing to support layer, as a result of a decrease in ICP as discussed earlier, leading to an increased concentration gradient across the selective layer membrane.

Effect of membrane vibration on water flux during FO of Mao juice

Unlike water, Mao juice contains a range of soluble and non-soluble components potentially creating ECP and fouling during FO. The properties of fresh Mao juice used in the present work are shown in Table 1. It is worthy to note that properties of Mao juice vary depending on several factors such as season, ripening stage, and extraction method. The initial TSS of fresh extracted Mao juice was approximately 12 °Brix. The water flux during Mao juice concentration using FO with varying membrane vibrating amplitudes and the final TSS of concentrated Mao juice were observed, and the results are shown in Figure 2. The water flux decreased and TSS content increased with processing time. In addition, the average water flux is also presented in Figure 1. Generally, the average water flux of the second stage was lower than that of the first stage. The result also revealed that vibration of the membrane at 0.6 mm amplitude resulted in the highest water flux ratio for both stages, where 1.6 corresponded to the water flux improvement of 70%. The water flux reduction during FO was attributed to several factors including ICP in the support layer, and ECP on the feed side, as well as membrane fouling (Li *et al.,* 2012). The water flux during the FO of Mao juice was much lower than that of distilled water, since ECP was absent at the feed side during the FO of water. In contrast, the ECP developed rapidly during the FO of juice, and continued to increase with processing time due to an increase in TSS. As a result, the osmotic pressure difference across the selective membrane decreased which led to lower water flux. In addition, the membrane fouling developed was also possibly responsible for flux reduction during the FO of Mao juice. The membrane fouling during the membrane processing of fruit juice is generally the result of the deposition of polysaccharides cell-wall composition such as pectin, cellulose, lignin, and hemicellulose on the membrane surface, and the consolidation of fouling layer (Domingues *et al.,* 2014; Kim *et al.,* 2019).

Table 1. Physical and chemical properties of fresh juice and concentrated Mao juice.

Properties	Fresh Mao juice (12 ± 0.5 °Brix)	FO-Mao juice* (35 ± 0.5 °Brix)
pH	3.3 ± 0.1[a]	3.3 ± 0.1[a]
a_w	0.998 ± 0.001[a]	0.957 ± 0.001[b]
L*	8.96 ± 0.01[a]	0.41 ± 0.01[b]
a*	39.05 ± 0.01[a]	2.74 ± 0.01[b]
b*	14.90 ± 0.06[a]	0.68 ± 0.01[b]
ΔE (colour difference)	-	39.82 ± 0.01
Chroma	41.79 ± 0.03[a]	2.82 ± 0.10[b]

Values are mean ± SD of triplicates (n = 3), and values in rows with different superscript letters are significantly different ($p < 0.05$).

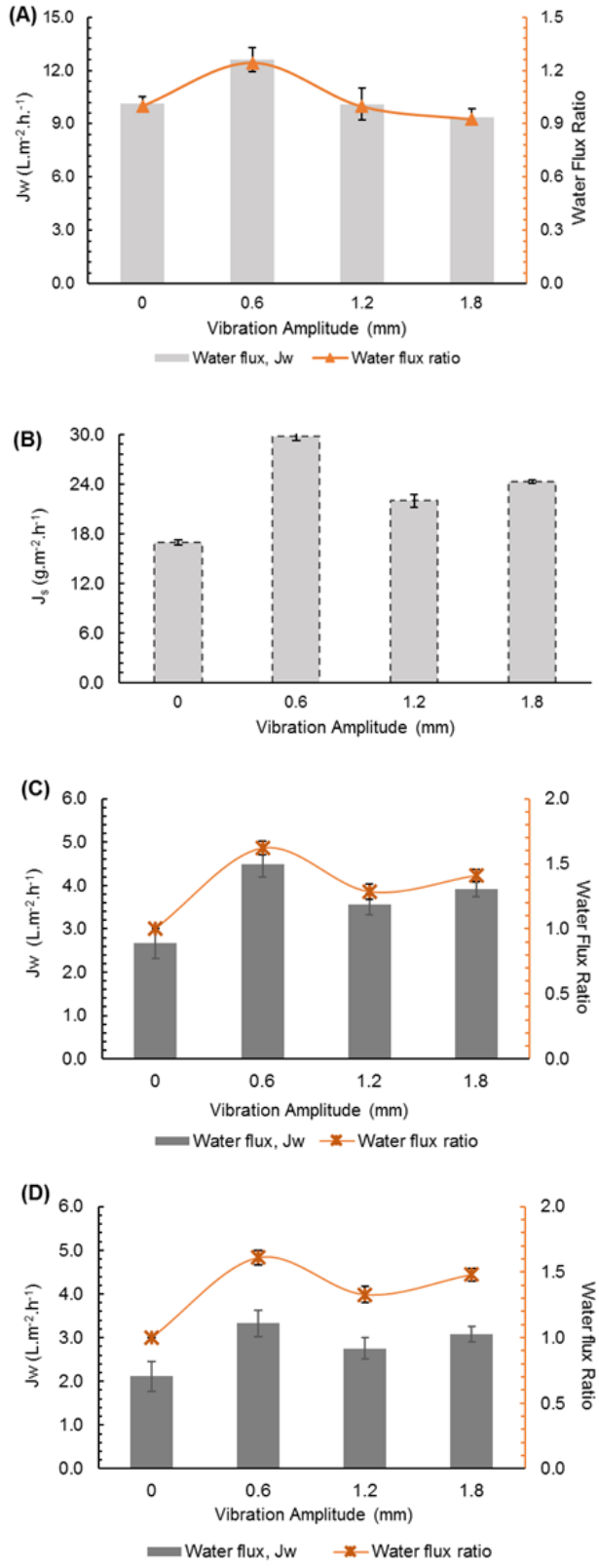

Figure 1. (A) Average water flux (J_w) and (B) reverse solute flux (Js) during the FO of distilled water, average water flux, and water flux ratio during the FO of Mao juice, at varying amplitudes of membrane vibration and constant frequency of 50 Hz; and total soluble solid content of Mao juice at (C) initial TSS = 12 °Brix for stage I, and (D) initial TTS = 20 °Brix for stage II.

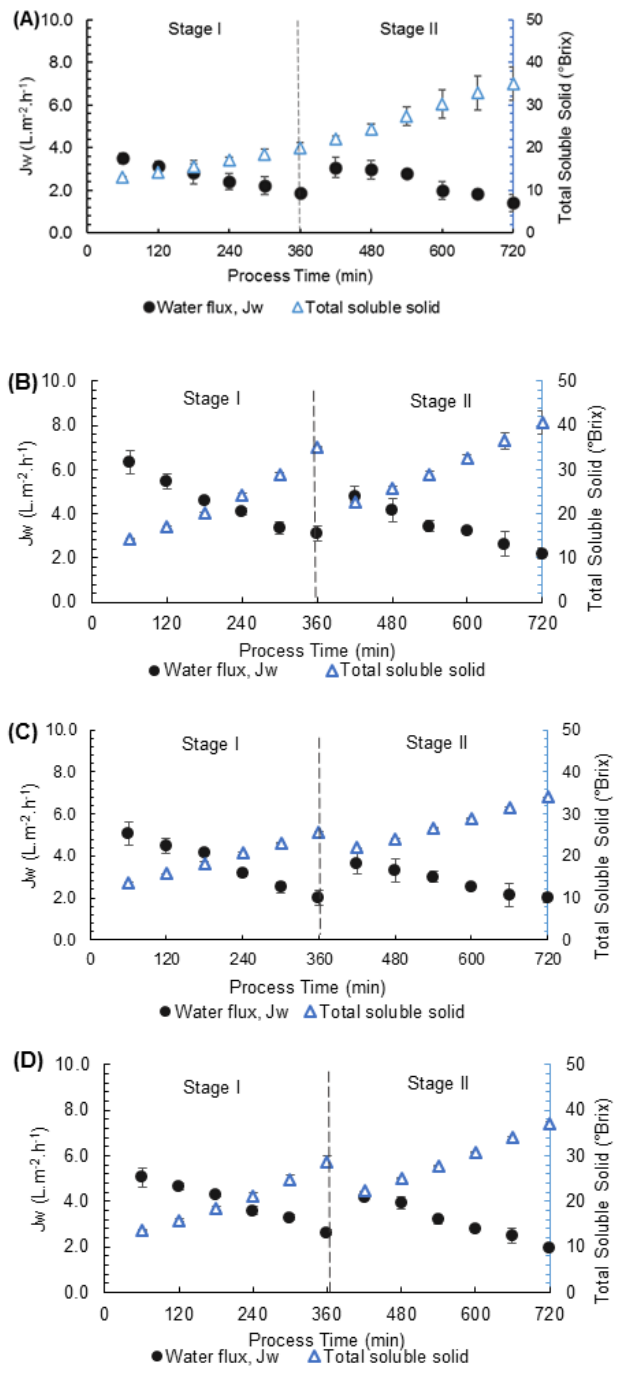

Figure 2. Water flux during the FO of Mao juice at varying amplitudes of membrane vibration for (A) Control, (B) 0.6 mm, (C) 1.2 mm, and (D) 1.8 mm, at a constant frequency of 50 Hz, and total soluble solid content of Mao juice with initial TSS = 12 °Brix for stage I and initial TTS = 20 °Brix for stage II.

Effect of membrane vibration on water flux recovery during the forward osmosis of Mao juice

It was hypothesised that membrane vibration could reduce membrane fouling during the FO of Mao juice. In FO, the impact of membrane fouling and cleaning is reflected through the water flux of fouled membrane. The percentage of water flux

recovery after hydrodynamic washing of fouled membrane is presented in Figure 3. It was discovered that the water flux recovery in FO was much higher as compared to a pressure driven membrane process such as RO (Chun *et al.,* 2017). It is important to note that the water flux recovery of the FO membrane fouled under vibrating conditions was higher as compared to membrane fouled under un-vibrating condition. The highest water flux recovery was found at a vibration amplitude of 0.6 mm. The water flux was recovered up to 95 and 75%, in the first and second stages of Mao juice concentration, respectively. Lower water flux restore level observed in the second stage of concentration was possibly due to an increase in fouling resistance as feed concentration increased (Sioutopoulos *et al.,* 2019). These results also suggested that membrane vibration could partially reduce membrane fouling generated during FO of Mao juice. As reported in the previous studies, Mi and Elimelech (2010) and Lee *et al.* (2010) found that most FO fouling was reversible, and could be removed by hydraulic shear stress, e.g., water flushing. The severity and reversibility of membrane fouling varies depending on several parameters, e.g., feed property, membrane property, and operating condition. In fruit juice application, membrane fouling is generally caused by polysaccharides, the hydrophilic component in fruit juice. This component presents a tendency absorption on hydrophilic active layer of membrane (Chun *et al.,* 2017). The results of the present work suggested that the FO membrane fouling due to the components present in Mao juice could not be completely removed. Therefore, proper cleaning methods such as osmotic backwashing, oxidant, acid, and surfactant cleaning should be carried out.

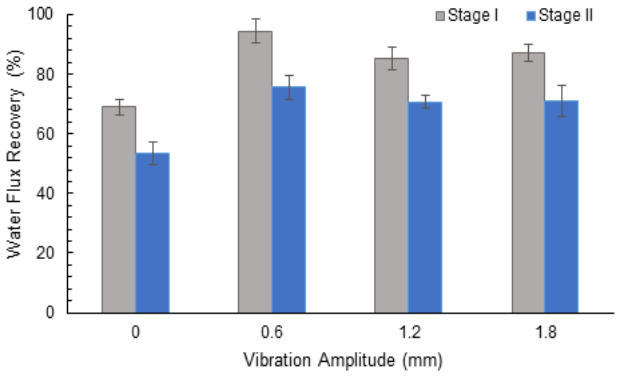

Figure 3. Water flux recovery after hydrodynamic washing of membrane fouled during the FO of Mao juice at different amplitudes of membrane vibration, at a constant frequency of 50 Hz, and initial TSS = 12 °Brix for stage I and initial TTS = 20 °Brix for stage II.

Water flux reduction analysis

The significant effect of ICP, ECP, and fouling on water flux reduction during FO of distilled water as well as Mao juice were analysed. The theoretical water flux of distilled water calculated with the assumption of the absent of ICP, ECP, and fouling using NaCl 4 M as draw solution, was 88.7 $L.m^{-2}.h^{-1}$. This value then was used to analyse the significant effect of flux reduction during FO as presented with the term FR. In FO of distilled water, the water flux was much lower than the theoretical water flux because of ICP effect, as described in the previous section. For instance, the water flux of distilled water of FO without and with membrane vibration at amplitude of 0.6, 1.2, and 1.8 were 11.4, 14.2, 11.4, and 10.6% of the theoretical water flux, corresponding to the FR of 88.6, 85.8, 88.6, and 89.4%, respectively. The result suggested that the ICP would also play a major role in flux reduction during FO of Mao juice. In addition to ICP, ECP and fouling were generated during FO of Mao juice leading to further reduction of the water flux. Please note that the FR due to ICP during FO of Mao juice were assumed to be the same as those of FO of distilled water, since the same operating conditions e.g., vibrating amplitude, draw solution concentration were employed. Generally, the FR due to ECP and fouling were much lower than those of FR due to ICP, and the FR of ECP were much higher than those of fouling. For instance, the FR of the first stage (membrane vibration at amplitude of 0.6 mm) due to ICP was 85.8%, while FR due to ECP and fouling were only 13.4 and 0.8, respectively. In addition, the FR due to fouling of stage I were lower than those of stage II, suggesting that less fouling developed during FO of lower feed concentration. In addition, higher FR due to ECP was found at stage I as compared to stage 2. Higher FR due to ECP effect was observed using membrane vibration at 0.6 mm as compared to other conditions, possibly due to higher water flux resulting in the greater convective water flow drag solute from the bulk solution to the surface of membrane active layer (Geraldes *et al.,* 2001). From these results, it can be concluded that ICP was the main factor responsible for water flux reduction during FO of Mao juice, followed by ECP and fouling, respectively.

The results of this primary study suggested that vibration of membrane is a promising technique in enhancing water flux during FO of Mao juice. Regarding the similarity of their properties, this technique could be possibly applied for the concentration of other juices or beverages. To prevent breakage, a commercial-sized scale of

vibrating FO system should be carefully designed similarly to those UF and MF systems, which are nowadays available in the membrane market (Sriniworn *et al.,* 2016).

Chemical and physical properties of Mao juice

Change in total phenolic compound, anthocyanin, and ascorbic acid during FO

Total phenolic compound, anthocyanin, and ascorbic acid are major bioactive compounds found in Mao juice. Therefore, the capability of FO in the concentration and preservation of these components are of great interest. The FO process was carried out using two stages, and each stage was run continuously for approximately 6 h. The degradation profiles of these selective components as indicated by the percentage of normalised phytochemical content (C/C_i) are shown in Figure 4. Generally, the degradation of selective phytochemical compounds during FO was more significant in the first stage of concentration (12 to 20 °Brix), and less degradation was observed afterward.

Total phenolic content increased with processing time or TSS content. The total phenolic content rapidly degraded during concentrations of up to 15 °Brix, and the degradation slightly decreased afterward. At the end of the FO concentration (35 °Brix), 82.3% total phenolic compound was preserved. Since no heat treatment was applied during fresh Mao juice preparation prior to FO, polyphenol oxidase usually found in fruit was expected to present in the juice. As a result, the degradation of total phenolic during FO was possibly due to an enzymatic oxidation. The thermal stability of phenolic compounds found in Mao juice has been previously studied (Sripakdee *et al.,* 2015). Phenolic compounds in Mao juice were preserved approximately at 90.3, 88.9, and 85.7% after 60 min of heating at 60, 80, and 100°C, respectively. If evaporation was operated at a 100°C for 1 h to achieve the final product with a TTS of 35 °Brix, FO possibly did not spoil the preservation of total phenolic compound as compared to thermal concentration process.

Anthocyanins are bioactive compounds with desirable properties. Anthocyanins found in the Mao fruit are cyanidin, malvidin, pelargonidin, and delphinidin (Jorjong *et al.,* 2015). These compounds are responsible for colour as well as for the antioxidant properties of the fruit. Unfortunately, anthocyanins are not stable and tend to degrade easily during food processing and storage. Some parameters that could affect its stability are pH, oxygen, enzyme, ascorbic acid, and heat treatment

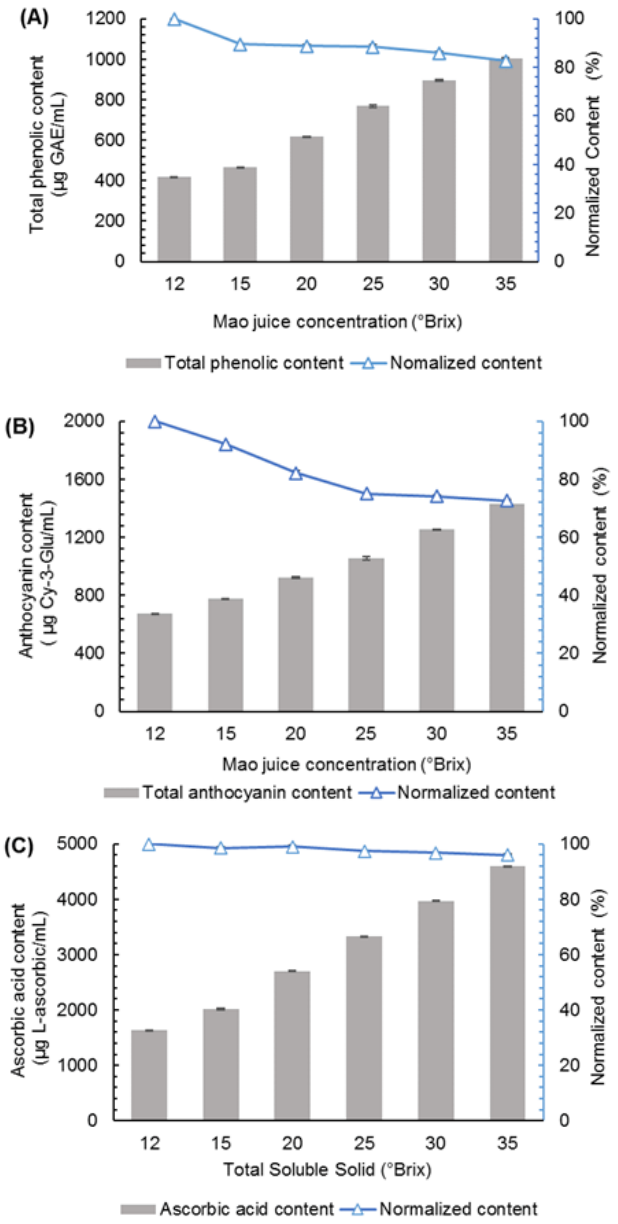

Figure 4. The phytochemical and normalised content (C/C_i) (%) of total phenolic compounds (A), anthocyanin (B), and ascorbic acid (C) during the FO of Mao juice, at a constant vibration amplitude of 0.6 mm and 50 Hz. The C_i is the initial phytochemical content, and C is the phytochemical content of the concentrated juice.

(Wang and Xu, 2007; Mohideen *et al.,* 2015). For instance, the half-life of anthocyanin found in blackberry juice heating at 60, 70, 80, and 90°C were 16.7, 58.8, 4.7, and 2.9 h, respectively (Wang and Xu, 2007). In FO, the anthocyanin degraded as TSS or processing time increased. At the end of FO (35 °Brix), 72.6% anthocyanin was preserved. The degradation of anthocyanin during the FO of Mao juice was possibly due to an increase in dissolved oxygen during the circulation of the juice stream, and the presence of enzymes leading to an enhanced

oxidation reaction (Persic *et al.,* 2017; Weber and Larsen, 2017).

Ascorbic acid is one of the major reducing agents of the essential phytonutrients which is essential for living cells, and could act as the main contributor to antioxidants in vegetable and fruit (Sripakdee *et al.,* 2015). Unfortunately, it is the most sensitive among vitamins. It can be destroyed under oxygen, light, and heat exposure. However, ascorbic acid is more stable in acidic environments (pH < 6) such as in lemons, oranges, and grape juice (Belajová *et al.,* 2017). In the present work, ascorbic acid degraded slightly with increasing processing times or TSS content during the FO of Mao juice, as shown in Figure 4. The degradation of ascorbic acid was less as compared to the total phenolic and anthocyanin. The FO could preserve at least 90% of ascorbic acid in Mao juice. This could possibly be due to the juice characteristic (pH = 3.3) and low temperatures (ambient temperature) during FO of Mao juice.

Regarding thermal and non-thermal concentration, a number of factors are found to affect degradation of bioactive compounds in fruit juices. For thermal concentration processes, vacuum evaporation at low temperatures is usually employed to reduce its negative impact on the degradation, as well as for thermal efficiency of the process. In non-thermal FO processes, operating conditions that reduce possible oxidation should be avoided e.g., oxygen and enzyme. Increasing permeate flux and membrane area are preferable to reduce concentration time and the negative impact on phytochemical degradation.

Colour, pH, and water activity of fresh Mao juice and Mao juice concentrate

Colour, pH, and a_w of fresh Mao juice and Mao juice concentrate were studied, and the results are summarised in Table 1. Following concentration, removal of water leads to an increase in TSS content. As expected, a_w of concentrated juice prepared by FO and evaporation were lower as compared to fresh juice, while the pH of fresh and concentrated juice did not show any difference. Fresh and concentrated Mao juice were different in the visual component of the juice as indicated by different Hunter parameters, ΔE, and Chroma. Generally, L*, a*, and b* of fresh juice were much higher than those of concentrated juice. This could be due to a higher concentration of darkening components in the concentrate. The ΔE (colour difference) is a combination of change of three components, i.e., L*, a*, and b* value. The results indicated the magnitude of colour difference between fresh and concentrated juice. This result is similar to that of sterilised mulberry juice (You *et al.,* 2018). Chroma value is generally used to indicate the vivid colour of the juice in which higher Chroma represents higher levels of colour. A significantly lower Chroma value of the FO was observed as compared to fresh Mao juice. The less vivid colour of concentrated juice was due to a lower b* parameter (blueish), which is similar to those found in blueberry juice samples (You *et al.,* 2018).

Conclusion

The effect of membrane vibration on the water flux of TFC membrane during the FO of distilled water as well as Mao juice were studied. In addition, the phytochemical contents of Mao juice during the FO and physical properties of the concentrated were also analysed. The following are the highlights of the research findings. Membrane vibration was an effective technique to enhance water flux during FO in comparison to the membrane without vibration integration. In the FO of distilled water, membrane vibration could enhance water flux due to the reduction of ICP, and vibration at 0.6 mm amplitude resulted in the highest water flux water enhancement (25%) as compared to un-vibration condition. In the FO of Mao juice, membrane vibration at 0.6 mm amplitude enhanced water flux up to 70%. The ICP, ECP, and fouling were responsible for flux reduction during FO of Mao juice. However, the ICP was the main factor influencing the water flux reduction during the FO of Mao juice. Although vibration is a promising technique to enhance membrane permeate flux, its combination with other parameters should be further studied. In addition, intensive study of membrane fouling and support layer structure under vibration is required for future work. Concentration of Mao juice (35 °Brix) by FO could preserve the total phenolic compounds, anthocyanin, and ascorbic acid up to 82.7, 72.6, and 95.9%, respectively. However, raw juice pre-treatment for enzymatic inactivation should be further studied to reduce its impact on bioactive degradation.

Acknowledgement

The present work was financially supported by the Thailand Research Fund under the Royal Golden Jubilee PhD Program (PHD/0190/2557) and Graduate School, Prince of Songkla University, Thailand.

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