



ELSEVIER

Journal of Membrane Science 150 (1998) 99–110

Journal of
MEMBRANE
SCIENCE

A study of the direct osmotic concentration of tomato juice in tubular membrane – module configuration. I. The effect of certain basic process parameters on the process performance

Konstantinos B. Petrotos^{a,*}, Peter Quantick^a, Heracles Petropakis^b

^a*School of Applied Science and Technology, University of Lincolnshire and Humberside, 61 Bargate str, DN34 5AA, Grimsby, NE Lincolnshire, UK*

^b*Department of Chemical Engineering, Laboratory of Food process Engineering, School of Engineering, Aristotle University of Thessaloniki, 54006 University Campus, Thessaloniki, Greece*

Received 11 March 1998; accepted 7 July 1998

Abstract

A novel tubular module was used to investigate the direct osmosis concentration process in the case of tomato juice. This module, was constructed, according to given specifications, by PCI UK and consisted of an external stainless steel shroud accommodating, internally, a set of two identical RO membrane tubes having no support lengthwise and properly sealed at their ends. The process performance was measured in terms of water permeation flux and its response to changes of the process parameters was experimentally assessed and established. The process parameters which were investigated in the course of this study were: the kind of osmotic medium, the viscosity of osmotic medium, the osmotic medium concentration, the juice temperature, the juice flow rate, the juice concentration and the membrane thickness. Sodium chloride brine was found to be the best osmotic medium, among the six which were tried, and this was due to its very low viscosity. The above parameter appears to be of paramount importance regarding the effectiveness of an osmotic medium. Higher osmotic medium concentrations yielded to higher osmotic permeation rates. Increasing the juice temperature was found to markedly increase the permeation flux. However, only a slight enhancement of flux was observed by increasing the juice flow rate. Moreover, higher juice concentrations up to approximately 12° Brix led to a lowering of the osmotic flux. Finally, as far as the membrane thickness was concerned, a strong trend was revealed for exponential increase of permeation by shifting towards lower membrane thicknesses. This trend however needs to be further investigated as an inadequate number of experimental points were obtained due to lack of additional membranes. © 1998 Elsevier Science B.V. All rights reserved.

Keywords: Concentration polarisation; Osmosis; Reverse osmosis; Tomato juice; Concentration

1. Introduction

The basic principle of osmotic concentration was applied by Popper et al. [1] under the name dialysis, to obtain grape juice concentrates.

Beaudry and Lampi [2,3] then renamed the same process “direct osmosis” describing its basic principles: Direct osmosis demands the concentrating juice to be in intimate contact with the osmotic agent across a very thin (25–100 μm overall thickness), semipermeable and water selective membrane with a molecular weight cut-off of 100 Da. Any water

*Corresponding author.

solution with higher osmotic pressure than the juice and with a solute which does not pass through the membrane can be used as an osmotic medium and as no substantial hydraulic pressure is employed in this process the driving force producing water transport through the membrane is virtually the difference in osmotic pressure across the membrane. An innovative design, ensuring a consistently high water flux and minimal fouling, was employed by Beaudry and Lampi (Osmotek, Corvallis, Oregon, USA) in the construction of their direct osmosis cell. The whole invention of Osmotek, has been covered by a USA patent, granted to Herron et al. [4] under the no: 5 281 430, which is currently the only available source of information about this new process. In the disclosure section of this patent document there is very brief reference to application of this novel process in the concentrating of tomato juice but a thorough study of the application of direct osmosis in the concentration of the most concentrated vegetable juice is lacking. On the other hand, the concentration of this juice (tomato juice) by using reverse osmosis techniques has been extensively studied and information is available in a series of scientific papers [5–12].

Therefore, the purpose of the present work is to investigate the effect of the process parameters on the direct osmosis concentration flux of tomato juice which is concentrated in bulk worldwide and furthermore to provide a further insight into this new and not very well known process.

2. Materials and methods

A novel direct osmosis tubular module was constructed, according to our specifications, by the membrane systems manufacturer Paterson Candy Industries UK. This system consisted of a stainless steel shroud which provided containment to a set of two reverse osmosis membrane tubes, 1.22 m in length and with an external surface of approximately 0.1 m^2 . These tubes were properly sealed at both their ends by using rubber gaskets similar to those used in RO modules and were self-supported lengthwise. The tomato juice was circulated through the systems inside the bore of the tubes and the tube arrangement and connection was in series with the assistance of a *U*-bend element. The osmotic medium was recircu-

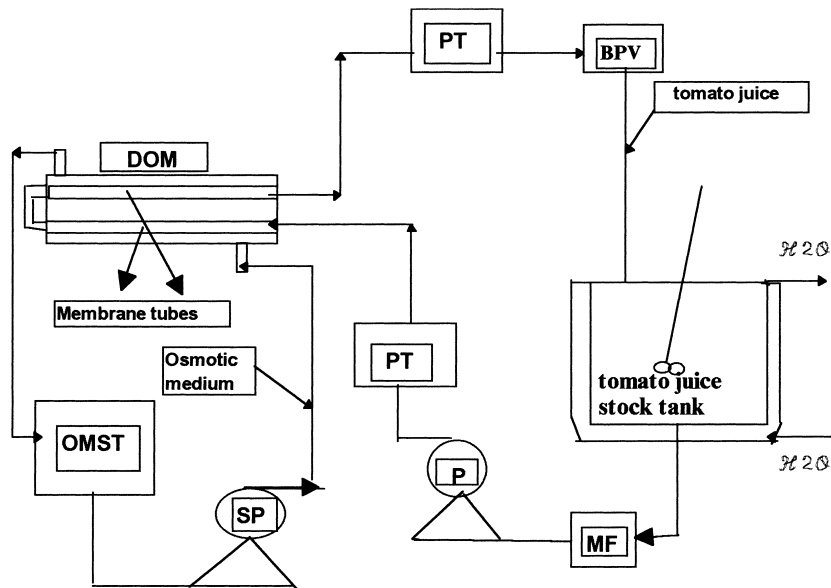
lated each time outside the tubes in the shroud. The overall arrangement of the DO experimental rig is illustrated in Fig. 1. Two different membranes were used in the course of the experimentation: the AFC99 commercial reverse osmosis membrane which is a thin film composite aromatic polyamide membrane with nominal sodium chloride rejection 99%. Two different overall thicknesses of this membrane were tried: 600 and 500 μm . Additionally the company constructed especially for the needs of the research a novel thinner membrane (400 μm) where the difference in thickness was due to the use of a thinner backing material while the top active layer was identical to that of the commercial AFC99. The external diameter of the membrane tube was approximately 2.6 cm and the membrane active layer was located on the inside of the tube. The thickness of the membrane active layer was in all three membranes 140 μm , while the thickness of the porous substrate was 460, 360 and 260 μm from the thicker to the thinner membrane. The water permeation coefficient was the same for all membranes and according to the supplier had the value $A=1.125 \times 10^{-3} \text{ m}^3/\text{m}^2 \text{ h}$. The water coefficient *B* was not available by the supplying company but its value could be based on a 99% rejection of NaCl [13]. All the membranes were supplied by Paterson Candy Industries UK.

The cleaning of the membranes was performed according to the membrane manufacturer specification, by using the P3 Ultrasil 10 alkaline detergent which was supplied by HENKEL UK.

The raw material of the direct osmosis experiments was prepared by mixing commercial double concentrated tomato paste (28–30° Brix) with soft water, obtained from the Lab boiler house water purification unit. The mixing was done at a ratio (water:tomato paste) 5.35:1. This mixture, approximately 11 kg, was then left agitated for at least 1 h at 30 rpm in order to be homogenised. The tomato paste which was used for the reconstitution of the juice was supplied by GERBER UK in 1 kg tins and was of Greek origin.

Sodium chloride of technical Grade was supplied by UK manufactures as well as calcium chloride, calcium nitrate, glucose, sucrose and polyethylene glycol 400 (PEG400). All these materials were used for the preparation of osmotic media.

The experimental procedure was consisted of the following steps:



MF = Magnetic flowmeter - MAGFLO MAG 3000 by DANFOSS Co
P = Positive displacement pump - RANNIE PISTON PUMP
(cap. 10 litres/min)
SP = Stroke pump. cap. 10 litres / min
PT = Pressure transducer - supplied by DRUCK LTD - UK.
DOM = Direct osmosis module.
OMST = Osmotic medium stock tank- PLASTIC BUCKET cap. 15 LITRES
BPV = Back pressure valve.
TOMATO JUICE STOCK TANK- cap. 50 litres

Fig. 1. Direct osmosis experimental rig.

- Soft water and tomato paste at the above mentioned ratio were put into the juice stock tank and left agitated for at least 1 h.
 - Preweighed quantities of water and osmotic medium solute at the required ratio were put in the plastic osmotic medium stock tank and the stroke pump was put in operation with the output pipe emptying the material back into the plastic tank. The osmotic medium was in this way recycled and mixed for at least 1 h, to become homogenised.
 - After this first hour had elapsed the output pipes of the pumps were connected to the module and the tomato juice pump was put in operation. When the first quantity of tomato juice was returned to the stock tank after passing through the module, the back pressure valve was set immediately to the right position to ensure a back pressure of about 3 bar and the stroke pump of the osmotic medium was set in operation.
 - The normal run lasted 5 h and during this time the osmotic medium was sampled at regular time intervals, every 15 min for the first 90 min of operation and every 30 min for the rest of the operation.
 - The collected samples were then analysed by using a refractometer and the refractive index was interpreted into concentration.
 - From the drop of the concentration of the osmotic medium in the 5 h run the water which had passed through the membrane was calculated by using a material balance and obtaining consequently the permeation flux.
- For the refractive index measurements, the high precision refractometer RFM340 was used which was supplied by Stanley & Bellingham, UK.

Viscosity measurements for the osmotic media were conducted by using a BOHLIN CS Rheometer operated at the stress viscometry mode.

3. Results and discussion

3.1. Comparison of the effectiveness of several osmotic media in the direct osmotic concentration of tomato juice

A total of six different osmotic media were used in order to ascertain the one presenting the optimum performance. These were sodium chloride solution, calcium chloride solution, calcium nitrate solution, sucrose solution, glucose solution and polyethylene glycol solution with a molecular weight of 400 Daltons (PEG400). The experimental conditions used along with the average concentration of the osmotic media are listed in Table 1. The data of Table 2 indicate that salt solutions perform better as osmotic media than carbohydrate solutions and the PEG400. Also, the best performing osmotic medium among all tested was NaCl brine. The viscosity of the osmotic media was measured by an automatic Bohlin stress viscometer and the values of their respective osmotic pressures were calculated using literature data.

The driving force for the separation was then calculated each time by subtracting an element equal to 8.7 bar corresponding to the osmotic pressure of the tomato juice at 4.3° Brix (equation given by Dale et al. [14]) and adding the tomato juice overpressure (2 bar). The driving force ($\Delta\Pi - \Delta P$) along with the viscosity values of the osmotic media and the calculated flux values are all presented in Table 2. Statistical processing of the data of flux versus ($\Delta\Pi - \Delta P$) proved that

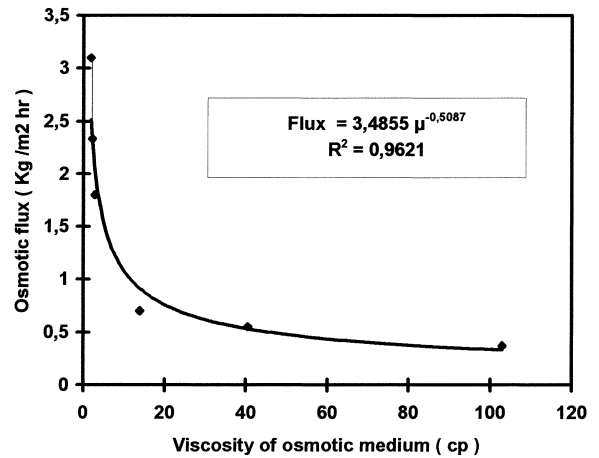


Fig. 2. Correlation of the direct osmosis flux with osmotic medium viscosity in direct osmosis concentration of tomato juice.

there is only little correlation between these two parameters (the correlation coefficient $R=0.10$ is close to zero). This means that the performance of the process of direct osmosis is dependent on the diffusion of water from the membrane active layer through the membrane backing material and osmotic medium polarised layer, which is in turn dependent on the physical properties of the used osmotic solution. On the other hand graphical representation of the flux values against the measured viscosity values of the osmotic solutions (Fig. 2), disclosed a strong exponential dependence between the two parameters, where the flux value appears to be inversely proportional to the square root of the viscosity. This signifies once again the importance of using a low viscosity osmotic medium in direct osmosis applications to enhance performance. Lower viscosity of an osmotic medium besides its direct positive effect on eliminating

Table 1

243Experimental conditions for the six experiments employing different osmotic media

	Solution					
	NaCl	CaCl ₂	Ca(NO ₃) ₂	Glucose	Sucrose	PEG400
Tomato juice flow rate (l/h)	502	510	498	500	501	504
Osmotic medium flow rate (l/h)	565	554	572	552	570	555
Osmotic medium concentration (average) (w/w%)	22.24	29.07	29.0	62.86	58.29	49.97
Osmotic pressure data calculated according to or obtained from	[18]	[24]	[23,26]	[26]	[26]	[25]

Tomato juice pressure: 2.95 ± 0.1 , tomato juice temperature: 26°C, osmotic medium pressure: 0–2 pulsing, osmotic medium temperature: 25°C, membrane type: AFC99 – overall thickness 400 μm.

Table 2

Values of viscosity of the used osmotic media, the corresponding driving force and flux

Osmotic medium	NaCl 22.24% (w/w)	CaCl ₂ 29.07% (w/w)	Ca(NO ₃) ₂ 29.00% (w/w)	PEG400 49.97% (v/v)	Sucrose 58.29% (w/w)	Glucose 62.86% (w/w)
Viscosity in cp (25°C)	1.9	2.3	2.9	14.0	40.5	103.0
$\Delta\Pi - \Delta P$ (bar)	280.3	548.0	176.3	303.0	134.8	337.5
Flux (kg/m ² h)	3.10	2.33	1.80	0.70	0.55	0.37

the resistance to mass transfer through the osmotic medium polarized layer (external polarisation) also implies higher diffusivity D according to the classical equation of Wilke and Chang and its correction for concentrated solutions [15]. This is expected to reduce the resistivity K of the solute in the porous membrane backing material [16] lowering, the so called, internal polarisation [17] and leading to higher osmotic fluxes. From the practical point of view the interpretation of the results, presented in Table 2, is that choosing of osmotic medium, to be used in direct osmosis applications, is based mainly on its mass transport properties (viscosity, diffusivity).

3.2. The tomato juice direct osmosis flux in relation to the osmotic medium concentration

The effect of the osmotic medium concentration on the direct osmosis flux was investigated by using two

different membranes, the AFC99 with a thickness of 600 μm and the AFC99 with a thickness of 400 μm . In both cases the osmotic medium used was sodium chloride brine and the experimental conditions practically the same (the relevant values are presented in Table 1). In both cases the flux value was calculated at five respective brine concentrations. Data for the dilution of the brine (5 h run) along with flux values in both cases are presented in Table 3.

After plotting the calculated flux values against average brine concentration in both cases (Fig. 3), a linear correlation between these two parameters was disclosed, with the flux increasing in accordance with the brine concentration. After this, the calculation of the overall mass transfer coefficient (flux/driving force) was carried out by calculating the driving force as the difference between the osmotic pressure of the brine (calculated according to Perry and Chilton [18]) and the osmotic pressure of the tomato juice

Table 3

Data for the dilution of the osmotic brine and calculated osmotic flux values in direct osmosis concentration of tomato juice with AFC99 (600 μm) and AFC99 (400 μm) membranes at different brine concentrations

Experiment no.	Initial brine quantity (g)	Initial brine concentration (% NaCl)	Final brine quantity (g)	Final brine concentration (% NaCl)	Average brine concentration (% NaCl)	Calculated direct osmosis flux (kg/m ² h)
<i>Membrane AFC99 600 μm overall thickness</i>						
1	11 788	5.96	12 000	5.85	5.91	0.446
2	11 782	9.95	12 011	9.76	9.86	0.460
3	11 782	14.96	12 056	14.62	14.79	0.560
4	11 802	19.63	12 110	19.13	19.38	0.620
5	11 767	23.33	12 083	22.72	23.02	0.640
<i>Membrane AFC99 400 μm overall thickness</i>						
1	11 796	5.83	12 595	5.46	5.65	1.61
2	11 782	9.59	12 695	8.90	9.25	1.85
3	11 782	14.56	12 889	13.31	13.94	2.24
4	11 796	19.47	13 072	17.57	18.52	2.58
5	11 778	23.59	13 300	20.89	22.24	3.10

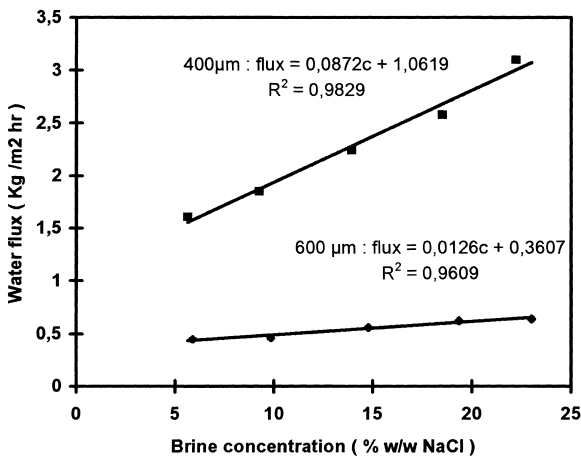


Fig. 3. Correlation between osmotic medium concentration and direct osmotic flux in tomato juice concentration by direct osmosis.

(calculated according to [14]) adding a 2 atm element to take into account the average hydraulic overpressure of the tomato juice. These values of the mass transfer coefficient were then plotted against the average brine concentration and an exponential reduction of this coefficient was revealed as the brine concentration was increased (Fig. 4). Also, a remarkable closeness of the exponent to -1 in both cases indicates that the mass transfer coefficient is inversely proportional to the brine concentration. This sharp

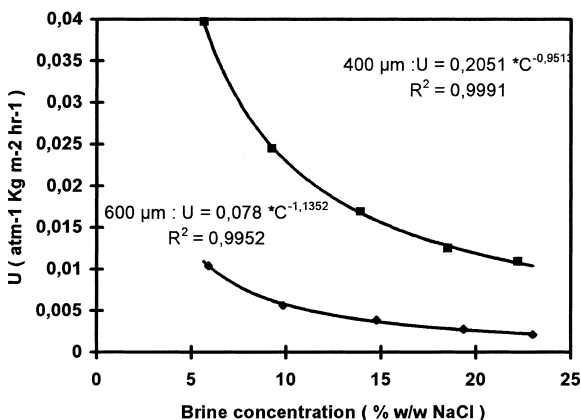


Fig. 4. Correlation between osmotic medium concentration and overall mass transfer coefficient in tomato juice concentration by direct osmosis.

reduction of the overall mass transfer coefficient with increasing brine concentration was further analysed. As there was no difference between the tomato solutions used in the respective experiments and the used hydrodynamic conditions of flow in each set of five experiments were the same for both fluids (tomato juice and brine), the observed reduction can be attributed to changes on the physical properties of the brine affecting the resistance of the brine side film coefficient and potentially the diffusion through the membrane backing material. However, data taken from [15] for the diffusion coefficient of water in sodium chloride solutions indicate rather an increase than a reduction of this coefficient over the range of brine concentrations used in this specific experimentation. In addition the use of the same membrane in each one of the five respective experiments with different concentrations of osmotic brine makes obvious that the resistance in water transfer through the skin of the membrane was the same in any case, as well as the thickness, tortuosity τ and porosity ϵ of the backing material (porous matrix supporting the membrane). Therefore following the analysis given in a series of literature references [16,17,19] along with the resistance to mass transfer exerted by the membrane skin, the resistivity $K = \tau t / D_s \epsilon$ of the backing material to water transfer should be roughly the same in all five experiments given that D_s appears fairly constant with concentration, if not increasing at higher salt concentrations [15]. This was verified by calculating the resistivity K , using a simplified version of an equation proposed by Loeb et al. [16]

$$K = (1/\text{flux}) \ln(\Pi_{\text{hi}}/\Pi_{\text{low}}), \quad (1)$$

where K is the resistivity of membrane support layer ($\text{m}^2 \text{h/kg}$), flux the water flux through the membrane ($\text{kg/m}^2 \text{h}$), Π_{hi} the osmotic pressure of osmotic brine (bar) and Π_{low} is the osmotic pressure of tomato juice (bar).

This calculation is presented in Table 4 and from the obtained values of K it is apparent that this is fairly constant along the whole range of the tested concentrations of brine. From this analysis, it has to be eliminated the possibility that the physical properties of the osmotic brine are affecting the mass transfer resistance of water through the membrane and consequently the overall mass transfer coefficient U . This means, in turn, that the dependence of the overall mass

Table 4
Calculation of the resistivity K at various brine concentrations

Average brine concentration (% NaCl)	Π_{hi} (bar) ^a	Π_{low} (bar) ^b	FLUX (kg/m ² h) (data from Table 3)	K (m ² h/kg) from Eq. (1)
5.65	48.0	8.7	1.61	1.06
9.25	84.5	8.7	1.85	1.23
13.94	142.6	8.7	2.24	1.25
18.52	216.1	8.7	2.58	1.25
22.24	293.5	8.7	3.10	1.14

^aData calculated from [18].

^bRef. [14].

transfer coefficient on the osmotic brine concentration is solely created by the influence of the osmotic brine physical properties on the film mass transfer coefficient at the side of the brine. However, from the three physical properties (density, diffusivity and viscosity) which are expected to be changed with concentration, the density of the sodium chloride solution undergoes only a small change over the tested range of concentrations and the diffusivity D remains practically constant, thus leading to the conclusion that the significant parameter for the reduction of U coefficient is the viscosity of the osmotic solution. This physical property increases by approximately 60% over the tested range of concentrations. Observing the curves of Fig. 4 a sharper reduction of the U coefficient with the thinner membrane (400 μm) is concluded. This can be better visualised by calculating the first order derivative of U versus C , which is $dU/dC = -0.195/C^{1.9513}$ for the 400 μm membrane and $dU/dC = -0.089/C^{2.1352}$ for the 600 μm membrane. The coefficient (-0.195) for the 400 μm membrane is more than double the coefficient (-0.089) in the case of the 600 μm membrane. This marks a more substantial contribution of the osmotic medium film coefficient in the formation of the overall mass transfer coefficient in the case of a thinner membrane, because of the reduction of the membrane resistance due to its lower thickness. The higher limit of tried concentration of NaCl brine was approximately 23% (w/w) NaCl. This concentration, which was close to the one of the saturated solution, gave the optimum performance due to the highest osmotic strength of the osmotic medium. This is also the higher concentration limit which can be achieved by electrodialysis, a method cheaper than conventional evaporation and also

well-known, concerning the reconcentration of the osmotic medium [20].

3.3. The effect of temperature on the direct osmosis flux

The effect of temperature on the direct osmosis flux was investigated in a course of five respective experiments which were conducted at five different temperatures (approximately 26°C, 36°C, 46°C, 52°C, 58°C respectively) within the range between 26°C and 60°C. The experimental conditions for these five experiments were: juice flow rate: 506 \pm 4 l/h, brine flow rate: 565 \pm 5 l/h, tomato juice pressure: 2.95 \pm 0.1 bar, brine pressure: 0–2 bar pulsing, initial brine concentration: 23% (w/w) NaCl, membrane type: AFC99 (500 μm).

The calculated values of the direct osmosis flux, from the osmotic brine dilution, are graphically represented versus juice temperature in Fig. 5. From this graph, it is obvious that an increase to the temperature caused an enhancement to the direct osmosis performance. This enhancement is expressed quantitatively by a 64% increase in terms of permeation flux as the temperature rises from 26°C to 60°C. It must be considered, however, that a fluctuation of the average brine concentration of less than 0.4% between respective runs, introduces a small error in calculating the flux value which, being less than 2%, was not corrected. Wrolstad et al. [21] reported the same positive effect of temperature on the direct osmosis flux in the case of concentration of raspberry juice but in their case only two temperature settings were tested (17°C and 27°C). Beaudry and Lampi [3] speculated on the same dependence of direct osmosis flux on

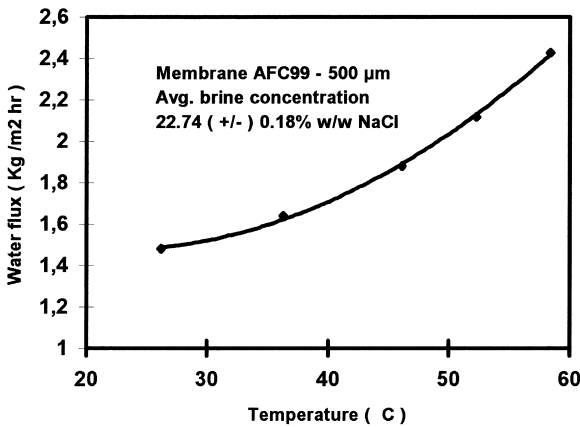


Fig. 5. The effect of temperature on the tomato juice direct osmosis flux.

temperature without, however, providing data to support their claim. According to Beaudry and Lampi [3] the increase of temperature lowers the viscosity and increases the diffusion coefficients of the fluids involved in the process, thus positively affecting the permeation flux. However, an attempt to fit the direct osmosis flux–temperature data to the well-known Arrhenius form of dependence which is valid in other membrane processes was not successful. Despite the fact that the temperature improved the performance of the direct osmosis concentration process, this positive element must be balanced with the negative effect which appears when the tomato juice is exposed at temperature for a considerable time. However, it must be noted that with conventional evaporative concentrators operating under vacuum the temperature of the product approaches 72–75°C, a value far greater than the higher tested value in this study (approximately 60°C).

3.4. The effect of tomato juice flow rate on the direct osmosis concentration flux

The effect of the tomato juice flow rate on the direct osmosis concentration flux was disclosed in a course of five respective experiments conducted at juice flow rates of 109, 216, 317, 419 and 502 l/h, respectively. The rest of the experimental conditions were: juice temperature: 25.5±0.5°C, juice pressure: 2.95±0.5 bar, brine flow rate: 560±28 l/h, brine pressure: 0–2 bar pulsing, and membrane type: AFC99 (500 µm).

The brine dilution data for these experiments are presented in Table 5 along with the calculated values of the permeation flux. From the calculated values of the permeation flux, which are presented against tomato juice flow rate in Table 5, it can easily be concluded that the effect of the juice flow rate on the direct osmosis flux is very smooth as an almost five-fold increase in flow rate (from 109 l/h up to 502 l/h) caused only a 32% increase of permeation flux. It must be noticed, however, that the average brine concentration between runs presents a small fluctuation which was not considered significant (Table 5). According to Beaudry and Lampi [3], the flow rate of the liquid to be concentrated by direct osmosis affects the resistance to mass transfer of the polarised layer adjacent to the membrane surface and consequently the value of the permeation. However, Merlo et al. [10] as well as Ishii et al. [6] and Pepper et al. [8] in their study on reverse osmosis concentration of tomato juice and in alignment with this present study, did not find any significant correlation between flux and juice flow rate. This similarity between the results obtained by Merlo et al. [10] and the ones obtained in this present investigation can be explained because, despite the

Table 5

Experimental data for the dilution of the osmotic brine (NaCl) for each one of the five experimental runs conducted to reveal the effect of the direct osmosis flux on the tomato juice flow rate

Experiment no.	Juice flow rate (l/h)	Initial brine quantity (g)	Initial brine concentration (% NaCl)	Final brine quantity (g)	Final brine concentration (% NaCl)	Average brine concentration (% NaCl)	Calculated direct osmosis flux (kg/m ² h)
1	109	11 783	23.56	12 338	22.50	23.03	1.12
2	216	11 770	23.48	12 359	22.36	22.92	1.19
3	317	11 780	23.70	11 951	23.36	23.53	1.38
4	419	11 784	23.62	12 437	22.38	23.00	1.32
5	502	11 798	23.61	12 530	22.23	22.92	1.48

fact that the two processes (reverse osmosis and direct osmosis) are different to each other, the nature of the two boundary layers is basically the same (same kind of liquid). This strange behaviour of the tomato juice, according to Pepper et al. [8] can be attributed to the rheological nature of the juice along with a theory known as “tubular pinch effect” which applies to colloidal suspensions [22]. As the tomato juice has got a considerable amount of pulp in colloidal suspension as well as certain macromolecules, this smooth increase of the direct osmosis flux with flow rate can also be explained by reference to this phenomenon.

3.5. Correlation between the tomato juice concentration and direct osmosis flux

The relationship between the direct osmosis flux and the concentration of the tested tomato juice was investigated using five respective values of juice concentration within the range 4.3–11.7° Brix. The investigation was limited to a maximum of 11.7° Brix due to problems of using higher concentration of high viscosity because of the unsupported membranes. The membrane which was used was the AFC99 at 500 μm and the osmotic medium was brine sodium chloride at 23% (w/w) NaCl. The temperature was 26°C for the tomato juice and 25°C for the brine. By observing the calculated flux values against the concentration of the tomato juice a substantial reduction of the flux value with increasing juice concentration was concluded. This reduction appears to be almost linear and reflects both the reduction of the driving force due to higher juice osmotic pressure at high concentrations and the reduction of the overall mass transfer coefficient due to increasing juice viscosity at higher juice concentrations. The linear formula relating the direct osmotic flux to juice concentration was found to be:

$$\text{flux} = -0.0989 \times (\text{juice concentration in } ^\circ \text{ brix}) + 1.8884, \quad R^2 \cong 0.99.$$

By calculating the overall mass transfer coefficient U , as the ratio of observed flux value divided by the difference in osmotic pressure between the osmotic medium and tomato juice and graphically representing the U -value against the corresponding average juice concentration the conclusion was a linear reduction of

the overall mass transfer coefficient with juice concentration. More specifically a 2.7-fold increase of tomato juice concentration (from 4.3 to 11.7° Brix) depressed the U -value to 53% of its initial value for juice at 4.3° Brix. The values for the osmotic pressure of tomato juice were calculated using the equation proposed by Dale et al. [14] while values for osmotic brines of sodium chloride were obtained from [18]. This significant reduction of flux observed with higher juice concentrations presents the direct osmosis concentration process as rather impractical where very thick and viscous juice is concerned.

3.6. The effect of the membrane thickness on the performance of the direct osmosis process

Three membranes having the same active layer and different thicknesses of backing material were used in order to ascertain if there is any effect of the membrane thickness on the performance of the direct osmosis. The overall thickness of these membranes were 600, 500 and 400 μm . The osmotic medium was NaCl brine (approximately 23%) in all cases and the experimental conditions for the three respective experiments were: juice temperature, 26±0.5°C, juice flow rate, 510±2% l/h, juice pressure, 2.95±0.1 bar, brine temperature, 25°C, brine flow rate 562±1%, brine pressure 0–2 bar pulsing. In Fig. 6 is illustrated the concentration reduction of the osmotic brine over the period of five hours for each one of the three

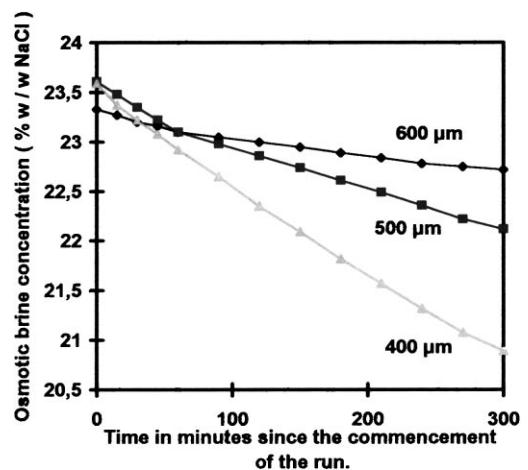


Fig. 6. The concentration reduction of the osmotic brine (NaCl) over the five hour period for three different membrane thicknesses.

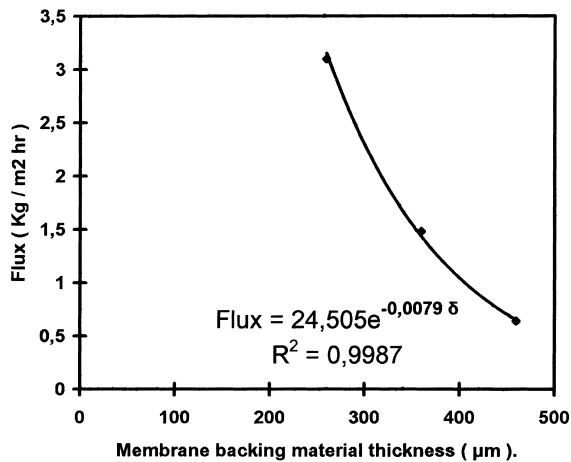


Fig. 7. Correlation of the tomato juice direct osmosis flux with the membrane backing material thickness.

membranes. From the slope of the three curves, a faster reduction of the brine concentration versus time as we shift from the 600 to the 400 µm membrane is apparent. Also, a graphical representation of the calculated flux values against membrane backing material thickness, illustrated an exponential trend for flux increase as the thickness was reduced, leading to a smaller overall membrane thickness with the active membrane layer being the same (Fig. 7). Unfortunately, a refusal of the membrane manufacturer to provide even thinner novel membranes did not allow investigation into whether this trend is consistent at yet lower membrane thicknesses. It is also worth reporting that such a trend is not in agreement with the claim by Beaudry and Lampi [3] that the direct osmosis flux is linearly correlated with membrane thickness. However, the general conclusion of Herron et al. [4] that the thickness of the membrane backing material can dramatically impair the direct osmosis flux was found fully aligned with the experimental evidence of the present investigation. Similar effect of the thickness of membrane backing material on the obtained direct osmosis flux was described by Loeb et al. [16] who, studying non-food systems, concluded that: clearly, the porous fabric of the membranes they used decreased the osmotic permeation excessively moreover, a separate investigation on the same matter, involving thinner flat membranes is under way at

the moment, and is expected to elucidate the exact form of dependence between direct osmosis flux and membrane thickness.

4. Conclusions

Several parameters believed to affect the performance of the operation of direct osmosis concentration of tomato juice were studied in the course of the present investigation. Initially, a comparison carried out by using six different osmotic media, including sodium chloride brine, calcium chloride brine, calcium nitrate brine, sucrose solution, glucose solution and polyethylene glycol 400 solution, disclosed the most effective among them was the sodium chloride solution. Additionally, in the present experiments, the measured osmotic flux values with different osmotic media were not in correlation to the corresponding overall osmotic pressure difference but to the viscosity of the osmotic medium used each time. Accordingly, this was considered to be clear evidence of the importance of using low viscosity osmotic media in direct osmotic applications. Also, the increase of the osmotic medium concentration, despite the fact it resulted in higher osmotic fluxes due to increased osmotic pressure at high concentrations, led to a reduced overall mass transfer coefficient. This overall mass transfer coefficient was found to be inversely proportional to the osmotic medium concentration and the parameter which was responsible for this particular relationship was identified to be the viscosity of the osmotic medium. Concerning juice temperature, as it was rising, exhibited a positive effect on the direct osmosis concentration flux. However, the well-known and anticipated Arrhenius relationship, which exists between flux and temperature in other membrane processes was not found to be valid in this present case. On the other hand, the juice flow rate did not substantially affect the direct osmosis flux and this was attributed to the rheological particularities of the tomato juice which caused a phenomenon previously identified as the tubular pinch effect. Regarding the increase of the tomato juice concentration, this had a reducing effect on the direct osmosis flux. The increasing juice osmotic pressure at higher concentration along with the changes of the physical properties of the juice (higher viscosities and lower diffusivities at

high concentration) both contributed to the observed reduction. Finally, a parameter of paramount importance for the operation of direct osmosis concentration was proved to be the overall thickness of the used membrane, as a trend of an exponential increase of flux with reducing overall thickness was revealed for a certain range of membrane thicknesses. However, additional work is under way in order to ascertain whether this trend is consistent at even lower membrane thicknesses.

Acknowledgements

The authors of this paper wish to express their sincere thanks to the European Union which kindly funded this research through the AIR programme. The contribution of Dr. A. Merry and Mr. A. Nightingale from PCI UK company is also acknowledged by the authors.

Appendix

Spreadsheets for the figures

Fig. 2

Viscosity (C_p)	Flux ($\text{kg}/\text{m}^2 \text{ h}$)
1.9	3.1
2.3	2.33
2.9	1.8
14	0.7
40.5	0.55
103	0.37

Fig. 3

Concentration	Flux 600	Flux 400
5.65		1.61
5.91	0.446	
9.25		1.85
9.86	0.46	
13.94		2.24
14.79	0.56	
18.52		2.58
19.38	0.62	
22.24		3.1
23.02	0.64	

Fig. 4

% NaCl	U (600)	U (400)
5.65		3.97E-02
5.91	1.04E-02	
9.25		2.45E-02
9.86	5.60E-03	
13.94		1.69E-02
14.79	3.87E-03	
18.52		1.25E-02
19.38	2.78E-03	
22.24		1.09E-02
23.02	2.11E-03	

Fig. 5

Temperature ($^{\circ}\text{C}$)	Water flux ($\text{kg}/\text{m}^2 \text{ h}$)
26.2	1.48
36.3	1.64
46.1	1.88
52.3	2.12
58.4	2.43

Fig. 6

Time (min)	600 μm	500 μm	400 μm
0	23.33	23.61	23.59
15	23.27	23.48	23.37
30	23.2	23.35	23.22
45	23.16	23.22	23.08
60	23.1	23.1	22.92
90	23.05	22.98	22.65
120	23	22.86	22.35
150	22.95	22.74	22.09
180	22.89	22.61	21.82
210	22.84	22.49	21.57
240	22.78	22.36	21.32
270	22.75	22.22	21.08
300	22.72	22.12	20.89

Fig. 7

Membrane backing material thickness	Flux
260	3.1
360	1.48
460	0.64

References

- [1] K. Popper, W.M. Camirand, F. Nury, W.L. Stanley, Dialyzer concentrates beverages, *Food Eng.* 38(4) (1966) 102–104.

- [2] E.G. Beaudry, K.A. Lampi, Membrane technology for direct-osmosis concentration of fruit juices, *Food Technol.* 44(6) (1990) 121.
- [3] E.G. Beaudry, K.A. Lampi, Osmotic concentration of fruit juices, *Flussiges Obst.* 57 (1990) 652–656; 663–664.
- [4] J.R. Herron, E.G. Beaudry, C.E. Jochums, L.E. Medina, US Patent 5 281 430 (1994).
- [5] R.L. Merson, G. Paredes, D.B. Hosaka, Concentrating fruit juices by reverse osmosis, in: *Ultrafiltration Membranes and Applications*, Plenum Press, New York, 1980, p. 405.
- [6] K. Ishii, S. Konomi, K. Kojima, M. Kai, Development of a tomato juice concentration system by reverse osmosis, in: A.F. Turbak (Ed.), *Synthetic Membranes: vol. II – Hyperfiltration and Ultrafiltration Uses*, ACS Symposium Series, vol. 154, ACS, Washington, DC, 1981, p. 1.
- [7] A. Watanabe, T. Ohtani, S. Kimura, Performance of dynamically formed Zr(IV)-PAA membrane during concentration of tomato juice, *Nippon Nogeikagaku Kaishi* 56(5) (1982) 339.
- [8] D. Pepper, A.C.J. Orchard, A.J. Merry, Concentration of tomato juice and other fruit juices by reverse osmosis, *Desalination* 53 (1985) 157–166.
- [9] S. Gherardi, R. Bazzarini, A. Trifiro, A. Lo Voi, D. Palamas, Pre-concentration of tomato juice by reverse osmosis, *Industria Conserve* 61(2) (1986) 115–119.
- [10] C.A. Merlo, W.W. Rose, L.D. Pedersen, E.M. White, Hyperfiltration of tomato juice during long term high temperature testing, *J. Food Sci.* 51(2) (1986) 395–398.
- [11] C.A. Merlo, W.W. Rose, L.D. Pedersen, E.M. White, J.A. Nicholson, Hyperfiltration of tomato juice: pilot plant scale high temperature testing, *J. Food Sci.* 51(2) (1986) 403–407.
- [12] S. Cross, 1988. Achieving 60° Brix with membrane technology, Paper presented at the Annual Meeting, Institute of Food Technologists, New Orleans, LA, 19–22 June 1988.
- [13] PCI, Personal communication (1998).
- [14] M.C. Dale, M.R. Okos, P. Nelson, Concentration of tomato products: analysis of energy saving process alternatives, *J. Food Sci.* 47(6) (1982) 1853–1858.
- [15] R.E. Treybal, *Mass Transfer Operations*, 3rd ed., McGraw-Hill Chemical Engineering Series, Tokyo, Japan, 1982, p. 36.
- [16] S. Loeb, L. Titelman, E. Korngold, J. Freiman, Effect of porous support fabric on osmosis through a Loeb–Sourirajan type asymmetric membrane, *J. Membr. Sci.* 129 (1997) 243–249.
- [17] K.L. Lee, R.W. Baker, H.K. Lonsdale, Membranes for power generation by pressure – retarded osmosis, *J. Membr. Sci.* 8 (1981) 141–172.
- [18] R.H. Perry, C.H. Chilton, *Chemical Engineer’s Handbook*, 5th ed., McGraw-Hill Chemical Engineering Series, Tokyo, Japan, p. 17–41.
- [19] S. Loeb, Pressure-retarded osmosis revisited: the prospects for osmotic power at the Dead Sea, in: *Proceedings of the Euromembrane 1995*, vol. 2, 1995 II-1 to II-11.
- [20] R.E. Lacey, S. Loeb, *Industrial Processing with Membranes*, Robert E. Krieger, Huntington, NY, pp. 300–310.
- [21] R.E. Wroldstad, M.R. McDaniel, R.W. Durst, N. Micheals, K.A. Lampi, E.G. Beaudry, Composition and sensory characterization of red raspberry juice concentrated by direct-osmosis or evaporation, *J. Food Sci.* 58(3) (1993) 633–637.
- [22] M. Cheryan, *Ultrafiltration Handbook*, Technomic, Lancaster, PA, 1985, pp. 118, 192.
- [23] A. Apelblat, Activity and osmotic coefficients in electrolyte solutions at elevated temperatures, *AIChE J.* 39(5) (1993) 918–923.
- [24] R.N. Goldberg, R.L. Nutall, Evaluated activity and osmotic coefficients for aqueous solutions: the alkaline earth metal halides, *J. Phys. Chem. Ref. Data* 7(1) (1978) 291.
- [25] N.P. Money, Osmotic pressure of aqueous polyethylene glycols. Relationship between molecular weight and vapor pressure deficit, *Plant Physiol.* 91 (1989) 766–769.
- [26] R.C. Weast, *Handbook of Chemistry and Physics*, 55th ed., CRC Press, New York, 1974, pp. D-215, D-231, E-1.