

# **In-line-flocculation - Filtration as Pre-treatment to Reverse Osmosis Desalination**

A. H. Johir, C. Khorshed, S. Vigneswaran<sup>\*</sup>, and H. K. Shon

*Faculty of Engineering, University of Technology, Sydney, P.O. Box 123, Broadway, NSW 2007, Australia*

*<sup>\*</sup> Corresponding author (Tel.: +61295142641, Fax: +61295142633 Email: s.vigneswaran@uts.edu.au)*

## **Abstract**

In this paper the performance of single and dual media filters with in-line flocculation have been examined as pretreatment to seawater reverse osmosis (SWRO). A comparison of filter performance was made between single medium filter (80 cm) consisting of sand or anthracite, and dual media filter consisting of sand (40 cm at the bottom) and anthracite (40 cm on top). Short terms (6 hours) experiments were conducted with in-line coagulation followed by direct filtration. Filtration velocities of 5 m/h and 10 m/h were used. The performances of these filters were assessed in terms of turbidity removal, head loss build-up, and organic compound removal in terms of Molecular Weight Distribution (MWD). The efficiency of the filter as pretreatment was evaluated in terms of Silt Density Index (SDI) and Modified Fouling Index (MFI). It was found that the turbidity removal was high and all the filters produced more or less same quality water. There was a slower buildup of head loss for coarser filter medium. A post treatment of reverse osmosis after an inline-flocculation-dual media filtration showed lower normalized flux decline ( $J/J_0$ ) (0.35 to 0.22 during the first 20 hours operation) while, seawater without any pretreatment showed steeper flux decline (0.18 to 0.11 at first 20 hours operation) in RO.

*Keywords: Deep bed filtration, In-line flocculation, Desalination, Fouling Index.*

## **1. Introduction**

Water scarcity is becoming a looming problem through out the world especially in the arid regions such as Southern Europe, Middle East, North Africa, many states in United States and Australia. Meeting water demand is posing a great challenge. To produce potable drinking water from seawater, many desalination technologies including reverse osmosis have been successfully used. Among the problems with seawater desalination using reverse osmosis (RO), membrane fouling is the most important one as it deteriorates the performance of RO membranes and increases the energy. Membrane fouling is generally categorized into four groups: (a) inorganic fouling (including scaling), (b) particles/colloids fouling, (c) biological fouling and (d)

organic fouling. Inorganic fouling is caused by metal hydroxides and carbonates which precipitate on and in the membranes due to changes in water chemistry. Particulate fouling is due to the accumulation of suspended solids or colloids in the feed water accumulate on the surface of the membrane. Biological fouling results from the formation of a biofilm caused by the attachment and metabolism of biological matter which includes microorganism and macroorganism. Organic fouling is very common with surface waters containing natural organic matters (NOM.). Organic compounds consist of humic acid, fulvic acid, polysaccharides, and aromatic compounds. Organic compounds are also an energy source for microorganism. On the other hand it is very difficult to prevent fouling from colloidal, organic and biological matters.

Pretreatment is required to prevent the membrane fouling, increase the life of RO membrane and to maintain a constant permeate flux. Conventional pre-treatments are coagulation/ flocculation, deep bed filtration etc. Generally the feed water criteria for RO desalination are turbidity of less than 2 NTU and Silt Density Index ( $SDI_{15}$ ) of less than 3. Bonnelye et al (2004) [1] showed that coagulation followed by dual media filtration produced good quality feed water to RO. In-line filtration is commonly used pretreatment system in RO desalination [2]. In this study in-line flocculation-filtration process was investigated as pretreatment to RO. The efficiency of this pretreatment was carried out in-terms of SDI, MFI, headloss buildup, turbidity removal efficiency and organic matter removal in terms of Molecular Weight Distribution (MWD).

## **2. Materials and Methods**

In this study, seawater was collected from Chowder Bay, Mosman, Sydney. The average turbidity, pH and Dissolved Organic Carbon (DOC) values of seawater were 0.82 NTU, 8.1 and <1 mg/l respectively. Average silt density index with 15 minute ( $SDI_{15}$ ) and 10 minute ( $SDI_{10}$ ) were 6.12 and 8.75 respectively. The molecular weight (MW) distribution of the organic matter found in the seawater ranged from 105 to about 1220 daltons with the highest fraction at 105–390 daltons. The weight average molecular weight ( $M_w$ ), number average molecular weigh ( $M_n$ ) and polydispersity ( $\rho$ ) of the organic matter found in the seawater were 1090, 730, and 1.5 respectively.

Short term in-line flocculation and filtration experiments were carried out using transparent acrylic filter columns. The internal diameter of the filter columns were 4.50 cm and

length 120 cm. The columns were filled with filter media (Anthracite and Sand) up to a depth of 80 cm. The filtration velocities tested were 5 and 10 m/h. The coagulant used was  $\text{FeCl}_3$  at a dose of 1mg/l. The experimental set-up is shown in Fig 1. Down-flow filtration was employed and coagulant was added using a dosing pump. An overflow outlet was provided above the filter bed to maintain a constant head. The effluent samples were collected for analysis from the outlet at the bottom of the column.

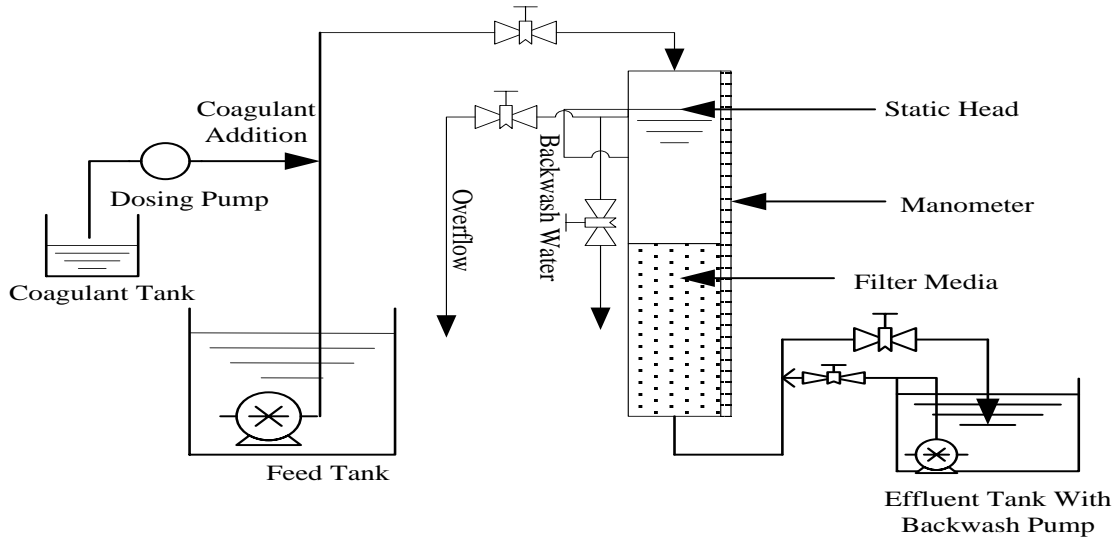


Fig 1 Schematic Diagram of Media (Anthracite/Sand/Dual) Filter System

The anthracite and sand used in this study were obtained from James Cumming & Sons P/L, Australia and Riversands P/L respectively and their properties are given in Table 1.

Table 1 Physical properties of Anthracite and Sand

Parameter	Anthracite	Sand
Effective Size (mm)	1.0-1.1	0.55-0.65
Uniformity	1.30	<1.5
Acid Solubility	1%	<2%
Specific Gravity	1.45	2.65
Bulk Density ( $\text{kg/m}^3$ )	660 to 720	1500

The pre-treatment efficiency was assessed in-terms of Silt Density Index ( $\text{SDI}_{10}$ ) and Modified Fouling Index (MFI). The modified fouling index (MFI) was established by Schipper and Verdouw et al. [3], [4] to evaluate membrane fouling. The SDI/MFI experimental set-up is

shown in Fig 2. In each experiment new membranes (with pore size of 0.45 $\mu$ m and diameter of 47mm) were used to avoid the residual fouling. The raw seawater and treated water were pressurized at 200 kPa using N<sub>2</sub> gas.

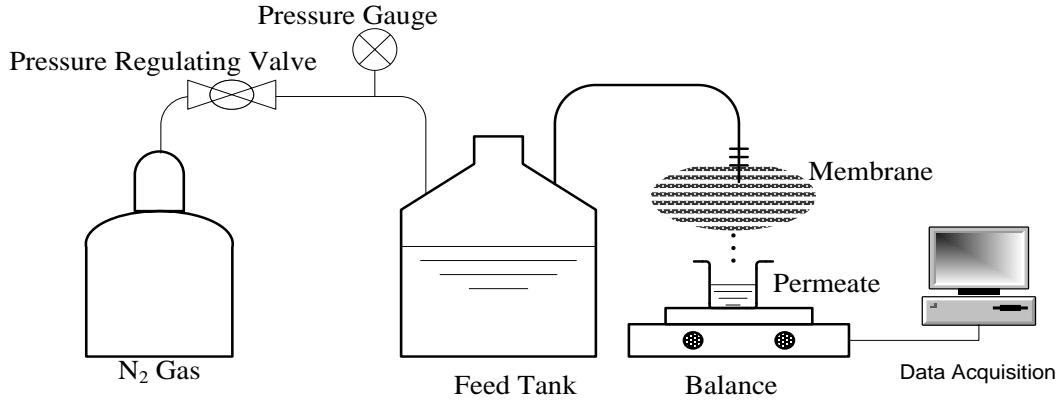


Fig 2 MFI experimental set-ups

The slope of the plot of  $t/V$  vs  $V$  gives the MFI value [5], [6].

$$\frac{t}{V} = \frac{\eta R_m}{\Delta P A} + \underbrace{\frac{\eta \alpha C_b}{2 \Delta P A^2}}_{\text{Slope (MFI)}} V$$

Where,

- $V$  total permeate volume (l)
- $R_m$  membrane resistance (m/kg)
- $t$  filtration time (s)
- $\Delta P$  applied trans-membrane pressure (Pa)
- $H$  water viscosity at 20°C
- $\alpha$  specific resistance of the cake deposited
- $C_b$  concentration of particles in feed water (mg/l)
- $A$  membrane surface area (m<sup>2</sup>).

The  $t/V$  versus  $V$  plot normally shows three phases of fouling namely (i) blocking filtration, (ii) cake filtration without compression and (iii) cake plugging and/or cake compression. The first sharp increase in slope is attributed to membrane pore blocking followed by cake filtration, which is the linear region of the curve. The MFI is defined as the gradient (tan

$\theta$ ) of this linear region of  $t/V$  vs.  $V$  plot normalized to standard reference values of 2 bar (207±3 kPa) trans-membrane pressure.

Silt density index (SDI) is the most common technique to quantify the performance of the pretreatment system in excluding colloidal particles. The SDI calculation procedure is described in the American Standard Testing and Method (ASTM D4189-95). The SDI value is calculated from the following equation:

$$SDI = [1 - (t_i - t_f) / T] * 100$$

where:

$t_i$  = initial filter time to filter a volume of water of 500 mL

$t_f$  = final filter time to filter the same volume of water of 500 mL

$T$  = elapse time (normally 15 min; in some experiments 10 min was used)

The recommended  $SDI_{15}$  should be less than 3 for RO desalination.

The Molecular Weight Distribution (MWD) of the organic matter in the influent and effluent were measured using High Pressure Size-exclusion Chromatography (HPSEC, Shimadzu, Corp., Japan) with SEC column (Protein-pak<sup>TM</sup> 125, Waters, Milford, USA). The absorbance of elutes were detected UV responses at 254 nm. The number average molecular weight ( $M_n$ ) and weight average molecular weight ( $M_w$ ) distributions were calculated using following equations.

$$M_w = \frac{\sum_{i=1}^n (N_i M_i^2)}{\sum_{i=1}^n (N_i M_i)}$$

$$M_n = \frac{\sum_{i=1}^n (N_i M_i)}{\sum_{i=1}^n (N_i)}$$

$$\rho = M_w / M_n$$

where,

$M_w$  = weight average molecular weight,  $M_n$  = number average molecular weight,  $\rho$  = polydispersity,  $N_i$  is the number of molecules having a molecular weight  $M_i$  where  $i$  is an incremental index over all molecular weight present.

The pre-treatment efficiency was also assessed in-terms of reverse osmosis (RO) flux decline. Cross-flow Seawater Reverse Osmosis (SWRO) experimental set-up used in this study is shown in Fig 3. In this study, RO flux decline was measured both for the pretreated effluent (by in-line flocculation-filtration) and seawater. These feeds had a conductivity of 48.9 mS/cm

and 50.1 mS/cm and turbidity of 0.23 NTU and 0.62 NTU respectively. The characteristics of RO membranes used in this experiment are given Table 2. The raw seawater and pretreated seawater were pressurized at 6000 kPa under 28 °C Temperature controller was used to keep constant temperature. The total volume of feed seawater before and after pre-treatment used was 5 L and the retentate was continuously recirculated to the feed tank. The feed was changed every 24 h of operation.

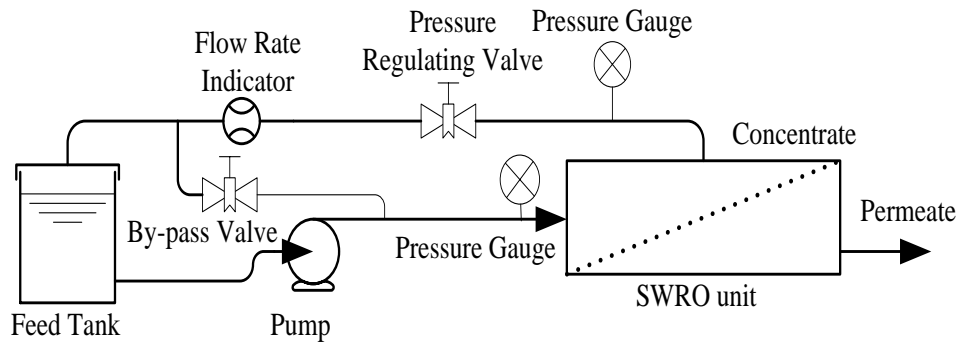


Fig 3 Schematic drawing of cross-flow SWRO unit used in this study

Table 2 Characteristics of RO membrane used

	Material	MWCO* (dalton)	Contact angle(°)	Zeta potential at pH 7 (mV)	PWP**at 6000 kPa (m/d)
SR	Aromatic polyamides	100	35	- 21	2.94

\* MWCO: molecular weight cut-off. \*\* PWP: pure water permeability

### 3. Results and Discussion:

The single and dual media filtration processes with in-line coagulation were evaluated for turbidity removal efficiency at 2 different velocities of 5 and 10 m/h. Fig 4 the present the turbidity removal at different filtration velocities and filter media. The average turbidity of raw seawater was 0.82 NTU. The result showed that finer filter media and dual media filter resulted in a higher turbidity removal efficiency of 70% (Fig 4).

Fig 5 shows the head loss development as a function of filtration time. Higher the filtration velocity and finer the grain size faster was the headloss development.

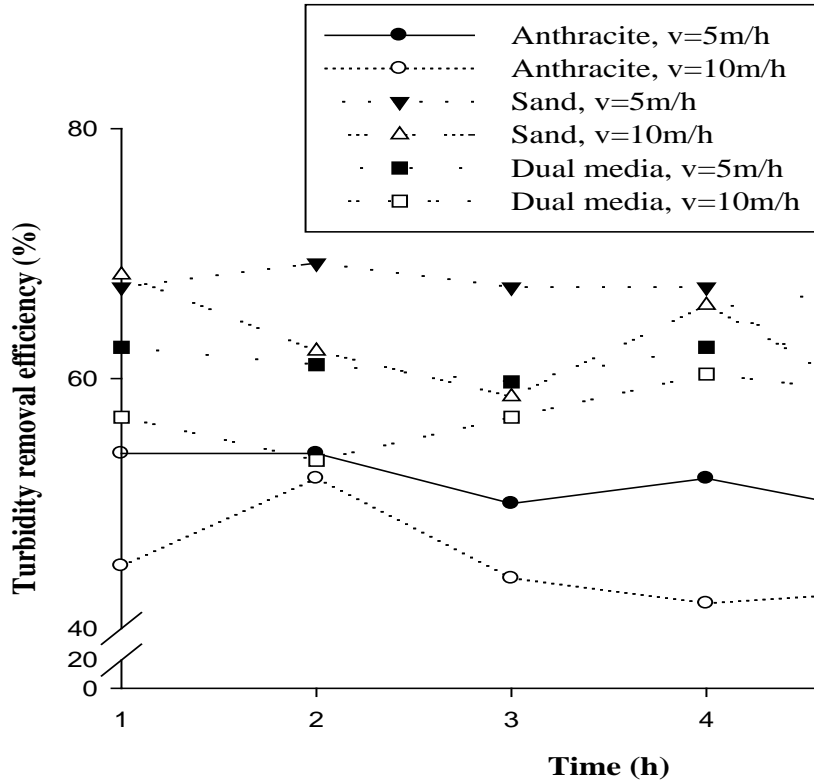


Fig 4 Effect of Filter media and filtration velocity on turbidity removal (total filter depth = 80 cm, avg. seawater turbidity = 0.82 NTU, FeCl<sub>3</sub> dose=1 mg/l)

For better understanding of membrane fouling, it is necessary to know the range of organic matter removed from seawater with and without pretreatment. The MWD of SWOM was measured for the prefiltered effluents. The MWD of the seawater ranged from 105 to about 1220 daltons with the highest fraction at 105–390 daltons. The weight average molecular weight ( $M_w$ ), number average molecular weight ( $M_n$ ) and polydispersity ( $\rho$ ) were 1090, 730 and 1.5 respectively. Fig 6 shows the MWD of SWOM before and after pretreatment. It is found that in-line flocculation with dual media filtration removed the majority of SWOM, while other filters also gave rise to good results in terms of SWOM removal efficiency. In-line flocculations with FeCl<sub>3</sub> (1 mg/l dose) effectively removed large SWOM. This phenomenon is explained by the complexation of Fe [7]. However the relative intensity of the smallest SWOM (at 390) was not reduced, showing that in-line flocculation was not effective in removing small molecules. The  $M_w$  (Weight average molecular weight),  $M_n$  (Number average molecular weight) and  $\rho$  (Polydispersity) of SWOM before and after pretreatment are presented in Table 3. Fig 6 and

Table 3 show that in-flocculation with dual media filtration resulted in better removal of SWOM.

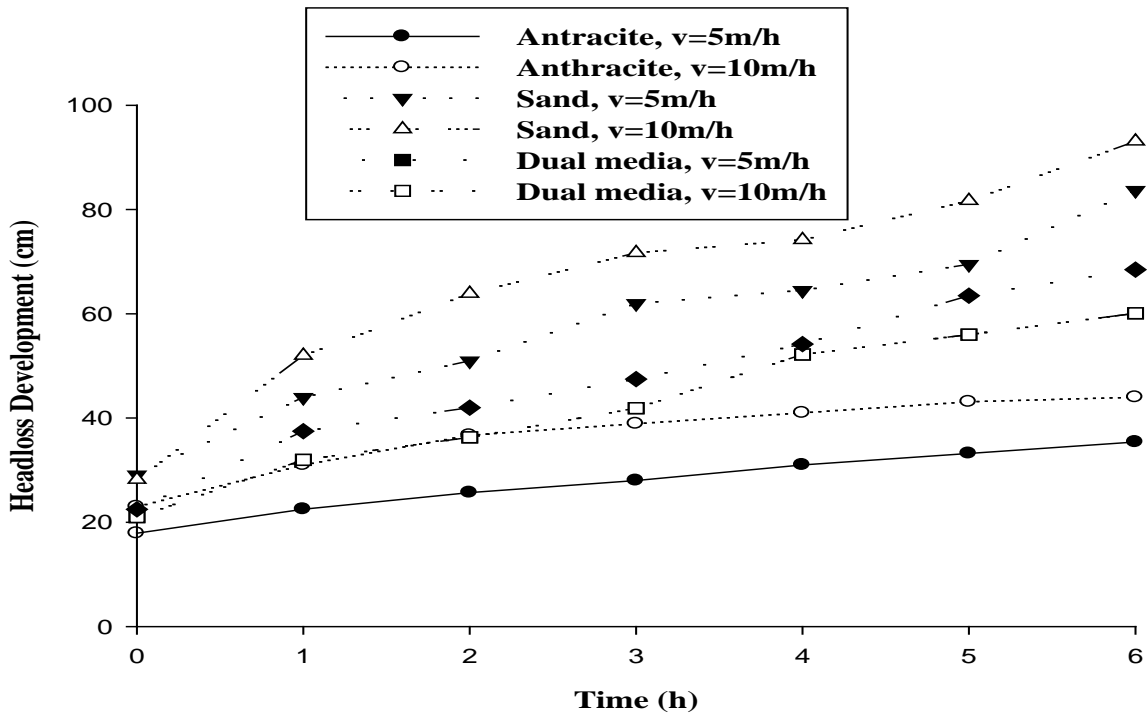


Fig 5 Effect of filter media and filtration velocity on headloss development (total filter depth = 80 cm, FeCl<sub>3</sub> dose=1 mg/l, average turbidity of seawater = 0.82 NTU)

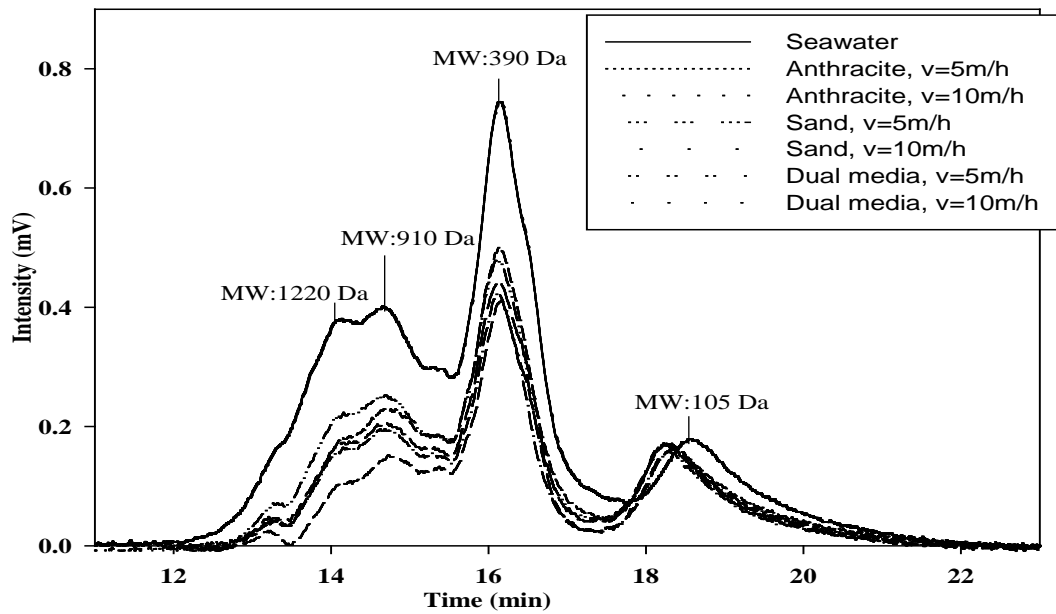


Fig 6 MWD of SWOM with and without pretreatment



Table 3  $M_w$ ,  $M_n$  and  $\rho$  of SWOM before and after in-line flocculation and filtration:

Parameter	Seawater	Sand filter (v=5m/h)	Sand filter (v=10m/h)	Anthracite (v=5m/h)	Anthracite (v=10m/h)	Dual Media (v=5m/h)	Dual Media (v=10m/h)
$M_w$	1090	1015	945	845	885	970	850
$M_n$	730	610	556	540	550	560	475
$\rho$	1.50	1.65	1.70	1.6	1.6	1.70	1.8

Modified fouling index (MFI) and Silt density indexes (SDI) were calculated to assess the fouling reduction of these pretreatment. The average MFI and  $SDI_{10}$  of raw seawater was  $256s/l^2$  and 8.75 respectively, while after pretreatment with contact flocculation-filtration, the  $SDI_{10}$  and MFI value reduced to 2.4-4.8 and 0.77-2.95  $s/l^2$  respectively (Table 4). In this study the filtrate SDI was taken after an elapse time of 10 minute instead of 15 minute because the water of 10 L volume was filtered before 15 min as the flow was too fast. Dual media filter as pretreatment resulted in highest fouling reduction (Table 4).

Table 4 MFI and  $SDI_{10}$  values after different in-line flocculation-filtration (seawater avg. MFI =  $256 s/l^2$ ,  $SDI_{10} = 8.75$ , pressure = 200 kPa, membrane pore size =  $0.45 \mu m$ , membrane dia. = 47 mm)

Filter Media	Velocity (m/h)	MFI (After 1.5 h)	MFI (After 3 h)	$SDI_{10}$ (After 1.5 h)	$SDI_{10}$ (After 3 h)
Anthracite	5	2.1	1.65	4.62	3.72
Anthracite	10	2.95	1.9	4.81	3.63
Sand	5	1.4	1.25	3.09	2.82
Sand	10	0.9	1.22	1.95	2
Dual media	5	1.2	0.8	N/D	1.87
Dual media	10	0.83	0.77	2.43	N/D

A post treatment of reverse osmosis after an inline-flocculation-dual media filtration showed a normalized flux decline ( $J/J_0$ ) from 0.35 to 0.22 during the first 20 hours (Fig 7). After that there was no significant decline in flux. On the other hand, seawater without any pretreatment showed steeper flux decline in RO and continued even after 3 days of operation.

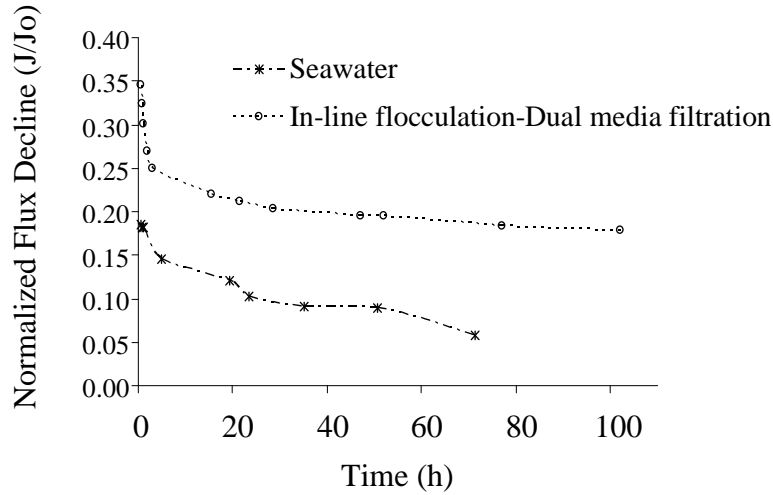


Fig 7 Temporal variation trend of filtration flux with and without pretreatment (SR membrane, crossflow velocity = 0.5 m/s, initial pure water flux = 2.94 m/d at 6000 kPa, total feed volume: 5 L, J is filtration flux at a given time and  $J_0$  is pure water filtration flux).

#### 4. Conclusion

In this study, the effectiveness of in-line flocculation-filtration as pretreatment was examined using single media and dual media filter in terms of headloss development, turbidity removal efficiency, SDI, MFI and DOC removal efficiency. The lowest headloss development of 35 cm was found for anthracite filter operated at 5 m/h of filter velocity. The result showed that finer filter media (sand) and dual media filter with filtration velocity of 5 m/h exhibits 70% of turbidity removal efficiency. The average MFI and  $SDI_{10}$  of raw seawater was  $256s/l^2$  and 8.75 respectively, while after pretreatment with contact flocculation-filtration, the  $SDI_{10}$  and MFI value reduced to 2.4-4.8 and 0.77-2.95  $s/l^2$  respectively. The weight average molecular weight ( $M_w$ ) was reduced from 1090 (for seawater) to 850 after pretreatment with dual media filter with in-line flocculation. A post treatment of reverse osmosis after an inline-flocculation-dual media filtration showed a normalized flux decline ( $J/J_0$ ) from 0.35 to 0.22 during the first 20 hours after that there was no significant decline in flux. On the other hand, seawater without any pretreatment showed steeper flux decline (0.18 to 0.11 during first 20 h) in RO. This decline continued even after 3 days of operation. Thus, in-flocculation and filtration process can be one of the practical and economical pretreatment methods in reducing RO membrane fouling.

## **Acknowledgements**

This work was funded by DEST-International Science Linkages Competitive Research Grant (CG110188)

## **References:**

- [1] V. Bonnelye, M.A. Sanz, JP. Durand, L. Plase, P. Gueguen, P. Mazounie, Reverse osmosis on open intake seawater: pre-treatment strategy, *Desalination*, 167 (2004) 191–200.
- [2] B. Tenzer, A. Adin, M. Priel, Seawater filtration for fouling prevention under stormy conditions, *Desalination*, 125 (1999) 77-88.
- [3] J.C. Schippers and J. Verdouw, The Modified Fouling Index, a Method of Determining the Fouling Characteristics of Water, *Desalination*. 32 (1980) 131-148.
- [4] J.C. Schippers, H.C. Folmer, J. Verdouw and. H.J. Scheerman, Reverse Osmosis for Treatment of Surface Water, *Desalination*. 56 (1985) 109-119.
- [5] S.F.E. Boerlage, M.D. Kennedy, M.P. Aniyé, E.M. Abogrean, G. Galjaard, J.C. Schippers, Monitoring particulate fouling in membrane systems, *Desalination*, 118 (1998) 131-142.
- [6] S.F.E. Boerlage, M.D. Kennedy, A.C. Paul, Bonne, G. Galjaard, J.C. Schippers, Prediction of flux decline in membrane systems due to particulate fouling, *Desalination*, 113 (1997) 231-233.
- [7] H.K. Shon, S. Vigneswaran, H.H. Ngo, R. Ben Aim, Is semi-flocculation effective as pretreatment to ultrafiltration in wastewater treatment?, *Water Research*, 39 (2005) 147–153.