

Influence of the process parameters on hollow fiber-forward osmosis membrane performances

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Abstract

Continued efforts are made in improving the performance of the low-cost forward osmosis (FO) membrane process which utilizes naturally available osmotic pressure of the draw solution (DS) as the driving force. Selection of a suitable DS and development of a better performing membrane remained the main research focus. In this study, the performance of a hollow fiber forward osmosis (HFFO) membrane was evaluated with respect to various operating conditions such as different cross-flow directions, membrane orientation, solution properties, and solution flow rates (Reynolds number). The study observed that operating parameters significantly affect the performance of the FO process. FO comparatively showed better performance at counter-current orientation. NaCl, KCl, and NH₄Cl were evaluated as DS carrying common anion. Properties of the anionic part of the DS were found important for flux outcome, whereas reverse solute flux (RSF) was largely influenced by the properties of DS cationic part. FO was operated at different DS and feed solution (FS) flow rates and FO outcome was assessed for varying DS and FS Reynolds number ratio. FO showed better flux outcome as Re ratio for DS and FS decreases and vice versa. Results indicated that by adjusting FO processes conditions, HFFO membrane could achieve significantly lower specific RSF and higher water flux outcome. It was observed that using 2 M NaCl as DS and deionized water as FS, HFFO successfully delivered flux of 62.9 LMH which is significantly high compared to many FO membranes reported in the literature under the active layer-DS membrane orientation mode.

Keywords: *Hollow fiber forward osmosis (FO) membrane, flux, RSF, hydrophilicity, boundary layer effects, DS, flow rate*

1. Introduction

Recently, in the last decade, forward osmosis (FO) technology has shown its future potential for various water desalination and separation processes. Using the natural osmotic pressure potential of draw solution (DS), it is gaining popularity over other thermal and membrane

based desalination techniques which consume extensive energy for water separation processes. In a very short span of time, the low cost FO process has been studied for various useful applications [1-15]. FO rapid evaluation for such a high number of applications indicates that the low energy FO desalination concept is really being overwhelmingly welcomed by the research community and industry.

Membrane and DS characteristics greatly affect the FO performances in such a way how these two facilitate water molecules movement through the membrane surface. In earlier FO studies, low performance ratio (PR), a ratio of actual flux to the theoretical flux, has highlighted some serious issues in FO process which are closely linked with the suitable DS selection and FO membrane characteristics [10, 16-19]. A wide range of both inorganic and organic DS have been evaluated for FO process which indicated varied performance outcome in term of flux and reverse solute flux (RSF) [20-24]. However, in selecting a suitable DS selection, DS recovery and its separation from the permeated water are still considered as the most challenging issue for FO process. DS separation and permeate recovery from the diluted DS are complex and energy intensive steps and if not properly addressed, may mitigate the real potential of FO process.

Due to the nature of FO process, system design and its operation, mostly, asymmetric FO membranes were developed by casting a very fine FO membrane active layer (AL) on a thick porous support layer (SL). This asymmetric membrane structure usually help developing concentration polarization (CP) progression in the SL. CP causes lowering of the net available osmotic pressure gradient available for osmosis which results in lower permeate flux and low PR for FO process. Internal concentration polarization (ICP) is considered as the most critical barrier in getting better performances from any particular FO membrane systems [25-27]. To overcome RSF and FO PR issues which seem directly linked to the FO membrane characteristics, various research groups focussed their attention to develop a very thin membrane AL on a highly porous support layer [18, 28-30]. Some of their work was focussed on AL improvement [31] while the others aimed their activities to improve FO outcome through SL modification [30, 32-34]. Following Hydration Technology Innovations (HTI) work, who initially produced their commercial flat sheet membrane, some other companies also introduced their flat sheet membranes and used them for various applications [1-3, 5]. Yip et al. [35] pioneered TFC- FO flat-sheet membrane using PS support.

Later, various other research groups also endeavoured to fabricate a suitable FO membrane with better operational performances. FO performance was evaluated for new kinds of FO membranes as hollow fiber FO (HFFO) membrane [36], polybenzimidazole (PBI) nanofiltration membrane [37], crosslinked layer-by-layer (xLbL) FO membrane [29], layer-by-layer polyelectrolyte applied on a PES hollow fiber substrate [38], high flux FO membranes by chemically crosslinked layer-by-layer polyelectrolytes [39], novel poly (amide-imide) FO hollow fiber membranes with a positively charged selective layer [28], TFC FO membrane for PRO [40], FO membrane with sulfonated polyphenylenesulfone (sPPSU) as the supporting substrate [41], Cellulose acetate nanofiltration hollow fiber membranes for FO [42] cellulose acetate membranes for forward osmosis with an ultra-thin selective layer[43] and others[33, 44-46].

Some other unique and distinctive nanofiltration membranes, carbon nano tubes[47], aquaporin membranes [48] are also used for forward osmosis applications. Although, most of the AL of flat sheet membranes were initially prepared using cellulose acetate (CA) and cellulose tri acetate (CTA) followed by Poly amide (PA) thin film composite (TFC) materials

[39], for hollow fiber membranes, most of the attempts were made with PA while few also used CTA active layer.

For enhanced FO performance results, along with continued efforts to select a suitable DS [23] and improve membrane characteristics, side by side, effect of various FO operating parameters such as temperature [49, 50], flow direction, membrane orientation [26, 51, 52], flow rate or velocity [17, 53, 54], viscosity [55] has also been evaluated in some earlier studies which demonstrated fluctuating FO performances.

On the whole, results have shown that HFFO membranes mostly showed far better results for water flux for FO operations. In an earlier study (Tahir et al. 2013), HFFO membrane was also evaluated under the same conditions for different fertilizer DS and compared with HTI's CTA membrane outcome. It was seen that although in this study, HFFO gave better results but not as par with other published work showed with HFFO membranes [18, 40, 42, 56].

It is also interested to note that in most of the above cited literature, some of the FO performances were not evaluated on similar operating conditions thus it is very difficult to track any real fluctuation in FO performances which may lead you towards a suitable direction for improvement. Looking into the variations of these performance outcome, Cath et al. [55] emphasized the importance of following some standard protocols to carry out FO experiments with a set of pre-defined operating conditions.

Beside all these continued efforts to improve FO performances, very few attempts were made to understand how the water and solute molecules transport across AL and SL of the membrane is affected. These areas were not properly explored in depth thus insufficient published literature is available on these important issues. Water transport patterns within the membrane and SL structure may provide solution to flux, RSF and CP issues. The water transport phenomenon is directly linked with the DS and feed solution (FS) properties and membrane characteristics as well. Both effect each other during osmosis process through membrane pores.

However, overall, from the varying results of the previously cited work, we hypothesized that along with the membrane characteristics and DS properties, specific operating conditions also exhibit very important roles in delivering improved performances with flat sheet and HFFO membranes. From this, it is hypothesized that even FO process and operating conditions may have more pronounce effects on FO performance.

This study evaluated performance of hollow fiber forward osmosis membrane for varying operating parameters especially the flow rates and membrane orientation.

The purpose of this study was to evaluate role of some FO process operating parameters expected to significantly affect the FO performance outcome. HFFO membrane was evaluated to assess the effects of membrane orientation, changing DS and varying DS and FS flow rates and to understand how and to what extent these parameters may affect the FO performance of the same membrane. FO membrane was tested for AL-DS and AL-FS orientation. NaCl, KCl and NH₄Cl were evaluated as DS to evaluate how a cationic or anionic part of DS modify FO outcome. Later, FO was operated at varying DS and FS flow rates and FO performances were assessed for varying DS and FS Reynold no. (Re) values.

2. Materials and Methods

The bench scale FO system, similar to one used in a previous study, was used to evaluate performance of HFFO membrane. HFFO lumens were supplied by Samsung Cheil Industries, Korea which were used to prepare different size modules. FO lumens were supposed to be made up of inside polyamide (PA) thin film composite (TFC) active layer on hydrophilic sulphonated polysulphone (SPSf) substrate. Most of the experiments were completed using modules with hollow fibre lumens membrane area of 0.04 m^2 , whereas other modules with different membrane area were used to re-confirm the results outcome.

NaCl was used as a primary DS for most of the results outcome. Some other monovalent DS carrying chloride (Cl) as their common anionic part such as KCl and NH_4Cl were also used for supplementary studies to evaluate how cationic and anionic parts of the DS behave differently for HFFO tests. All these chemicals were supplied by Chem-Supply, Australia. Water permeation flux was calculated by recording the DS/FS tank mass changes in unit time using a weighing balance.

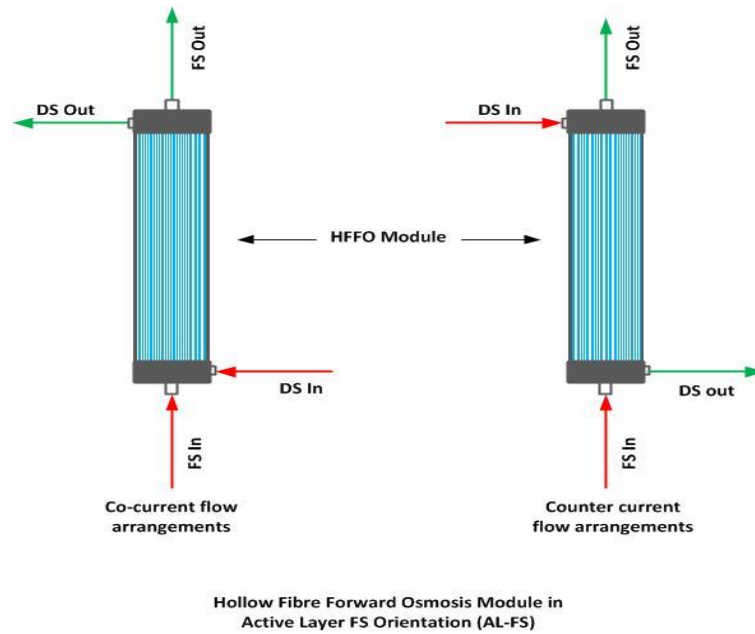
The temperatures of the DS and FS were maintained at $25 \pm 0.5^\circ\text{C}$ using a water bath linked with a temperature controller. FO tests were completed on AL-DS and AL-FS membrane orientations. For AL-FS membrane orientation, FS was flown through the fibre and DS was flown in the shell outside the fiber. Similarly, for AL-DS membrane orientation, FS was flown in the shell area outside the fibers whereas DS was flown inside the FO hollow fiber. In AL-FS, water permeates through the FO fiber in In/Out (I/O) direction whereas AL-DS uses Out/In (O/I) direction for water permeation. H270G-BNDL conductivity meter was used to regularly monitor FS conductivity to evaluate any reverse solute flux (RSF). Conductivity curves were drawn with all DS for their low and high concentrations. RSF of the FS was then evaluated using these standard conductivity curves for each DS.

3. Results and discussion

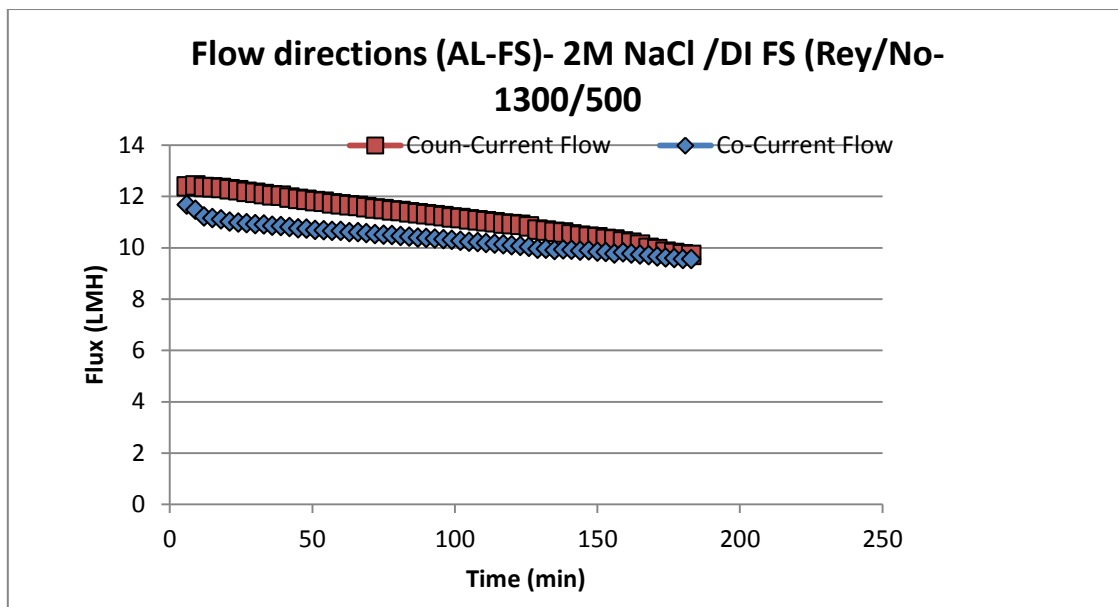
Effect of various operating parameters as flow direction, type of DS, membrane orientation and varying DS and FS flow rates on FO performances was evaluated. Some of these parameters affected FO flux marginally whereas others showed significantly enhanced results as discussed later.

3.1. Effect of flow direction on FO performance

Fig. 1 compares HFFO flux outcome when FO was operated in two different flow arrangements. In co-current flow arrangement, DS and FS enter the FO module from same side and leave it from the other side. Contrary to that, in counter-current flow arrangement, DS and FS enter the FO module in opposite sides of the module, follow different flow directions and leave the module from opposite directions Fig. 1 (a). The experiments were performed at DS and FS flow rate represented Re of 1300 and 500 respectively. Fig. 1(b) compares flux outcome when the flow directions were changed from co-current to counter-current in HFFO operation. HF membrane shows higher flux at counter-current flow arrangements. In counter-current orientation the net osmotic pressure ($\Delta\pi$) at the different DS and FS entering points (inlet) is comparatively high than in co-current mode. The high flux outcome suggests that for most part of the HFFO module, the FO driving force i.e., $\Delta\pi$ is slightly high in counter-current arrangement than in co-current arrangements. DS and FS meet HF cell membrane with comparatively higher $\Delta\pi$ than in co-current arrangement. However, as due to higher flux and high water permeation rate, the DS is diluted quickly with time, $\Delta\pi$ also reduces faster in counter-current arrangements. As the test continued, flux nearly became equal after some time and later due to rapid dilutions, FO in co-current



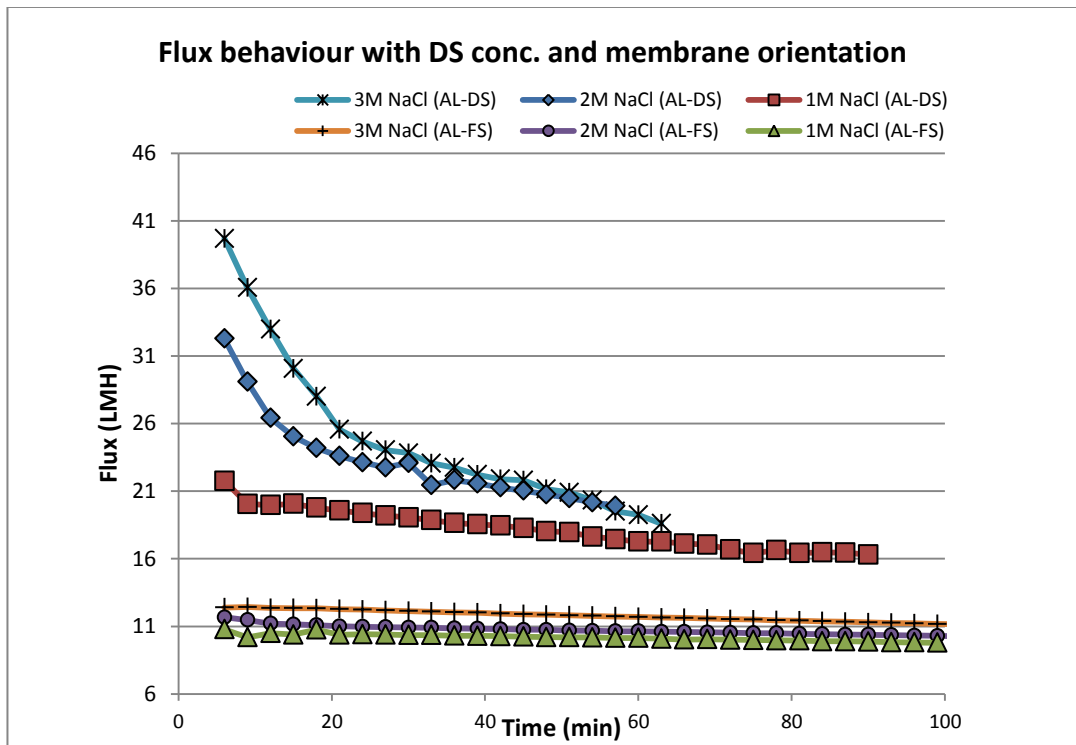
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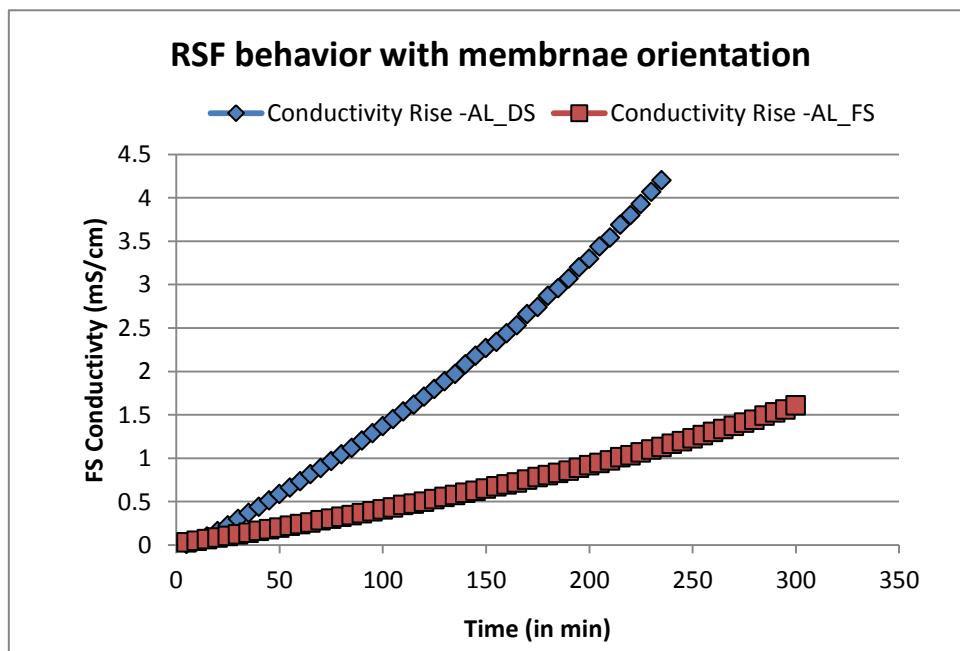
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Fig. 1. a) Showing DS and FS flow directions in Co-current and counter current arrangement for FO experiments b) FO performance for different flow directions in AL-DS membrane orientation

arrangement showed comparatively lower flux for counter-current arrangements. However, it is noticed that for a longer run both set of flow arrangements nearly give the same permeated water. Jung et al. [53] also confirms that flow direction does not have any important effect on the FO flux performance in a small module.



a)



b)

Fig. 2. a) Effect of changing membrane orientation on flux outcome for 1M, 2M and 3M NaCl DS b) Effects of RSF on FS conductivity with time

3.2 Effect of membrane orientation on FO performance

Fig. 2(a) displays flux outcome when HFFO membrane module was operated for two different membrane orientations i.e., AL-FS and AL-DS. For AL-FS, PA rejection layer was facing FS whereas for AL-DS, membrane active layer was facing DS. The results indicated an immense increase in flux outcome on AL-DS membrane orientation. Comparison to AL-FS membrane orientation, HFFO membrane delivered upto 202%, 293% and 340% higher flux with AL-DS orientation for 1M, 2M and 3M NaCl DS respectively. Flux did not vary significantly for AL-FS orientation as it showed an increase of just 8% when DS concentration was changed from 1M to 3M. Whereas at AL-DS orientation, the same DS

concentration change showed 82% rise in flux. By increasing DS concentration, the flux rises more in AL-DS membrane orientation although this rise was not linear for higher DS concentrations. These difference in flux for AL-DS and AL-FS orientation reveal that in AL-FS orientation, CP builds quickly in the membrane SL, reduces the available osmotic pressure difference ($\Delta\pi$) at the membrane interface thus demonstrate lower flux outcome [26, 46, 57, 58]. The flux rise was small in AL-FS orientation as with the increasing DS concentration, CP also increases sharply and thus flux did not rise as expected. These results clearly indicate enhanced performance by HFFO membrane at AL-DS orientation.

However, from the previous theories [25, 59, 60] suggest that concentrative and dilutive ICP starts once the water started to permeate through the membrane. With time a dilutive and concentrated stream develops in the AL and SL which enhance ICP effects. However, in Fig. 2, in AL-FS orientation, flux immediately showed very low flux beside that at that time the adjacent stream at the membrane AL-and SL were not diluted or concentrated enough to represent the severe effect of CP. Some additional studies are required to highlight reasons for the low FO flux outcome from the start of the test in these two membrane orientations. FO processes involve concurrent contact of both DS and FS on the top and bottom membrane surfaces and water transport through the semi-permeable membrane is based on the $\Delta\pi$ available on the membrane surface. From the above results we suggest that CP build up may not be the only parameter resulting huge difference in FO performance or even if it there, it does not support the concepts of dilution and concentration of DS and FS layers near the membrane AL and SL.

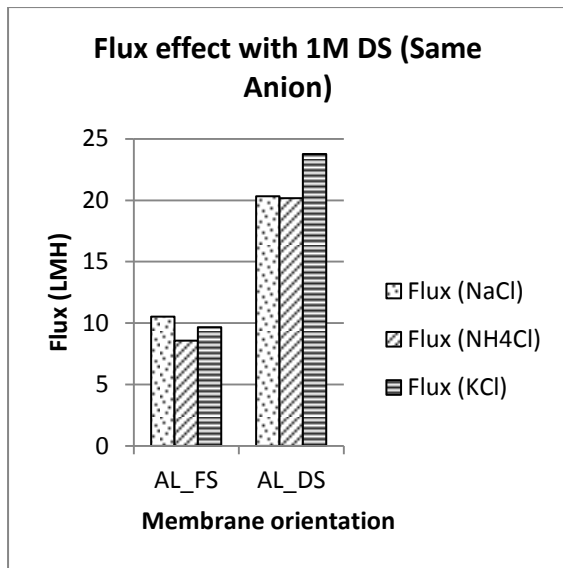
FO membrane substrate physio chemical properties as hydrophilicity, porosity, pore size, pore-size distribution play very important roles in the overall FO performance [39]. Han et al.[61] has highlighted role of physicochemical properties of the substrates. By using a sponge type hydrophilic sulphonated polyether ketone (SPEK) support structure in the fabrication of TFC flat-sheet FO membrane, water flux of 50 LMH and 35 LMH was achieved using DI as the feed and 2 M NaCl as the DS for AL-DS and AL-FS membrane orientation respectively. DS and membrane AL and SL develop some associations in different ways in both AL-DS and AL-FS orientations to build this phenomenon that leave huge gap in flux outcome for FO results.

Fig. 2(b) represents FS conductivity rise due to RSF in FO process for both type of membrane orientations. It is observed that in comparison to FO operation at AL-FS orientation, reverse salt passage was high in AL-DS orientation. Conductivity of the FS rises more quickly in AL-DS orientation. The same DS molecules behave differently on both side of the membrane indicate the important role of support layer (SL) for the resultant flux. Support layer properties such as its charge, hydrophilic or hydrophobic nature seem more important for this behaviour. We suggest that the most important of these is hydrophobic or hydrophobic properties of the membrane. This is similar to hydrophobic membranes used for membrane distillation which beside carrying large pore size (0.2 micron) [62] don't allow water molecules (0.28 nm size) to permeate through membrane surface. Membrane or AL charge plays secondary roles in this diffusion processes. Fig. 2(a) and 2(b) confirm that the DS approached closer to FO membrane's active layer in AL-DS orientation and thus it facilitates the diffusion of the salt through the membrane. Again for FO operations with DI feed, Donnan equilibrium theory for solute transfer seems unable to provide reasons for this high salt flux.

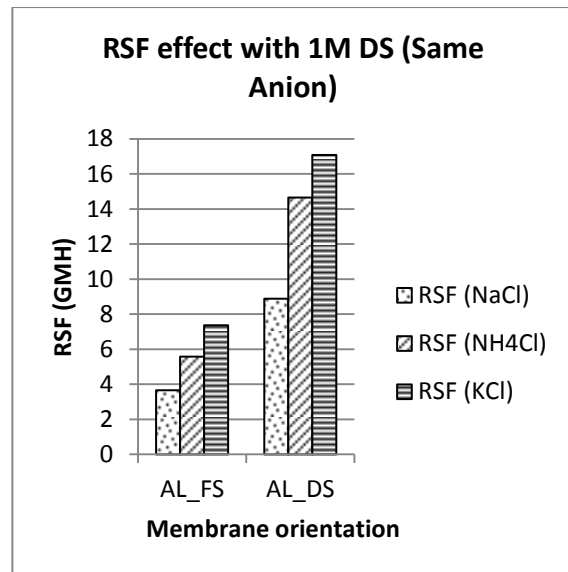
In FO, water permeation (flux) and salt molecules diffusion (RSF) take place in opposite directions. Water molecules move from FS side to DS side whereas solute movement is from DS side to FS side. Due to this opposite flow directions of molecules, water flux and RSF movement should apparently effect each other, resist other stream movement and cause slowing down of their movement. Contrary to that, rather, in this case, it was observed that RSF increases as the water flux increases in AL-DS orientation. This further confirms that movement of DS and FS stream does not affect other's stream movement as different forces might be responsible for water flux and RSF which to some extent, are independent of each other's movement or don't effect the performance of the other at all. They either use different pores for diffusion with diverse phenomenon for their water molecule movement.

We also noticed that in AL-DS membrane orientation, water flux declines distinctly in the initial run and then slope declines. This sharp flux drop does not follow the usual flux declining patterns of other flat sheet FO experiments [20, 25, 26, 63] and HF in AL-FS orientation (This study). There may be two reasons for that. Firstly since the membrane used in these experiments was carried high surface area (0.04 m^2) which is 20 times more than membrane areas used in most of the flat sheet FO studies, more water permeates in unit time which quickly dilute the DS, reduces $\Delta\pi$ and thus force flux to decline sharply at the start. Later as the flux decreases with time, it depresses the DS rapid dilution which reduced the flux slope as in the later period, $\Delta\pi$ does not change too quickly.

Along with high flux outcome at AL-DS orientation, HFFO also showed high RSF at AL-DS membrane orientation. Due the high RSF, salt concentration in the FS increases rapidly and this cause reduction in available osmotic pressure ($\Delta\pi$) which causes flux to decrease with time. Chou et al. [36] further revealed that compared to flat sheet FO, as HFFO membrane takes little more time to build-up salt concentration in substrate and to develop steady ICP, hence HF membrane flux declines sharply at the initial stages of the test run. When a steady ICP is developed in the substrate, HFFO membrane then starts giving linear flux.



a)



b)

Fig. 3. Showing flux and RSF for 1M NaCl, NH₄Cl and KCl DS. These three DS have different cationic parts but same anion i.e., Cl⁻

3.3 Effect of DS type on FO performance

Three DS NaCl, NH₄Cl and KCl were used to evaluate effect of various DS carrying common anion but different cationic parts for flux and RSF. Fig. 3 shows flux and RSF outcome for HFFO when 1M NaCl, NH₄Cl and KCl were evaluated. It shows that both flux and RSF increases for these three DS when FO is operated in AL-DS orientation. Further, it is observed that flux did not change significantly for these DS both in AL-FS and AL-DS orientations. However, RSF changes markedly for these three DS for both AL-FS and AL-DS orientations. Salt transport through the membrane is influenced by both charge effects and size effects[64]. However, still the RSF outcome is contrary to that as Na⁺ with higher hydrated radii showed lower RSF than K⁺ and NH₄⁺ which carry nearly the same hydrated radii.

3.4 Effect of flow rate on HFFO performance

In order to find out effects of changing flow rates on FO performance, FO was operated for varying FS and DS flow rates adjusted by pump valves. These experiments were done for both AL-FS and AL-DS membrane orientation. DS concentration was changed from 1M to 5M NaCl whereas DI water is used as FS. A set of two flow rates representing Re 200/500 and 600/1600 for DS and FS were selected for these tests. First number of the fraction represents DS Re whereas the other represents FS Re.

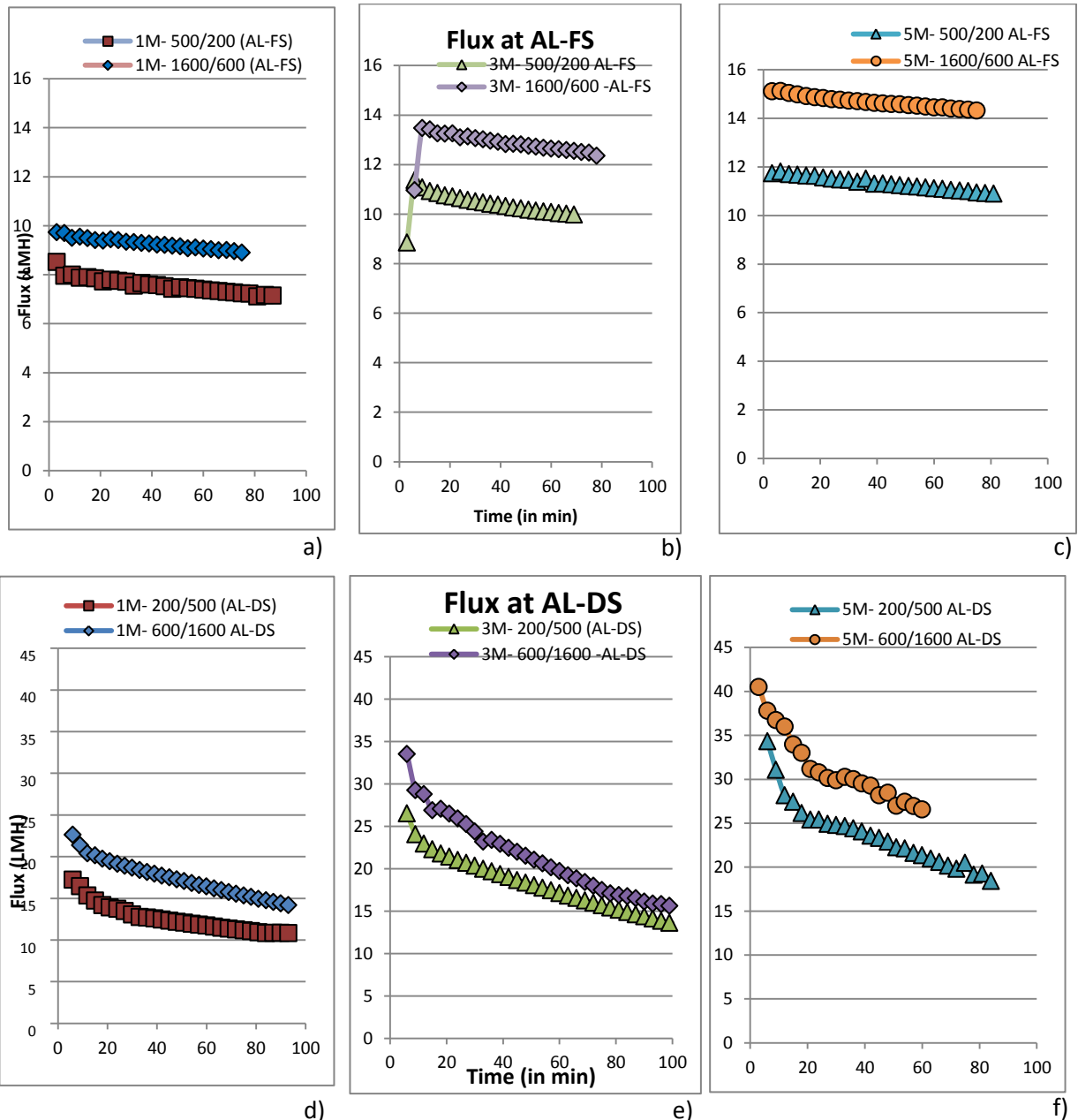
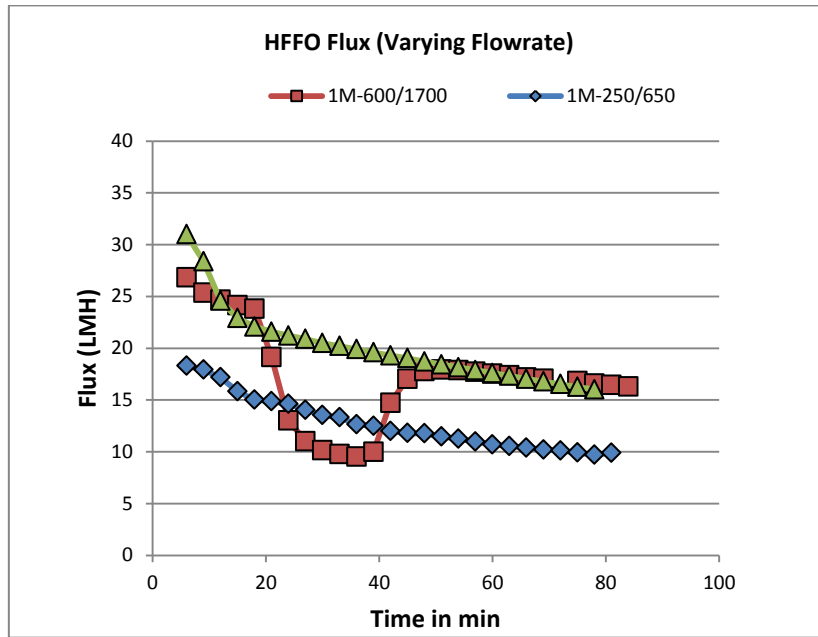
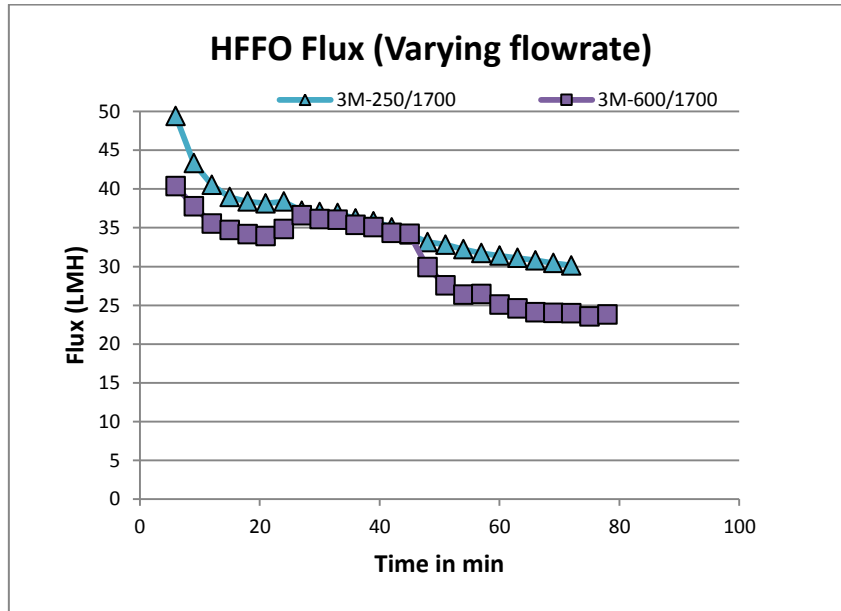


Fig. 4. Effect of changing DS/FS flowrate on HFFO membrane water flux. a, b and c) On AL-FS orientation for 1M , 2M and 3M NaCl DS respectively and d, e and f) On AL-DS orientation for 1M , 2M and 3M NaCl DS. Legends show Re for DS and FS respectively.



a)



b)

Fig. 5. Effect of changing flow rate of only one stream (either DS or FS) on FO flux a) Flux dropping pattern with 1M NaCl DS when during the test run FS Re was reduced from 600 to 250 and then restored to 600 b) Flux elevation pattern when DS Re was reduced from 600 to 250 and then restored back to 600. Both tests were performed at AL-DS membrane orientation.

Fig. 4 represents HFFO membrane flux outcome for different flow rates. FO showed enhanced flux outcome for AL-FS and AL-DS membrane orientations when flow rates were changed from one set to another. In AL-FS membrane orientation water flux increases by 22%, 18.46% and 28.8% for 1M, 3M and 5M DS concentrations. Similarly, FO membrane showed 37.56%, 31.71% and 17.98% higher flux for 1M, 3M and 5M DS concentration for the above given Re values.

3.5 Effect of changing DS and FS flow rate

Effect of flow rates representing various Re on FO flux was further evaluated when some of the tests were started with a specific set of flow rates and after short test run, the flow rate of either FS or DS was changed to reflect a new set of Re. values for that stream. After short test run at revised Re, both DS and FS flow rates were restored to the initial values. Fig. 5 presents the flux outcome when flow rate of only FS and DS streams were changed during FO test. Fig. 5 (a) shows flux when HFFO membrane was initially operated at the Re. nos. of (DS) 600/ FS (1700) using 1M NaCl DS against DI feed. After about 20 minutes, FS flow rate was reduced to reflect Re. no. of 650. It was observed that by reducing FS flow rate, FO flux was reduced sharply. Overall about 60% of the flux decline was observed by reducing FS Re. no. from 1700 to 650. After 20, minutes when the flow rate of FS was again increased to restore initial Re. no. for FS, flux restored back to its normal position. This result was further compared with two other FO outcomes Fig 5(a), one when HFFO was operated for 1M NaCl DS at different Re. nos of 250/650 and with 3M NaCl DS operated at lower Re. no. of 250/650. It was noticed that 1M NaCl DS at higher Re. no. (600/1700) operation showed better flux output as 3M NaCl DS gives at lower Re.no. (250/650).

HFFO membrane was further evaluated for another set of test using 3M NaCl DS against DI feed Fig. 5(b). FO test was initially started for DS and FS Re. no. of 600/1700 respectively. Opposite to the above pair of tests, the flow rate of the DS was reduced to represent Re. no. of 250. It was noticed that by reducing the DS flow rate, FO quickly showed increase in flux and an about 8% flux increase was noticed. However after 20 minutes when the flow rate was reduced to the initial start-up value, flux reduced back by 20.2%. Again, a reference test was run with 3M NaCl DS at Re. no. of 250/1700 and flux outcome was compared. with the respectively . From this we may deduce that the HFFO membrane comparatively gives high flux outcome when DS flow rate is reduced. On the other hand, when FS flow rate is decreased, the resultant flux is also decreased.

Same HFFO membrane showed interesting variation for flux while adjusting DS and FS flow rates and membrane orientation. This huge flux difference is evaluated below in the light of Some theories are being presented to Hereunder we try to present some approaches how the water and salt molecules would cross the membranes which induce high difference in flux for these varying operation parameters.

Comparing permeate flow mechanism through membranes as MF, UF, RO, NF and FO, it is clear that MF or UF does not show any rejection of water molecules and small solutes cross the membrane. Both these follow the same permeation rate and are readily rationalized by the filtration-type mechanism or a pore-flow membrane. PSF support layer usually showed pore sizes of 20-25 nm [65]. Due to the large pore size of membrane SL, it is suspected that the PSF support layer don't offer any molecular resistance to salt permeation and direct salt diffusion occur. As the FO support layer exhibited properties of an ultrafiltration membrane thus the membrane's AL appears mainly responsible for the salt rejection and water permeation in FO process.

For NF, RO and FO, water molecules pass through the membrane easily as they carry size smaller than the membrane pore and the same membrane hinder the passage of all larger size solutes particles [66]. RO, NF and FO seem to follow solution diffusion mechanism and from membrane classification by pore size, the transition between a pore-flow and a solution

diffusion mechanism seems to occur with membranes having very small pores [67]. Solution diffusion theory does not properly explain this permeation process through the membrane. Water or solute ions should overcome mechanical and electrostatic barriers to flow through membranes.

Ignoring the pore flow issues, for the same kind of SL, all FO performances should rely on the properties of AL the membrane. However, FO has shown higher flux with modified SL having highly hydrophilic nature [39]. We therefore deduce that by increasing hydrophilicity of the membrane AL and SL, the boundary layer can be made become thin and weak which helps water molecules come closer to membrane surface to permeate towards the other side which results higher flux.

FO membranes were designed to use solution - diffusion mechanism and superior separation performance of solute from water which require adequate pore size control to allow water molecule carrying an average size of 0.28 nm pass through the membrane while retain all ions and salts from both DS and FS side. Surprisingly, beside that the most of the solutes also have hydrated radii of sizes close to water molecule size; they are retained by FO membranes. This indicates that either membrane carry unique pore structure allowing water molecules to pass easily and retain majority of solutes on either side of the membrane. The question is still unanswered if water molecules pass the membrane using size greater than its molecular size only or it also passes from the other pore. FO membrane performances suggest that instead of following solution-diffusion or pore-flow theory, water or salt molecule diffuse through the membrane using their particle and membrane charge. Membrane charge help to hold certain particles and ion while facilitate others to cross quickly regardless of the size.

Cellulose acetate (CA) membranes have pore size of 3.88 Å [66] which is sufficiently large for water molecules to pass through, but the results with all CA and cellulose triacetate FO membranes record very low flux. Membranes form a boundary layer with liquids streams in contact with and these boundary layers on either side of the FO membrane induce significant resistance for water permeation and salt diffusion. The transport resistance of this boundary layer may be a function of many factors such as shape of the interface, wettability of the surface to the liquid, hydrophobicity or hydrophilicity, velocity of the liquid at the interface [68]. Overcoming resistance of the boundary layer, how far and how quickly the liquid water penetrates into the membrane is the most important factor in the overall transport property of a porous membrane. The water penetrated into the pores of the membrane and its support layer can be hardly affected by stirring of the bulk water or turbulence due to water flow.

NF membranes having good rejections to divalent ions and most organic solutes, but rejection of monovalent ions in the 20-50% range [67]. Different solute molecules varying hydrated molecule sizes cross the membrane showing that for small solutes, even more dia is not so relevant but the selective permeability of these membranes suggest that solute follow some other diffusion process. Potassium ions diffuse rapidly across cell membranes through proteins called K^+ channels and these channels use diverse mechanisms of gating (the processes by which the pore opens and closes), but they all exhibit very similar ion permeability characteristics [69].

In a negatively charged membrane pore, positively charged hydrogen atom enters first whereas in positively charged membrane pores, oxygen atom enters first. In a negatively charged membrane pore, hydrogen atom tends to go closer to the pore wall, giving rise to higher electrostatic interactions with the wall. Thus, the water molecules in the negative

pore experience a higher mean force due to wall-water electrostatic interaction compared to a positively charged pore. Water molecules move in the form of chain and probability of water chain to be broken is lowest in the negatively charged pore which causes a higher osmotic flux through a negatively charged membrane.

Further, in AL-DS membrane orientation (DS flows inside the hollow fiber and FS flows in shell outside fiber), DS moves within HFFO making swirling movements that helps create some vacuum along the inner walls of the hollow fiber which help pull water molecules to permeate quickly thus HFFO membranes in AL-DS orientation deliver high flux at high flow rates especially when the DS flow rate is increased in comparison to FS flow rate. The flux did not vary too much by changing DS and FS flow rates of the flat sheet FO membranes [18, 33, 40, 41, 53].

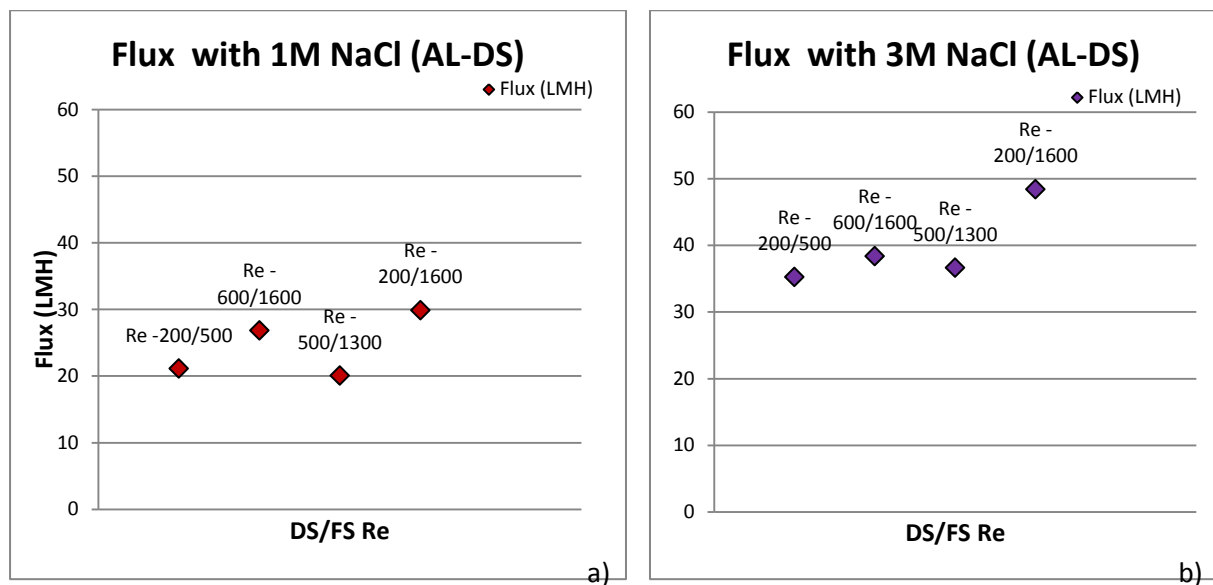


Fig. 6. Effect of changing DS and FS flow rates on FO flux performance with a) 1M NaCl DS b) 3M NaCl DS

No concrete theories have been yet found authenticating how a fingle-like or sponge type substrate strucures aparently faciliate water permeation throught the membrane. Their strucures does not seem provide any support in either the pore-flow or solution diffusion transport mechanism. Most of the nanoporous membranes do not show a uniform pore distribution and geometry. From the fluctuating flux results for HFFO in AL-DS orientation at different DS and FS flowrates, we deduce that various set of DS and FS flow rate values develops some pressure inside the hollow fiber. This assists membrane structure to stretch and modify. Irregular shaped pores are changed to uniform size pores which allow water molecules easily pass through the membrane and thus high fluxes are obtained. We conclude that with the new pore shape arrangements, the pore structures also becomes align with each other, symmetrical throughout the membrane depth and thus resembles uniform structures close to the CNT membrane structures which have shown higher water flux [47, 65].

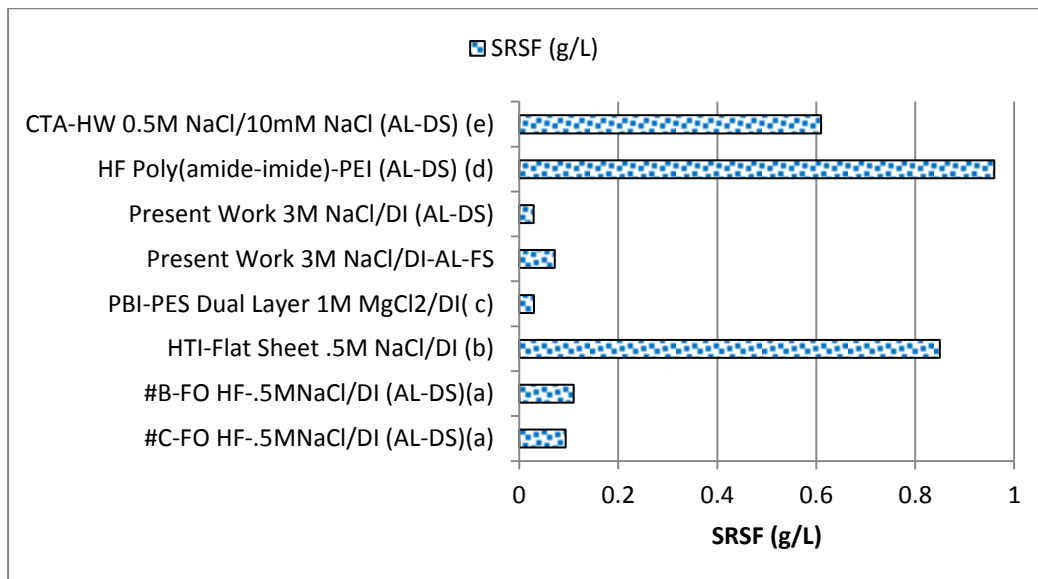


Fig. 7. SRSF comparison-Present work with literature data (a)[36], (b)[58], (c)[56], (d)[28], (e)[70]

It was observed that there is a particular set of FS and DS flow rate which help get highest flux through the FO membrane. Other higher or lower DS and FS flow rates showed comparatively lower flux. From this we may deduce that membrane pores, being made of an elastomeric properties material, change shapes with pressure and flow and at some specific values of these process parameters, they form good arrangement of the pore shapes which facilitates both water and solute flux. By changing operating parameters from these optimum values, the pores again changes back to disorder structure and which don't show the same enhanced performance. This further confirms the importance of interfacial polymerization (IP) process of the membrane making step where the polymeric material develop various pore sizes, structures and arrangements of the membranes.

3.6 Effect of changing flow rates of FO performance in AL-DS

Jung et al [53] emphasized the role of flow rate affecting the mass transfer within the external CP layer to optimize FO operations in terms of energy consumption and production recovery. Looking into the above results, FO was further operated in AL-DS membrane orientation for varying DS and FS flow rates representing different Re. no. sets and results are summarized in Fig. 6. It was found that HFFO gave better results at DS and FS Re. no. values 200/1600 for both 1M and 3M DS. It was also noticed that the flux increases with the decrease in DS/FS Re. no ratio. For 1M NaCl DS, flux is increased by 41% Fig. 6(a) whereas 3M NaCl DS showed a flux increase of 37% Fig. 6(b) when DS/FS Re. no were increased from 200/500 to 200/1600, which reflects DS and FS Re.no reduced from 0.4 to 0.125.

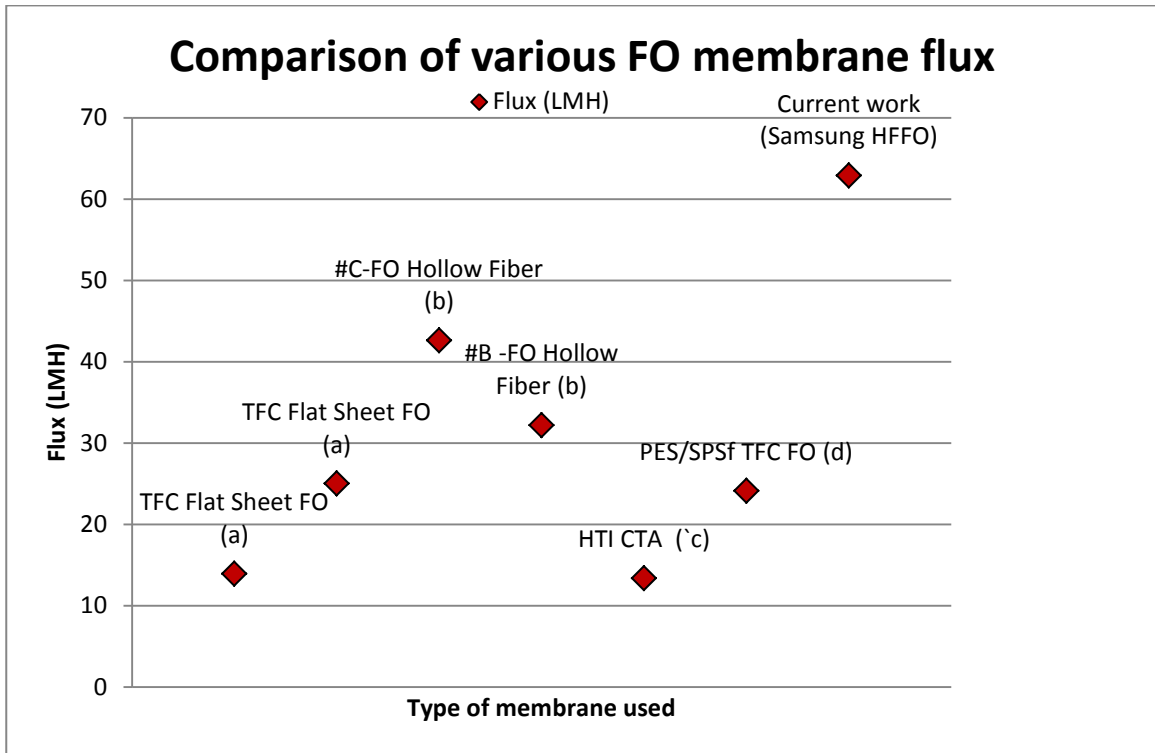


Fig. 8. Comparison of flux outcome used by various researchers (a)[34], (b) [36], (c) [26], (d) [39]

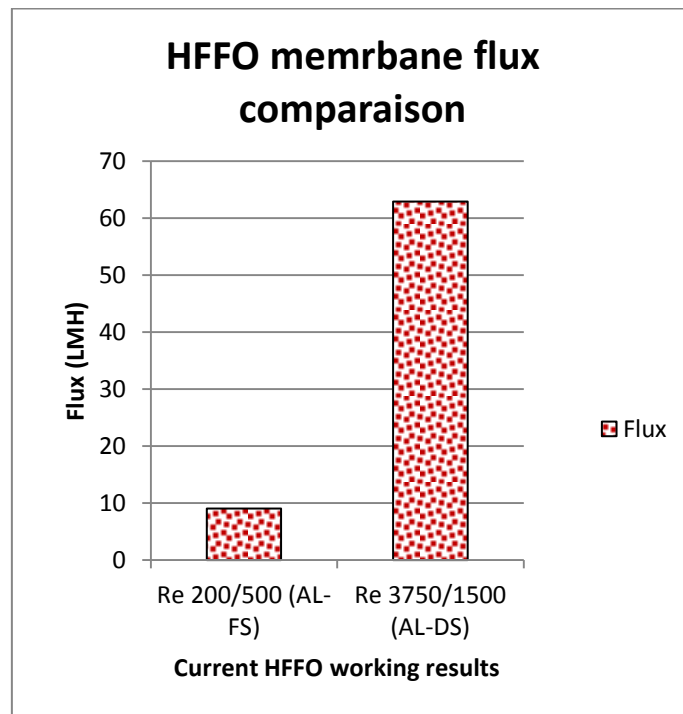


Fig. 9. Flux outcome showing effect of operating conditions.

3.7 Comparison of SRSF and flux performances

Specific reverse solute flux (SRSF) was evaluated for 3M NaCl DS in AL-FS and AL-D membrane orientation and a comparison with the earlier published work is presented in Fig. 7. The comparison shows that with some adjustments in the FO process conditions, HFFO membrane gave the lowest SRSF even wither operated at higher DS concentration for both AL-FS and AL-DS membrane orientation.

Following Fig. 6 results, HFFO was further tested at various other DS and FS flow rates. HFFO showed remarkably high flux of 62.9 LMH when FO in AL-DS orientation was operated at Re. no. 3750/1500 and a comparison with some earlier research work is presented in Fig. 8. It showed that the current work results stand alone in this comparison. The variation in these FO flux performances is reflected mainly due to the changes in operating parameters and not for the changing membrane characteristics. Finger- like structure of the support layer and sponge like support layer structure have been evaluated in the literature which showed some variation in FO performances. However, none of these are unable to explain how these structure enhance changes in FO water flux as both these SL structures don't advocate water permeation through he support structure.

Furthermore, FO membrane flux outcome for 2M NaCl DS against DI water FS was evaluated for two different operating conditions i.e., for Re. no 200/500 at AL-FS membrane orientation and Re. no 3750/1500 at AL-DS membrane orientation and results are presented in Fig. 9. The results indicate that a flux increase of about 511% was achieved by just manipulating FO process conditions which help transform membrane pore structure, facilitate DS come closer to membrane, reduce the boundary layer effect and expedite water molecule transport through the membrane AL and SL pores.

4. Conclusions

HFFO membrane was evaluated to assess the effects of some of the operating conditions in terms of water flux and RSF. Parameters including membrane orientation, DS properties, cross-flow directions, and cross-flow rates were evaluated. It was observed that operating parameters significantly affect the performance of the FO process. Main findings are summarized as follows:

- (1) Properties of the anionic part of the DS were found important for flux outcome whereas RSF was largely influenced by the properties of DS cationic part.
- (2) Results indicated that by adjusting FO processes conditions, HFFO membrane achieve significantly lower specific RSF and higher water flux outcome.
- (3) FO operation at varying DS and FS Re ratio showed better flux outcome as Re ratio for DS and FS decreases and vice versa.
- (4) FO operation under the AL-DS orientation at varying DS and FS cross-flow rates markedly showed enhanced performance outcome. It was observed that using 2M NaCl as DS and DI water as FS, HFFO successfully delivered water flux of 62.9 LMH at DS/FS Re of 3,750/1,500.

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