Zero Waste Discharge in Wastewater Reclamation System for Small Cities

G. Naidu, M. A. H. Johir, S. Shanmuganathan, A. Listowski and S. Vigneswaran

Abstract Presently, wastewater reclamation plants (WRPs) are widely adopting reverse osmosis (RO) as a polishing treatment in the final operation stage. Sydney Olympic Park Authority's (SOPAs) WRP adopts a treatment operation of combined biological treatment, microfiltration and lastly RO. RO advantage is in maintaining a consistently good quality standard of water. Nevertheless, major limitations of using RO in SOPAs WRP are (a) high cost of RO and fouling susceptibility and (b) production of substantially large volume of wastewater RO concentrate (WWROC). The aim(s) of this study were to evaluate methods to overcome both these limitations by (i) investigating the integration of nanofiltration (NF) prior to RO to reduce cost and fouling issues; and (ii) evaluating the performance of membrane distillation (MD) for treating WWROC to achieve zero waste discharge. The results of this study highlighted (i) blending NF and RO permeate, acquired from a hybrid two-stage NF–RO process resulted in a water quality suited for irrigation. Utilizing NF prior to RO, reduced RO membrane fouling was more cost-effective compared to direct RO application. The hybrid NF–RO removed most of the micropollutants and therefore achieved reuse water quality that was safe to be applied for irrigation; and (ii) MD displayed minimal flux decline (13–15%) at 85% water recovery of WWROC and achieved freshwater with high quality (10–15 μS/cm, 99% ion rejection). The organic contents of micropollutants in WWROC were effectively reduced with granular activated carbon (GAC). This enabled to achieve good quality water (micropollutants-free) by MD with the potential of reuse.

Keywords Nanofiltration · Membrane distillation · Wastewater reclamation plant

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1 Introduction

The application of reverse osmosis (RO) as a final polishing treatment is an increasing trend in many wastewater reclamation plants [1, 2]. RO is beneficial in attaining a consistent water standard. Nevertheless, the application of RO is still challenged by membrane fouling and scaling issues, high energy needs, low rejection of several micropollutants and the generation of high volume of wastewater RO concentrate (WWROC) at an average of 20–25% of the feed stream volume [3, 4].

For instance, Sydney Olympic Park Authority's (SOPAs) WRP in Australia treats wastewater (biologically treated sewage effluent and stormwater) using a series of treatment processes (biological treatment and microfiltration followed by RO) for removal of particulate matter and pathogens and minimizing total dissolved solids and micropollutants. The RO, at 80% water recovery, operates at 55 m^3/h volumetric feed flow. As a result, an average of 300 kL of WWROC/day is discharged from this WRP.

Fouling in RO is predominantly attributed to the adhesion of suspended solids, inorganic ions as well as dissolved organic carbon (DOC) and on the surface of the membrane which subsequently blocks the membrane pores, invariably compromising its performance. In this regard, a pretreatment strategy which is cost-effective could potentially reduce membrane fouling. This will minimize the requirement of RO membrane cleaning and membrane replacements, effectively reducing RO operational costs. A number of approaches have been evaluated for reducing membrane fouling, such as air scouring the membrane surfaces of submerged membrane system. Although aeration effectively reduces deposition of solids' particles on the membrane surface through air scouring [5], irreversible membrane fouling due to DOC deposition inside the membrane pores, remains a challenge. In this regard, previous studies have highlighted that the incorporation of suspended media adsorbents in a membrane reactor system was effective in reducing membrane fouling through (i) mechanical scouring; and (ii) sorption of DOCs. Moreover, the mechanical scouring on the membrane surface by adsorbents in the membrane reactor helped to reduce membrane fouling and averted transmembrane pressure (TMP) build-up [5]. It must be highlighted that although the membrane adsorption hybrid system does exhibit effectiveness for DOC removal, it is also important to remove micropollutants present in the secondary effluent. Removal of micropollutants can be achieved through more advanced membrane treatments, namely NF or RO. In this regard, the advantages of NF over RO are low operational pressure and high filtration flux. Comparatively, operation and maintenance costs of RO are higher than that of NF. Moreover, NF shows promising performance in rejection of micropollutants and selective inorganic ions [6]. NF is considered as a feasible substitute to RO.

Another major issue with RO is that the WWROC generated comprises of salt and inorganic ions at higher levels compared to the original wastewater including micropollutants. That may be toxic or bioaccumulative [4]. Discharge of WWROC containing micropollutants into natural water sources may potentially increase the risk of eco-toxicological. As such, it is necessary to treat WWROC prior to discharge

[1–4]. Much attention is being focused on methods to treat WWROC in WRPs sustainably. In this regard, rather than treating and subsequently discharging WWROC, a more favourable option would be to obtain zero waste discharge of WWROC. Membrane distillation (MD) is a potential method to achieve zero waste discharge of WWROC [7]. Thermal MD process operates by vapour pressure difference (created by temperature difference) through heated feed solution and cold permeate (distillate), across a hydrophobic membrane. The moderate feed temperature requirement (50–70 °C) enables heat waste or solar integration. The mass transfer mechanism of vapour in MD potentially offers high ion rejection. This promises addition high-quality water while concentrating the WWROC, systematically reducing the volume of WWROC.

The aim(s) of this study were to evaluate methods to overcome limitations of wastewater treatment in WRPs by (i) investigating the integration of NF 'dual membrane—hybrid system' as an alternative low cost system prior to RO and (ii) evaluating the performance of MD for treating WWROC to achieve near-zero waste discharge.

2 Materials and Methods

2.1 Micro Filtration (MF)–GAC Hybrid System Followed by NF System

2.1.1 MF–GAC Hybrid System

In the membrane hybrid system setup used [8], a flat sheet MF membrane module was immersed in the reactor tank (10 L). The flat sheet MF membrane was made up of polyvinylidene fluoride (PVDF) (A3 Membrane Company, Germany) with a surface area of 0.2 m^2 and pore size of 0.14 mm. The reactor tank was continuously fed with biologically treated sewage effluent (BTSE) using hydraulic pumps to control the inflow and outflow as well as maintain a constant BTSE volume in the reactor. An air flow rate of 1.5 m^3/m^2.h was used to keep the GAC particles in suspension in the reactor [2, 5]. An initial 2 g/L GAC was added into the reactor.

2.1.2 Dual Membrane System

Nanofiltration (NF) was used as a polishing stage with the effluent from the MF–GAC hybrid system (as described in Sect. 2.1.1) as feed (Fig. 1). A flat sheet NF membrane (NTR729HF) made of polyvinylalcohol/polyamides with a molecular weight (MW) cutoff of 700 Da was used. The effective membrane area was 68 cm^2. Prior to each experiment, the NF system was cleaned with 0.1 M NaOH solution followed by 0.1 M HNO_3 solution for 2 h and then, Milli-Q water for 1 h to remove impurities

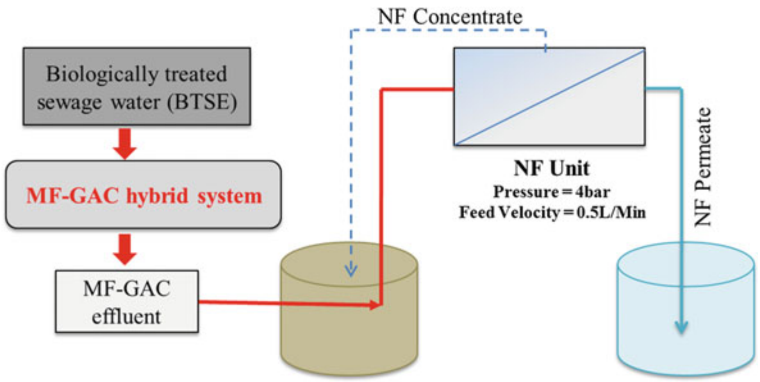

Fig. 1 Experimental schematic diagram of dual membrane system

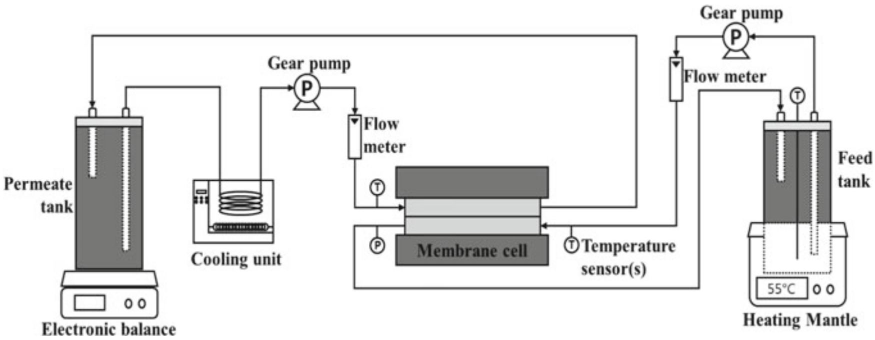

Fig. 2 Experimental setup of DCMD

from the unit. The NF feed flow was set at 0.5 L/min at pressure of 400 kPa (4 bar). The NF concentrate and permeate samples were collected for DOC, inorganic anions (Ca, Mg) and micropollutants analysis.

2.2 Membrane Distillation (MD)

A bench-scale direct contact MD (DCMD) was used in this study (Fig. 2) [7]. A hydrophobic polytetrafluoroethylene (PTFE) flat sheet membrane (General Electric, US) was used with a 40 cm^2 effective membrane area. The temperature of the feed and permeate solutions were set at 55 and 25 °C, respectively. The feed and permeate solutions (2 L each) were channelled with a gear pump into the membrane cell at a feed and permeate flow velocity of 1.1 m/s in a countercurrent mode.

Table 1 Ionic composition of BTSE and WWROC from SOPA

Ions	BTSE (mg/L)	WWROC (mg/L)
Ca^{2+}	30.5	88.2
Mg^{2+}	12.1	72.0
Na^{+}	100.4	445.2
K^{+}	18.2	63.4
SO_4^{2-}	50.3	198.1
Cl^{-}	225.4	605.3
PO_4^{3-} as total P	2.1	2.8
F^{-}	0.9	2.7
NO_3^{-} as N	5.3	7.8

2.3 Feed Solution

In this study, two feed solutions (biologically treated sewage effluent, BTSE and wastewater reverse osmosis concentrate, WWROC) were used, which was obtained from SOPAs WRP. BTSE was used for the experiments with MF–GAC hybrid system followed by NF system. The pH and dissolved organic carbon (DOC) of BTSE was 7.05–7.11 and 3.6–7.7 mg/L, respectively. Meanwhile, WWROC was used for the experiments with DCMD. The DOC, turbidity and pH of the WWROC were 58–60 mg/L, 0.11 NTU and 8.01–8.10, respectively. The ionic composition of BTSE and WWROC are listed in Table 1.

2.4 Granular Activated Carbon (GAC) as Adsorbent

Coal-based GAC (MDW4050CB, James Cumming & Sons Pty Ltd) was used and its particle size was in the range of 425 and 600 μm. The average pore diameter and surface area of GAC were 30 Å and 1000 m^2/g, respectively.

2.5 Analysis

The DOC was characterized by liquid chromatography with organic carbon detection (LC-OCD), while the inorganic ions was detected using inductively coupled plasma-mass spectrometry (ICP-MS, Agilent 7500, Agilent, USA). The micropollutant concentrations were detected by high-performance liquid chromatography (HPLC) with tandem mass spectrometry detector. The MD membrane autopsy was carried out using water contact angle and scanning electron microscope (SEM) and energy-dispersive X-ray spectroscopy (EDS).

3 Results and Discussion

3.1 MF–GAC Hybrid System Followed by NF System

3.1.1 Performance of MF–GAC Hybrid System

The DOC removal capacity of the MF–GAC hybrid system correlated to its operating settings such as flux and rate of GAC replacement. The results showed that the best performance of MF–GAC hybrid system was obtained at low filtration flux of 2.5 L/m^{2}h, achieving a DOC removal rate of more than 80%. In comparison, higher filtration flux (10 L/m^{2}h) compromised the DOC removal efficiency (40–60%) despite a higher GAC replacement rate of 30%. Low filtration flux and high GAC replacement rates are not sustainable. As such, a combination of average filtration flux (5 L/m^{2}h) and GAC replacement (10%) were identified as the most suitable operation condition for MF–GAC hybrid systems [8].

3.1.2 Performance of MF–GAC Hybrid System Followed by NF System

The MF–GAC hybrid system was effective in removing major organic compounds. Hydrophobic and hydrophilic (especially humics)-based organic compounds were removed. This subsequently reduced membrane fouling. The NF operation showed minimal TMP increase and flux decline when the MF–GAC hybrid system was used as pretreatment. The DOC removal by NF was excellent (95%). In terms of the different organic fractions removal, lower rejection of LMW neutrals (84%) was observed compared to humics/building blocks (98%). This was because NF organic rejection correlated to molecules size and charge. In terms of micropollutant removal, MF–GAC followed by NF hybrid system is enabled to reject the majority of the micropollutants. However, the removal of inorganic ions was ineffective with MF–GAC hybrid system alone, while on the other hand, a significant rejection of divalent (Ca, Mg and SO_4) were observed with NF (60–99%).

3.1.3 Product Water Quality Evaluation for Irrigation

The treated BTSE (through MF–GAC) contained minimal micropollutants. Therefore, the prospect of using the treated BTSE for irrigation is favourable for maximizing water reuse in arid and semi-arid regions. However, the MF–GAC-treated BTSE may not be suitable for irrigation in view of the high sodium adsorption ratio (SAR) value, in which, the Na and Cl contents are higher than the maximum allowable limits for sensitive crops. In this regard, the subsequent polishing step of using NF was effective in removing all micropollutants as well as divalent cations (Ca, Mg). However, the inorganic ion evaluation showed that the SAR value of the NF permeate was still above 14 (above the SAR safety levels for irrigations). The Cl and Na concentrate in the NF permeate were 202 and 110 mg/L, respectively. These levels are significantly above the maximum allowable levels for sensitive crops. In comparison, RO process can remove all the inorganic ions below critical levels. However, it would also remove essential ions which are important plant nutrients. In view of this, the potential of blending NF and RO permeate with feed water was considered in this study.

The results highlighted that blending feed water (10%) with RO permeate (90%) enabled to achieve a SAR value of 6 with reduced contents of Cl (40 mg/L) and Na (15.5 mg/L). This condition would enable to rectify soil infiltration issues, while reducing toxicity due to the presence of excess Cl and Na. More importantly, the blending of NF and RO permeate was a more economically sustainable option instead of using 90% RO permeate with 10% raw feed water. For instance, blending an equal portion of NF and RO (50% NF and RO permeate) enabled to achieve a SAR value of 8 with Cl and Na concentrations of 109 and 57 mg/L, respectively.

3.2 Direct Contact Membrane Distillation

3.2.1 DCMD as a Treatment Option for WWROC

WWROC treatment with DCMD enabled to achieve a considerably good quality distillate (freshwater) and an 85% water recovery, with only a small permeate flux reduction (Fig. 3). This was attributed to the low NaCl content in WWROC (only 3 g NaCl/L), resulting in the minimal effect of polarization compared to seawater ROC. At high water recovery, the WWROC volume was significantly reduced, ultimately leading to a highly saturated WWROC. This was a suitable condition to precipitate and crystallize ions with low dissolving capacity such as Ca, Sr, Mg and F. The precipitation was beneficial to reduce the inorganic ion in WWROC-MD brine. Consequently, the ability of MD to achieve concentrated WWROC enabled to meet a close to zero waste discharge and salt recovery potential from WWROC.

3.2.2 GAC Pretreatment for WWROC

In WWROC, the high DOC concentrations (40–60 mg/L) lead to significant MD membrane organic deposition. This result in prevalent membrane hydrophobicity reduction by up to 70%, based on contact angle (CA) reduction from 139° to 20° (Fig. 3). Chemical cleaning could not restore the MD membrane hydrophobicity, indicating irreversible organic fouling on MD membrane and membrane wetting in long-term MD operations. Therefore, GAC as pretreatment to reduce WWROC organic contents is necessary prior to MD operation. The results showed that a simple GAC sorption was effective in reducing 46% of organic content in WWROC. The GAC pretreated WWROC did not severely affect membrane hydrophobicity and this was restored by chemical cleaning. Moreover, apart from organic compounds, the

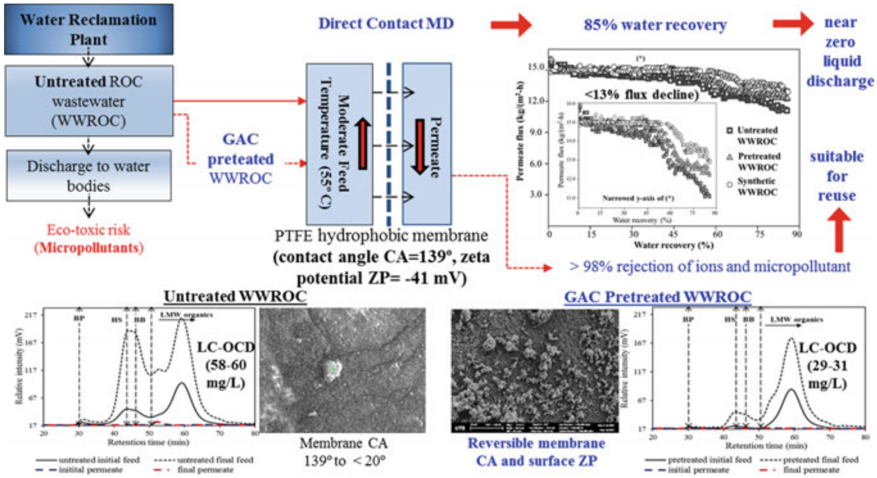

Fig. 3 Performance of DCMD with WWROC using GAC pretreatment

GAC was found to be an effective pretreatment in adsorbing hydrophobic micropollutants as well as hydrophilic micropollutants. This enabled to obtain high removal of major micropollutants, ensuing permeate (freshwater) of good quality, suitable for reuse.

4 Conclusion

The dual membrane hybrid system (MF–GAC followed by NF system) was an effective treatment strategy for removing DOC, micropollutants and selective divalent inorganic ions. Utilizing treated BTSE for irrigation was a favourable option for maximizing water reuse in arid and semi-arid regions. Blending the treated BTSE permeate (MF–GAC followed by NF) and RO permeate were a cost-effective option, given that RO is more expensive than NF. The blending approach is an added advantage as RO only needs to treat 50% of the NF permeate. Moreover, the effective organic reduction (more than 85%) using the dual MF–GAC hybrid system was beneficial in minimizing RO fouling issue. The blending proportion of NF–RO permeates can vary depending on the type of membranes used, soil type, salt tolerance of crops, salts in the soil solution and wastewater (feed) characteristics (Fig. 4).

Meanwhile, DCMD showed promising results for treating wastewater reverse osmosis concentrate (WWROC). DCMD achieved 85% water recovery with minimal flux reduction (13–15%) and obtained good quality freshwater (10–15 μS/cm, 99% ion rejection). A simple GAC pretreatment prior to DCMD was useful in minimizing organics in WWROC and micropollutants. This enhanced the reuse potential of the freshwater permeate produced in MD. Selective ion precipitation of the concentrated

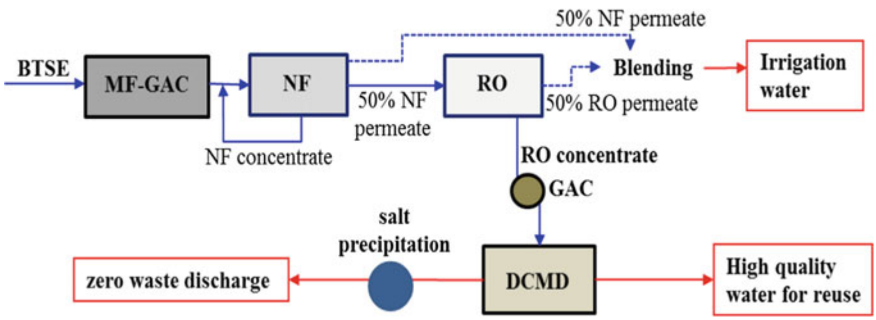

Fig. 4 Potential of using treated BTSE by the approach of blending NF–RO permeate and achieving high-quality water reuse and zero waste discharge of ROC through DCMD operation

MD WWROC must be evaluated to obtain a near zero waste discharge in WRPs (Fig. 4).

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