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Brackish water reverse osmosis (BWRO) operation in feed flow reversal mode using an ex situ scale observation detector (EXSOD)

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ABSTRACT

The feasibility of operating brackish water reverse osmosis (BWRO) desalination in a feed flow reversal (FFR) mode so as to mitigate mineral scaling was evaluated with feed solutions containing calcium, sodium, sulfate and chloride ions. The premise of the FFR approach is to enable dissolution of surface mineral crystals, that form first at the BWRO plant tail elements, by periodic exposure to the undersaturated feed solution. Automatic triggering of FFR was accomplished by integrating an Ex-situ scale observation detector (EXSOD) with the BWRO plant controller. The EXSOD system consisted of a small high pressure plate-and-frame RO cell, with its membrane surface digitally imaged in real-time, to monitor the onset and development of membrane mineral scaling. During normal feed flow (NFF) operation, the EXSOD monitored a concentrate side-stream from the BWRO plant tail element, signaling the BWRO plant controller to initiate FFR when the specified scaling threshold was reached. Once the plant feed flow was reversed, the EXSOD feed stream was switched from the BWRO plant concentrate to the plant's permeate in order to dissolve mineral crystals formed on the EXSOD membrane during NFF operation, Successful operation of automated FFR, whereby scale formation was prevented, required adjustment of the EXSOD operating conditions such that the saturation index in the EXSOD RO cell was at a level that would not result in scale detection significantly before or after scaling would occur in the RO plant. Although the onset of mineral scaling was detected at the level of appearance of the first observable mineral crystal, the results suggest that it would be more practical to trigger FFR based on a threshold that considers both the mineral crystal number density and fractional surface scale coverage. Current work is continuing to demonstrate the broad applicability of FFR mode of RO mineral scale-free operation (with reduced or no antiscalant addition) over a wide range of solution compositions and operating conditions.

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1. Introduction

Arid and semi-arid areas with limited available water resources are challenged to find water sources that are more local to reduce the pressure on existing supplies, many of which are needed for agriculture. Reverse osmosis (RO) and/or nanofiltration (NF) membrane desalination of inland local water sources have been pursued in various regions of the world as a potential approach to generating these new water supplies. Due to difficulties involved in brine disposal/management and related expenses, there is a premium on recovering as much of the feed-water as possible, thereby reducing the volume of generated desalination concentrate (i.e., brine). The costs of environmentally responsible brine disposal methods at inland sites can be prohibitively expensive ($1-\frac{2}{m^3}$ of brine) [6]. As a result, it is often economically infeasible to develop such ground water sources unless very high recovery (\geq 90%) is attained [6].

High recovery in membrane desalination processes is often limited by scaling of sparingly soluble salts when they concentrate above their solubility limits in the highly concentrated brine. Among the various mineral salts that are associated with membrane scaling, calcium sulfate dihydrate (gypsum) and calcium carbonate are most commonly encountered in desalination of brackish surface and ground waters. Calcium carbonate scaling can often be mitigated by adjusting the water feed (prior to RO desalination) to acidic conditions (typically at pH < 6). Although calcium sulfate

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Fig. 1. Illustration of the reversal of the axial concentration polarization profile as the result of feed-flow reversal. (a) As the feed water flows through a series of one or more membrane elements, the concentration of the retentate stream and at the membrane surface increases axially along each membrane element. In this illustration the saturation index for the mineral scalant (Sl_g) is allowed to increase above the saturation limit to a value of ~2.3. Upon the onset of mineral scaling at the tail elements or just prior (i.e., at a time less than the crystallization induction time on the membrane, τ_{ind}) the feed flow is reversed (b) such that the solution concentration at the membrane surface at the module entrance (previously the exit or end L) is now undersaturated with respect to the mineral scalant saturation index due to the reversal of the concentration polarization profile.

scaling is nearly pH insensitive, partial success of its scale prevention has been achieved via antiscalant dosing of the RO feed [10] which adds to the overall water production cost.

Membrane scaling is of immense practical importance since such fouling significantly degrades membrane performance. Many processes to enhance product water recovery entail relatively high chemical costs because of (1) the necessity to reduce the concentration of the scaling ions significantly below the scaling thresholds, and/or (2) the addition of one or more different processes that can increase the complexity of the desalination process. These processes were reviewed by Van der Bruggen et al. [13] and the technical issues concerning the integration of RO desalination with chemical demineralization were presented in recent work by Rahardianto et al. [9,4].

Recently, a feed flow reversal (FFR) process was proposed for operating RO desalting of brackish water avoids the formation of mineral scale even under supersaturated solution conditions at the membrane surface [5,8]. In the FFR process, the feed flow is reversed just prior to or at the onset of scaling at the tail elements of the RO system. Upon FFR, the tail elements become the lead elements, and the lead elements become the tail elements. As a result, the axial concentration polarization profile is reversed and correspondingly the solution saturation profile (Fig. 1). When the feed flow is reversed, the tail elements (in which the brine stream is supersaturated and the rate of scaling is highest) [10] are now exposed to the undersaturated feed solution; this enables dissolution of mineral scale crystals that may have formed in the tail elements. At the same time, the solution concentration increases along the RO membrane train (due to concentration polarization) toward the now tail elements; thus, the driving force for scale formation increases in these membrane tail elements. Once mineral scale in the tail elements begins to form, the flow is again reversed. This process of periodic RO FFR continues so as to ensure scale-free desalting operation without the need for antiscalant treatment. It has been postulated that this feed-flow reversal scheme is feasible when FFR is triggered prior to the onset of membrane surface mineral scaling, i.e., $t_{\rm R} < t_{\rm S}$ where $t_{\rm R}$ is the period of FFR and t_{S} is the induction time for the initiation of scaling in the RO system. Although it was argued that upon reversing the feed flow the scaling induction clock is effectively re-zeroed, previous work did not demonstrate control of the FFR process via direct monitoring of the onset of surface crystallization. It is also noted that studies have shown that the induction time for crystallization on membrane surfaces is a function of both the local level of solution supersaturation at the membrane surface, the membrane surface topography and chemistry and flow hydrodynamics.

Monitoring of flux decline as a way of monitoring scale formation is a relatively insensitive approach to detecting the onset of mineral scaling [12,11]. However, recent development of an online ex situ scale observation detector (EXSOD) [3] has made it possible to detect the early stages of mineral crystal formation on the membrane surface. Such a detection scheme can be exploited for automating the transition to the FFR mode of operation. The approach for detecting the onset of scale formation relies on the use of a plate-and-frame RO cell with near dark-field illumination and an optical window that enables digital imaging of the membrane surface. The appearance of surface crystals is detected by on-line image analysis of the recorded images and the evolution of the surface number density and size of mineral crystals, as well as the percent of surface area covered by scale are analyzed in real-time. The emergence of direct scale detection makes it feasible to connect this monitor to a side-stream from the tail element of an RO spiralwound desalination plant, in order to determine the precise timing of the onset of scale formation (i.e., quantified by the observed crystallization induction time or threshold surface scale coverage). This type of detection would then trigger feed flow-reversal to ensure that membrane productivity does not decline and that permeate production is maintained at the desired rate.

In order to evaluate the possibility for automating the transition to the RO FFR mode of operation, the present study focused on demonstrating the benefit of integrating direct scale observation of a side-stream from an RO brackish water desalination plant. A series of tests were carried out, focusing on gypsum scaling, with the level of gypsum supersaturation within the EXSOD membrane cell being set at below, above and equal to the supersaturation level at the membrane surface of the last spiral-wound element of the RO train. The above modes of operation were evaluated with respect to triggering feed-flow reversal and the potential impact on permeate productivity and FFR frequency.

2. Experimental

2.1. Materials and model solutions

Feed solutions were prepared using analytical grade calcium chloride dihydrate (Carlo Erba; Rodano, Italy) and anhydrous sodium sulfate (Frutarom, Ltd., Akko, Israel) dissolved in equimolar amounts (Table 1) in deionized water $(2-10 \,\mu\text{S/cm})$. The feed solution pH was maintained at about 7.6. The feed solution was analyzed

 Table 1

 Composition of reference model solution.

Mineral salt ions	Concentration (mM
Ca ²⁺	10
Na ⁺	20
SO4 ²⁻	10
Cl-	20
TDS (mg/L)	2,529
Gypsum saturation index (at 25 °C)	0.519
pH	7.6

periodically for chloride and calcium ion levels using argentometric titration (chloride) and colorimetric titration (calcium). The degree of supersaturation with respect to gypsum (the sole sparingly water soluble mineral salt in the feed) was quantified in terms of the gypsum saturation index (SI_{σ}), defined as

$$SI_g = \frac{(Ca^{2+})(SO_4^{2-})}{K_{sp,g}}$$
(1)

where $K_{sp,g} = (Ca^{2+})_{eq}(SO_4^{2-})_{eq}$ is the solubility product for gypsum and where (Ca^{2+}) , $(Ca^{2+})_{eq}$ and (SO_4^{2-}) , $(SO_4^{2-})_{eq}$ are the activities of the calcium and sulfate ions, in the solution and at equilibrium, respectively. The saturation index for gypsum was determined using the OLI Stream Analyzer [2]. The monitored concentrate RO stream conductivity was correlated with the calcium ion concentration and the gypsum saturation index. These correlations were derived using the OLI software [2] resulting in the following expressions

$$[Ca^{2+}] = \left(\frac{G}{185.1}\right)^{1.1815} \tag{2}$$

$$SI_g = 60.8 [Ca^{2+}] - 0.161$$
 (3)

in which *G* is the solution conductivity (mS), $[Ca^{2+}]$ is the calcium ion concentration (M), and SI_g is the gypsum saturation index. The above correlations cover the SI_g range of 0.93–2.8 for a calcium chloride/sodium sulfate concentration range of 18–48.7 mM which covered the range relevant in the present study. It is noted that the feed solution was undersaturated ($SI_g = 0.52$; Table 1) with respect to gypsum in order to avoid gypsum bulk precipitation.

The BWRO unit was equipped with ESPA1 spiral wound elements (Hydranautics, Inc., Oceanside, California) having a water permeability ranging from 3 to $4 \text{ Lm}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$ depending on location in the train and the age of the membranes, which have been in use for periods of up to about 3 years. The EXSOD RO cell also utilized a newer batch of the ESPA1 membranes (L_p of about 5.5 Lm⁻² h⁻¹ bar⁻¹) which were fitted into the EXSOD RO cell as a flat sheet membrane.

2.2. Flow reversal manifold and EXSOD monitor

A pilot-scale brackish water RO unit (Tambour Ecology, Ltd., Akko, Israel) equipped with a Programmable Logic Controller (PLC; Unitronics M-90, Airport City, Israel) was used for the flow reversal experiments. This system was capable of operating up to 25 bar at flow rates up to 3 m³/h, using either 2.5 inch or 4 inch (diameter) spiral wound RO elements. In the present study, the feed pressure and feed flow rate were in the range of 12–20 bar and 800–1200 L/h, respectively. A train of six 2.5 inch by 40 inch ESPA1 (Hydranautics, Inc., Oceanside, California) spiral wound elements were installed in this pilot plant. Membrane elements were inserted into three pressure vessels in series and attached to a manifold that allowed for flow reversal (Fig. 2). Feed and permeate flow rates were monitored using a rotameter (Model KSK-1999HK3200; Kobold Messring, Nordirng, Germany) and turbine-type (Model 8031; Burkert Fluid Control Systems, Ingelfigen, Germany) flowmeter, respectively. Differential pressure was monitored using a pressure transducer (Model LD301; Smar, Sertaozinho, Brazil). Concentrate-side pressure monitoring was enabled by a pressure gauge connected via a valve manifold to several locations along the feed line (feed pump exit, 1st PV feed entrance, and feed entrance and exit for the last pressure vessel). The feed and concentrate conductivities were continuously monitored during the course of the experiments using conductivity probes (Model CD75-C; Seko Rieti, Italy and Burkert Model 8220; Burkert Fluid Control Systems, Ingelfingen, Germany, respectively). Pressures, temperatures, flow rates and conductivities of concentrate and permeate streams were monitored online and recorded with an online data logger (Field Logger, Novus Electronics Ltd., Porto Allegre, Brazil).

The EXSOD scale monitor consisted of a plate-and-frame RO cell with a channel height of 2.7 mm, width 2.5 cm and length 7.5 cm. The RO cell was transparent with provisions for near dark-field illumination of the membrane surface and surface imaging system with on-line image acquisition as described previously [3]. Pressure was monitored by a pressure transducer (PX 303-500G5V Omega, Stamford, CT), permeate flux was measured using a digital flow meter (Model 1000, Fisher Scientific, Pittsburgh, PA), feed and permeate conductivities were monitored with an on-line conductivity meter (Model WD-35607-30, Oakton Research, Vernon Hills, IL) and pH was measured with a pH meter (Model pH 110, Oakton Research, Vernon Hills, IL). All process monitors were interfaced with a PC-based data acquisition system.

The EXSOD monitor was installed on the concentrate side stream of the BWRO pilot unit (Fig. 2). A backpressure needle valve (V-8) was placed on the EXSOD's exit, in order to control flow rate through the EXSOD system. During cleaning cycles (i.e., washing of the system with a permeate stream), solenoid valves were opened to direct the permeate stream (from the BWRO plant) through the EXSOD unit. The status of each valve during normal flow and flow reversal operation is listed in Table 2. The exit streams (permeate and concentrate) from the EXSOD monitor were returned to the BWRO pilot feed tank during both the scale detection mode and during the wash cycle (i.e., FFR for the pilot system).

During normal flow operation, as shown in Table 2, valve V-1 connects the feed and pressure vessel PV1, valve V-2 connects pressure vessels PV3 and V-3, and valve V-4 remains closed. In the



Fig. 2. Flow reversal manifold with the EXSOD monitor mounted on the concentrate stream with computer controlled valves to switch between the concentrate and permeate side streams as alternate feeds to the EXSOD monitor. NFF and FFR indicate normal direction and reverse feed flow direction, respectively, with the arrows indicating the corresponding flow directions.

Table 2
Valve configuration during normal flow and flow reversal ^a .

	V-1 (three way valve)	V-2 (three way valve)	V-3	V-4	V-5	V-6	V-7	V-8	V-9
Scale detected Normal flow	Feed > PV1	PV3 > V-3	N/A	Close	Open	N/A	Close	N/A	Close
Scale removed Flow reversal	Feed > V-2	V-1 > PV3	N/A	Open	Close	N/A	Open	N/A	Open

^a Valve V-3, V-6, V-8, are adjustable valves and are not adjusted during flow reversal process.

EXSOD subsection of the system (Fig. 2) valve V-5 is opened, valves V-7 and V-9 are closed. Additionally valves V-6 and V-8 are adjusted to set the required operating conditions (pressure and flow rate) inside the EXSOD cell. Once scale is detected and flow reversal is initiated, the following valves are readjusted: valve V-1 is connected to the feed and valve V-2, valve V-2 is connected to valve V-1 and pressure vessel PV3, and valve V-4 is opened, essentially connecting pressure vessel PV1 and valve V-3. In the EXSOD subsection, valve V-5 is then closed and valves V-7 and V-9 are opened. It is noted that the positions of valves V-6 and V-8 stay unaltered during the cleaning cycle and are open at the same setting as during normal operation to avoid having to reset on each cycle. It is noted that a high pressure does not develop in the EXSOD monitor during this part of the cycle since valve V-9 is kept open.

The solute concentration at the membrane surface in the EXSOD RO cell was calculated using the previously published finiteelement numerical concentration polarization solution for this cell [7]. Subsequently, given the solution concentration at the membrane surface at the exit region of the EXSOD RO cell, the corresponding gypsum saturation index (Eq. (1)) was calculated using the correlation (Eq. (3)) derived from [2]. For the BWRO spiral-wound membrane element, the concentrations of the scaling species at the membrane surface were estimated using the concentration modulus calculated by IMS software provided by Hydranautics [1], given the feed conductivity (see Eqs. (2) and (3)) and the measured permeate flux and feed flow rates for the given experimental run.

In each experiment with the BWRO pilot plant, the operating conditions were set to attain a predetermined initial saturation index at the membrane surface for the tail element (PV 3), based on knowledge of the feed water composition, permeate flux and recovery in the six element train of spiral-wound RO modules. From the concentrate stream (exiting from the tail RO element), a sidestream was diverted to the EXSOD monitor where the pressure (using valve V-6, Fig. 2) and flow rate (using valve V-8, Fig. 2) were adjusted to obtain a specified initial level of supersaturation at the EXSOD membrane surface. Monitoring of mineral salt scaling and the triggering of FFR using the EXSOD image capture and analysis system and control software were then evaluated for three different scenarios (Table 3), whereby the initial gypsum saturation index (at the EXSOD cell exit region) was set higher (Table 3, Run 1), lower (Table 3, Run 2) or similar (Table 3, Run 3) to the initial SI_g at the tail (sixth) element of the BWRO plant.

Table 3

Experimental conditions for gypsum RO membrane scaling experiments.

Experiment	Initial gypsum saturation index at the BWRO membrane surface (module) ^a	Initial gypsum saturation index at membrane surface (EXSOD)
Run 1	2.11	2.37
Run 2	2.9	2.57
Run 3	2.37	2.0

Note: Concentration polarization modulus (i.e., $CP = C_m/C_b$ where C_m and C_b are the concentrations at the membrane surface and in the bulk of the brine stream, respectively).

^a Last tail element.

2.3. Flow reversal control of the BWRO plant

The onset and progression of scale formation during the operation of the BWRO plant was monitored by the EXSOD system via real-time membrane surface image capture and on-line image analysis using specially designed image analysis software tools. Captured images were stored and compared in order to eliminate false detections. Upon the emergence of mineral crystals, their number, size and surface area coverage were continuously monitored. In order to initiate a control action (i.e., feed flow reversal for the BWRO plant and permeate washing of the EXSOD RO cell or return to normal operation and EXSOD scale detection), a threshold level was set for the accumulation of mineral scale quantified in terms of fractional surface area coverage (FSAC) by scale or crystal number density (CND, i.e., #/cm²). The EXSOD software control system operates such that, when the set threshold is reached a low voltage signal is generated as: (a) an input to the BWRO PLC to initiate flow-reversal, and (b) a trigger to the EXSOD control system to initiate a permeate wash cycle in the EXSOD cell. Return to normal flow operation (i.e., when PV1 is the lead element) can be triggered by following the state of scale removal in the EXSOD monitor RO cell or a fixed wash time. In the current work, the BWRO was operated in the reverse flow mode for a period of 5 min less than the time it took to reach the specified mineral scaling threshold. It was found that the above period of washing of the EXSOD cell with a BWRO permeate side stream was effective in removing (via dissolution) surface crystals from the EXSOD membrane surface, consistent with previous reporting of the ability to remove gypsum crystals by dissolution via exposure to an undersaturated solution [12]. The above FFR/normal feed flow (NFF) was repeated automatically under the full-control of the EXSOD and BWRO plant PLC.

3. Results and discussion

A continuous RO operation in the FFR mode requires cleaning of the EXSOD membrane during each FFR cycle by dissolving the formed surface mineral crystals, preferably using the low salinity permeate stream from the RO plant. Therefore, a preliminary test was first carried out to assess both the capability of the EXSOD monitor to detect the onset and development of mineral salt scale on the EXSOD membrane surface and evaluate the repeatability of monitoring after each EXSOD membrane cleaning cycle. In this test (Run 1, Table 3), the BWRO operation was set such that the initial gypsum saturation at the membrane surface of the tail element of PV3 was 2.1 in the NFF mode (Fig. 2). At the above condition, the BWRO plant could be operated for a period of about 60 h without any noticeable flux decline from the third module (i.e., PV3), thus enabling testing of the EXSOD scale monitoring capability using a side-stream from PV3 with the EXSOD cell operating such that the initial SI_g at the test section of the membrane surface was ~2.4. This higher EXSOD initial SIg, relative to the condition at PV3 (in the BWRO plant), was selected in order to first evaluate the EXSOD performance at an accelerated rate of scale development, without concern for scaling of the BWRO plant over the ${\sim}25$ h EXSOD scaling/cleaning test period (Fig. 3; Run 1, Table 3). The results of this test run indicated that the flux from PV3 of the BWRO plant



Fig. 3. (a) Demonstration of EXSOD monitoring of mineral scale formation at an initial SI_g of 2.37 at the EXSOD membrane test section (Run 1, Table 3). A concentrate side stream from PV3 (Fig. 2), at an initial SI_g of 2.11 at the tail element in PV3, was the feed stream to the EXSOD cell. In the five cycles, the EXSOD feed was switched from the concentrate from PV3 to the permeate from the BWRO plant once the percent surface scale coverage of the membrane, at the EXSOD test section, reached 7.3%, 6.5%, 1.8%, 25.8%, and 13.2%. The EXSOD cleaning period is indicated by the horizontal dotted line. The BWRO plant operated in normal feed flow (NFF) such that the initial SI_g at the PV3 exit was 2.11 (Table 3). The onset of scaling in PV3 was detected only after ~60 h of operation in normal feed flow mode. (b) Crystal number density in the EXSOD monitor.

(operating at normal forward flow for the entire test period) was essentially unchanged, indicating lack of adverse impact of scaling on the BWRO over the period of the test. Once the percent scale coverage in the EXSOD membrane test section reached the prescribed threshold, the FFR signal triggered switching of the EXSOD feed to the permeate side-stream (from the BWRO plant) in order to dissolve the gypsum crystals. This process was carried out over five cycles. The threshold for triggering cleaning of the EXSOD (or "mock" BWRO FFR) was varied from about 1.8% to 25.8% surface scale coverage which corresponded to a range of crystal number density (CND) of 86–753 crystals/cm².

Gypsum scale, which developed in the EXSOD membrane cell test section (quantified by the mineral scale FSAC and CND; Fig. 3a and b), was effectively removed by dissolving the gypsum crystals with the permeate stream over a cleaning period five minutes shorter than the scaling period (Section 2.3). This was found to be the case by direct visual monitoring of the membrane surface, as well as by monitoring the permeate flux and conductivity from the EXSOD cell. Similar results were obtained (not shown) when the RO plant (tail element) and EXSOD cell were both operated at a gypsum SI of 1.9. The above test clearly demonstrated that the EXSOD monitor returned to its initial condition after each cleaning step although the temporal scale development pattern (as indicated by both the fractional scale coverage and crystal number count) varied somewhat from cycle to cycle (Fig. 3). This behavior is not surprising given that the mineral scaling process is governed by a random nucleation process.



Fig. 4. Demonstration of BWRO plant operation in normal feed flow mode (Run 2, Table 3). (a) Permeate flow from PV3 of the BWRO plant and fractional area coverage by scale in the EXSOD membrane test section. (b) Crystal number density in the EXSOD membrane test section and total permeate flux for the EXSOD monitor. Initial gypsum saturation was 2.9 and 2.57 at the BWRO membrane surface of PV3 and the EXSOD membrane test section, respectively (Table 3).

It is worth noting that above results suggest that when the gypsum saturation index in the EXSOD monitor is higher than that in the BWRO tail element, FFR cycles may be unnecessarily triggered. Conversely, if the scale monitor is operated such that the solution saturation at the EXSOD membrane surface is lower than in the BWRO plant, the onset of mineral scale formation in the BWRO plant may not be detected sufficiently early (i.e., prior to significantly measurable flux decline). In particular, under rapid scaling conditions (e.g., at high solution supersaturation; [11]), late detection of scale formation would be more pronounced when the EXSOD monitor is set to operate at SI below that of the tail element of the BWRO plant. Such an example is provided in Fig. 4 (see also Table 3, Run 2) in which the tail element (element 6 of PV3) SI of the BWRO plant was 2.9 (in NFF operation) and at 2.6 for the EXSOD cell. In this test, the EXSOD monitor indicated about 5% percent scale coverage (within the monitored membrane region) at t = 1.6 h, while flux decline for permeate production from PV3 was of 8% (Fig. 4). It is noted that at the above operating point (i.e. at t = 1.6 h) the EXSOD monitor indicated a CND of 175 crystals/cm² with the appearance of the first gypsum crystal detected at t = 0.35 h. Detection of the onset of mineral scale is feasible even when the solution saturation index is lower in the EXSOD cell relative to the BWRO plant tail element; however, the scaled area or CND thresholds for triggering FFR would have to be set at low levels. Notwithstanding, even under such settings flux decline in the BWRO would already be significant (e.g., \sim 4% flux decline in the BWRO when the EXSOD monitor indicates 1% scale coverage).

Effective control of FFR operation with the EXSOD monitor is possible even if the EXSOD cell operation is adjusted such that the initial SI_g in its membrane test section is below that of the RO plant,



Fig. 5. Demonstration of BWRO plant operation in normal feed flow and feed flow reversal (Run 3, Table 3). (a) Permeate flow from PV3 of the BWRO plant and fractional area coverage by scale in the EXSOD membrane test section. (b) Crystal number density in the EXSOD membrane test section and total permeate flux for the EXSOD monitor. Initial gypsum saturation was 2.37 and 2.0 at the BWRO membrane surface of PV3 and the EXSOD membrane test section, respectively.

provided that the rate of mineral scaling is not excessively rapid and that $SI_g > 1$. The results from such an operational mode (see Fig. 5; Run 3, Table 3) lead to scale detection (in terms of both the mineral scale FASC and CND) that is more sensitive than can be inferred from the observed flux decline (Fig. 5a). For example, when the BWRO plant and the EXSOD monitor operated such that their gypsum saturation indices were 2.4 and 2.0, respectively, 17% of the EXSOD monitored area scaled within a 6 h period in normal BWRO plant flow (i.e., from PV1 to PV3, Fig. 5a), with a corresponding 3% flux decline from the BWRO plant tail (PV3) module. Over the same period, flux decline of 8% was recorded for the EXSOD cell (Fig. 5b). In the above test case, feed flow reversal (i.e., from PV3 to PV1, Fig. 5a) was triggered at a threshold scale coverage of 17% (in the EXSOD monitored area). During the FFR period, permeate flux from PV3 (now the lead element) was higher than during NFF operation (i.e., $PV1 \rightarrow PV2 \rightarrow PV3$) since in the FFR operation, water feed entered the BWRO through PV3; thus, the concentration polarization modulus (i.e., $CP = C_m/C_b$, where C_m and C_b are the salt concentrations at the membrane surface and in the bulk, respectively) increased along the brine-side from PV3 toward PV1 (Fig. 2). Upon return to normal feed flow (t = 12 h) the PV3 permeate flux was back to its initial NFF scale-free level (i.e., at t=0 h). Similarly, the EXSOD permeate flux returned to its initial level (i.e., at t = 0 h).

Although triggering of flow reversal based on a threshold percent of scaled membrane area (in the EXSOD monitored area) is clearly a practical approach, it entails uncertainties at low threshold

values. As the scaling threshold is lowered the error in determining the scaled surface area will also increase, given smaller crystal size during the initial phases of surface crystallization [11]. On the other hand, the mineral scale CND is a more accurate measure of the evolution of membrane scale since the identification (and count) of evolving crystals can be completed with a higher degree of certainty than surface area measurements. For example, for the first and second FFR cycles shown in Fig. 5b, the onset of crystallization (i.e., the appearance of the first gypsum crystal) was detected at a time of 0.18 h and 0.33 h, respectively. It is important to recognize, however, that while an increase in the CND does correlate with increasing mineral scale FSAC, it is possible for similar scale area coverage to evolve with different mineral scale CND, depending on the rate of growth (i.e., size) of the evolving crystals. Therefore, it may be more appropriate to set a threshold for triggering the FFR based on considerations of both the mineral scale crystal count (i.e., CND) and surface scale coverage (i.e., FASC). It may also be reasonable to set the threshold for triggering FFR based on a critical time derivative of the above two parameters, with a safety threshold imposed by a maximum allowable mineral scale FASC and/or crystal count.

As discussed previously, there are a number of alternatives for setting the threshold for triggering FFR depending on the desired sensitivity and frequency of FFR cycles. Clearly, triggering FFR using an ex situ direct scale observation RO cell can be based on monitoring the evolution of surface scale coverage, crystal number density, permeate flux decline, conductivity or a combination of the above parameters and their temporal derivatives. In general it can be seen that the onset of mineral scaling can be determined more accurately by the evolution of the mineral scale CND than by the mineral scale FSAC. The temporal derivatives of scale coverage can be a sensitive indicator of the development of surface scale as indicated by a marked rise of the rate of scale growth (i.e., slope of the mineral scale FSAC-time profile) that occurs when there are very few crystals on the membrane surface ($\sim 20 \text{ crystals/cm}^2$ at t = 0.5 h; Fig. 4a and b). In the case of Run 2 (i.e., Fig. 4), using the more sensitive indicators (slope of mineral scale FSAC or CND) for triggering FFR would lead to more frequent triggering. Furthermore, where saturation indices are particularly high and scale onset is particularly rapid (as in Run 2, Fig. 4), using a surface coverage threshold is too insensitive for triggering FFR. This suggests that the sensitivity of the indicator chosen should be set based on the intended supersaturation conditions to be reached in the last element of the RO train, using the most sensitive indicators for the highest supersaturations. Practical implementation of the present approach to FFR operation and its control via a scale monitor would require direct real-time evaluation of the solution saturation at the membrane surface in the desired RO plant element. Operational FFR mode will require that the saturation index for the mineral scalant of concern is properly set in the EXSOD system relative to the BWRO plant. Current efforts are ongoing to develop both rapid experimental protocols and a theoretical framework for choosing the appropriate "scale onset" indicators as a function of the operating conditions of the RO plant.

4. Conclusions

An ex situ scale observation detector (EXSOD) was integrated with a brackish water RO (BWRO) pilot plant to evaluate the feasibility of operating the RO system in a cyclic normal feed flow (NFF)/feed flow reversal (FFR) operation with the ultimate goal of enabling scale-free operation of the BWRO plant in the absence of antiscalant feed dosing. The use of the EXSOD system with its online image analysis and control capability proved to be effective for scale detection, and as an actuator for initiating feed flow reversal in the RO plant. Effective NFF/FFR operation will require optimal setting of the saturation index for the mineral salt scalant in the EXSOD cell relative to the RO plant tail element. Scaling tests have shown that when the saturation index (SI) for the mineral scalant (gypsum in the present study) in the EXSOD RO cell is lower than in the tail element of the RO plant, the detection of the onset of mineral scaling can be delayed and thus creating a risk of RO plant scaling. When the gypsum SI was higher in the EXSOD cell, relative to the RO plant tail element, scale in the EXSOD cell was detected in advance of the onset of scaling in the RO plant. This latter condition would result in a high frequency of flow reversal cycles. Practical deployment of NFF/FFR operation would necessitate minimization of the frequency of FFR cycles while maintaining constant permeate productivity. Further studies are ongoing to establish the criteria for optimal NNF/FFR operation as controlled by direct ex situ scale detection.

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