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APPLICATION OF ULTRAFILTRATION TO APPLE JUICE CLARIFICATION

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Abstract: Raw depectinized apple juice was clarified in a laboratory scale ultrafiltration (UF) system using ceramic tubular membranes (Tech-Sep Carbosep) with a molecular weight cut-off of 300,000, 50,000, and 30,000 daltons. The experiments have been carried out over a wide range of transmembrane pressures (100-400 kPa), temperatures (293-328 K), and feed flow rates (100-900 mL/min). Permeate flux significantly decreased with time until a steady state was established. The steady-state permeate flux reached a maximum at a transmembrane pressure of about 200 kPa. Higher permeate flux was obtained at higher temperatures due to lower permeate viscosity. A linear relationship between steady-state permeate flux and feed flow rate in logarithmic coordinates was obtained and the slope of these lines ranged from 0.22 to 0.31.

Key words: ultrafiltration, apple juice, clarification, ceramic membrane, permeate flux.

Introduction

Application of ultrafiltration (UF) to the clarification of fruit and vegetable juices has been extensively studied during the last 15 years. Heatherbell et al. (1977) introduced UF to clarify apple juice and obtained a stable clear product. UF has also been employed for the clarification of pear (Kirk et al., 1983), orange and lemon juices (Capannelli et al., 1992, 1994), starfruit juice (Sulaiman et

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al., 1998), kiwifruit juice (Wilson and Burns, 1983), guava juice (Chan and Chaing, 1992), pineapple juice (Jiraratananon et al., 1997), passion juice (Jiraratananon and Chanachai, 1996), etc. Besides that, UF has been used in conjunction with ion-exchange resins to deacidify passion juice (Lue and Chiang, 1989) and to debitter grapefruit juice and grapefruit pulp (Hernandez et al., 1992).

Compared to the conventional clarification process, UF can bring the following benefits: (1) completely eliminate the use of diatomaceous earth, thereby reducing production costs and the problem of waste treatment; (2) improve product quality, especially the clarity of the juice; (3) increase the product yield; (4) reduce labor costs; and (5) allow enzyme recovery for potential reuse, thereby reducing total enzyme consumption to about 1/3 (Rösch, 1985).

Membrane clarification of juices has been tested using tubular modules (Alvarez et al., 1996; Padilla and McLellan, 1993), flat membrane modules (Sheu et al., 1987), spiral wound modules (Wu et al., 1990), hollow fiber modules (Constenla and Lozano, 1996) and dead-end or stirred batch cells (Riedl et al., 1998, Sulaiman et al., 1998). These studies have been carried out by using ceramic membranes (Alvarez et al., 1996), metallic membranes (Thomas et al., 1986; Barefoot et al., 1989), and polymeric membranes made of polysulfone, polyamide, fluoropolymers, and polypropylene (Riedl et al., 1998).

The main problem in practical applications of UF is a reduction in permeate flux through the membrane caused by fouling of the membrane surface. In order to control membrane fouling during juice clarification, several flux enhancement methods have been proposed such as periodic gas backwashing with air or N_2 (Su et al., 1993), permeate backwashing and pulsating entry flow (Ben Amar et al., 1990, Padilla and McLellan, 1993).

The main objective of this work is to study the effect of operating parameters such as transmembrane pressure, feed flow rate through the module, and temperature on the permeate flux in apple juice ultrafiltration. The experiments have been carried out using commercial ceramic tubular membranes in a batch mode of operation with total retentate recycle. Ceramic membranes afford many advantages over polymeric membranes such as thermal and chemical tolerance, resistance to abrasion and high mechanical strength.

Material and Methods

The schematic view of the experimental setup employed for the clarification of apple juice is shown in Fig. 1. The raw depectinized juice was pumped from the reservoir to the module by a rotary pump and the flow rate was controlled with a laboratory made rotameter. Temperature in the system was adjusted by passing juice from a bypass line through the thermostat bath. Permeate was collected in a reservoir placed on a Tehtnica model ET-1111 digital balance. The mass of permeate collected was measured with an accuracy of ±0.1 g every minute for periods of 120 min. Transmembrane pressure was controlled by the back-pressure valve. The feed flow rate through the module ranged between 100 and 900 mL/min.

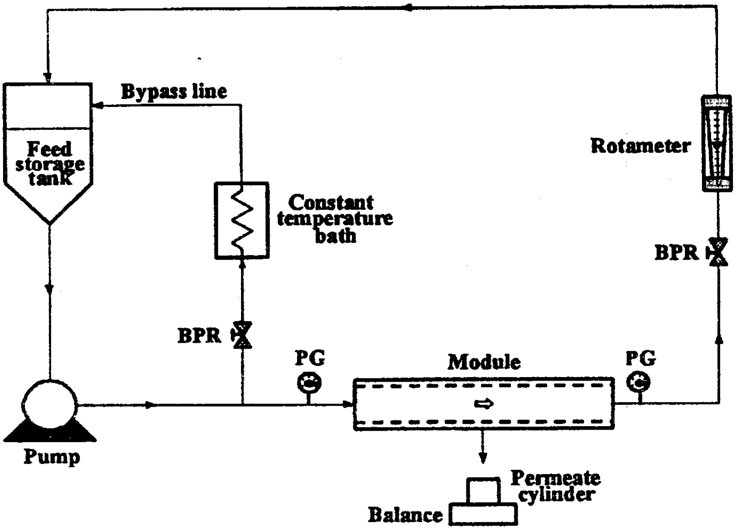

Fig. 1. - Schematic view of the experimental setup used in this work
(PG - pressure gauge, BPR - back-pressure regulator)

The experiments were performed with three tubular inorganic ultrafiltration membranes produced by Tech-Sep (Rhône-Poulenc Group, Miribel, France), type Carbosep M9, M8, and M7. These anisotropic membranes are made with an inner diameter of 6 mm and a molecular weight cut-off of 300,000, 50,000 and 30,000 daltons, respectively. Carbosep membranes are composed of thin permselective skin of zirconium oxide and titanium dioxide supported by a porous carbon substructure. The membranes were installed inside a cylindrical stainless steel module with an effective membrane length of 225 mm and an effective membrane area of 42.4 cm^2. The membrane was cleaned after each experiment with a hot 1 wt % solution of NaOH and 1 mg/L NaClO solution for about 30 minutes. The acid cleaning was not used because no improvement in permeate flux recovery was observed.

Results and Discussion

The cumulative permeate volume as a function of time for Carbosep M7 membrane at different pressures, temperatures and feed flow rates is shown in Fig. 2. Similar dependencies were obtained for M7 and M8 membranes, but for the sake of brevity these dependencies are omitted from the figure. The permeate volume collected during ultrafiltration of apple juice increased with time but at a decreasing rate. Besides that, the permeate volume collected at any time increased with transmembrane pressure, temperature and feed stream flow rate in the investigated range of operating parameters. The lower permeation rates for apple juice in comparison with those of water were due to the membrane fouling by the suspended solids.

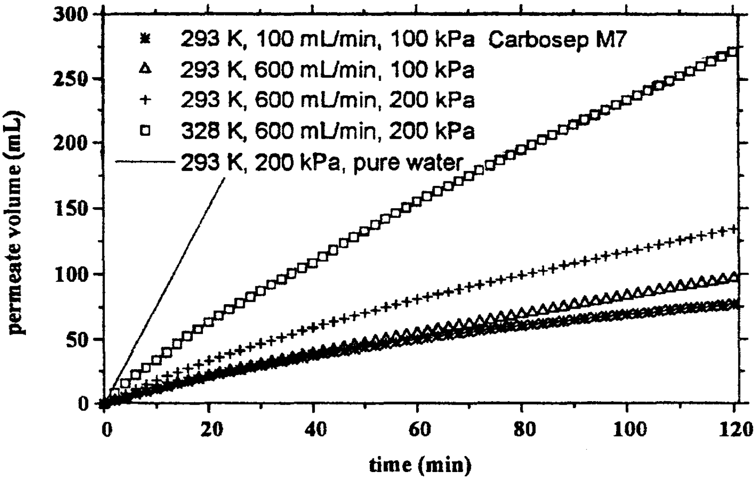

Fig. 2. - Permeate volume vs. time for ultrafiltration of apple juice and pure water using Carbosep M7 membrane (every second data point is shown)

The permeate volume collected during pure water ultrafiltration increased linearly with time. Membrane resistances estimated from the slopes of these lines were 0.671, 0.654, and 3.96 x 10^{13} l/m for Carbosep M7, M8, and M9 membrane, respectively.

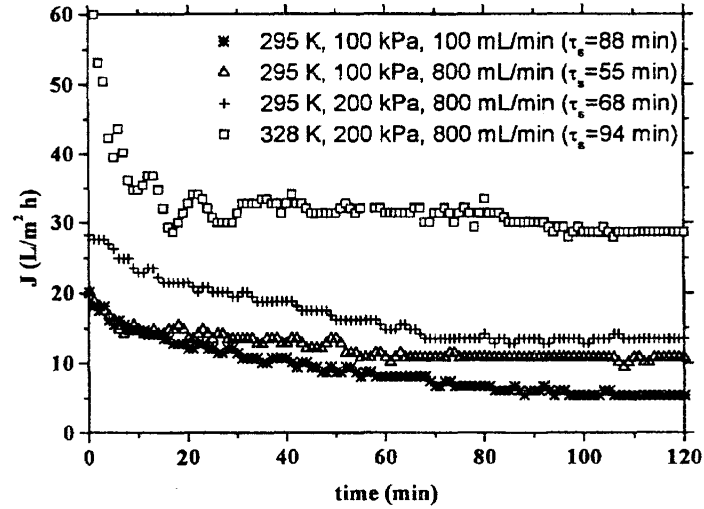

Fig. 3. - Variation of permeate flux with time for Carbosep M7 membrane (the time τ_s at which the steady state was established is shown in brackets)

Experimental data corresponding to the permeate flux decline using the Carbosep M7 membrane are presented in Fig. 3. Flux values were obtained by numerical differentiation of the mass versus time data collected during each experiment. A significant permeate flux decline was observed during the first 20 min of operation, followed by a progressive stabilization of the flux, leading to a stationary value. For operating conditions as in Fig. 3, the steady state was established after 55-94 min of operation and the steady-state permeate fluxes were 27-53 % of their initial values and only 14-19 % of the pure water fluxes. It should be noted that the initial permeate flux is independent on the feed stream flow rate.

The effect of transmembrane pressure, TMP, on steady-state permeate flux, J_s is shown in Fig. 4. Permeate flux increased initially with applied transmembrane pressure, and then decreased with continued increase in the transmembrane pressure. The point at which the permeate flux was maximal was the optimum transmembrane pressure. In our experiments this optimum occurred at 160-240 kPa and decreased with feed stream flow rate. The optimum TMP of 160 kPa for Carbosep M8 membrane at 800 mL/min is in good agreement with 157 kPa reported by Kirk et al. (1983) for pear juice ultrafiltration, 154 kPa reported by Sulaiman et al. (1998) for starfruit juice ultrafiltration, and 140-145 kPa reported by Rao et al. (1987) for apple juice ultrafiltration using polysulfone hollow fibers.

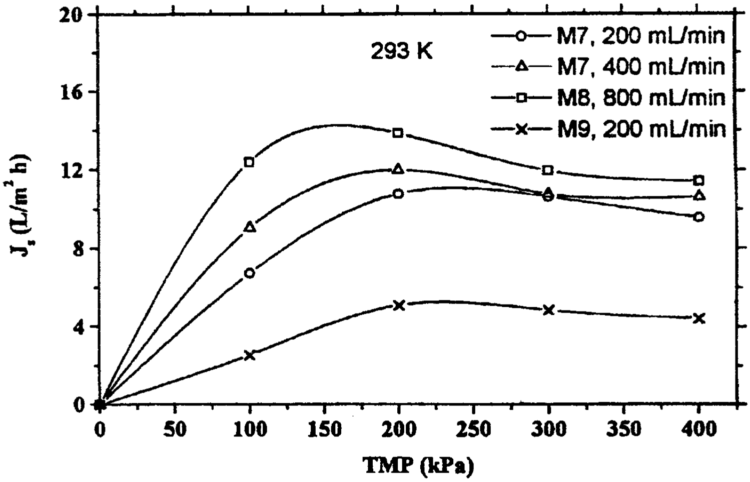

Fig. 4. - Effect of transmembrane pressure on steady-state permeate flux at 293 K using Carbosep M7, M8 and M9 membranes at different feed stream flow rate

The bell-shaped permeate flux-pressure behavior for UF of apple juice is in contrast to what many investigators have found for protein solutions (Do and Elhassadi, 1985) and colloidal inorganic oxide dispersions (Vladisavljević et al., 1992; 1995). Permeate flux of protein solutions and colloidal inorganic oxide dispersions continued to increase but at a decreasing rate with transmembrane pressure until it reached a constant limiting value.

The possible explanation of these results could be that pectic substances and suspended juice solids have physical characteristics different from proteins and inorganic oxide particles. Proteins can be regarded as rigid globular molecules. When a gel layer of protein builds up, the spherical shape leaves spaces for passage of the permeate. On increasing transmembrane pressure these spaces never become completely closed, thus allowing the permeate to pass through to give a flux plateau above the optimum TMP. On the other hand, apple juice haze particles are chain-like aggregates of polymerized polyphenol and/or polyphenol-protein complexes (Beveridge and Tait, 1993). At a sufficiently high transmembrane pressure, these large aggregates are disrupted into much smaller particles forming a less porous and more resistive gel. Similar to that, pectic substances are chain-like macromolecules of galacturonic acid units aggregated by hydrogen bond bridges. Kirk et al. (1983) have suggested that when pectic gel layer is compressed, these bridges could collapse leading to the closure of interstitial spaces between the chains. They reported that the pectic gel layer is elastic as evidenced by the partial restoration of permeate flux upon gradual release of the transmembrane pressure.

Fig. 5 indicates that log(steady-state permeate flux) increases in proportion to log(feed flow rate) for all three Carbosep membranes, i.e.:

$$J_s = AQ_f^B \qquad (1)$$

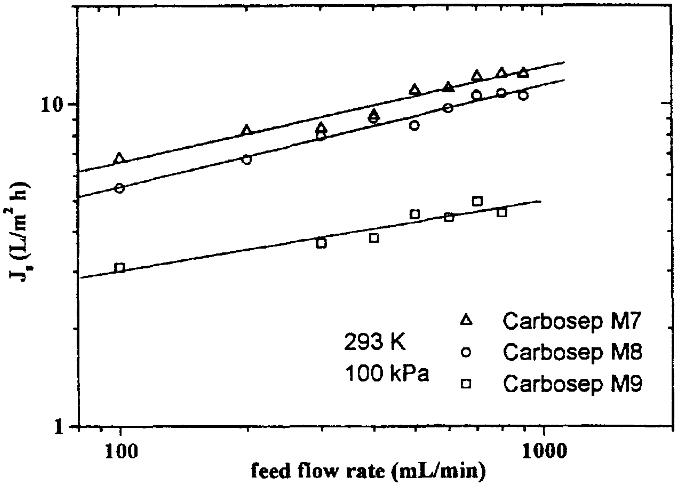

Fig. 5. - Effect of feed flow rate on steady-state permeate flux for all three membranes

The power law parameters A and B in Eq. (1) were obtained by the nonlinear least-squares regression of the plots in Fig. 5 (Table 1). The exponent on feed flow rate ranges between 0.22 and 0.31 which is in good agreement with 0.33 anticipated by Lévêque (1928) for heat transfer in laminar flow channels. However, the values of B in Table 1 are much lower than 0.54 obtained by

Charcosset and Choplin (1996) for UF of pectine solutions under laminar flow conditions using Carbosep M8 membrane.

Tab. 1. - Values of parameters A and B in permeate flux vs. feed flow rate correlations (Eq. 1) determined using the least-squares regression analysis method

Membrane	t (K)	Δp (kPa)	A*	B*	Correlation coefficient
Carbosep M7	293	100	1.72	0.29	0.978
Carbosep M8	293	100	1.30	0.31	0.987
Carbosep M9	293	100	1.10	0.22	0.945

*With J_s in $L/(m^2h)$ and Q_f in mL/min.

The effect of temperature on permeate flux for the three Carbosep membranes at a transmembrane pressure of 200 kPa and a feed flow rate of 800 mL/min is illustrated in Fig. 6. As expected, the permeate flux increased with increasing temperature, which is due to a decrease in the permeate viscosity. For Carbosep M9 membrane, the permeate flux increased linearly with temperature at a rate of 0.23 $L/(m^2h)$ for each 1 K. For Carbosep M7 and M8 membrane, an exponential increase of the permeate flux was observed.

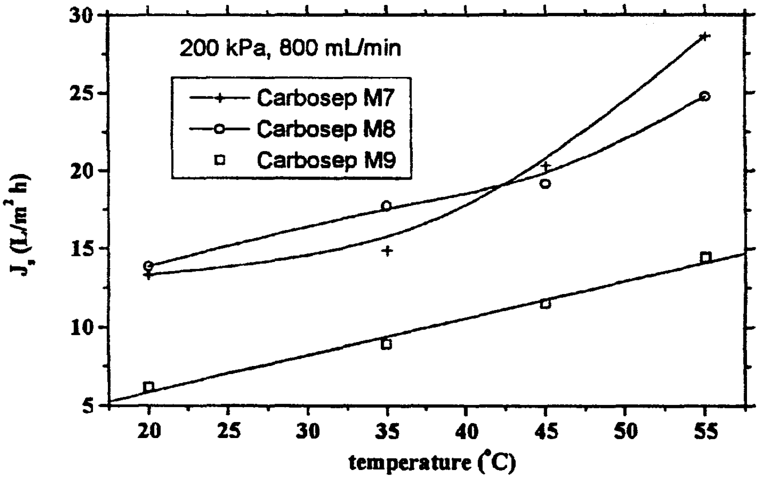

Fig. 6. - Effect of temperature on steady-state permeate flux for all three membranes

Conclusion

Ceramic tubular UF membranes with 300,000, 50,000, and 10,000 dalton molecular weight cut-off were successfully used to clarify depectinized apple juice. Decline in permeate flux over time was attributed to the formation of a layer of retained juice solids on the surface of the membrane that increased hydraulic resistance. The steady-state permeate flux increased with transmembrane pressure until it reached a maximum value at a TMP of about 200 kPa and then decreased with further increase in TMP. The observed decrease in permeate flux after reaching a maximum value was probably due to the disruption of large aggregates of haze particles into much smaller particles forming a less porous and more resistive gel layer. The steady-state permeate flux was proportional to the feed flow rate raised to powers ranging between 0.22 and 0.31.

REFERENCES

1. Alvarez, V., Andres, L.J., Riera, F.A., Alvarez, R. (1996): Microfiltration of apple juice using inorganic membranes: process optimization and juice stability, Can. J. Chem. Eng, Vol. 74, p. 156-162.
2. Barefoot, S.F., Tai, H.Y., Brandon, S.C., Thomas, R.L. (1989): Production of microbiologically stable apple juice by metallic membranes ultrafiltration, J. Food Sci, Vol. 54, p. 408-411.
3. Ben Amar, R., Gupta, B.B., Jaffrin, M.Y. (1990): Apple juice clarification using mineral membranes: fouling control by backwashing and pulsating flow, J. Food Sci., Vol. 55, p.1620-1625.
4. Beveridge, T., Tait, V. (1993): Structure and composition of apple juice haze, Food Structure, Vol. 12, p. 195-198.
5. Capannelli, G., Bottino, A., Munari, S., Ballarino, G., Mirzaian, H., Rispoli, G., Lister, D., Maschio, G. (1992): Ultrafiltration of fresh orange and lemon juices, Lebensm.- Wiss. u. - Technol., Vol. 25, p. 518.
6. Capannelli, G, Bottino, A., Munari, S., Lister, D.G., Maschio, G., Becchi, I. (1994): The use of membrane processes in the clarification of orange and lemon juices, J. Food Eng., Vol. 21, p. 473-483.
7. Chan, W.Y., Chaing, B.H. (1992): Production of clear guava nectar, Inter. J. Food Sci. Technol., Vol. 27, p. 435.
8. Charcosset, C., Choplin, L. (1996): Ultrafiltration of non-newtonian fluids, J. Membr. Sci., Vol. 115, p. 147-160.
9. Constenla, D.T., Lozano, J.E. (1996): Predicting stationary permeate flux in the ultrafiltration of apple juice, Lebensm. - Wiss. u .- Technol, Vol. 29, p. 587-592.
10. Do, D.D., Elhassadi, A.A. (1985): A theory of limiting flux in a stirred batch cell, J. Membr. Sci, Vol. 25, p. 113-132.
11. Heatherbell, D.A., Short, J.L., Strubi, P. (1977): Apple juice clarification by ultrafiltration, Confructa, Vol. 22, p. 157-169.
12. Hernandez, E., Couture, R., Rouseff, R., Chen, C.S., Barros, S. (1992): Evaluation of ultrafiltration and adsorption to debitter grapefruit juice and grapefruit pulp wash, J. Food Sci., Vol. 57, p. 664-666.
13. Jiraratananon, R, Chanachai, A. (1996): A study of fouling in the ultrafiltration of passion fruit juice, J. Membr. Sci., Vol. 111, p. 39-48.
14. Jiraratananon, R., Uttapap, D., Tangamornsuksun, C. (1997): Self-forming dynamic membrane for ultrafiltration of pineapple juice, J. Membr. Sci., Vol. 129, p. 135-143.

15. Kirk, D.E., Montgomery, M.W., Kortekaas, M.G. (1983): Clarification of pear juice by hollow fiber ultrafiltration, J. Food Sci., Vol. 48, p. 1663-1666.

16. Lévêque, M.A. (1928): Les lois de la transmission de chaleur par convection, Ann. Mines Rec. Mem. L'Explotas Mines, Vol. 13, p. 201.

17. Lue, S.J., Chiang, B.H. (1989): Deacidification of passion fruit juice by ultrafiltration and ion-exchange processes, Int. J. Food Sci. Technol., Vol. 24, p. 395-401.

18. Padilla-Zakour, O., McLellan, M.R. (1993): Optimization and modeling of apple juice cross-flow microfiltration with a ceramic membrane, J. Food Sci., Vol. 58, p. 369-388.

19. Rao, M.A., Acree, T.E., Cooley, H.J., Ennis, R.W. (1987): Clarification of apple juice by hollow fiber ultrafiltration: fluxes and retention of odor-active volatiles, J. Food Sci., Vol. 52, p. 375-377.

20. Riedl, K., Girard, B., Lencki, R.W. (1998): Influence of membrane structure on fouling layer morphology during apple juice clarification, J. Membr. Sci., Vol. 139, p. 155-166.

21. Rösch, E., Experience with ultrafiltration in the 1984 season, Confructa, Vol. 30, p. 27-30.

22. Sheu, M.J., Wiley, R.C., Schlimme, D.V. (1987): Solute and enzyme recoveries in apple juice clarification using ultrafiltration, J. Food Sci., Vol. 52, p. 732-736.

23. Su, S.K., Liu, J.C., Wiley, R.C. (1993): Cross-flow microfiltration with gas backwash of apple juice, J. Food Sci., Vol. 58, p. 638-641.

24. Thomas, R.L., Westfall, P.H., Louvieri, Z.A., Ellis, N.D. (1986): Production of apple juice by single pass metallic membrane ultrafiltration, J. Food Sci., Vol. 51, p. 559-563.

25. Vladisavljević, G.T., Milonjić, S.K., Nikolić, D., Pavasović, V.Lj. (1992): Influence of temperature on the ultrafiltration of silica sol in a stirred cell, J. Membr. Sci., Vol. 66, p. 9-17.

26. Vladisavljević, G.T., Milonjić, S.K., Pavasović, V.Lj. (1995): Flux decline and gel resistance in unstirred ultrafiltration of aluminium hydrous oxide sols, J. Colloid Interface Sci., Vol. 176, p. 491-494.

27. Wilson, E.L., Burns, D.J.W. (1983): Kiwifruit juice processing using heat treatment techniques and ultrafiltration, J. Food Sci., Vol. 48, p. 1101-1105.

28. Wu, M.L., Zall, R.R., Tzeng, W.C. (1990): Microfiltration and ultrafiltration comparison for apple juice clarification, J. Food Sci., Vol. 55, p. 1162-1163.

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PRIMENA ULTRAFILTRACIJE ZA BISTRENJE SOKA JABUKE

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Rezime

Bistrenje depektinizovanog matičnog soka jabuke uspešno je vršeno ultrafiltracijom kroz keramičke cevne membrane proizvedene od cirkonijum-oksida i titan-dioksida na podlozi od poroznog ugljenika. Eksperimenti su vršeni uz potpunu recirkulaciju retentata i sa protocima napojne struje u modulu od 100-900 ml/min. Fluks permeata u početku opada sa vremenom, ali se posle određenog vremena uspostavlja stacionarno stanje. Stacionarni fluks permeata, J_s dostiže maksimum pri transmembranskom pritisku od oko 200 kPa, nakon čega opada sa daljim porastom pritiska. To se može objasniti deformacijom čestica mutnoće u sloju gela, raskidanjem krupnijih lančastih agregata otvorene strukture i formiranjem sitnijih čestica koje se gušće pakuju u sloju gela, što dovodi do smanjenja poroznosti i propustljivosti sloja. U log-log koordinatnom sistemu stacionarni fluks permeata linearno raste sa porastom protoka napojnog soka, pri čemu nagib ovih pravih varira od 0,22 do 0,31.

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