Desalination and Water Treatment
www.deswater.com

doi: 10/5004/dwt.2012.2462

42 (2012) 87–93
April

Techno-economical approach of GAC and microfiltration as a coagulant-free pre-treatment of seawater desalination

Matan Beery[a], Ji Jung Lee[b], Byung Soo Oh[b], Joon Ha Kim[b,*], Jens-Uwe Repke[c]

[a]*Chair of Process Dynamics and Operation, Berlin Institute of Technology (TU-Berlin), Strasse des 17. Juni 135, Berlin, Germany*
[b]*Department of Environmental Science and Engineering, Gwangju Institute of Science and Technology (GIST), Gwangju, 500-712, Korea Tel. +82-62-970-3277; Fax: +82-62-970-2434; email: joonkim@gist.ac.kr*
[c]*Institute of Thermal, Environmental and Natural Products Process Engineering, TU Bergakademie Freiberg, Leipziger Strasse 28, 09596 Freiberg, Germany*

Received 20 October 2010; Accepted 3 August 2011

ABSTRACT

Membrane filtration is lately becoming a popular process for the pre-treatment of seawater in reverse osmosis (SWRO) desalination. The common practiced method of controlling membrane fouling and reducing the treatment costs usually involves a short coagulation step prior to the membrane filtration. In this work, a study of the feasibility of a coagulation free microfiltration (MF) as pre-treatment for SWRO desalination from a technical, economical and environmental point of view was performed. The experimental part included filtration of seawater from the Yellow Sea in Korea both with and without granular activated carbon (GAC) pre-treatment using a laboratory scale MF plant and different constant fluxes in outside-in dead-end mode. The results show that a coagulant and GAC free, stand-alone microfiltration using low fluxes and intense chemical cleanings is technically possible. When compared to a state of the art coagulation-MF, such a process could be economically and environmentally favourable when accounting for the lower energy demand and the relinquishment of the sludge treatment system.

Keywords: SWRO pre-treatment; Microfiltration; GAC; Coagulation; Yellow Sea; Sludge treatment

1. Introduction

RO membranes used for seawater desalination are highly susceptible to fouling due to organic/inorganic, biological and particulate matter often present in the sea. Fouling of the RO membranes has several negative effects which decrease the plant's economical and environmental efficiencies. Such effects include reduction in production rate, higher energy and chemical consumptions, frequent membrane cleanings and replacements, increase in the plant's downtime etc. An effective pre-treatment of the seawater is therefore a key issue for the long-term operational success of an SWRO plant.

A current trend is the replacement of the traditional granular media filters by microfiltration or ultrafiltration (UF) membranes as the main pre-treatment process step. The membrane based pre-treatment can provide a more reliable, higher quality RO feed especially when dealing with difficult waters which is prone to temporal and seasonal fluctuations in quality and temperature. The membrane pre-treatment's long-term economical and environmental benefits have not yet been proven but show a promising potential in the near future [1].

*Corresponding author.

When choosing a membrane pre-treatment most of the particulate-, bio-, colloidal- and organic fouling is relocated from the RO membranes to the pre-treatment membranes and must be controlled in order to maintain efficient working conditions. The usual way of confronting this problem is by using pre-membrane strainers, frequent backwashes (usually twice per hour) with or without air scouring and performing daily and monthly chemical cleanings (usually one daily chemically enhanced backwash and one monthly cleaning in place event). Additionally a small dose of coagulant is often introduced into the MF/UF feed water as it contributes to reducing the irreversibility of the fouling formed during filtration [2,3] and allows to work with higher fluxes and less frequent membrane cleanings. The use of a coagulant however, also means it will be present in the membrane backwash waste stream and that it must be considered for its environmental impact. In certain regions of the world's desalination market (like Australia, California or the southern European countries) where green policy making has a strong foothold, the release of untreated coagulant rich backwash waters into the sea would be very unlikely. The broader impacts of using coagulation with a membrane filtration including the waste treatment and disposal process should therefore be carefully considered in the overall economical and environmental life cycle assessment of the plant [4].

In a few documented cases a coagulant-free membrane filtration for SWRO pre-treatment was shown to be technically possible in long-term operation. This was usually the case when low fluxes and/or frequent chemical cleanings were used [5]. A systematic evaluation of the membrane filterability of coagulant free seawater under consideration of the economical and environmental impacts is therefore called for.

Since it was previously suggested that the combination of membrane filtration with an activated carbon pre-treatment could be a promising improvement of the filtration performance (especially concerning the removal of organic matter [6,7]) it was decided to technically explore this method as well, again without coagulation, as a comparable case for the stand-alone MF.

2. Experimental

2.1. System description

The experimental system used is composed of a 20 l feed tank, a frequency controlled feed pump, a membrane (MF) module, flow and (digital) pressure meters on the feed side, a permeate tank located on a digital scale and a backwash system. The process flow sheet of the system is given in Fig. 1. A photo of it can be seen in Fig. 2 with the feed pump on the left and the membrane module on the right. The system was operated in dead end mode by completely closing the cross flow valve, V1. The pressure measurements were only taken on the feed side of the membrane but because the permeate flow rate was low and open to the atmosphere it can be speculated that this pressure is only slightly higher than the trans-membrane pressure. The membrane module used had 10 hollow fibers of a microfiltration PVDF membrane with an outside-in vertical configuration. The main module's characteristics are given in Table 1.

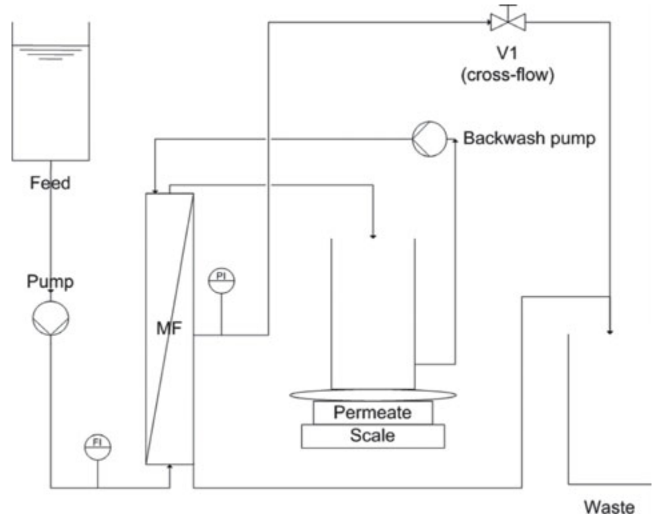

Fig. 1. The microfiltration-module experimental system.

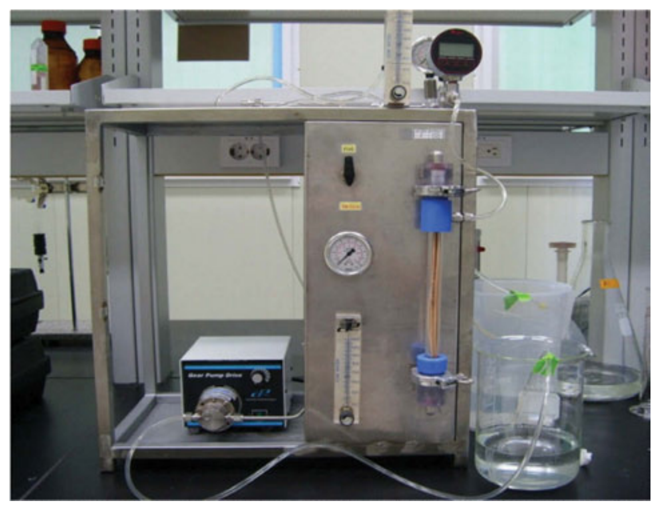

Fig. 2. A photo of the experimental system.

2.2. Experiments

Even though constant pressure filtration is more often seen in laboratory experiments, constant flux filtration is the mode of operation chosen by most industrial applications. Since the two modes have different fouling

Table 1
Technical data of the membrane module

Type	Pore size	Material	Fiber length	Diameter (in/out)	Number of fibers	Effective surface area	Mechanical strength	Operation mode
Cleanfil-S hollow fiber (Kolon)	0.1 µm	PVDF	0.25 m	0.8/2.0 mm	10	135 cm²	>25 kgf/fiber	Dead-end (constant flux)

Table 2
Raw seawater quality parameters

Temp	pH	DO	Turbidity	TOC	TSS
18.5°C	7.86	5.97 (mg l^{-1})	4.78 (NTU)	9.2 (mg l^{-1})	49 (mg l^{-1})

dynamics [8] and because it was aimed to mimic a realistic system in this work, constant flux filtration was chosen for the experiments. The flux was calculated by a computer using the read outs from the digital scale (precision: 0.01 g) and corrected for temperature using Eq. (1) [9]:

$$J_{\text{corrected}} = J \cdot \left(1.784 - 0.0575 \cdot T + 0.0011 \cdot T^2 - 10^{-5} \cdot T^3\right) \tag{1}$$

with J being the measured flux in l m^{-2} h^{-1} and T the temperature in °C. The pressure was manipulated accordingly by changing the feed pump's frequency.

Before using a new module a stabilization of the membrane with deionized water filtered at 1 bar for 6 h was performed, as well as a short chemical backwash (100 ppm NaOCl for 2 min) to make sure the membrane was free and clean.

Two kinds of waters were tested in the experiments: Surface seawater collected from Mokpo at the shore of the yellow sea in Korea and the same water pre-filtered with a 50 cm bed of granular activated carbon (GAC) in an acrylic glass column at a loading rate of 7 m h^{-1} (rapid granular media filtration). More information regarding this specific filtration apparatus can be found elsewhere [10]. The raw seawater quality parameters are given in Table 2. The characteristic parameters of the activated carbon are listed in Table 3. The turbidity of the GAC filtrate was 0.43 NTU.

Since one of the goals of this study was to inspect the system's feasible operation range in close to real conditions, the experimental regime for both the raw and pretreated waters was set as follows: Four cycles of 20 min filtration followed by 1 min of backwash were performed at the lowest possible flux, then a 2 min chemical backwash (100 ppm NaOCl) was performed followed by a cleaning in place (soaking over night in 0.5% NaOCl). The pure water permeability of the membrane was tested after every filtration, backwash and cleaning. The entire run was then repeated with a slightly higher flux. The permeate turbidity was measured in every filtration cycle.

Table 3
Granular activated carbon properties

Parameter	Value	Deviation
Particle size	12 × 30 Mesh	
Bulk density	0.51 (g l^{-1})	±0.03
Specific area	100 (m² g^{-1})	±0.5
Suspended solids reduction potential	92.9%	±0.5%
Turbidity reduction potential	93%	±5%
Fixed carbon	95%	±5%
Maximum ash content	7%	±3%

3. Results and discussion

3.1. Technical feasibility

The results of the raw seawater filtration are given in Fig. 3. The first working flux was achieved at 400 l m^{-2} h^{-1} with a pressure level of around 0.1–0.15 bar. At time points 20, 40 and 60 backwashes with fluxes of 1000 l m^{-2} h^{-1} took place which proved to be ineffective as the pressure levels did not drop back to previous levels in the next filtration cycles. At some point around t = 70 min the pressure began to rapidly increase going from 0.15 to 0.3 bars in a single filtration cycle. At the end of that cycle a CEB and CIP were performed and the filtration was commenced at a slightly higher flux: 450 l m^{-1} h^{-1}. The pressure levels were 0.25–0.3 bar. The backwashes in this case were performed at 1350 l m^{-2} h^{-1} and proved to be slightly more effective in restraining the pressure increase during filtration.

The results of the GAC filtrate filtration are shown in Fig. 4. In this case the lowest flux was achieved at 300 l m^{-2} h^{-1} with an initial pressure of 0.1 bar. The pressure increase shows a more distinct trend than in the raw water filtration. The backwashes (again performed at 1000 and 1350 l m^{-2} h^{-1}) were much more effective as one

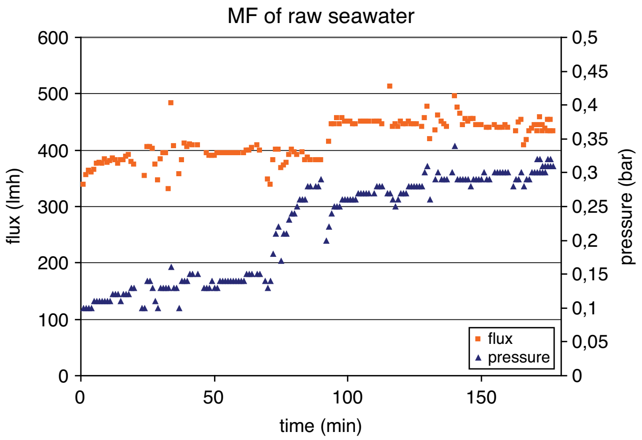

Fig. 3. Flux and pressure development during membrane filtration of raw seawater.

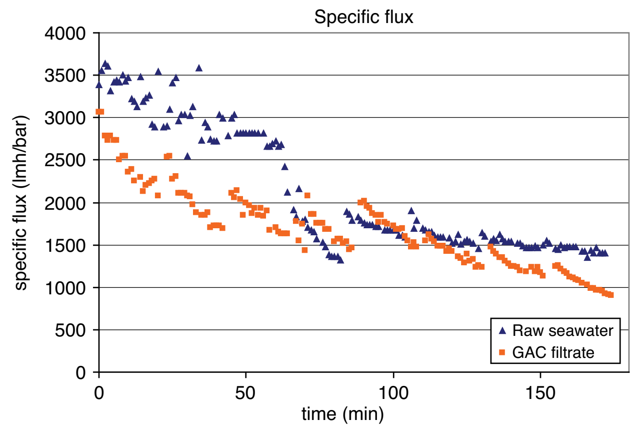

Fig. 5. Specific flux of the raw seawater microfiltration and the GAC filtrate microfitraion.

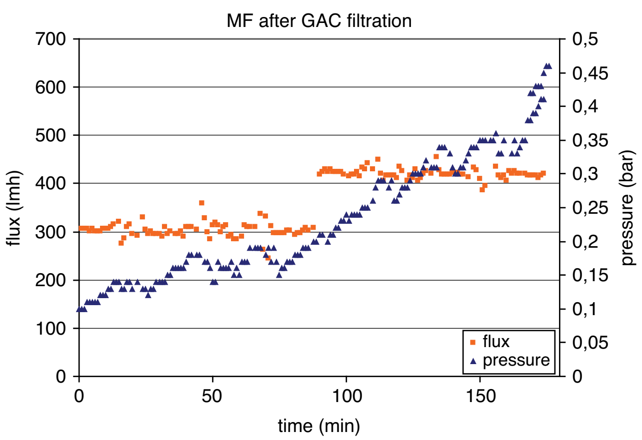

Fig. 4. Flux and pressure development during membrane filtration of GAC-filtered seawater.

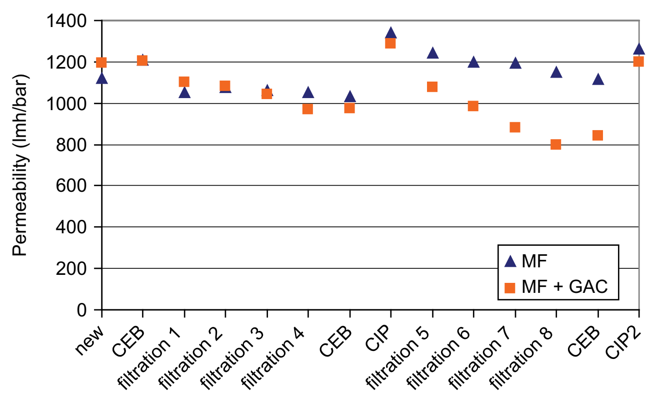

Fig. 6. Pure water permeability measured after backwashes/cleanings for raw seawater and GAC filtrate microfiltration.

can see from the typical saw-tooth shape of the pressure development. This could indicate a reversible fouling layer that is not as strongly attached to the membrane surface as the one formed in the raw seawater filtration. After the chemical cleaning was performed the initial pressure was raised to 0.2 bar and the resulting flux, 420 $l\ m^{-2}\ h^{-1}$, was maintained during the next four filtration cycles. The final pressure reached was higher than the one in the raw seawater case: 0.46 bar.

A way of bringing all these results together and being able to compare the feasibility of the different systems is by using the specific flux, an indicator which is reciprocally proportional to the overall filtration resistance, R_{tot}:

$$\frac{J}{\Delta p} = \frac{1}{\mu R_{tot}} \quad (2)$$

As it shows on Fig. 5, the specific flux of raw seawater filtration is highly erratic during the first filtration runs showing massive fluctuations and a sharp decrease during the fourth filtration cycle. None the less after the chemical cleaning it was fairly stable at around 1500 $l\ m^{-2}\ h^{-1}\ bar^{-1}$, indicating a feasible working region using these operation conditions. The specific flux of the GAC pre-treated water on the other hand shows a constant decline despite the chemical cleanings, a phenomena which could be attributed to a persistent physical plugging and jamming of the pores by the GAC fine particles. Despite the fact that the backwashes in this case are more effective than in the raw water case, the constant decline in specific flux proves this pre-treatment process to be less promising.

After each backwash and chemical cleaning the pure water permeability (PWP) was tested using deionized water filtration at constant flux and constant pressure. As it can be seen from Fig. 6, the pure water permeability of the membrane in the GAC case has decreased more rapidly than in the raw seawater case, probably due to pore jamming by the GAC particles.

It can also be seen that the backwashes at the end of the filtrations are not very effective in restoring the permeability in both cases and that even the CEB's have a very minor effect when compared to that of the CIP's which restore the permeability to levels similar or even better than the ones measure for the new membranes. At a first glance, one could suspect it is the lack of coagulation that is causing this persistent fouling however, some preliminary tests using coagulated seawater have also shown similar poor backwash effectiveness (not shown here due to lack of sufficient measurements). The reason why the backwashes were so ineffective could therefore be explained by two reasons: First, the largest amount of foulants accumulate at the far end of the fibers [11], whereas the most of the backwash water leaves at the beginning of the fibers (bottom of the module). Second, the removal of a fouling layer on the fibers' outer surface requires sufficient shear forces which are not available in this system. This is the reason why outside-in vertical systems often apply air scouring as an additional effective cleaning mechanism that creates these required shear forces.

When comparing the filtrate turbidity of the different filtrations runs (Fig. 7) one can observe similar values with a slight advantage for the raw seawater filtration: An average permeate turbidity of 0.18 NTU as opposed to 0.19 in the GAC case. A surprising result as the GAC filtrate is expected to be of better quality. The cause could be the fine activated carbon particles which are going through the membrane pores and raising the permeate turbidity.

Although more long-term pilot tests are required to determine the lasting effects of fouling and membrane cleaning, one can already make some preliminary conclusions from the current results. First, it appears that a coagulant free, raw seawater microfiltration is feasible when working with specific fluxes of 1500 l m^{-2} h^{-1} bar^{-1}. Assuming an overall average working pressure of 0.3 bar this would mean fluxes of 450 l m^{-2} h^{-1}. Second, the use of GAC filtration as a pre-treatment seems to have negative results on the MF membrane filtration (both in terms of filterability and of permeate quality) due to the breakup of small carbon pieces from the media. Therefore the raw seawater filtration is the only process of the two which seems technically prudent and is the one chosen for the further analysis described in the next section.

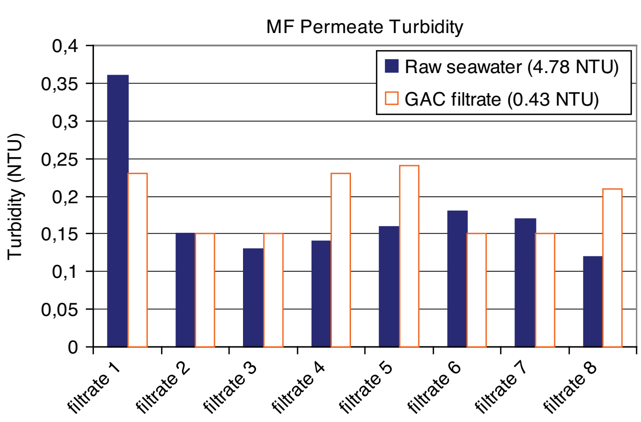

Fig. 7. Permeate turbidity measured during microfiltration cycles of raw seawater and GAC filtrate.

3.2. Economical/environmental assessment

Using some basic assumptions regarding the process while taking trade-offs between different capital and operation costs into account allows the formulation of a basic economical assessment for a large coagulation free pre-treatment process. Since an in depth look into such a system's economical and environmental pros and cons is desired, a parallel assessment was performed on a typical state of the art membrane pre-treatment system which includes inline coagulation. The missing information was taken from the literature [1,4,5,12,13]. The different cost factors and process parameters are shown in Table 4.

The capital costs spent on the larger membrane surface area in the coagulant free case (i.e. more modules and racks) are somewhat reduced by the savings on the coagulation and sludge processing systems. On the operation side, the savings on energy, sludge treatment, coagulation and dewatering chemicals are more than enough to compensate for the larger membrane replacement and cleaning costs associated with a coagulant-free process. Further more, since the higher energy consumption is also associated with larger green house gas emissions, the environmental impact of climate change (which was shown before as being the most dominant one in SWRO plants [4]) can also be taken into account in this analysis with the use of carbon certification. Assuming that changes in CO_2 emissions are tied to fiscal bonuses or penalties (as in the case of the European Carbon Trading Scheme) and that the electrical power source is coal-based, additional $22,300 could be saved yearly by relinquishing the coagulant. This number is currently not very significant but because it is highly depended on the power source as well as on the price of the CO_2 certificates (a price of $17.17 ton^{-1} was used as recorded in March 2010 by the European Energy Exchange [14]), it could be very meaningful in the near future.

Eventually, assuming 5% interest over a 20 y life span, the total cost of the treated water would be very similar in both cases: 2.02 and 2.03 cents m^{-3} in the coagulant and coagulant-free cases accordingly. The breakdown of

Table 4
Cost analysis: SWRO membrane pre-treatment with and without coagulation

General parameters	Membrane + coagulation	Membrane only
SWRO capacity (m^3 d^{-1})	200,000	200,000
RO recovery	45%	45%
Pretreatment recovery	90%	90%
Pretreatment capacity (m^3 d^{-1})	493,827	493,827
Avg. pretreatment flux (lmh)	600	450
Capital costs		
Membrane area (m^2)	34,294	45,725
Area per module (m^2)	50	50
Number of modules	686	915
Module price ($)	1800	1800
Cost of membranes ($)	1,234,800	1,647,000
Racks ($)	1,400,000	1,750,000
Microscreens ($)	4,850,000	4,850,000
Additional plant equipment ($)	3,500,000	3,500,000
Cleaning system ($)	2,100,000	2,625,000
Electrical and control ($)	100,000	100,000
Footprint ($)	1,900,000	2,375,000
Coagulation system ($)	115,000	0
Sludge processing system ($)	1,200,000	0
Install. & commissioning ($)	330,000	396,000
Overhead and contingencies	7,528,410	7,759,350
Tot. Capex ($)	24,258,210	25,002,350
Operation costs		
Avg. pressure (bar)	0.5	0.3
Electricity price ($ k^{-1} W^{-1} h^{-1})	0.1	0.1
Energy demand (kW)	346.1	207.7
Energy demand (kWh m^{-3})	0.0187	0.0113
Energy cost ($ y^{-1})	337,062	203,679
CO_2 certifications ($ y^{-1})	22,300	0
Coagulation chemicals ($ y^{-1})	57,800	0
Cleaning chemicals ($ y^{-1})	310,000	620,000
Dewatering chemicals ($ y^{-1})	28,500	0
Membrane replacement ($ y^{-1})	246,960	329,400
Sludge disposal ($ y^{-1})	182,000	0
Labor ($ y^{-1})	125,000	125,000
Utilities and maintenance ($ y^{-1})	196,443	191,712
Tot. Opex ($ y^{-1})	1,506,065	1,469,791

the costs is shown in Fig. 8. As it can be seen the additional capital and chemical costs in the coagulant-free case can be levelled out by the savings in energy and sludge treatment. Losing the coagulant will also result in environmental advantages associated with a smaller carbon footprint and elimination of the sludge waste stream.

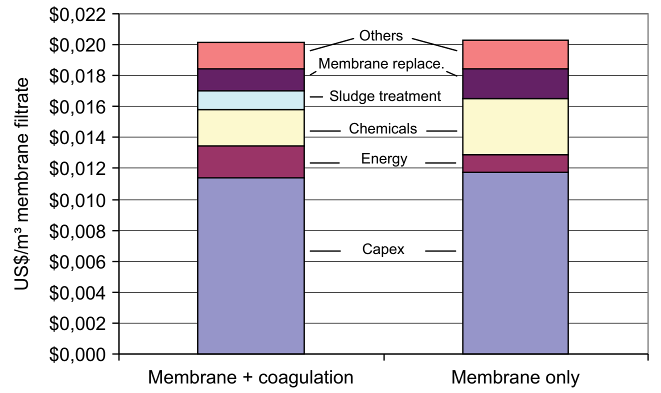

Fig. 8. Cost breakdown for MF-SWRO pre-treatment with and without coagulation in $ m^{-3} permeate.

4. Conclusions

An effective coagulant free microfiltration of seawater is possible when using low fluxes and intensive chemical cleaning. A flux of 450 l m^{-2} h^{-1} showed very limited growth in filtration resistance which indicates moderate fouling build up. In the filtration of the water pre-treated with GAC compared to that of raw seawater, the fluxes were smaller for the same pressure levels, the backwashes proved to be more effective and the pure water permeability declined more rapidly. Furthermore the filtrate turbidity was slightly higher in the GAC case. All of this implies that the main source of fouling in the GAC case was particulate matter (probably activated carbon) that was partially or completely blocking the pores and at times also going through the membrane. The economical analysis showed that under certain circumstances, coagulation free, low flux membrane filtration could be comparable in costs to that employing inline coagulation while having the environmental benefits associated with lower carbon emissions and reduced sludge waste production. Further long-term pilot experiments using large scale modules are, however, required in order to make an exact design decision.

Acknowledgements

This research was supported by the following programs:

- Plant Technology Advancement Program funded by the Ministry of Land, Transport and Maritime Affairs of the Korean government (SeaHERO).
- Elsa Neumann scholarship from the city of Berlin, Germany (NAFÖG).

References

[1] N. Voutchkov, Pretreatment Technologies for Membrane Seawater Desalination, 2009 AWA.

[2] H.-J. Yang and H.-S. Kim, Effect of coagulation on MF/UF for removal of particles as a pretreatment in seawater desalination, Desalination, 249 (2009) 45–52.
[3] K.A. Bu-Rashid and W. Czolkoss, Pilot tests of Multibore UF membrane at Addur SWRO desalination plant, Bahrain, Desalination, 203 (2007) 229–242.
[4] M. Beery and J.-U. Repke, Sustainability analysis of different SWRO pre-treatment alternatives, Desalin. Water Treat., 16 (2010) 218–228.
[5] M. Busch, R. Chu and S. Rosenberg, Novel trends in dual membrane systems for seawater desalination: minimum primary and low environmental aspect treatment schemes, IDA World Congress, Dubai, 2009.
[6] R. Zhang, C. Khorshed, S. Vigneswaran and J. Kandasamy, Submerged microfiltration coupled with physcio-chemical processes as pretreatment to sea water desalination, Desalin. Water Treat., 11 (2009) 52–57.
[7] H.K. Shon, S.H. Kim, S. Vigneswaran, R. Ben Aim, Sungyun Lee and J. Cho, Physicochemical pretreatment of seawater: fouling reduction, and membrane characterization, Desalination, 238 (2009) 10–21.
[8] J. Kim and F.A. Digiano, Fouling models for low-pressure membrane systems, Sep. Purif. Technol., 68 (2009) 293–304.
[9] S. Allgeier, B. Alspach and J. Vickers, Membrane Filtration Guidance Manual, United States Environmental Protection Agency, 2005.
[10] M. Beery, J.-J. Lee, J.-H. Kim and J.-U. Repke, Ripening of granular media filters for pretreatment of seawater in membrane desalination, Desalin. Water Treat., 15 (2010) 29–34.
[11] W.J.C. van de Ven, Towards Optimal Saving in Membrane Operation, PhD dissertation, University of Twente, 2008.
[12] M. Wilf, L. Awerbuch, C. Bartels, M. Mickley, G. Pearce and N. Voutchkov, The Guidebook to Membrane Desalination Technology, Balaban, L'Aquila, Italy, 2007.
[13] Affordable Desalination Collaboration (ADC online: www.affordabledesal.com), Dow SW30HR-380 NPV Spreadsheet, 2006.
[14] European Energy Exchange (online: www.eex.com), Emissions rights spot, March 2010.