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Complete sustainability in electrodialysis reversal desalination: reusing tertiary-treated municipal wastewater as feed in the concentrate stream and electrodes rinsing water

Maung Thein Myint[a,*], Abbas Ghassemi[a,b], Nagamany Nirmalakhandan[c]

[a]*Institute for Energy and the Environment, New Mexico State University, MSC WERC, P.O. Box 30001, Las Cruces, NM 88003, USA*
Tel. +1 575 646 2073; Fax: +1 575 646 5474; email: mmyint@nmsu.edu
[b]*Department of Chemical Engineering, New Mexico State University, Las Cruces, NM 88003, USA*
[c]*Department of Civil Engineering, New Mexico State University, Las Cruces, NM 88003, USA*

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ABSTRACT

Electrodialysis reversal (EDR) has been innovative based on unplanned indirect potable water reuse to replace the same amount of raw water withdrawn from brackish (sea) water source and conserving 15–20% of freshwater by reusing tertiary-treated municipal wastewater (TTMWW) as feed in the concentrate and electrode stream while brackish groundwater (sea water) remains feed into dilute stream. By substituting TTMWW, our analysis shows the resulting total dissolved solid (TDS) concentration in the waste stream lower than that in the original source of brackish groundwater (sea water) which fed into dilute stream. The waste from EDR system with TTMWW as feed in concentrate and electrode streams can be recharged back into groundwater (sea water) source which do not add the TDS concentration. In fact, it reduces the TDS concentration by dilution. New significant findings are: (1) 17,500 m^3/d of freshwater could be saved with the data from 1990; the saving would be increased in 2011 and much more in future due to the increasing installation of ED/EDR and (2) cost reductions by enhancing the life of membrane and electrodes; and by reducing current required to attract ions and antiscalant due to low concentration of TDS and low scale causing ions in the TTMWW which fed into concentrate stream.

Keywords: EDR; Electrode rinsing water; Feed in concentrate; Fouling; Scaling; Unplanned indirect potable water reuse

1. Introduction

Scarcity of water, rapid population growth, and global warming have made water reuse popular to conserve and to extend water supplies especially in semi-arid regions. Communities in the semi-arid regions value water reuse for nonportable use to meet their water requirements with no significant health effects [1,2]. Indirect portable water reuse (IPWR) has been transferred into high-quality recycled water for potable use by using the tertiary water treatment technologies [2,3]. Highly tertiary-treated municipal wastewater (TTMWW) effluents are discharged into surface water and/or groundwater sources to increase the drinking water supply which is defined as IPWR [1,2]. The IPWR has been successfully practiced and appreciated [2–4].

Unplanned IPWR has naturally taken place for centuries; the secondary-treated municipal wastewater

*Corresponding author.

(STMWW) effluent is discharged back into surface water and subsequently pumped, treated, and reused as potable water by downstream customers [2,3]. This unplanned IPWR practice is adopted and engineered in water-scarce region to increase the potable water resource by injecting TTMWW into inland aquifer to improve water quality and quantity [5]; offshore aquifer to prevent sea water intrusion into groundwater [6,7]; and dam supplementation [2,3]. Moreover, [5] recommended reusing the STMWW effluent into surface water reservoir to increase feed water capacity in municipal drinking water treatment plant.

In contrast to IPWR, direct potable water reuse (DPWR) is defined as highly tertiary-treated wastewater effluent is directly distributed or diluted with the potable water source without passing through the dilution of any surface water or groundwater [2]. Although a community in Windhoek's Goreangab of Namibia practices DPWR [2,8], DPWR is used only at a few places, primarily due to public perception and support.

Natural surface water is less energy intensive than TTMWW water. The trivial cost for tertiary treatment of STMWW is much lower than the cost paid for dam water or desalinated water [9]. Surface water continuously decreases in arid and semi-arid zone through evaporation, and thus creating a potential for TTMWW to replace surface water. In the past several years, the western USA has suffered and is suffering moderate to severe drought. The water level in the rivers of these regions has decreased that subsequently reduce the water level of the reservoirs downstream [2,10]. For example, Lake Mead lost 46% of its water [2,11]. These losses are typically replaced by pumping and desalting brackish groundwater that carries total dissolved solid (TDS) > 1,000 mg/L [12]. Continuous reliance on pumping groundwater becomes unstainable due to the diminished supply (refilling/recharging) and higher demand (pumping).

The membrane technologies (electrodialysis reversal [ED/EDR] and reverse osmosis [RO]) are dominated in treating STMWW into TTMWW for water reuse. Both of these technologies, however, waste about 15–35% (15–20% in ED/EDR [13] and 25–35% in RO) of feed water as brine (RO) or concentrate (EDR) depending on water recovery rate; TDS removed (volume of product water produced) rate over the effective membrane area and current (voltage) applied [14]; equipment and membrane life; chemical dosing; allowable annual capital and operation cost; water chemistry of feed water [15]; and demineralization degree. Waste disposal costs could range from 5 to 33% of desalination cost [16,17]; in the case of inland sites, estimates show that it can be in the order of 15% of the cost of desalination [18]. The cost for concentrate disposal by evaporation pond is 1.18–10.04 $/m^3 [19].

To be sustainable and cost-effective, waste has to be reused as resource [20]. The reuse of waste into resource has to accentuate unharmful to the public health [21,22]. The TTMWW is widely accepted to use as IPWR and unplanned DPWR; we hypothesize that TTMWW can be used as feed in concentrate stream and electrode compartment (EC) in EDR to save the 15–20% of fresh brackish groundwater (sea water), and to improve the EDR desalination. The *objective* of this article is to study the feasibility of reusing TTMWW as feed in concentrate stream and EC of EDR by comparing three case studies—(1) brackish groundwater is fed into dilute, concentrate, and electrode streams, (2) only brackish groundwater (or) in case (3) only sea water is fed into dilute stream, and TTMWW is fed into concentrate stream and EC.

2. Suitability of EDR in water reuse of TTMWW

Even though TTMWW is used as feed in concentrate steam in EDR, TTMWW does not directly contact the dilute product water. There are two flow streams—dilute and concentrate streams—that are separated by ion exchange membrane in EDR. Hydraulic and polarity are reversed at 15–30 min intervals, circling between dilute and concentrate streams to clean out the scaling deposits on membrane surface of concentrate stream. Dilute stream produces as drinking water at effluent after *Escherichia coli* treatment. Concentrate attached in membrane surface of dilute stream from previous cycle of polar reversal is washed out with dilute water for 30 s [23] in each hydraulic stage just after polarity and hydraulic reversal.

2.1. Electrode rinsing water in EDR

Oxygen (O_2) and hydrogen (H^+) ions are produced in anode compartment, and hydrogen (H_2) gas and hydroxyl (OH^-) ions are produced in cathode compartment in the presence of current [16,23]. A small amount of water is required to rinse off these off-gases and OH^- from electrodes to prevent deposits on the surface of electrodes. The 0.5 L/hr of feed water was used to rinse off both cathode and anode ECs that have effective surface area of 0.008 m^2 of each in the ED to treat the brackish groundwater (BGW) with 6,000–36,000 mg/L of TDS in the product flow rate of 3.6–9.6 L/hr with the applied potential 36–40 V [24]. The electrode rinsing water flow rate of 0.2 L/hr was used in 0.008 m^2 of effective area of cathode and

anode electrodes for each for treating 2,120, 3,020, 4,260, and 4,800 mg/L of brackish groundwater in 1.1–5.7 L/hr of dilute flow rate with 25 V applied potential [25].

By using TTMWW as feed in EC, more opportunity to create acid environment in anode (positive electrode) compartment and less scaling potential in cathode (negative electrode) compartment due to less scale causing ions in TTMWW. Oxygen (O_2) and hydrogen (H^+) ions are produced in anode compartment in the present of current from the following equation [23]; H^+ tends to lower the pH of electrode rinsing water and create acid environment.

$$2H_2O = O_2 + 4H^+ + 4e^- \quad E^\circ = 1.229\ \text{V} \tag{1}$$

where E° = the standard potential which refers to the normal hydrogen electrode at the standard state.

Hydrogen (H_2) gas and hydroxyl (OH^-) ions are produced in cathode compartment in the existence of current [23] as the following equation:

$$4H_2O + 4e^- = 2H_2 + 4OH^- \quad E^\circ = -1.656\ \text{V} \tag{2}$$

The OH^- tends to increase the pH of electrode rinsing water and creates alkaline or scaling environment by reacting with Na^+, Ca^{2+}, and Mg^{2+} from feed water. Since TTMWW contains less amounts of Na^+, Ca^{2+}, and Mg^{2+} (Table 1), there is a less chance to react with OH^-, and less opportunity to get scaling in electrodes. If there is any scale occurring in the cathode electrode in the present polar circle, this scale can be reduced in acid environment created by the anode electrode in the next polar circle. The acid environment dose does not have enough capacity to react with all the scales if the feed water contains a lot of scales forming ions and compounds.

2.2. Availability of MWW

Each drop of wastewater may become a drop of water in reuse. Each drop of drinking water may result in a drop of wastewater. A total of 110–150 Mm^3/d of MWW is available in the USA with higher flow rate available in densely populated areas and with a large disparity in effluent discharge rates [26,27]. The EPA reveals that a total of 33,852 National Pollutant Discharge Elimination System permitted discharges were presented in 2007; 17,864 were public owned and operated wastewater treatment plants in the lower 48 states [26,27].

Table 1
Composition of ions in STMWW, brackish groundwater, and sea water

Ions Reference	STMWW (eq/L) [36]	BGW (eq/L) [32]	Sea water (eq/L) [37]
Na^+	0.01060 [36,44]	0.0074	0.4675
Ca^{2+}	0.00334	0.0254	0.0205
Mg^{2+}	0.00322	0.0154	0.1049
K^+	0.00180 [44]	0.0000	0.0102
Sr^{2+}	0.00000	0.0000	0.0002
Cl^-	0.00670	0.0096	0.5443
HCO_3^-	0.00660 [44]	0.0035	0.0017
CO_3^{2-}	0.00000	0.0000	0.0005
SO_4^{2-}	0.00644	0.0323	0.0501
NO_3^-	0.00010	0.0000	0.0000
Fe^{2+} or Fe^{3+}	0.00000	0.0000	0.0000
Mn^{2+}	0.00000	0.0000	0.0000
Br^-	0.00000	0.0000	0.0008
F^-	0.00000	0.0001	0.0000
Sum of positive ion	0.01896	0.0482	0.6033
Sum of negative ion	0.01984	0.0455	0.5975
Average of positive and negative ions	0.01940	0.0468	0.6004
Total dissolved solid (g/L)	0.90340	2.9690	34.8967

BGW = brackish groundwater.
STMWW = secondary-treated municipal wastewater.

3. Methods

3.1. Case study 1: classical EDR desalination

In the classical approach, the same water (i.e. brackish groundwater) is used to feed all streams that include dilute, concentrate, and ECs. Due to the attraction of ions from dilute into concentrate stream along the flow path, ion concentration at the effluent of concentrate stream is always higher than concentration at its influent and feed water as shown in Table 2 and Fig. 1. If the concentrate is chosen to dispose back into surface water or ocean, more ions are added into the original source.

3.2. Case studies 2 and 3: TTMWW as feed in concentrate stream and EC

The TTMWW is proposed to use as feed into concentrate stream and EC only as shown in Table 2 and Fig. 1; however, brackish groundwater (Case study 2) and sea water (Case study 3) are fed into dilute streams. The TTMWW was obtained from STMWW after pretreatment with ultrafiltration (UF) with 73% of water recovery rate. The UF (membrane pore diameter 0.001–0.02 μm [28–30]) with module ZW-10 showed to remove all of the *E. coli* (1.1–1.5 μm wide and 2.0–6.0 μm long) from STMWW [31]. The UF also remove 33.8, 94.6, and 98.6% of COD, turbidity, and total suspended solid, respectively, and permeate from UF is suitable for EDR. As the concentrate from UF contains total coliform, this concentrate has to pass through coliform treatment as shown in Fig. 1. Mass balance diagrams of EDR that use TTMWW as feed in concentrate stream and EC for both brackish groundwater and sea water are shown in Fig. 1 and Table 2.

4. Results and discussion

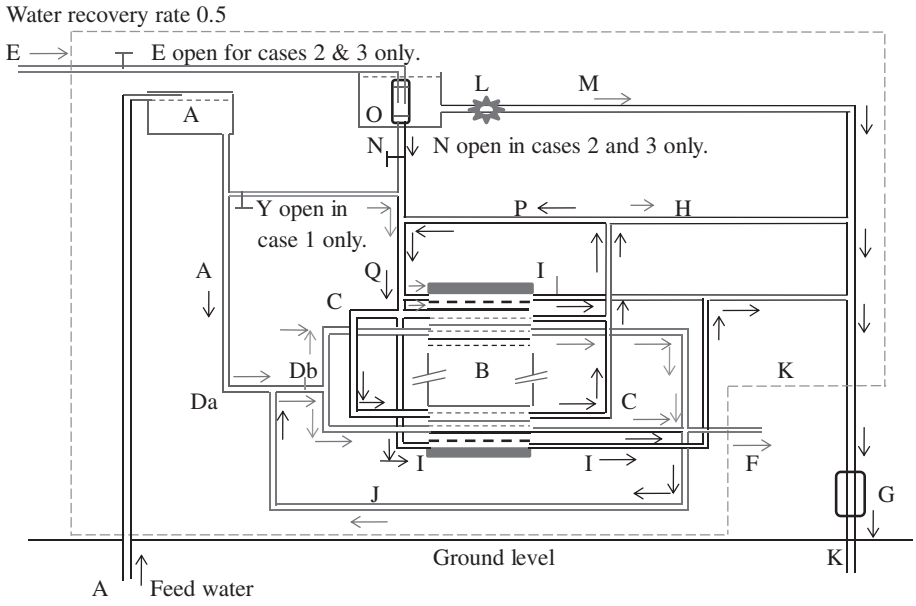
4.1. Results

The results of three case studies are shown in Fig. 1, Tables 2 and 3. Fig. 1 is the flow diagram of EDR. Fig. 1 calculates mass balance of mass flow rate and ion concentration in each point including concentrate stream; ion concentration in concentrate stream requires the $CaSO_4$ saturation level and mean-ion-residence-time in concentrate (MIRTc) to be pinpointed. Table 2 shows flow rate and ion concentration in the key points for the three case studies. Table 3 compares ion concentrations and $CaSO_4$ saturation level in concentrate of EDR for the three case studies.

Table 2
Comparison of flow and concentration in each point (Fig. 1) of three case studies

	A	E	C	Db	F	H	I	J	M	K
(1) Brackish groundwater from [32] *was fed to dilute and concentrate streams and EC*										
Flow (m^3/s)	2.315	NA	1.312	1.312	1.157	1.157	0.000	0.154	NA	1.158
TDS (eq/L)	0.047	NA	0.050	0.045	0.006	0.087	0.047	0.014	NA	0.087
(2) *Brackish groundwater from* [32] *was fed to dilute; TTMWW was fed into concentrate and EC*										
Flow (m^3/s)	1.157	1.312	1.312	1.312	1.157	1.312	0.000	0.154	0.354	1.312
TDS (eq/L)	0.047	0.019	0.019	0.047	0.006	0.053	0.019	0.009	0.019	0.053
(3) *Sea water was fed into dilute*; *TTMWW was fed to concentrate stream and EC*										
Flow (m^3/s)	1.157	1.312	1.312	1.312	1.157	1.312	0.000	0.154	0.354	1.312
TDS (eq/L)	0.600	0.019	0.019	0.600	0.009	0.540	0.019	0.084	0.019	0.540

A: feed water.
C: concentrate stream.
Da, Db: dilute stream.
E: secondary-treated MWW effluent.
F: dilute product water.
H: concentrate from EDR.
I: electrode rinsing water (ERW).
J: off-spec flow.
K: total waste flow from system.
M: concentrate from UF.



A - feed water flow rate
B - electrodialysis reversal
C - concentrate stream
Da, Db - dilute stream
E - secondary treated MWW
F - dilute product water
G - recharge by surface spreading
H - concentrate from EDR
I - electrode rinsing water (ERW)
J - off-spect flow
K - total waste flow from system
L - coliform and virus treatment
M - concentrate from UF
N - tertiray (UF) treated MWW (TTMWW)
O - UF with 73% recovery
P - Concentrate recycle into feed of concentrate
Q - Feed of concentrate stream

Fig. 1. Mass flow diagram of innovative EDR.
Case 1: brackish groundwater is fed in dilute, concentrate, and EC while $E = 0$ and brackish groundwater is supplied from A to Da and Db; to Y and Q. Valve Y is open.
Case 2: TTMWW as feed in concentrate and EC through E, O, N, and Q; brackish groundwater is fed to dilute through A to Da and Db. The valve Y is closed.
Case 3: TTMWW as feed in concentrate and EC through E, O, N, and Q; sea water is fed to dilute through A to Da and Db. The valve Y is closed.

Table 3
Comparison of ions concentration and saturation level in concentrate streams of three case studies

Case studies	1	2	3
Reference	[32]	Calculated values	
K^+ (mol/L)	0.0000	0.0018	0.0070
Na^+ (mol/L)	0.0095	0.0117	0.4098
Ca^{2+} (mol/L)	0.0244	0.0125	0.0102
Mg^{2+} (mol/L)	0.0155	0.0087	0.0519
Cl^- (mol/L)	0.0165	0.0132	0.4815
HCO_3^- (mol/L)	0.0052	0.0076	0.0060
SO_4^{2-} (mol/L)	0.0313	0.0149	0.0257
TDS (mg/L)	5,482	3,406	31,288
$CaSO_4$ saturation (%)	152[a] (147)	33	100
MIRTc (min)	13.4	10.2	7.7

[a]Literature value from [32].
() – calculated value.
MIRTc: mean ions residence time in concentrate.

4.2. Advantages and disadvantages

In the Case study 1, the $CaSO_4$ saturation level in concentrate stream is 152% [32] which is more than the design limit (150% from [23]), and antiscalant is required to add in the concentrate stream to prevent $CaSO_4$ scaling on the surface of membrane. However, in the Case studies 2 and 3, the $CaSO_4$ saturation level in concentrate streams is 33 and 100%, respectively, that are less than the design limit, and antiscalant is not required to add in concentrate stream. Case studies 2 and 3 save the cost of antiscalant and pumping facilities.

In the classical EDR desalination, TDS concentration at the effluent of concentrate stream from the system is always higher than that in the influent (Fig. 1 and Table 2). The concentrate is required to dispose off in evaporation pond, inject into ground, or sea. Disposal in evaporation pond loses water resource into air environment; injection into ground (outfall back into sea) increases the TDS concentration in the original water resource. None of these disposal methods solves the problem at the point of origination and reassign the problem for the future generations.

4.2.1. The system maintains sustainability when it does not destroy the resource for the future generation

This occurs when the TDS concentration wastes from the EDR desalination is less than that is fed into the system; the water flow rate taken from the groundwater (or sea water) is sent back with less TDS concentration. Our proposed EDR desalination meets these criteria and is shown in Fig. 1 and Table 2.

The fresh brackish groundwater (sea water) is saved from reusing and substituting TTMWW as feed in concentrate stream and EC. Based on information available from IDA Desalting Plants Inventory, the capacity of ED/EDR installed plants expanded from 7.5 million L/d in 1955 to more than 750 million L/d in 1992 [23]. Since, there are more than 1,000 ED/EDR plants (with the capacities of $100\,m^3/d$ installed) around the world in 1990 according to the IDA desalting plant Inventory report [33], the fresh brackish groundwater (sea water) saving could reach to $17{,}500\,m^3/d$ with the data from 1990; the saving will increase much more in future due to increasing installation of ED/EDR.

4.3. Chemical saving in using TTMWW as feed in concentrate stream

In classical EDR, due to the same feed water (2,969 mg/L from [32] is used our study in Table 1) is fed into concentrate, dilute, and EC, acid and antiscalant are required to add in concentrate to avoid $CaCO_3$ and $CaSO_4$ scaling in surface of membrane to gain a higher water recovery and a higher TDS removal rates [34,35]; acid is required to add in feed of electrode rinsing water to avoid the scaling in electrodes especially in high potential scaling feed water. All these problems can be minimized when TTMWW is used as feed in concentrate and electrode rinsing water due to the less scaling causing ions and less TDS containing in TTMWW than in brackish groundwater as shown in Table 1. Lower scale causing ion and lower TDS concentrations in feed of concentrate steam subsequently reduce these concentrations in the concentrate stream and disposal outlet as shown in Tables 1 and 2 and Fig. 1.

4.4. Cost reduction

Due to the ample quantity and uniform quality availability of TTMWW, it is not necessary to recirculate the concentrate from effluent into influent of concentrate stream in EDR to gain higher water recover rate by reducing the amount of waste. The MIRTc is low without recirculation [13]. Lower MIRTc (13.4 min in case 1; 10.2 min in case 2; and 7.7 min in case 3 as shown in Table 3) further reduces the scale potential in the surface of membrane due to lower contact time between scale causing compounds and membrane surface. These subsequently reduce the membrane damage and increase the membrane life time. The electrode' life span is based on ionic composition of the source water and the quantity of current supplied to the electrode; higher percent of chlorides containing source water and larger quantity and frequency of current application deteriorate the life span of electrode [23]. The TTMWW carries less chloride (0.0067 eq/L from [36]) than brackish groundwater (0.0096 eq/L from [32]) and sea water (0.5443 eq/L from [37]) as shown in Table 1; TTMWW enhances the life of electrode.

4.5. Less desalting power consumption by substituting TTMWW as feed in concentrate of EDR

In the classical EDR, the same kind of feed water is fed into dilute and concentrate streams. Ions are attracted from dilute into concentrate streams, and the ion concentration in concentrate stream is much higher than that in dilute stream according to water recovery and demineralization rates. The power supply must be high enough to attract the ions against the concentrations difference. Ions are likely to diffuse back from concentrate into dilute stream against the

current direction if concentration ratio between concentrate and dilute streams more than 150 [23]. Therefore, the higher the concentration ratio between concentrate and dilute stream, the higher power required to desalt ions from dilute into concentrate streams. This higher power application also deteriorates the life span of electrode [23].

The desalting power can be saved in the improved EDR where TTMWW is used as feed in concentrate stream due to the lower TDS concentration in feed (TTMWW) of concentrate stream and higher TDS concentration in the feed (brackish ground water or sea water) of dilute stream. Ions from higher TDS concentration (dilute stream of EDR fed with brackish groundwater or sea water) naturally migrate into lower TDS concentration (concentrate stream of EDR fed with TTMWW) across the ion exchange membrane [38]. Due to efficiency loses in the higher ion concentration by current shortcutting, the efficiency can be increased by operating ED/EDR at lower ion concentrations between dilute and concentrate streams by reusing TTMWW.

4.6. Technical challenge

There are three main technical challenges when TTMWW is used as feed in EDR desalination—(1) fouling by colloidal materials; (2) fouling by natural and synthesis organics; and (3) biological growth [33]. Colloids develop an effective negative charge at the surface of their bound water layer when colloids interact with water. The negative charged colloids are attracted by direct current toward the anion exchange membrane. When the colloids (diameter 0.001–1 μm [39–41]) approach the surface of membrane (pore diameter 0.001–0.002 μm [42]) in EDR, the electric field and electrostatic magnetism to ion exchange sites at the surface of membrane are likely to seize the colloidal deposit in place [33]. The periodic polar reversal (hydraulic and direct currents) in EDR drives off the deposit. The silt density index in 5 min ($SDI_{5\,min}$) is the typical parameter used to identify the potential colloidal fouling. If $SDI_{5\,min} < 12$, there is not likely to be colloidal fouling; if $SDI_{5\,min} > 16$, colloidal fouling is likely [32]. If there is serious colloidal fouling in EDR, the membrane stack can be disassembled and washed out the colloidal deposits in membranes and spacers. However, there is a labor cost such as 100 man-hour of supervised unskilled labor is required to clean out a typical 1,135 m^3/d EDR plant [33].

Membranes in EDR are made of ion exchange resins spread in sheet form that are subjected to potential internal resin fouling by organics. Experiments show that molecular weight of internal resin fouling organics ranges from 250 to 700 [33]. Internal fouling is defined as fouling causing substances that have penetrated inside the pore space of membrane. Internal resin reversible fouling in acrylic-based anion exchange membranes can be fully removed by flushing with a 5% sodium chloride salt solution. However, the flushing will not remove irreversible fouling in membranes that have to be replaced. Average annual anion and cation membrane replacements are 11% and 1.7%, respectively [33].

The TTMWW still contains considerable amounts of phosphorus (P), nitrogen (N), and residual organic material. The warm moist environment fastened with spacers and membranes in concentrate steam of EDR desalination is excellent for biological growth. The spacers and surface of membranes with the nutrients from TTMWW in the environment of pH 6–9, are perfect for biofilming of bacteria, fungi, and microalgae.

The biological growth can quickly result in biofilm accumulation on the surface of membranes and spacers; biofilm can lead to biofouling. The organics and TDS in concentrate can be combined to aggravate mineral scaling. Biological growth can be prevented in EDR membrane with the effective residuals of chlorine, chloramines, and chlorine dioxide. The literature data shows that EDR membrane can tolerate chlorine residuals up to 0.5 mg/L, chloramines (total chlorine) residuals up to 1.0 mg/L, and chlorine dioxide exposure of 100,000 mg/L hours without shortening the life of membrane [33].

Pretreatment and chemical adding can be used to control the biological growth. Pretreatment consists of processes that remove the harmful materials upstream of EDR before TTMWW supplied into the concentrate stream. Controlling biological growth in TTMWW reuse by chemical biocides, chlorination (NaOCl adding), and monochloramine (NH_2Cl) is widely reviewed everywhere [27] and will not recited in here. Chemical treatment, however, relates to cost and secondary by-product uncertainty in the future; one way to control biological growth in concentrate is to operate EDR in low bio-residence time without recirculation of concentrate into feed of concentrate stream. The *EDR membrane* has demonstrated the ability to *handle* continuous microfiltration pretreated STMWW effluent to produce high quality of treated-water pilot plant studies [4] and field scale in the Canary Islands [43].

5. Conclusion

In the analysis, EDR desalination is improved by reusing TTMWW as feed in concentrate and electrode rinsing compartments. The TTMWW is technically

feasible as feed in concentrate and ECs. Reusing TTMWW is more sustainable and cost-effective due to less scaling causing ions and TDS are in TTMWW. At least, 17,500 m^3/d of freshwater could be saved with data from 1990; the saving increases in 2011 much more in future due to increasing installation of ED/EDR. Based on this proof-of-concept finding, the lab scale experiments will be conducted in the near future to evaluate the impact of public acceptance from water saving by reusing TTMWW. Technical advance is important, however, public acceptance factor is also critical in reusing TTMWW as feed in concentrate and electrode streams.

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