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Proportionality of solute flux and permeability to water flux and permeability in forward osmosis process

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ABSTRACT

Several forward osmosis (FO) experiments were carried out using different concentrations of sodium chloride (NaCl) aqueous solutions as draw solution (DS) and freshwater as feed solution. The experiments were carried out at different temperatures using a symmetric cellulosic membrane. The study investigates the relationship between water flux and DS solute flux (the reverse solute diffusion) across the membrane. The solute flux is one of the important factors that determine the practicality of FO processes. The results show that both water flux and solute flux increase as the solute concentration difference across the membrane increases, as well as with the temperature increase. It is found that water flux is proportional to solute flux, and a similar trend was also found between water permeability and solute permeability. Proportionality factors (k_J) and (k_E) are suggested to represent the flux and the permeability proportionalities, respectively. The k_J was calculated experimentally and found to be almost constant and approaching the saturation concentration of the solute, NaCl in this case.

Keywords: Forward osmosis; Membranes; Water treatment; Desalination

1. Introduction

Clean water and renewable energy are major concerns worldwide due to water scarcity, the environmental impacts of energy use, pollution, and resource depletion. As an indication of the importance of water as a global resource, it is estimated that about 3.5 million people die annually from the shortage of water supply and sanitation [1]. Separation of fresh water from saline or wastewater by desalination processes of seawater and treatment of industrial or municipal wastewater, appear to be as the only solution for this problem. The cost of refining and separations represents a large portion of the total production cost. Thus, selection of an appropriate separation technology is essential for economic effectiveness and feasibility. Membrane separation processes play a growing part in applications such as seawater desalination, industrial and municipal wastewater treatment, gas separation, mining and petroleum industries, and biomedical engineering. Membrane processes are attractive due to their simplicity (no phase change), applications diversity at the ambient temperature, and the relatively low energy needs, which have a major impact on greenhouse gases reduction. Although the operational cost for these processes is considerably low, the capital cost largely depends on the unit specific productivity, which is normally determined by a critical process parameter, the system permeability to the solvent (water); as the permeability increases, the required membrane area, consequently the unit size and cost, decreases.

Forward osmosis (FO), which is the engineered application of the natural phenomenon of osmosis, if cleverly implemented, is one of the unique and potentially promising

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membrane separation technologies that can provide the solution to the global need for affordable clean water and inexpensive sustainable energy. In FO processes, water is naturally transferred from a feed solution (e.g., seawater, wastewater) towards a draw solution (e.g., concentrated brine), across a semi-permeable membrane, driven by a net differential pressure (NDP) obtained from the osmotic pressure difference. Despite the increasing interest in FO industrial applications, there are some technological barriers that have yet to be overcome in order to achieve efficient and high productivity processes. Improving these processes can be achieved by increasing the permeability of water. The water permeability depends on the membrane microstructure, the physical properties of the solutions (feed and draw), the molecular properties of the solutes, and the operational conditions. Having a better understanding of these parameters will conquer the practical problems and open new horizons for future developments. This can be achieved after systematized experimental/theoretical studies of the mass transfer phenomena across the membrane in order to generate a predictive mechanistic understanding.

The principle of FO is currently utilised for some industrial purposes such as concentrating of fruit juice and dairy products and in some medical applications such as haemodialysis, where wastes and excess water are removed from the blood. It has also been used for pharmaceuticals dehydration and for controlled release drug delivery systems. The FO is increasingly applied in industrial wastewater treatment and seawater or brackish water desalination [2–4]. Other unique areas of FO current research include the pressure retarded osmosis (PRO) for electricity generation, which employs the salinity gradient between saline (e.g., seawater) and freshwater (e.g., river water) [5]. Several recent reports stated that the PRO is closer than ever to being a practical choice for renewable energy production [6–8]. Additionally, FO technology has a promising application to increase the availability of water for irrigation through extraction of freshwater from brackish or wastewater sources. In such applications, fertilizers can be used to prepare a concentrated draw solution (DS) and brackish groundwater or wastewater is used as feed water (FW). The diluted fertilizer solution is subsequently applied to plantings through a fertigation distribution network.

One of the major problems facing the FO process practicability is the need for high productivity membranes. The synthesis of high-performance FO membrane is still in the early stage of its development. An ideal FO membrane should possess high water flux as well as high solute rejection and good chemical stability. The commercial FO membranes are still limited in terms of both manufacturers and choice of membrane chemistries. The symmetric membranes, which are composed from one homogenous layer, outperform the asymmetric ones, as adding porous supports decreases water permeation excessively [9]; however, the current available FO membranes are composite (asymmetric) and made of a multi-layered structure (active skin, intermediate, and support layers). These composite membranes have been optimized in terms of the porous support layer, while the active skin tends to have low water permeability and limited solute retention. However, without the support layer the membrane becomes too weak, which means that there are still significant opportunities to further improve the FO membrane performance.

The academic interest in FO technology over the past two decades has increased resulting in enormous publications in this field [10]. The main trends of the current research in this field are membranes and modules development, investigations on new draw solution and regeneration processes, and optimizing the operational conditions. An ideal DS should provide high osmotic pressure, low viscosity, easy recovery, cost-effectiveness, non-toxicity, and safety [11]. The FO process alone is unable to directly produce pure water, as water should be regenerated from the diluted DS. The DS solute recovery, especially when prepared DS solutes are utilized, is a key factor affecting process feasibility. Additionally, the low specific energy consumption is a major consideration to select the DS and the type of the regeneration processes.

This study investigates the FO process using a dialysis membrane available in the market having an average pore diameter of about 1.8 nm. This membrane is symmetric composed of one active layer. Symmetric membranes do not endure internal solute concentration polarization, which may occur in the substrate layer of asymmetric membranes [9,12]. This study attempts to provide better understanding of the relationship between water flux and solute flux, and between water permeability and solute permeability, at different DS concentrations and operational temperatures. The experimental work of this study utilizes sodium chloride (NaCl) aqueous solutions as DS, while freshwater as FW.

1.1. Theory of FO

Osmosis is a natural phenomenon by which water is transferred across a semi-permeable membrane from a feed water (FW) having low osmotic pressure (low solute concentration, such as wastewater) towards a draw solution (DS) having high osmotic pressure (high solute concentration, such as brine or seawater).

The FO process can be schematically represented as shown in Fig. 1. In this process, water molecules (represented as continuous and dotted arrows) transfer across the membrane driven by the net differential pressure (NDP). The NDP is obtained from the osmotic pressure difference, ΔΠ, resulting from the solute concentration difference. Water can transfer through the membrane pores and by diffusion through the membrane material. This transfer is faced by another diffusion transport of solute molecules (represented as spheres) driven by the concentration difference [9,13].

At any steady state condition, where the DS and the FW are both assumed to be continuous phases, the value of water flux from the FW side towards the DS side has a corresponding reverse value of solute flux. This can be mathematically represented as follows:

$$J_s = k_J J_w \tag{1}$$

where J_s is the solute mass flux with units of, for example, g/m^2 h, and J_w is the volumetric water flux with units of, for example, L/m^2 h, while k_J is a flux proportionality factor.

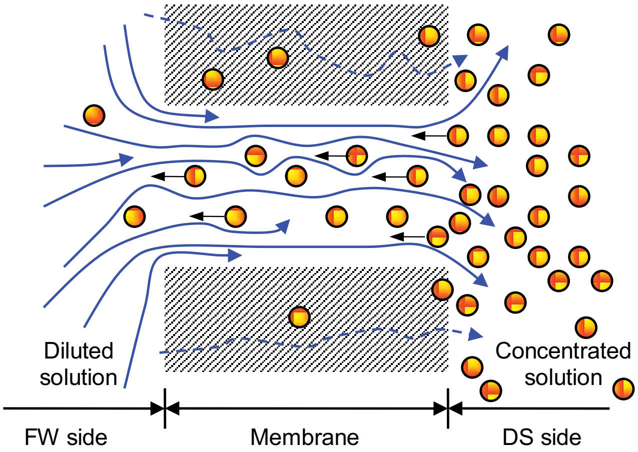

Fig. 1. Schematic representation for the FO process across a pore of a symmetric membrane using an aqueous solution as DS and pure water as FW.

The flux proportionality factor has units of concentration (e.g., g/L) and represents the extent of solute-water mutual transfer across the membrane. In pressure-driven membrane separation processes, such as reverse osmosis (RO), the factor k_J is the same as the solute concentration in the permeated fluid, while in FO it represents the solute concentration inside the membrane. Eq. (1) indicates that solute flux in FO process increases proportionally as water flux increases; this is supported by the experimental work of this study. From the parameter k_J, the amount of the solute transferring across the membrane can be estimated. For example, a value for k_J equal to 35 g/L indicates that a number of solute molecules equivalent to 35 g cross a specific membrane area at specific time upon a number of water molecules equivalent to one litre crossing the same area at the same period.

Water volumetric flux and solute mass flux can be defined using the following phenomenological relationships:

$$J_w = A_w\left(\Pi_{\mathrm{DS}} - \Pi_{\mathrm{FW}}\right) \tag{2}$$

The water flux is driven by the osmotic pressure difference across the membrane ($\Pi_{\mathrm{DS}} - \Pi_{\mathrm{FW}}$), where A_w is a water permeability coefficient (variable and depends on the system conditions) with units of, for example, L/m^2 h bar, and:

$$J_s = B_s\left(c_{\mathrm{DS}} - c_{\mathrm{FW}}\right) \tag{3}$$

The DS solute flux is driven by the concentration difference ($c_{\mathrm{DS}} - c_{\mathrm{FW}}$), where B_s is a DS solute permeability coefficient with units of m/h.

Hence, by substituting both Eqs. (3) and (2) into Eq. (1), the following relationship can be obtained:

$$B_s\left(c_{\mathrm{DS}} - c_{\mathrm{FW}}\right) = k_J A_w\left(\Pi_{\mathrm{DS}} - \Pi_{\mathrm{FW}}\right) \tag{4}$$

The osmotic pressure, Π, is the property of the solutions, which results from the dissolution of solutes in an aqueous solution. In physical terms, it is the hydrostatic pressure produced by a solution in a compartment separated by a semi-permeable membrane from another of lower concentration. Several relationships have been developed to predict the osmotic pressure of the solutions. One of the most commonly used formulas is the van't Hoff equation for ideal solutions [14]:

$$\Pi = i_v \frac{c}{M_{\mathrm{wt}}} R_g T \tag{5}$$

where i_v is the van't Hoff factor and refers to the number of moles of the dissociated entities when one mole of the solid solute is dissolved (e.g., for glucose $i_v = 1$ and for NaCl $i_v = 2$), c is the weight concentration of the solute, M_{wt} is the molecular weight, R_g is the universal gas constant, and T is the thermodynamic temperature (formally called absolute temperature).

By substituting Eq. (5) into Eq. (4), the following relationship can be obtained for the case of ideal binary solution on both sides of the membrane:

$$B_s\left(c_{\mathrm{DS}} - c_{\mathrm{FW}}\right) = k_J \frac{i_v R_g T}{M_{\mathrm{wt}}} A_w\left(c_{\mathrm{DS}} - c_{\mathrm{FW}}\right) \tag{6}$$

This can be simplified to the following:

$$B_s = k_E A_w \tag{7}$$

where:

$$k_E = k_J \frac{i_v R_g T}{M_{\mathrm{wt}}} \tag{8}$$

The new factor, k_E, is a permeability proportionality factor, and is a function of temperature has units of pressure (e.g., bar). Eq. (7) shows that solute permeability increases proportionally with water permeability, while Eq. (8) shows the effects of the molecular weight of the solute and the temperature on the factor k_E. For example, with low operational temperature and high molecular weight of the DS solute, a lower solute permeability can be obtained.

In spite of the simplicity of van't Hoff equation, it has a limited application when more precise results are needed. In ionic solutions, due to a phenomenon called ion pairing, a certain number of the positive and the negative ions will randomly come together and form ion pairs. This reduces the total number of free particles in solution, and consequently decreases the osmotic pressure from its ideally estimated value by Eq. (5). However, Eq. (6) can be developed further by incorporating osmotic coefficients that account for the deviation of the solvent from the ideal behaviour [15].

2. Experimental methods

The FO is investigated in this study using the set-up illustrated in Fig. 2. The osmotic cell is made of two PVC cubical shape (20 cm × 20 cm × 20 cm) compartments separated by a square flat sheet membrane. The surface area of the membrane is estimated to be 400 cm^2 (20 cm × 20 cm). The membrane is supported by a perforated stainless steel sheet (0.55 mm thick with 79% open

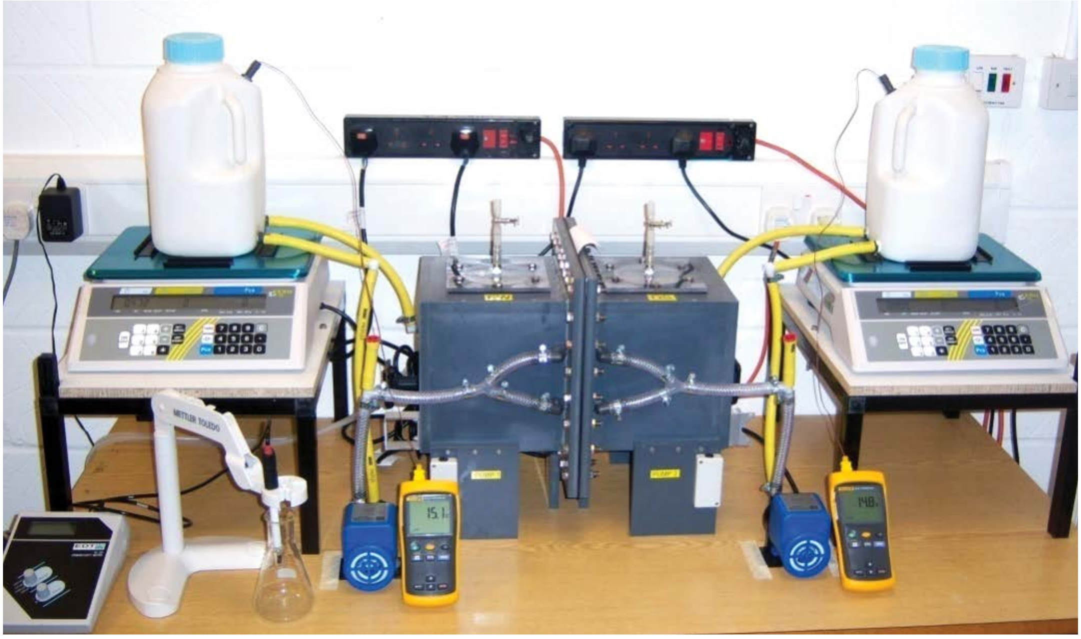

Fig. 2. The FO bench-scale experimental setup.

area) to prevent membrane bending. Each compartment is connected to separate plastic container placed over a digital scale. The total fluid volume capacity of each side is ~10 L when filled to a zero level, situated at the middle of each fluid container.

Measurements of solute concentrations were taken using an electrical-conductivity meter model SevenMulti manufactured by Mettler-Toledo (Switzerland). The corresponding osmotic pressure readings were calculated using stream analyzer software, OLI software, which predicts the properties of solutions via thermodynamic modeling based on experimental data.

Each compartment is equipped with an external small centrifugal pump providing good fluid inter-circulation (14 l/min) in order to minimise concentration polarization effects. The Reynolds number of the fluid flow inside the FO cell is estimated to be 1309, which refers to a laminar flow alongside the membrane. Flexible plastic pipes were used to connect all the cell components and pumps. The cell inputs and outputs are located near the membrane surface in order to get good horizontal cross-flow alongside the membrane. Furthermore, each compartment is equipped with an immerged stainless steel coil to circulate a cooling/heating media fed from a separate water bath circulator; this kept the DS and the FW temperature constant within ±0.5°C during experiment. At the start of each run, the system was thoroughly cleaned by circulating/flushing pure water for 1–2 h.

In practice, FO bench-scale experiments utilizing small area membranes require continuous circulation of the DS and the FW. With experiment progress, the collected DS is diluted and increases in volume, while the FW is concentrated and decreases in volume. The readings of weight and concentration were recorded as function of the experiment time. The experiment time in such bench-scale experiments is equivalent to the residence time in larger size modules. For example, a period of 300 min in this bench-scale study is equivalent to a residence time of 300 min in continuous operation FO module.

A dialysis cellulosic membrane-type Visking-DVT01350 supplied by Medicell International Ltd., (UK) was used to carry out the experiments. According to the manufacturer datasheet, this membrane is made of natural cellulose (cotton linters). It is fabricated by dissolving cellulose in special inorganic solvents, the polymer then reformed by taking away the solvent to form the membrane as a flat sheet. This membrane is highly resistant to organic solvents, elevated temperatures, and extremes of pH. The name code of this membrane refers to its molecular weight cut-off (MWCO). A membrane with MWCO of 1,350 Da is estimated to be having a mean pore diameter of 1.8 nm, according to the following relationship between the molecular weight and the molecular diameter [16]:

$$D_p = 0.066 M_{\mathrm{wt}}^{0.46} \tag{9}$$

where D_p is the approximate diameter of the molecule in nano-meters and M_{wt} is the molecular weight in g/mol.

3. Results and discussion

Several FO experiments were carried out using NaCl aqueous solution as DS and pure water as FW, (1) using four different DS concentrations at constant temperature of 25°C, and (2) using one DS concentration at three different temperatures of 13°C, 25°C, and 32°C.

3.1. Experiments at constant temperature

In this part of the experimental work, four runs were carried out, each using a specific DS concentration. The four initial DS solute concentration were symbolized as L (low), M (medium), H (high), and S (saturated). Due to the procedure of the initial setting, small quantities of the solute diffused to the FW side. The initial and final concentrations of the DS and FW are given in Table 1.

The results of water flux vs. the net differential pressure (NDP) are shown in Fig. 3. As predicted, water flux increases as the osmotic pressure and the concentration difference across the membrane increases.

In Fig. 4, the results of the solute flux are plotted as function of the NDP. Similar to water flux, solute flux or diffusivity increases as the NDP, or the concentration difference, increases.

In Fig. 5, solute flux is plotted against water flux, where their proportional relationship is shown; solute flux increases as water flux increases. The trend of the increase is found to be almost linear in the range of these experiments. This linear trend represents the flux proportionality factor, k_J, according to Eq. (1). Although, the calculated value of k_J was fluctuated between 200 and 500 g/L, due to the fluctuations in experimental readings, it is estimated as an average of 322.7 g/L. As mentioned earlier, the k_J represents the average concentration of the solute inside the membrane material and pores; it is found here to be near the saturation concentration of NaCl, which is about 362 g/L at 25°C.

The calculations of water permeability, A_w, by applying Eq. (2), are plotted in Fig. 6, as a function of the NDP. It shows that water permeability decreases as the NDP increases, in contrast to the trend of water flux. This can be explained by the decrease of the ability of water molecules to pass through membrane pores and among solute molecules and their hydration spheres in concentrated solutions, while water flux increases due to the increase of the osmotic driving force, the NDP.

Fig. 7 shows the solute permeability, B_s, as a function of the NDP, where B_s decreases as the NDP increases. This decrease of solute permeability in concentrated solutions can be explained by the decrease in the diffusivity or the ability of the solute molecules to move in concentrated solutions, while the increase of the solute flux across the membrane is due to the steeper concentration gradient between membrane sides.

By drawing B_s against A_w, as shown in Fig. 8, a certain proportionality trend is obtained representing the permeability proportionality factor, k_E. The results show that the solute permeability increases with water permeability, and decreases with solute concentration; at higher NDP, or higher solute concentration, is the lower solute permeability and water permeability.

3.2. Experiments at different temperatures

In this section, three FO experiments were carried out at three different temperatures, 13°C, 25°C and 32°C, using NaCl aqueous solution as DS and pure water as FW (the experiment for the 25°C is borrowed from the previous section). The DS and the FW in these experiments were at similar controlled temperatures. Similarly to the previous set, the FW side was contaminated initially with some solute diffusion. Table 2 shows a summary of the initial and the final concentrations of this part.

Fig. 9 shows water flux as function of the NDP. As expected, water flux increases as the temperature increases, due to, for example, lower solution viscosity, as well as with the NDP increase.

Fig. 10 shows solute flux across the membrane as a function of the NDP. It can be seen that solute flux, similar to water flux, increases as the temperature increases. It can be noted that the NDP values increase at higher temperatures; although, the concentration was similar in these experiments. This can be attributed to the increase n osmotic pressure with temperature increase.

Fig. 11 shows solute flux vs. water flux. The experimental results show a proportionality, although the fluctuations in experimental results, especially at higher temperature.

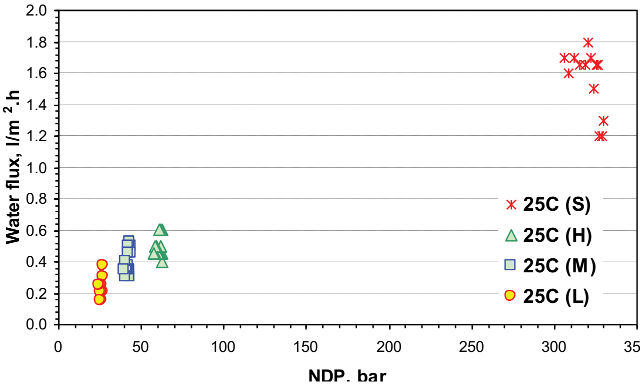

Fig. 3. Water flux as a function of NDP in several FO experiments at 25°C using different concentrations of NaCl aqueous solutions as DS.

Table 1
Initial and final conditions of the FO experiments at constant temperature of 25°C

Experiment symbol[a]	DS concentration, NaCl (g/L)		FW concentration, NaCl (g/L)		Experiment period (min)	DS initial osmotic pressure (bar)
	Initial	Final	Initial	Final		
L	35.48	33.78	1.180	2.575	300	28.0
M	54.98	52.20	0.683	2.946	300	44.3
H	77.30	74.02	1.660	4.950	300	63.8
S	306.35	291.60	0.600	8.550	240	330.3

[a] Symbols refer to DS initial concentration: L: Low; M: Medium; H: High; S: Saturated.

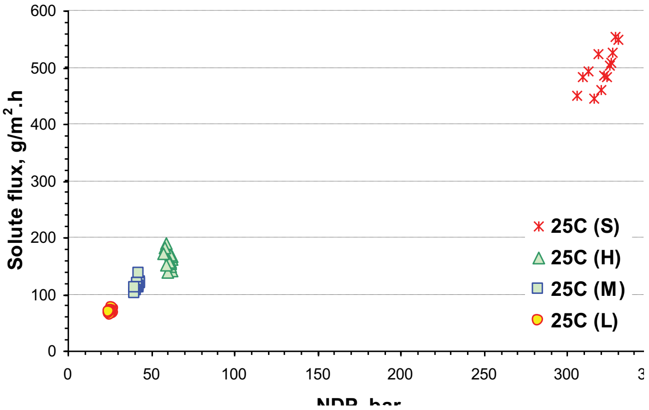

Fig. 4. Solute flux as a function of NDP in several FO experiments at 25°C using different concentrations of NaCl aqueous solutions as DS.

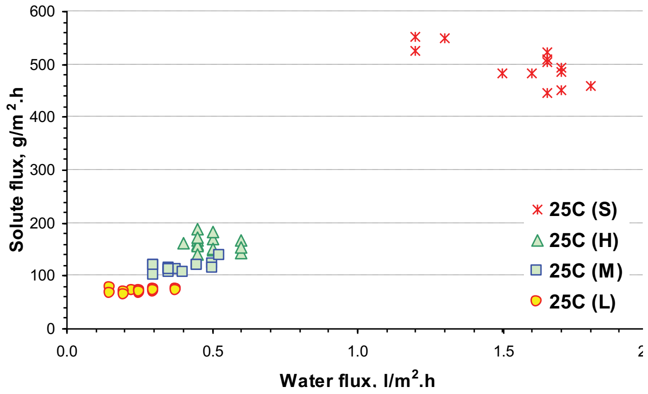

Fig. 5. Solute flux as a function of water flux in several FO experiments at 25°C using different concentrations of NaCl aqueous solutions as DS.

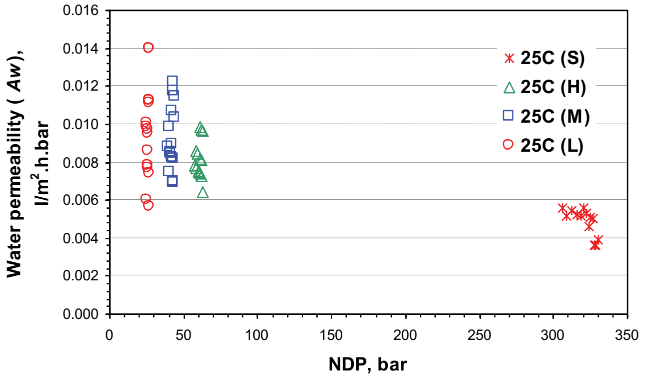

Fig. 6. Water permeability as a function of the NDP in several FO experiments at 25°C using different concentrations of NaCl aqueous solutions as DS.

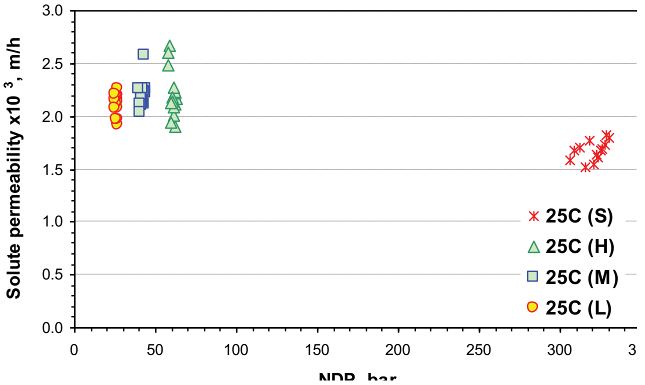

Fig. 7. Solute permeability as a function of NDP in several FO experiments at 25°C using different concentrations of NaCl aqueous solutions as DS.

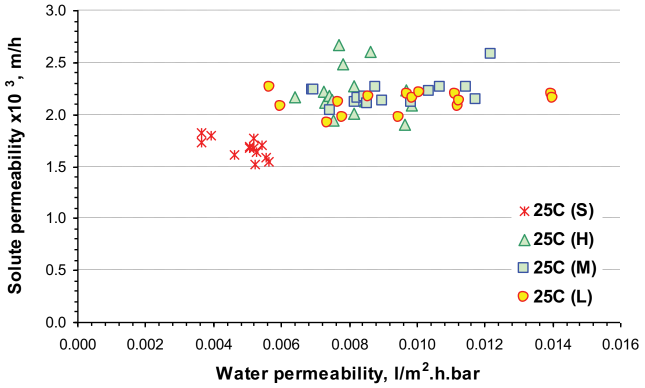

Fig. 8. Solute permeability as a function of water permeability in several FO experiments at 25°C using different concentrations of NaCl aqueous solutions as DS.

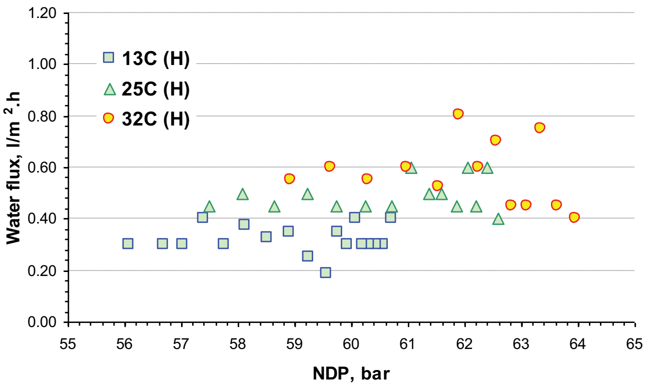

Fig. 9. Water flux as a function of the NDP in several FO experiments at different temperatures using NaCl aqueous solutions as DS.

Fig. 12 shows water permeability as a function of the NDP. It can be concluded that the water permeability increases as the operational temperature of the FO process increases. This can be explained by the decrease of the solution viscosity at higher temperatures, which accordingly results in an increase in water permeability.

The solute permeability, which is the product of dividing solute flux by the concentration difference, is plotted in Fig. 13 as a function of NDP and in Fig. 14 as a function

Table 2
Initial and final conditions of the FO experiments at different temperatures

Experiment temperature	DS concentration, NaCl (g/L)		FW concentration, NaCl (g/L)		Experiment period (min)	DS Initial osmotic pressure (bar)
	Initial	Final	Initial	Final		
13°C (H)	77.48	74.30	1.746	4.880	420	62.0
25°C (H)[a]	77.30	74.02	1.660	4.950	300	63.8
32°C (H)	77.45	74.22	1.550	4.700	240	65.2

[a] The letter H refers to that we used similar initial concentration for the DS as in experiment H of Table 1.

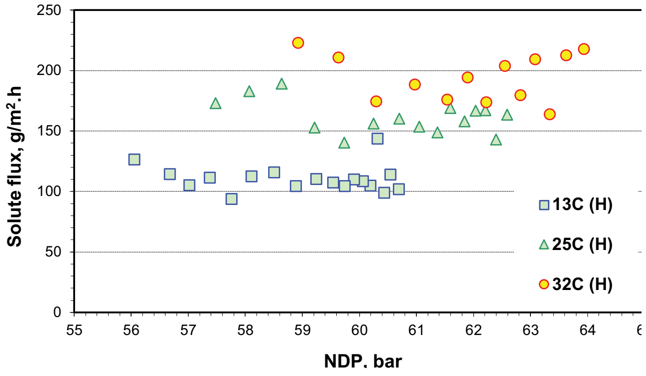

Fig. 10. Solute flux as a function of NDP in several FO experiments at different temperatures using NaCl aqueous solutions as DS.

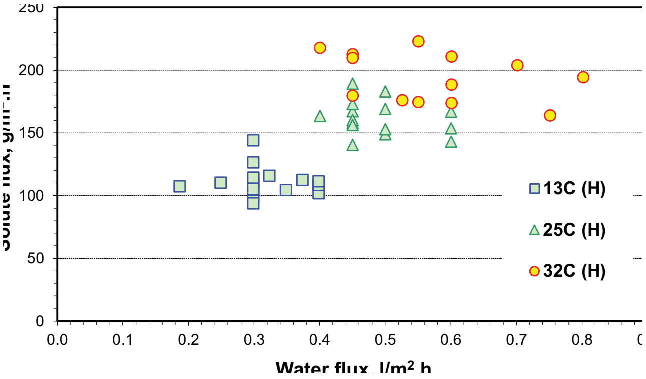

Fig. 11. Solute flux as a function of water flux in several FO experiments at different temperatures using NaCl aqueous solutions as DS.

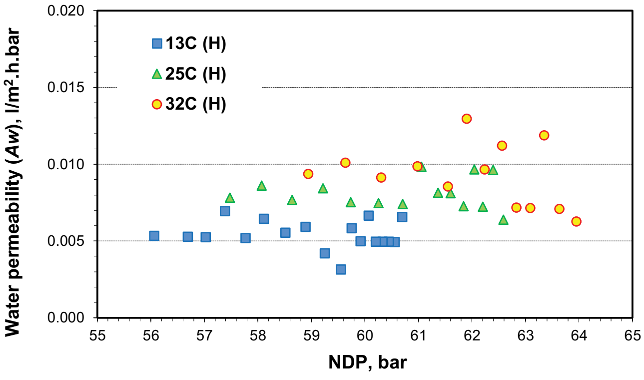

Fig. 12. Water permeability as a function of the NDP in several FO experiments at different temperatures using NaCl aqueous solutions as DS.

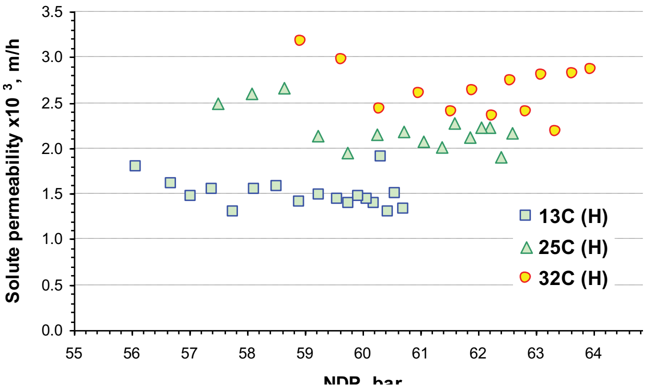

Fig. 13. Solute permeability as a function of NDP in several FO experiments at different temperatures using NaCl aqueous solutions as DS.

of water permeability. The solute permeability increases as the temperature increases, and in a proportionality with water permeability. The relationship between solute permeability and water permeability is characterized by the factor k_E, as described in Eqs. (7) and (8).

4. Conclusions

Several FO experiments were carried out using different concentrations of NaCl aqueous solutions as DS and freshwater as FW. A first part of experiments was carried out using different DS concentrations at a constant temperature (25°C), while the second part was carried out using similar DS concentration at three different temperatures (13°C, 25°C and 32°C). The membrane used is a symmetric cellulosic membrane composed of one finely porous layer; the advantage of such membranes in FO applications is the absence of the internal concentration polarization that may occur inside the substrates of asymmetric membranes.

The membrane used is a readymade dialysis membrane with about 1.8 nm mean-pore diameter. The results of water and solute flux obtained from this study indicate that this dialysis membrane, or similar, is potentially suitable for use in other FO applications, especially when the

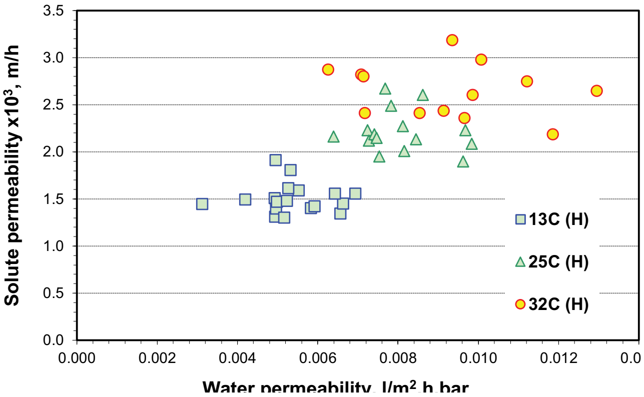

Fig. 14. Solute permeability as a function of water permeability in several FO experiments at different temperatures using NaCl aqueous solutions as DS.

FW used is industrial or municipal wastewater, where the diffusion of DS solutes to FW is of less concern.

The results show that both water flux from the FW to the DS and the opposite solute flux from the DS to the FW increase with the increase of the solute concentration difference, or the osmotic pressure difference, across the membrane. This proportionality is characterized by a suggested proportionality factor, k_J. It is found that the value of this factor approaches the value of the saturation concentration of NaCl, the solute used in this case.

The results also show that both water permeability and solute permeability decrease with the increase of the solute concentration difference, or the osmotic pressure difference, but increase with temperature increase. It is found that solute permeability is proportional to water permeability, and another permeability proportionality factor, k_E, is suggested. It is explained that this factor increases with the operational temperature increase and decreases with the increase in the molecular weight of the DS solute.

Experiments carried out at different temperatures using similar range of DS solute concentration show that both of the flux and the permeability of water and solute increase with the increase in temperature. This is explained by the physical properties of solutions and the relationships between temperature and solute diffusivity or solution viscosity.

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