

Concentration of Orange Juice Using Forward Osmosis Membrane Process

Khalid W. Hameed

Biochemical Engineering Department- Al-Khwarizmi College of Engineering- University of Baghdad

Abstract

Forward osmosis (FO) process was applied to concentrate the orange juice. FO relies on the driving force generating from osmotic pressure difference that result from concentration difference between the draw solution (DS) and orange juice as feed solution (FS). This driving force makes the water to transport from orange juice across a semi-permeable membrane to the DS without any energy applied. Thermal and pressure-driven dewatering methods are widely used, but they are prohibitively energy intensive and hence, expensive. Effects of various operating conditions on flux have been investigated. Four types of salts were used in the DS, (NaCl, CaCl₂, KCl, and MgSO₄) as osmotic agent and the experiments were performed at the concentration of the salts in the DS ranged (3.5 – 20% by wt), the temperature of DS ranged (20 – 50°C), and the flow rate of the FS and DS ranged (1 – 4 lit/min). It was observed that the optimum operating conditions are: concentration of salt = 20% by wt for CaCl₂, temperature of DS = 50°C, and the flow rate of FS = 4 lit/min where at these conditions the maximum flux was obtained equal to 13.2 lit/m².h or the total volume of the water transferred from the juice (during 3 hours and membrane area of 0.0135 m²) was 0.535 lit. NaCl performed much higher efficiency as osmotic agent than the others salts up to the concentration of 15.2%, but after 15.2% the CaCl₂ was the best.

Key Words: Forward osmosis, reverse osmosis, fruit juice, osmotic pressure, draw solution.

Introduction

Membrane separation processes have become one of the emerging technologies in the last few decades especially in the separation technology field. They offer a number of advantages over conventional separation methods in a wide variety of applications such as distillation and evaporation. Membrane processes can be easily scaled up due to their compact and modular design; they are

able to transfer specific components selectively; they are energy efficient systems operating under moderate temperature conditions ensuring gentle product treatment [1].

Osmosis is the transport of water across a selectively permeable membrane from a region of higher water chemical potential to a region of lower water chemical potential. It is driven by a difference in solute concentrations across the membrane

that allows passage of water, but rejects most solute molecules or ions [2]. Water molecules will therefore move from one solution to another to achieve maximum mixing, i.e. equilibrium. Thermodynamically, the strength of this mixing tendency is measured by the solution's "osmotic potential," or "osmotic pressure." The osmotic potential is high for concentrated solutions and low for dilute solutions, and is roughly proportional to the molar concentration of dissolved species. Osmotic pressure is the pressure that must be applied to a solution to prevent a net transfer of water into the solution across a semi-permeable membrane [3].

By applying a pressure in excess of the osmotic pressure, pure water flows from the high solute concentration side through a membrane to the low solute concentration side and thus, the separation of water from the solution is achieved. This is the reverse of the normal osmosis process and termed as reverse osmosis (RO) [4, 5]. In wastewater treatment applications where the solvent is usually water and the solutes are the contaminants, the semi-permeable membrane allows the flux of water across the membrane but rejects contaminants. In such a system the wastewater, or feed, is passed on one side of the membrane and an osmotic agent (OA), such as salt water, is passed on the other. The OA can use any solute as long as it can produce an osmotic pressure that is higher than that of the feed and the solute used is well rejected by the membrane [6].

Forward osmosis (FO) is emerging membrane separations technologies that have the potential to be innovative, sustainable, and affordable alternatives to reverse osmosis (RO) and electrodialysis reversal (EDR) because of its ability to utilize the green energy available in natural systems [7]. The term forward osmosis is used to refer

to normal osmotic processes is that will occur on its own, without any form of external pressure or push. Our system adapts the FO theory and the advantages include:

- Elimination of external pressure
- Reduce cost by eliminating large pressure pumps and pressure exchanger systems
- Pressure generated in draw solution can be utilized in place of external pressure
- Supports the objective of small-scale power generation
- Availability of FO membranes that can be used to develop PRO situations [8].

Compared to RO, FO systems and the principle of osmotic pressure have a wide range of applications in the areas of wastewater treatment and water purification systems; seawater desalination and brackish water processes; concentration of solutions of food products, pharmaceutical solutions and chemical streams; and power generation [9, 10, 11].

Traditionally, orange juice has been concentrated using a thermal process. Such a process results in a loss of flavor top notes, color degradation, and a cooked taste. The citrus industry compensates for the product degradation through essence recovery, careful process control and blending to produce a good quality concentrate which, although readily distinguishable from fresh juice, has received broad consumer acceptance. The membrane process was designed to produce a concentrate juice with fresh juice flavor and commercial levels of stability [12].

Eq. (1) shows the relationship between water flux across the membrane and both the hydrostatic and osmotic pressure differentials across the same membrane [13]. This equation is stated in the form most relevant to RO as follows:

$$F_w = A_C (\Delta P - \Delta \pi) \quad \dots(1)$$

Where:

F_w = Total water flux across the membrane (*lit/m².h*)

A_C = Membrane flux resistance constant (*lit/m². h.kPa*)

ΔP = Hydrostatic pressure (kPa)

$\Delta \pi$ = Opposing osmotic pressure potential (kPa) which is equal to the difference between osmotic pressure of the draw solution and the feed solution

In FO, the hydrostatic pressure supplied is zero and the same governing equation can be rearranged to Eq. (2):

$$F_w = A_C \Delta \pi \quad \dots(2)$$

As a result of Eq. (2) it can be seen that the membrane can be configured such that no hydrostatic pressure exists across the membrane and thus no pressure housing and/or support is required. This allows the membranes to operate in soft bags packed within water walls.

In most cases some hydrostatic pressure is still present as a result of the act of supplying the membrane with a flow of liquid. This flow is required for both sides of the membrane and should be near to balanced (i.e. ΔP still zero across the membrane). In this situation the hydrostatic pressure could be in either the forward or opposing direction relative to the intended water flux direction, but in either case will be negligible in comparison to the osmotic pressures [14]. A_C dependent on such membrane characteristics as membrane thickness, partition (sorption) coefficient of water into the membrane, and diffusivity of water within the polymer membrane phase [15].

The aim of the present work is to concentrate the orange juice by low cost method and retain the juice with

its properties compared with the other methods such as reverse osmosis (high pressure required) or evaporation (caused loss of some properties of the juice such as its vitamins, flavor, color degradation ...etc). In this research Cellulose triacetate membrane was proposed for forward osmosis, fresh orange juice as feed solution (FS), and salt solution (sodium chloride, potassium chloride, calcium chloride, or magnesium sulfate) as draw solution (DS). The parameters studied were: concentration of salt in DS in the range of (3.5 – 20% by weight), temperature of DS in the range of (20 – 50°C), and volumetric flow rate of DS and FS in the range of (1 – 4 lit/min) were studied on the effect of flux of water from FS to DS.

Experimental Work

1. Materials

Orange juice:

The orange juice (feed solution) was obtained from fresh natural orange of local market, then the juice was filtered using filter paper to remove suspended solids, fiber, coarse pulp and pieces of orange.

Draw solution:

The concentrated solution on the permeate side of the membrane is the source of the driving force in the FO process. The draw solution was prepared by smelting one of the following salts in the distilled water: sodium chloride (NaCl), calcium chloride (CaCl₂), potassium chloride (KCl), or magnesium sulfate (MgSO₄).

Pumps:

Two pumps were used. Each of 150-720 gal/h, 11.4-54.6 lit/min. One used to the draw solution and the other used to the feed solution.

Membrane:

Cellulose triacetate (CTA) sheet membrane (Hydration Technology Inc. Albany, OR) have been used in the forward osmosis (FO) process.

Refractometer:

Digital Refractometer was used to measure the sucrose content in the orange juice. It is calibrated by depending on the literature given by Randle [16].

Conductivity meter:

Digital conductivity meter was used to measure the conductivity of permeate solution.

2. Experimental Procedure

The experiments were run on a bench-scale laboratory system. A schematic diagram is presented in Fig. (1). The separation cell of membrane unit was built of two rectangular channels with the dimensions 15 cm long×9 cm wide×2 cm deep (the area of membrane = 0.0135 m²) on both sides of the membrane. The volume of draw solution (DS) is 15 lit (in order to its osmotic pressure does not affected

significantly with water transferred and then the change in flux with time will be small) and they were run in a closed loop. The FS flows on the active layer of the membrane. In order to increase the mass transport on both sides of the membrane, mesh spacers made of polypropylene were inserted within both channels; also these mesh support the membrane and protect it from deformation. The temperature of both solutions was controlled using heating coil holding thermostat, and the volumetric flow rate of both solutions was controlled using rotameter. The pressures have kept on the FS side about 1.1 bar and on the DS side about 1.05 bar using gauge pressures mounted on above the box which contain the membrane as shown in Fig. (1).

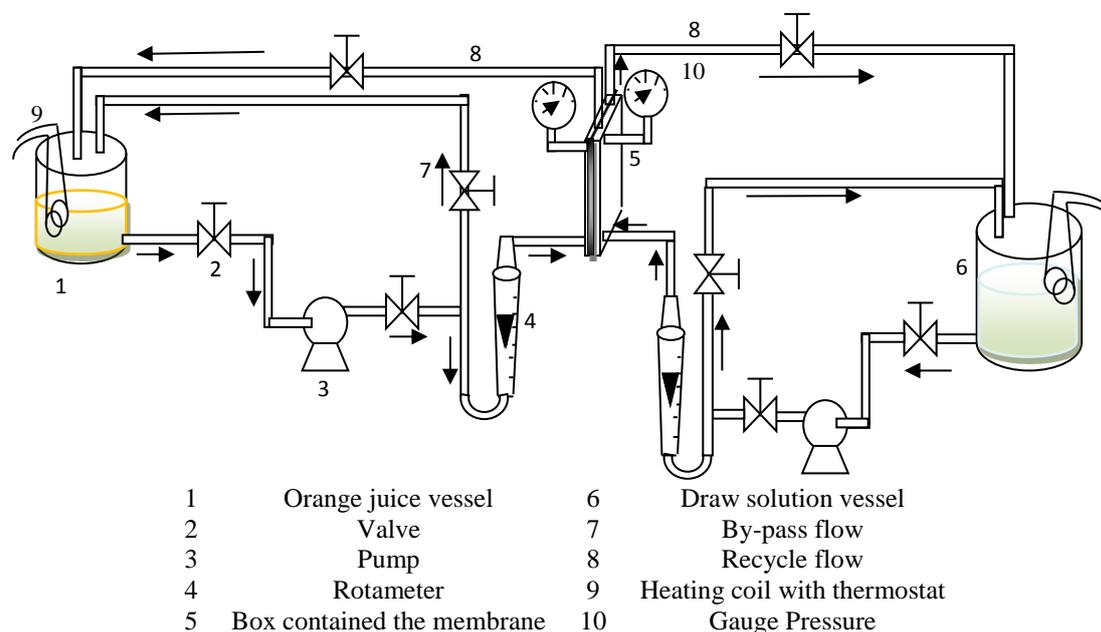


Fig. 1, Schematic diagram of the laboratory scale forward osmosis membrane unit system

The time of experiment was three hours. Water flux (permeate) into the DS was measured by the elevation in the DS volume during a selected period of time and compare with the reduction in the FS volume for checking where the vessels of the DS and FS are

graduated and calibrated accurately with volume.

Dividing the water transferred by the area of membrane per time gives the flux in (L/m².h). After each experiment, FS return to its original volume by adding the distilled water

instead of the water lost and then reused again for three another experiment, after that FS (orange juice) is eliminated and new juice is used. DS also is return to its original concentration by adding a suitable amount of salt and checking its concentration using conductivity meter.

Also the fouled membrane is backwashed using salt solution in the juice circuit and fresh water in the DS circuit and operate for 15 min then washed for 30 min by circulating using RO water in each side.

Results and Discussion

1. Effect of Draw Solution Concentration

Fig. (2) shows the effect of draw solution (DS) concentration on the flux of water through the membrane for four types of salts (NaCl, CaCl₂, KCl, and MgSO₄). The experiments were conducted at the same conditions (temperature of feed and draw solution are 20°C, and volumetric flow rate of feed and draw solution are 1 lit/min). As shown in Fig. (2) the flux of water increases with increasing of salt concentration and the sodium chloride salt (NaCl) has the more osmotic pressure than the other salts up to 15.2% by wt, after this concentration the calcium chloride salt (CaCl₂) exhibits more osmotic pressure and then more flux than the other salts. Also magnesium sulfate (MgSO₄) has less osmotic pressure than the other salts, where before concentration of 7.4% gave negative flux (from draw solution to feed solution).

The high osmotic pressure is obtained when the osmotic agent is highly soluble in water and has low molecular weight [5], and since NaCl has lower molecular weight (58.5) and higher solubility (357 g/lit at 20°C) than the other salts [17] it gave more osmotic pressure and then more flux

than the others salts. Nevertheless CaCl₂ (its molecular weight = 111 and its maximum solubility at 20°C = 294g/lit [17]) gave osmotic pressure greater than NaCl after concentration of 15.2%.

Based on Eq. (2), the increase in flux should be linear with the osmotic pressure difference. Fig. (2) however, shows a non-linear phenomenon, especially at higher driving forces. This phenomenon is attributed to internal concentration polarization, most likely due to microporosity at the membrane permeate side.

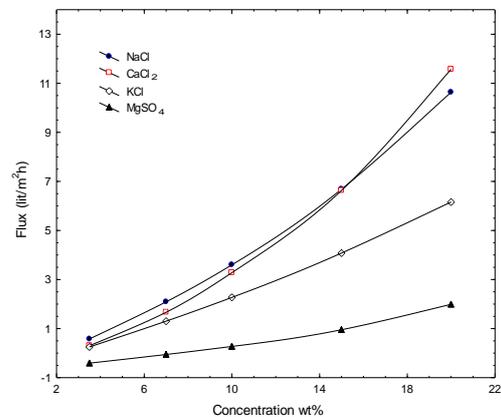


Fig. 2, Effect of DS concentration on the flux of water at temp of FS and DS = 20°C, Flowrate of FS and DS = 1 lit/min

2. Temperature Effect

Fig. (3) shows the effect of temperature of draw solution (DS) on the osmotic pressure difference between the FS and DS and then on the flux of water. The experiments of the Fig. (3) are achieved at the concentration of DS is 10% by wt, temperature of FS is 20°C, and flow rate of FS and DS are 1 lit/min. The results in Fig. (3) indicate that with increasing temperature of DS leads to increase the flux of water from FS to DS through membrane because of the viscosity decreases with increasing temperature which increases the diffusion rate of water through the membrane and thus, its water permeability coefficient and lead to

increase of the osmotic pressure of the DS and then increase the flux of the water. An increased diffusion coefficient for the DS will increase the mass transfer coefficient, reducing the impact of the external concentration polarization (ECP) modulus. The effect is similar for the internal concentration polarization (ICP) modulus, where an increased diffusion coefficient reduces solute resistivity. However, since both the ECP and ICP moduli are exponential functions of the permeate water flux as well, the temperature effect on these phenomena will be lessened, as an increase in the water permeability coefficient of the membrane will increase flux.

The viscosity of a solution is a measure of the fluid's resistance to flow or deformation [18]. Also the osmotic pressure of the solution increased with increasing temperature according to the Van't Hoff equation [5, 19]:

$$\pi = iRCT \quad \dots(3)$$

Where:

- π = Osmotic pressure (kPa)
- i = dissociation factor (van't Hoff factor)
- R = gas constant (8.314 kJ/kmol.K)
- C = molar concentration (kmol/m³)
- T = absolute temperature (K)

For three salts (NaCl, CaCl₂, and KCl) with increasing the temperature of DS gave slight increasing in water flux, while for MgSO₄ salt with increasing the temperature of DS gave slight decreasing in water flux.

Fig. (4) and (5) show the effect of concentration of NaCl and CaCl₂ respectively as osmotic agent (OA) on the flux of water through membrane at different temperatures. Again, by increasing the concentration of OA the flux of water increases and also increases with increasing the temperature of DS as shown in Fig. (3). From Fig. (3), (4), and (5) it can be

seen that the temperature plays a minor role in osmosis driven processes compared with the concentration of the DS.

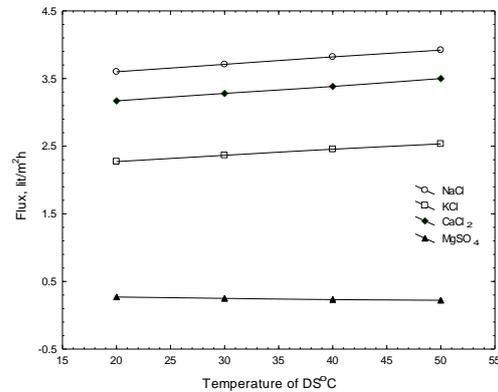


Fig. 3, Effect of Temperature of DS on the flux of water at Concentration of salt in DS = 10%, flow rate of FS and DS = 1 lit/min and at temperature of FS = 20°C

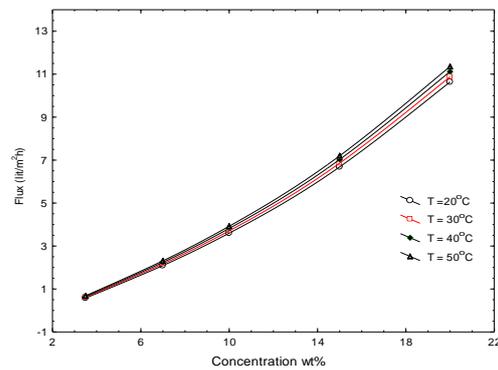


Fig. 4, Effect of concentration of NaCl on the flux of water at different temperature of DS and at temp of feed = 20°C, flow rate of feed and DS = 1 lit/min

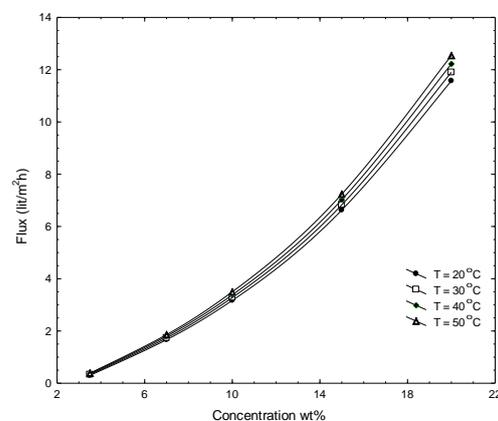


Fig.5 Effect of concentration of CaCl₂ on the flux of water at different temperature of DS, temp of FS = 20°C, flow rate of feed and DS = 1 lit/min

3. Flow Rate Effect

Fig. (6) and (7) show the effect of flow rate of FS and DS on the flux of water respectively. Fig. (6) explains that the flux of water increases by increasing the flow rate of FS while from Fig. (7) the flux decreases by increasing the flow rate of DS for all four salts. Increasing the flow rate of FS prevents the concentration buildup in the solution at the vicinity of the membrane surface, thus reducing the accumulated solute on the surface of the membrane and lead to increase the water flux. This behavior contradicts the case of increasing the DS flow rate.

Fig. (6) and (7) illustrate that the flow rate has slight effect on the flux compare with the effect of concentration of the DS.

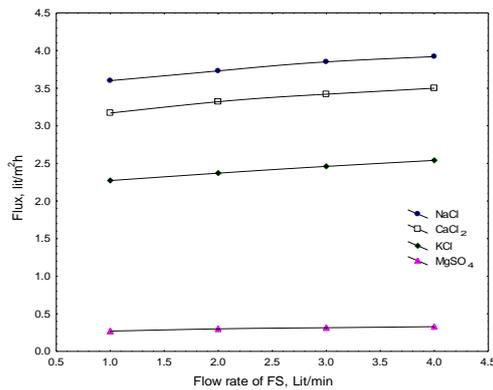


Fig. 6, Effect of flow rate of FS on the flux of water at concentration of DS = 10% by wt, Flow rate of DS = 1 lit/min and temperatures of FS and DS = 20°C

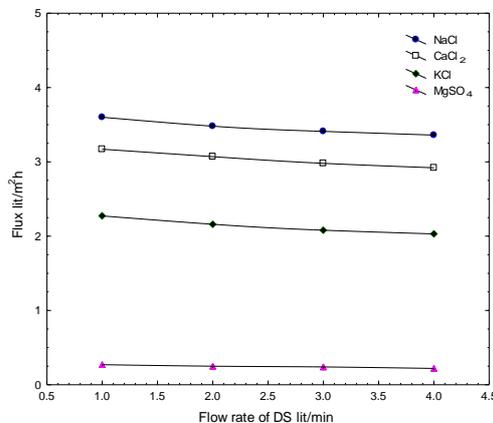


Fig. 7, Effect of flow rate of DS on the flux of water at concentration of DS = 10% by wt , flow rate of FS = 1 lit/min and temperature of FS and DS = 20°C

4. Membrane Flux Resistance Constant

Fig. (8) shows the osmotic pressure against concentration of sucrose [20]. After calibration of refractometer by depending on the literature given by Randle [16] the sucrose content in the orange juice was measured where the refractive index was equaled to 1.3539 that gives the sucrose content in the juice = 13.9% by wt [16], then from Fig. (8) the osmotic pressure of orange juice = 1180 kPa.

Fig. (9) shows the osmotic pressure against concentration of NaCl solution [20]. At concentration of NaCl = 10% by wt, from Fig. (9) the osmotic pressure is evaluated to be 8700 kPa.

From Fig. (3) or (4), at 10% NaCl by wt, Temp = 20°C for FS and DS , and flow rate = 1 lit/min for FS and DS, The flux of water = 3.605 lit/m²h.

The membrane flux resistance constant A_C is calculated at 20°C from Eq. (2):

$$F_w = A_C \Delta\pi$$

$$3.605 = A_C (8700 - 1180)$$

$$\rightarrow A_C = 4.8 \times 10^{-4} \text{ lit/m}^2\text{h.kPa}$$

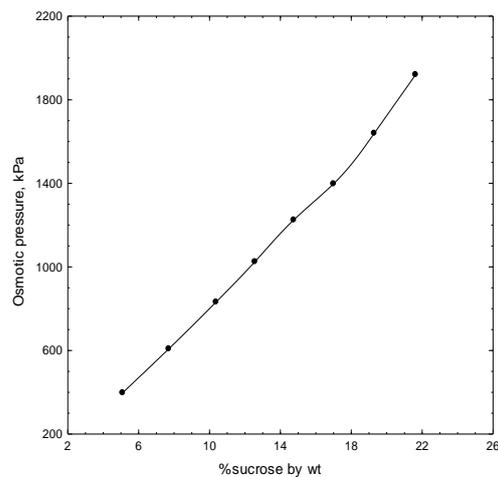


Fig. 8, Osmotic pressure of the sucrose against concentration

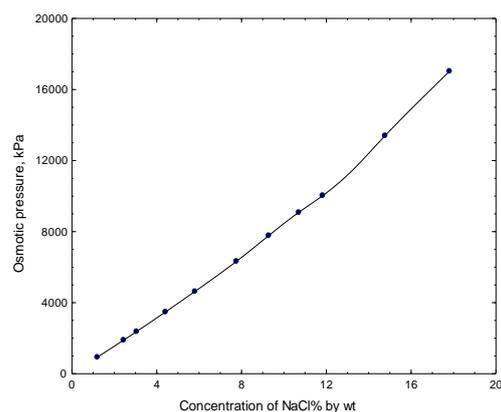


Fig. 9, Osmotic pressure of the Sodium chloride against concentration

Conclusion

The obtained results in this research demonstrate the applying of forward osmosis (FO) process for the concentration of orange juice. The draw solution concentration is a serious parameter that influences water flux in the FO process where it has the biggest effect on the water flux compared with the others variables. Temperature and flow rate were found to have a minor effect on the permeate water flux, where as increased the temperature leads to little increase in water flux for salts (NaCl, CaCl₂, and KCl) and little decrease in water flux for MgSO₄ salt. Also increasing of flow rate of FS or DS leads to little change in the flux, so it can be execute the FO membrane process at ambient temperature and with small scale pump. NaCl salt gave higher osmotic pressure up to concentration of 15.2% by wt than the other salts (KCl, CaCl₂, MgSO₄) that used in the draw solution, but CaCl₂ gave higher osmotic pressure than NaCl when the concentration exceed 15.2%.

This technology can be applied for other fruit juices.

Nomenclature

| | |
|----|-----------------|
| FS | Feed solution |
| DS | Draw solution |
| FO | Forward osmosis |
| RO | Reverse Osmosis |

| | |
|------------|---|
| DS | Draw solution |
| OA | Osmotic agent |
| EDR | Electrodialysis reversal |
| A_c | Membrane flux resistance constant ($lit/m^2 \cdot h \cdot kPa$) |
| C | Molar concentration ($kmol/m^3$) |
| F_w | Total water flux across the membrane ($lit/m^2 \cdot h$) |
| i | Dissociation factor (van't Hoff factor) |
| R | Gas constant (8.314 kJ/kmol.K) |
| T | Absolute temperature (K) |
| ΔP | Hydrostatic pressure (kPa) |
| π | Osmotic pressure (kPa) |

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