(This article was initially submitted to National Chemical Engineering Congress and reviewed by JOTCSB editorial staff).

Concentration of Skim Milk and Dairy Products by Forward Osmosis

Ayça Hasanoğlu[1]*, Kübra Gül[1]
[1]Yıldız Technical University, İstanbul, Turkey

Abstract: The concentration of liquid foods such as fruit-vegetable juices and milk by removing its water content is an important chemical process in terms of concentration of food nutrition, shelf-life and stability extension, decrease of the bacterial activities and cost reduction of transportation and storage. Milk can be consumed as concentrated milk by removing its water content at certain rates. Eliminating the water content in milk at certain proportions is also an essential step during several dairy product manufacture processes such as cheese, yogurt and milk powder production. In this study, milk concentration is carried out by means of forward osmosis using membrane contactors as an alternative process to conventional evaporation processes. In this process, milk is circulated through the shell side of the hydrophobic membrane contactor while a draw solution of $CaCl_2$ is circulated through the lumen side. Thus water is transferred from the milk solution to the brine solution because of the activity difference between these two solutions. In forward osmosis the effect of process parameters such as feed and draw solution rates, draw solution concentration, temperature on water fluxes was investigated and it was found that water can be removed efficiently and rapidly using this process. The concentration was carried out until the milk volume reduced in half and flux values at investigated process conditions were found to be in the range of 155-387 mL. h^{-1} m^{-2} .

Keywords: Forward osmosis; membrane contactor; milk concentration.

Submitted: September 30, 2016. **Revised:** November 21, 2016. **Accepted:** December 30, 2016.

Cite this: Hasanoğlu A, Gül K. Concentration of Skim Milk and Dairy Products by Forward Osmosis. JOTCSB. 2017;1(1):149–60.

*Corresponding author. E-mail: aycameric@yahoo.com.

INTRODUCTION

In order to provide microbiological and chemical stability, milk is generally concentrated using multi-stage evaporators. When the focus is to prepare concentrated milk to be consumed, the dry matter content in milk should be increased from 8-12% to 20-25% [1]. Since the volume is reduced, stock and transfer costs decrease in this way. On the other hand, in most dairy product production such as cheese, yogurt, and milk powder, milk is used as its concentrated form by removing its water content at certain rates. For example, in milk powder process milk is concentrated until 50% dry matter content before it is sent to spray dryer [2]. Ramirez *et al.* (2006) made an analysis of energy consumption and energy efficiency for the dairy industry in four European countries and indicated that milk concentration, together with drying, can be considered as the most energy intensive operations of the dairy industry [3]. In general, in the dairy industry, evaporation is mainly done in falling film evaporators employing multiple stage evaporators [3]. However, conventional thermal evaporation can result in a loss of aroma compounds and color and taste degradation in liquid foods due to the thermal effects [4]. Therefore, non-thermal alternative concentration techniques have been gaining importance recently. The use of membrane separation techniques in liquid food concentration have several advantages over conventional separation processes including improved product quality, easy scale-up and lower energy consumption [4]. In the literature, several liquid food concentration studies such as fruit juices using forward osmosis have been reported [4-7], however milk concentration was not encountered using this process. Although there are reported studies of milk processing with the use of membrane processes such as reverse osmosis and ultrafiltration [8, 9], there are few works in the literature focused on the use of membrane contactors for milk concentration. Hausmann *et al.* proposed membrane distillation which employs membrane contactors for milk and whey processing [10, 11]. In their study, water transfer through the membrane is achieved by a temperature difference provided on both sides of the hydrophobic membrane contactor. They studied skim milk and whey concentration by membrane distillation where the driving force is the temperature difference between two sides of the membrane. They concluded that the retention of dissolved solids was found to be close to %100, indicating the potential use of the membrane contactors in dairy product processing. On the other hand, several reports in literature indicate that that forward osmosis has several advantages over membrane distillation [12, 13]. In forward osmosis the driving force for the water transfer through the hydrophobic membrane contactor is provided by activity difference of water between two sides of the hydrophobic membrane contactor. Alves and Cohelso (2005) carried out a comparative study of forward osmosis and membrane distillation for orange juice concentration and concluded that the fluxes are lower in membrane distillation due to the temperature gradient created between the bulk and the membrane interface, which reduces the driving force for water transport [12]. Dinçer *et al.* (2016) carried out a comparative study of forward osmosis and membrane distillation for black mulberry juice concentration [13]. They indicated that forward osmosis was generally superior in comparison to membrane distillation process especially in maintaining nutrient content and volatile components in the feed. In this study, forward osmosis process is proposed for milk concentration. The effect of important process parameters that influence milk processing by forward osmosis such as feed flow rate, draw solution flow rate, draw solution concentration, feed/draw solutions volume ratios, and temperature on water fluxes was investigated. Thus optimum conditions for the process were obtained. The studies in the literature and the obtained results in this work lend credence to potential use of the osmotic distillation in dairy processing.

MATERIALS AND METHODS

Chemicals

$CaCl_2$ was purchased from Carlo Erba in its anhydrous powder form. The milk used in the experiments was prepared by using 9 wt.% dry milk powder purchased from local brand of Pınar. NaOH and sodium azide were purchased from Sigma Aldrich.

Apparatus

X50 Liqui-cel® 1.7×5.5 Minimodules purchased from Membrana GmbH were used as membrane contactors. The membrane contactors have 7400 microporous polypropylene fibers which provide 0.58 m^2 contact surface.

Experimental procedure

In forward osmosis, the aim is to transfer the water molecules from the feed solution with higher activity to the draw solution with lower activity where the driving force is the activity difference of water molecules between two sides of the membrane. The transfer of the water through the membrane pore is achieved by three steps: (1) Vaporization of water in feed-membrane interface on the boundary layer, (2) the transport of the water vapor through the membrane pore to the other interface, (3) the condensation of water in brine- membrane interface on boundary layer. Since the water molecules in milk have higher activity than the ones in brine, they transfer to the brine side. Thus milk is concentrated by continuous removal of its water content. The illustration of mass transfer can be seen in Figure 1. The membrane material is polypropylene, which shows hydrophobic characteristics and thus the pores cannot be wetted by the water. The water is evaporated at the pore mouth; it diffuses through the pore and then condensates on the brine side boundary layer so only vapor form of water can be transferred through the pores.

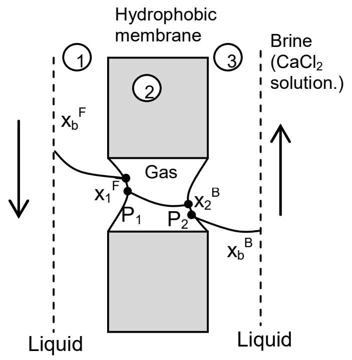

Figure 1 Mass transfer through the membrane.

The experimental setup for forward osmosis is illustrated in Figure 2. The feed solution, which is milk in this case, was circulated through the shellside of the membrane by a peristaltic pump while the brine solution was circulated through the lumen side of the membrane by another peristaltic pump. Both the temperatures of feed and brine were controlled using a thermostatic water bath. Every 10 minutes, the volume reduction in milk was recorded and the experiments were carried out until the dry matter in milk is reached to 20% wt from 9% wt.

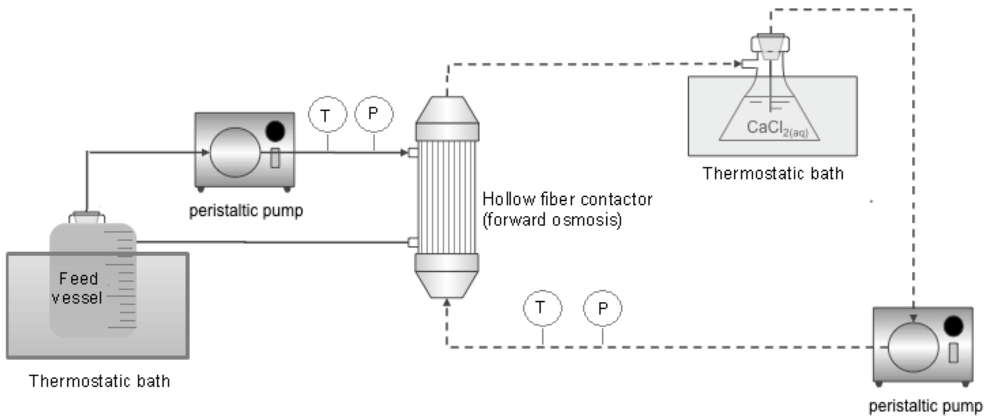

Figure 2: Experimental setup of forward osmosis process.

The process parameters and operating conditions studied in forward osmosis process are given in Table 1. The experiments were carried out to investigate the effect of the velocities of feed and brine solutions, brine concentration, feed/brine volume ratio, and temperature on the fluxes. The experiments that investigate the parameters apart from the temperature were carried out at 25°C.

Table 1: Operating conditions and process parameters of forward osmosis.

Parameter/Condition	Value/Range	Unit
Feed volume	400	mL
Temperature	25-40	°C
Solution flowrates	800-2000	mL min^{-1}
$CaCl_2$ concentration	3-5	M
V_{feed}:V_{brine}	1:1, 1:2, 1:3, 1:4	Volume ratios

RESULTS AND DISCUSSION

Since forward osmosis is a process in which the driving force is the activity difference between two sides of the membrane, the concentration of the brine is an important process parameter which affects directly the magnitude of this driving force. The experiments were carried out using 3, 4 and 5 M $CaCl_2$ brine solutions to investigate the effect of brine concentration on water fluxes. In Figure 3, the volume reduction of feed and water fluxes with time is given at different brine concentrations.

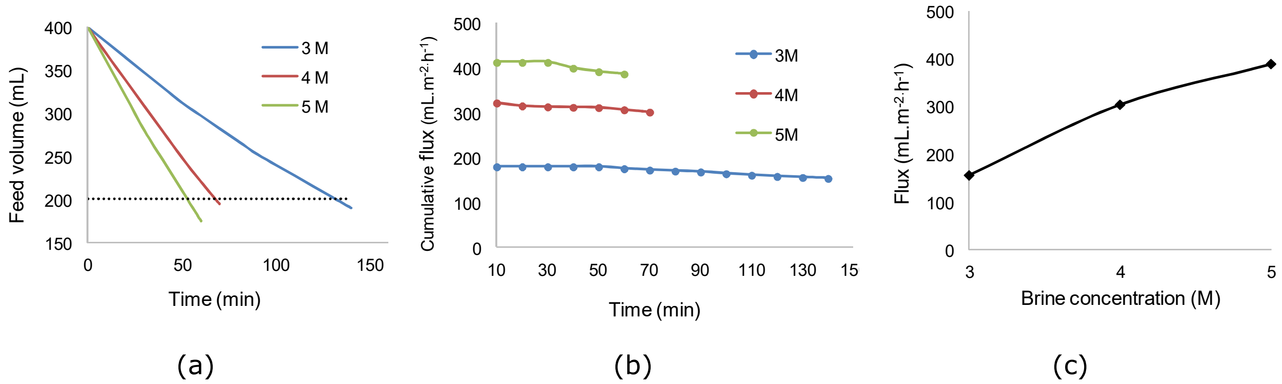

Figure 3: The effect of brine concentration on water permeation: (a) The volume change of milk with time, (b) cumulative water fluxes with time, (c) the fluxes at different brine concentrations.

As can be seen from Figure 3, fluxes increase with increasing brine concentration. As the brine concentration increases, the activity difference between two sides of the membrane increases and consequently the process becomes more rapid and operation time decreases for the required volume reduction. As can be seen from Figure 3a, the operation time for a volume reduction by half using 4 M brine is relatively closer to the operation time attained with 5 M brine. Thus considering the $CaCl_2$ consumption, an optimum concentration of 4 M brine was chosen for the further experiments. In Figure 3, it can be observed that fluxes tend to decrease slightly with time. This tendency is resulted from the decrease of activity difference with time because of the dilution of brine with the permeation of water continuously. Kujawski *et al.* investigated concentration of red grape juice by forward osmosis using $CaCl_2$ as draw solution [14]. They measured the change of the driving force difference in water activities between feed and draw solutions as a function of time of experiment using an initial draw solution of 50 wt% $CaCl_2$ and juice concentration of 5°Brix. They concluded that in extended time of experiment activity difference decreases continuously because of the dilution of the brine and concentration of the juice by time. Hence it is important to operate at brine volumes that is not affected by the dilution dramatically. Thus different feed volume to brine volume ratios including 1:1, 1:2, 1:3 and 1:4 were studied to investigate optimum ratio and results are presented in Figure 4.

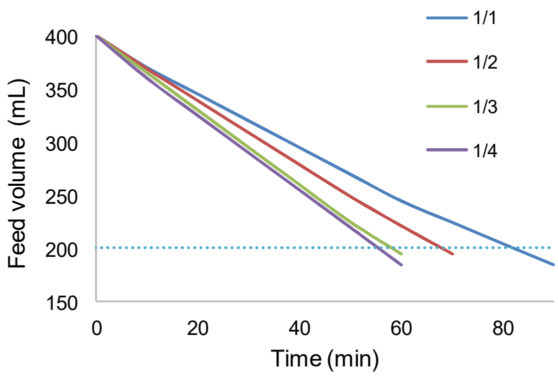

Figure 4: Feed volume reduction with time at different feed/brine volume ratios.

As can be seen from Figure 4, as the brine volume increases compared to the feed volume, the dilution of the brine becomes less significant and thus, operation time for a volume reduction by half decreases. However as the brine volume increases the consumption of $CaCl_2$ increases significantly, thus an optimum ratio should be considered for a more economic operation. Considering the optimum operation time and $CaCl_2$ consumption, the ratio of feed to brine was chosen as 1:2 in all experiments.

In order to investigate the effect of the feed velocity on fluxes, experiments were carried out at different feed velocities. The velocity of feed was changed as 800, 1200, 1600 and 2000 $mL.min^{-1}$ while the brine solution velocity was kept at 1600 $mL.min^{-1}$. The volume reduction and fluxes are presented in Figure 5 at various feed velocities.

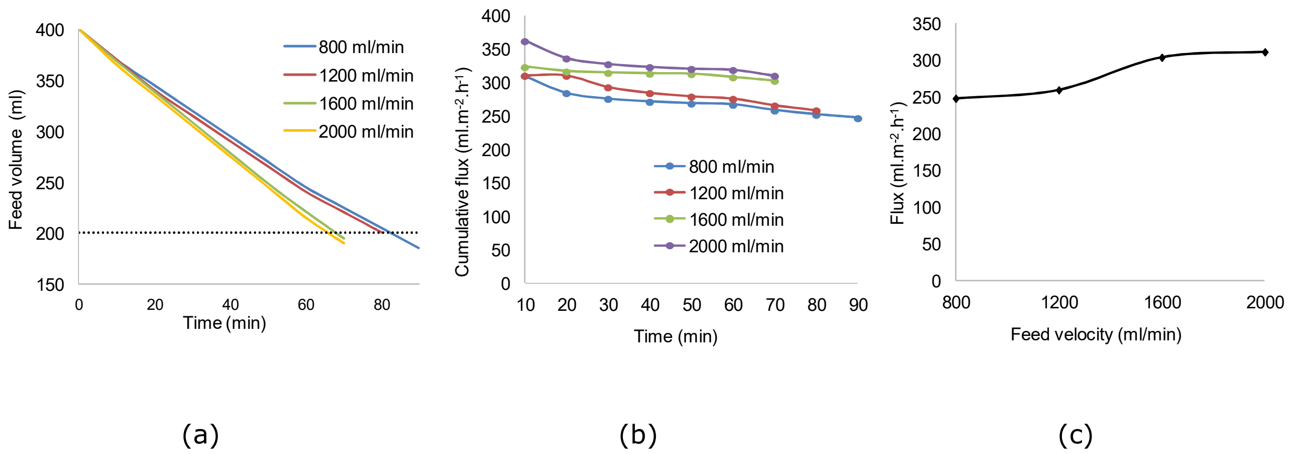

Figure 5: Effect of feed velocity on flux (a) The volume change of milk with time (b) Cumulative fluxes with time (c) Fluxes at different feed velocities.

As can be seen in Figure 5, as the feed velocity increases fluxes increase slightly and the operation time reduces accordingly. This is resulted from increase of the Re numbers and the decrease of the feed boundary layer thickness which presents resistance for the mass transfer. Since the mass transfer through the membrane requires the transfer of water molecules from feed boundary layer, membrane pores and brine boundary layer respectively, the overall mass transfer coefficient is expressed as follows according to resistance-in-series model:

$$K = \left(\frac{1}{K_f} + \frac{1}{K_m} + \frac{1}{K_d}\right)^{-1} \quad \textbf{(Eq. 1)}$$

where $1/K_f$, $1/K_m$ and $1/K_d$ are the transport resistances of feed boundary layer, membrane and draw solution boundary layer respectively. Thus, in forward osmosis process, it is important to define the resistances in order to describe the mass transfer through the membrane. After investigating the effect of the feed velocities, experiments were carried out at different brine velocities at fixed feed velocity in order to investigate the effect of brine velocity on fluxes. The brine velocities were in the range of 800-2000 mL.min^{-1} while feed velocity was kept at 1600 mL.min^{-1}. The results are presented in Figure 6.

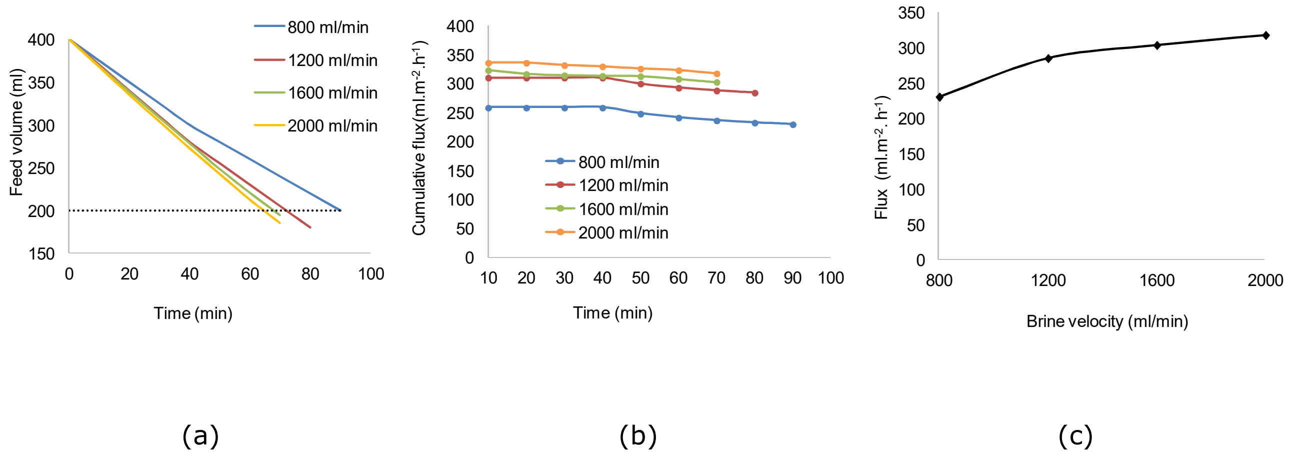

Figure 6: Effect of brine velocity on flux (a) The volume changes of milk with time, (b) Cumulative fluxes with time, (c) Fluxes at different brine velocities.

Figure 6 indicates that water fluxes increase with increasing brine velocity. At 800 mL.min^{-1} brine velocity, the operation time is longer. At this velocity value brine boundary layer presents a larger resistance for the mass transfer. As the velocity was increased to 1200 mL.min^{-1} operation time decreased however further increase in brine velocities did not affect the fluxes and consequently operation times significantly. Thus it can be concluded that although both feed and brine resistances affect the mass transfer, the mass transfer is dominated by feed side resistance and the brine boundary layer resistance affects the overall resistance less than the feed side. In general, increasing the feed or draw solution flow rate improves the water trans-membrane flux in osmotic distillation. This phenomenon can be attributed to the reduction in the related hydrodynamic boundary layer thickness [5]. Dova *et al*. (2007b) studied osmotic concentration of several model fruit juice solutions containing sucrose and glucose using a brine solution of NaCl [15]. They developed a model to analyze the resistances of water transport through the membrane and determined the magnitude of the two fluid resistances, namely the one on the side of osmotic medium containing salt solutions and the one on the side of the feed liquid containing sugar molecules. They showed that increasing the velocities of each sides affect the overall resistance and a detailed comparison of the two resistances indicated that feed side resistance was greater than the respective resistance of the osmotic medium. In this study also the effect of feed boundary layer resistance was found to be more dominant. This can be attributed to the difference in the magnitude of the tendencies of formation of fouling layers on each side. Since milk contains large protein and lactose molecules, the fouling is expected to be more severe on feed side. Thus change in hydrodynamic conditions on feed side affects the water flux more significantly.

The experiments were carried out at different temperatures in the range of 25-40 °C to investigate the effect of the temperature on the fluxes. In some experiments both feed and brine solution were heated to the same temperature while in some of the experiments the temperature of feed was kept slight higher than that of brine to generate a temperature difference between two sides of the membrane to enhance the driving force. The results are presented in Figure 7.

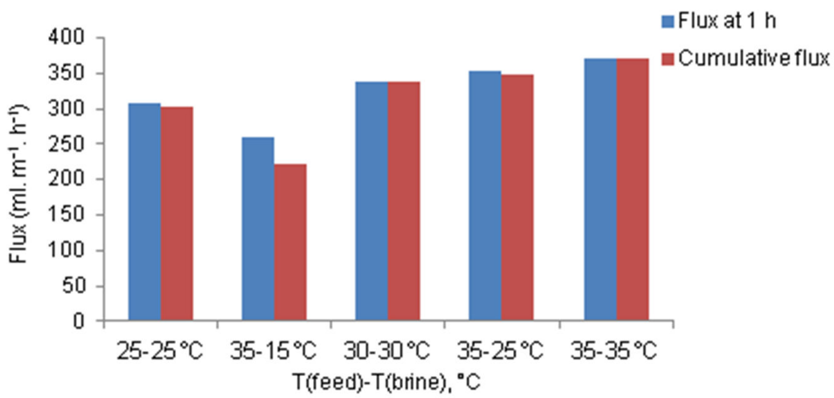

Figure 7: Water fluxes at various temperatures of brine and feed solutions.

When the feed and brine solutions are used at the same temperature, fluxes increase with increasing temperature. On the other hand, when the feed and brine solution have different temperatures, the temperature difference between two sides of the membrane gets closer during the experiment because of the evaporation-condensation steps of the water molecules as they release the latent heat to the brine solution. By this way, the temperature difference between the solutions decreases because of the temperature polarization and the system acts as at unique temperature. The fluxes attained at feed and brine temperatures of 35-25 °C respectively were slightly higher than the fluxes attained at 35-35 °C indicating a very small temperature difference may enhance the driving force. Nevertheless, the experiments carried out at feed and brine temperatures of 35-25 °C respectively have higher fluxes than the experiments carried out at 35-15 °C of feed-brine temperatures. Although the temperature difference is higher in the latter case, the total thermal energy given to the system is higher in the former case thus the fluxes are higher at 35-25 °C of feed and brine compared to the case at 35-15°C. In general, increasing the process temperature positively affects the fluxes. The viscosity of solutions to be processed decreases with increasing temperature and the diffusion coefficients increase with the temperature, and consequently water fluxes transmembrane increase. Similar results were obtained about the effect of temperature in literature. Thanedgunbaworn *et al.* (2007) reported that increasing bulk temperature from 25 to 35 °C, flux increased by 20% during the osmotic concentration of fructose [6]. Babu *et al.* (2006) indicated that increasing the process temperature from 25 to 45°C enhanced the water flux by 78% when they concentrated the pineapple juice by osmotic distillation [16]. These results show that temperature is an important process parameter in osmotic distillation.

CONCLUSION

In this work, several process parameters on water flux were investigated and optimum conditions were determined for the concentration of milk by forward osmosis. As the draw solution concentration increased, the fluxes increase due to the increase of the water activity difference on both sides of the membrane. When the effect of the different feed and draw solution flow rates were compared, the impact of the feed velocity was found to be slightly higher than that of brine velocity indicating that the feed side resistance is more significant especially when the viscosity increases in feed boundary layer during operation. As the temperatures of feed and brine increase, fluxes increase generally. A very small temperature difference between feed and draw solution generated around room temperatures enhances the fluxes. These results show that fluxes obtained at mild temperatures were satisfactory for a reasonable operation time. The flux values at investigated process conditions were found to be in the range of 155-387 mL. h^{-1} m^{-2} while the operation time was in the range of 50-120 min for the concentration process carried out until the volume reduction by half. In general, the concentration of milk was performed efficiently using forward osmosis process. Based on the results obtained, forward osmosis can be considered as an alternative process to conventional evaporation.

ACKNOWLEDGEMENTS

The authors acknowledge the support of the Scientific and Technological Research Council of Turkey (Project TÜBİTAK 115M137).

REFERENCES

1. Van Den Berg J. C. T.. Evaporated and condensed milk. In: Milk Hygiene, A monograph from WHO, Geneva, Switzerland: WHO Monograph series No:48, 1962. 321-345 p.

2. Liu DZ, Dunstan DE, Martin GJO. Evaporative concentration of skimmed milk: Effect on casein micelle hydration, composition, and size. Food Chemistry. 2012 Oct;134(3):1446–52. DOI: 10.1016/j.foodchem.2012.03.053.

3. Ramirez C, Patel M, Blok K. From fluid milk to milk powder: Energy use and energy efficiency in the European dairy industry. Energy. 2006 Sep;31(12):1984–2004. DOI: 10.1016/j.energy.2005.10.014.

4. Jiao B, Cassano A, Drioli E. Recent advances on membrane processes for the concentration of fruit juices: a review. Journal of Food Engineering. 2004 Aug;63(3):303–24. DOI: 10.1016/j.jfoodeng.2003.08.003.

5. Sant'Anna V, Marczak LDF, Tessaro IC. Membrane concentration of liquid foods by forward osmosis: Process and quality view. Journal of Food Engineering. 2012 Aug;111(3):483–9. DOI: 10.1016/j.jfoodeng.2012.01.032.

6. Thanedgunbaworn R, Jiraratananon R, Nguyen MH. Mass and heat transfer analysis in fructose concentration by osmotic distillation process using hollow fibre module. Journal of Food Engineering. 2007 Jan;78(1):126–35. DOI: 10.1016/j.jfoodeng.2005.09.023.

7. Zambra C, Romero J, Pino L, Saavedra A, Sanchez J. Concentration of cranberry juice by osmotic distillation process. Journal of Food Engineering. 2015 Jan;144:58–65. DOI: 10.1016/j.jfoodeng.2014.07.009.

8. Wenten IG, Khoiruddin. Reverse osmosis applications: Prospect and challenges. Desalination. 2016 Aug;391:112–25. DOI: 10.1016/j.desal.2015.12.011.

9. Keogh MK, Murray CA, O'Kennedy BT. Effects of ultrafiltration of whole milk on some properties of spray-dried milk powders. International Dairy Journal. 2003 Jan;13(12):995–1002. DOI: 10.1016/S0958-6946(03)00123-7.

10. Hausmann A, Sanciolo P, Vasiljevic T, Ponnampalam E, Quispe-Chavez N, Weeks M, et al. Direct Contact Membrane Distillation of Dairy Process Streams. Membranes. 2011 Jan 4;1(4):48–58. DOI: 10.3390/membranes1010048.

11. Hausmann A, Sanciolo P, Vasiljevic T, Kulozik U, Duke M. Performance assessment of membrane distillation for skim milk and whey processing. Journal of Dairy Science. 2014 Jan;97(1):56–71. DOI: 10.3168/jds.2013-7044.

12. Alves VD, Coelhoso IM. Orange juice concentration by osmotic evaporation and membrane distillation: A comparative study. Journal of Food Engineering. 2006 May;74(1):125–33. DOI: 10.1016/j.jfoodeng.2005.02.019.

13. Dincer C, Tontul I, Topuz A. A comparative study of black mulberry juice concentrates by thermal evaporation and osmotic distillation as influenced by storage. Innovative Food Science & Emerging Technologies. 2016 Dec;38:57–64. DOI: 10.1016/j.ifset.2016.09.012.

14. Kujawski W, Sobolewska A, Jarzynka K, Güell C, Ferrando M, Warczok J. Application of osmotic membrane distillation process in red grape juice concentration. Journal of Food Engineering. 2013 Jun;116(4):801–8. DOI: 10.1016/j.jfoodeng.2013.01.033.

15. Dova MI, Petrotos KB, Lazarides HN. On the direct osmotic concentration of liquid foods: Part II. Development of a generalized model. Journal of Food Engineering. 2007 Jan;78(2):431–7. DOI: 10.1016/j.jfoodeng.2005.10.011.

16. Babu BR, Rastogi NK, Raghavarao KSMS. Effect of process parameters on transmembrane flux during direct osmosis. Journal of Membrane Science. 2006 Sep;280(1–2):185–94. DOI: 10.1016/j.memsci.2006.01.018.

Türkçe Öz ve Anahtar Kelimeler

İleri Osmozla Yağsız Süt ve Süt Ürünlerinin Derişiklendirilmesi

Ayça Hasanoğlu, Kübra Gül

Öz: Meyve-sebze suları, süt gibi su içeren sıvı gıdaların suyunun giderilerek derişiklendirilmesi, gıdadaki besinsel değerin yoğunlaştırılması, raf ömrü ve stabilitenin arttırılması, bakteriyel faaliyetlerin azaltılması, depolama ve taşınım maliyetlerinin düşürülmesi açısından önemli bir kimyasal süreçtir. Süt, içerdiği suyun belli oranlarda uzaklaştırılmasıyla derişik süt olarak kullanılabilir. Aynı zamanda yoğurt, peynir, süt tozu, peynir altı suyu tozu gibi süt ürünlerinin yapımında sütteki suyun belli bir orana kadar uzaklaştırılarak derişiklendirilmesi şarttır. Bu çalışmada derişik süt üretimi, geleneksel buharlaştırma prosesine alternatif olarak membran kontaktörlerin kullanımıyla ileri osmoz süreci ile gerçekleştirilmiştir. Bu süreçte süt hidrofobik membran kontaktörün gövde kısmından geçerken, çekme çözeltisi olarak kullanılan $CaCl_2$ çözeltisi kontaktörün iç kısmından geçmektedir. Böylece suyun aktivitesi yüksek olan süt çözeltisinden, aktivitesinin düşük olduğu $CaCl_2$ çözeltisine geçişi sağlanmıştır. İleri osmoz sürecinde besleme ve çekme çözeltisi hızı, çekme çözeltisi konsantrasyonu, sıcaklık gibi işletme parametreleri incelenmiş ve suyun bu süreçle hızlı ve etkin bir şekilde sütten uzaklaştırılabildiği görülmüştür. Sütün hacmi yarıya düşene kadar derişiklendirme işlemi yapılmış ve incelenen süreç şartlarında akı değerleri 155-387 mL. h^{-1} m^{-2} aralığında bulunmuştur.

Anahtar kelimeler: İleri osmoz; membran kontaktör; sütün derişiklendirilmesi.

Sunulma: 30 Eylül 2016. **Düzeltme:** 21 Kasım 2016. **Kabul:** 30 Aralık 2016.