Effects of draw solution and operating factors on forward osmosis for lime juice concentration

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Abstract Forward osmosis (FO) is a process that concentrates liquid food by slow water-transport which requires lower pressure, and the product from this process has similar quality to natural one. This study aimed to find suitable operating factors in FO process including temperature (10 - 30°C), circulation flow rate (200 – 350 ml/min), and concentration of NaCl (1 – 4M) for concentrating lime juice. By using commercial flat-sheet Reverse Osmosis membranes installed in a 3×8×12 cm membrane module with a 45 cm2 of cross-section area at the feed solution side, in order to study the effect of differences draw solution temperatures (10 to 30°C) on water flux value in a laboratory-scale. The result of the study found that using lower-temperature of draw solution achieves lower water flux value because of increasing viscosity values that also decreasing water flux. Moreover, increasing the circulation flow of draw solution is also increasing water flux and reducing membrane fouling during the process. Therefore, the suitable condition of Forward Osmosis is using 3M NaCl as Draw solution at 25°C, Synthetic lime at 20°C and operated with the circulation flow of draw solution side as 350 ml/min which result in a maximum average water flux is 2.33 l/m^2h at 90 minutes of operating time.

Keywords: Forward Osmosis, Lime Concentration, Temperature effect, Water Flux, Circulation flowrate

Introduction

Lime is one of the citrus plants that are important for both household and industrial economies in Thailand and are used as an ingredient in various foods and beverages. During drought or summer season often lacking lime period that was the lowest products and poorest quality yields of lime. Therefore, this issue was solved by preservation in lime concentrate form to facilitate storage, transportation, and distribution of the product as an economical operation. However, the thermal processes commonly applied for concentrating juice products, such as evaporation, have been reported to have detrimental impacts on the negative sensory and nutritional values of food products (Jesus *et al.,* 2007; Sant'Anna *et al.*, 2012).

Therefore, Forward osmosis (FO) process should be applied in the lime juice concentration process by this process archives without negative impacts on the sensory or nutritional values of juice products. In addition, FO process used more energy efficient as a concentration process than thermal evaporation and pressure-driven membrane processes, (Chekli *et al*., 2016). FO or another name. "Direction osmosis" is a diffusion phenomenon of water or solute through a membrane. (Semipermeable Membrane) with the Concentration Gradient. these is water diffuse from a low concentration solution called a "feed solution" to a high concentration solution called "Draw Solution" by Osmotic Pressure (Nagy, 2018). The water diffusion in this phenomenon was slow or fast depending on the osmotic pressure and applied external factors for improving process which has related physical and chemical factors such as pressure, temperature, viscosity, and the concentration of feed and draw solution within the forward osmosis process (Kim *et al.*, 2019), etc.

In this research, we investigated the possible improvement for lime juice concentration by FO process with the temperature-driven. This research focused on the physical factors of the draw solution that affecting the efficiency of the FO process in lab-scale which are temperature, circulation flow rate, and concentration. By using commercial flat-sheet Reverse Osmosis membranes because of these factors were directly related to osmotic pressure that achieved accelerating the diffusion of water in feed solution side to draw solution side and reducing the time of lime juice concentration with FO process. Lastly, there result also suggested improvements of FO process for lime juice concentration.

Materials and methods

Raw Material

Synthetic Lime solution was mixed with organic compounds in Lime juice refer to the most proportion of the organic chemicals in lime juice table (Tressler and Joslyn, 1961) that include AR Grade of Citric Acid Anhydrous (5.97 g./L.), Ascorbic Acid (0.03 g./L.) and Sucrose (3.2 g./L.) using as feed solution. the initial feed solution has the ph of feed solution equal 2.25±1 and the acidity of feed solution equal 0.02 g/100ml.. The draw solution was used 1, 2, 3 and 4M NaCl.

Forward osmosis module

The Forward osmosis experiments were proceeded with a polyamide thin-film composite RO membrane. The experiments were set using a 90 cm^2 surface area membrane with two sides in the 3x8x12 cm module as show in Figure 1. 6 L. container of NaCl as draw solution were fed by pass about 200 mL. of solution to the module by peristatic pump. A 3L. container of Synthetic Lime as feed solution were put in the container where the module had been set up inside

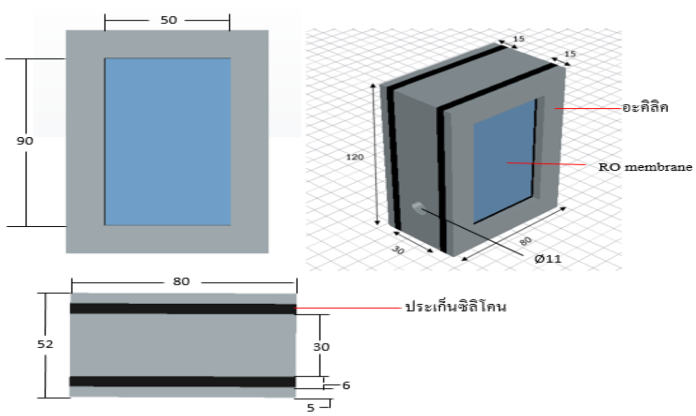

Figure 1. Forward osmosis module in experiments (unit; Milimeter)

Forward osmosis experiments

Forward osmosis experiments were carries out in three experiments. First, the effect of draw solution concentration on FO at 1,2,3 and 4M NaCl as draw solutions by fixed circulation flow rate at 300 ml/min for operating. Second, the effect of circulation flow rate on draw solution with 3M NaCl as draw solutions. Third, the effect of differential temperature on feed and draw solutions. The both temperature in first and second experiments were maintained in room temperature at around 30±1°C. the third experiment was separated the temperature control of feed and draw solutions which set up feed solution at 10 and 20 °C and draw solution at 10, 15, 20 and 25 °C, respectively.

Water Flux

Water flux (Nayak C. *et al.*, 2011) was calculated according to the following equation:

$$J = \frac{V_2 - V_1}{A \times t}. \qquad (1)$$

in which J is water flux. V_1 and V_2 are the draw solution volume at initial and monitoring times, respectively. A is the membrane surface area and t is the osmosis times (operating time).

Total Acidity

The total Acidity as Citric acid was calculated following to (A.O.A.C., 2012) method.

pH Brix Viscosity and salinity

The pH and salinity of sample was measured by pH meter; AMT03 AMTEST, Amtrast USA Inc., UAS. 2.7. Brix analysis
The °Brix of feed solution were measured by Brix refractometer; MR32ATC, Milwaukee, Massachusetts, USA. The viscosity of feed and draw solutions were measured by Viscometer; RV DV III, AMETEK Brookfield, Massachusetts, USA

Results

Effect of draw solution concentrations

The experiment 1 were resulted show in Figure 3. that 3M NaCl got higher water flux than other concentrations of NaCl solutions as Draw solution in equal temperature condition. The maximum average of water flux in 1, 2, 3 and 4 M NaCl at 1 hours of operating were 251.34 ,466.07 ,707.15 and 97.30 mL/m^{2}h, respectively. But the maximum water flux of 4 M NaCl was 114.31 mL/m^{2}h at 30 minutes of operating time and then the water flux decreasing to 0 mL/m^{2}h after 120 minutes of operating time but were found to continue to undergo forward osmosis, but 2M and 3M NaCl the water flux decreased until the forward osmosis was terminated at a time of 180 min in 1M 2M and 4M NaCl solution exception of 3M NaCl could be perform forward osmosis even after 3 hours of systemic operation.

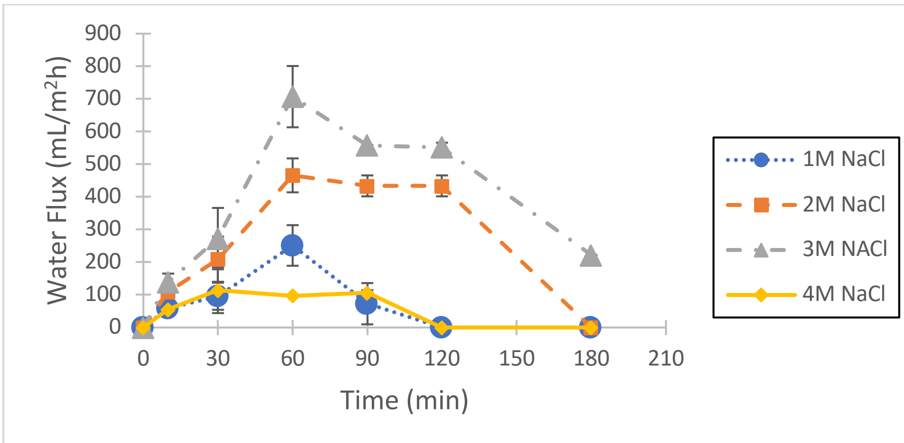

Figure 2. Water flux of concentrations of NaCl solution in 1, 2, 3 and 4 M as draw solution using a 300 ml / min. as draw solution flowrate

Effect of draw solution circulation flow

In Experiment 2, the results showed as Figure 4. that the effect of various flowrate of draw solution applied in FO was found the water flux in low to high flowrate were 456.93, 467.70, 707.15, and 903.09 mL/m^{2}h at 60 minutes that applied flowrate as 200, 250, 300 and 350 mL /min., respectively. According to the results of the experiment that increasing the flowrate with suction flow assist increases the water diffuse from the feed solution to draw solution.

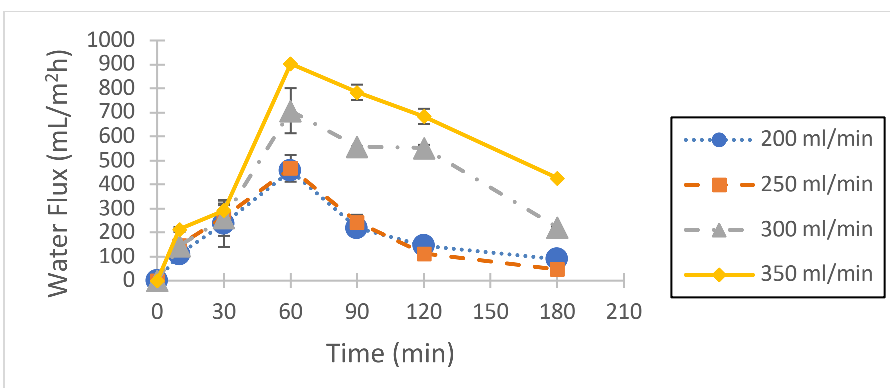

Figure 3. Water flux of 3M NaCl as draw solution with 200, 250, 300 and 350 ml./min as draw solution flowrate

Effect of draw solution and feed solution temperature

Before start the experiment 3, the viscosity of the draw and feed solutions was analyzed. Synthetic lime juice and NaCl solution at temperatures 10, 15, 20 and 25°C with the Brookfield viscometer was shown in Table 1. It was found that the viscosity of both solutions increased when the solution was at low temperature, corresponding to the viscosity theory of the solution Which describes the relationship between temperature and the viscosity of the solution is inversely proportional to the viscosity is the anti-flow force of the solution Therefore, high temperature operation results in better FO performance than low temperature operation (She *et al.*, 2016; Tang *et al.*, 2010).

Table 1. Viscosity of feed and draw solution at 10, 15, 20 and 25°C

Sample	Temperature (°C)	Viscosity (cP)	Shear stress (N/m^2)
Synthetic Lime (Feed)	25	1.92	4.70
	20	2.08	5.09
	15	2.24	5.48
	10	2.34	5.71
3M NaCl (Draw)	25	2.34	5.71
	20	2.46	6.03
	15	2.62	6.42
	10	2.78	6.81

The results of the operating of the synthetic lime juice at 10°C with NaCl solution at 10, 15, 20 and 25° C that shown as Figure 5. were found NaCl at 10 and 15 °C had the highest average water flux over 30 minutes of operating, where the maximum average water flux of the solution. NaCl at 10 and 15°C were 1,227.78 and 1,555.56 mL / m^{2}h, respectively. The FO characteristic of this first part of the system was a high-water flux from 10 to 30 minutes after 30 minutes of operation. System flux will be gradually reduced to complete a full 180 minutes of cycle. The system operation with solution NaCl at 20 and 25°C had the highest average water flux over 90 minutes of operating, where the highest mean water flux of NaCl solution at 20 and 25°C was 1,777.78 and 2,500.00 mL / m^{2}h in respect of These two FO system characteristics are that during 10-90 minutes the water flux will gradually increase up to 90 minutes, after which the water flux decreases so rapidly to the water flux is zero for the full 180 minutes of cycle.

The results of the operating experiment by controlling the temperature of synthetic lime juice at 20°C with NaCl solution at 10, 15, 20 and 25°C. NaCl at 10°C has the highest average water flux over 30 minutes of operating. The maximum average water flux of the solution. NaCl at 10 °C is equal to 1,518.52 mL / m^{2}h, the FO formation characteristic of this part of the system is similar to the FO formation characteristic of the experiment. Synthetic lime juice was obtained at 10 °C with NaCl solution at temperatures 10 and 15 °C before the part in the solution operation. NaCl at 15 and 20 ° C had the highest average water flux over 60 minutes of operation, where the maximum average water flux of the solution. NaCl at 15 and 20 °C were 1,777.78 and 1.833.33

mL / m^2h respectively. The FO characteristic of this system is that the water flux will increase gradually up to 60 minutes. A total of 180 minutes was completed. NaCl at 25 ° C has the highest average water flux over 90 minutes of operation. NaCl at 25 ° C is equal to 2,333.33 mL / m^2h, the FO-inducing behavior of the system in this section is similar to the FO-inducing behavior of the experiment, the temperature of synthetic lime juice was given to 20°C with NaCl solution at 15 and 20°C, but the water flux will increase up to 90 minutes of operating. After that, the water flux of the system will gradually decrease until the complete one cycle of operation.

The experiment was operating by FO system synthetic lime juice to 25°C with NaCl solution at 30°C. The maximum average water flux was within 90 minutes of operation where the maximum average water flux of the solution was obtained. NaCl at 25°C is 1,111.11 ml / m^2h, the FO characteristic of this part of the system is similar to the FO characteristic of experiment 2. Gradually decrease until the complete cycle of the system.

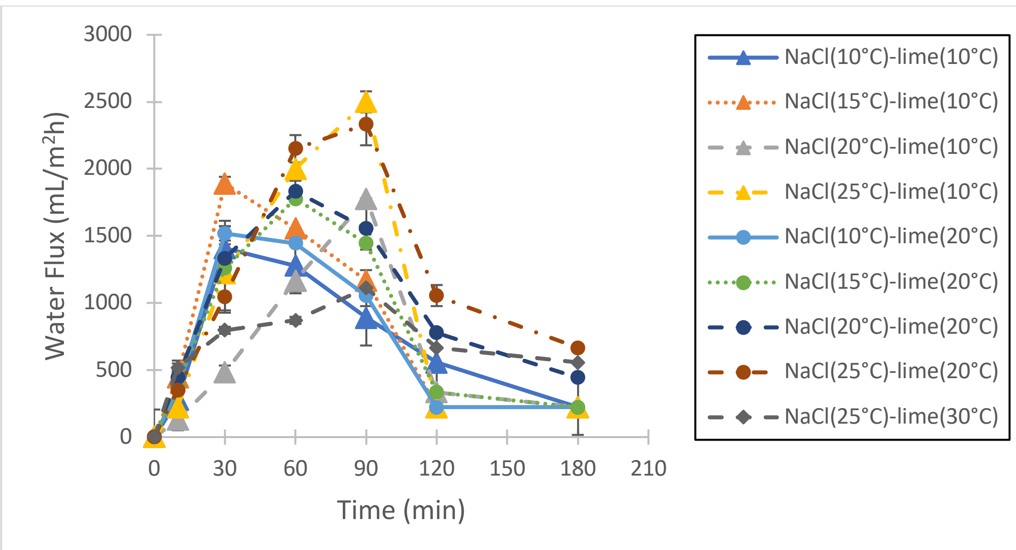

Figure 4. Water flux of 3M NaCl as draw solution at 10, 15, 20, 25 and 30°C and synthetic lime as feed solution at 10 and 20°C using 350 ml./min as draw solution flowrate (draw temp./feed temp.)

Feed Solution characteristics

In synthetic lime solution, the feed solution is slighly change chemical characterized that show it Table. 2 but the water in feed solution is changed by volume. From experiment results 1-3 the factor that gives the highest water flux value in experiments was found that the initial feed solution volume decreased was 3 liters after 3 hours of operation, it was reduced feed volume to 2.95 liters (3M NaCl was used as the pulling solution), 2.94 liters (the draw solution flow rate was 350 ml./min.) and 2.85 liters (controlled temperature of feed solution at 20 °C and draw solution at 25 °C) respectively. It is assumed that this FO system can partially removing water in synthetic lime juice without degradation. Thus, the FO system has the potential to be applied in fresh lime juice.

Table. 2. Acidity of feed solution before and after operating FO in the best conditions of experiments (Exp.)

Time (min)	Acidity of feed solution (g/100 ml.)		
	Exp. 1	Exp. 2	Exp. 3
0	0.02	0.02	0.02
180	0.02 ±0.01	0.03 ±0.01	0.03 ±0.01

Draw Solution characteristics

The draw solution characteristics that shown in Figure 5. demonstrated results in terms of the pH and salinity of the draw solution. In all experiments, there was a change in this value, it was found that as the operating time increased, the pH of the draw solution decreased. Moreover, the salinity of draw solution was found that when operating for a long time, the salinity decreased which is related to the pH of the draw solution.

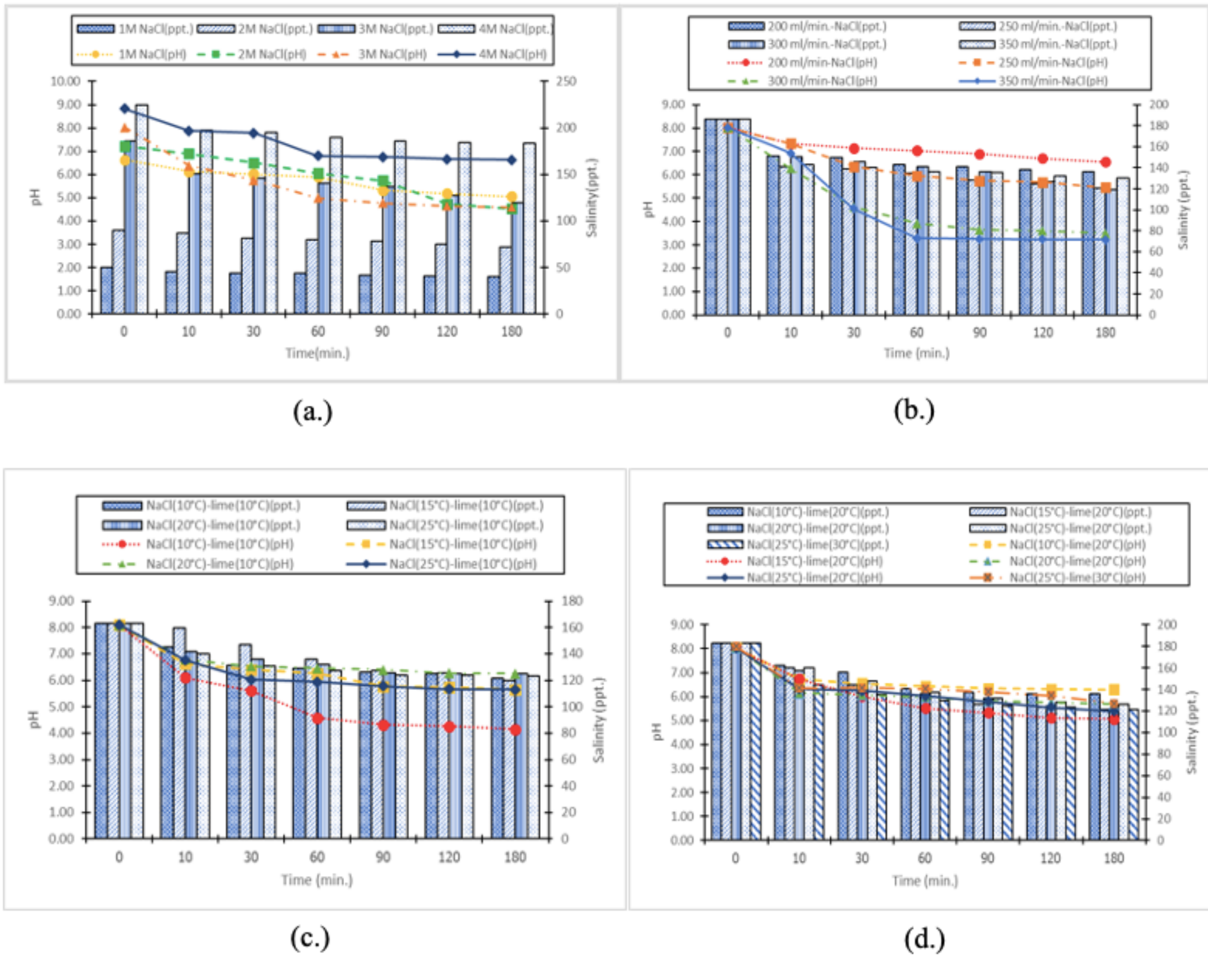

Figure 5. pH and Salinity of draw solution (a.) pH of draw solution experiment 1 (b.) pH of draw solution experiment 2 (c.) pH of draw solution in experiment 3 in synthetic lime at 10°C (d.) pH of draw solution in experiment 3 in synthetic lime at 20°C

Discussion

The results of first experiments were same as increasing beetroot water concentration by forward osmosis was found that the mean water flux in the system was 12.40 L / m^2h (Nayak *et al.*, 2011), which was less than that of research by Kim *et al.*, (2019) using NaCl solution. The 2M and 3M NaCl were draw solutions for increasing grapefruit concentrations by forward osmosis process. The average water flux values of forward osmosis process were 13.2 and 18.4 L / m^2h, respectively. Using a high-concentration draw solution will not always improve the efficiency of forward osmosis but also increases rapid fouling from the draw solution even though it gives a high flux value (Wang *et al.*,2016). The efficiency of the forward osmosis process depends on other factor such as the chemical composition, concentration and viscosity of the sample solution that also affect forward osmosis (Kim *et al.*, 2019; Nayak *et al.*, 2011). Therefore, the best condition in this experiment was using 3M NaCl that give a high-water flux as 707.15 mL/m^2h

The results of the second experiments were found that increasing the flowrate with suction flow assist increases the water diffuse from the feed solution to draw solution. Due to the high solution flowrate made the turbulent flow that creates a shear on the membrane surface, thereby reducing membrane fouling from Internal Concentration polarization in the membrane surface (Boo *et al.*, 2013). Moreover, the result was consistent with the experimental of Kim *et al.*, (2019), that increasing the flow rate of the draw solution improves the efficiency of the FO process.

The results of third experiments demonstrated that operating at high temperatures as well as solvents is the preferred method for significantly improving the efficiency of the FO system. In addition, this method reduces the viscosity of the two solutions, which is friction force and diffuses the particles in the solution, resulting in high temperature-controlled operation of both solutions with an average water flux. It was higher than the temperature-controlled system of the two solutions (Zhao *et al.*, 2012) because the highly viscous substance also had a high resistance to water diffusion. This occurs when the solution is in a low temperature state. Where temperatures have inverse viscosity relationships. That is why the system operation at low temperatures of both solutions is less efficient than the system that controls the high temperature of the solution.

The draw solution characteristics similar to the research of Saengrungnapaphan *et al.* (2019). Since Synthetic lime juice includes Citric acid and Ascorbic acid, it will break down in solutions that Proton (H^+) is smaller than water molecule during the FO process. H^+ particles pass through membrane pore with water molecules that results a rapid decrease in the pH of the draw solution. the research showed that the membrane material polyamine in this experiment allowed the H+ particles from the synthetic lime or feed solution side pass to the draw solution side (Saengrungnapaphan *et al.*, 2019).

Thus, the efficiency of the FO system in increasing the concentration of synthetic lime juice. It was found that the factors that increase the efficiency of the FO system were operation with increased flowrates, high temperatures and a suitable concentration of the draw solution. According to the high flow rate increases, it reduces membrane fouling due to shear force on the membrane surface that preventing accumulate some particle on the membrane surface. Less operating the system with a highly concentrated draw solution not only yields more water flux values over a period of time, but there will also be a chance of faster fouling in the system as a result of Concentration polarization on the membrane surface. The temperature of feed solution and the draw solution is one of factor that influences the efficiency of the FO system due to the inverse temperature relationship with the viscosity, which is the flow resistance of the solution. If operating the system with high solution temperature, it will reduce the viscosity of the solution and increase the efficiency of the FO system. Not only, the performance of the FO system depends on the factors of the draw solution but also the efficiency of the FO system depends the factors of the feed solution can also affect the performance of the FO system.

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