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Renewable energy powered membrane technology: Impact of intermittency on membrane integrity

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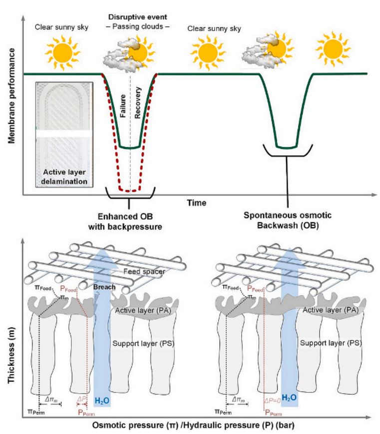
HIGHLIGHTS

- Spontaneous osmotic backwash is reliable in batteryless PV-NF/RO
- Frequent shut-down events (1000 cycles) did not affect the membrane integrity
- High-pressure increase sequence did not affect membrane integrity
- Membrane integrity loss occurred under enhanced OB with permeate backpressure

GRAPHICAL ABSTRACT



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ABSTRACT

Directly coupled photovoltaic-powered nanofiltration/reverse osmosis is a sustainable and cost-effective solution to brackish water desalination in remote areas. Intermittent operation of the system may cause physical membrane damage and loss of membrane integrity. The potential causes of integrity loss during intermittency include the sudden spontaneous restart process, frequent shut-down events, and osmotic backwash (OB) cleaning with controlled permeate backpressure. A bench-scale crossflow system powered by a solar array simulator was used to perform periodic fluctuation on filtration experiments that in this case cause intermittency. A wide range of feed pressure increase rates (0.17 to 2 bar/s) during the start-up process, up to 1000 shut-down events, and additional permeate backpressure up to 4 bar to enhance OB were applied. Results show that no significant membrane performance deterioration was observed at the highest feed pressure increase rate (2 bar/s), and when the shut-down event number increased to 1000 implying the robustness of NF/RO membranes and spontaneous OB cleaning. When increasing permeate backpressure to 2–4 bar to enhance the OB process, membrane integrity loss of both membranes was observed. This demonstrates the reliability and robustness towards fluctuations, intermittency, and spontaneous OB cleaning in a directly coupled photovoltaic-powered nanofiltration/reverse osmosis system if permeate backwash is avoided.

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1. Introduction

1.1. Intermittent operation of directly coupled photovoltaic-powered membrane systems

Directly coupling solar energy to nanofiltration/reverse osmosis processes provides the most cost-effective solution for safe drinking water issues in off-grid remote areas where solar energy is abundant [1,2]. These systems are preferably operated batteryless for robustness and lower capital costs, and hence a reduced water cost and low payback period [3]. For this scenario, the operating pressure and flow rate are unstable and fluctuate with solar irradiance fluctuations, which results in an intermittent operation that affects both permeate quantity and quality [4,5]. Such a system was reported to tolerate large fluctuations and intermittency [6], and no deterioration of membrane performance was observed in one-day operations [7–9]. However, the effect of such energy fluctuations on the integrity of the system, particularly the membrane elements that exhibit extreme and variable operational conditions, requires investigation to enable long-term operation.

1.2. Membrane integrity loss in nanofiltration/reverse osmosis

The standard performance (permeability and selectivity) or status of a membrane in perfect condition is known as membrane integrity [10,11]. Testing for membrane integrity loss is an effective means to monitor small changes in membrane material that lead to performance reduction. In nanofiltration/reverse osmosis (NF/RO) systems, the loss of membrane integrity can be due to two reasons. The first one is associated with the failure of the membrane system components, such as broken O-seals by mechanical stress inside the pressure vessel [12], and leaking in the glue line caused by permeate backpressure or membrane element telescoping due to fouling [13]. The second reason is associated with defects on the membrane surface, such as the formation of pinholes (breaches), increase in pore size, and deformation of the active layer caused by cleaning chemicals or in contact with the chlorine residual or other oxidants [14–18], as well as the delamination of the active layer from the support layer and forming blisters/imprints against the feed spacer due to permeate backpressure [12,19].

Such imperfections in the surface of NF/RO membrane elements can result in significant bacteria and viruses passage, resulting in compromised water quality and increased microbial regrowth after the filtration [20], as well as a decrease in salt retention accompanied by a flux increase [21]. This means that once the membrane integrity is lost, the safety of the produced water is at risk.

1.3. Scenarios resulting in membrane integrity loss due to intermittent operation

During intermittent operation, and particularly during the occurrence of the osmotic backwash (OB), which is a water backflow from the permeate to the feed driven by the osmotic pressure difference across the membrane, three scenarios may cause the loss of membrane integrity: (i) the sudden spontaneous restart-up process during fluctuation; (ii) frequent shut-down events, and (iii) OB with controlled enhancement of its cleaning efficiency via additional permeate backpressure. The harsh start-up process with a high rate of feed pressure increase may damage membranes due to hydraulic shock or excessive pressure/flow [19]. During shut-down events due to low solar irradiance (insufficient power to operate the pump), a spontaneous OB process occurs [22]. Such permeate backflow with frequent shut-down events may potentially cause membrane delamination [12]. This is aggravated if additional permeate backpressure is applied to further improve OB cleaning efficiency, particularly in the absence of a feed spacer. Scenario (iii) may not be inevitable for PV-NF/RO systems in intermittent operation compared to scenarios (i) and (ii), but it is relevant if OB is used as a cleaning strategy for scaling/fouling [23–25], when the cleaning efficiency would still require an additional permeate backpressure to enhance the OB volume/flux.

The above scenarios, mainly (i) and (ii), resulted from intermittent operation, may not only be limited to batteryless PV-NF/RO systems but also can apply to other NF/RO designs, such as in the case of multi-cyclic operation of semi-batch NF/RO [26], batch NF/RO [27], and feed reversal flow NF/RO [28], used to improve energy efficiency and membrane cleaning.

1.4. Membrane integrity monitoring

NF/RO membrane integrity monitoring methods can be classified as direct and indirect methods, which have been discussed in several reviews [29–33]. Direct integrity monitoring includes standardised pressure- and marker-based methods [34]. Pressure-based methods, such as vacuum- and pressure decay tests [11,30,35], are very sensitive for detecting breaches in NF/RO membrane sheets, O-ring defers, and glue-line failures [36]. Marker-based methods consist of adding particles, microorganisms, or molecules in the feed water that do not interfere or interact (adsorb) with the membrane and can be easily quantified. MS2 bacteriophage (MS2 phage) with a size of 25 nm, is the most frequently used marker to detect membrane breaches in NF/RO systems, even though such a surrogate is not desirable in full-scale drinking water plants which requires special expertise [20]. Instead, molecular fluorescent markers, such as fluorescein, uranine, eosin B, lissamine green B, and rhodamine WT, can achieve a maximum log removal value (LRV), which is the ratio of the log to the base 10 of the feed concentration divided by permeate concentration, of 4–6 log [37]. Other current and potential online surrogates of membrane integrity testing, such as laser-induced breakdown-detection (LIBD) for particles that can be used for the detection of membrane breaches down to 20 nm [38,39], can be used for high-pressure membranes such as NF/RO.

Indirect methods, which can be performed without interrupting system operation, such as total organic carbon (TOC), electrical conductivity (EC), and specific ion content (such as sulphate), can be used for real-time assessment of membrane integrity loss [30,40–42]. With such methods, the productivity is preserved, while the reliability of the NF/RO elements is improved [12]. However, the sensitivity of such conventional and low-cost methods depends on the feed water quality and the size of the membrane breaches [37]. For example, EC monitoring alone is insufficiently accurate to detect small breaches (pinholes) with an area of 0.3 to 1.2 μm^2 [21]. Such indirect methods (TOC, EC, sulphate monitoring) can monitor a maximum LRV of 3 log, which is 4 log lower than what can be achieved with direct methods such as MS2 phage [31]. In full-scale plants, such measurements will be averaged over typically large membrane areas (modules) and the localization of pinholes is very difficult.

However, in off-grid and remote areas, a simple and reliable method for membrane integrity monitoring should be adopted for decentralized PV-NF/RO systems. Direct methods will require manual collection of water samples with special pre-treatment and transport, which is not compatible with remote areas with large distances and limited access to reliable laboratories. In this case, indirect monitoring using inline EC and flowrate sensors, which have a lifetime of about 10–15 years [43,44], to indicate changes in permeability and salinity retention caused by membrane integrity loss.

To establish whether the three aspects during intermittency, sudden restart-up process, frequent shut-down events (regular OB cleaning), and enhanced OB via additional permeate backpressure, cause membrane integrity loss in directly coupled PV-NF/RO membrane systems used for brackish water desalination, three specific research questions were investigated: (i) does the sudden restart-up process with different feed pressure increase rates after fluctuations cause NF/RO membrane integrity loss?; (ii) does the number of shut-down events (OB cleaning frequency) increase to a point causing NF/RO membrane integrity loss?; and (iii) how does the enhanced OB process via additional permeate backpressure affect NF/RO membrane integrity?

2. Materials and methods

2.1. Bench-scale crossflow filtration system powered by solar array simulator

A bench-scale crossflow NF/RO system for testing flat sheet membranes was used for the membrane integrity experiments. The schematic of the system is shown in Fig. 1. The system was powered with a solar array simulator (SAS) (Chroma, 62050H-600s) in which voltage and current were simulated from the desired solar panel to a water pump (Wanner Engineering, Hydra Cell P200, Germany) with a DC motor (Baldor Electric, VP3428D, Germany) under controlled solar irradiance fluctuation. Other system components have been described elsewhere [4], except for a peristaltic pump (Minipuls 2, Gilson, France) implemented on the permeate side to provide additional permeate backpressure (up to 5 bar) for the enhanced OB scenario.

A feed spacer (90° diamond shape, thickness 0.56 ± 0.05 mm, hole size 3 by 3 mm) was used in the module, which has a feed flow channel of 0.19 m (L)·0.025 m (W)·0.0007 m (H) with $4.7 \cdot 10^{-3}$ m^2 effective membrane area. The hydrodynamics of the module are comparable to the spiral wound module, and hence allowing investigation on a bench scale [45]. The image of the feed spacer with a ruler is shown in the supporting information (SI).

2.2. Experimental design and filtration protocol

Periodic fluctuation experiments consisted of three different scenarios suitable for the above research questions: (i) sudden restart-up process where different pressure increase rates (0.17– 2 bar/s) were used to achieve the targeted applied pressure of 10 bar by adjusting the opening of the pressure control valve. This was performed in 3 cycles, each cycle 15 min of filtration at 10 bar and 3 min pump-off (shut-down event) followed by the restart-up process; (ii) frequent shut-down events with 1000 cycles, where each cycle consisted of 5 min filtration at 10 bar and 3 min pump-off (shut-down event). After 100 cycles during the day, which was selected as a worst-case scenario and as accelerated testing on membrane damage due to shut-down events, the system was off at night (intermittent operation) before this was repeated for 10 days; and (iii) OB with additional permeate backpressure during the shut-down event. For this, a peristaltic pump with different flow rates (manually turned on once the shut-down event starts) was used to induce different permeate backpressures. The filtration duration was 15 min 10 bar, followed by 5 min of enhanced OB process and reset to initial operation conditions (10 bar).

The filtration protocol was adopted from prior work for these experiments [4], starts with membrane soaking in 10 mM NaCl solution for 1 h, followed by membrane compaction at 10 bar applied pressure for 1 h to determine the pure water permeability, then a set-point adjustment (10 bar at 800 W/m^2) to achieve a stable and original membrane performance for 30 min prior the periodic fluctuant filtration experiment with OB, and ending with membrane inspection (pure water permeability determination after experiment) and system cleaning.

2.3. Membrane choice

Commonly used commercial flat sheet nanofiltration membrane NF270 (DuPont Water, FilmTec™, USA) and reverse osmosis membrane BW30 (DuPont Water, FilmTec™) were chosen to cover a wide range of NF/RO membrane performance. NF270 has a higher pure water permeability (15 ± 2 L/m^2h.bar) with lower salt retention (41 ± 2 % at 10 mM NaCl with 1 mM $NaHCO_3$) than that of BW30 (4 ± 1 L/m^2h.bar; 95 ± 2 % at 10 mM NaCl with 1 mM $NaHCO_3$). Detailed characteristics of both membranes were reported in previous studies [22,23]. NF270 membranes have a smoother, more negatively charged, and hydrophilic surface than BW30 membranes [24]. The active layer of NF270 is a semi-aromatic piperazine-based polyamide with a thickness 25 ± 5 nm, while

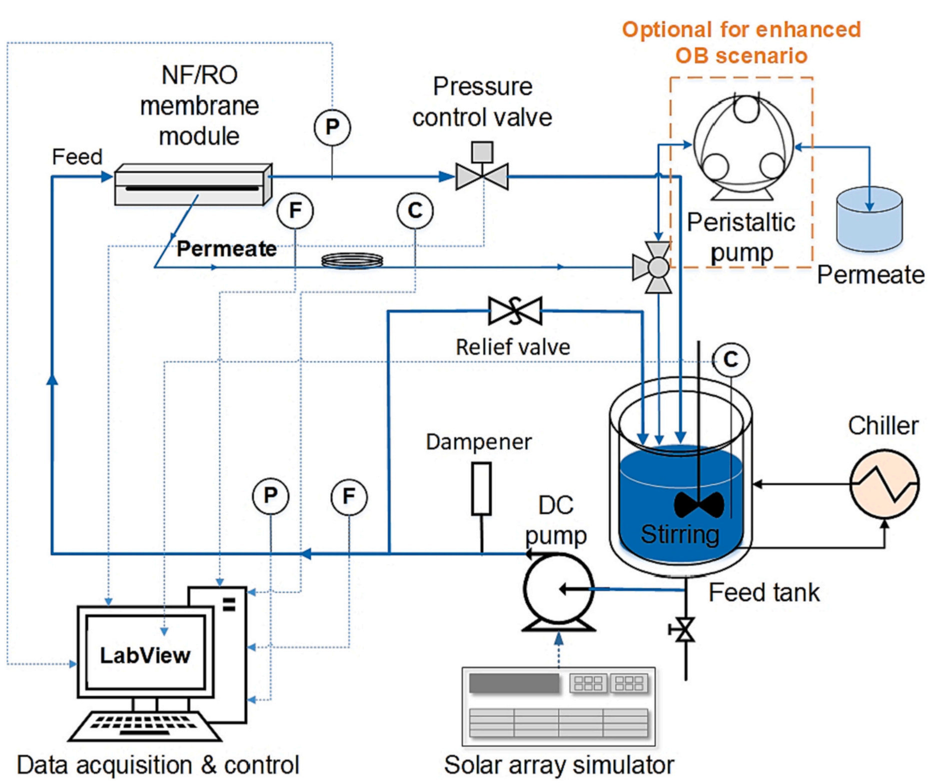

Fig. 1. Schematic of the bench-scale crossflow membrane system powered by solar array simulator with a peristaltic pump on the permeate side for permeate backpressure.

BW30's active layer is a fully aromatic polyamide with a thickness 233 ± 88 nm [46]. The amide groups in polyamide are vulnerable to frequent contact with cleaning chemicals and especially active chlorine species [47], which can result in permselectivity reduction and membrane lifetime shortening [48]. Chemical integrity is not investigated in this work, although a brackish water RO module can be used for up to 10 years if proper cleaning is carried out [49]. However, the cleaning of systems in remote locations is yet to be investigated as usage and disposal of cleaning agents in these situations is not trivial.

2.4. Solution chemistry and water quality analysis

Feed solutions were prepared with salinities of 1 and 10 g/L NaCl with 1 mM $NaHCO_3$, as synthetic brackish water with a natural water buffer. The resulting solution pH was 7.8 to 8.0. Stock solutions of 1 M NaCl, prepared from sodium chloride salt (EMSURE®, Merck Millipore, purity ≥99.5 %, Germany) and 100 mM $NaHCO_3$ (pH 8.2 ± 0.1) prepared from $NaHCO_3$ salt (Bernd Kraft, purity ≥99.7 %, Germany) were freshly prepared every two days. Milli-Q water (EC < 0.1 μS/cm, resistivity >18.2 MΩ/cm) produced by a Milli-Q® Reference A+ system (Merck Millipore, Germany) was used to prepare stock and feed solutions.

Common water quality parameters, pH and EC, in feed solutions and permeate samples were measured using a combined pH/Cond meter (WTW, pH/Cond 3320, Germany) with separate pH (WTW SenTix® 81, Germany) and EC (WTW TetraCon® 325, Germany) probes.

2.5. Membrane integrity monitoring parameters

The flux and permeate EC were monitored before and after the periodic fluctuation experiments. The permeate flux J_v was calculated by Eq. (1).

$$J_v = \frac{Q_p}{A} \tag{1}$$

where Q_p is the permeate flow rate, L/h; A is the effective membrane area, in this study, $4.7 \cdot 10^{-3}$ m^2.

Loss of membrane integrity is indicated when both flux and permeate EC increase out of the original range of membrane performance (>5 %) at the same operating pressure (10 bar). Following integrity experiments, the membrane coupon was taken out gently from the filtration cell for visual inspection. It should be noted that flux and EC monitoring may not provide sufficient resolution for quantifying the trend of integrity loss [30,50], but these parameters are fast and reliable qualitative indicators for integrity loss [30]. The maximum variation of measured parameters (such as flow rate, EC, and pressure) under a stable state was used to estimate the absolute error of the measured parameters, while the maximum calculated variation of parameters (such as flux) was used to estimate the absolute error of calculated parameters. The data in this paper have been adapted from the PhD thesis of Cai [51].

3. Results and discussion

The three integrity loss scenarios, (i) frequent shut-down, (ii) harsh restart-up, and (iii) enhanced OB with backpressure, are illustrated in Fig. 2. Integrity of NF/RO membranes was investigated when the OB occurred, followed by a relatively fast performance recovery resulting from harsh start-up pressure. Frequent OB and enhanced OB with applied permeate backpressure were also considered when investigating the membrane integrity. The start-up process was the first investigated scenario.

3.1. Membrane integrity with harsh start-up pressure scenario

During intermittent operation, sudden start-up processes (indicated with different pressure increase rates) may cause membrane integrity loss. Such damage is caused by forces due to sudden changes in pressure. For this, membrane suppliers recommend a proper start-up process to maintain membrane integrity, which is not always possible during intermittent operation. The applied pressure is defined as the feed pressure provided by the pump, used as a driving force to the membrane coupon, and measured by the pressure sensor. The applied pressure increase rate is defined as a rate (bar/s) to reach the target applied pressure (in this study, 10 bar) for membranes. Note that the transmembrane pressure is the applied (feed side) pressure minus the permeate side pressure. The membrane manufacturer recommends a soft start-up process with a pressure increase rate of <0.7 bar/s to prevent membrane damage due to hydraulic shock or excessive pressure [19]. Different feed pressure rising rates covering a wide range (from 0.17 to 2 bar/s) were applied during the sudden start-up process, as shown in Fig. 3A.

The start-up process with different rates of pressure increase ranging from 0.17 to 2 bar/s exhibited a duration ranging from 5 to 60 s to reach 10 bars (Fig. 3B). The soft start-up time window (30 to 60 s) is within the

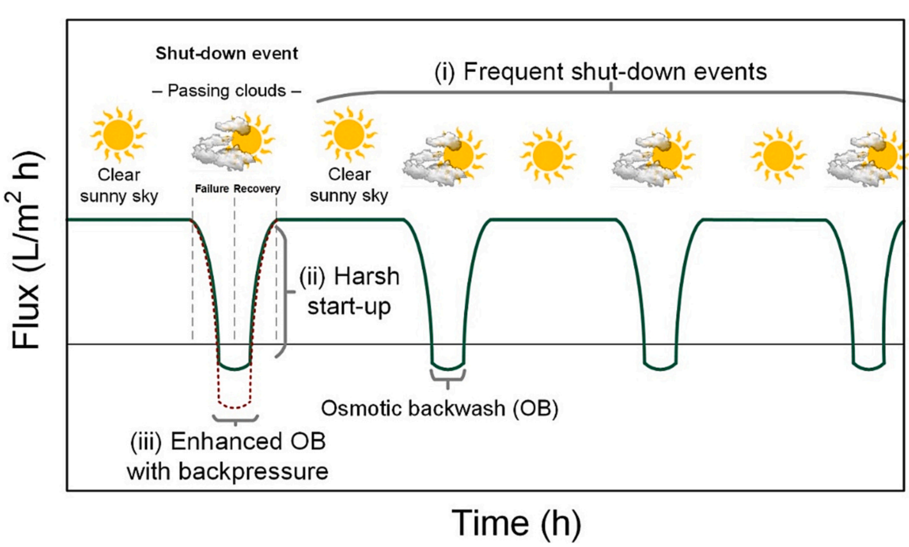

Fig. 2. Schematic of the three integrity loss scenarios in the example of flux as a performance indicator over time. The clouds result in reduced irradiance, which causes the pump to shut down due to power loss.

typical recovery time (30 to 350 s) of the PV-NF/RO system operated with real solar energy fluctuations [6], indicating that the system will not normally operate out of the recommended start-up window during realistic fluctuations. At a start-up time window ≤15 s, the membrane integrity loss may result from the sudden and harsh start-up process (start-up pressure increase rate >0.7 bar/s, the orange zone). To evaluate if membrane integrity loss occurs during the harsh start-up process, the performance of NF270 and BW30 in terms of flux and permeate EC was investigated after the occurrence of shut-down and OB (Fig. 4).

The flux reduction to 0 L/m^2 h for both membranes (NF270 and BW30) is associated with insufficient effective pressure to overcome the osmotic pressure (8.7 bars for 10 g/L NaCl) and achieve the separation process. This underlines the occurrence of the OB characterised by a water backflow driven by the osmotic pressure difference between the permeate and the feed compartments [22]. While permeate EC was expected to increase during spontaneous OB due to disruption of the concentration polarisation (CP) layer [52], a slight decrease in EC permeate was observed, which could have resulted from the sudden water backflow, the low permeate volume, or even the sucked back air that affected the operation of the EC sensor.

For both membranes, flux and permeate EC remained at the same level after applying a harsh start-up process (>0.7 bar/s), indicating that the integrity of both membranes was maintained after the intensive hydraulic shock. These results imply that both membranes in the bench-scale crossflow system can handle solar energy fluctuations, even at a low (fast) recovery time (≤15 s) resulting from a harsh pressure rise (>0.7 bar/s) to achieve the set point pressure of 10 bars. It should be noted that at the pilot scale, increasing the pressure beyond the soft start-up sequence recommended by the membrane manufacturer can induce a hydraulic shock leading to telescoping and/or fiberglass shell cracking of the membrane element [19]. It was not possible to investigate this risk with the bench scale methodology in this study.

In the next section, membrane integrity will be investigated in the frequent shut-down events (frequent solar energy fluctuations) scenario.

3.2. Membrane integrity in the frequent shut-down events scenario

A shut-down event is when the solar panel is not producing enough electricity due to low solar irradiance, causing the pump to shut down. This can result in a spontaneous OB, implying one shut-down event means one OB process. This section investigates whether the number of shut-down events (*i.e.* the number of spontaneous OB cleaning) increases to a point causing membrane integrity loss. Over 8000 min of operation, 1000 shut-down events were applied to verify the impact of frequent spontaneous OB processes on membrane performance. This integrity evaluation was carried out without a feed spacer, which resulted in a more significant stress on the membranes. Results are presented in Fig. 5, while the performance with the feed spacer and the results of 100 cycles are shown in Fig. S2.

Regardless of NaCl concentration (1 or 10 g/L), flux and permeate EC of NF270 and BW30 after 1000 shut-down events were similar to the first 10 cycles when operated in a worst-case scenario without a feed spacer. Similarly, with the feed spacer, no significant difference in membrane performance was observed for either membrane after 100 shut-down events (see Fig. S2). These results reflect the resilience of the process after 1000 shut-down events (OB cleaning) at the bench-scale crossflow filtration system and indicate that no membrane integrity loss has occurred. Such a periodic cleaning strategy was shown to be effective for fouling mitigation in RO spiral wound modules [53]. However, it should be noted that such frequent OB cleaning, combined with chemical cleaning, could accelerate membrane deterioration.

To put such shut-down events in perspective, the actual number of events is highly dependent on both system design and weather, where the level of fluctuation determines if a shut-down occurs causing intermittency. On a perfect sunny day there will be no shut-down, while complete cloud coverage will not experience shut-down either, provided the minimum irradiance for pump operation is available. On a mixed day, any number of clouds can cause any number of shut-downs. As a rough estimate, 1000 shutdowns may reflect operation over about one year. This confirms the reliability and robustness of the spontaneous OB cleaning process, at least in bench-scale membrane systems.

To further challenge membrane integrity, OB cleaning was enhanced with a permeate backpressure, which is investigated in the following section.

3.3. Membrane integrity with enhanced osmotic backwash scenario

Enhanced OB via additional permeate pressure may increase the OB cleaning efficiency, but it may adversely affect the membrane integrity. The membrane manufacturer reported that when the permeate pressure is higher than the feed pressure by >0.3 bar (this a negative transmembrane pressure), the membrane may delaminate and form blisters against the feed spacer, while the pattern of the feed spacer could be visibly imprinted on the membrane surface [19]. To investigate whether this scenario causes membrane integrity loss, a peristaltic pump was implemented on the permeate side to provide permeate pressure as an additional driving force to enhance the OB process during the shut-down event, and a feed spacer was used to be comparable with the spiral wound module in practice. The selected backpressure range (2, 4 bars) was selected based on the osmotic pressure range (1.8, 4.5 bar) of the permeate (2.2 g/L NaCl in BW30 permeate and 5.6 g/L NaCl in NF270 permeate when filtrating 10 g/L NaCl). The membrane performance

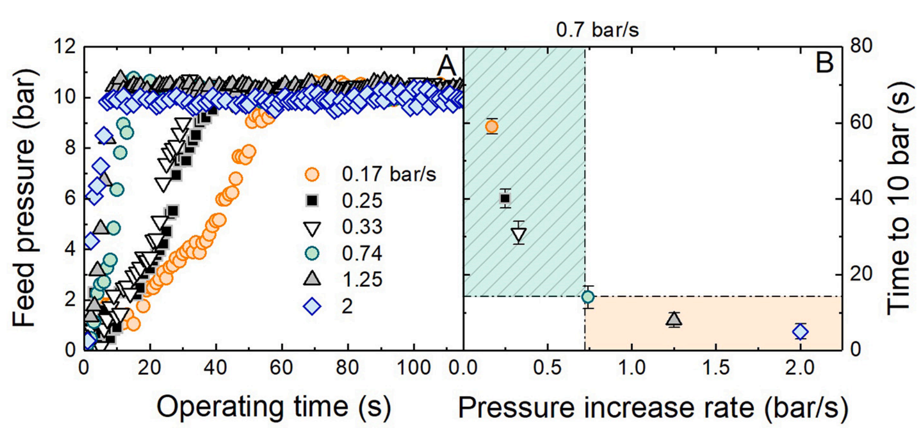

Fig. 3. (A) The sudden start-up processes with different sequences of pressure rise (0.17 to 2 bar/s) for both membranes (10 g/L NaCl, 1 mM $NaHCO_3$; 21 ± 1 °C); (B) time to achieve 10 bar applied pressure as a function of average start-up pressure increase rate; the green zone with pattern is soft start-up window recommended by membrane manufacturer while the orange zone may cause membrane integrity loss.

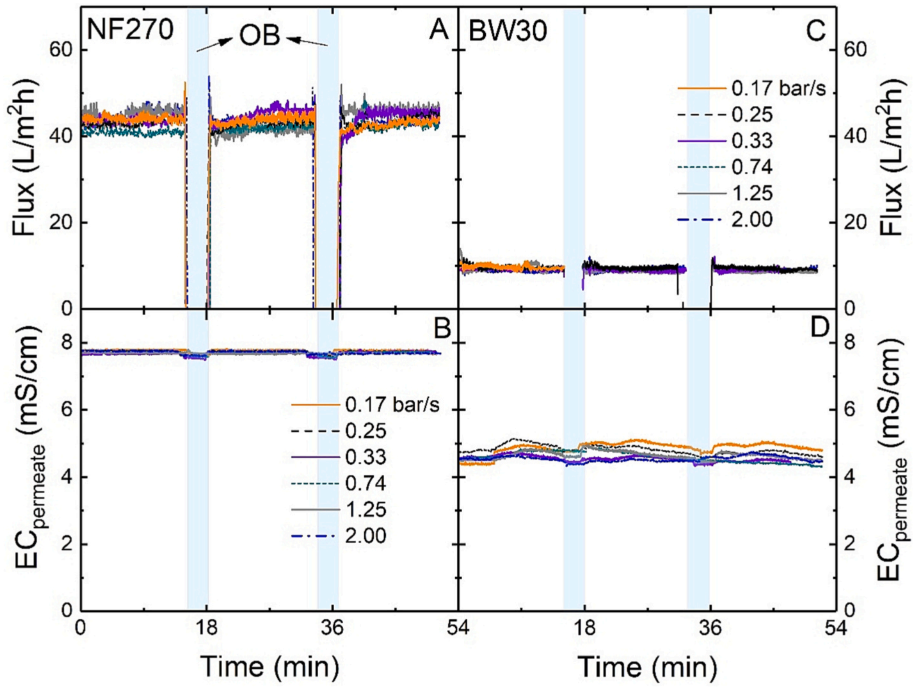

Fig. 4. (A, C) Membrane flux and (B, D) permeate EC with different applied pressure increase rates as a function of operating time; (A, B) for NF270 membrane, (C, D) for BW30 membrane. Three cycles: each cycle 15 min of 10 bar filtration with 3 min of OB (10 g/L NaCl, 1 mM $NaHCO_3$).

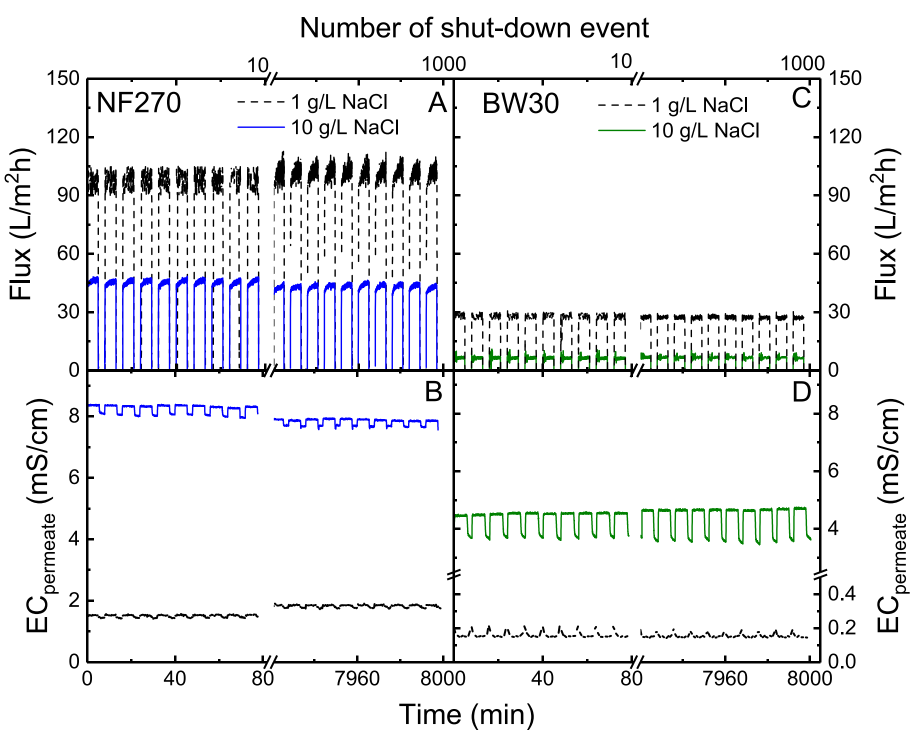

Fig. 5. (A, C) Membrane flux and (B, D) permeate EC with different salinities as a function of operating time. (A, B) for NF270 membrane and (C, D) for BW30 membrane. (1, 10 g/L NaCl, 1 mM $NaHCO_3$; 21 ± 1 °C).

with the feed spacer before and after the enhanced OB is shown in Fig. 6. The effect of enhanced OB with backpressure on membrane integrity without the feed spacer is shown in in Figs. S3 and S4.

After applying 2 bar permeate backpressure during the shut-down event, both flux and permeate EC of the BW30 membrane increased abnormally, while that of the NF270 membrane remained unchanged. When 4 bar of permeate backpressure was applied to the NF270 membrane, an abnormal increase of flux and permeate EC was observed. These results indicate that enhanced OB during shut-down events will cause membrane integrity loss if the backpressure is high enough for a given membrane. In this case, NF270 was more robust than BW30. Clearly, if an enhanced OB is implemented, this requires a careful control strategy to prevent membrane integrity loss [54].

When investigating the damage caused, an analysis of the permeate

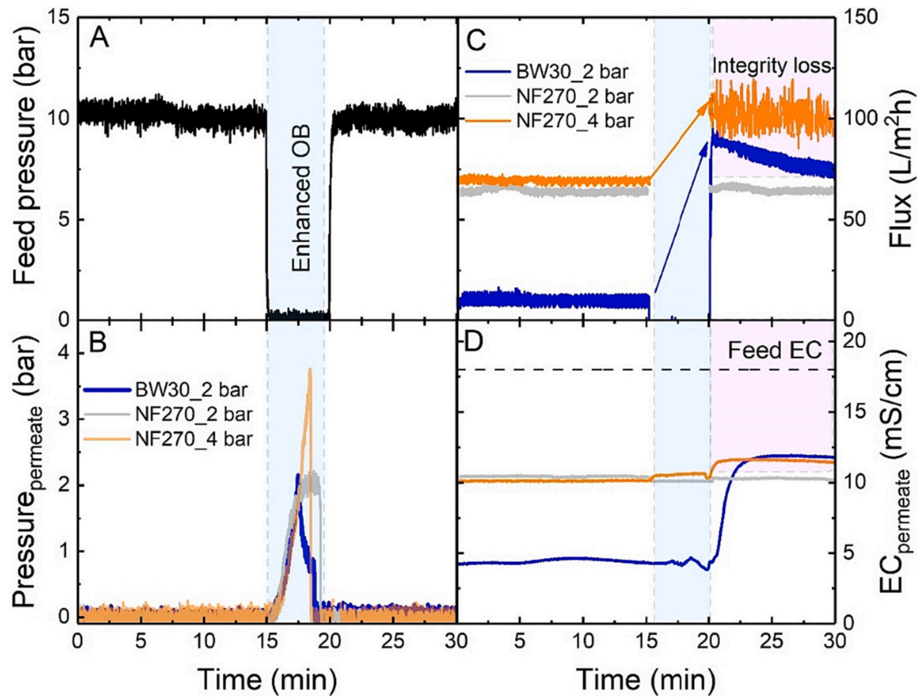

Fig. 6. Membrane performance operated at 10 bar with enhanced OB processes via additional permeate backpressure (2, 4 bar) as a function of operating time: (A) applied pressure, (B) permeate backpressure, (C) flux, and (D) permeate EC. 5 min of enhanced OB, with feed spacer (10 g/L NaCl, 1 mM $NaHCO_3$; 21 ± 1 °C).

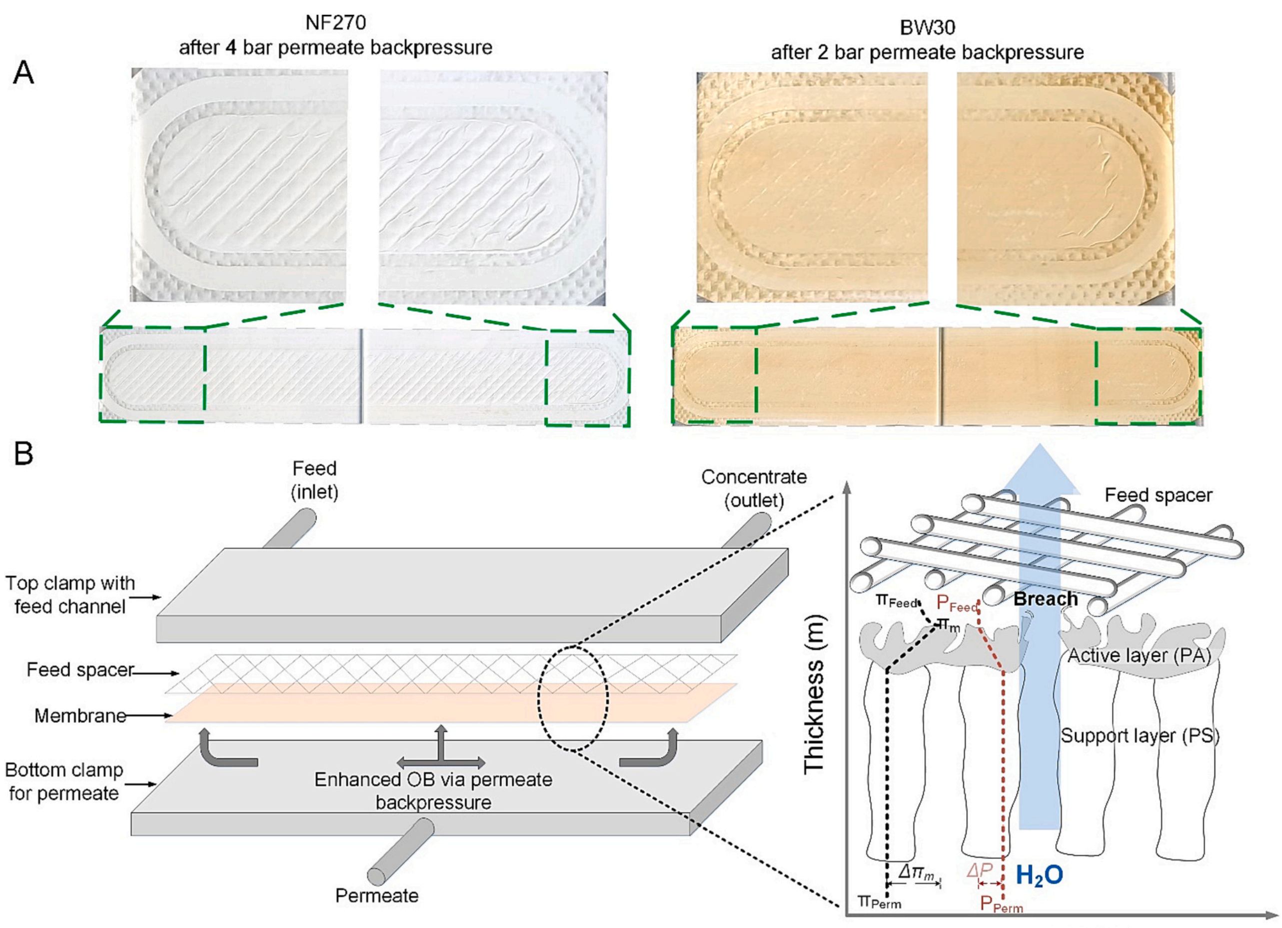

Fig. 7. (A) Optical images of NF270 and BW30 membranes with compromised integrity (1000 cycles). The applied backpressure was 4 bar (NF270) and 2 bar (BW30) with experiments from Fig. 6. (B) Proposed mechanism of enhanced OB via additional permeate backpressure causing membrane integrity loss.

pressure (Fig. 6B) shows that the permeate backpressure of membrane integrity loss dropped sharply after the peak. This pressure release is most likely due to the formation of cracks/imprints on the membrane surface against the feed spacer as shown in Fig. 7A. These images indicate a membrane integrity loss resulting from the enhanced OB process with additional permeate backpressure. The NF270 membrane surface had more visible imprints of the feed spacer and some blisters against the feed spacer than the BW30 membrane, probably due to the thinner active layer of NF270 and higher permeate backpressure than that of BW30 membranes. Such physical damage was reported to affect the roughness of the membrane surface at the nanoscale [55]. In the absence of a spacer (Fig. S3) this damage is more significant.

Enhanced OB cleaning with permeate backpressure was the only scenario that resulted in membrane integrity loss observed by physical membrane active layer damages/deformation. The proposed mechanism is shown in Fig. 7B.

The mechanism that enhanced OB causes membrane integrity loss: high permeate backpressure pushes the membrane active layer against the feed spacer, leaving the imprints/cracks of the feed spacer and causing physical damage to the NF/RO membrane active layer. This delamination of the membrane active layer against the feed spacer is typical in NF/RO spiral wound modules operated with excessive permeate backpressure [12,19]. When filtration/desalination restarts, water flow, and salt ions could easily pass through the ruptured/broken membrane active layer, causing abnormally high flux and permeate EC. Without a feed spacer, the deformation and subsequent damage were more significant.

4. Conclusions

This work investigated three main scenarios that could potentially cause membrane integrity loss during intermittent operation caused by fluctuation in irradiance in directly coupled PV-NF/RO systems, namely (i) restart-up processes, (ii) shut-down events, and (iii) enhanced OB. Major findings can be summarised as follows.

The membrane performance was maintained when applying a wide range of feed pressure rates (up to 2 bar/s) during the restart-up process, validating the reliability of fluctuations and NF/RO membranes. After 1000 shut-down events (*i.e.* 1000 spontaneous OB), the integrity of NF/RO membranes was maintained, demonstrating the reliability and robustness of the NF/RO membranes and spontaneous OB process. Membrane integrity loss was observed after enhanced OB via additional permeate backpressure during shut-down events. The high backpressure induced imprints/breaches of NF/RO membrane active layer against the feed spacer, which is confirmed by visual inspection. Therefore, a permeate back-pressure strategy is not recommended to enhance OB cleaning.

These findings emphasise the reliability and robustness of the PV-NF/RO system operated under energy fluctuations where spontaneous OB may occur during intermittent operation.

CRediT authorship contribution statement

Yang-Hui Cai: Writing – review & editing, Visualization, Validation, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Youssef-Amine Boussouga:** Writing – review & editing, Writing – original draft, Data curation. **Andrea I. Schäfer:** Writing – review & editing, Supervision, Resources, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supporting information includes i) images of the feed spacer and its installation on the feed channel; ii) the effect of 100 shut-down events on NF/RO membrane performance with feed spacer; iii) membrane integrity with enhanced OB and without spacer. Supplementary data to this article can be found online at https://doi.org/10.1016/j.desal.2024.117504.

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