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Direct fertigation with brackish water by a forward osmosis system converting domestic reverse osmosis module into forward osmosis membrane element

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ABSTRACT

Forward osmosis has found numerous applications in water treatment, water reuse, and other sectors e.g. desalination, concentration of wastewater, landfill leachate treatment, controlled drug release, emergency water treatment kit, etc. because of its low pressure operation and low fouling. The present paper demonstrates that the fertilizers can function as draw solution and be employed for drawing water from the brackish water. The proposed process eliminates the recovery step and the diluted draw solution can directly be utilized in fields for fertigation. We have selected fertilizers as a draw solution which provides nutrients to the crops for their basic growth. The domestic reverse osmosis membrane element has been converted to forward osmosis membrane element in a novel approach. The performance of each fertilizer is accounted in terms of experimentally obtained flux by comparing it with theoretically estimated flux. It has been found that there is a significant difference in experimentally obtained flux and theoretically estimated flux because of internal concentration polarization due to the presence of draw solution on permeate side and low availability of the effective membrane surface area. We have also investigated reverse draw solute diffusion for each fertilizer and the increase in concentration of feed solution to understand the movement of ions across the membrane. The reverse salt diffusion was observed in all three fertilizers studied with water as feed, however it was absent when sodium chloride feed solution was used. This shows that the presence of salt on feed side of the membrane suppresses the reverse salt diffusion on account of Donnan exclusion.

Keywords: Forward osmosis; Fertilizer; Draw solution; Reverse solute diffusion; Brackish water

1. Introduction

Water is an essential element of any living being and thus basic for survival of life. The demand of water has been increasing at rapid rate due to growth of population and industrialization, and it has led to serious concerns on global sustainability in terms of water, energy, climate change, and environment [1,2]. Water and energy are correlated and intertwined with

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each other because production of water is energy intensive process and production of energy requires water. Hence, adequate water supply and affordable energy sources become essential in today's world to sustain public health, industrialization, and national prosperity. It has been estimated that population of world will increase from current population [3–7].

Water is also very important for agricultural land to mitigate increasing food demand. It has been estimated that around 70% of water from total consumption is being used for irrigation of agricultural land followed by industrial use (21%) and domestic use (9%) [8]. Thus, sustainable supply of water for agriculture is very important to the well-being of population.

A significant part of the world is occupied by arid/barren land/desert where, the economies have thrived on account of oil reserve but the water availability is a grave concern. They have to depend on desalination i.e. converting saline source of water—seawater or ground water into fresh water. Ground water is the major source of water, but there has been a considerable depletion in ground water table due to unpredictable rainfall and uneven use of ground water. So, sources of fresh have been vanished gradually due to climate change and it has led to development of water reuse and purification system as alternative water sources.

In particular, membrane separation processes such as sea water desalination and wastewater treatment are alternative sources of water production and they attract the researcher due to its cost effectiveness and excellent water quality in comparison to that produced by conventional water production technology [9,10]. Membrane-based desalination has overtaken the thermal desalination owing to its lower capital and operating cost when the stand-alone system is considered.

Among all the membrane separation processes reverse osmosis is one of the most sound and efficient method for the production of clean water. However, it has certain limitations as well as disadvantages such as it is energy intensive process because of high hydraulic pressure required to overcome osmotic pressure of the source water. Other problems in RO, such as concentration polarization and membrane fouling, lower feed recovery rate. Besides that the disposal of the brine after RO is a serious environmental issue [11,12].

Forward osmosis (FO) has gathered attention of researchers world over to address the problems of RO e.g. high pressure requirement, fouling, and concentrate management. FO is an osmotic pressure-driven process that takes advantage of osmotic pressure for transport across the membrane and therefore very low hydraulic pressure is required to drive the water across membrane. Thus forward osmosis process is less energy intensive. Moreover, FO has low tendency for fouling because of lower trans-membrane pressure, easier fouling removal, and higher water recovery as compared to the pressure-driven process like reverse osmosis [13,14].

FO is still facing some critical challenges. Concentration polarization is a common and inevitable phenomenon in both pressure-driven and osmotically driven membrane processes; concentration polarization is caused by the concentration difference between the feed solution and the draw solution through FO membrane. External concentration polarization (ECP) occurs at the surface of dense active layer of the membrane and Internal concentration polarization (ICP) occurs within the porous support layer of the membrane. Membrane fouling is also an inevitable phenomenon in all membrane processes. Lower membrane fouling implies more product water, less cleaning, and longer membrane life, thereby reducing operational and capital costs. However, membrane fouling in osmotically driven membrane processes is different from pressure-driven processes due to low hydraulic pressure being employed former. Membrane fouling in FO was originally studied by Cath and co-workers where they have reported FO might be low fouling since no sign of flux decline in their studies [15]. On the other hand, membrane fouling may influence the solute rejection of the FO membrane. Valladares and co-workers observed that organic foulants on the membrane surface (active layer) could enhance the negative charge property and hydrophilicity of the surface and also increase the absorption capacity for hydrophilic compounds. These changes can improve the rejection for many trace organic contaminants [16].

In osmotically driven membrane processes, reverse diffusion of the solute from the draw solution through membrane to the feed solution is also causing problem. It should be minimized in the development/design of FO process.

1.1. Membrane development

In last decade, many researchers have tried to make FO membrane material e.g. cellulose acetate or ethyl cellulose by dip coating and phase inversion, Polysulfone-polyamide by phase inversion, and interfacial polymerization PSF nanofiber support polyamide by electrospinning and interfacial polymerization [17–19].

FO has been attempted for numerous applications such as brine concentration, wastewater treatment and osmotic membrane bioreactor, liquid food concentration and pharmaceutical applications. There are two major factors that affect the FO process, membrane, and draw solution. Numerous attempts have been made for the development of the different draw solutions e.g. Ammonia and Carbon dioxide, Glucose, Magnesium chloride, Potassium nitrate and sulfur dioxide, Magnetic nanoparticles, 2-mithylamidazole-based solute, Stimuli responsive hydrogel, polyelectrolytes, hexavalent phosphazene salts, etc. [20–30].

All the draw solutes listed have some drawbacks and required some energy for their recovery; moreover it makes the process lengthy and tedious. So, to eliminate energy requirement for recovery of water from draw solution one can use fertilizers as draw solute for FO because it can be directly used for fertigation. Fertigation is the method in which crops nutrients are supplied in the dissolve form to the crop through proper irrigation system. This concept offers several benefits: firstly, the cost of desalination will be low because no recovery of draw solute required after process, secondly it will provide nutrient-rich water to the crops [31–33].

Initially, concept of fertilizer-driven FO was developed by moody which was reported in his article [34]. Later, this concept is not much more exploited. So the aim of this study is to develop FO-based fertigation process using different fertilizers as draw solute which may provide primary nutrients required for growth of any crop.

1.2. Theoretically estimated flux

Theoretical flux can be obtained due to osmotic pressure difference of feed and draw solution in ideal FO process, which is given by following equation [35,36]:

$$J_w = A\,\sigma\,(\pi_{ds} - \pi_{fs}) \tag{1}$$

where J_w is the water flux, A is the membrane permeability coefficient, σ is the reflection coefficient, π_{ds} and π_{fs} are the osmotic pressure of draw solution and feed solution, respectively.

Thin film composite forward osmosis membranes with novel hydrophilic supports for desalination have been demonstrated [37]. Thin film composite forward osmosis membranes based on polydopamine-modified polysulfone substrates showed enhancements in both water flux and salt rejection [38].

Here, we have selected fertilizers which will provide primary nutrients such as nitrogen, potassium, and magnesium. The study includes difference in osmotic pressure among the taken draw solutions and water flux obtained for each of them. We have also studied reverse draw solute flux for each draw solution. Thus, the present work demonstrates the novel approach of using forward osmosis for direct fertigation by removing the tedious and lengthy process of recovery of water from draw solution.

2. Experimental

2.1. Materials

All the chemicals used in the experiment are listed in Table 1.

2.2. Experimental setup

The bench scale experimental setup is given in the Fig. 1. We have fabricated a novel forward osmosis element where the spiral wound thin film composite RO membrane element has been converted into forward osmosis membrane element. The Dow make domestic RO membrane element (0.6 m^2 area) was used in the experiment. The blockage from center tube was removed. Also, an additional hole in membrane housing was made. The flexible pipe was used to connect the center tube with the membrane housing center hole and thus two inlets, two outlets systems were fabricated.

Experimental setup of FO consist of spiral wound module provided with two inlets and two outlets for feed and draw solution in which draw solution will pass through perforated center tube and feed solution will pass on the normal feed side of the membrane element. For FO process, both the solutions were passed through module in counter-current fashion. Here, we have used only one pump to create flow rate of the feed solution, while draw solution was supplied from draw solution column as shown in the Fig. 1 to save the cost of energy and to avoid the back pressure generated on account of pump that may retard the flow of water from feed side to draw solution side.

2.3. Specification of draw and feed solution

For FO performance test, draw solution of each fertilizer was prepared in RO water with concentration 2 M. DI water and NaCl solution (2,000 ppm) were used as feed solution. Here, it must be mentioned that DI water as a feed solution was used only for flux comparison assessment; because the flux with DI water will be high as compared to flux with NaCl because of higher differential osmotic pressure.

Table 1
Different chemicals used in experiment with their specifications

Name of fertilizers	Chemical formula	Molecular weight	Purity (%)	Supplier
Ammonium sulfate	$(NH_4)_2SO_4$	132.1	99.5	RFCL Ltd
Magnesium nitrate	$Mg(NO_3)_2 \cdot 6H_2O$	256.41	99	SD Fine chem. Ltd
Potassium chloride	KCl	74.55	99	Himedia laboratories Pvt. Ltd
Sodium chloride	NaCl	58.4	99.9	Fisher scientific

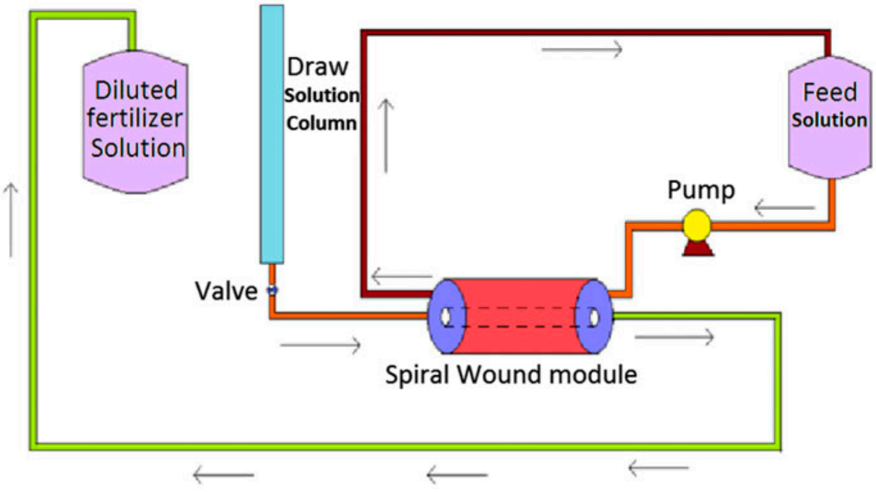

Fig. 1. Schematic setup of experiment.

2.4. Analysis of concentrations of feed and draw solutions

Concentrations of the feed solution and draw solution were found by different analytical methods. Sodium (Na^+) and potassium ion (K^+) concentration were assessed by flame photometer. Magnesium ion (Mg^{++}) concentration was found by titration with EDTA (ethylene diamine tetra acetate) using EBT (Eriochrome black T) as an indicator. Concentration of sulfate ion (SO_4^{2-}) was measured by gravimetric analysis using methyl orange and HCl as indicator and barium chloride as reagent ($BaCl_2$).

2.5. Osmotic pressure determination

The osmotic pressure of draw solution and feed solution were found experimentally by vapor pressure osmometer. Vapor pressure osmometer gives molality of solution in mol/kg. The osmotic pressure can be found by substituting molality in the equation $\pi = M\rho RT$ (M: Molality, ρ: density, R: universal constant (0.08206 L atm/mol K), T: absolute temperature in (°K)).

The above equation is another form of Morse equation [39].

3. Results and discussion

3.1. Osmotic pressure measurement of draw and feed solution

The molalities of solutions in mol/kg were determined by vapor pressure osmometer and that was used to find osmotic pressure. The results are shown in Table 2.

Higher the osmotic pressure, higher will be osmotic potential. We can notice that osmotic pressure of all the fertilizers taken as draw solution is considerably higher than that of the feed solution NaCl and even higher than seawater which has the osmotic pressure of ~28 atm.

3.2. Comparison of flux with NaCl and DI water of different fertilizer solutions

Table 3 indicates the experimental flux with water and NaCl solution with different fertilizer solutions.

It can be seen from Table 3 that the flux with DI water is more as compared to flux with NaCl with all three draw solutions. The flux with NaCl is the highest in case of magnesium nitrate draw solution as the

Table 2
Osmotic pressure of feed and draw solution

Substance	Molality (mol/kg)	Osmotic pressure determined by Osmo-meter (in atm)
NaCl	0.069	1.6938
$Mg(NO_3)_2 \cdot 6H_2O$	7	205.26
$(NH_4)_2SO_4$	5.4	148.19
KCl	4.6	124.31

osmotic pressure is the highest. The ratio of flux with NaCl to flux with DI Water ranges from 0.4162 to 0.6311.

Theoretical flux was calculated by Eq. (1), where A (membrane permeability co-efficient) was 5.32×10^{-12} m/Pa s and reflection co-efficient σ was 0.93. Thus, theoretical flux for potassium chloride, ammonium sulfate, and magnesium nitrate are 61.47, 73.44, and 102.05 μm/s respectively. However, the actual flux is significantly lower than theoretical flux.

There are certain limiting factors that reduce the flux:

1. (1) ICP due to high concentration of draw solution.
2. (2) ECP due to gradually increase in feed water concentration during the experiment.
3. (3) The draw solution passes by gravity in the center tube and the outlet of the draw solution pipe is heightened to ensure the movements of draw solution inside the membrane element on permeate side. However, due to resistance inside the membrane element on account of thin permeate spacer and narrow passage; the draw solution may not be able to reach the entire surface area of membrane. Back pressure from draw solution side may retard the flow of permeate as it has to counter an additional pressure gradient. Therefore, back pressure was not given on permeate side.

These are the reasons why the theoretical flux is significantly higher than experimental flux.

It is also noted that highest experimental flux was obtained in case of ($Mg(NO)_3$–NaCl) followed by potassium chloride and ammonium sulfate. It represents nonlinear relationship between osmotic pressure difference and experimental water flux, while in case of draw solution and DI water, relationship between experimental water flux and osmotic pressure remains constant as shown in Fig. 2.

3.3. Reverse draw solute flux measurement

Reverse draw solute flux is inevitable phenomena in case of membrane separation process, especially when there are two solutions kept in intimate contact with the help of membrane and separation is carried out based on concentration difference or chemical potential difference. Reverse draw solute flux may cause significant drawback for FO process in terms of ICP and that will lead to a considerable decline in the pure water flux and it will also deteriorate quality of the feed solution so, it is necessary to measure the performance of fertilizer draw solution in terms of reverse draw solute flux. We have investigated the reverse solute flux for each fertilizer by measuring particular ion concentration of initial sample (before experiment) and final sample (after experiment) in both case DI water and NaCl (feed solution) by different analysis method that has been already described in the earlier section. We have also measured the passage of the feed solution from the membrane of feed side to draw solution side by the same analytical method. Reverse draw solute flux and

Table 3
Experimental flux with water and NaCl solution with different fertilizer solutions

Draw solution	Initial concentration of draw solution (mg/l)	Final concentration of draw solution (mg/l)	Flux with DI Water (μm/s)	Flux with NaCl solution (μm/s)	Ratio Flux with NaCl/Flux with DI water
KCl	65,200	39,400	5.7215	3.6111	0.6311
$Mg(NO3)_2 \cdot 6H_2O$	49,300	32,500	9.3429	3.8889	0.4162
$(NH_4)_2SO_4$	76,800	42,800	6.7737	3.5555	0.5249

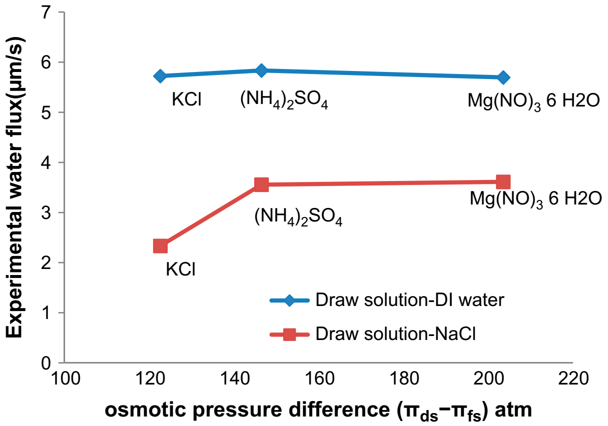

Fig. 2. Experimentally obtained flux for different fertilizers (2 M Concentration) as a function of osmotic pressure.

passage of the feed solution in terms of ion concentration in initial and feed solution are given in the Table 4.

From the data of the Table 4, we can observe that reverse draw solute diffusion is significant when DI water was used as the feed solution with different fertilizer draw solution and it is observed that reverse draw solute diffusion is highest in case of magnesium nitrate and lowest in case of ammonium sulfate. While no reverse draw solute diffusion was observed when NaCl used as a feed solution with different fertilizer as a draw solution.

However, if we look the trend of the sodium ion concentration (Na^+), the sodium concentration in draw solution increases in ammonium sulfate and magnesium nitrate, whereas sodium concentration decreases in case of potassium chloride fertilizer. This is because of potassium ion present on draw solution side pushes the sodium ion across the membrane on account of Donnan exclusion and it is also seen in case of $Mg(NO_3)_2$–DI water. Sodium ion concentration increases in feed, which is obvious because of passage of water toward draw solution side. Fig. 3 demonstrate the Donnan exclusion phenomenon, where magnesium ions diffuse from the draw solution side to feed side to balance charge on either side of the membrane. The passage of sodium ion through membrane offsets by the passage of magnesium ion from draw solution side to feed side.

Table 4
Different ions concentration in feed and draw solution

Experiment	Sample	Concentration (mg/l)	Concentration (mg/l)	Concentration (mg/l)
1	Draw solution (Magnesium nitrate–Water)	Na^+(i) = 275	Mg^{2+}(i) = 45,068	NO_3^-: 232,850
		Na^+(f) = 196.6	Mg^{2+}(f) = 14,200	NO_3^-: 72,460
	Feed (Magnesium nitrate–Water)	Mg^{2+}(i) = 0	NO_3^-: 0	Na^+: 0
		Mg^{2+}(f) = 474	NO_3^-: 2,450	Na^+: 0
2	Draw solution (Magnesium nitrate–NaCl)	Mg^{2+}(i) = 51,200	Na^+(i) = 373	NO_3^-: 261,265
		Mg^{2+}(f) = 15,600	Na^+(f) = 432	NO_3^-: 79,600
	Feed (Magnesium nitrate–NaCl)	Mg^{2+}(i) = 0	Na^+(i) = 1.016.1	NO_3^-: 0
		Mg^{2+}(f) = 0	Na^+(f) = 2.005.3	NO_3^-: 0
3	Draw solution (Ammonium sulfate–Water)	$(SO_4)^{2-}$(i) = 204,352	Na^+(i) = 58.98	NH_4^+: 76,625
		$(SO_4)^{2-}$(f) = 69,628	Na^+(f) = 0	NH_4^+: 26,100
	Feed (Ammonium sulfate–Water)	$(SO_4)^{2-}$(i) = 0	Na^+(i) = 0	NH_4^+: 0
		$(SO_4)^{2-}$(f) = 12.36	Na^+(f) = 432.52	NH_4^+: 4
4	Draw solution (Ammonium sulfate–NaCl)	$(SO_4)^{2-}$(i) = 188,902	Na^+(i) = 98.3	NH_4^+: 70,838
		$(SO_4)^{2-}$(f) = 78,692	Na^+(f) = 235.9	NH_4^+: 29,510
	Feed (Ammonium sulfate–NaCl)	$(SO_4)^{2-}$(i) = 0	Na^+(i) = 1016.1	NH_4^+: 0
		$(SO_4)^{2-}$(f) = 0	Na^+(f) = 1769.4	NH_4^+: 0
5	Draw solution (Potassium chloride–Water)	K^+(i) = 32,980.5	Na^+(i) = 275.24	Cl^-: 30,030
		K^+(f) = 24,081	Na^+(f) = 98.3	Cl^-: 21,920
	Feed (Potassium chloride–Water)	K^+(i) = 0	Na^+(i) = 0	Cl^-: 0
		K^+(f) = 15.705	Na^+(f) = 13.56	Cl^-: 14.30
6	Draw solution (Potassium chloride–NaCl)	K^+(i) = 65,749	Na^+(i) = 2,950	Cl^-: 59,850
		K^+(f) = 37,244	Na^+(f) = 490	Cl^-: 33,820
	Feed (Potassium chloride–NaCl)	K^+(i) = 0	Na^+(i) = 1,016.1	Cl^-: 0
		K^+(f) = 0	Na^+(f) = 1,769.4	Cl^-: 0

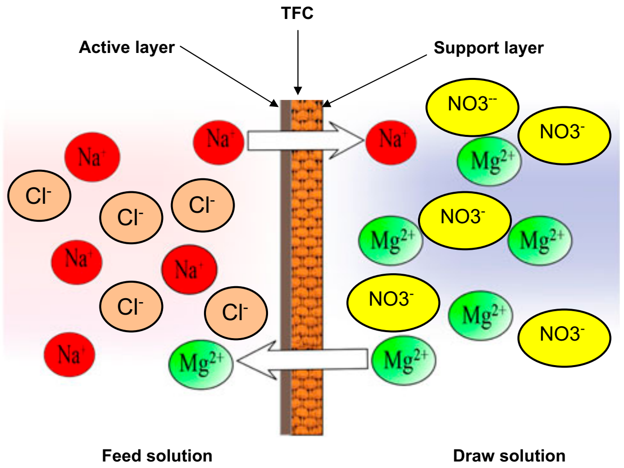

Fig. 3. Donnan exclusion.

4. Conclusion

We have tried to design low energy consuming FO-based desalination process by converting the domestic RO membrane element into FO membrane element and we have come to certain outcomes as listed below:

(1) The use of fertilizers as a draw solution leads to elimination of second step i.e. recovery of water from draw solution which is a major challenge in FO-based separation process because it is very tedious and require high energy for separation of water from draw solution.

(2) The ratio of flux with NaCl solution to flux with DI water ranges from 0.4162 to 0.6311 in the fertilizers taken for study.

(3) Theoretical flux is significantly higher than experimental flux because of external and ICP and inaccessibility of the draw solution to entire membrane area.

(4) The reverse salt diffusion was observed in all three fertilizers studied with water as feed, however it was absent when sodium chloride feed solution was used. This shows that the presence of salt on feed side of the membrane suppresses the reverse salt diffusion on account of Donnan exclusion.

(5) This work opens the opportunity for future work to study the bottlenecks of the system and address them to exploit such application at larger scale.

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