



Review Paper

How Operational Parameters and Membrane Characteristics Affect the Performance of Electrodialysis Reversal Desalination Systems: The State of the Art *

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HIGHLIGHTS

- Effects of operating factors and membrane characteristics in ED/EDR were reviewed
- Voltage, flow rate, and temperature are the main effective factors in ED/EDR process
- Membrane characteristics, feed composition, and pH also influence ion removal
- Effects of flow rate on the desalination rate and product quality were classified

ABSTRACT

Operating parameters and membrane characteristics strongly affect the performance of electrodialysis reversal systems. The most impactful factors are applied voltage, flow rate, temperature, initial feed composition, and ion exchange membrane characteristics; the pH of the feed also has an effect, although to a lesser extent. To determine more precisely how all of these factors impact performance – especially as measured by desalination rate and the quality of the desalinated water – several valuable studies have been conducted. However, no comprehensive paper has been published, and some researchers have reported different findings from differently designed electrodialysis reversal systems. Therefore, to synthesize previous research and reconcile superficially disparate findings, this review discusses, analyzes, and summarizes the results from the literature. Such a review can help researchers and technicians optimize electrodialysis reversal systems and improve the quality of desalted water.

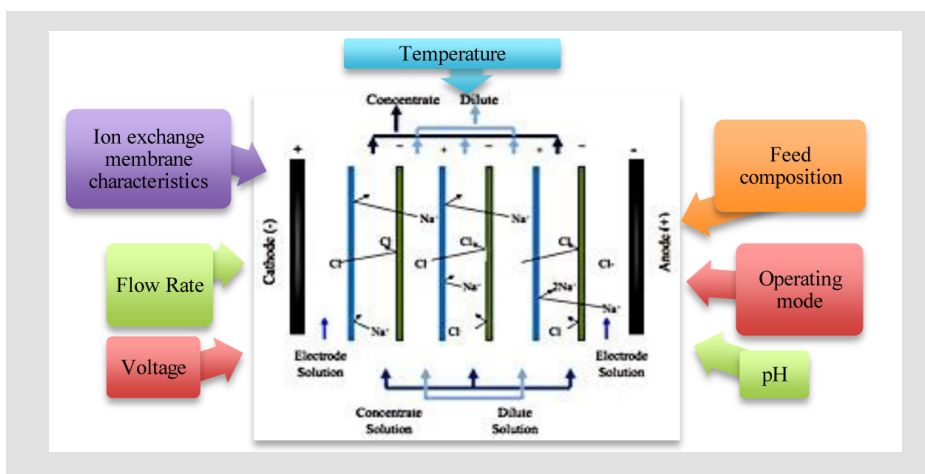
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1. Introduction

Electrodialysis / electrodialysis reversal (ED/EDR) is a membrane-based desalination process in which ions are transferred through ion exchange membranes (IEMs) under the influence of an applied electrical field [1]. The technology is very effective in desalinating brackish water with low to moderate levels of salinity, and it also has several advantages compared to

other desalination technologies: it needs fewer pretreatment steps [2]; is relatively resistant to scaling, especially from silica [3]; can remove a wide range of ions regardless of their size [4]; is scalable and can be used both at very large and very small scales, making it a feasible approach in small and remote communities [5–7]; and has other applications, such as acid and base production and waste water treatment [8]. All of these characteristics have

GRAPHICAL ABSTRACT



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fostered the deployment of approximately 2.43 million m³/day of installed capacity from ED/EDR and electrodeionization (EDI) desalination systems [9]. In EDR, the basic ED process is enhanced by periodically changing the direction of ion transport by reversing the polarity of the electrodes. Additionally, when the polarity is reversed (typically every 15-30 minutes), automatic valves switch the flows of the dilute and concentrate streams through the cells [10]. The periodic change in polarity is done to prevent scaling and fouling problems on the surface of ion exchange membranes, and the polarity reversal adds a self-cleaning feature to the system [11,12]. However, polarity reversal reduces the lifetime of the electrodes [13].

Since ED/EDR is very extensively used, there is a great deal of interest in optimizing its performance, which is strongly affected by multiple factors. The most impactful factors are applied voltage, flow rate, temperature, initial feed composition, and IEM characteristics; a lesser but still significant effect can be observed from pH. To determine more precisely how all of these factors impact performance – especially desalination rate and the quality of the desalinated water – several valuable studies have been conducted. However, no comprehensive paper has been published, and some researchers have reported different findings from differently designed electro dialysis reversal systems.

Therefore, to synthesize previous research and reconcile superficially disparate findings, this review discusses, analyzes, and summarizes the results from the literature. Such a review can help researchers and technicians optimize electro dialysis reversal systems and improve the quality of desalted water.

2. Review and discussion

The EDR process is affected by controllable factors such as applied voltage, flow rate, temperature, membrane characteristics, and pH. Each of these factors is discussed in sequence in this paper. The EDR process can also be influenced by noise factors such as concentration polarization, ambient environmental temperature, fouling, and electrolysis effects. The noise factors are prohibitively difficult or costly to control, so optimization research has focused primarily on the controllable factors, along with the operational factors and feed composition. The results from such research are discussed in the following sections.

2.1. Voltage

Applied voltage significantly affects the rate of separation in the EDR process. Its effect on ion transport is very direct, as shown in the Nernst-Planck equation (Equation 1):

$$J_i = -D_i \left(\frac{dC_i}{dx} + z_i C_i \frac{F}{RT} \frac{d\phi}{dx} \right) + C_i V \quad (1)$$

where J_i , D_i , C_i , F , R , T , ϕ , z_i , and V are ion flux, ion diffusion coefficient, ion concentration, Faraday constant, molar gas constant, temperature, applied electrical potential, ion valence, and velocity, respectively [14]. This equation shows that the electro-migration of ions is affected by the electrical charge of the ions, the ion diffusion coefficient, temperature, voltage, and the concentration of the ions.

Since applying greater voltage increases current density, higher levels of voltage increase flux for all ions, resulting in a decrease in ion concentration on the membrane surface and a greater concentration gradient for each ion in the diffusion boundary layer, finally resulting in limiting current conditions at higher voltages. In line with this, researchers have frequently reported that voltage positively affects the removal amount [3, 15–19].

For example, Karimi and Ghassemi (2015) report the positive effect of applied voltage on the removal of different ions through pilot scale experiments. According to their findings, the removal of different ions, regardless of their characteristics and initial concentrations, is close to zero at very low applied voltages, especially at higher feed flow rates and lower temperatures [19]. At very low values of voltage, the applied voltage is not sufficient to overcome the resistance of the membranes and cause ion transfer; therefore, the removal of ions at very low voltage values is close to zero [20]. However, at very high applied voltages, the percentage of ion approaches 100%, especially at lower feed flow rates and higher temperatures [19]. Sadrzadeh et al. (2008) also show the positive effect of higher voltage on the separation percentage of Pb²⁺ in the ED process [21], although previous work shows that voltage variation has less of an effect on the removal of Pb²⁺ at higher voltage levels [22]. Overall, the positive effect of voltage on ion removal is well demonstrated.

However, as shown in Equation 1, the electromigration of ions is affected by many interacting factors, including ion mobility, which itself is affected by

ion characteristics such as diffusion coefficient and ion charge. Additionally, the effect of voltage can be mediated by interactions between counter-ions and the fixed and surface charges of the IEM. Therefore, since diverse ions have varied diffusion coefficients and may respond dissimilarly to the fixed and surface charges of the IEMs, various levels of voltage have different effects on different ions. The following paragraphs present examples from the literature, focusing on how ion mobility and ion-membrane interactions mediate the effects of voltage.

The interaction between voltage and ion mobility has been demonstrated in several investigations. For instance, Kabay et al. (2006) report that voltage change from 5 to 10 volts affects the removal of monovalent ions more than the removal of divalent ions, especially when a mixture of monovalent and divalent ions is present in the solution [16]. As expected, the removal percentage of monovalent Na⁺ experienced the maximum variation, and the removal percentage of K⁺ experienced the lowest variation.

This effect can be explained by differences in the mobilities of the involved ions. If the diffusivities of ions are assumed to be independent of each other under the dilute solution assumption, which is valid in the work of Kabay et al. (2006), then the mobilities of the ions used in the mixtures exhibit the order of K⁺ > Ca²⁺ > Mg²⁺ > Na⁺ [19], as calculated on the basis of the ions' diffusion coefficients, electrical charges, and temperature.

Faster removal of K⁺ ions compared to the removal of Na⁺ ions is also shown in the work of Demircioglu et al. (2002) [17], whose results, using batch desalination experiments for NaCl and KCl at the same operating conditions, confirm the positive effect on ion removal from the higher mobility of K⁺ ions compared to Na⁺ ions at the same applied electrical conditions.

Like ion mobility, ion concentration can change the effect of voltage. An example of this phenomenon is demonstrated in the work of Banasiak et al. (2007), which states that the kinetics of removal of F⁻ is more affected by voltage changes than is the kinetic removal of NO₃⁻. This effect could be attributed to ion mobility; however, Banasiak et al. explain that the observed effect is due to different initial concentrations of the two ions [18]. NO₃⁻ has a small hydrated size, and in the research of Banasiak et al., it also had a higher initial concentration, which caused its better removal even at lower voltage. The F⁻ ions, in contrast, had lower initial concentrations and experienced more concentration polarization at lower applied voltage compared to NO₃⁻ ions.

Other observable differences in ion removal are expected due to interactions between counter-ions and the fixed and surface charges of ion exchange membranes. For example, Balster et al. (2005) show that calcium transport through a cation exchange membrane (CEM), which has a positively charged coating, is low at lower current densities, but increases strongly at higher current densities [23]. This effect occurs because the 2+ charge of the calcium ions experiences repulsion with the positive surface charge of the CEM. At low voltages, this repulsion impedes calcium transport, but at higher applied voltages (higher current densities), the driving electrical force overcomes this repulsion, significantly reducing the influence of the repulsion on calcium ion transport.

To summarize, voltage has a positive effect on ion transport; however, its effect is influenced by factors such as ion diffusivity, ion charge, ion initial concentration, and interactions between counter-ions and the fixed and surface charges of IEMs.

2.2. Flow rate

Flow rate can affect the rate of ion removal in two ways. First, increased flow velocity can decrease the thickness of the diffusion boundary layer, producing a positive effect on the rate of ion removal. Second, increased flow velocity can decrease ion residence time in the EDR stack, causing a negative effect on the rate of ion removal. Research and theory have been unsettled about which effect is dominant and whether increased flow rate has an overall positive or overall negative effect on ion removal.

Theoretically, a positive effect on ion removal could occur because increasing the velocity increases the mixing of the solution, decreases the thickness of the diffusion boundary layer [24–28], and increases the dilute concentration on the membrane surface [15]. Consequently, the electrical resistance in the boundary layer would decrease at higher current density [29], resulting in higher rates of ion removal. This phenomenon can be explained using the current efficiency equation as follows:

$$\eta = \frac{zFQ\Delta C_i}{IN_{cp}} \quad (2)$$

where η , z , F , Q , ΔC_i , I , and N_{cp} are current efficiency, ion charge, Faraday constant (C.eq⁻¹), flow rate (cm³.s⁻¹), desired ion concentration difference

between feed and dilute streams ($\text{mol}\cdot\text{cm}^{-3}$), electrical current (A), and number of cell pairs, respectively. As depicted from Equation 2, by increasing the flow rate, the current efficiency should consistently increase, thereby increasing ion removal.

Conversely, a negative effect on ion removal could occur because at higher feed velocities, the ions might not have enough time to pass through the membrane, and could leave the ED stack before being transferred from the dilute stream to the concentrate stream [19].

Experiments, like theory, have produced conflicting results. Negative, positive, null, and both positive and negative effects on ion removal have been reported from increases in the feed flow rate.

Several researchers, utilizing continuous electro dialysis processes, have reported an adverse effect of increased velocity on ion removal and separation performance. For example, Aponte and Colon (2001) examine NaCl recovery from urine using the ED process, and report that at lower flow velocity – which causes higher residence time – greater NaCl removal from urine was achieved [30]. Additionally, Ali et al. (2010 and 2013) observe higher flow rates' adverse effects on total salinity removal and the removal of iron ions [20, 31]. Lee et al. (2013) also show that the removal of cationic species, such as Ca^{2+} , Mg^{2+} , and Na^+ , decreases as the feed flow rate increases [32]. Their results depict that the negative influence of higher flow rates is slightly more severe for the removal of Na^+ than for the removal of the divalent ions Ca^{2+} and Mg^{2+} , results which can be attributed to the lower mobility of the Na^+ ions. This means that the removal of Na^+ ions, compared to other ions, is more strongly affected by velocity changes. Numerous other researchers also report the negative effect of velocity on the separation of different ions [21, 22, 33, 34].

Conversely, some researchers, using batch EDR processes, have reported positive effects on ion removal from increased flow rate [3, 29]. Nevertheless, these studies do not recommend desalination at higher feed flow rates due to high hydraulic capital and operating costs [3].

In findings that obfuscate matters further, several investigators have reported both positive and negative effects from increased velocity, or have found no effect. Demircioglu et al. (2002) do not report any significant effect of flow velocity on the removal of K^+ and Na^+ ions in their experiments [17]. Sadrzadeh and Mohammadi (2009) report both positive and negative effects of velocity on current efficiency in sea water treatment using a small ED system at different flow rates and different feed concentrations [35]. They observe that by increasing the flow rate, the current efficiency increases up to a maximum point, after which it decreases as flow rate increases further. This observation explains that at lower flow rates, the removal of ions (ΔC) does not severely decrease with increases in the flow rate, due to the positive effect of velocity on the mass transfer rate. However, at higher flow rates, the adverse effect of flow rate on the ion residence time is severe, so the beneficial effects of flow rate (as shown in Equation 2) cannot compensate for the decreased ΔC in the equation. Therefore, the adverse effect of flow rate on current efficiency is seen after a specific flow rate. Additionally, Karimi and Ghassemi (2015) report the nonlinear effect of velocity at different levels, describing the effect qualitatively and quantitatively by developing a nonlinear model [36]. This nonlinearity is explained by the dual effect of velocity on ion removal, with positive effects from reducing the concentration boundary layer and improving mass transfer rate, but negative effects from the reduced residence time in the ED stack. Karimi and Ghassemi describe that, at lower flow rates, the positive effect of velocity on ion removal is dominant, while at higher flow rates the adverse effect of velocity on ion removal predominates [36]. They experimentally show that there can be a limitation to velocity's positive effect on ion flux in the ED/EDR batch process, which means that the desalination rate cannot be increased indefinitely by increasing the feed velocity [36].

Flow rate has also been reported to have different effects on different ions, but these effects have been explained by other factors. For instance, Walker (2010) reports a positive effect of feed velocity variation on the removal of sulfate and sodium, but an insignificant and barely detectable effect on calcium and chloride removal. This difference is explained by differences in the initial concentrations [3].

Many of the disparate results about the effects of flow rate on ion removal can be resolved by considering that the effect of superficial velocity (feed flow rate) has been experimentally studied through two approaches, one of which is more valid than the other. The first approach uses a continuous desalination process that does not recycle the dilute and concentrate streams; the second approach uses a batch desalination process in which dilute and concentrate streams are recirculated. The experimental studies conducted for the influence of superficial velocity using the batch ED process [3, 29] mostly report a positive effect of superficial velocity on the total removal of ions, especially at the beginning of electro dialysis. This effect can be considered as the effect of the number of solution passages through the electro dialyzer rather than the effect of velocity if the effect of desalination duration is not

taken into account: by increasing the superficial velocity of the feed solution in a batch desalination system, not only is the thickness of the boundary layer decreased, increasing ion transfer, but the solution is subjected to an increased number of passes through the desalination system. Consequently, when the same desalination period is considered for the same superficial velocity, the effect of the number of passages is ignored, confounding the results. Therefore, it can be concluded that the continuous experiments, which consistently report negative effects from higher flow rates, are more reliable for investigating the effect of superficial velocity.

Overall, it can be concluded that an increased flow rate has negative effects on ion removal in the ED/EDR process. In practice, though, this consideration can be balanced by flow rate's positive effect on the rate of desalination. On the one hand, increasing the flow rate can adversely affect the quality of the desalted water, but on the other hand, increasing the flow rate can increase the rate of production. Therefore, an optimal flow rate should be chosen based on the required quality and quantity of the desalted water.

2.3. Temperature

As feed temperature increases, theory suggests that ion separation increases due to the effects of temperature on ion mobility and the electrical resistance of the solutions. The Nernst-Einstein equation (Equation 3) shows that ion mobility – and thereby ion separation – improves with higher temperatures as a result of increased ion diffusivity [27].

$$u_i = \frac{|z_i|FD_i}{RT} \quad (3)$$

In Equation 3, u_i , z_i , F , D_i , R , and T are ion mobility, ion electrical charge, Faraday constant, ion diffusion coefficient, molar gas constant, and temperature, respectively. The diffusion coefficient changes linearly with temperature and inversely with viscosity, as shown by Equation 4 [37]:

$$D_i(T) = \frac{D_i(T_0) \times T \times \mu(T_0)}{T_0 \times \mu(T)} \quad (4)$$

where $D_i(T)$ and $D_i(T_0)$ are diffusion coefficient at temperature T and reference temperature T_0 , and where $\mu(T)$ and $\mu(T_0)$ are solution dynamic viscosity at temperature T and reference temperature T_0 , respectively.

Numerous empirical works have confirmed the theoretically expected results from changes in the temperature of the feed stream. Karimi and Ghassemi (2015) report that increased temperature positively impacts the removal of various ions [19, 38] and reduces the specific energy consumption of EDR [5]. Karimi (2015) also demonstrates that the removal percentage of ions is a function of the temperature in the form of the Arrhenius equation [38]. Additionally, Sadrzadeh et al. (2008), while using the ED process at different operating conditions, report that increased temperature positively affects the separation percentage of Pb^{2+} ions. This effect is explained by how increased temperature reduces resistance in the solution phase [21, 22], increases conductivity, and increases the ion diffusion constant through the membrane [22]. Other researchers have also shown the positive effect of temperature on the removal percentage of ions [42, 49-53].

Overall, since increased temperature improves the ion transfer rate and reduces the energy consumption of ED, using warmer feed water can reduce the cost and improve the performance of the ED process. However, to avoid incurring extra expenses, the feedwater should be heated using waste thermal sources.

2.4. Ion type

Ion charge and ion diffusivity can influence the performance of ED/EDR in terms of the removal of target ions. Ion charge affects ion removal through the interactions between counter-ions and fixed charged groups in the IEM matrix. Ion charge affects both separation and energy consumption in the ED/EDR process. For separation, divalent ions, such as Ca^{2+} and Mg^{2+} have a higher removal rate than monovalent ions, such as Na^+ [3]. This finding is attributed to the greater mobility of Ca^{2+} and Mg^{2+} compared to Na^+ ions in a mixed feed solution [32]. However, Zhang et al. (2011) reported the easier removal of monovalent ions using different types of IEMs [44]. This different behavior of the ED process can be attributed to the IEMs' characteristics, which will be discussed in Section 2.5 of this paper.

From results such as those reported by Zhang et al., we can see that ion type often affects ED/EDR performance. However, research conducted by Mohammadi et al. (2005) reports that the performance of ED in the removal of zinc, lead, and chromium is independent of the types of ions and IEM, as long as the IEMs have a minimum ion exchange capacity. In this research, ion removal is mostly a function of the operating conditions [39]. The ions

involved have different hydrated sizes, which theoretically could affect their removal, but the authors report that no such effects occurred.

2.5. Ion exchange membrane characteristics

Ion exchange characteristics are especially important in the ED/EDR process, and these characteristics can be controlled to some extent in the manufacturing process. Electrical resistance, ion exchange capacity, permeability, permselectivity between ions with and without the same electrical charge, and water content are particularly influential. The effects of these membrane characteristics are difficult to study independently because the characteristics correlate with each other. However, this paper isolates the effects of these features to the extent possible.

The electrical resistance of ion exchange membranes can be changed by changing fixed charge groups or membrane charge density, strongly affecting ion selectivity through interactions with different counter-ions. For example, Balster et al. (2005) show that the membrane charge density affects calcium ion transport through different commercial IEMs. IEMs with higher conductivities and charge densities have greater calcium transport. Balster et al. also relate that IEMs with lower charge densities have a greater tendency to remove monovalent ions selectively, and find that membrane charge density affects the removal rate of multivalent ions more than the removal rate for monovalent ions [23].

Ion exchange capacity (IEC), another key membrane characteristic, can be increased to produce higher membrane conductivity, less membrane resistance, and less potential drop. Although increasing the IEC can improve control over the counter-ion pathways in the membrane phase [45], higher IECs can cause more swelling in the IEM, resulting in less effective Donnan exclusion. This results in lower permselectivity (a higher diffusion rate of co-ions in the membrane) [42]. An example of the positive effect of higher IEC on the passage of the counter-ions in the ED process is shown by Mohammadi et al. (2004) using a typical CEM with a higher ion exchange capacity which demonstrates better removal of lead in the ED process.

Membrane permeability is another feature that affects the differentiated transfer of different ions in the membrane phase. In general, the mobility of the co-ions is less than the mobility of the counter-ions due to the Donnan exclusion. Additionally, the transfer of co-ions in the membrane phase is affected by the interactions with counter-ions. Therefore, the diffusion of co-ions in the membranes is much less than the diffusion of counter-ions [46].

If an IEM has higher porosity, its water content will be higher, so the movement of ions and the conductivity of the membrane will be elevated due to the presence of the continuous water channels. On the other hand, if an IEM has lower porosity, its fixed charge density will be low, which results in lower permselectivity for counter-ions. Higher porosity and larger pore size distribution can result in less effective Donnan exclusion, which means co-ions can enter the membrane phase like counter-ions. Therefore, in order to have a membrane with higher transport number for counter-ions and better permselectivity, an IEM with lower porosity, smaller pore size distribution, and higher fixed charge density is recommended [47]. The porosity of the membrane can be controlled during its synthesis by changing the drying time and thickness of the casting film [48].

Permselectivity between counter-ions through an IEM is determined by the ions' different affinities with the IEM and their different migration speeds through the membrane [49]. Permselectivity between ion *A* and ion *B* can be defined as shown in Equation 5 [49]:

$$P_B^A = \frac{t_A/t_B}{c_A/c_B} \quad (5)$$

where *t* and *C* are the transport number and concentration of ions in the membrane phase, and subscripts *A* and *B* are two ions: cations for CEM and anions for AEM, respectively. To standardize the permselectivity term for IEMs, Sata (2000) recommended using Cl⁻ and Na⁺ as reference ions for anions and cations, respectively [49], in order to establish a baseline for comparing the transport of other counter-ions with the same charge.

The permselectivity value for two counter-ions significantly depends on the preparation method used to fabricate the IEMs; different fabrication methods result in different ion exchange characteristics. The permselectivity of an IEM for different counter-ions can be improved via either surface or bulk modification [50]. One of the investigated methods to change the permselectivity of the IEMs is increasing the cross-linkage; however, increasing the cross-linkage can result in higher electrical resistance in the membrane. Therefore, it would be better to apply tight layers on the surface of the IEMs [49]. For instance, Sata et al. (1996) showed that the permselectivity of SO₄²⁻ vs. Cl⁻ is higher for membranes without polypyrrole modification compared to the anion exchange membranes (AEMs) with polypyrrole modification. They also showed that the transport number of SO₄²⁻ ions is

controlled by the polypyrrole layer, and is independent of the IEC of the membranes [51].

The hydrophilic feature of CEMs is also affected by a hydrophobic polypyrrole coating, which increases permselectivity for monovalent ions. This elevated permselectivity for monovalent ions occurs not only because of the smaller hydrated size of monovalent ions, but also because of the affinity of the composite membrane to the less hydrated counter-ions [52].

One of the other investigated methods that could affect the permselectivity of the membranes for different counter-ions is applying a polycation layer on CEMs in order to decrease their divalent cation permeability through intense repulsion between divalent cations and the polycation layer [53–55]. Furthermore, applying a polyanion layer on the AEM surface results in a strong repulsion force between divalent ions and the polyanion layer, which improves the permselectivity of monovalent anions, such as Cl⁻, against divalent anions, such as SO₄²⁻ [56]. Additionally, the applied polyionic layer not only decreases the permselectivity of multivalent ions, but also the permselectivity of the monovalent ions with lower hydrated radii, due to stronger electrostatic repulsion. For instance, the application of a polyanionic layer on AEMs can increase P_{Cl}^F because the hydrated radius of Cl⁻ ions is smaller than that of F⁻ ions, causing a stronger electrostatic repulsion between Cl⁻ ions and the polyanionic layer on the membrane [57]. There are some other methods to change the permselectivity of the IEMs, such as changing the hydrophobicity and hydrophilicity of the membrane, adding photoresponsive groups to the membrane to control the permeability of anions using photoirradiation, or applying a thermally responsive IEM. These methods were extensively discussed in the literature and are beyond the scope of this paper [49].

The mobility of the counter-ions and co-ions in the IEMs are affected by the nature of the polymer and the distribution of the functional groups in the volume where ions can move, a volume which is almost equal to the water content of the membranes. Therefore, the mobility of the ions is higher for membranes with more free water per charged site [46]. Consequently, the water content of the IEM affects the ion traffic and the ion pathways in the membrane phase by providing larger transfer channels for both the co-ions and counter-ions. Therefore, since high water content in IEMs increases permeability for both co-ions and counter-ions and decreases permselectivity for counter-ions, higher permselectivity for counter-ions can be achieved by IEMs with lower water content [45].

2.6. Feed Composition

Feed waters can be composed of different ions at different concentrations, and such differences can strongly affect the performance of ED/EDR systems. The removal rate of each ion and the total ion removal in the ED/EDR process are strongly contingent on the feed water's ion concentration and ionic composition.

Ion concentration, for its part, is positively associated with ion removal rate and total ion removal. As reported by Banasiak et al. (2007), who investigate how initial NaCl concentration affects performance, the removal rate increases at higher initial concentrations of NaCl [18]. Additionally, Karimi (2015) shows that the total removal of both monovalent and divalent ions increases at higher initial concentrations, because more ions are affected by the applied voltage at the same time [36, 38]. This result is further supported by Mohammadi et al. (2004), who report that increasing the initial feed concentration improves the removal percentage of Pb²⁺ using electro dialysis [22]. However, Mohammadi et al. find this effect to hold only across a limited range of concentration: increasing the initial concentration of Pb²⁺ from 100 ppm to 500 ppm had a positive effect on the removal percentage of Pb²⁺, but increasing the initial concentration to more than 500 ppm did not produce significant further effects. From these results, it can be concluded that increases in ion concentration tend to produce higher removal rates and higher total levels of removal, albeit in a manner inversely proportional to the initial ion concentration.

Although removal rates and total levels of removal increase with higher ion concentrations, Ali et al. (2010) show that the removal percentage of ions decreases as the initial concentration of ions increases between 500 and 3000 ppm. In this research, according to the equation for removal percentage of salt, increasing the initial concentration decreases the total calculated removal percentage of ions. However, the authors explain that at the same hydrodynamic and electrical conditions, the removal of ions was slightly increased by increasing the initial concentration, especially at higher applied voltages [31].

Ion composition also plays a large role. Above and beyond the impacts expected from ion concentration or the individual characteristics of constituent ions, mixtures of ions can interact to produce effects distinct from those observed in the presence of only a single ion type, especially when the mixture is composed of ions with different electrical charges. For instance,

when a single salt is present in the feed solution, monovalent ions are removed more readily than divalent ions [48, 54]. In contrast, when both monovalent and divalent ions are present in the feed solution, divalent ions have a higher removal rate [44, 48]. This result can be attributed to the strong electrostatic interactions between monovalent and divalent ions with opposite charges. Therefore, divalent ions are removed more, while electrostatic interactions impede the movement of monovalent ions: in mixed feed solutions, the monovalent cations are attracted to the divalent anions, and a similar phenomenon occurs for monovalent anions. This attraction "anchors" monovalent ions, hindering their removal [38]. However, this effect is most noticeable at lower voltages, and disappears as voltages reach higher levels [48, 55].

Multiple researchers have experimentally investigated the impacts of ion composition and concentration, and a broad range of findings are available. For example, Kabay et al. (2006) investigate the effect of feed composition in the EDR process using different 0.01 N binary mixtures for monovalent and divalent ion removal at room temperature and a constant flow rate of 1.6 L.min⁻¹. They show that at a lower voltage, monovalent cations are removed more efficiently in the presence of only monovalent anions than under conditions where divalent anions are present in the feed solution, because the monovalent cations are attracted by divalent anions more strongly, affecting their removal rate. However, this effect disappears at higher voltages [59]. It is shown that the removal of monovalent ions is greater than the removal of divalent ions when a single salt is present in the feed solutions [58], while monovalent ions are removed less than divalent ions when both monovalent and divalent ions are present in the feed solution, as reported by Lee et al. (2013) [32]. Karimi (2015) also shows that Na⁺ and Cl⁻ are removed better than divalent ions, such as Ca²⁺ and SO₄²⁻, when the mono salts of Na⁺ and Cl⁻ are used. However, the removal of divalent ions is greater when the multi-ionic solution is used as feedwater. This behavior is justified by the electrostatic interaction between ions, which is more impactful at lower applied voltages than higher applied voltages [38].

While Banasiak et al. (2007) were investigating the effect of the initial concentration of NaCl in the feedwater, they also demonstrated the efficiency of EDR processes for removing F⁻ and NO₃⁻ ions from brackish water. Their results confirm that, at higher initial concentrations of NaCl in the feed solution, the rate of removal is greater [18]. Ali et al. (2010) show that the total removal percentage of ions decreases as the initial concentration of ions increases. It is explained that at the same hydrodynamic and electrical conditions, the removal of salt is the same, while according to the equation for removal percentage of salt, increasing the initial concentration decreases the total calculated removal percentage of ions [31]. Karimi (2015) shows that the total removal of monovalent and divalent ions increases with increases in the initial concentration, because more ions are affected by the applied voltage at the same time [36, 38]. Additionally, Mohammadi et al. (2004) report that increasing the initial feed concentration improves Pb²⁺ removal using electrodialysis [22]. They explain that increasing the initial concentration of Pb²⁺ from 100 ppm to 500 ppm has a positive effect on Pb²⁺ removal, while increasing its initial concentration to more than 500 ppm does not significantly increase the removal of Pb²⁺ [22].

2.7. pH

Depending on specific conditions, pH may affect the performance of ED by impacting important parameters such as current efficiency, concentration polarization, and energy consumption. In general, acidic, low-pH environments result in preferential removal of anions, while alkaline, high-pH environments result in preferential removal of cations. This effect is explained by competition between OH⁻ and anions, and between H⁺ and cations [60].

The effect of pH on the ion separation of Pb²⁺ and NO₃⁻ was investigated by Abou-Shady et al. (2012), who focus on how pH affects current efficiency, concentration polarization, and energy consumption [60]. Additionally, these researchers show that pH variations influence the distribution of NO₃⁻ and Pb²⁺ ions on the membrane surface due to competition between the adsorption and ion exchange phenomena, thereby affecting the removal of those ions.

Despite the preferential removal of anions in low-pH environments, low pH levels reduce the removal of both monovalent ions (e.g., Na⁺ and K⁺) and divalent ions (e.g., Mg²⁺ and Ca²⁺) due to competitive removal between H⁺ and other cations, as revealed by the work of Kabay et al. (2003), who investigate the removal of these ions at pH levels of 2, 4, and 6 [58]. Additionally, at pH levels less than 4, higher energy consumption is required to remove ions, regardless of whether the ions are monovalent or divalent. As reported in this research, changes in pH do not affect the competitive separation of monovalent and divalent ions [58].

2.8. ED/EDR operating mode

The ED/EDR process can operate at different modes, such as batch, feed-and-bleed, and continuous. In a batch ED/EDR process, all three streams – feed, concentrate, and electrode rinse solutions – are circulated during the process operation. The batch mode can be used when a high degree of the desalination is desired; however, recirculating the concentrate stream for a long time can increase the scaling potential if the concentration of the less-soluble salts exceeds the saturation concentration [61]. Additionally, although increasing the flow rate can increase the rate of desalination (as discussed in Section 2.2), a higher flow rate can increase the pumping cost without improving the desalination rate if the flow rate exceeds a specific value in the batch mode. Therefore, an optimum flow rate should be considered in the batch ED/EDR process. In the feed-and-bleed process, either a fraction of the product or the whole product is discharged when the desired degree of desalination is achieved, while the concentrate stream and electrode rinse solution are recirculated.

In the continuous ED/EDR process, all three streams are single path, and they are discharged or sent to another stack for further treatment. However, in some continuous ED/EDR processes, only a portion of the concentrate stream is drained, while a big portion of it is recirculated. The drained amount of the concentrate is replaced by a portion of the feed. Recirculating a big fraction of the concentrate in the process increases total recovery. In general, the electrode rinse solution can be recirculated after passing a degasifier in all three modes, because its composition is not significantly affected by the process.

The selectivity of membranes under different operating modes has been explored through work such as that of Zhang et al. (2011), who compare the performance of the ED process in terms of the selectivity of AEMs using the two operating modes of batch and feed-and-bleed. Their experimental results show that SO₄²⁻ and HCO₃⁻ ions remain longer in the dilute solution in the feed-and-bleed mode compared to the batch mode [44]. Although this result was reportedly due to the different mobilities of ions in different operating modes, the mobility of ions can be attributed to the feed composition and ion characteristics rather than the operating modes.

In general, the choice of the operating mode mostly depends on the application of the process and the required quality and quantity of the product water, as well as the number of stages and many other operating factors.

2.9. Energy consumption in the EDR process

In general, the design, capital cost, and operating cost of the EDR process can be significantly affected by fixed parameters, such as the stack geometry and process application, as well as operating parameters, such as the desired quantity and quality of the product water, the membrane type, the applied voltage and current density, the feed water composition, the desired recovery rate, and the operating mode [62]. The total operating cost also includes the pumping cost, which is a function of the flow rate, the salinity of the feed water, the efficiency of the pump, the plant size, and the desired pressure. On the basis of the aforementioned factors, Tsiakis and Papageorgiou accurately formulated the operating energy and cost [63].

The capital cost of an ED/EDR desalination plant includes the cost of the ED/EDR stacks, a cost that, in turn, encompasses the costs of membranes, electrodes, electrical equipment, and hydraulics, such as pumps, valves, and plumbing. The largest portion of the capital cost is devoted to the membrane area required for the desired production capacity. This portion of the capital cost is influenced by the membranes' prices and the lifetime of the membranes; membrane lifetime depends heavily on the operating conditions and the quality of the feed water [62]. Lee et al. (2002) show the total cost of an ED plant as a function of current density and feed water salinity, and divide total cost into membrane costs and desalination costs [62]. The detailed breakdown of the capital and operating costs of the ED/EDR process has been evaluated in previously published papers [8, 63]. For example, Tsiakis and Papageorgiou (2005) formulated the specific energy consumption, pumping, and capital costs of an ED/EDR process. Based on the fact that the separation energy of the ED process is mainly affected by the operating parameters, which are the focus of this paper, this portion of the energy consumption in the ED/EDR plants is presented as follows in Equation 6 [63]:

$$E_{D,s} = \frac{u_s \delta a (c_s^d - c_s^f)^2 z^2 F^2}{\xi^2 L_s \beta} \left[\frac{\delta \ln \frac{c_s^d c_s^f}{c_s^d - c_s^f}}{\Lambda (c_s^d - c_s^f)} + (\rho^a + \rho^c) \right] \quad s = 1, \dots, S \quad (6)$$

where $E_{D,s}$, u , δ , a , C , z , F , ξ , L , β , Λ , and ρ are separation specific energy consumption (kWh.m⁻³), linear flow velocity in a stage (m.s⁻¹), thickness of a desalting cell (m), correction factor for effective membrane area, concentration (kEq.m⁻³), electrochemical valance, Faraday constant, current efficiency, calculated flow path required in each stage (m), effective area of membrane, equivalent conductance (m³.kEq⁻¹), and membrane resistance,

respectively. The subscript s is ED stage number, and superscripts c , d , A and C represent concentrate cell, dilute cell, and AEMs and CEMs, respectively [63]. As depicted in Equation 5, the separation specific energy consumption of ED/EDR is affected not only by the concentration and the quantity of the feed and product streams, but also by the feed composition. Kabay et al. (2006) report that the specific energy consumption for sulfate salts is less than that for some other salts due to sulfate salts' higher removal at low voltage [59]. Kabay et al. showed in their experiments that the specific energy consumption is affected mostly by applied voltage, feed pH, and the valences of ions [58]. The same research also showed that the specific energy consumption is not significantly affected by feed velocity variation [16], which shows that a significant portion of the energy consumption in an ED system is attributable to separation rather than hydraulics [16].

In addition to the effects of operating parameters on the removal of ions, the IEM structure and its physicochemical properties also influence the transport mechanism of the ions [64].

3. Conclusions

As discussed in this paper, increasing some parameters, such as applied voltage and temperature, improves the performance of the ED/EDR process. Up to a certain level, these two parameters can be increased to improve the desalination rate. However, when these parameters are elevated, they can increase costs and may damage the system. For increased temperature, the concern of heightened costs can mostly be avoided if a waste thermal source is accessible and if temperature can be kept to a level at which the membranes are not damaged.

Apart from applied voltage and temperature, the other main determinant parameter for ED/EDR performance is feed flow rate (superficial velocity). This factor has a dual effect on the electro dialysis process, making it more complicated than the other main parameters: increasing the flow rate increases the desalination rate and product quantity, but reduces the quality of the desalted water. Therefore, to minimize the cost of the process, it is crucially important to choose the optimum flow rate based on the required quality and quantity of the desalted water.

Secondary factors that nevertheless can significantly affect the performance and cost of ED/EDR are the characteristics of the IEM and the ion composition of the feed water. In choosing the type of IEM, decision makers should consider the quality of the feed water, the purpose for applying ED/EDR, and the number of electrical and hydraulic stages.

For a higher rate of desalination, ED/EDR operators can utilize higher applied voltage, higher temperature, and higher flow rates; higher flow rates must be used carefully, though, because the optimum flow rate must allow the required degree of desalination per ED/EDR stage. Flow rates are strictly limited in continuous ED/EDR systems, where increasing the flow rate decreases the quality of the product water and can require the use of more desalination stages. However, if increasing the quality and quantity of the product (desalted water) is a central objective, batch electro dialysis processes or a series of ED stacks can be employed. These processes can be operated at higher flow rates and largely avoid reductions in product quality, since increases in flow rate also increase the number of passes through the ED/EDR stack. Nevertheless, even in batch processes, the maximum flow rate is limited by the adverse effect of lower ion residence time at higher flow rates, which may increase hydraulic and electrical costs without improving the quality of the desalted water.

4. References

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