Mild desalination demo pilot: New normalization approach to effectively evaluate electrodialysis reversal technology

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ABSTRACT

Key performance indicators for characterization of nanofiltration performance are well developed, similar key performance indicators for electrodialysis reversal are however underdeveloped. Under the E4Water project Dow Benelux BV and Evides Industriewater BV operate a pilot facility to compare both technologies for their application to mildly desalinate a variety of brackish water streams. Normalized pressure drop, normalized current efficiency and normalized membrane resistance proved to be a useful tool to interpret process performance and to initiate a cleaning procedure if required. The availability of these normalized key performance indicators enables optimization and process monitoring and control of electrodialysis reversal independent of the continuously changing conditions of the feed water.

1. Introduction

Six large chemical companies work together in a European partnership to set a new paradigm in water management in the chemical industry. One of the case studies comprises a demonstration pilot plant at the Evides water treatment plant for Dow Terneuzen (the Netherlands), where Dow aims to maximize water reuse in its processes. As the region is scarce of fresh water sources [1], alternative sources of water are evaluated on their applicability to furnish reusable water at affordable cost for industry or other sectors. At present various streams, originating from process industry, rural areas, and municipalities, are discharged to the river Scheldt. These streams have in common that their salinity content is too high for direct usage. Therefore the salt content needs to be reduced – these streams require so-called "mild desalination". The degree of desalination will vary, depending on the final application of the produced water. Hence, technologies have been selected for the pilot facility, which can effectively remove the majority but not all of the salinity present in raw water streams, aiming at a final salinity characterized by an electrical conductivity of less than 1 mS/cm, suitable for reuse in industry or agriculture. The selected mild desalination technologies are nanofiltration (NF) and electrodialysis reversal (EDR).

Although both technologies use membranes to separate salt ions from water, their theories and working principles are different. NF is a pressure driven process in which a feed solution is forced through a semi-permeable membrane while partly retaining ions and other components. The pressure difference (ΔP) over the membrane is needed to overcome the osmotic pressure created by the difference in salt concentration at both sides of the membrane. As NF membranes are less tight than reverse osmosis (RO) membranes a certain fraction of the salt ions also pass the membrane. Hence slightly saline water is produced, as well as a concentrate comprising more salt than the product. The EDR process uses an electrical potential gradient across a stack of alternating cation and anion exchange membranes. Cation exchange membranes (CEMs) have fixed negatively charged groups thus allowing transport of cations, while anion exchange membranes (AEMs) have fixed positively charged groups thus allowing the transport of anions. Movement of cations and anions are opposite of each other within an electrical potential gradient ($\Delta \psi$). Placing cation and anion exchange membranes between this potential

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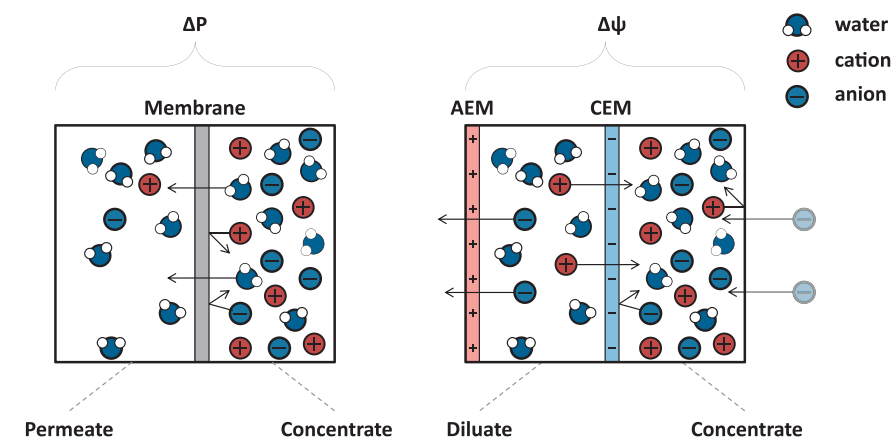 E-mail address: ckgroot@dow.com (N. Groot).

Fig. 1. Schematic representation of the desalination principle of nanofiltration (left) and electrodialysis (right).

gradient creates compartments which are depleted and enriched of ions, i.e. diluate and concentrate compartments respectively (Fig. 1). Within EDR one cell pair consists of a CEM and AEM and diluate and concentrate compartment, with an electrodialysis stack being capable of having up to 600 cell pairs.

The demonstrator at Dow/Evides comprises of a robust pre-treatment followed by parallel EDR and NF pilots and has a capacity of approximately 40 m^3/day mild desalinated product. NF and EDR are used to treat three different water streams (cooling tower blow down, collected rain water, and wastewater treatment effluent). The demonstrator was installed to run for a thirty months period with the objective to evaluate the two mild desalination technologies (with their required pre-treatment) and translate these results to a full scale concept. Over such a long period a wide variety of tests are performed under continuously changing conditions. It is therefore of utmost importance to evaluate results on an independent basis, in other words using performance indicators, which can be applied irrespectively of varying circumstances. While pressure driven membrane processes, like RO and NF, have well established normalized key performance indicators (KPIs), similar KPIs are yet undeveloped for EDR. Previous pilot work comparing EDR and RO makes use of normalized product flow as a performance parameter for RO [2,3] and total dissolved solids (TDS) or electrical conductivity (EC) in the product water are used as a performance parameter for EDR without normalization towards feed conditions. In both referred cases fouling/scaling occurred which was cleaned periodically [2] or triggered by an increased EDR product conductivity [3]. Other parameters commonly used to determine EDR performance are stack resistance [4,5], current efficiency [4,5], specific energy consumption [4–6], potential drop across an ion exchange membrane [6] and individual removal efficiencies [5,7,8]. The effect of temperature and feed salinity is acknowledged but not formally corrected for [4,5]. Monitoring the potential drop across an ion exchange membrane (or even better the membrane resistance) is an approach useful for research purposes but less practical for process monitoring and control in a full-scale plant. On the other hand, mathematical description of the EDR process has reached a high level of maturation. Modeling of EDR is currently used for process simulation and design rather than for process monitoring & control [9–12], nevertheless the relations between feed parameters such as temperature and salinity and process parameters e.g. solute recovery, current efficiency and specific energy consumption are well known. This enables calculation of for example the membrane resistance and data normalization to acquire normalized key performance indicators (KPIs) similar to the KPIs used in pressure driven membrane filtration. Typical KPIs used in NF/RO processes are normalized pressure drop (NPD), normalized salt

passage (NSP), and mass transfer coefficient (MTC). These KPIs can be used for process monitoring and control due to their clear relation with either fouling phenomena or membrane integrity [9]. In this article the development and application of equivalent KPIs for EDR is presented. A theoretical background is given in the materials and methods section to provide the basis of relevance for the KPIs developed covering the critical aspects of the EDR process. Applicability of the developed KPIs is further illustrated with the results obtained in the pilot facility.
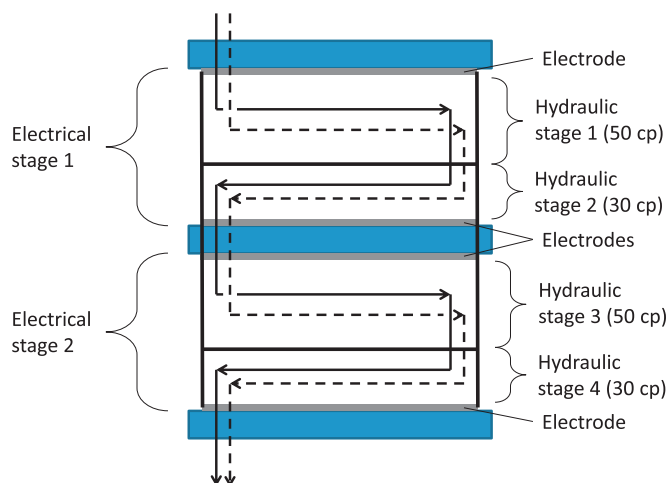
2. Materials and methods

2.1. Demonstrator setup and water sources

The pilot facility is located in Terneuzen, Netherlands. The pilot treatment train comprises of a robust pre-treatment including a flocculator, lamella separator and ultrafiltration module followed by paralleled NF and EDR pilots. Capacity of the plant is approximately 40 m^3/day mild desalinated product. The NF pilot consists of two arrays: a first array of two pressure vessels with each two modules and a second array of one pressure vessel with two modules. The concentrate is partly recycled to be able to achieve a high recovery. The modules used in the pilot are Dow Filmtec NF 90-4040. The EDR stack used in the pilot is the Aquamite 3 AQ 3-1-50/35 by General Electric. The installation is alternately supplied with three different water sources: 1) Cooling tower blow down water (CTBD) consisting of filtered surface water 4–5 times concentrated in a natural draft counter current cooling tower at a full-scale power plant located next to the Dow premises in Terneuzen, the Netherlands. Cooling tower blow down water has a typical conductivity ranging from 2.3 to 3.5 mS/cm. CTBD water contains various chemicals including sulfuric acid used for pH adjustment, phosphates as corrosion inhibitor, scale inhibitors, dispersants and biocides to prevent microbiological growth; 2) Spuikom-water which is collected rain water from the Dow Terneuzen production site and has a typical conductivity of 1.5–3.0 mS/cm; 3) Wastewater treatment effluent which has a typical conductivity of 1.0–1.5 mS/cm [13].

2.2. EDR pilot

The Aquamite 3 AQ 3-1-50/35 (General Electric) EDR stack has a total of 4 hydraulic stages divided over 2 electrical stages consisting both of 50 and 35 cell pairs (cp), which is schematically represented in Fig. 2. Temperatures, pressures and conductivity of EDR feed, concentrate and diluate are measured and recorded by on-line instruments.

Fig. 2. Schematic representation of the EDR stack flow (GE Aquamite 3 AQ 3-1-50/35).

Fig. 3. Picture of EDR pilot set−up (above) and schematic drawing of pilot set-up (below).

In the EDR pilot, desalination starts by applying a voltage difference between the outer electrodes and the center electrode. The EDR desalinates the feed-water continuously (once-trough), while the concentrate stream is recirculated over the EDR stack (feed and bleed). Part of the feed water is fed to the concentrate stream, so-called concentrate make-up and part of the concentrated water stream is discharged. The applied voltage of the different electrical stages can be set separately, tuned to the water quality and the desired degree of 'mild desalination'. The concentrate make-up is adjustable and determines together with the feed water stream the EDR recovery (Fig. 3).

Polarity reversal is achieved by changing the polarity of the electrodes and simultaneous changing of concentrate and diluate flows through the EDR stack. This mode of operation cleans the membrane by releasing colloidal particulates from the ion exchange membranes during polarity reversal. More information of EDR fouling can be found in corresponding reference [14]. Alternating the polarity of the electrodes results in a temporary off-

Table 1
Specifications and used settings for EDR with CTBD water.

Parameters	Unit	Setting
Model	[–]	General Electric AQ 3-1-4-50// 35BAS 50 HZ
Stack type	[–]	Mark IV-2,2″
Hydraulic stages	[–]	4
Electrical stages	[–]	2
Fresh water feed	[m³/h]	2.3
Product flow	[m³/h]	1.6
Concentrate blow down	[m³/h]	0.7
Voltages	[V]	49/39
Recovery	[%]	70
Antiscalant Genesys LF dosage to concentrate	[mg/l]	52
HCl dosage to electrode	[mg/l]	99.6

spec diluate-product production, caused by mixing when switching from concentrate and diluate. Specifications and applied settings of the EDR pilot are shown in Table 1.
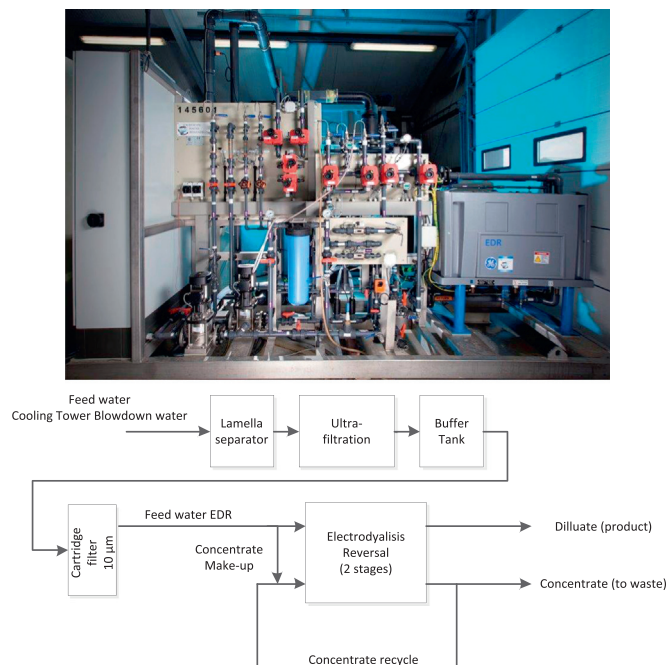
Despite polarity reversal fouling of the EDR stack by mineral scale, biological matter, particles and (insoluble) organic components might occur. Deposits can build up in the spacer filled channel and on the membrane surface during operation resulting in loss in process performance such as reduced efficiency in terms of salt removal. When fouling is too severe, a complete recovery of the initial process performance might be too difficult. Therefore a preventive Cleaning-In-Place (CIP) should be performed in due time to assure optimal process performance. During the CIP process the normal desalination operation of the EDR unit is intermitted. The cleaning chemicals are pumped into the EDR system and are recirculated over the stack. The applied CIP procedure consists of two steps; 1) salt CIP (5 wt% NaCl+NaOH to pH 8–10.5) to remove organic foulant and 2) acid CIP (5 wt% HCl, pH < 1.2) to remove scale formation [15]. This procedure is repeated three times during a complete CIP including 0.5 h soaking times between steps. Prior to start-up and in between CIPs the EDR is flushed with demineralized water to remove remaining traces of chemical solutions.

2.3. Derivation of KPIs

KPIs are typically used in pressure driven membrane filtration to normalize process data while compensating for differences in ambient variation such as feed temperature and salt content. Normalization of performance indicators allows comparison of the actual performance of a process to a given reference performance while the influences of operating parameters and change in the conditions of the feed water are taken into account. Normalization of membrane performance data is used in literature in order to monitor process performance independent of varying conditions [16–20]. Monitoring of KPIs allows early identification of potential problems (e.g. scaling or fouling) and can be used to set trigger values for cleaning. Typical KPIs used in NF/RO processes are NPD, NSP and MTC. These KPIs are first described in the following paragraphs to illustrate the principle and similar KPIs for EDR are subsequently derived.

2.3.1. NF
2.3.1.1. Normalized pressure drop. Pressure drop across the feed spacer of the membrane module is a method to describe the fouling in the feed spacer. Normalization for flow and temperature results in the normalized pressure drop (NPD) [20,21]:

$$NPD = \Delta P_{ACT} \cdot Q_{CF\Delta P} \cdot T_{CF\Delta P} = (P_{FEED} - P_{CONCENTRATE})$$

$$\cdot \left(\frac{(Q_{FEED,REF} + Q_{CONCENTRATE,REF})/2}{(Q_{FEED} + Q_{CONCENTRATE})/2} \right)^{1.6} \cdot \left(\frac{\eta_{T,REF}}{\eta_{T,ACT}} \right)^{0.4} \tag{1}$$

Where NPD is the normalized pressure drop, ΔP_{ACT} the actual pressure difference, $Q_{CF,\Delta P}$ the flow correction factor, $T_{CF,\Delta P}$ the temperature correction factor, P_{FEED} the feed pressure, $P_{CONCENTRATE}$ the concentrate pressure, $Q_{FEED,REF}$ the reference feed flow rate, $Q_{CONCENTRATE,REF}$ the reference concentrate flow rate, Q_{FEED} the feed flow rate, $Q_{CONCENTRATE}$ the concentrate flow rate, $\eta_{T,REF}$ the viscosity at reference temperature and $\eta_{T,ACT}$ the viscosity at actual water temperature.

2.3.1.2. Mass transfer coefficient.
The mass transfer coefficient (MTC), describes the permeability of the membrane and is inversely proportional to the amount of pressure needed to press water through the membrane [16,20,22]. A high MTC indicates a better permeability resulting in a low feed water pressure.

$$MTC = \frac{Q_{PERMEATE} \cdot T_{CFKW}}{A_{mem} \cdot NDP \cdot 3600}$$

$$= \frac{Q_{PERMEATE} \cdot e^{\left(3754 \cdot \left(\left(\frac{1}{T+273.15} \right) - \left(\frac{1}{25+273.15} \right) \right) \right)}}{A_{mem} \left(\left(\frac{P_{FEED} + P_{CONCENTRATE}}{2} \right) - \left(\frac{\Pi_{FEED} + \Pi_{CONCENTRATE}}{2} - \Pi_{PERMEATE} \right) \right)} \tag{2}$$

Where MTC is the mass transfer coefficient, NDP the net driving pressure, T_{CFKW} the temperature correction factor, $Q_{PERMEATE}$ the flow rate permeate, A_{MEM} the membrane surface area, T the temperature, P_{FEED} the feed pressure, $P_{CONCENTRATE}$ the concentrate pressure, Π_{FEED} the osmotic pressure feed, $\Pi_{CONCENTRATE}$ the osmotic pressure concentrate and $\Pi_{PERMEATE}$ the osmotic pressure permeate.

2.3.1.3. Normalized salt passage.
The normalized salt passage (NSP) is used to describe the effectiveness for salt being retained by the membrane and is normalized for temperature and permeate flow rate [16,20,21].

$$NSP = SP_{ACT} \cdot \frac{Q_{PERMEATE}}{Q_{PERMEATE,REF}} \cdot T_{CFKW} = \frac{C_{PERMEATE}}{C_{FEED} \cdot (LN((1/(1-Y))/Y))}$$

$$\cdot \frac{Q_{PERMEATE}}{Q_{PERMEAAT,REF}} \cdot e^{\left(3754 \cdot \left(\left(\frac{1}{T+273.15} \right) - \left(\frac{1}{25+273.15} \right) \right) \right)} \tag{3}$$

where NSP is the normalized salt passage, SP_{ACT} is the actual salt passage, $Q_{PERMEATE,ACT}$ the actual permeate flow, $Q_{PERMEATE,REF}$ the reference permeate flow, T_{CFKW} the temperature correction factor, $C_{PERMEATE}$ the total dissolved solids in the permeate, C_{FEED} the total dissolved solids in the feed, Y the recovery and T the Temperature.

2.3.1.4. KPI analysis.
The Key Performance Indicators (KPIs) are used in the operation of the NF pilot to identify (i) spacer fouling with the NPD, (ii) salt leakage with NSP and (iii) membrane fouling with MTC (Table 2).

Increase or decrease with 10–15% of the KPI [20,23], is normally used for process control to determine the need of cleaning in NF/RO systems. The cleaning limit for the NPD equals a 15% increase compared to results from design software of the membrane supplier.

2.3.2. EDR
2.3.2.1. Normalized pressure drop.
The pressure drop along the flow path length in an electrodialysis stack can be used as a KPI for spacer fouling similar to NPD in NF and depends typically on spacer type, feed composition, flow rate and temperature. CIP can be initiated based on the pressure drop development across the stack e.g. upon increase by 15% from the conditions at start-up. Pressure drop of laminar flow through a tube is given by Poiseuille's law:

$$\Delta P = \frac{8 \cdot \eta \cdot L \cdot Q}{\pi \cdot (d/2)^4} \tag{4}$$

where ΔP equals pressure drop, η equals feed viscosity, L equals length of the channel, Q equals flow rate and d equals hydraulic diameter. The pressure drop has to be normalized for temperature and flow rate, which then leads to:

$$\Delta P_0 = \Delta P \cdot \frac{\eta(T_0) \cdot Q_0}{\eta(T) \cdot Q}$$

$$\eta(T) = 2.414 \cdot 10^{-5} \cdot 10^{247.8/(T-140)} \tag{5}$$

A more precise empiric relation for viscosity as function of concentration and temperature should be used [9] when larger variations in feed salt concentration are present. A further appropriation of the normalization for deviations caused by e.g. non-laminar flow and spacer effects is made by calibration of the equation using a tuning parameter (a), to be obtained specifically for the EDR system used.

$$\Delta P_0 = \left(\Delta P \cdot \frac{10^{247.8/(T_0 - 140)}}{10^{247.8/(T-140)}} \right)^a \cdot \frac{Q_0}{Q} \tag{6}$$

For the EDR pilot used in this work T_0 is set at 25 °C and Q_0 at 1.7 m³/h, the tuning parameter for the EDR stack in the pilot is 0.42 and was determined during the start of the pilot trials. Finally the following formula for normalized pressure drop (NPD) is obtained:

$$NPD = \Delta P \cdot \left(\frac{36.88645}{10^{247.8/(T-140)}} \right)^{0.42} \cdot \frac{1.7}{Q} \tag{7}$$

2.3.2.2. Normalized current efficiency.
Current efficiency (CE) is a measure of how effective ions are transported across the ion exchange membranes for a given electrical current. CE takes into account all the undesirable phenomena occurring in the EDR stack, such as the non-ideal permselectivity of membranes or physical leakage [24]. CE can be normalized into a KPI similar to the NSP in NF, giving an indication of membrane integrity. CE is expressed as the amount of actual ion transport to the theoretical amount, based on Faraday's law and given by the following equation for n

Table 2
KPI analysis NF including cleaning limits.

KPI	Unit	Cause	Consequence	Cleaning limit
Normalized pressure drop	bar	Fouling of feed-spacer	Increased energy consumption and risk of membrane module damage	> 1.6 Maximum given by supplier 2.1 [19]
Normalized salt passage	%	Damage of membrane/scaling	Reduced salt rejection	Used for membrane replacement not used as cleaning limit
Mass transfer coefficient	m/s Pa	Scaling/fouling at the membrane	Increased energy consumption	< 1.9

salts in solution:

$$CE = \frac{F \cdot \left(Q_{feed} \sum_{i=1}^{n} |z_i| \cdot C_i - Q_{diluate} \sum_{i=1}^{n} |z_i| \cdot C_i \right)}{2 \cdot I \cdot N_{cellpairs}} \tag{8}$$

where Q_{feed} and $Q_{diluate}$ are the volumetric flows entering and exiting the diluate compartment respectively; z_i the charge of ion i; and C_i the concentration ion i. For practical purposes Q_{feed} and $Q_{diluate}$ are assumed to be equal, thus neglecting solute transport through the ion exchange membranes, the concentration is derived from online electrical conductivity data (EC), through a correction factor (cf). Temperature correction to 25 °C is implicitly taken into account by the on-line EC measurement. This methodology however does not take into account changes in relative ion composition necessitating regular sampling and chemical analysis for validation of cf.

$$CE = \frac{F \cdot Q \cdot \left(EC_{feed} - EC_{diluate} \right) \cdot cf}{2 \cdot I \cdot N_{cellpairs}} \tag{9}$$

Compensation of temperature, flow rate and the electrical conductivity resulted in a CE in the range of 75–90% and was found to be linear dependent on the electrical conductivity of the diluate and type of diluate during the start of the pilot trials. For example, the average current efficiency of 'spuikom water' is described by the following correlation between 0.075 and 0.15 mS/cm:

$$CE_0 = f(EC) = -32.511 \cdot EC + 84.897 \tag{10}$$

The current efficiency is made relative to the initial current efficiency thus obtaining the normalized current efficiency (NCE):

$$NCE = \frac{CE}{CE_0} \tag{11}$$

2.3.2.3. Normalized membrane resistance. Membrane resistance in EDR relates the ion flux through the membrane to the applied potential difference over the membrane and can be normalized into a KPI similar to MTC in NF which relates mass flux through the membrane to the applied pressure difference. Organic matter can interact with the ion selective membranes used in EDR causing fouling. Inorganic fouling, such as for example scale formation, can be internal or external, but will in both cases reduce the transport of ions through the membranes and result in an increase in the membrane resistance [25,26]. Membrane resistances R_{CEM} and R_{AEM} are a component of the total stack resistance indicated in Eq. (5). The overall resistance of the electrodialysis stack is considered to be the sum of the resistance of the cation exchange membrane (R_{CEM}), anion exchange membrane (R_{AEM}), bulk solution in the concentrate compartment (R_C), bulk solution in the diluate compartment (R_D), electrode rinsing solution (R_{ERS}), the boundary layers of the diluate near the CEM and AEM ($R_{fCEM,D}$ and $R_{fAEM,D}$) and the boundary layers of the concentrate near the CEM and AEM ($R_{fCEM,C}$ and $R_{fAEM,C}$).

$$R = \left(R_D + R_{fCEM,D} + R_{CEM} + R_{fCEM,C} + R_C + R_{fCEM,C} + R_{AEM} + R_{fAEM,D} \right) \cdot N_{cellpairs} + 2 \cdot R_{ERS} \tag{12}$$

The overall potential difference across an electrodialysis stack can be written as [11,12]:

$$E = E_{el} - \left(E_j + E_{Don} \right) \cdot N_{cellpairs} + R \cdot I \tag{13}$$

where E is the overall potential difference; E_{el} the thermodynamic potentials and overpotentials of the electrodes; E_j is the overall junction potential difference across the boundary layers in one cell pair; E_{Don} is the Donnan potential difference across the membranes in one cell pair; $N_{cell pairs}$ the overall number of cell pairs; R the overall resistance of the membranes, bulk solutions, boundary layers and electrode rinsing solutions; and I is the electrical current supplied to the electrodialysis stack. Rearranging (Eqs. (12) and 13) results in the following equation which relates the membrane resistance to the electrical current (I) and applied potential difference (E):

$$R_{CEM} + R_{AEM} = \left(\frac{E - E_{el} + \left(E_j + E_{Don} \right) \cdot N_{cellpairs}}{I} - 2 \cdot R_{ERS} \right) \cdot \frac{1}{N_{cellpairs}} - \ldots \\ \left(R_D + R_{fCEM,D} + R_{fCEM,C} + R_C + R_{fCEM,C} + R_{fAEM,D} \right) \tag{14}$$

The influence of current on the thermodynamic potentials and overpotentials of the electrodes (E_{el}) is assumed to be neglectable compared to the applied potential difference and fixed at 1.8 V, thus assuming an overpotential of 0.25 V for the cathode and anode. The junction potentials across the boundary layers (E_j) are neglected [12], while the Donnan potential difference across the membranes is calculated for ideal solutions and ideal ion exchange membranes, i.e. $t^- = 1$ and $t^+ = 1$ for AEM and CEM, respectively. Eq. (6) is then simplified to:

$$E_{Don} = \frac{R \cdot T}{F} \cdot \ln \frac{C_{fIEM,1}}{C_{fIEM,2}} \tag{15}$$

where $C_{f,1}$ and $C_{f,2}$ denote the local concentrations facing side 1 and side 2 of the ion exchange membrane (IEM). The local concentrations can be estimated using Fick's law of diffusion:

$$J = -D \cdot \frac{\partial C}{\partial x} = -\frac{D}{\delta} \cdot \left(C - C_{fIEM} \right) \tag{16}$$

where J is the ionic flux, D is the diffusion coefficient, δ the boundary layer thickness, C the concentration in the bulk and C_{fIEM} the concentration at the IEM. The ionic flux of a monovalent ion is related to the electrical current by the following equation under the assumption of 100% CE.

$$J = \frac{I}{A \cdot F} \tag{17}$$

Here A is the membrane surface area. The thickness of the boundary layer can be calculated using the Sherwood number which is related to channel characteristics and hydrodynamic

Table 3
KPI analysis EDR including cleaning limits.

KPI	Unit	Cause	Consequence	Cleaning limit
Normalized pressure drop	(bar)	Fouling of spacer	Increased energy consumption and risk of EDR stack damage	> 2.1 [29] – 2.3 Maximum given by supplier 3.4 [30]
Normalized current efficiency	(–)	Scaling/fouling/leakages at the membranes	Reduced salt rejection Increased energy consumption Risk of membrane damage	< 0.9
Normalized membrane resistance	(–)	Scaling/fouling at membranes		> 1.1–1.5

conditions by the following correlation:

$$Sh = a \cdot Re^b \cdot Sc^c \tag{18}$$

where a, b and c are constants depending on channel characteristics, Re is Reynolds number and Sc is the Schmidt number. The constants a, b and c for the EDR stack used in the pilot are equal to 0.29, 0.61 and 0.33, respectively.

Combining Eqs. 15 to 18 allows calculation of the Donnan potentials for AEM and CEM using solely electrical current and bulk concentration:

$$E_{Don} = \frac{R \cdot T}{F} \cdot \ln \frac{C_D - \frac{I \cdot h_D}{A \cdot F \cdot \left(0.29 \cdot Re^{0.61} \cdot Sc^{0.33}\right) \cdot D}}{C_C + \frac{I \cdot h_C}{A \cdot F \cdot \left(0.29 \cdot Re^{0.61} \cdot Sc^{0.33}\right) \cdot D}} \tag{19}$$

Here h denotes the channel height.

The resistances of the bulk solutions (R_{ERS}, R_D and R_C) can be calculated based on the electrical conductivity (κ) by:

$$R = \frac{h}{\sigma \cdot \kappa} \tag{20}$$

where σ denotes the spacer shadow effect, which accounts for the fraction of the compartment (i.e. the spacer), which is accessible for current (estimated at 0.8).

The resistance of boundary layers is dependent on the concentration gradient and thus the electrical current (Eq. 21). The electrical conductivity is proportional to the concentration, so the resistance of the boundary layers is calculated by integration of the electrical conductivity over the boundary layer thickness and assuming a linear concentration gradient:

$$R_f = \frac{1}{\sigma} \cdot \int_0^\delta \frac{1}{\kappa} = \frac{1}{\sigma} \cdot \left(\frac{1}{\kappa - \kappa_{fIEM}}\right) \cdot \ln\left(\frac{\kappa}{\kappa_{fIEM}}\right) \tag{21}$$

Here κ_{fIEM} is the electrical conductivity of the solution at the IEM, which is calculated using the local concentration (C_{fIEM}) and converted into electrical conductivity.

Using (Eqs. (19), (20) and 21) allows enables Eq. (14) to be rewritten thus making the membrane resistance only dependent on parameters available by online measurement:

$$R_{CEM} + R_{AEM} = \frac{E - 1.8}{I \cdot N_{cellpairs}} + \frac{2 \cdot R \cdot T}{I \cdot F} \cdot \ln\left(\frac{EC_D \cdot cf - \frac{I \cdot h_D}{A \cdot F \cdot \left(0.29 \cdot Re^{0.61} \cdot Sc^{0.33}\right)}}{EC_C \cdot cf + \frac{I \cdot h_C}{A \cdot F \cdot \left(0.29 \cdot Re^{0.61} \cdot Sc^{0.33}\right)}}\right)$$

$$- \frac{2 \cdot h_{ERS}}{N_{cellpairs} \cdot \sigma \cdot EC_{ERS}} - \dots$$

$$\left(\frac{h_D}{\sigma \cdot EC_D} + \frac{h_C}{\sigma \cdot EC_C} + \frac{2}{\frac{\sigma \cdot I \cdot h}{cf \cdot A \cdot F \cdot \left(0.29 \cdot Re^{0.61} \cdot Sc^{0.33}\right)}}\right)$$

$$\cdot \left(\ln\left(\frac{EC_D}{EC_C}\right) + \ln\left(\frac{EC_C + \frac{I \cdot h_C}{cf \cdot A \cdot F \cdot \left(0.29 \cdot Re^{0.61} \cdot Sc^{0.33}\right)}}{EC_D - \frac{I \cdot h_C}{cf \cdot A \cdot F \cdot \left(0.29 \cdot Re^{0.61} \cdot Sc^{0.33}\right)}}\right)\right)$$

The used EDR stack consists of four hydraulic stages and two electrical stages. The electrical conductivity of each stream exiting a stage in the EDR stack is calculated using mass balance principles. Calculation of the membrane resistances is feasible by using electrical current, applied potential drop and electrical conductivity of each electrical stage. The logarithmic mean concentration over the flow channels is used to account for concentration differences between channel inlet and outlet.

$$EC = \frac{EC_{in} - EC_{out}}{\ln\left(\frac{EC_{in}}{EC_{out}}\right)} \tag{22}$$

The influence of temperature on the resistance of the ion exchange membranes was not assessed, normalization towards a temperature is therefore omitted. Compensation for solute temperature, flow rate and the electrical conductivity resulted in an area resistance for CEM and AEM of 100–400 Ω cm². Lower salt concentrations result in increased membrane area resistance [27], which might explain that the obtained area resistance is one order of magnitude higher than given by supplier [28]. The dependency of membrane resistance to the electrical conductivity of the feed flow rate was determined during the start of the pilot trials and could be related to each other by the following equation (for the first stage):

$$R_{CEM,0} + R_{AEM,0} = 0.0184 \cdot EC^{-0.712} \tag{23}$$

Finally membrane resistance is made relative to the initial membrane resistance for ease of use resulting in the normalized membrane resistance (NMR):

$$NMR = \frac{R_{CEM} + R_{AEM}}{0.0184 \cdot EC^{-0.712}} \tag{24}$$

2.3.2.4. KPI analysis. The Key Performance Indicators (KPIs) are used in the operation of the pilot to identify (i) spacer fouling with the NPD, (ii) stack leakage with NCE and (iii) membrane fouling with NMR and NCE.

Limits for performing a CIP of the EDR stack were established (Table 3) and include a relative increase or decrease of the KPI by 10–15% [23] compared to start-up, which is analogous to the membrane cleaning procedure of a NF/RO system. Based on the pilot results the cleaning limits of the normalized pressure drop and membrane resistance were increased due to practical experience obtained during our research.
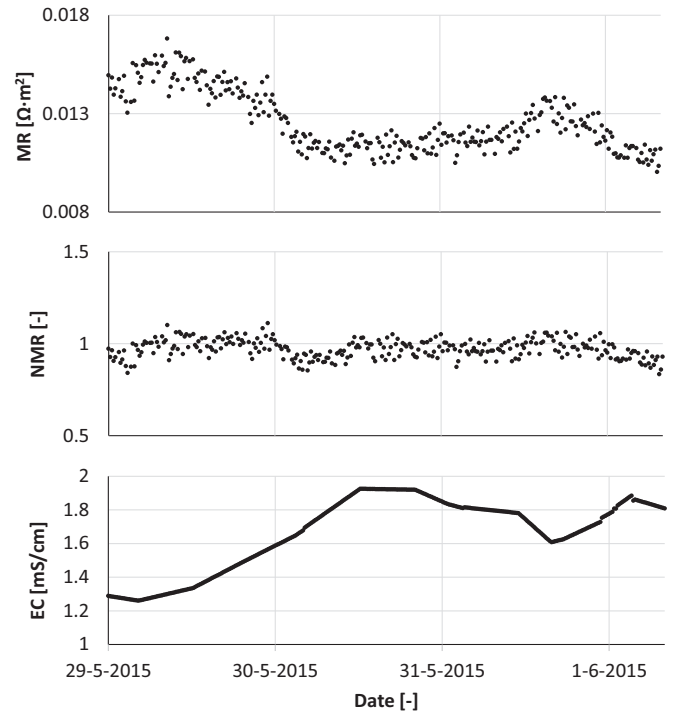
Fig. 4. Membrane resistance (top), normalized membrane resistance (middle) and electrical conductivity of the feed water during four days of EDR operation on Spuikom water.

3. Results and discussion

3.1. Validation of key performance indicators

The membrane resistance (R_{CEM} and R_{AEM}) in the first electrical stage of the EDR stack and pressure drop during four days of normal operation using Spuikom water (collected rain water) were shown in respectively Figs. 4 and 5 together with NMR and NPD to illustrate the effect of normalization. Only short periods of the 30 month run, showing the most variation in the defined KPIs, were
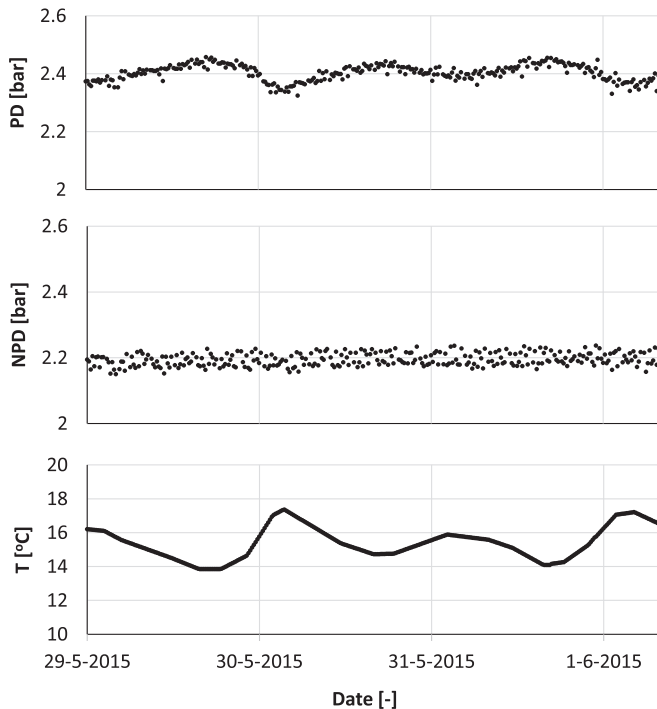

Fig. 5. Pressure drop (top) and normalized pressure drop (bottom), time period of four days for the EDR treatment of Spuikom water.

presented in these and following figures for reasons of clarity. The variations in electrical conductivity (1.3–1.9 mS cm^{-1}) and temperature (13–18 °C) of Spuikom water were the result of rainfall and influence of ambient temperature.

Variations of 50% in the calculated membrane resistance emphasize the need for normalization as can be seen in Fig. 4. The dependency of the membrane resistance on the salt concentration was non-linear resulting in higher resistivity at lower concentrations [27] which was also indicated by the presented data. The required normalization was therefore performed using the obtained correlation (Eq. (23)) at the beginning of pilot operation (approx. one year before the data presented in Fig. 4). When normalizing for feed water conductivity, the normalized membrane resistance results in a constant value indicating no membrane fouling and an NMR after one year of operation similar to initial value, i.e. NMR ≈ 1 (Fig. 4, middle graph).

The pressure drop across the EDR stack was influenced by feed water temperature as shown in Fig. 5. The pressure drop increases at decreasing temperature due to increase of viscosity of the feed water. A varying temperature results in a trend, which was difficult to interpret from a process control point of view. Normalization eliminates the effect of temperature resulting in a stable normalized pressure drop trend which indicates no fouling of the spacer.

3.2. Key performance indicators for process monitoring and control

Results of Cooling Tower Blowdown water desalination with varying feed water conductivity of 2.8–3.2 mS/cm and product water conductivity of 0.3–0.7 mS/cm, were used to illustrate the effectiveness of the developed KPIs on process control, i.e. trigger to initiate various CIP treatments. NCE, NMR and NPD are shown in Fig. 6. The gaps in data represent the down time of the EDR system due to CIP, maintenance, unavailability of feed water or malfunctions. The broad band of the NPD signal is caused by the reversal operation.

During the two weeks of EDR operation an increase in NPD is noticed thereby exceeding the maximum threshold, CIP was therefore executed resulting in a decrease of NPD. A second CIP was executed because the NPD still exceeded, although in lesser extent, the threshold value. The second CIP restored the NPD to
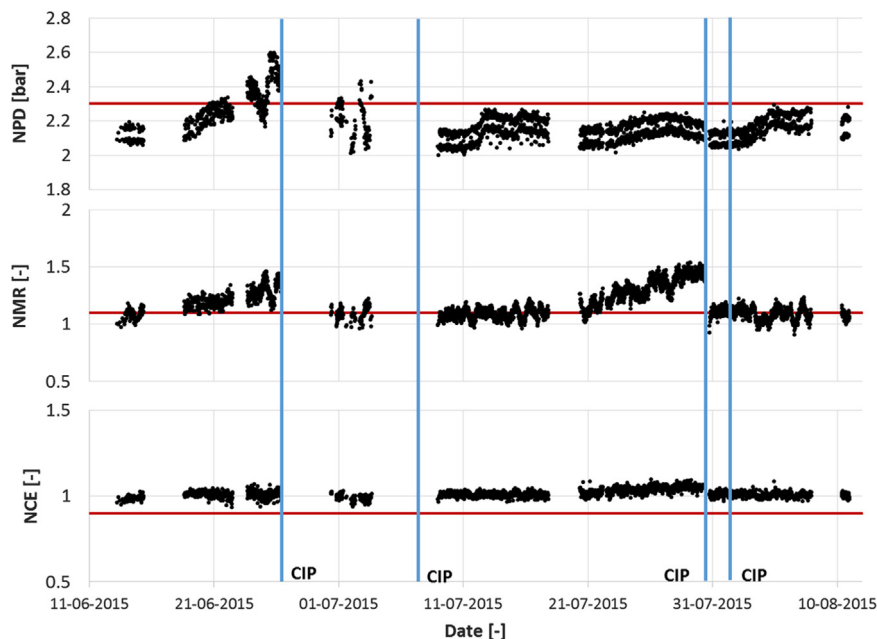

Fig. 6. Normalized pressure drop, normalized membrane resistance and normalized current efficiency from June 2015 till August 2015 for EDR treatment of cooling tower blow down water. Vertical lines represent a CIP.

the original value. These results show the effectiveness of NPD as indicator for fouling in EDR despite temperature variations between 20 and 28 °C.

The NCE remains stable at approximately unity during operation (Fig. 6, bottom graph), therefore no effect was found nor expected after applying a CIP. This NCE therefore indicates that the integrity of the membranes of the EDR stack were not compromised, thus continuing efficient desalination in terms of current utilization.

Stabilization of the NMR is expected at around 1 for EDR operation when no fouling processes were taking place. It is shown in Fig. 6 (middle graph) that the NMR starts at approximately 1.0 followed by a gradual increase which was also visible in the NPD. The performed CIP resulted in a decrease of the NMR to approximately unity. After stable operation of 10 days the NMR started to increase gradually again, however no increase in NPD was observed. The performed CIP's brought the NMR back to its original value, while the NPD appears unaffected. The NMR was therefore a helpful KPI to detect a structural increase of the resistance and a valuable next to the NPD to detect the presence of fouling. Furthermore, performing CIP's led to a comparable NMR obtained at startup of the pilot with a new EDR Stack. Therefore it can be stated that these CIP's were effective in restoring the membrane resistance and the developed KPIs showed to be very useful in controlling the EDR process.

4. Conclusions

In analogy to KPIs for pressure driven membrane processes like NF, well founded KPIs have been developed and successfully applied for the assessment of the performance of ED and EDR technology.

Based on electrochemical theories and supported by practical results in a full-scale pilot facility a set of strong KPIs were put together. Despite some scattering, the presented KPIs, normalized pressure drop, normalized current efficiency and normalized membrane resistance showed to be a useful tool to monitor and evaluate EDR process performance and initiate cleaning protocols.

The availability and use of normalized key performance indicators enables optimization and monitoring EDR performance independent of varying conditions.

A clear distinction can be made between membrane and spacer fouling by using NPD and NMR. Also membrane deterioration can be detected by following the NCE. KPIs similar to those already used for RO and NF are now also available for EDR.

Acknowledgments

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