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Review

Fouling of ion exchange membranes used in the electrodialysis reversal advanced water treatment: A review

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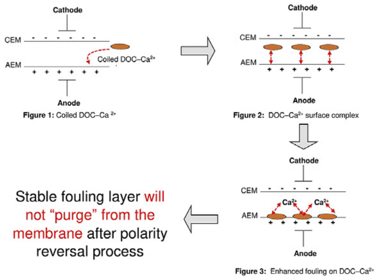
HIGHLIGHTS

- Self−reversal in EDR fails some times to get rid of foulants.
- Groundwater contains DOC−Ca^{2+} complexes alone with free Ca^{2+} ions.
- Ternary DOC−Ca^{2+} −membrane complexes responsible for irreversible fouling in AEMs.

GRAPHICAL ABSTRACT



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ABSTRACT

Electrodialysis self-reversal (EDR) technology has attracted in the treatment of water for domestic and industrial uses. The self-reversal consists of a frequent reversal of the direction of current between the EDR-cell electrodes to combat fouling of ion exchange membranes (IEMs). Irrespective of the EDR self-cleaning processes, the role of natural organic matter and their complexing ability with metal ions on IEMs fouling is partially understood. The objective of this review is to identify the research gaps present in the elucidation of IEM fouling routes. The common IEMs' foulants are identified, and several fouling mechanisms are briefly discussed. The effectiveness of self-cleaning mechanisms to reduce IEMs fouling is also be discussed. Dissolved organic carbon (DOC) possesses high chelation which forms metal complexes with di and trivalent cations found in water. The role of ternary complexes, e.g. $M^{2+/3+}$−DOC and membrane surface, on membrane fouling via surface bridging, are also addressed. Finally, mitigation methods of IEMs membrane fouling are also discussed.

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1. Introduction

Recently, UNICEF reports have confirmed that over 740 million people around the world do not have adequate safe water supply and over 2.5 billion people have access to merge water supply (World Water Assessment, 2019). Conventional water treatment technologies were primarily designed to reduce the risks of waterborne diseases by removing or chemically inactivating viruses, bacteria, and protozoa (Kumar Karn and Harada, 2002; Rappaport and Smith, 2010; Teodosiu et al., 2018). The advanced water treatment processes include technologies to desalinate seawater and remove contaminants from polluted water bodies (Song et al., 2019). Examples for advanced water treatment processes are membrane-based processes, advanced oxidation, and adsorption on activated carbon and other substrates.

Membrane-based technologies are used in water purification and desalination since 1960 (Fane et al., 2011). Membrane water treatment is a process that removes unwanted constituents from water. A membrane is a barrier that allows certain substances permeate while blocking the others. Water treatment facilities use various types of membranes and processes to clean surface water, groundwater, and wastewater to produce clean water for industry and drinking. The driving force behind membrane technologies is the pressure gradient, chemical potential, temperature, or current flux to pass liquids, gases, particulates, molecules and ions. In dialysis and forward osmosis, the concentration gradient of chemical potential is used as the driving force. Temperature-driven membrane process includes membrane desalination. To date, water desalination is best achieved either by reverse osmosis or electrochemical treatment methods. The drawback of reverse osmosis is overtreatment of feed water, high capital cost and large waste generation.

In contrast, most of the electrochemical treatment methods do not need to comply with stringent feed water chemistry, and they are low energy-intensive. In principle, electrochemical treatment methods are divided into three categories: conversion methods such as electro-oxidation, electro-reduction, and electrochemical destruction; separation methods such as electro-flotation, electrodialysis, electrodeposition, electro-filtration and electrodeionization; combined methods such as electrocoagulation, electrocatalysis, electro-disinfection, electro flotation-coagulation, electro-Fenton processes, photo-electrocatalysis, and sonoelectrocatalysis (Sillanpaa and Shestakova, 2017). In common, all electrochemical processes initiate at the electrodes while passing a direct current through the electrolyte solution. Out of the methods stated above, electrodialysis (ED) is particularly attractive in water desalination, which initially used for brackish water (TDS>3000 mg L^{-1}), seawater or brine desalination (Haddad et al., 2019). Later on, the ED technology is used for industrial applications. For example it is used to de-acidify complex solutions like sweet and acid whey generated from dairy products (Dufton et al., 2020; Lemay et al., 2020; Talebi et al., 2020), and de-odorize herring milt hydrolysates from the fishery industry (Todeschini et al., 2020). In the latter case, odorous compounds trimethylamine (pK_a 9.80), dimethylamine (pK_a 10.70) and trimethylamine oxide (pK_a 4.66) resulted from hydroxylates were removed by ED technology. The amines protonated at feedwater pH results in arresting them at the ion-exchange membranes (IEMs). The lignin yields a negative charge due to feedwater pH (black liquor pH 12.2) (Haddad et al., 2017). The negatively charged lignin selectively partitioned at IEMs removing them. Both in lignin and amines, the charge separation occurs by the polarization of electron cloud can occur under the electric field. Recently treatment of industrial discharges like polymer flooding produced water from oil and gas industry (Sosa-Fernandez et al., 2020), pig manures (Shi et al., 2020; Vineyard et al., 2020), as well as discharges enriched with heavy metals (Jin and Zhang, 2020) and municipal wastewater, are carried using ED technology.

Presently there is a growing interest in using ED methods for water desalination due to their inherent advantages. However, in some instances, the electrocoagulation methods are used to remove water hardness with limited success. The electrocoagulation operates with a sacrificing Al or Fe anode; the Al may yield undesired contaminants such as Al^{3+} resulting in water unsuitable for consumption. Further, the efficiency of removing monovalent cations in water by electrocoagulation methods is very low. Although electrocoagulation methods used to treat fluoride-rich water, the treated water may require additional treatment steps to remove fine particulates, which induces added operational cost. At the same time, electrocoagulation methods are energy-intensive, and frequent replacement of sacrificing anodes is necessary. Alternatively, in ED methods, no chemicals are added by the system, and the treated water is not entirely demineralized; hence, it restores natural water signatures. In ED, cation and anion exchange membranes are arranged alternatively between inert electrodes so that low (1000–5000 mg L^{-1}) and high (15,000 mg L^{-1}) salinity streams are separated (Al-Amshawee et al., 2020). Upon prolong operation, the ion exchange membranes tend to foul and scale. By reversing the current in specific time intervals, the membrane scaling can minimize (American Water Work Association, 1995). Despite the elegant cell arrangements of electrodialysis self-reversal (EDR), however, the fouling of IEMs cannot be eliminated.

Presently, we reviewed the mechanisms of the fouling of the IEMs used in EDR systems for drinking water treatment. Membrane fouling occurs due to organic, biological and inorganic contaminants in water. Membrane fouling mechanisms by inorganic contaminants often designated as scaling are reasonably known. However, the role of dissolved organic carbon (DOC) on membrane fouling is debatable. We discussed the efficiency of the self-cleaning process of IEMs in the presence of DOC. A new fouling mechanism is postulated via the formation of ternary complexes (metal ion–DOC–membrane surface). Fouling (and scaling) of IEMs are extensively studied in conventional ED systems, and the allied mechanisms are almost the same as in EDR (Langevin and Bazinet, 2011; Persico et al., 2017a,b). Research needs in membrane fouling specific to EDR technology are also identified. However, the role of ternary complexes in IEM fouling requires particular attention since it also perturbed scaling processes. Further, we discussed the current status of EDR technology regarding self-cleaning mechanisms, scaling, membrane fouling, in-situ and ex-situ membrane cleaning methods. Finally, new strategies adopted for the development of foul retarded EDR system are also discussed.

1.1. History and evolution of EDR systems

Both ED and EDR processes use IEMs to separate ions from aqueous solutions under the electrical flux driving force (Katz, 1979; Shaffer and Mintz, 1980). The ED process has widely been used in desalination before introducing EDR technology. Desalination setup of ED comprises of an array of alternating cation and anion permeable membranes (collectively called IEMs), which lead to the separation of product and brine compartments (Katz, 1982). Fig. 1 illustrates the ED process (Galama et al., 2014; Zrelli, 2017; Saadat et al., 2018).

The first commercial ED unit was installed in the Arabian Desert in 1954 (Katz, 1979). One of the significant obstacles found in the ED process is the scale formation due to concentration polarization, particularly on the concentrate side of IEMs (Kenneth, 1961; Giuffrida and Parsi, 1967; Thompson Brewster et al., 2017; Dufton et al., 2018; Lemay et al., 2019; Andreeva et al., 2020; Barros et al., 2020). The antiscalants, such as acids and polyphosphates, show a partial success in the prevention of scale formation on membranes. Katz et al. (1979) reported the ineffectiveness of the

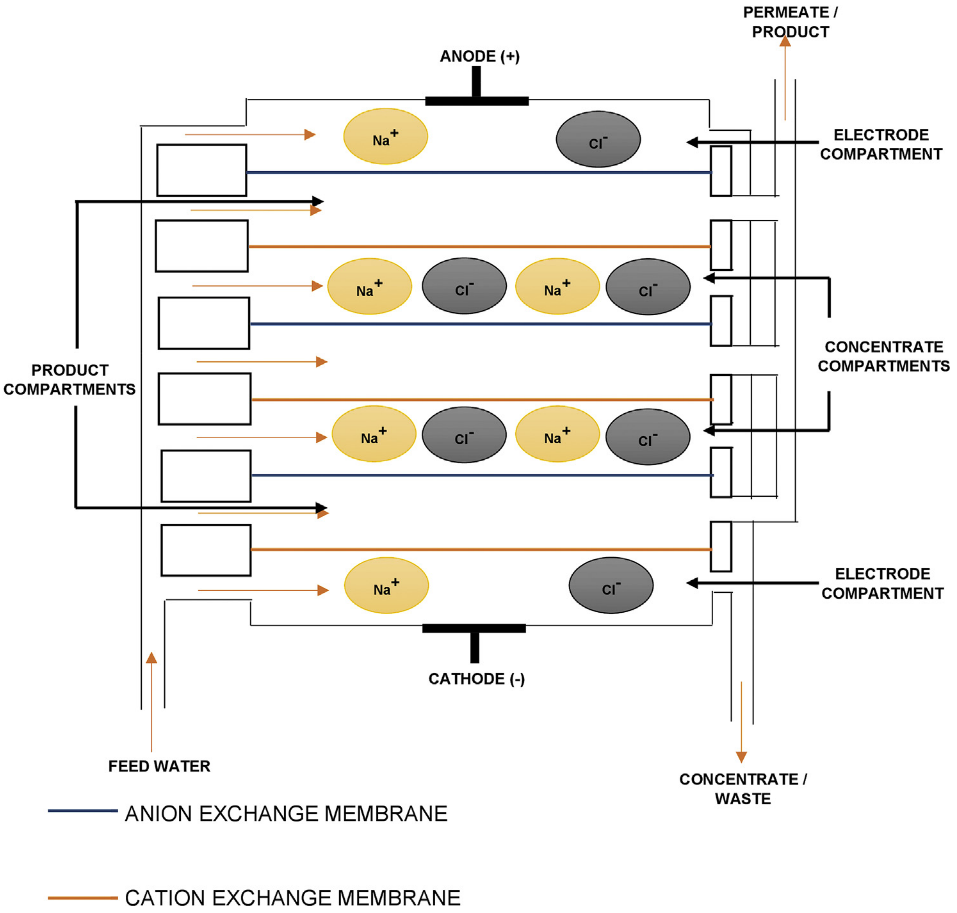

Fig. 1. Typical ED/EDR system used for desalination. Aqueous NaCl is used as feedwater (Galama et al., 2014; Zrelli, 2017; Saadat et al., 2018).

chemicals added to re-dissolve calcium salts, which caused poor flow distribution in membrane array due to localized blockages. The EDR technology is in operation since 1960 to overcome scaling and fouling problems experienced in ED (Katz, 1979; Pilat, 2003). In EDR, the polarity of the electrodes reverses by changing the direction of the direct current (DC) field at a fixed point in time domain (Pilat, 2003). Typically, the polarity of the electrodes reverses 2–4 times per hour in EDR systems. Due to the success, virtually all desalination units are designed incorporating the polarity reversal process since 1974 (Katz, 1979).

1.2. "Self-cleaning" process in EDR systems

The IEMs used in EDR comprise a specific charge. Therefore, charged particles in the feed water attract IEMs and then deposit on the membrane surface as well as inside the channels. They increase electrical resistance, decrease perm-selectivity and membrane alteration (Mikhaylin and Bazinet, 2016; Park et al., 2003). Fouling can be categorized as colloids fouling, organic fouling, scaling, or bio-fouling (Mikhaylin and Bazinet, 2016). Colloidal particles contain a net variable surface charge allowing them to migrate towards anion (AEM) or cation exchange membrane (CEM) of EDR stacks. Examples of common colloid particles found in water are silica, iron oxide, aluminium oxide, and organic colloids (Lee et al., 2003; Mondor et al., 2009; De Jaegher et al., 2020). Membrane fouling by organic compounds occurs due to carbohydrates, proteins, aromatic substances, humic substances, oil, and antifoaming agents present in feed solution (Lee et al., 2009; Tanaka et al., 2012). These substances attract to IEMs by electrostatic, chemical or hydrophobic forces (Jermann et al., 2007; Mikhaylin and Bazinet, 2016). Scaling is a different type of fouling which occurs by depositing sparingly soluble substrates along membrane channels/pores. The carbonates and sulfates of Mg^{2+}, Ca^{2+}, and Ba^{2+} are examples for scale-forming salts (Mikhaylin and Bazinet, 2016). Next category is bio-fouling, in which a biofilm develops on IEMs. Bio-fouling includes several steps; interactions of bacteria with the surface by adhereing into charged particles, production and excretion of extracellular substances, and continuous growth in developing microcolonies (Matin et al., 2011; Vaselbehagh et al., 2017; Herzberg et al., 2020).

Application of direct current across ED systems initiates concentration polarization and water splitting on IEMs (Lemay et al., 2019, 2020). In the presence of an electric field, as ions/charged particles move across the IEMs, a concentration gradient at the membrane–water interface is created. When the concentration of ions in the dilute ED compartment depletes, a limiting current density condition attained to water splitting (Rubinstein, 1977; Simons, 1979; Mani, 1991; Lemay et al., 2019, 2020; Todeschini et al., 2020). Cifuentes-Araya et al. (2013) have shown inhibition of scaling when water splitting occurs.

Due to the polarity reversal, the product compartments converted into brine compartments and vice versa (Valcour Jr, 1985; Lai et al., 2014; Shi et al., 2019). This polarity reversal process creates a "self-cleaning" mechanism in EDR systems to overcome scaling and fouling problem found in ED systems (Ramanan et al., 2013). Fubao et al. (1985) reported that the polarity reversal process permits pH changes, which initiate removing $CaCO_3$ scale and $CaCO_3$ seeds from the membrane surface. Accordingly, $CaSO_4$ scale, charged organic groups, colloids, and bacteria are removed from the EDR stack by "purge" due to reversal of polarity, as shown in Fig. 2a and Fig. 2b.

Presently, EDR treatment plants in different scales are widely used in water desalination, brine minimization and wastewater reclamation (Allison, 1995; Zhao et al., 2019). Several examples illustrate the success of the EDR systems due to their self–cleaning mechanism. Turek et al. (2003) reported an EDR system used to desalinate brackish water generated by reversed osmosis (RO) concentrate, and it functions without any scaling despite the 359% of $CaSO_4$ saturation and high Langelier index value, i.e. 2.2. In a similar study, the purification of low salinity coal–mine water with supersaturated $CaSO_4$ solution (175% of $CaSO_4$) did not form any scaling on IEMs (Turek et al., 2006). These observations show the efficiency of self-cleaning during the EDR process. Shi et al. (2019) reported EDR methods to recover nutrients from the pig manure slurry enriched with Ca^{2+} and Mg^{2+}. They reported no apparent deposition of particulates on the EDR membrane, indicating a significant reduction of membrane fouling. Pilat (2003) reported that an EDR system used for desalinating surface waters of middle Asia with 1000–4000 mg L^{-1} TDS and 600–1400 mg L^{-1} hardness could withstand without scaling EDR membranes. Allison et al. (1995) reported that a large number of EDR treatment plants are in operation for several months to treat turbid water (~0.5 NTU) without membrane fouling. Chao and Liang (2008) have reported a long-lasting EDR system used for treating wastewater with high organic solutes, colloidal particles, and microorganisms. All the above mentioned EDR systems showed less-fouling due to self-cleaning mechanisms.

2. Effect of foulants on EDR process

This section comprises information about the influence of potential inorganic (i.e. cations and anions) scalants and organic (i.e. natural organic matter (NOM) and synthetic organic matter) foulants related to IEMs fouling. A summary of related fouling mechanisms is shown in Table 1. In the discussion, Ca^{2+} and Mg^{2+} ions are considered as cationic foulants, whereas silicate and borate are treated as anionic foulants. Especially these cations and anions are capable of forming ion pairs; the relative proportion of ion pairs is strongly pH-dependent, and their role in scaling is not examined, systematically. The natural organic matter can form metal-organic complexes which pay way to hypothesize an interesting mechanism for membrane fouling.

2.1. Effect of cations (Ca^{2+}, Mg^{2+})

Ca^{2+} and Mg^{2+} are membrane scaling ions common in water. The water with excess hardness is typically treated by EDR technology. The Ca^{2+} and Mg^{2+} ions can foul IEMs, especially CEMs. Interestingly, even a small amount of scaling is sufficient, because further crystallization by nucleation would, later on, aggravate the fouling problems. Formation of supersaturated substrates such as $CaCO_3$, $Mg(OH)_2$, $CaSO_4$, and $MgCO_3$ induce scaling of IEM membranes in the ED concentrate compartments (Korngold et al., 1970; Zhao et al., 2019). When the EDR process is in progress, the concentration of the solute in the dilute compartment decreases rapidly reaching a zero-concentration condition. The resistance of the dilute compartment steadily increased as a result of ions depletion; to compensate this effect water splitting occurs on CEM generating OH^- and H^+ ions. The OH^- may penetrate CEM, i.e. hydroxide leaking, which results scaling due to formation of partially soluble metal hydroxides in the dilute compartment of AEM. Hydroxide leaking from concentrate through CEM can foul membranes by scaling (Casademont et al., 2008a,b, 2010; Cifuentes-Araya et al., 2014; Kroupa et al., 2015; Kozaderova et al., 2020). Cifuentes-Araya et al. (2014) explained minor hydroxide scaling within the dilute side of the AEM due to OH^- leakage. Although EDR has a self-cleaning mechanism which may disrupt accumulation of ions, some EDR systems have reported IEMs scaling due to Ca^{2+} and Mg^{2+} ions. Further the ion pairs as $CaCO_3^0$ induces no charge, and therefore, self-reversal process deems

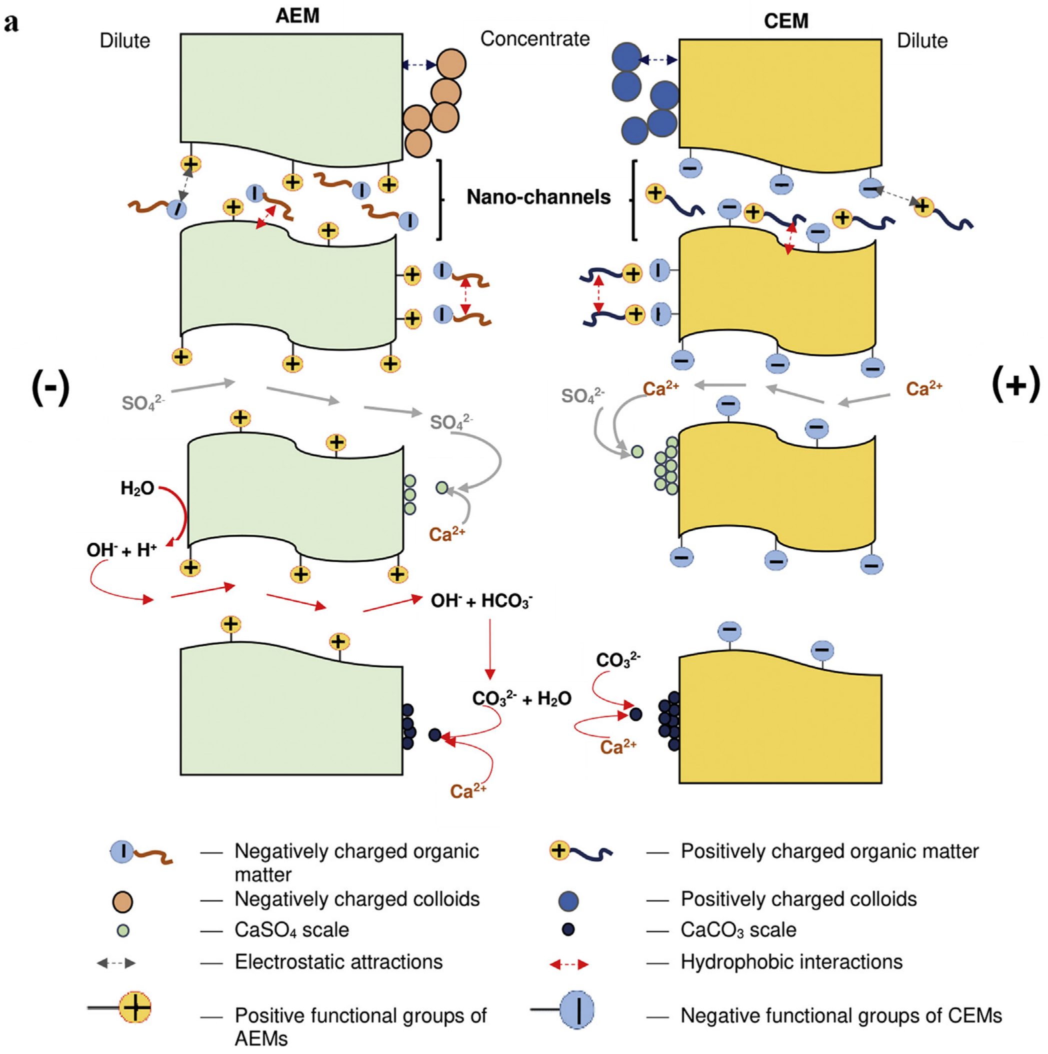

Fig. 2. Postulated mechanisms for **a)** colloidal fouling, organic fouling, and scaling **b) IEMs** “self-cleaning” mechanism to remove colloidal foulants, organic foulants and $CaSO_4$ scales. Polarity reversal is used to purge and dissolve $CaCO_3$ scales on the membrane (Fubao et al. (1985).

ineffective in scale removal. This section provides information about reported scaling effects on several ED/EDR systems.

In a typical EDR plant used for the treatment of RO concentrate, Zhao et al. (2019) reported an increase in resistance for water flux of the EDR system within the first 8 h of operation. The resistance of the EDR IEMs for water flow increased with the level of TDS in the concentrate. The scale forming potential on the IEMs increases because of the supersaturating conditions prevails in solution. In another study, Goodman et al. (2013) observed a $Ca_3(PO_4)_2$ scale on the IEM surface during municipal wastewater desalination. As the concentration of CO_3^{2-} and PO_4^{3-} increases in the concentrated stream, scale formation and a pH increase could be observed. Pilat et al. (2003) discussed a “home system” to desalinate water. The styrene-divinylbenzene membrane shows preferred precipitation of divalent cations at crystallization centres on the membrane. Concurrent occurrence of Mg^{2+} and Ca^{2+} exerts a mutual effect in membrane scaling. When both Ca^{2+} and Mg^{2+} occur together, scaling occurs preferably along with Ca^{2+} mediated pathway. Cifuentes-Araya et al. (2014) observed that hydrated Ca^{2+} ions show lower friction than hydrated Mg^{2+} with the negative sites of CEMs when penetrating them over microchannels. The higher mobility and lower hydration radius of Ca^{2+} when compared to Mg^{2+} increase the Ca^{2+} migration rate. Conversely Dufton et al. (2018) observed $Mg(OH)_2$ ($K_{sp,\ Mg(OH)2} = 5 \times 10^{-12}$ $mol^3\ L^{-3}$ at 25 °C) as a dominant scalant over $CaCO_3$ ($K_{sp,\ CaCO3} = 3.7 \times 10^{-9}$ $mol^2\ L^{-2}$ at 25 °C) due to $K_{sp,\ Mg(OH)2} < K_{sp,\ CaCO3}$. The reduction of $[OH^-]$ by Mg $(OH)_2$ precipitation inhibits $CaCO_3$ formation by decreasing HCO_3^-/CO_3^{2-} formation ($HCO_3^- + OH^- \rightarrow CO_3^{2-} + H_2O$) (Mikhaylin et al., 2014; Mikhaylin and Bazinet, 2016). However, only limited research is available identifying the influence of $[Mg^{2+}]/[Ca^{2+}]$ ratio on the degree of IEM fouling. Casademont et al. (2008a,b) showed that Ca^{2+} in $Ca(OH)_2$ enhanced scaling, however in the presence of Mg^{2+}, it has a tendency to form $CaCO_3$ as a scalant. Further studies require to clarify the role of $[Mg^{2+}]/[Ca^{2+}]$ratio in IEM fouling specially in EDR systems. To resolve the competition between $Ca(OH)_2$ and $Mg(OH)_2$, activation energies of respective reaction pathways of the rate-limiting steps require calculation.

2.2. Effect of anions and colloidal silica

Natural waters contain anions as SO_4^{2-}, CO_3^{2-}, PO_4^{3-}, OH^-, and

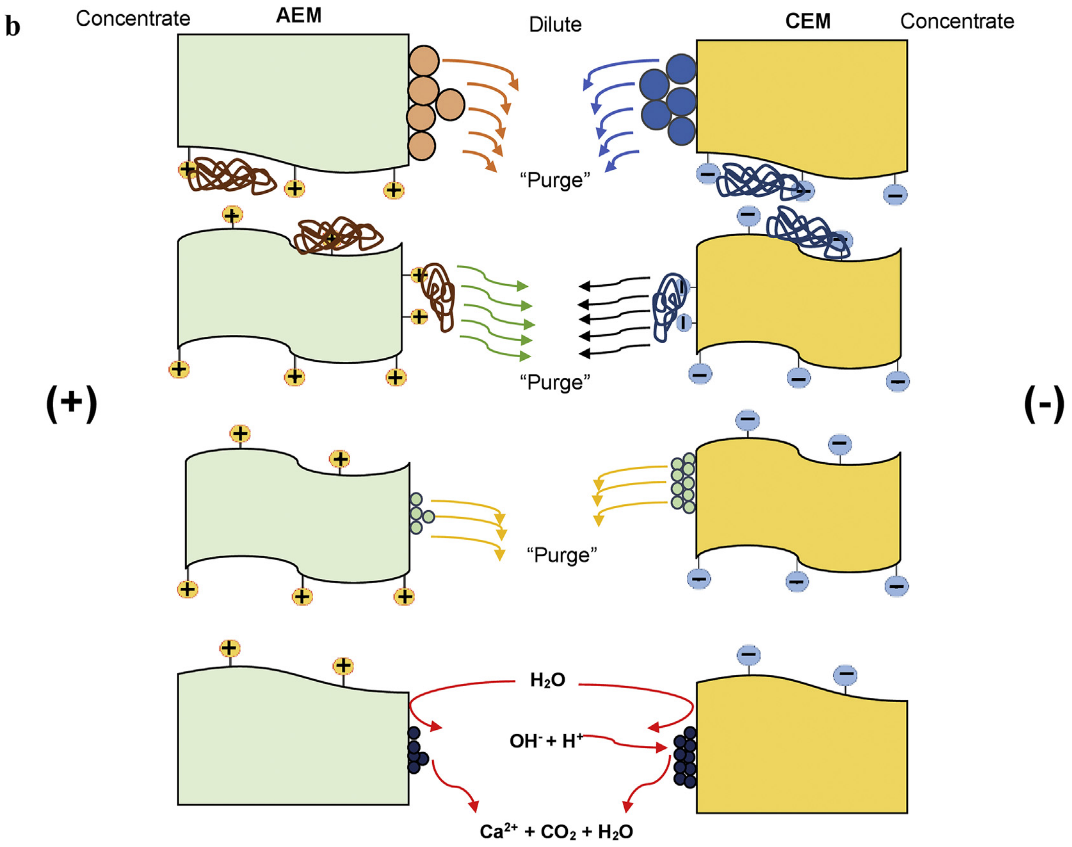

Fig. 2. (*continued*).

borates. Among them, SO_4^{2-}, CO_3^{2-}, PO_4^{3-}, and OH^- foul CEM forming sparingly soluble solids as $Ca(OH)_2$, $Mg(OH)_2$, $CaSO_4$, $CaCO_3$, or $Ca_3(PO_4)_2$ (Bazinet et al., 2000; Ayala-Bribiesca et al., 2006; Shee et al., 2008; Wang et al., 2011; Goodman et al., 2013). The membrane scaling by sparingly soluble mineral phases are reviewed in section 2.1. This section focuses more on membrane fouling caused by colloidal silica and borates.

The surface charge of colloid silica is negative at pH values typical of natural water, i.e. 6–8. The pH_{zpc} of silica surface is around 2. As a result, silica particulates move towards AEM to form electrostatic interactions with the positively charged sites of the membrane surface (Lee et al., 2003; Lee and Moon, 2004). Lee et al. (2003) reported silica sol fouling on AEM as irreversible. Mondor et al. (2009) observed a dark colouration on AEM surface due to colloid silica deposits and natural organic matter which is unable to remove by chemical cleaning (irreversible fouling). However, the role of organic matter in silica precipitation requires further exploration. Internal IEM fouling is also possible with colloid silica particles. For example, Guolin et al. (2008) observed a channel blockage by colloid silica which declines a membrane exchange efficiency. Therefore, at high pH, colloidal silica forms reversible or irreversible deposits on the surface as well as interiors of AEMs and cartridge filters. At pH values as high as 9.23, boron shows the following equilibrium (Schmitt-Kopplin et al., 1998).

$$B(OH)_3\,(aq) + OH^-\,(aq) \rightleftharpoons B(OH)_4^-\,(aq) \qquad (1)$$

The borate ions ($B(OH)_4^-$) migrate towards AEMs forming a precipitate on the membrane. Furthermore, $B(OH)_4^-$ ions, as well as $B(OH)_3$, can form complexes with the polar end of humic acids ($B(OH)_4^-$–HA complexes) (Banasiak and Schäfer, 2009). Although inconclusive, the electrostatic repulsive gradient may offset by the formation of chemical bonds between B and –COOH sites Schmitt-Kopplin et al. (1998) described that $B(OH)_4^-$ ions form complex with cis-diol groups to form stable $B(OH)_4^-$ –HA complexes. Fig. 3 (Schmitt-Kopplin et al., 1998) illustrates the six-membered ring formed by $B(OH)_4^-$ ions with 1,3-diols. Further, borates tend to form polyanionic borate species at high concentrations. Examples are $B_3O_3(OH)_5^{2-}$, $B_3O_3(OH)_4^-$, $B_4O_5(OH)_4^{2-}$, and $B_5O_6(OH)_4^-$. These ions form complexes with single hydroxyl groups. At neutral pH values $B(OH)_3$ tend to form transient hydrogen-bonded structures with carboxylate groups within and between humic acid molecules as in Fig. 4 (Banasiak and Schäfer, 2009). All those complexes are negatively charged. Therefore, they have the potential of fouling AEMs. Fouling behaviour of $B(OH)_4^-$–HA complexes on ED systems are also examined to a certain extent (Banasiak and Schäfer, 2009). However, the effect to AEMs in EDR system by the same complexes should be investigated in detail. Additionally, the presence of boron as triborates ($B_3O_4(OH)_3^-$) and tetraborates ($B_4O_5(OH)_4^{2-}$) at elevated pH produce insoluble salts with Ca^{2+} and Mg^{2+}. These salts act as severe membrane scalants (Dydo and Turek, 2013).

Therefore, particular attention should pay when raw water is enriched with major anions and silica to avoid IEM membrane fouling in EDR reactors.

2.3. *Effect of natural organic matter*

The natural organic matter (NOM) is ubiquitous in terrestrial environments (Aiken, 2002; Matilainen et al., 2010). Organic substances like proteins, polysaccharides, amino sugars, nucleic acids, humic acids, fulvic acids, cell components are IEM foulants (Ang and Elimelech, 2007; Mikhaylin and Bazinet, 2016). Mechanisms

Table 1
A summary of potential inorganic and organic foulants to EDR-IEMs with their corresponding fouling mechanisms.

Foulant	Surface charge	Membrane of affection	Mechanism of fouling	References
Inorganic cations e.g.: Ca^{2+}, Mg^{2+}	Positive	CEM & AEM	Scale formation	(Pilat, 2003; Casademont et al., 2008a,b, 2010; Goodman et al., 2013; Cifuentes-Araya et al., 2014; Mikhaylin et al., 2014; Mikhaylin and Bazinet, 2016; Dufton et al., 2018; Zhao et al., 2019; Kozaderova et al., 2020)
Inorganic anions e.g.: SO_4^{2-}, CO_3^{2-}, PO_4^{3-}, OH^-	Negative	CEM & AEM	Scale formation	(Pilat, 2003; Casademont et al., 2008a,b, 2010; Goodman et al., 2013; Cifuentes-Araya et al., 2014; Mikhaylin et al., 2014; Mikhaylin and Bazinet, 2016; Dufton et al., 2018; Zhao et al., 2019; Kozaderova et al., 2020)
Colloidal silica	Negative (pH 6–8)	AEM	Colloidal deposition	(Lee et al., 2003; Guolin et al., 2008; Mondor et al., 2009)
Boron as $B(OH)_4^-$	Negative (as $B(OH)_4^-$ at pH 9)	AEM	Deposition of DOC–Borate complexes	(Schmitt-Kopplin et al., 1998; Banasiak and Schafer, 2009)
Boron as $B_3O_4(OH)_3^-$ and $B_4O_5(OH)_4^{2-}$	Negative	CEM	Scale formation	Dydo and Turek (2013)
Organic matter e.g.: humic acid, sodium dodecyl sulfate, sulfadiazine, and tetracycline	Negative	AEM	Precipitation, electrostatic interactions, hydrophobic interactions, and formation of complexes or chelation	(Korngold et al., 1970; Allison, 1995; Lee et al., 2002a,b; Tanaka, 2007; Bukhovets et al., 2010; Goodman et al., 2013; Persico et al., 2017a,b; Shi et al., 2020)

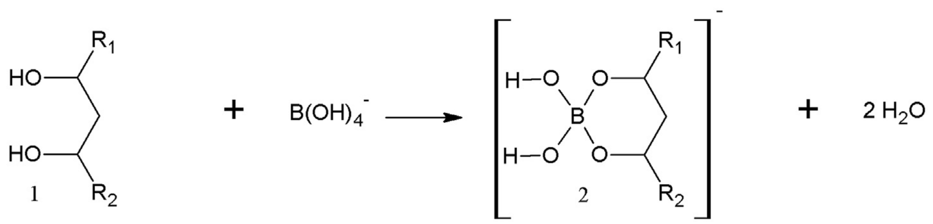

Fig. 3. Formation of a stable six-membered bidentate ester with $B(OH)_4^-$ and HA (Schmitt-Kopplin et al., 1998).

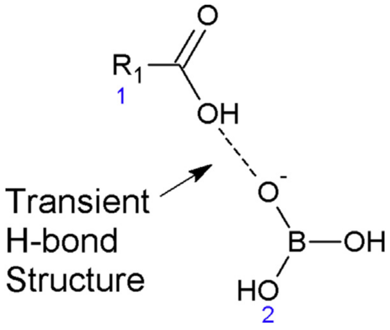

Fig. 4. Transient hydrogen-bonded structure between humic carboxylates and boric acid at neutral pH (Banasiak and Schäfer, 2009).

behind IEM fouling in EDR include precipitation, complexation, or aggregation in the presence of chelating agents at specific pH and temperature (Strathmann, 2010; Haddad et al., 2017; Bdiri et al., 2019). Fouling mechanism varies with the nature of NOM and the type of the IEMs. We discuss factors affecting IEM fouling by NOM; these factors are molecular weight fractions, feed solution pH, NOM solubility, molecular charge, macromolecular structure, surface roughness and pore size of nanochannels.

Allison explained the effect of molecular weight on the organic fouling in IEMs (Allison, 1995). Accordingly, total organic carbon (TOC) removal efficiency from EDR is around 35%, and the removed portion of organic matter attributes to low molecular weight ionized fractions (200–300 Da). The medium-weight (250–700 Da) organics caused internally reversible fouling in IEMs, and that can easily remove by chemical cleaning (i.e. salt flush). However, high molecular weight fractions of organic molecules (>1 kDa) render irreversible fouling on IEMs, which is not removed by chemical cleaning. Therefore, the gradual deposition of high molecular weight fractions of organic moieties tends to foul IEMs. For example, Haddad et al. (2017) observed the surface precipitation of lignin on CEM. The protonation of phenolic functional groups on the lignin surface enhances self-aggregation into lignin colloids via attractive van der Waals forces that result in the formation of a deep fouling layer on the membrane surface. Further, the AEMs fouling occurs due to negative charge on high molecular weight organics. Korngold et al. (1970) also reported that the increase of the molecular weight of the organic fraction increases the resistance of IEMs for water permeation. Zhao et al. (2019) observed reduced IEM fouling on EDR when treated the concentrates resulted from RO. They noted the abundance of neutral, low molecular weights (LMWs) in RO concentrates, which are not attracted by IEMs in EDR. In contrast, Bukhovets et al. (2010) reported severe AEM fouling due to the presence of LMW organic anions and the low mobility inside the membrane. Haddad et al. (2017) explained the fouling mechanism considering the hydrophilicity of LMW organic acids. In typical natural water pH, i.e. pH 5–7, acidic lignin groups readily deprotonate and form electrostatic complexes with positively charged quaternary ammonium sites ($^+N(CH_3)_3$) sites on AEM which facilitate internal membrane fouling. However, lignin is a cross-link polymer with a hydrophobic backbone structure. Therefore, weak electrostatic complexing of NOM may promote hydrophobic interactions with the backbone structure of the lignin. Detailed investigations into hydrophobic interactions between lignin backbone moieties and membranes are required, and such assessment may shed light in identifying irreversible fouling. Other than the NOM molecular size factor, the solution pH also plays a significant role in membrane fouling. Organic acids such as humic substances deprotonate/protonate depending on the pH. Further, amino acids in proteins carry different charges depending on their isoelectric points, which changes with the solution pH. Therefore, electrostatic interactions between NOMs and IEMs highly depend on the pH. For example, Persico et al. (2017a,b) explained a fouling mechanism by peptides on CEM at three pH values ($pH = 2$, 6, and 10). The highest membrane fouling is noted at pH 6 while virtually no fouling occurs at pH 10; at pH 2 the degree of membrane fouling is somewhat intermediate. At extremely acidic conditions, i.e. pH 2, the concurrent protonation of sulfonate on the membrane and acidic groups on the natural organic matter reduces membrane fouling significantly. At basic conditions, i.e. pH 10, the same arguments are valid for deprotonated anionic groups in the reduction of membrane fouling. Membrane fouling at pH 6 is due to the formation of a monolayer between protonated amino groups and sulfonate sites on CEM by electrostatic interactions. Intense irreversible membrane fouling does occur due to hydrophobic interactions between peptide and membranes. The hydrophobic segments on peptide tend to lean away on CEM surface facilitating peptide−peptide interactions to form deep secondary fouling layer on the membrane. When the pH of a peptide solution varies, Persico et al. (2016) observed a reversed order in AEM fouling noting highest fouling at pH 10 and no fouling at pH 2. Around pH ~6, AEM fouling by peptide occurs moderately. In peptides due to the presence of electron-withdrawing amino groups, the acidity of carboxylic groups is high (typically pK_a 2.1), and the basicity of amino groups is low (pK_a 9.8). Therefore, at pH 6, the carboxylic groups on peptides readily deprotonate, creating negatively charged organic moieties. However, the amino groups remained unchanged. In peptides the number of carboxylic sites is higher than amino sites. When pH 6, therefore the peptides are net negatively charged. Therefore, at pH 6, the interactions between peptides and AEM are attractive but less significant. Similarly, around pH 2, the peptide molecules are net positively charged due to protonation of both amine and carboxylic groups. Bacher et al. (2017) reported that fouling of AEM due to a tannin based product, which is used as a coagulant during the pretreatment. They reported when increasing pH from 5 to 6.5, the charge of tannins become negative, resulting in excessive AEM fouling. Further, the excess tannin amount used in the pretreatment step results fouling. Lee et al. (2002a,b) proposed a mechanism attributes to AEM fouling caused by humic substances having a large molecular weight and a high negative charge density. They also reported the influence of pH on AEM fouling and the importance of electrostatic interactions on fouling. Therefore, the application of an electrical field and a reversal of polarity phases in EDR should contain a great impact on fouling mechanisms and further research into the area is required. Other than pH, the solubility of organic substances also affects on membrane fouling. Generally, the less soluble organic molecules in water lead severe fouling (Bukhovets et al., 2010; Bdiri et al., 2018, 2019). Confirmation and the nature of charge distribution of a foulant have a significant impact on determining membrane fouling mechanisms, particularly for peptides due the presence adjacent charges with opposite signs (i.e. COO^- and $^+NH_3$) that could neutralize (Persico et al., 2016).

Membrane characteristics have also influenced the fouling of IEMs. The anion exchange membranes are mostly affected by organic fouling. They carry positive sites; for example, $-NH_3^+$, $-NRH_2^+$, $-NR_2H^+$, and $-NR_3^+$ groups. Thus, negatively charged molecules/moieties easily attract towards them (Xu, 2005). For example, sodium humate and anionic polyamides readily foul AEMs (Korngold et al., 1970; Guolin et al., 2008). Although fouling in AEM is prominent, few studies are reported on CEM fouling by NOM. For example polyphenolic substances (e.g. anthocyanin) caused fouling on CEM due to their positive charges at specific pH values (Bdiri et al., 2018). Further, Bukhovets et al. (2010) reported on CEM fouling due to amino acids because of their variable charge. Chemical composition of the membrane also plays a pivotal role in deciding the IEMs fouling by organics. Irreversible fouling occurs in styrene-divinylbenzene based AEM due to hydrophobic interactions between benzene rings of foulant and the membrane. However, in acrylic-based membranes, no fouling is reported (Allison, 1995; Tanaka, 2007). Bukhovets et al. (2010) reported fouling due to negatively charged amino acids as a result of stacking interactions between membrane components. Bdiri et al. (2020) also discussed the importance of stacking interactions between the polymer matrix of the AEM and the foulant (e.g. anthocyanin).

Further, membrane fouling is affected by the roughness of the membrane (Shi et al., 2019; Persico et al., 2020). As shown in Persico et al. (2020) increased surface area of IEMs allows strengthening the membrane fouling. Another important membrane characteristics for fouling is the pore morphology of nanochannels. Persico et al. (2020) observed severe fouling with membranes containing narrower pore channels. The confined narrow pore channels can intensify the frequency of foulant interactions with the membrane when compared to full pore analogues. In conclusion, the foulant and membrane characteristics are vital in proposing a membrane fouling mechanism.

2.4. *Effect of synthetic organic matter*

Many anthropogenic compounds such as detergents, dyes, and non-oxidizing biocides are present as trace contaminants in natural waters (Önder et al., 2007; Aleksic et al., 2010; Shi et al., 2020). They are potent IEM foulants (Allison, 1995). In section 2.3, the current status of the organic fouling mechanisms of IEM membranes was discussed. The essentials of IEM fouling by synthetic organic pollutants are discussed below.

Shi et al. (2020) examined membrane fouling by sulfadiazine (SD) and tetracycline (TC) found in manure of pigs vaccinated with antibiotics. At pH 5.5 (feed solution), TC binds on AEM; however, no SD detects on AEM membrane because of the small size, SD permeates through the membrane nanochannels. However, due to restricted motion within the nanochannels, SD sorbed on the interior surface of the AEM creates a negatively charged amino groups (i.e., $R-NH_2$, pK_a 2.5 and $R'R''-NH$ pK_a 6.5) that can exchange with AEM exchange sites. Therefore, some of the deprotonated SD may retain inside AEM through H-bonding, which induces fouling by SD to some extent. In TC, the amide (pK_a 3.3), phenolic (pK_a 7.7) and amine (pK_a 9.7) functional groups carry different charges which may self-neutralize to some extent, thus dominating H-bonding interactions in membrane fouling. The large size of TC prevents permeation it through the AEM membrane. The feedwater pH (pH 5.5) favours TC binding on AEM, which subsequently promotes AEM fouling. When compared to SD, the TC preferably retains with concentrates which favour membrane fouling. In another study, Allison et al. (1995) reported fouling caused by sodium dodecyl sulfate, a commonly used laundry detergent. In the pretreatment step, the charged antiscalants, i.e. polyacrylate and hexametaphosphate, are also used. They observed fouling on AEMs due to the antiscalants (Lindstrand et al., 2000). Also examined the AEM fouling by anionic surfactants such as sodium octanoate and sodium dodecylbenzene sulfate used in the pulp industry. The increased concentration of foulants due to concentration polarization increased the resistance of water flow through IEM. In this case, the self-reversal mechanism of EDR is ineffective in removing foulants from the membrane. The effect of charge reversal in IEM membrane fouling mechanisms due to synthetic organic pollutants requires further investigation.

2.5. Effect of co-existence of metal ions and organic matter

Different molecular fractions of NOM and synthetic organic matter can chelate metal ions ubiquitous in water (Jermann et al., 2007; Schaefer et al., 2009; Shi et al., 2014; Makehelwala et al., 2019; Zhang et al., 2019). Natural water and wastewaters contain divalent and trivalent metal ions (e.g. Ca^{2+}, Mg^{2+}, Ba^{2+}, Al^{3+}, Fe^{3+}) and organic substances (e.g., humic acid, fulvic acid, proteins, polysaccharides). Sections 2.1-2.4 reviewed the fouling potentials of them as individual candidates. Understanding the interactions between metal ions and natural organic matter require elucidating the mechanism of membrane fouling. Section 2.5 discusses the role of $M^{2+/3+}$—DOC complexation on IEM fouling. It is a well-established that DOCs are capable of reducing crystal growth rates, especially calcite growth (Reddy, 1978; Reddy and Hoch, 2000, 2001; Amjad et al., 2002; Leenheer and Reddy, 2008). The nature of DOC influences the growth rate of an inorganic crystal.

Ang et al. (2007) reported fouling mechanism of membranes with bovine serum albumin (BSA) and Ca^{2+} complexes. Ca^{2+}—BSA forms a complex with the reduced charge that enhanced the stability on IEMs. The free Ca^{2+} is not bridging efficiently with adjacent organic moieties to form stable fouling layers (Ang and Elimelech, 2007). However, in the presence of Ca^{2+} —DOC complexes enhanced membrane fouling occurs due to charge reduction of Ca^{2+} by a bridging mechanism with the surface. Ren et al. (2008) also reported enhanced CEM fouling with Ca^{2+} in the presence of amino acids. The feed solution pH ($<pH_{IEP}$) provides a net positive charge on CEM which favours membrane fouling. In the presence of amino acids, severe CEM fouling is observed. In this case, the EDR self-cleaning process seems ineffective due to the reduced charge of Ca^{2+} ion in Ca^{2+}—amino complexes. In this way, the DOM seems triggering inorganic or organic membrane fouling in the presence of di- or trivalent cations. Dufton et al. (2019) have shown an intimate association of calcium and phosphorus with lactic acid in a fouled AEM. However, the exact molecular configurations of calcium or phosphorous with lactic acid require further investigation. The XRD data show an amorphous calcium phosphate phase; however, elucidation of fine structures of calcium—organic complexes using synchrotron analysis is vital in understanding membrane fouling (Trudel et al., 2011). In conclusion, it is essential to identify fouling sequence which would provide detailed information about fouling mechanisms by metal ion—organic—membrane ternary complexes.

2.6. A hypothetical mechanism to elaborate AEM fouling in the EDR systems by DOC—Ca^{2+} complexes

A hypothetical mechanism is proposed to understand IEM membrane fouling by DOC—Ca^{2+} complexes. Initially, electrostatic interactions arise within the double layer region between reduced charge DOC—Ca^{2+} complexes and positively charged AEM surface (due to positively charged active sites, for example, $-NH_3^+$ sites). Then hydrophobic interactions, hydrogen and chemical bonding initiate and promote ternary complexing between DOC—Ca^{2+} and AEM surface sites. Free Ca^{2+} in raw water can give rise to the intermolecular bridging of DOC—Ca^{2+} with surface sites, which allow the formation of a stable fouling layer on or inside AEMs channels. According to this hypothesis, the polarity reversal phase permits foulants to “purge” from the membranes only if they are sufficiently negative to repel from the negative electrode. However, the negative charge of the foulant is reduced due to metal ion—DOC complexation; hence it may not purge from the membrane. Unpurged foulants may lead to severe membrane fouling by forming stable fouling layers. Further research is required to clarify the proposed fouling mechanism.

3. Chemical cleaning process on EDR

Self-cleaning process of the EDR largely controls the fouling due to scaling, colloids, and natural organic matter. Despite this fact, during EDR operation, foulants are built on the membrane surface, channels and spacers (Bisselink et al., 2016). In-situ and ex-situ cleaning processes are carried out to prevent efficiency loss in EDR performance. This section comprises a brief account of chemical cleaning carrying out in EDR.

3.1. In-situ cleaning process

In-situ cleaning or clean-in-place (CIP) carries out cleaning without dismantling EDR to remove foulants from the stack membranes (Bremer et al., 2006; Tamime, 2009; Tuck et al., 2019). In CIP, chemical agents such as acids, alkali, salts, and oxidants flushed to membranes. Further, CIP protocols are applicable to remediate reversible fouling due to physical adsorption of foulants onto membranes (Shi et al., 2019). Acids like HCl are frequently used in CIP to eliminate multivalent cationic salts such as $CaCO_3$, $CaSO_4$, and $MgCO_3$ (Van der Hoek et al., 1998; Valero and Arbós, 2010; Garcia-Vasquez et al., 2016). Alternatively, alkali solutions like NaOH are used to cleave organic foulants such as polysaccharides and proteins (Garcia-Vasquez et al., 2016).

Further, strong oxidants (e.g. NaClO and Cl_2) are used to achieve complete cleaning by the degradation of protein functional groups present in bacteria, thereby preventing the biological growth (Dydo and Turek, 2013). Salts remove proteins due to salting-out effect (Bdiri et al., 2019). The IEMs fouling due to proteins occurs mainly due to electrostatic interactions (Haddad et al., 2019). Salt solutions (e.g. NaCl) can destabilize the electrostatic interactions detaching solutes from the membranes (Langevin and Bazinet, 2011; Persico et al., 2017a,b). The proteins can interact with the membrane at both polar and non-polar regions. Persico et al. (2017a,b) used NaCl to remove electro-statistically adhered proteins at polar regions in the membrane. In contrast, sodium dodecyl sulfonate removes proteins bound with the membrane through hydrophobic forces. The American Water Works Association recommends the use of 2—5% HCl, 3—5% NaCl and NaOH (pH 8—10), and 10—50 mg L^{-1} Cl_2 for in-situ cleanings of membranes (Murry, 1996; Talebi et al., 2019).

Several studies are carried out with acid-CIP to assure optimal performance in EDR (Tanaka, 2007; Shi et al., 2014). Shi et al. (2014) used HCl to acquire effective membrane cleaning to remove protein-like substances. In acidic pH, amino functional groups in proteins become positively charged. Generation of a loose fouling layer attributes to electrostatic repulsion forces between positively charged amino acids and active sites on anion membrane (Tanaka, 2007). This mechanism provides a satisfactory explanation for the removal of protein on the membrane by acid-CIP process. In acidic pH carboxylate and phenolate groups in humic acid substances protonate. As such, the neutralization of humic substances helps to eliminate them from the AEM surface (Guo et al., 2015; Shi et al., 2019). Guo et al. (2015) observed considerable amounts of Ca^{2+} and Ba^{2+} with anionic polyacrylamide (APAM) in the cleaning solution after treating fouled IEMs by oily wastewater. The formation of chelation complexes between APAM and inorganic metal ions attribute to the above observation.

Base-CIP has also played a significant role in restoring the IEMs performance. Guo et al. (2015) reported NaOH as an excellent defouled agent for fouled CEM due to APAM. Functional groups of organic foulants are deprotonated and become negatively charged in alkaline pH (i.e. $pH = 11$). The resultant repulsive interactions between foulants and fouling layer would loosen the latter to pass into cleaning solution (Lee and Elimelech, 2006; Ang et al., 2011; Wang et al., 2014; Guo et al., 2015). Deterioration of the IEMs by NaOH cleaning solution has also been examined (Garcia-Vasquez et al., 2014; Xia et al., 2019). For example, Garcia-Vasquez et al. (2014) reported about obstacles originated with strong alkaline conditions, which changes the physicochemical properties of IEMs. In this study, polyvinyl chloride polymer binder was used in AEM preparation. Application of strong-alkali agents led dehydrochlorination on polyvinyl chloride into to polyene structure. Cracks and fractures are observed on AEM after CIP treatment which may be due to modifications that occurred in the polymer network. Also, a high degree of AEM degradation was observed. AEMs contain quaternary ammonium sites, whereas CEMs contain sulphonate sites. The enhanced dehydrochlorination by quaternary ammonium sites facilitate AEM degradation; however, such observations are minimal in CEMs (Garcia-Vasquez et al., 2014). Haddad et al. (2016) noted holes on AEM surface after alkali treatment of CIP (NaOH solution at pH 12 and dilute black liquor solution at pH 12). However, the exact mechanism of IEM degradation by alkali conditions is not clear todate.

In most circumstances, CIPs have included successive additions of acid and then a base or first an acid, then a base according to the feed solution pH (Garcia-Vasquez et al., 2014). These cleaning cycles initiate inflate-deflate sequencing in IEMs because of the rapid changes as Cl^- and OH^- on the polymeric material. Subsequent elimination (by NaOH) and addition (by HCl) permits a mechanical push of polymer from the IEMs. This influence is observed only in AEMs because negatively charged OH^- and Cl^- can only migrate towards AEMs (Garcia-Vasquez et al., 2014). In some cases, they observed only partial restorations in membrane performance by application of CIP cycles. For example a base-CIP cycle used in the recovery and concentration of ammonia from swine manure by ED, is only facilitated in partial restoration of membrane integrity (Mondor et al., 2009).

In addition to acid-base CIP approaches, salt flushing and addition of chemical oxidants are used in numerous EDR systems. Allison et al. (1995) reported the effective use of salt flushing to remove medium molecular weight organics while treating water using EDR. Oxidants are another type of cleaning agent found in CIP. Garcia-Vasquez et al. (2014) stated NaClO as the most frequently used oxidant. HClO, ClO^-, and Cl_2 exist in an aqueous NaClO that are capable of oxidizing both organic and inorganic foulants. These oxidants could also disulfonate or deaminate, sulfonate and ammine sites on the IEMs. It affects the membrane perm-selectivity due to destruction of IEMs active sites (Garcia-Vasquez et al., 2014). Garcia-Vasquez et al. (2014) explained the destruction process by a radical oxidation mechanism which is profoundly affected by the feed solution pH. Saline solutions (seawater) are also used as a CIP agent in cleaning IEM membranes fouled with waste solutions in the food industry. However, intensified membrane fouling is observed after CIP cleaning, possibly due to following factors (Bdiri et al., 2018). Waste solutions in food industry comprise polyphenols, polysaccharides, amino acids, and proteins, and the chelates can form complexes with divalent and trivalent ions such as Ca^{2+}, Mg^{2+}, Fe^{3+}, etc. ubiquitous in seawater to form colloids to foul fouled-membranes (Bdiri et al., 2018). A water-ethanol mixture acidified with H_2SO_4 is used in CIP to overcome the above difficulty experienced with saline solutions. The main drawback when using the water-ethanol mixture in CIP is the necessity of removing ethanol from the solution by a distillation owing to its environmental concerns (Bdiri et al., 2018).

Applications of CIP built as a multiple-step process includes successive addition of different cleaning agents. For example, Bisselink et al. (2016) reported a two-step CIP process which includes a salt CIP (5 wt% NaCl and NaOH in pH 8–10.5) and acid CIP (5 wt % HCl, $pH < 1.2$) steps to remove organic foulants and to scale assuring optimal performance in EDR.

3.2. *Ex-situ cleaning process*

Irreversible internal fouling of IEMs due to DOC sorption on membranes sometimes compels complete shutting down of EDR plants. Chemical cleaning methods such as CIP cannot remove irreversible fouling of membranes (Lee et al., 2002a,b; Jermann et al., 2007). Ex-situ cleaning is carried out by dismantling the EDR stack and manual cleaning. When compared to RO, nanofiltration or micro-filtration, the fouled membranes in EDR can easily clean by disassembling the stacks (Valero and Arbós, 2010). In EDR plants, routinely the ex-situ cleaning of membrane is carried out in every eight months (Allison, 1995). For an EDR plant of 47, 320 L h^{-1} capacity, for example, the cost for manual cleaning is around $16 900, and the task required 100 working hours (Allison, 1995). Ex-situ cleaning process played a significant role in the restoration of process performance in EDR systems when they subject to irreversible internal fouling. However, this is a costly process, and it requires well-trained persons to avoid incorrect reassembling. However, some EDR systems did not restore complete performance after the ex-situ cleaning step (Allison, 1991).

4. Development on EDR systems towards fouling prevention

This section reviews the development of IEMs by various membrane-surface modifications, pretreatment processes, and process optimization mitigating fouling problems.

4.1. *Development of ion exchange membranes*

The degree of IEMs fouling has been examined as a function of surface charge, hydrophilic or hydrophobic balance, and surface roughness (Mikhaylin and Bazinet, 2016). Developing AEMs by changing the resin material were reported in late 1979. In Texas (USA), styrene-divinylbenzene AEMs were used to demineralize municipal water which contains anionic organic compounds with membrane fouling potential. To membrane fouling by anionic organic compounds, acrylic-AEMs were successfully substituted with styrene-divinylbenzene-AEMs (Tanaka, 2007). The acrylic membranes do not have binding sites to trap organic foulants. However, benzene rings present in styrene-divinylbenzene provide selective sites (due to stacking-π forces) to trap organic foulants and thereby increased membrane fouling. As a result, acrylic-based AEMs became popular in EDR technology (Grebenyuk et al., 1998; Park et al., 2003). Modified the surface of AEM to repel negatively charged organic foulants. Modification to styrene-divinylbenzene AEM was carried out by coating a high molecular weight (500, 000 Da) surfactant (Mulyati et al., 2012, 2013). This method allowed the formation of a surface layer with enhanced negativity requires to repel foulants from IEM. Fernandez-Gonzalez et al. (2017) also found an improved fouling resistance in polyethene AEMs when coated with negatively charged nanocomposites made with poly (2,6-dimethyl-1,4-phenylene oxide) and oxidized multi-walled carbon nanotubes ($CNTs\text{-}COO^-$) or sulfonated iron oxide ($Fe_2O_3{-}SO_4^{2-}$) nanoparticles. Another interesting approach in the prevention of fouling is to convert membrane chloromethyl styrene units → carboxylic acid groups. The negative charge on carboxylates repels anionic foulants from the surface enhancing antifouling properties of the membrane (Kusumoto et al., 1975). Other than chemical-based surface modifications, Tamersit et al. (2018) studied the protection of the AEM using an ultrafiltration membrane barrier before EDR treatment. It rejects proteins by approaching AEM.

4.2. Pretreatment processes

Elimination of potential foulants from the raw water by a pretreatment process is an effective prevention method of membrane fouling. By adding strong oxidants such as ClO^-, O_3, H_2O_2 and MnO_4^-, the organic matter in the feed water of the EDR destructs which prevent microbial growth (Allison, 1991; Harries et al., 1991; Heshka, 1992; Hughes et al., 1992; Goodman et al., 2013). Addition of scale-inhibitors like polyacrylate and sodium hexametaphosphate has been studied intensively (Heshka, 1992; Turek and Dydo, 2003; Turek et al., 2006). Yeh et al. (2016) used an array of pretreatment processes, i.e. coagulation, flocculation, sedimentation and fibre filtration. Other than chemical addition, filtration methods, including activated carbon and sand filters, have been successfully used to remove organic matter from feed water (Huang et al., 2007; Taylor, 2011; Venzke et al., 2017). Pressure driven membrane processes, i.e. ultrafiltration also reported as a pretreatment method before feed water to EDR. However, it is noted that the pressure-driven membranes also have a high fouling risk (Huang et al., 2007; Chen et al., 2014; Wang et al., 2013).

4.3. Optimization of process conditions

Innovative modifications to the EDR process itself is a novel concept in reducing fouling potential. For example, hydrodynamic parameters like flow rate and turbulence require to control fouling. Further, process parameters like current density and frequency of polarity reversal are also crucial in the prevention of membrane fouling (Grossman and Sonin, 1972, 1973).

High flow velocities allow the destruction of deposited colloids and adhered organic foulants by scrubbing them from the membrane surface (Johnson and Hill, 1976). Spacers in the EDR system maintain flow velocities inside membrane stacks; substantial attention is paid to design spacers maintaining high flow velocities (Kim et al., 1983; Allison, 1995; Korngold et al., 1998; Messalem et al., 1998; Długołęcki et al., 2010). Another way of eliminating fouling from IEMs is the creation of turbulence in EDR compartments (Booster et al., 2004). Further, several studies have also been carried out by applying a pulsed electric field (PEF) on IEMs to disturb the formation of gel layers on the membrane surface (Lee et al., 2002a,b; Cifuentes-Araya et al., 2013, 2014; Sayadi et al., 2015; Haddad et al., 2019; Lemay et al., 2019, 2020; Dufton et al., 2020; Sosa-Fernandez et al., 2020). Fouling and scale prevention by PEF occur due to restored ions on the membrane surface during pause lapsing cycles which prevent the concentration polarization markedly. Several researchers examine the effect of PEF on pulse & pause duration as well as pulse-pause frequency in fouling prevention (Lemay et al., 2019; Dufton et al., 2020; Sosa-Fernandez et al., 2020). Another way of reducing fouling in EDR–IEMs is lowering the polarity change frequency; hence a low pulsation removes foulants from IEMs (Lee and Moon, 2002). Formation of fouling layers can perturb by applying a disturbance, mechanically. For example, an ultrasonic or a magnetic field can inhibit the nucleation rate of scale growth due to mechanical disturbance (Sayadi et al., 2015). Booster et al. reported an elegant method by applying a small pressure difference across the membranes to breakdowns scaling layers (Booster et al., 2004). However, internal membrane fouling layers cannot be removed by this method. Also, the optimal frequency for the application of pressure difference should predetermine to assure the required particle size. Otherwise, particles with larger size will not remove from the IEM surfaces. Another parameter used to mitigate fouling is applying a high current high flux (Lee et al., 2013). High current density split water which changes pH and enhances the scale formation on IEMs found in concentrated compartments. Therefore, applying current density should not exceed the limiting value (limiting current density, LCD) even though it facilitates enhanced permeate flux.

5. Conclusions

Polarity reversal in the EDR permits a self-cleaning mechanism to remove scaling, colloidal deposition, and charged organics from the IEMs. EDR systems are efficient in removing hardness, salinity, charged organics, and even turbidity from water even with high saturation and Langelier indexes. On the other hand, EDR is incapable of removing uncharged components as bacteria in raw water.

Despite a self-cleaning mechanism, several EDR systems experience severe irreversible fouling mainly due to DOC and associated metal ion complexes. Irreversible fouling can be removed by cost-ineffective ex-situ cleaning through disassembling in EDR stack systems. “How fouling occurs in EDR systems even with the presence of a polarity reversal phase?” is unanswered to date due to the lack of information available on fouling mechanism in the presence of a polarity reversal phase.

A handful of information is available to show the co-existence of DOC with Ca^{2+} in raw water to form DOC–Ca^{2+} complexes. How EDR performs under this condition? And the effects of polarity reversal on the removal of DOC–Ca^{2+} complexes from the membrane require a detailed study in elucidating fouling mechanism. The postulated mechanism of IEM membrane fouling by DOC–Ca^{2+} complexes requires validation by experimental and theoretical data.

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.

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