Role of various physical and chemical techniques for hollow fiber forward osmosis membrane cleaning

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Abstract

Fouling is an inevitable phenomenon with most of the water treatment systems. Similar to RO, NF and other membrane based systems, fouling also seriously affects the performance of low cost forward osmosis (FO) systems and disturbs the overall efficiency of these systems and various cleaning practices have been evaluated to restore their designed performances. This study evaluates the performance of various physical and chemical cleaning techniques for hollow fiber forward osmosis (HFFO) membrane. HFFO membrane was subjected to various fouling conditions using different brackish ground water qualities and model organic foulants such as alginate, humic acid and bovine serum albumin. Results indicated the physical cleaning gives different results for inorganic and combine fouling flux restoration and the crossflow rates play an important role in cleaning membrane surfaces in active layer –feed solution (AL-FS) and active layer-draw solution (AL-DS) membrane orientation. The higher crosssflow Re values at any particular area seem important for the cleaning. With hydraulic flushing, the flux performances of HFFO were recovered fully when operated in AL-FS orientation as high shear force help detach all scaling layers from the surface, however the lower shear force did not fully restore the flux for the FS membrane in AL-DS orientation. Chemical cleaning was planned for the fouled HFFO membrane and HCl and NaOH were used in various combination sequences. It was found that HCl did not clean membrane used for AL-DS orientation for combine fouling. HCl cleaning (at pH 2) was found to be more effective for removing inorganic scale whereas NaOH cleaning (at pH 11) for a similar period successfully restored the flux for all the membranes used for FS with inorganic and/or organic foulants. EDTA was also evaluated for its cleaning performances and it was found that compared to NaOH, EDTA cleaning (1 mM concentration at pH 11)

showed superior results in terms of membrane cleaning as it helped to successfully restore the membrane flux in a very short time.

1. Introduction

Membrane performance can be seriously decreased as a repercussion of different types of fouling including inorganic, colloidal-, organic- and bio-fouling [1,2]. Membrane fouling is also an important and inevitable phenomenon in all membrane processes [3-5]. The fouling growths on the membrane surface seriously affect water flux and permeate quality and reduce the overall efficiency of water treatment systems [5]. Various efforts have been made in different directions to control fouling development risks including improvement in the membrane properties for flux resistance, setting optimum operating conditions and incorporating several pre-treatment techniques [6].

To get sustainable results from the membrane based systems, membrane cleaning is usually performed for the fouled membranes at different time intervals [7]. For the existing popular membrane systems such as reverse osmosis (RO), nanofiltration (NF) and ultra-filtration (UF), the membrane cleaning is usually initiated when a significant change is observed either in the membrane performance (decrease of permeate flux and/or salt rejection) or operating parameter (rise of trans-membrane pressure) to deliver the same designed water flux.

FO is a low pressure process and thought to be quite different from RO in terms of the reversibility and water cleaning efficiency [8,9]. Earlier FO studies indicated little or no fouling on FO, and only physical cleaning practices were experienced to restore flux through the FO membrane [4,10-12]. Various cleaning techniques such as normal flushing, osmotic backwash, high flow osmotic flushing and chemical cleaning have been evaluated to restore the water flux of the fouled forward osmosis (FO) membranes [5,10,13,14]. To decipher the mechanisms of fouling and chemical cleaning, it is always important to apprehend the foulant-membrane, foulant-foulant, and foulant-cleaning agent interactions. The choice for any physical cleaning technique or specific chemical cleaning plan depends on lowering the foulant-membrane along with increasing the foulant-cleaning agent interaction and foulant-foulant interactions [1,14].

Physical cleaning methods depend upon mechanical forces to dislodge and remove foulants from the membrane surface. The forces of the interaction between the membrane surface and the attached fouling layer are important in understanding the effectiveness of the physical cleaning phenomena. Usually, being a low pressure process, flat sheet FO fouling is assumed as reversible using physical cleaning methods possibly due to less compact fouling layer [12]. Cleaning chemicals restore the membrane performances by changing the morphology of the foulants, or altering the surface chemistry of the fouling layer. Consequently, proper selection of chemical cleaning agents relies on our mechanistic understanding of the foulants particularly the chemical reactions between the foulant and the cleaning chemicals [15,16]. Various studies have evaluated these interaction using atomic force microscopy (AFM) [12,17,18] which help evaluate the suitability of a particular chemical cleaning reagent or technique. Li and Elimelech [5] found that the cleaning efficiency is highly dependent on the solution pH and the concentration of the chemical cleaning agent.

For chemical cleaning of fouled membranes, five categories of cleaning agents are commonly used: acids, alkalis, metal chelating agents, surfactants, and enzymes [19]. Generally HCl, H_2SO_4 and citric acid are the main ingredients for the acid cleaning solutions whereas NaOH is mainly used for alkaline cleaning with some detergents. Sodium hypochlorite (NaClO) and ethylenediamine tetra acetic acid (EDTA) are also used in some cases. Various factors such as the concentration of the cleaning chemicals, contact time, pH and temperature play an important role in the effective cleaning of the membrane. Most of the commercial chemicals used in the market for membrane cleaning are proprietary and the exact concentration of various ingredients is never disclosed by their manufacturers.

The focus of this study was to evaluate the effectiveness of various physical and chemical techniques for cleaning the fouled hollow fiber FO (HFFO) membrane. Fouling experiments were done with different brackish ground water quality feed solutions and different concentrations of model foulants as alginate, humic acid (HA) and bovine serum albumin (BSA). Membrane cleaning was evaluated for both active layer-feed solution (AL-FS) and active layer draw solution (AL-DS) orientation. General chemical such as HCl, NaOH and EDTA were used to evaluate their cleaning potential.

2. Experimental

2.1.1. FO setup

Three identical bench scale hollow fiber FO set-ups, similar to the one reported in an earlier study [20] was used in this study. First set-up was used for fouling tests, rinsing, flushing, hydraulic flushing, second for chemical cleaning and third one for the performance check experiments. All of the fouling tests were performed for crossflow rates representing the Reynolds number (Re) of 1900 and 700 for the lumen side and shell side respectively. These Re values represent laminar flow through the lumen and shell sides. Flushing and performance checks were carried out at the same Re. Higher crossflow rates were used during hydraulic flushing representing Re of 2350/1450 for lumen side and shell side respectively.

Table 1: Detailed composition of various synthetic BGW qualities evaluated for the FO fouling study. Osmotic pressure is calculated using OLI Stream Analyzer 3.2 software

	BGW10	BGW35
Compounds/Concentration	mmol	mmol
$CaCl_2.2H_2O$	2.2	7.6
NaCl	63.5	222.5
$NaHCO_3$	1.1	3.9
Na_2SO_4	12.6	44.2
KCl	1.8	6.3
$MgCl_2.6H_2O$	19.4	68
π (atm)	5.35	18.56

2.1.2. Membrane used

HFFO lumens, supplied by Samsung Cheil Industries, Korea, were used to construct different modules for the study. The detailed specifications of these lumens have been presented elsewhere [21]. Each module carried membrane area of 25 cm^2. These HF modules were evaluated for AL-DS and AL-FS membrane orientation.

2.1.3.   Chemicals used

2.1.3.1.   Feed solutions and Draw solutions

This study used 2 M KCl fertilizer draw solution (DS) for all inorganic scaling and organic fouling experiments. Feed solution (FS) showing various brackish ground water (BGW) qualities representing total dissolved solids (TDS) of 10,000, and 35,000 mg/L were prepared from different analytical grade chemicals and reported as BGW10, BGW35, respectively in the study. Detailed chemical composition of the FS quality is given in Table 1.

Deionized (DI) water was used as FS for baseline (reference line) tests. Further, all other DS and FS for this study were prepared in DI water. Normal and hydraulic flushing was also carried out using DI water.

2.1.3.2.   Organic foulants

Model foulants such as alginate, HA and BSA organic foulants were used with various quality FS for the fouling studies.

2.1.3.3.   Cleaning chemicals

NaOH and HCl were respectively used as simple alkaline and acidic cleaning chemicals for the membrane flux restoration. NaOH was used for the organic fouling removal/cleaning whereas HCl was used to clean membranes for inorganic scale deposition [1,14,22]. Dilute solutions were prepared from these laboratory grade chemicals supplied by Chem-supply, Australia. NaOH cleaning was carried out at pH 11 whereas HCl solution at pH 2 was used for FO membrane cleaning. pH was regularly monitored and maintained during these tests. EDTA, a metal chelating agent in 1 mM concentration was also evaluated at pH 11 (adjusted with NaOH) for comparing its effectiveness for the HFFO membrane chemical cleaning process.

2.1.4.   Measurement of water flux

Water flux was evaluated by continuously measuring the loss of FS tank weight placed on a weighting balanced directly connected to a computer. FS readings were recorded after a fixed time interval and this data was used to evaluate the FO water flux.
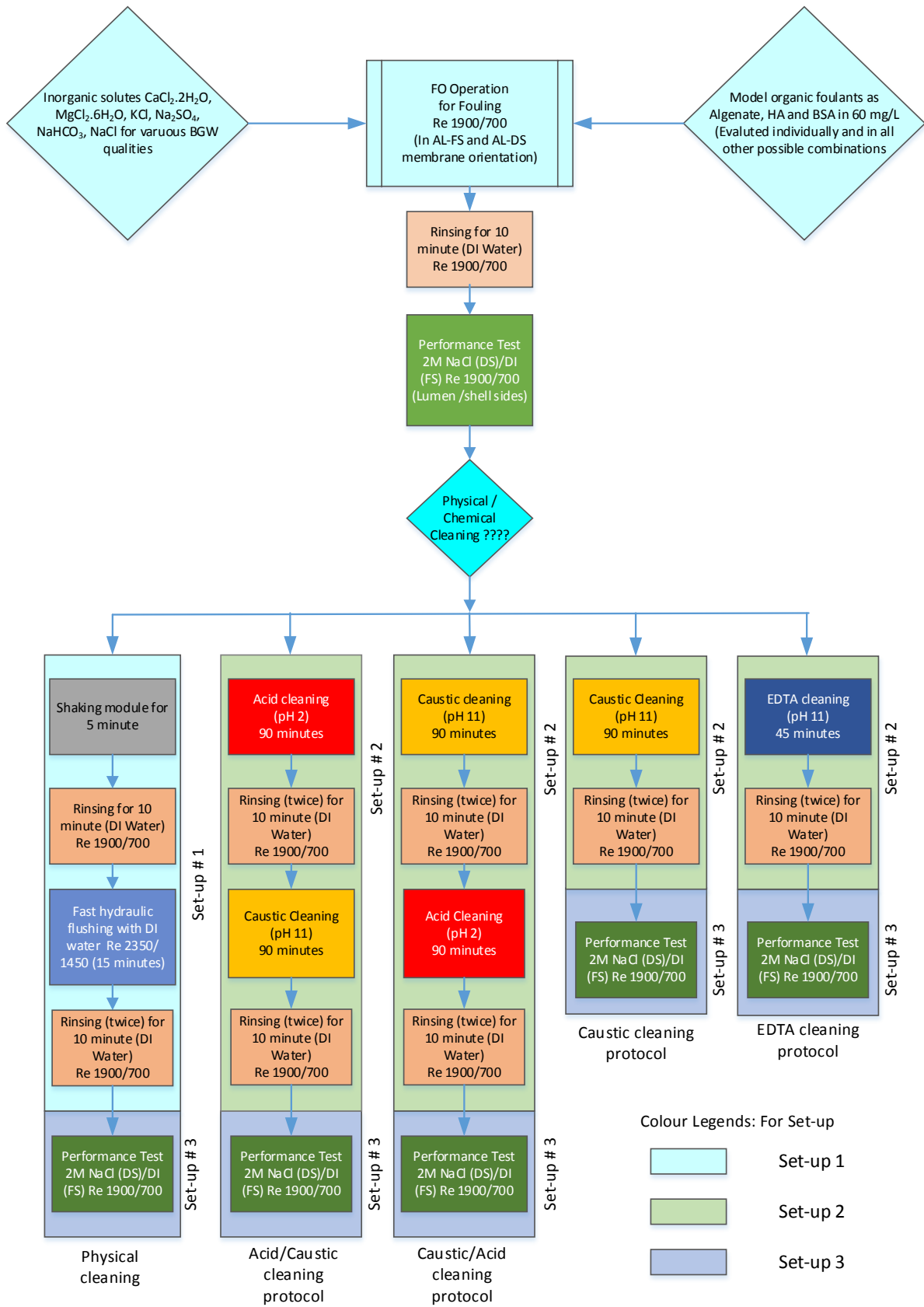
Inorganic solutes CaCl$_2$.2H$_2$O, MgCl$_2$.6H$_2$O, KCl, Na$_2$SO$_4$, NaHCO$_3$, NaCl for varuous BGW qualities

FO Operation for Fouling Re 1900/700 (In AL-FS and AL-DS membrane orientation)

Model organic foulants as Algenate, HA and BSA in 60 mg/L (Evaluted individually and in all other possible combinations)

Rinsing for 10 minute (DI Water) Re 1900/700

Performance Test 2M NaCl (DS)/DI (FS) Re 1900/700 (Lumen /shell sides)

Physical / Chemical Cleaning ????

Set-up # 1

Shaking module for 5 minute

Rinsing for 10 minute (DI Water) Re 1900/700

Fast hydraulic flushing with DI water Re 2350/1450 (15 minutes)

Rinsing (twice) for 10 minute (DI Water) Re 1900/700

Performance Test 2M NaCl (DS)/DI (FS) Re 1900/700 — **Set-up # 3**

Physical cleaning

Set-up # 2

Acid cleaning (pH 2) 90 minutes

Rinsing (twice) for 10 minute (DI Water) Re 1900/700

Caustic Cleaning (pH 11) 90 minutes

Rinsing (twice) for 10 minute (DI Water) Re 1900/700

Performance Test 2M NaCl (DS)/DI (FS) Re 1900/700 — **Set-up # 3**

Acid/Caustic cleaning protocol

Set-up # 2

Caustic cleaning (pH 11) 90 minutes

Rinsing (twice) for 10 minute (DI Water) Re 1900/700

Acid Cleaning (pH 2) 90 minutes

Rinsing (twice) for 10 minute (DI Water) Re 1900/700

Performance Test 2M NaCl (DS)/DI (FS) Re 1900/700 — **Set-up # 3**

Caustic/Acid cleaning protocol

Set-up # 2

Caustic Cleaning (pH 11) 90 minutes

Rinsing (twice) for 10 minute (DI Water) Re 1900/700

Performance Test 2M NaCl (DS)/DI (FS) Re 1900/700 — **Set-up # 3**

Caustic cleaning protocol

Set-up # 2

EDTA cleaning (pH 11) 45 minutes

Rinsing (twice) for 10 minute (DI Water) Re 1900/700

Performance Test 2M NaCl (DS)/DI (FS) Re 1900/700 — **Set-up # 3**

EDTA cleaning protocol

Colour Legends: For Set-up

Set-up 1
Set-up 2
Set-up 3

Fig. 1. Sequence of different physical and chemical membrane cleaning procedures used for this study

2.1.5. Performance checks

At the end of each fouling and/or cleaning tests, the performance checks were carried out for all modules with 2 M NaCl DS and DI water feed to reflect effects of fouling and/or cleaning on the HFFO membrane.

3. Results and discussion

3.1 Physical cleaning of FO membrane

On completion of the fouling experiments continued for 600 minutes, the performance of HFFO membranes was evaluated for scaling and fouling effects for varying combinations of FS qualities. Following Fig. 1 protocols, various types of simple physical membrane cleaning techniques such as normal flushing, vigorous physical and vibrator assisted shaking, osmotic backwash and hydraulic flushing and chemical cleanings using HCl, NaOH and EDTA were evaluated for the flux restoration of the fouled FO membranes.

3.1.1. Effects of simple flushing

The FO membrane fouled by combine foulants was subjected to simple flushing with DI water on both DS and FS sides. The same crossflow rates matching experimental conditions were used and flushing was done for 10 minutes. It was observed that normal flushing did not improve the performance of the fouled FO membrane used in the AL-FS and AL-DS orientations.

3.1.2. Effects of vigorous shaking

Similarly, FO modules fouled by combine foulants were removed from the FO unit, drained to half and then vigorous manual shaking was carried out to detach the attached fouling layer. During fouling experiments with HA carrying FS, FO module shown attachment of a brownish black layer on HF lumen. These black colour flaky layers were partially separated from HF lumen during vigorous shaking. However, when the FO module was placed on a high speed vibrator running at 1000 rpm, most part of the dark coloured fouling layer was detached from the HF lumen. . However, it was noticed that these techniques did not bring

any major improvements in relation to the restoration of FO module flux. This indicated that the initial fouling layers developed on HF lumen carried strong interactions between the combine foulants and the membrane surface which may not be weaken by the physical or vibrator assisted shaking. Vibrator effect could not successfully disturb foulant association with the lumen rather it just removed the later deposition on the fouled lumens. This further indicated that major flux decline resulted from the initial fouling layer whereas the subsequent fouling layers do not contribute significantly for flux decline as their separation from the membrane surface did not fully restore FO membrane flux.

3.1.3. Effects of fast hydraulic flushing for inorganic fouling

After the application of the simple physical cleaning procedures, the FO membrane was further evaluated to check the effectiveness of the hydraulic cleaning for all the inorganic scaling issues. The performance checks with various BGW quality FS indicated a flux loss of 3% and 5.6% for AL-FS and AL-DS orientation respectively.
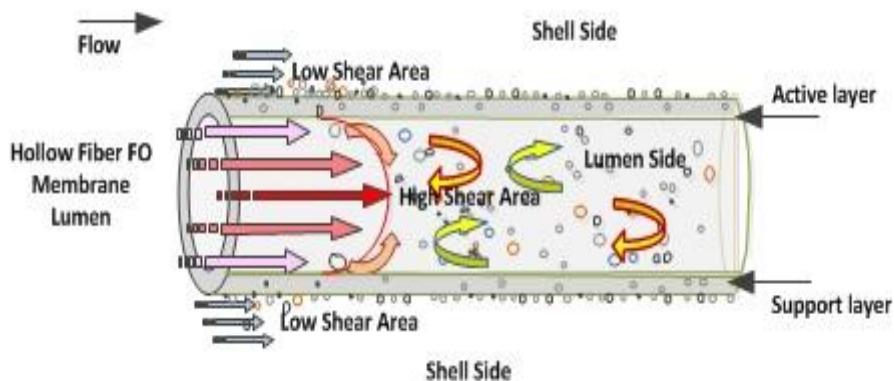
Fig. 2. Graphical presentation of the effects of crossflow shear force acting within and outside the HFFO lumens resulting different performances for hydraulic cleaning for the HFFO membrane used in AL-FS and AL-DS orientation.

Hydraulic flushing was used by operating FO for higher crossflow Re. of 2350 and 1450 for the lumen side and shell side respectively. DI water was used as FS and DS and flushing was continued for 15 minutes. It was found that for the FO modules used for fouling experiments with various BGW FS, the DI water flushing at the higher Re successfully restored the flux for modules used in AL-FS orientations. Hydraulic flushing helped dislodge and remove

foulants from the membrane surface droplets [23]. Unlike RO, the fouling layer is not compact and thus without using any chemicals, hydraulic flushing provides sufficient shear to weaken the fouling layer attachment with the membrane [12,24,25].

As opposed to the HFFO membrane used in the AL-FS orientation, the hydraulic flushing for 15 minutes did not provide a satisfactory outcome for the flux restoration of the FO used in the scaling experiments at the AL-DS orientation. Flushing was later extended to 30 minutes but only up to 50% of the lost flux was recovered.

Fig. 2 describes the varying effects of crossflow rates of different streams flowing within and outside the HF lumens in the HF module which plays a vital role in cleaning membranes through fast hydraulic flushing. When the crossflow rate was increased, higher flow rates produce swirling movements of the cleaning solution with excessive shear forces inside the HF lumens and these help to dislodge any loose scale deposit from the membrane surface. The flux restoration results with normal flushing using DI water further indicate that the inorganic scale does not penetrate inside the AL of membrane pores, rather it just builds up on the smooth outer surface of the AL of the membrane. Hence it is easily removed by hydraulic flushing. Due to lower pressure FO operations, the attached inorganic scale on the FO membrane does not compact to form a sticky and hard layer on the membrane surface. Thus, for HFFO used in AL-FS orientation with BGW quality FS, fast flushing representing high Re flow inside lumens helps fully restore flux for the fouled membranes.

However, with the current FO module design and the operating Re values, the same higher crossflow rates did not produce enough shear force in the shell side of the module which may assist to dislodge scale deposits from the outer surface of the HF lumens. Fast hydraulic flushing therefore did not show the same cleaning performance for HF membranes when used for AL-DS orientation. The same crossflow rates showing Re 1450 in the shell side and Re 2350 values in the lumen side were unable to produce enough shear force at the lumen outer surface which may help dislodge entire fouling layers. Results further suggest that due to comparatively larger SL pores, the BGW solute penetrates deeply into comparatively larger SL pores which are difficult to remove by fast flushing. The flow performances within the HF lumens and HFFO module shell sides are entirely different and these do not produce enough clipping on both sides of the membrane surface which resulted different cleaning effects for two sides of the HFFO membrane.

3.1.4. Effects of fast hydraulic flushing for organic fouling

HFFO modules used for fouling studies with Alginate, HA and BSA loaded DI or BGW FS were also subjected to similar fast hydraulic flushing. The results indicated that the fouling reduced the flux performances of the HFFO membranes by 9% and 49% for AL-FS and AL-DS orientation respectively. It was observed that the similar hydraulic flushing showed unproductive results for organically fouled membrane cleaning. For the membrane used in AL-FS orientation, normal flushing and fast flushing did not show any improvement for flux whereas for AL-DS orientation it was observed that it recovered 6.24% of the lost flux capacity (Fig. 3). This suggests that as the alginate, HA and BSA forms a gel type sticky fouling layer on the membrane surface [26] which requires additional force to dislodge the fouling layer from the membrane surface, the flux performance of the HFFO was not fully recovered for organic foulants loaded FS.
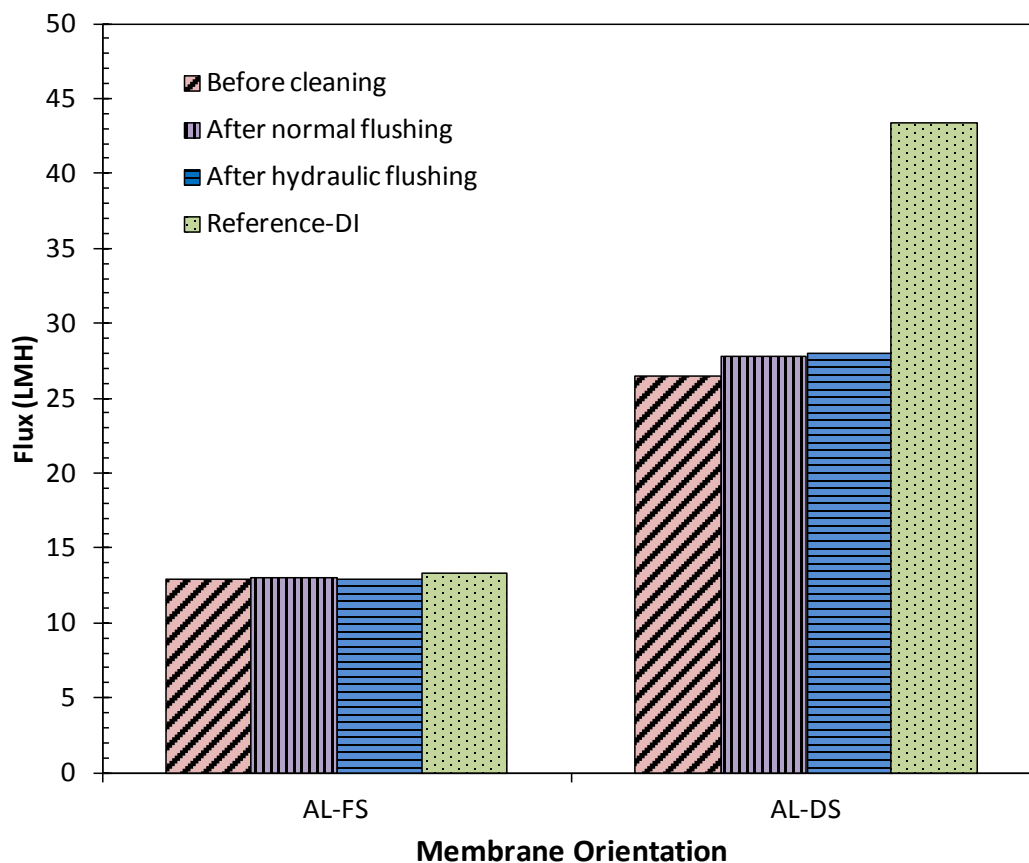
Fig. 3. Effect of physical cleaning of HFFO membrane for AL-FS and AL-DS orientation. 2 M KCl DS and 60 mg/L each of HA and BSA were used with BGW35 quality FS for the fouling test. Normal flushing was carried out at crossflow rates representing Re of 1900 and 700 whereas fast hydraulic flux used higher crossflow Re. of 2350 and 1450 for lumen side

and shell side respectively.

3.2 Chemical Cleaning

The effectiveness of the chemical cleaning was evaluated for the HFFO membrane where fast flushing did not restore the flux performance of the fouled membrane affected with combined fouling when operated in AL-DS orientation. Earlier studies have indicated various chemical cleaning protocols for different types of membranes using various acid and caustic solutions [1,5,6,22]. Cleaning efficiency varies with respect to the conditions applied during cleaning, namely, type of cleaning agent, cleaning solution pH, cleaning agent dose, cleaning time, crossflow velocity during cleaning, and cleaning solution temperature [6]. For low price and ease of availability, commonly used chemicals such as HCl and NaOH were evaluated in various combination sequences for HFFO membrane cleaning as indicated in Fig. 1 (acid only, acid-caustic, caustic-acid and caustic only). HCl was used at pH 2 whereas NaOH was used for pH 11.

Single chemical cleaning plan was extended for about 100 minutes for the following cleaning protocol; flushing with DI water 10 minutes, chemical recirculation 30 minute, chemical stay 20 minutes, chemical recirculation 30 minutes, rinsing twice with DI water (5 minutes each). Similarly, when HCl and NaOH cleaning chemicals were used together in different cleaning orders, one after the other, the total chemical cleaning time was extended to 190 minutes. After each acid or alkaline cleaning, both sides of the membrane were initially flushed twice with DI water and then evaluated with 2 M NaCl DS and DI water FS to record the flux outcome.

Furthermore, the membrane chemical cleaning was initially performed by circulating chemical solutions on both sides of the membrane. However, the outcomes showed that the chemical cleaning of only feed side successfully restored the membrane performance. Hence, later, most of the chemical cleaning experiments were just restricted to the FS side cleaning of the membrane (inner side of HF lumens in AL-FS orientation and outer side of HF lumens in AL-DS orientation) whereas DI water was circulated on the other side.

3.2.1   Acid (HCl) cleaning for HFFO membrane

FO membrane chemical cleaning was initially evaluated with HCl solution (pH-2). It was noticed the acid cleaning for 90 minutes successfully restored performances of the HF membranes used earlier for inorganic scaling studies at AL-FS and AL-DS orientation. Acid cleaning successfully restored FO flux as HCl easily dissolved out most of the scale forming solutes deposited on the membrane surface or trapped inside the porous support layer during FO operation in AL-DS orientation. Similarly, NaOH also solubilise inorganic salts and indicate better cleaning performance (Fig. 4).
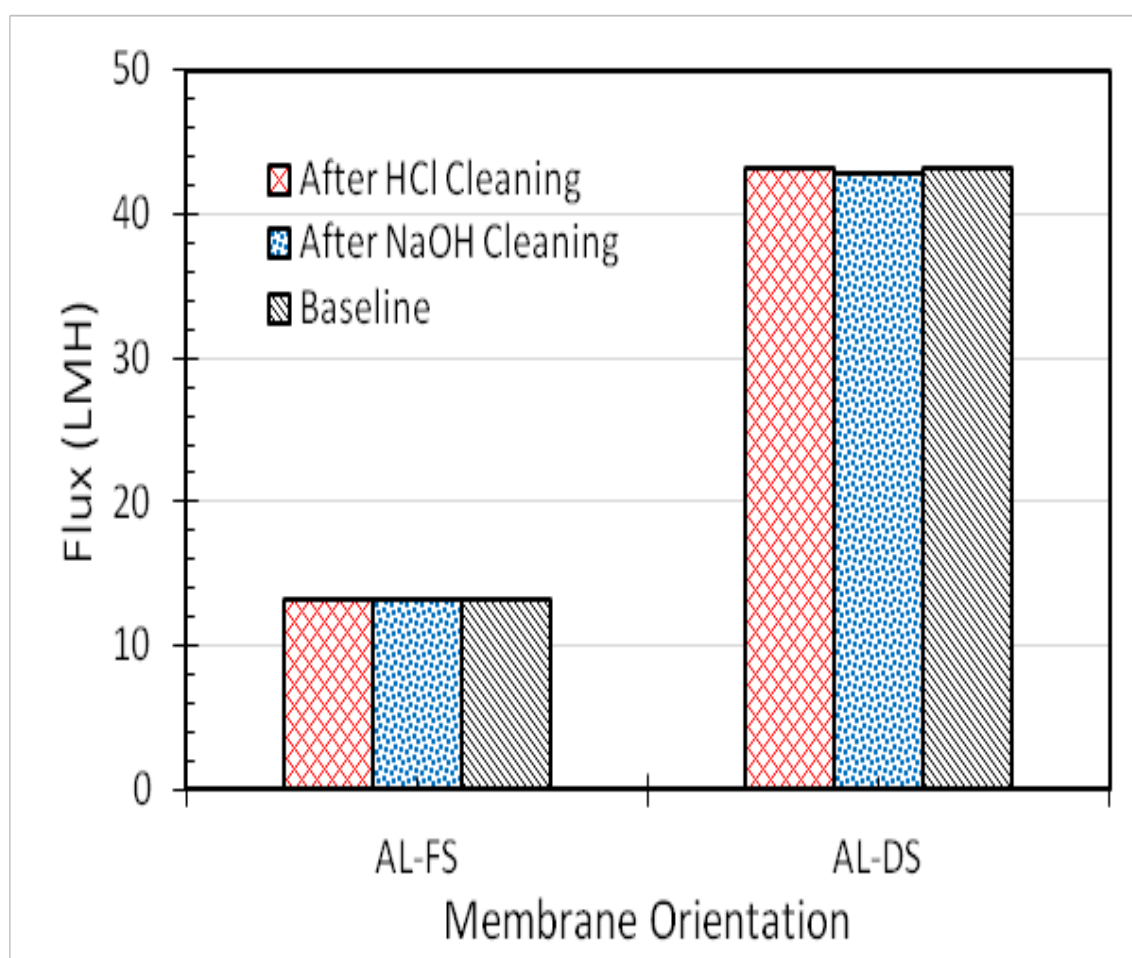
Fig. 4. Performances of HFFO membrane chemical cleaning using HCl and NaOH. BGW35 FS was used in fouling experiments.

Following Fig. 1 cleaning protocols, HCl was used alone and in other combinations with NaOH to clean fouled membrane operated with DI, BGW10, BGW20 and BGW35 quality FS carrying on 60 mg/L each of HA, BSA and alginate organic foulants. It was observed that

for FO operated for FS carrying organic foulants in DI water or various qualities of BGW, HCl cleaning alone did not produce good results to fully restore flux. However, when FO chemical cleaning was evaluated with various combinations, 100 min cleaning with NaOH fully restored HFFO membrane flux for FO membrane used FS carrying BGW 35 loaded with 60 mg/L of BSA, HA and alginate.
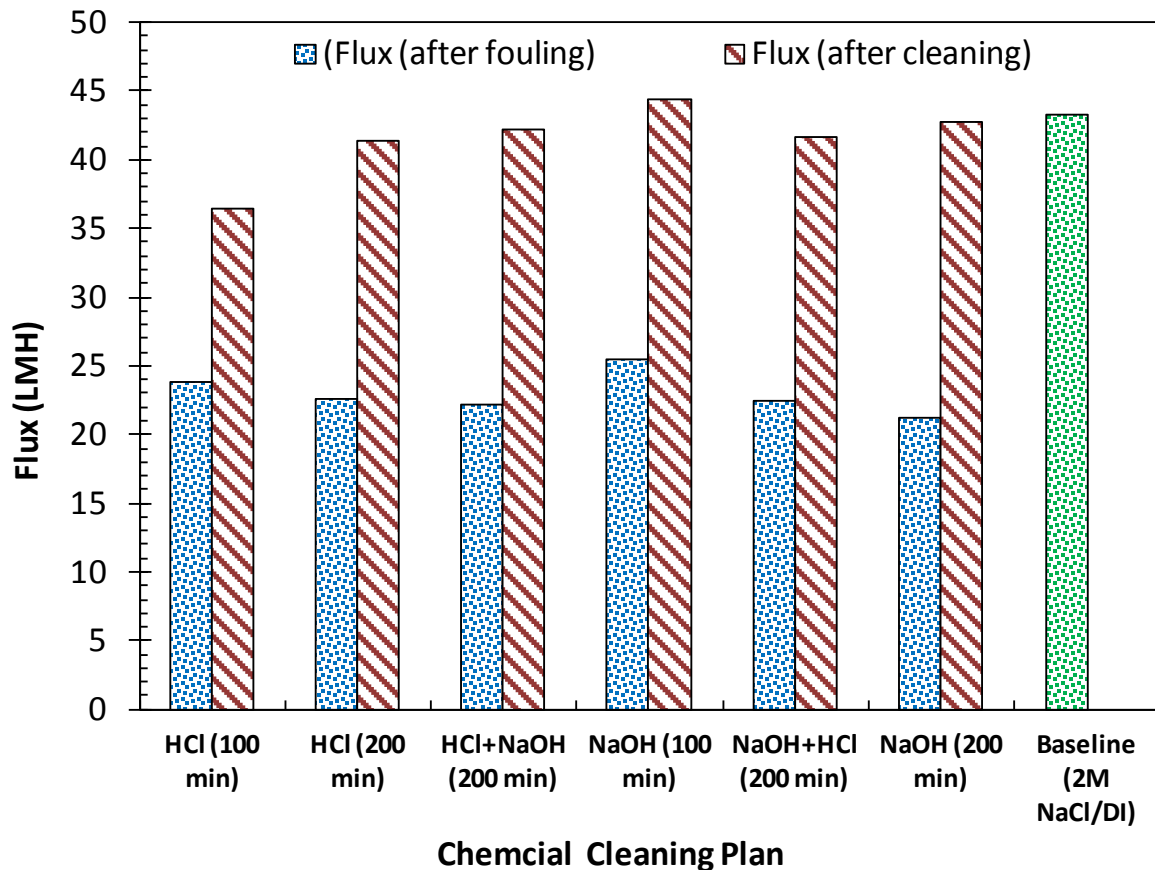

Fig. 5. Performances of various combinations of chemicals (HCl and NaOH) for HFFO membrane cleaning. Membrane was used for fouling experiments in AL-DS orientation.

3.2.2 Alkali (NaOH) cleaning for HFFO membrane

Fig. 5 summarizes the flux outcomes when the HFFO membrane was used with FS carrying on 60 mg/L each of Alginate, HA and BSA with BGW35 and then cleaned for fouling using various cleaning protocols. Among the above chemical cleaning sequences, for membranes fouled with combined foulants (inorganic or/and organic), the optimum results for chemical cleaning were achieved for 90 minutes of NaOH cleaning carried out at pH 11. Results

further indicated that for organically fouled membranes, among the various cleaning protocols, NaOH cleaning alone gave the best results to successfully restore the FO flux in the shortest period of time.

Based on these findings, few additional experiments for membrane cleanings were performed only with NaOH at pH 11 and the results are presented in Fig. 6. The FO membrane was evaluated for fouling using various combinations of different BGW quality FS with 60 mg/L each of Alginate, HA and BSA. Details of the FS quality used for fouling tests are given in

Membrane reference no	AL-FS	AL-DS
Table 2: Details of the FS used in the FO membrane combine fouling experiments. These fouled membranes were later used for NaOH cleaning		
1	Na-Alg.+DI	HA+Na-Alg.+ DI
2	HA+Na-Alg.+BSA+DI	Na-Alg.+ BSA+DI
3	Na-Alg.+B20	HA+ BSA+DI
4	Na-Alg.+B35	HA+Na-Alg.+ B20
5	HA+Na-Alg.+ DI	HA+B10
6	BSA+B10	HA+Na-Alg.+B10
7	HA+B35	HA+Na-Alg.+ BSA+ DI
8	HA+BSA+B10	Na-Alg.+ BSA+B35
9	HA+Na- Alg.+ BSA+B20	HA+Na-Alg.+ BSA+B35
10	Reference Baseline	HA+BSA+B35
11	HA+Na-Alg.+ B35	BSA+Na-Alg.+B35
12		Reference Baseline

Table 2. After fouling tests, the performance was checked with 2 M NaCl and then the membranes were subjected to cleaning with NaOH (pH 11). Fig. 6(a) presents few flux performances for NaOH cleaning for the FO membrane used in AL-FS orientation. The results indicated that for membranes used with FS with organic foulant, NaOH cleaning

uniformly show better outcomes using its two fold function of NaOH i.e., hydrolysis and solubilization. NaOH hydrolyses number of organic materials including polysaccharides and proteins [14]. It further increases negative charges of humic substances which made it easier to be removed from membranes. NaOH can change the configuration of natural organic matter and modify the fouling layer into a looser and more open structure which is easily removed with the shear force produced by normal flushing.

The results indicate that the chemical cleaning with NaOH successfully restored the FO membrane flux in 95% cases and delivered resultant flux with only ± 1% deviation. Some of the cleaning results show a higher flux outcome with NaOH, even higher than the baseline flux. This is similar to the higher flux performance for membranes treated with NaOH. Membrane treatment with NaOH adds more OH groups in the polymer chain which results in a decrease of the zeta potential of the membranes [27]. Membranes with lower zeta potential show improved flux performance for different membranes [28].

Similarly, for the HFFO membrane used in AL-DS orientation, NaOH chemical cleaning successfully restored membrane performance in 86% cases (Fig. 6(b)). Various FO membranes were used for fouling using different FS qualities as shown in Table 2. These membranes showed severe fouling effects as for various cases, the resultant flux in AL-DS orientation was dropped by 16 to 48%. A comparison of the resultant flux after NaOH cleaning with the baseline outcome indicates up to ±2% variation in flux (Fig 5(b)).

Similar to cleaning results for membranes used in AL-FS orientation, in some cases, FO membranes after cleaning showed an even higher resultant flux than the baseline outcome. The high negative charge of the membrane (lower zeta potential) helps quick and easy penetration of water molecules through the membrane. Higher flux outcome with a highly negatively charged membrane further indicates that the water permeation through the membrane starts with the association of the H^+ part of the water molecule and the negatively charged membrane and this helps water molecules to come closer and pass through the membrane pore.
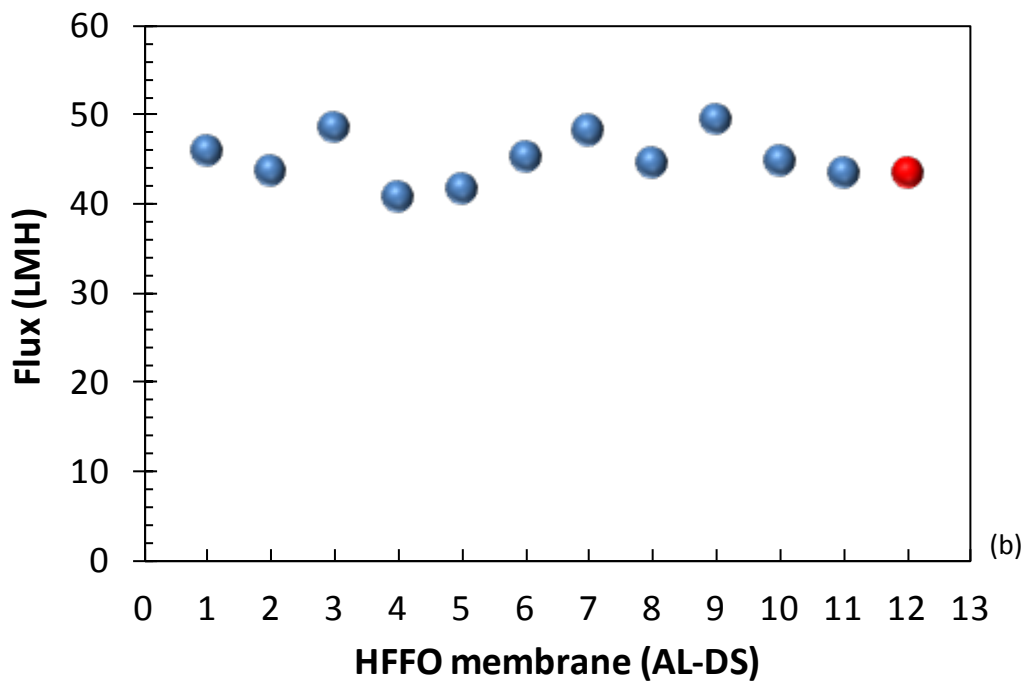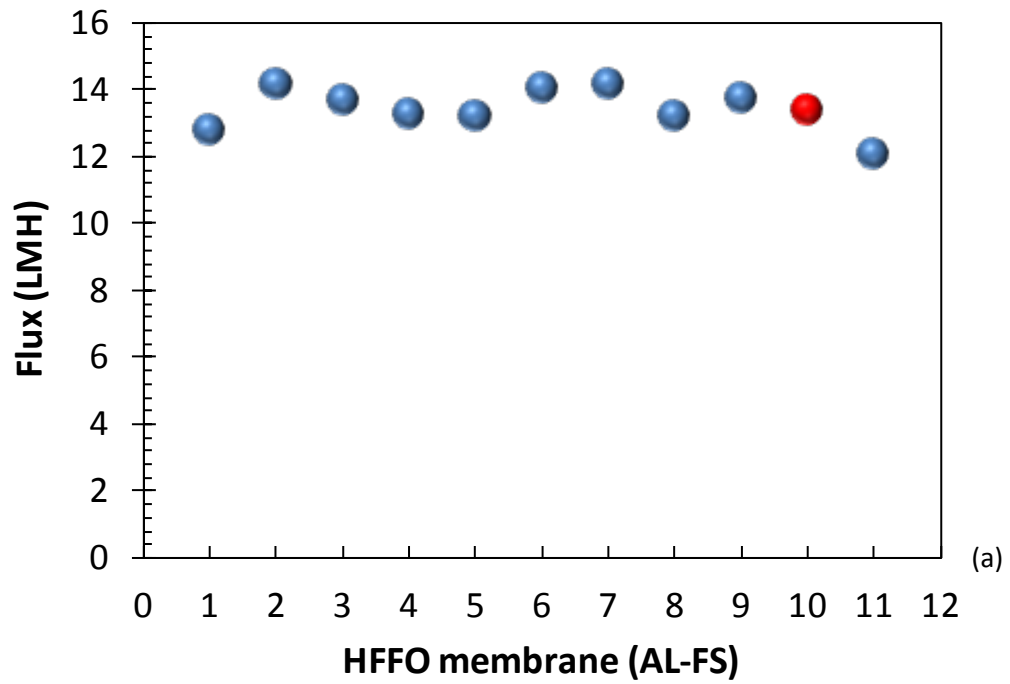
Fig. 6. Performances of FO membrane after cleaning a) resultant flux for membrane used at AL-FS orientation b) resultant flux for membrane at AL-DS orientation. Red circle shows the baseline flux.

3.2.3 Chemical cleaning with EDTA cleaning

EDTA was further assessed to evaluate the membrane cleaning efficiency. EDTA has been earlier used in various studies and it showed a remarkable ability in terms of membrane cleaning [5,6,29]. 1 mM EDTA was used for FO membrane cleaning with pH adjusted to 11 using NaOH. The following cleaning protocol was used; flushing DI water 5 minutes, chemical circulation 15 minute, chemical stay 5 minutes, chemical cleaning 10 minutes, rinsing twice with DI water (5 minutes).

After the fouling experiments using FS containing 60 mg/L of HA with BGW35, FO membrane cleaning was carried out separately with NaOH and EDTA and their cleaning performance in terms of time consumed and flux restoration is presented in Fig. 7. The comparison shows that the EDTA demonstrated better cleaning performance as it takes a lesser time of 45 minutes to clean the membrane fouling. The effectiveness of EDTA for membrane cleaning has been explained earlier. EDTA at high pH de-protonated all carboxylic groups and broke down the gel layer more quickly [6,30]. EDTA disrupts the fouling layer structure through ligand exchange between foulants and Ca^{2+} complexes which results in an increase of inter chain repulsion among foulant macro molecules leading to favourable conditions for the desorption of foulants from the fouling layer. EDTA works effectively as cleaner especially for scale compounds and metal oxides though solubilization and chelating [14] and helps the quick detachment of the fouling layer from the membrane surface and thus membrane cleaning is achieved in a short time. EDTA forms associations with the metallic ions in the fouling layer which serve to weaken their association with the membrane surface and reduce the adhesion forces between the fouling layer and membrane surface. NaOH cleaning efficiency was also checked after 45 minutes but the FO membrane showed a lower flux. NaOH cleaning was repeated again for further 45 minutes which helped fully restore the FO flux.
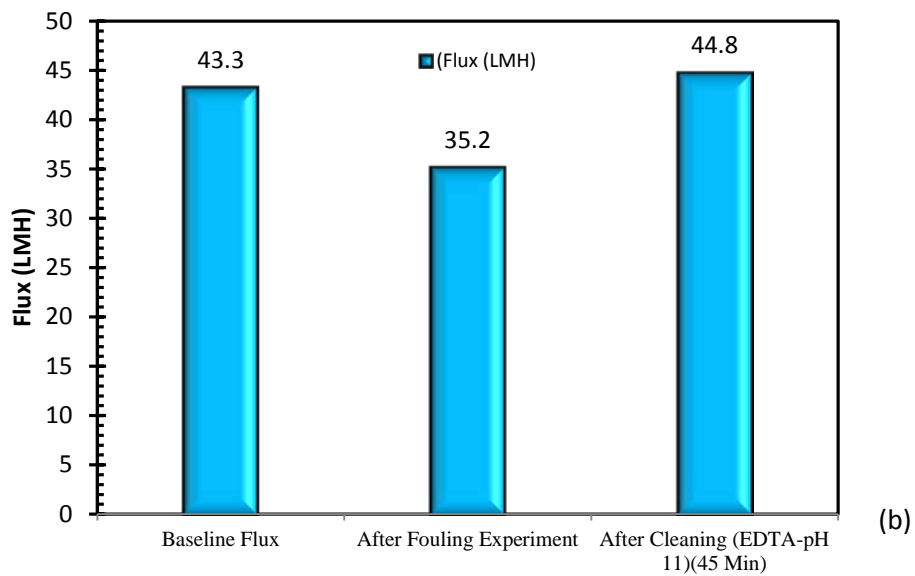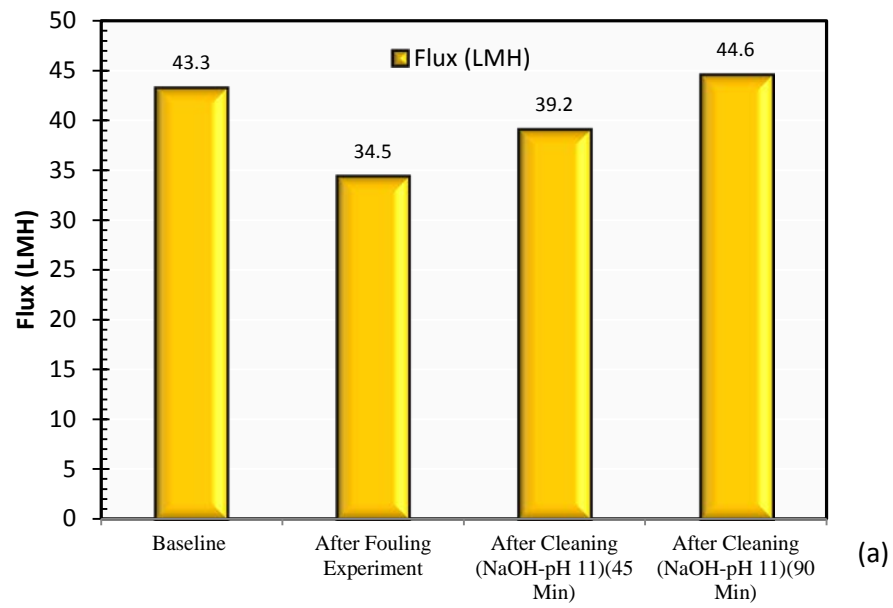
Fig. 7. Comparison of the performances of different chemicals for FO membrane cleaning a) Chemical cleaning using NaOH (pH 11) and b) Chemical cleaning using 1 mM EDTA (pH 11)

4. Conclusion

The HFFO membrane was assessed to evaluate various physical and chemical cleaning techniques for their cleaning efficiencies for membrane fouling which occurred as a result of different FS qualities carrying model organic foulants such as alginate, HA and BSA. The following conclusions can be drawn from the outcome of this study:

1) Hydraulic flushing helped clean HFFO membranes used in AL-FS orientation for fouling with FS with inorganic foulants whereas it did not fully restore the flux for the FS membrane in AL-DS orientation. The higher crossflow Re values at any particular area seem important for the cleaning.

2) HCl cleaning was found more effective for removing inorganic scale only whereas NaOH cleaning for a similar period successfully restored flux for all the membranes used for FS with inorganic and/or organic foulants.

3) Compared to NaOH cleaning, EDTA cleaning (1 mM concentration at pH 11) showed superior results in terms of membrane cleaning as it helped to successfully restore the membrane flux in a very short time.

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