



Review

Electrodialysis desalination for water and wastewater: A review

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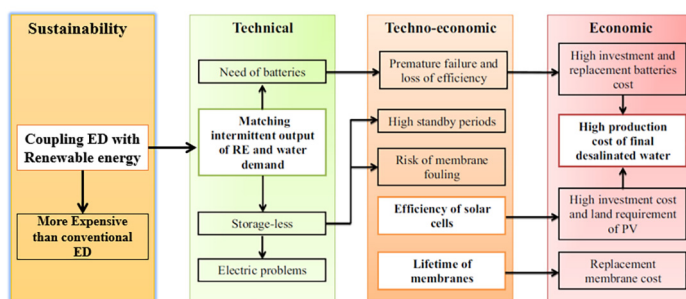


HIGHLIGHTS

- Anions and organics deposition restrict electro dialysis scaling up.
- The electro dialysis flexibility eases its coupling with renewable energies.
- Pilot scale studies bridge lab-scale electro dialysis application to full scale.
- Electro dialysis transfers waste treatment concept into water and nutrients recovery.

GRAPHICAL ABSTRACT

Sustainability and Performance Review of the Electro dialysis (ED) Desalination



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ABSTRACT

The principle of electro dialysis (ED) desalination was first presented in 1890 by Maigrot and Sabates, and its development into industrial scale started for more than 50 years ago. The operation of ED is driven by the development of ion exchange membranes produces high water recovery and does not require phase change, reaction, or chemicals. These advantages provide environmental benefits without the use of fossil fuels and chemical detergents. Whilst there are a number of reviews that have attempted to optimise ED performance for various applications, ED technology still has limitations involving scaling, membrane fouling, and permselectivity. In this paper, an extensive review of current studies on the process, principles, and setups of ED technology is given to deliver a comprehensive collection of all the main findings published on this technology so far. Also, it provides an overview of the possible sustainability approaches to be integrated with the ED process. The current developments and the sustainability of ED are critically examined for in-depth knowledge of what makes ED a promising desalination for potable water production. Finally, mathematical approaches to the design of ED process are briefly mentioned.

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1. Wastewater desalination

Water is necessary as a primary material for the life on planet earth. Today, the water environment is suffering from severe contamination, leading to severe destruction in the natural water courses. Therefore, a massive decreasing occurs in the growth of the environment, human sustenance, and economic status [1,2]. Domestic water and wastewater regulations for urbanisation usage have been enforced as the way to secure natural water sources from contamination. Despite that, deficient mediation of domestic wastewater still causes health issues, and environmental problems because of the poor removal of organic compounds, toxic and nontoxic matters, viruses, and pathogenic organisms [3–5]. Industrial wastewater contains heavy metals, oil, aromatic hydrocarbons, dyes, pesticides, and high organic and inorganic matters which comprises the most significant role in water pollution [2,6]. Accordingly, wastewater industries aim to sustain water resources by using high organic loading rate, low cost, simple, feasible and safe mediation process.

Desalination process is classified into membrane (non-phase change) and thermal (phase change) process. Membrane processes utilise membrane film as a physical barrier for pollutants separation from wastewater, while thermal desalination uses high energy to vaporise potable water from the feed and leave the contaminants as solids [7]. Mechanical vapour compression (MVC), thermal vapour compression (TVC), multi-stage flash desalination (MSF), multi-effect evaporation/distillation (MED), and solar desalination systems are examples of thermal desalination. Reverse osmosis (RO), and electro dialysis (ED) are instances of membrane desalination. After the year 2000, about 70% of the desalination plants are membrane processes because of their effectiveness and require lower energy and costs than thermal desalination. Globally, MSF, MED, ED, and RO are the dominant processes in providing fresh water for millions of people (see Fig. 1) [2]. Unfortunately, capacitive deionization (CDI) and membrane CDI (MCDI) are still in the laboratory scale [2]. Table 1 shows the approximate costs, benefits and drawbacks of different desalination technologies.

Since the 1950s, great advancements have occurred to the desalination technologies to achieve high organic loading rate, less fouling, higher flux and selectivity, and less costs. In fact, the growing global demand of fresh water is the main driving force of the rapid developments in the desalination technologies. The required energy 20 kWh in

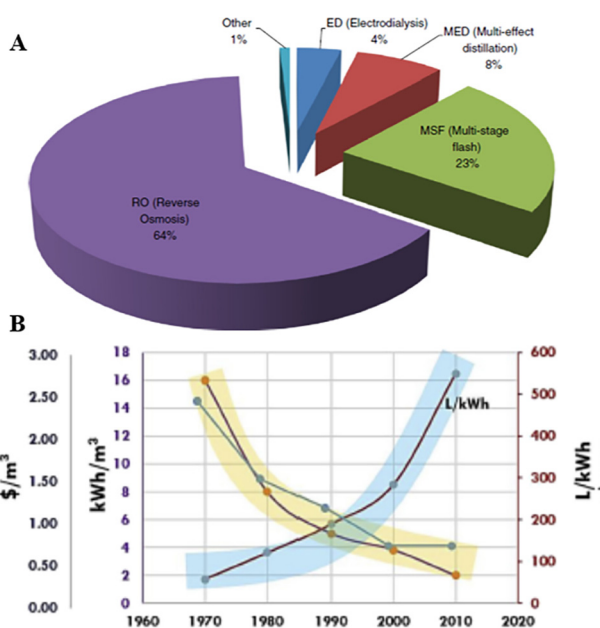


Fig. 1. A-Global desalination technologies share by capacity [16]; B- Development of desalination costs by year (adapted from ref. [7,17]).

Table 1
Desalination processes for water and wastewater.

Desalination Technology	Energy Consumption (kWh m ⁻³)	Advantages	Drawbacks	Ref.
Pressure-driven membrane operations	2–6	<ul style="list-style-type: none"> • Mature technology; • Consumes lower energy; • No phase changes; • Doesn't require chemical additives; • Easy to be scaled up and controlled; • Energy recovery devices are not required; • High removal of heavy metals; • Lower space requirement 	<ul style="list-style-type: none"> • Membrane fouling and durability; • High scaling; • Lower water recovery rates 	[8–10]
Forward Osmosis	Approximately 21	<ul style="list-style-type: none"> • No major scaling problems; • Does fast evaporation process 	<ul style="list-style-type: none"> • High energy consumption 	[11]
MSF	14–25	<ul style="list-style-type: none"> • Mature technology 	<ul style="list-style-type: none"> • High energy consumption; • Scaling issues 	[11]
MED	7–25	<ul style="list-style-type: none"> • Mature technology 	<ul style="list-style-type: none"> • Scaling issues; • Consumes high energy. 	[11]
MVC	20–25	<ul style="list-style-type: none"> • High removal rates of salt; • Able to generate high brine concentration; • Less susceptible to scaling; • High segregation of metals 	<ul style="list-style-type: none"> • Requires high capital costs; • Clogging and energy loss 	[11,12]
ED	1–12	<ul style="list-style-type: none"> • Able to handle high salinity up to 200 gm L⁻¹; • Low fouling; • Uses low-grade heat 	<ul style="list-style-type: none"> • Requires high energy; • Low water recovery 	[11]
Membrane Distillations	22–67	<ul style="list-style-type: none"> • 100% rejection; • No limitation on feed water; • Stationary operation without any moving parts; • Low energy consumption 	<ul style="list-style-type: none"> • Still in demonstration scale 	[9,13]
Adsorption Desalination	1.38			

1970 for 1 m^3 of fresh water production was minimised to 2.5 kWh in 2010 [7,14,15]. In addition, desalination cost got a significant reduction to less than USD 0.75 per m^3 in 2008 (as shown in Fig. 1). Many electro dialysis processes have been presented, based on the use of ion exchange membranes (IEMs), which remarkably increase the development of ED-related processes. Water Desalination report of 2017 showed a cumulative contracted capacity of desalination in 2016 of almost $100\text{ Mm}^3\text{ day}^{-1}$, with an average contracted capacity per year of $3\text{--}5\text{ Mm}^3\text{ day}^{-1}$ in the last 5 years. A slight part of the desalination growth is counted for the desalination of surface saline water, tertiary wastewater, and brackish water, with average capacities $50,000\text{ m}^3\text{ day}^{-1}$.

Desalination has been accepted as a promising process for wastewater treatment, but it still poses challenges (e.g., brine discharge, greenhouse gases emission, release of waste heat, prime quality electricity, high-grade thermal energy usage). The entire constructed desalination capacity is $47.6, 58, 65.2,$ and $74.8\text{ million m}^3\text{ day}^{-1}$ in the year 2007, 2008, 2011, and 2012, respectively, and it reached $97.5\text{ million m}^3\text{ d}^{-1}$ in the year 2015 (17% annual growth) [17]. Therefore, the energy demand has been increased as desalination capacity grows to fulfil the population need of fresh water. Desalination requires $10,000\text{ tons}$ of oil per year to mediate $1000\text{ tons (m}^3\text{)}$ per day of wastewater [7,18,19]. The high demand of energy for desalination process accelerates greenhouse gases emission such as sulphur dioxide (SO_2), nitrogen dioxide (NO_2), nitrite oxide (NO), and carbon monoxide (CO) [20]. Some desalination technologies are occupied with amounts of chemicals (e.g., cleaning chemicals, anti-corrosion, anti-foaming, biocides (chlorine) for controlling biological growth, and ...etc.) for saline water pre-treatment and post-treatment which deliver significant impacts to the environment [21,22]. Fortunately, the operation of ED does not require phase change, reaction, or chemicals. It just produces a desalinated stream and a concentrated stream. Hence, it is able to secure the healthy environment from contamination and prevents the extensive use of fossil fuels and chemical detergents.

For over 60 years, ED is an established technology in treating industrial wastewater, brackish water, municipal wastewater and used in drug and food industries, chemical processes [23–25], table salt production [26,27], electronics, biotechnology, heavy metals removal [28–30], and acids and bases production via its capability to remove ionic and non-ionic components under the effect of electric current. It has been driven by the development of IEM with enhanced electrochemical and physicochemical characteristics. The main advantages of ED are higher water recovery rates compared to RO, easy operation, long membrane lifetime, operation at high temperature, and unlike RO, it does not require extensive pre-treatment or post-treatment. Also, it can accomplish a selective separation of monovalent ions (e.g., NO_3^- , Cl^- , NH_4^+ , K^+ , Na^+) against multivalent ions (e.g., PO_4^{3-} , SO_4^{2-} , Mg^{2+} , Ca^{2+}) for the production of irrigation water, with the use of monovalent perm-selective IEM [26,31–35]. Researchers have reviewed the basic principles of ED in their articles [36–38], and it produced significant developments (e.g., electro deionisation EDI). Moreover, it originated photovoltaic (PV)-ED coupling system, thanks to the flexibility of ED process which can follow the oscillating behaviour of photovoltaic (PV) power generation [39]. Despite that, a lot of ED developments are still in the laboratory scale. Also, the global constructed capacity of ED desalination is only 4% (see Fig. 1).

There are about 16 studies specifically reviewed ED technology in different terms. Campione et al. (2018), Zourmand et al. (2015) and Galvanin et al. (2015) had discussed wide range of mathematical approaches as powerful tools to increase ED performance in terms of separation quality and energy saving [40–42], Scarazzato et al. (2018) and Andrew et al. (2018) applied ED application for a variety of wastewater [43,44], and Takagi et al. (2014) investigated IEMs [45]. Campione et al. (2018) found the higher cost of IEMs compared to RO membranes limits ED installation capacity, while other researchers [46] found IEMs fouling restricts ED scaling up and applications.

Furthermore, its application as a sustainable desalination has not been well described and endorsed. In fact, there is only one study reviewed the sustainability and the economic side of ED desalination combined with PV. It determined that the ED system is flawed by unsolved scientific problems and technical barriers [47]. Therefore, RO desalination with a slightly lower cost than ED has possessed 64% of the total constructed desalination [16].

In this paper, an extensive review of literature studies on the principles, setups and sustainability of ED process is given, with the aim of providing the reader with a comprehensive collection of all the main findings published on this technology so far. Unlike the review study of Fernandez-Gonzalez et al. [47], the current developments and the sustainability of ED are technically analysed for in-depth knowledge of what makes ED a promising desalination process for water and wastewater treatment. Finally, mathematical modelling approaches to the design and simulation of the ED process are presented.

ED applications have been so widely examined and mathematically simulated. However, a very little number of studies investigated and reported its weaknesses, while others didn't observe drawbacks in ED performance (e.g., "no fouling was observed"). This scientific paper systematically reviews ED principles and performance to show what makes it continuously fail to be a leading industrial desalination. Furthermore, it brings attentions to examine ED drawbacks that are unsolved since decades (e.g., IEMs fouling, high-cost IEMs ...etc). It's worth mentioning that drawbacks of IEMs were described in the 50s and the 60s as "decreasing membrane exchange capacity" [48]. Then, Smith et al. published a book chapter in 1972 acknowledging membrane fouling [49]. In contrast, Gregory et al. [50] reported in the same year that clogging was not a serious problem. To date, scaling up of ED desalination is limited due to IEM fouling [51].

The research strategy was to search for high-quality scientific papers published in scientific journals with the ScienceDirect database (see Fig. 2). In this research, we considered the period of publication 1950 to 2019 to track the evolution of electro dialysis. The following keywords were used:

- "Electrodialysis" AND "Wastewater";
- "Electrodialysis" AND "Industrial wastewater";
- "Electrodialysis" AND "Ion exchange membranes IEMs";
- "Electrodialysis" AND "Sustainability".

1.1. Electrodialysis for wastewater treatment

A voltage applied between cathode and anode electrodes passing through IEMs is used inside ED cell to separate charged species (i.e., ions) from uncharged matters and an aqueous solution. Therefore, ED is established as an electrically driven process. Inside ED stack, there are several anion exchange membranes (AEMs) and cation exchange membranes (CEMs), placed between cathode and anode electrodes. A spacer gasket is utilised inside ED stack to separate IEMs and create concentrate and dilute compartments. Electrodes are semi-separated by parallel membranes. Membranes work as a barrier to nutrients migration, which prevents or allows ions from passing in accordance with their electric charge. The electrolyte solution is circulated through electrode compartments, named as electrode rinse compartments.

When a feed solution enters ED stack, the applied electrical potential leads to a reduction reaction at the cathode, generating hydroxide ions in the cation compartment. To conserve electro-neutrality, a cation from the feed compartment will cross CEMs. Similarly, oxidation at the anode generates protons in the anion compartment, and anions will be recovered from the feed passing through CEMs. Hence, salts are removed from the feed solution to the concentrate compartment, producing dilute and concentrate effluents. For instance, cationic species like NH_4^+ , K^+ , and Na^+ are passing through CEM towards the cathode compartment. While, anionic components like PO_4^- , SO_4^- , and Cl^- are transferred to the anode section through AEM, which provides

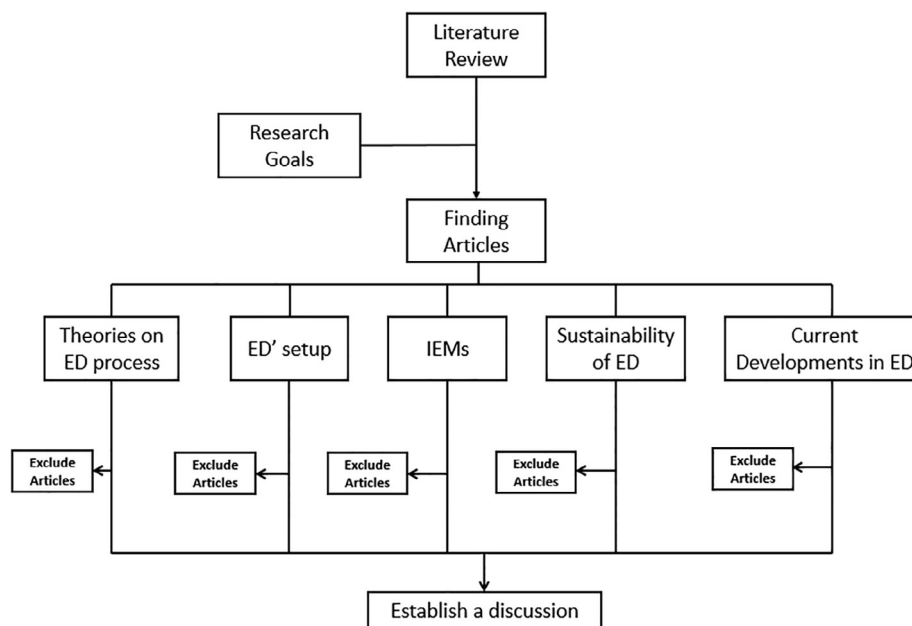


Fig. 2. Literature search process employed in this work.

electrical balance within the matrix [2].

Selective removal of specific components (e.g., Mg^{2+} , Ca^{2+} , Na^{+}) is achievable through utilising a selective IEM. Texas, New Mexico, California, and Arizona states have inhibited the use of groundwater and reclaimed water having high magnesium, calcium, and sodium rates for irrigation [52–54]. Therefore, ED with a series of selective membranes can remarkably reduce the unwanted ions concentration. In case of treating wastewater, ED is used for removing phosphorus, potassium, nitrogen, and organic and inorganic matters [55]. Several reports showed ED's high performance against iron compounds, cationic surfactants, nitrates, and divalent cations [26,56,57].

The principle of ED was found since 120 years, and its development into industrial scale started since 50 years ago [26,58]. Maigrot and Sabates presented the first model of ED for the first time in the year 1889, as a combination of electrolysis and dialysis for harmful substances removal that interferes in sugar manufacturing. It was employed for sugar syrup demineralisation in 1890 [59]. During 1900, Schollmeyer used the term 'Electrodialysis' in his patent for sugar syrup purification in a similar way [59,60]. After 1930, ion exchange materials based on polymer were manufactured for ED process. It was made in granules form, then membrane form. In 1939, Manegold and Kalauch employed ion selective membrane in ED stack comprising from three compartments [59]. Then, multi-sections ED was introduced in 1940 by Meyer and Strauss to reduce energy losses [60]. Later, Juda and McRae of Ionics Inc. in 1950, Wyllie and Patnode of Gulf Research in 1950, and Rohm in 1953 developed ED process to achieve low electric resistance, high selectivity, and stability [59,61]. Therefore, the rapid developments have qualified ED to be scaled up and used in the wastewater industry. It is proposed to use several hundred pairs of CEM and AEM in one ED stack arranged between electrodes for $100\text{--}20,000\text{ m}^3\text{ d}^{-1}$ with $1000\text{--}5000\text{ mg L}^{-1}$ salinity (total dissolved solids). Consequently, the first commercial ED was manufactured in the 1950s for brackish water desalination. In fact, the developments of ED application have not been only on wastewater treatment. In 1960, Asahi Co. demonstrated for the first time salt production via monovalent selective membrane [60]. Also, it has been applied in chemical, food, and drug industry as well as wastewater treatment, driven by IEMs evolution [62]. Fig. 3 shows ED's developments through time, from the first significant milestones to the recent applications existing so far (e.g., laboratory scale, pilot plant, commercialisation attempts). To date, ED is a worldwide recognised

technology and leading electro driven membrane process for salts production and ions separation using IEMs.

Driven by Donnan criteria of co-ion rejection, concentration gradient, and electrical neutrality, the applications of ED have been studied for rinse water treatment from plating industry of metals like Chromium [63], Cadmium [64], and Nickel [65]. Also, ED can recover and concentrate ions (e.g., $NH_4\text{-N}$, K , $PO_4\text{-P}$) from wastewater and supply the global demand of macronutrients [66–70]. This technique not only concentrates metals from rinse-waters, but it also maintains the quality of a plating bath [71]. Therefore, Ippersiel et al., (2012), Zhang et al., (2013), and Ledezma et al. (2015) have studied ED application to concentrate nutrients from waste streams [72–74]. Based on the use of IEMs, ED applications are still proposed among researchers to expand, promote, and optimise its application.

1.2. Electrodialysis setup

Frequently, ED system comprised of IEMs, power supply, auxiliary materials (Spacers, Electrodes, Gasket seal), and electro dialysis stack. The ED stack is closed with two end plates and compressed by bolts and nuts. Inside ED stack, there are series of IEMs, Electrodes, feed and concentrate compartments, spacers, and gasket gel. Also, it involves two electronic compartments, which converts ions current into an electrons current passing through IEMs, solutions, and the external electrical circuit. In the laboratory scale, ED includes few cell pairs, while, the industrial scale can reach up to several hundreds of cell pairs (e.g., 500 cell pairs). Structure of ED system consisting of dilute and concentrate compartments is shown in Fig. 4.

The repeating unit of ED, namely the cell pair involves CEM, dilute flow spacer, AEM, and concentrate flow spacer. AEMs and CEMs are arranged in an "Anode-C-A-C-A-Cathode" configuration between feed (dilute), and brine (concentrate) compartments. Along the surround of the channels, plastic spacers are installed with gaskets to guide the solutions flow, provide some mixing promotion in the channels, seal the channels, and keep a fixed intermembrane distance. The spacers create ducts for the hydraulic inlet, acting as manifolds for collecting/distributing the solutions from/to the channels. Thus, the feed channels are made by interposing between the membrane spacers. Accordingly, the applied electric potential gradient converts ions into electric current passing along ED stack. In the past, NaCl was used as the electrolyte, yet

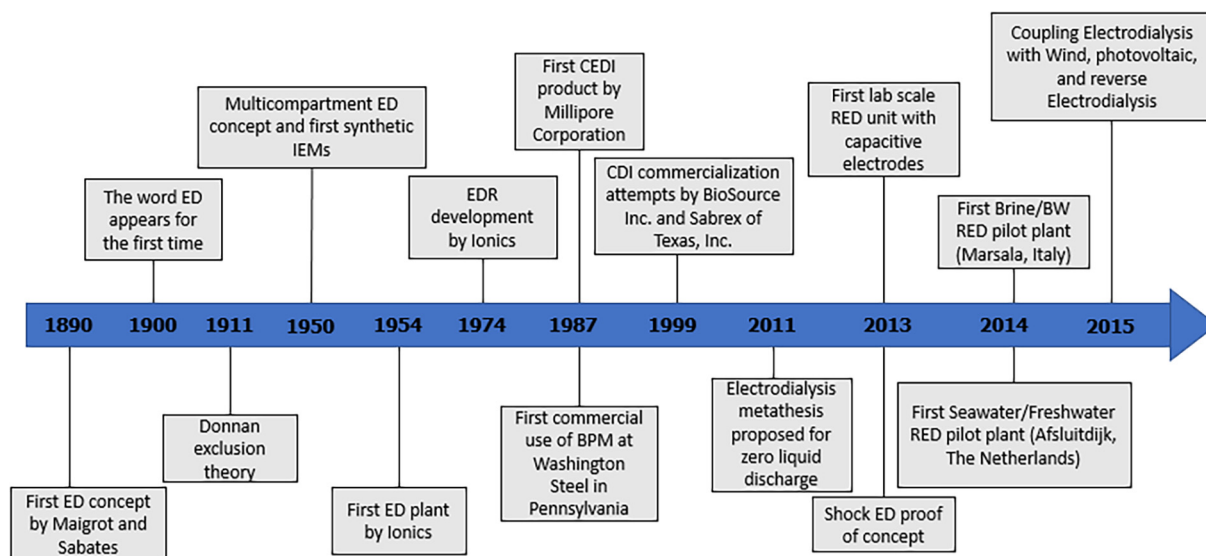


Fig. 3. Timeline of the most critical developments for ED and related processes (adapted from ref. [40]).

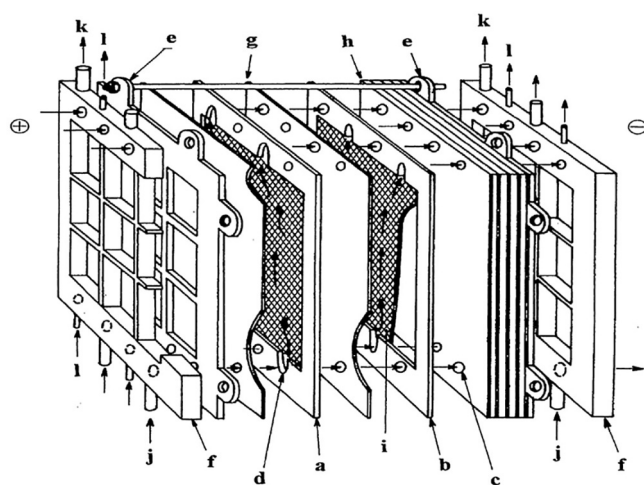


Fig. 4. Structure of electrodesalination bench system (adapted from ref. [38]). (a) Desalting cell; (b) concentrating cell; (c) duct; (d) slot; (e) fastening frame; (f) feeding frame; (g) cation exchange membrane; (h) anion exchange membrane; (i) spacer; (j) feeding solution; (k) desalted solution; (l) concentrated solution.

it produces Cl_2 in the anodic section [75]. Therefore, SO_4^{2-} is proposed to avoid the consequences to the electrode in the anode compartment, and 0.1 M Na_2SO_4 rinse solution is introduced to prevent toxic gas production [40].

ED system can be operated in batch mode to small-scale applications [75], or continuously to industrial scale [36]. Indeed, a single stack cannot produce the desired product characteristics, so a multistage system is implemented. Researchers use ED unit consisting of 2–3 chamber to carry out laboratory scale experiments. It can involve 10 membrane pairs in 5 compartments. In laboratory scale units, the active area of a membrane ranges 0.01–0.06 m^2 [76], while it reaches 1 m^2 for industrial scale [77]. The distance between membranes in ED stacks ranges between 0.3 and 2 mm [26,78]. The distance between the electrode and the end membranes is 0.001 m. Spacers made of polypropylene/plexiglass have an equal area to the effective area of the installed IEMs with 0.42–10 mm of thickness. A gasket is placed between each membrane and spacer with 1 mm of thickness. Thus, the channel would be 3 mm in thickness if the spacer and the gasket have 1 mm of thickness per each. The electrodes can be made of steel 314, titanium, titanium coated with ruthenium oxide, titanium plated with

iridium, platinum-plated iridium, titanium coated with titanium and ruthenium oxides ($70\text{RuO}_2/30\text{TiO}_2$), or graphite [65,79]. The feed (0.5–10 L), concentrate (0.5–10 L), and electrolyte (0.5–5 L) are circulated ($3.5\text{--}200\text{ L h}^{-1}$) through ED stack using a dual-head peristaltic pump. A power supply is installed to deliver a fixed current to the ED rig (e.g., 22 A and 36 V for NaCl removal, 65.6 A m^{-2} for molasses effluent, 39.1 A m^{-2} for the lignocellulosic effluent, 17.2 A m^{-2} for the sugar cane juice) [80]. It has been proposed to apply a higher voltage to the polarising electrode to deliver 1–1.5 V for each membrane pair (Cell) in ED stack [81]. Typically, the applied voltage to laboratory scale unit ranges from 7 to 30 V. The operation of the ED process can last to different durations (e.g., 24 h, 30 h, 55 h, 72 h).

Quality assurance can be done by using mathematical models examining ED performance in terms of water recovery (%), process capacity, and current efficiency (CE), as shown and explained in 1.3 Principles of Electrodialysis Application. While, quality control can be achieved experimentally by finding the effluent quality (e.g., using Hach spectrophotometer to find the chemical oxygen demand concentration, using ion chromatography to find ions concentration) as shown in 1.5 Feedwater & Discharge Quality. In 1986, Itoh et al. studied ED treatment for bio-refinery waste streams in terms of molecular weight and charge [82]. It was found that inorganic ions are easily separated from the feed, unlike organic ions. Dilute involves most of the organic material, while the brine contains inorganic ions. Mateusz et al. (2017) utilised a laboratory scale ED setup consisting of two chambers and having 1 anion exchange and 1 bipolar membrane for the production of Alpha-ketoglutaric acid [79]. The effective area of the membranes was 64 cm^2 , and the spacing thickness was 10 mm. Moreover, Merkela et al. (2017) used ED application to recover sodium hydroxide from the high alkaline solution [83]. Later, in 2018, Andrew et al. performed nutrients recovery from wastewater via pilot scale ED [44]. The rig had a total effective membrane area of 7.2 m^2 in 30 cell pair. In 72 h of functioning duration, it consumed $4.9 \pm 1.5\text{ kWh kgN}^{-1}$ for delivering $76 \pm 2\%$ of efficiency in terms of cations transport. The produced effluent is examined through ion chromatography to find the ionic concentration, and total organic carbon (TOC) analyser to measure the impurities.

1.3. Principles of electrodialysis application

Developed and refined dimensionless models of ED system were presented in the late 1960s and the 1970s. In 1968, Sonin and Probstin used laminar flow regime to develop an empirical equation for

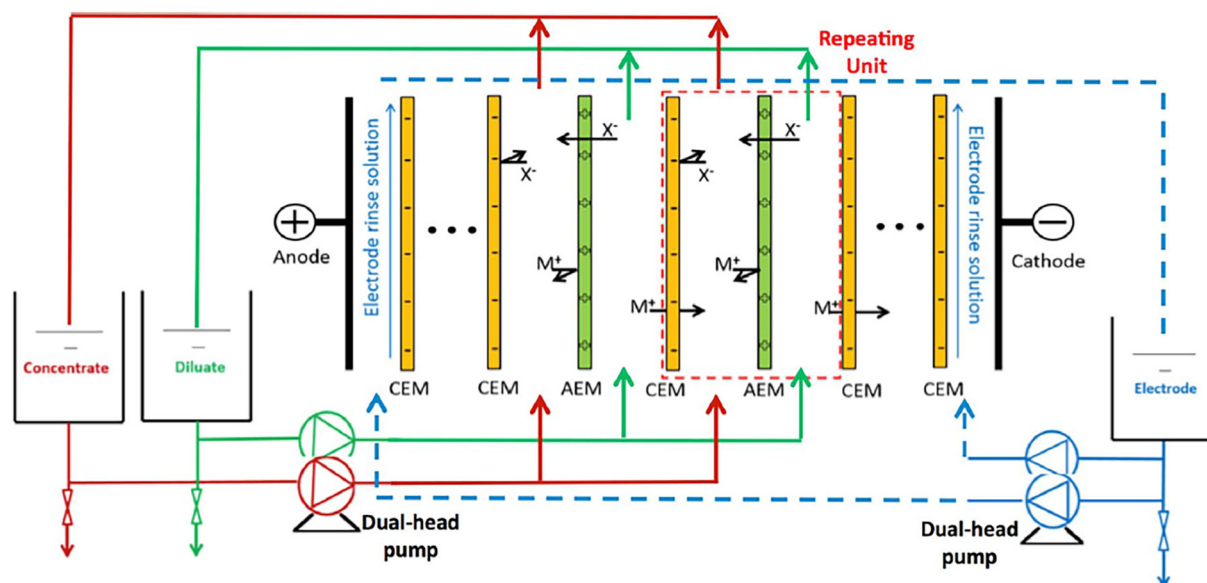


Fig. 5. A detailed scheme of the ED system (adapted from ref. [109]). X^- , and M^+ refer to negatively and positively charged ions pass through AEM and CEM to achieve charge balance between the cells.

dimensionless current density and used two regions (developing and fully developed) for concentration diffusion layers [84]. In 1971, Kitamoto and Takashima used a modified version of Peclet and Stanton numbers to present a dimensionless model that relates the current density to the applied voltage and solution flow hydrodynamics. Their results showed that the limiting current density has a weak relationship with the IEM type [85]. Leila et al. (2016) found the results of Kitamoto and Takashima are logical because diffusion is the controlling condition in the ionic mass transfer [86]. In 1972, Probst et al. studied zero and full polarisation. In addition, Probst et al. reported the most accurate correlation between the mean current density and the limiting current density for minimal levels of salt removal [87]. In 1977, Huang presented empirical models for ionic mass transfer at limiting current for laminar and turbulent flow [88]. Kuroda et al. (1983) found that Sherwood number (Sh) is independent of spacers type in the ED stack, and it's a function of $Re^{0.5}$ [89]. Remarkably, the empirical model presented by Sonin and Isaacson in 1974 was used by kraaijeveld et al. (1995) for the separation of amino acid [90,91]. In 2005, Fidaleo and Moresi compared their empirical results with the models of Sonin and Isaacson (1974) and Kuroda et al. (1983) and found that Sh is a function of $Re^{0.5}$ as Kuroda et al. (1983) had established, and not $Re^{0.3}$ [92].

In the last 8 years, several equations were reported for Sh by Shaposhnik and Grigorichul (2010) to explain the mass transfer rate in ED system, and to compare the ionic mass transfer rate between spacers free channels and channels having inert spacers [93]. Mostly, the developed Sh equations are mainly used to find the limiting current density. Fidaleo and Moresi (2005, 2006, and 2010) used empirical results of ED with various feeds to present a relationship between Re and Schmidt number (Sc), as independent variables and Sh as dependent variable [92,94,95]. In 2014, Mitko and Turek investigated concentration polarisation, and mass transfer coefficient in the ED stack by using Graetz-Leveque equation for laminar flow [96]. Later, Tadimetri et al. (2015) empirically established Sh equation to find the ionic mass transfer of Cl^- and Ca^{+2} from a sugar solution feed in a batch operated ED process [97].

Leila et al. (2016) critiqued the whole recent development in ED models because it was focused only on the limiting current density [86]. To the best of our knowledge, Probst et al. (1972) and Sonin and Probst (1968) investigated partial and full concentration polarisation impact for very small levels of salt removal besides the limiting current density condition. Despite studies have developed ED stack in

terms of membrane life, separation quality, and energy consumption, industrial EDs are still supplied with a higher/close voltage to the limiting current to receive the wanted performance without damaging the membrane films. Several studies reported that the use of high voltage leads to the rapid migration of ions through IEMs and results in ions accumulation [98]. Leila et al. (2016) found that using lower voltages than the limiting current density is acceptable to avoid the expected risks to the ED system [98]. Also, Meng et al. (2005) recommended applying a maximum current in range of 70 to 80% of the limiting current density [99].

The developed mathematical models have not been only conducive for understanding ED process, but also have been major functions in developing the engineering practices of the ED system. In 2018, Yu-Xiang et al. reported that the conventional ED is still playing essential roles above all the drawbacks (e.g., defects in the concentration diffusion and electromigration, and proton leakage of AEM) [100]. Also, Yu-Xiang et al. presented a model to find the concentration evolution, which can help to find the energy consumption. Finally, knowing a wide range of parameters in the ED system (e.g., power consumption, and current efficiency) is essential and vital for the application of an industrial ED scale.

The success of ED operation requires a mathematical expression as a powerful tool to examine and optimise its performance. A large number of literature introduced various ED mathematical models for salt removal, and it's based on mass balance of separation system [101], irreversible thermodynamics formalism [102–104], Stefan–Maxwell theory [90], Nernst–Planck equation [92,105–107], and semi-empirical models, i.e. involves empirical results besides the theoretical parameters [108]. Fig. 5 illustrates ED setup involving the dilute, cathode, anode, concentrate, CEM, AEM, and the movement of negative and positive ions, i.e. X^- and M^+ , in the ED stack.

Inside the ED stack, diffusion, migration, and convection are the three major forces which transport a wide range of negatively and positively charged ions [100]. Eq. (1) expresses diffusion and migration forces mathematically:

$$j_i = -D_i \frac{dC_i}{dx} + \frac{t_i I}{Z_i F}, \quad i = +, - \quad (1)$$

where, D_i ($m^2 s^{-1}$) is diffusion coefficient of i in solution, C_i (M) is the concentration of i , I (A) is the current, t_i is the transport number of i in membrane, and F is Faraday constant.

The ion diffusion coefficient (D_i) can be measured with the help of the Stokes-Einstein equation [100]:

$$D_i = \frac{RT\lambda_{i,m}^\infty}{|Z_i| F^2} \quad (2)$$

Herein, R is the universal gas constant, F is Faraday constant, T is the test temperature, and $\lambda_{i,m}^\infty$ ($S\cdot cm\ mol^{-1}$) is the limiting ionic conductivity.

Organics and inorganics leaving the dilute stream can be estimated with Eq. (3):

$$(A^- + X^-) = n_{OH^-} + M_{out}^+ - M_{in}^+ \quad (3)$$

where A^- and X^- are the equivalent number of organic and inorganic anions, which leave the dilute stream, n_{OH^-} indicates the equivalent number of hydroxyl ions that had entered the dilute compartment, M_{out}^+ represents the equivalent number of cations migrated from the dilute compartment by imperfect AEM selectivity, and M_{in}^+ is the equivalent number of cations that migrate to the dilute section due to the low selectivity.

The capacity of ED process (C_F) can be found with Eq. (4) [110]:

$$C_F = \frac{m_F}{N \cdot A \cdot t} \quad (4)$$

where t is the total time, N is the number of membrane pairs, A is the active surface area of membranes, and m_F is the mass of dilute stream.

Eq. (5) presents the mean flow velocity (V) inside a channel having inert spacers [111].

$$V = \frac{Q}{N \cdot \delta \cdot b \cdot \varepsilon_{sp}} \quad (5)$$

where δ is the spacer thickness (cm), Q is the volumetric flow rate ($mL\ min^{-1}$), ε_{sp} is the spacer porosity, and b is the compartment width. Xuesong et al. (2018) defined the linear velocity (u) in different equation [112]:

$$u = \frac{Q_d}{n \cdot h \cdot W} \quad (6)$$

where h and W are the height and the width of the feed chamber flow channel, n is the number of IEM pairs, and Q_d is the volumetric flow rate.

The hydraulic retention time (HRT) is estimated with Eq. (7):

$$HRT = \frac{n \cdot h \cdot W \cdot L}{Q_d} \quad (7)$$

where L represents the length of the feed flow channel in the ED stack.

The water recovery (%) is the ratio between the feed (Q_f) and final dilute (Q_d) which can be estimated with Eq. (8):

$$Recovery(\%) = \frac{Q_d}{Q_f} \times 100 \quad (8)$$

Dorota and Piotr (2018) used Eq. (8) in terms of mass to find zinc recovery (R_{Zn+2}) [113]. Also, they used a similar equation to find the retention percentage of iron.

Xuesong et al. (2018) used conductivity parameter ($mS\ cm^{-1}$) of the feed (C_f) and the dilute (C_d) to find the overall salt removal, Eq (9) [112]:

$$Conductivity\ cut(\%) = \left(1 - \frac{C_d}{C_f}\right) \times 100 \quad (9)$$

In order to evaluate the performance of ED process, Scarazzatoa et al. (2018), Tatiane et al. (2016), and Arthur et al. (2018) have used percent extraction ($E\%$), percent demineralization ($D\%$), and percent concentration ($PC\%$), according to Eqs. (10)–(12) [83,110,114]:

$$E\% = 1 - \frac{C_i^j}{C_0^j} \quad (10)$$

$$DR\% = 1 - \frac{EC_f}{EC_0} \quad (11)$$

$$PC\% = \frac{C_i^j}{C_0^j} - 1 \quad (12)$$

where C_0^j and C_i^j are initial and at a given time ion concentration, respectively. Solution conductivity at the beginning and the at a specific time are presented in the equation as EC_0 and EC_t , respectively. Merkela et al. (2017) used those equations in investigating the recovery of sodium hydroxide from the high alkaline solution [83], Benvenuti et al. (2016), and Scarazzatoa et al. (2018) employed it for studying ED performance towards chemical recovery (e.g., nickel and cyanide) [43,114].

Salinity (γ , mEq) is estimated for any solution/stream in ED stack with Eq. (13):

$$\gamma = \frac{m(K)}{39} + \frac{m(Na)}{23} + \frac{m(K)}{27.5} \quad (13)$$

where m is the mass of various ions. Thus, desalination rate (φ) can be found with Eq. (14):

$$\varphi = \frac{\gamma_0}{\gamma_d} \times 100\% \quad (14)$$

where, O and d represent feed and dilute, respectively. γ is the salinity amount. After estimating salinity, Eq. (15) can be applied to find the current efficiency (η):

$$\eta = \frac{F \cdot (\gamma_0 - \gamma_d) \cdot 10^{-3}}{It} \quad (15)$$

where, it is the total actual consumption of electricity, and F is Faraday constant ($96,485\ C\ mol^{-1}$).

The process of developing a mathematical model to perform a better understanding of the impact of various conditions on the ED process is considered a complex. Although, the current models have assisted industries to develop and built an industrial scale of ED. The electrical energy should be well determined to define the energy consumption of the process. Thus, the limiting current density is estimated to avoid inefficiency and predict the limiting current. The feed flow rate and its concentration impact the limiting current density [115,116]. Steven et al. (2011) mentioned that flowrate has a minor impact on the limiting current density at high salt concentrations [117], but it has a great effect at low salinity. Park et al. (2006) showed that high flow rates reduce the thickness of the diffusion boundary layer, leading to an increase in the limiting current density [118]. In 2010, Długołęcki et al. confirmed the findings of Park et al. and added that at high flow rates, the limiting current density is independent of the membrane performance [115]. In 2018, Yu-Xiang et al., reported an empirical equation to present the relationship between solution concentration and the limiting current [100]. The desalination energy required for the ED process is connected to the current density, which can be estimated with Eq. (16):

$$Current\ Density = \frac{I}{S_1} \quad (16)$$

where, S_1 is the membrane surface area of a single pair, and I is the current. The current efficiency (CE) can be defined as the ratio of moles transferred with time compared to the Faraday of electricity passed through ED cell [44]. Chindapan et al., (2009) reported a different form of the equation to estimate CE and defined it as a measure for the quality of i transport through ED stack [119]. Also, other researchers have presented different CE equation based on ED's application [79,113,120,121]. CE of ion (i^-) is measured with Eq. (17) [45,121]:

$$CE = \frac{Q_i}{Q_{applied}} \quad (17)$$

$$Q_i = \frac{|Z_i| \cdot F \cdot \frac{\Delta m_i(t)}{M_i}}{N} \quad (18)$$

$$Q_{\text{applied}} = \int_0^t Idt \quad (19)$$

where Q is the electric quantity, Z_i is the charge of i , N is the number of cells, $\Delta m_i(t)$ is the change of the mass of i , F is Faraday constant, M_i is the molar mass of i , and t is the total time. The total CE (%) can be found with Eq. (20):

$$TCE = \sum_i^n CE_i(\%) \quad (20)$$

where n represents the number of ionic species (e.g., NH_4^+ , Mg_2^+ , Ca_2^+ , Na^+ , K^+). Kenji et al. (2015) had investigated the transport of ions in IEMs by electrophoresis, molecular diffusion, and convection forces with using Nernst Planck equation [105]. The mathematical models were solved by Runge–Kutta–Gill method and the results successfully match the experimental data. Sizat et al. (1999), Chaabane et al. (2007), and Zabolotsky et al. (2002) used Nernst–Planck equation to explain the transport of ion along the axis perpendicular to the membrane surface [122–124]. However, the downstream of the dilute channel decreases in its compartment and increases in the concentrate compartment because of the osmotic pressure [105]. Therefore, an equal flow rate is required to eliminate possible flaws in ion concentration.

For years, researchers have used various models to understand ED process, and they found it consistence and close to the empirical results [125]. For instance, modelling the concentration change in the ED stack is a trend investigation, and it can be explained with ideal Donnan equilibrium and Nernst-Planck equation [126]. Remarkably, the modelling delivered good accuracy results compared to the experimental findings. Anyway, researchers have not made one generalized model to explain ED process, so they had to produce a specific model able to explain their ED setup, like Kaláb and Palatý [127], Ortiz et al. [101], Gong et al. [102], Fidaleo and Moresi [92,95,128], Kraaijeveld et al. [90], and Tanaka [129]. The most used theory is Maxwell-Stefan equations. Audrey et al. (2017) used a set of equations to find important parameters and found 2% error in the voltage measurement, 5% error in the current, and 6.4% error in the concentration level [80]. Moreover, Wright and Winter (2014) improved the analytical model of Ortiz et al. (2005) for studying batch ED performance in terms of power consumption, concentration, and operating time [101,130]. In 2015, Zourmand et al. used Nernst-Planck and Navier-Stokes equations to develop an isothermal, stationary, and bidimensional model to investigate ion mass transport in ED stack considering the polarisation phenomena [42]. The model predicted velocity distribution, electric potential, and concentration changes and it was solved by the finite element method and CFD [131].

1.4. Electrode materials

Studies have investigated the quality of various electrodes (e.g., activated carbons, alumina and silica nanocomposites, graphene, mesoporous carbons, carbon aerogel, carbide-derived carbons (CDCs), CNTs, and CNFs) [132–135]. Vermaas et al. (2013) investigated the use of capacitive electrodes instead of conventional ones [136]. It has an active layer of carbon (e.g., coal and wood) to adsorb/desorb ions, and current collector to release/capture free electrons. Thus, it converts an ionic current in the ED stack into an electric current passing through the external electrical circuit without a redox reaction. Moreover, it does not require toxic materials (e.g., Cl_2), but carbon layer saturation is their main drawback. Coal and wood are sent to a pyrolysis process to produce activated carbon with high porosity. Then, a poly (vinylidene fluoride) binds the activated carbon to maintain its structure. If we compare activated carbon with other electrode materials (e.g., carbon

aerogel, carbon cloth, carbon felt, carbon paper), we find activated carbon is more suitable because of its high surface area and pore size distribution.

Oxygen-depolarised cathode (ODC) is another electrode introduced to minimise the energy consumption of hydrochloric acid electrolysis [137]. It delivers oxygen reduction (i.e., hydroxyl ions generation) instead of hydrogen generation at the cathode and saves 30% of the energy demand. Other studies found the distance between the membrane layer and cathode, and current density has a major impact on the electrode performance.

1.5. Feedwater & discharge quality

The industrial scale of ED can process 2500–3000 mg L^{-1} of salinity. Thus, it has been applied widely to recover 85% of fresh water from low salinity feed, i.e. 3000 mg L^{-1} of TDS. The large scale of ED treats hundreds of ML day^{-1} , and it consumes high energy up to 6.4 kWh kg^{-1} for operating high salinity feed [30,33,34]. Burn et al., (2015) published a review study about desalination techniques and found that ED technology has been employed to process brackish water having salinity more than 15,000 mg L^{-1} [138]. Each stage of ED removes up to 50% of the contaminants, so several stages have been installed to deliver the required quality. Important factors are always considered for ED process like desalination ratio, water recovery, current efficiency, energy consumption, and operating cost. Also, feed salinity, and membrane type effect the efficiency of ED process.

In Morocco, ED is occupied for seawater desalination (e.g., 10, 6, 4, 2, 0.9 Ds m^{-1}) to deliver a high ratio of water recovery [139]. Sadyrbaeva (2016) had utilised ED to remove chromium (VI) from aqueous solution, and the process successfully removed 99.5% [140]. Another researcher treated wastewater having 2 to 6 μm of particle size with industrial scale ED. The efficiency increased 10 times when the electric current (150 V cm^{-1}) was applied, and then the pulse current (125 V cm^{-1}) improved the permeate rate [141,142]. In 2012, Buzzi et al. investigated the possibility of applying ED to treat acid mine drainage and recover water [143]. It was found that ED can remove metallic cations (e.g., Fe, Al, Mn, Pb, Zn, Cu) with greater than 97% of efficiency. Luiz et al. (2017) obtained 96% salt removal from bio-refineries wastewater by bench-scale batch operated ED. The wastewater had 380 g COD L^{-1} , 72.4 mS cm^{-1} conductivity, and 5600–572,000 PtCo colour level. Another group of researchers fed ED with cyanide-free copper plating wastewater, and it delivered 5 to 6 times concentrate stream with 80% extraction, and 90% demineralisation [43]. Luiz et al. (2017) and Scarazzato et al. (2018) did not observe membrane clogging, Buzzi et al. (2012) highlighted membrane clogging issue from ion precipitation at the IEMs surface, Luiz et al. (2017) considered fouling important if ED has been scaled up, and Valero et al. (2010) and Campione et al. (2018) considered ED technology is not affected by scaling phenomena, but it's important when the feed is rich in low solubility salts such as CaSO_4 and CaCO_3 . In short, such studies on membrane fouling in ED applications has not been enough to scale it up, therefore 64% of the industrial desalination is RO.

1.6. Ion exchange membrane (IEM)

IEMs are arranged in series and parallel between two electrodes inside the ED stack to separate two streams and perform a selective mass transport between the feeds. Moreover, IEMs are ion-selective membranes in which ion mass transport takes place by a concentration gradient i.e. osmosis, or electrical potential i.e. electro-osmosis. The performance of IEMs is based on the diffusion of ions, and electrostatic interactions (sorption) [144]. IEMs is a key component allowing ED to be utilised for chemicals conversion into electricity (energy conversion and storage), hydrogen production (water electrolysis), and chemicals synthesis (electrochemical reaction). It has been used in ED applications to deliver high desalination rate and high water recovery. However, it is

not preferred for the industrial scale due to high energy consumption, poor permselectivity and high resistance.

Conventional membranes are characterised based on the material and the pore dimension, while IEMs are based on the charged molecules interaction. Conventional membranes cannot be utilised for ED applications because they are not selective, and they are considered an ohmic barrier. Recently, a porous separator known as diaphragm has been used in an electrochemical cell, however, it does not deliver selective separation [59].

IEMs can be defined as thin polymeric films (e.g., polysulphone, polyethylene, polystyrene) having charged group (i.e., negative or positive charged ions). They are principally classified based on their charged groups into cation exchange membrane (CEM) and anion exchange membrane (AEM). CEM possesses negative charges (e.g. phosphonic acid ($-\text{PO}_3\text{H}^-$), phosphoryl ($-\text{PO}_3^{2-}$), carboxylic acid ($-\text{COO}^-$), sulfonic acid ($-\text{SO}_3^-$), $\text{C}_6\text{H}_4\text{O}^-$) covalently bonded to the polymer backbone of the membrane, which blocks negative and cations, and allows positive ions to pass [145]. Shortly, AEM has positive charges (e.g., quaternary amine ($-\text{NR}_3^+$), tertiary amine ($-\text{NR}_2\text{H}^+$), secondary amine ($-\text{NRH}_2^+$), ammonium ($-\text{NH}_3^+$), SR_2^+ , PR_3^+) and does the opposite work of CEM. These fundamental features were founded and reported by Donnan [146].

The manufacturing method and physical properties (e.g., hydrophilicity, surface charge distribution, electrical resistance, surface roughness) of AEM and CEM classify them into homogeneous and heterogeneous ion exchange membranes. In 1933, a sheet of polymer was wholly bonded with uniformly charges, known as homogenous IEMs, while heterogeneous IEMs have uncharged binding polymer. Homogeneous IEMs (e.g., Styrene-divinylbenzene-based membranes) are the most used type for table salt production [147]. Fabrication methods for homogeneous IEMs can be found in the review papers [36,148]. Fig. 6 illustrates the conventional structure of homogenous CEM under the nanoscale.

Sometimes, a supporting matrix is used for the IEMs to deliver the desired mechanical stability [150], but it reduces the conductivity. Besides, ED process generates complexes which increase the mechanical strength of IEMs but it reduces its permselectivity. Polytetrafluoroethylene (PTFE) has been introduced to increase the mechanical strength of membrane films (100 to 200 μm). The membrane thickness decides the potential ohmic loss, and it can be minimised by reducing the IEMs thickness. Bensmann et al. (2014) found the thin IEMs

produce low selectivity, and poor permeability to gases and have crucial mechanical properties [151]. A low ohmic loss by the IEMs thickness results in high efficiency of membrane area and electrode, and longer lifetime. Van der Stegen et al. (1999) has utilised 1 mm distance between the membrane and the cathode in the electrodialysis cell for sodium removal at a temperature of 80–90 $^\circ\text{C}$ and atmospheric pressure [152]. Other studies suggested pressing electrodes on each side of the membrane film to minimise the loss.

IEMs fouling is still a major drawback restricting ED from being scaled up and widely applied. The type of membrane clogging is determined by the molecular size (geometrical factor) of organic particles. In fact, large molecules remain on the IEMs surface, small molecules 200–700 Da result in internal membrane clogging, and smaller molecules move freely through the membrane pores and do not lead to fouling [153]. Kim et al. (2007) and Woźniak et al. (2014) have suggested to stream a feed into pre-desalination (e.g., ultrafiltration UF, microfiltration MF, nanofiltration NF) to remove heavy metals and organic matters before entering the ED stack [154,155]. A researcher reported using activated carbon and cleaning actions with NaOH solutions to prevent fouling from happening [156]. One of the other options to reduce a significant membrane fouling is using porous membrane instead of IEMs. It was invented in the 1950s as a solid electrolyte to replace the classical liquid electrolyte to be electrolyte and separator.

At present, a number of studies are occurring on combining ED process with external forces (e.g., microwave, magnetic field, ultrasonic, electric pulse) to reduce membrane blockage, increase desalination rate, and optimise water recovery [157]. Despite the introduced methods for reducing IEMs clogging, standard ED operation always need cleaning, and it leads to an increase in the operating costs. Therefore, studies had found the electrodialysis reversal (EDR) concept which has successfully reduced the tendency of IEMs fouling [40]. Furthermore, EDR contributes to reducing pre-treatments, cleaning procedures, and avoid pH controllers and presence of acids tanks. Turek et al. (2006) found that the polarity reversal concept can be applied in harsh circumstances like salt supersaturation [158]. Turek et al. effectively operated EDR under more than 175% supersaturation of CaSO_4 . More importantly, using EDR is the reason behind the long lifetime of brackish water ED industrial plants and IEMs lifetime reaches up to 10–15 years.

Recently, the pulsed electrical field (PEF) was investigated

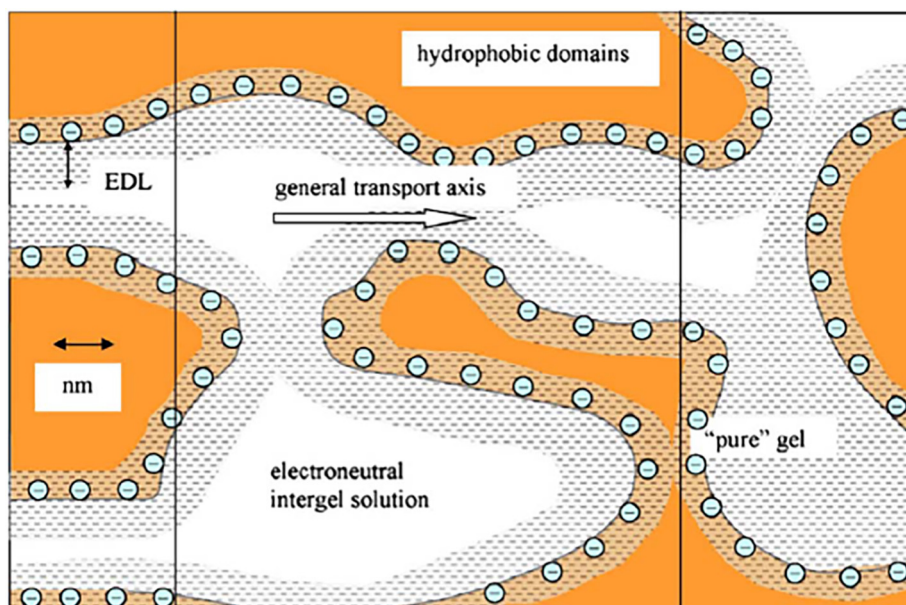


Fig. 6. Illustration of a homogeneous CEM, with the three different phases: polymeric hydrophobic, electroneutral solution and gel phase (adapted from ref. [149]).

[156,159]. PEF involves applying a discontinuous electric current to disturb the deposition of charged molecules on the IEMs surface. Sayadi et al. (2015) coupled an electronic device made by Ruiz et al. (2007) to the current generator to deliver PEF for brackish water treatment [160,161]. ED setup was operated under two different frequencies: 1 kHz (0.5 ms), as high frequency and 5 Hz (100 ms), as low frequency. PEF helped to prevent CaCO_3 formation on the membrane surface, reduce the membrane fouling, and improve ion transfer. The results of Sayadi et al. (2015) confirms the findings of Mishchuk et al. (2001) in terms of PEF advantages [162]. In contrast, Suwal et al. (2016) found that PEF doesn't affect migration rate and it disagrees with the findings of Sayadi et al. (2015) and Mishchuk et al. (2001) [163].

Furthermore, EDR and PEF were compared and delivered similar performances [163]. Yet, IEMs clogging is still a limiting factor for the wide construction of ED. It was found that humate and anionic surfactants are adsorbed on the AEMs and weaken its properties [37,164,165]. Also, most organic substances in wastewater have negative charges, so AEMs are always exposed to serious fouling. Lee et al. (2009) studied the possible AEMs clogging by sodium dodecyl benzene sulfonate (SDBS), bovine serum albumin (BSA), and humate [166]. Another researcher compared aliphatic and aromatic AEMs with various anion exchange groups and found the chemical composition of AEMs could affect the IEMs fouling type [167]. Moreover, new types of ionic, stable, selective, and conductive AEMs can have the potential of introducing very promising desalination for solving numerous industrial serious issues. In this respect, many researchers have been investigating AEMs surface modification processes in order to advance its antifouling properties [168–170]. In 1998, Grebenyuk et al. enhanced AEMs resistance against organic deposition by adding high molecular mass surfactants [168]. Other researchers used polydopamine and poly (sodium 4-styrene sulfonate) to improve antifouling, but their impact on AEMs properties was neglected [169,170]. The evolution in AEMs membranes can minimise the drawbacks of ED system, and using a cheap binding polymer reduces IEMs prices, but such studies on AEMs structure to prevent clogging remained scarce and have not delivered revolutionary results.

In summary, empirical investigations and review studies have introduced novel enhancements to improve ED concept (e.g., new IEMs fabrication methods, IEMs antifouling). EDR and PEF were investigated and found able to reduce membrane fouling, but they have not been used together in one ED stack. Researchers like Luiz et al. (2017) and Scarazzato et al. (2018) did not observe membrane clogging in their laboratory-scale ED system. Therefore, this review study agrees with Luiz et al. (2017) to use pilot-scale ED to widely investigate membrane fouling. Jiang et al. (2016) investigated membrane fouling of methionine separation using pilot-scale bipolar membrane electrodialysis, but studies are still largely scarce to significantly mitigate IEMs fouling. It is plausible that the slow advance of ED desalination may be attributed to a lack of functionally reducing its main drawbacks.

1.7. Sustainability in electrodialysis applications

Every year, desalination consumes more than 850 million tons of oil to treat more than 90 million $\text{m}^3 \text{day}^{-1}$, generating 76 million tons of CO_2 that expected to reach 218 million tons by 2040 [60]. Cleaner production is developed to prevent industrial pollution and promote benefits for human beings and the environment. It does encourage the use of natural resources efficiently to reduce serious risks in the environment. Moreover, the sustainability of ED desalination is assessed and impacted by water quality and availability, energy source, operation and maintenance costs, and geographical location. Integrating renewable energies with ED desalination improves water sustainability. Therefore, renewable energy has been considered as a practical solution to water scarcity [171,172].

Solar, wind and geothermal energies have been integrated with ED technology for brackish water and seawater desalination [60]. In 2011,

about 9% of the solar energy in the worldwide desalination was utilised for ED [173]. Coupling ED with PV has many advantages such as easy transportation and installation, no noise or direct pollution, environmentally friendly, feasible and limited maintenance cost [47]. Low scale pilot plants of ED coupled with photovoltaic (PV) cells to produce potable water have been installed in remote areas [174]. Based on the feed properties and the demanded recovery ratio, ED requires energy in the range of 6–11 kWh m^{-3} [175]. AlMadani (2003) investigated the ability of small-scale ED having 24 cell pairs driven by solar energy for desalinating groundwater having average salinity [176]. In 2018, Herrero-Gonzalez et al. used PV-ED system to produce HCL [177]. Ortiz et al. discussed the feasibility of PV-ED system without the need of energy storages [178]. In all the studies, it has been reported that PV-ED system is a promising method for the treatment of seawater and brackish water, especially in remote areas. In 2013, Uche et al. (2013) investigated the possibility of installing PV-ED system in Northern Chile because it possesses 7–7.5 $\text{kWh m}^{-2} \text{day}^{-1}$ solar radiation and rivers having above 4000 mg L^{-1} salinity, which makes it attractive to construct ED technology powered by solar energy [179]. ED process coupled with 8 PV panels produced 0.29 $\text{m}^3 \text{h}^{-1}$ within 0.35 h, but when the number of PV panels was reduced into 4, the duration increased to 0.45 h whereas the capacity reduced to 0.22 $\text{m}^3 \text{h}^{-1}$. Moreover, ED plant having 42 cell pairs coupled with a PV panel of 450 W peak power delivered 1000 L day^{-1} capacity and produced below 1000 ppm water salinity [39]. It is worth mentioning that the largest PV-ED system having 65 kWp PV array was installed in 1990 and located in Fukue City, Japan [173]. It delivers 200 $\text{m}^3 \text{day}^{-1}$ production capacity with 0.6–1 kWh m^{-3} energy consumption. A village scale PV-ED system was designed by Wright et al. to be energy efficient and cost effective for groundwater desalination [130]. Unlike ED powered by fossil fuels, PV-ED system for brackish water saves up to 0.724 $\text{kgCO}_2\text{-eq. m}^{-3}$ [180].

Empirical studies have been running to evaluate combining ED desalination with wind energy for potable water production [60]. Wind-powered ED has been found simple, economical and efficient (see Fig. 7). In 2016, Malek et al. studied wind-ED system for brackish underground water desalination characterised by 5000 mg L^{-1} of NaCl [181]. It produced 4.15 kWh m^{-3} at a wind speed of 10 m s^{-1} and 2.52 kWh m^{-3} at 2 m s^{-1} [181]. Another study found that wind speed of 2–8 m s^{-1} makes the wind-ED system capable of producing water capacity of 2.02–3.62 $\text{m}^3 \text{m}^{-2} \text{day}^{-1}$, which is higher than the reported PV-ED system capability (0.2–2.3 $\text{m}^3 \text{m}^{-2} \text{day}^{-1}$) for groundwater desalination [182].

Solar and wind energy are free, but they require obvious capital cost to harvest renewable energy and power ED desalination effectively. Studies found that conventional ED for brackish water treatment costs 0.45–0.78 € m^{-3} , while PV-ED costs 6.34–11.93 € m^{-3} , which makes the potable water produced by renewable-ED more expensive [47]. Fernandez-Gonzalez et al. (2015) predict that the cost of PV-ED system will be equal to grid-ED cost by 2025, after that, costs will continue increasing for grid-ED because of fossil fuels shortage and continue decreasing for PV-ED [47,183]. Another reason prevents PV-ED system from being largely commercialised, which is matching the serial output of PV energy with ED energy demand [184]. However, researchers have mentioned PV-ED system in their literature [176,182] for being stable and having a better performance than PV-RO. Also, PV-ED requires 50% less specific energy than PV-RO at 2000 mg L^{-1} , yet PV-RO received more attention than PV-ED [60]. One of ED main drawbacks is the generation of concentrated brines, which requires additional costly treatments.

In the early 1950s, ingenious way, known as reverse Electrodialysis (RED) was conceptualised by Pattle to generate energy by the salinity difference between brackish water and concentrated brines or river water and seawater [40,185]. Recently, researchers defined RED as an emerging renewable energy technology for electricity generation [186,187]. Inside RED stack, a chemical reaction driven by the salinity difference (driving force) between the two solutions occurs and

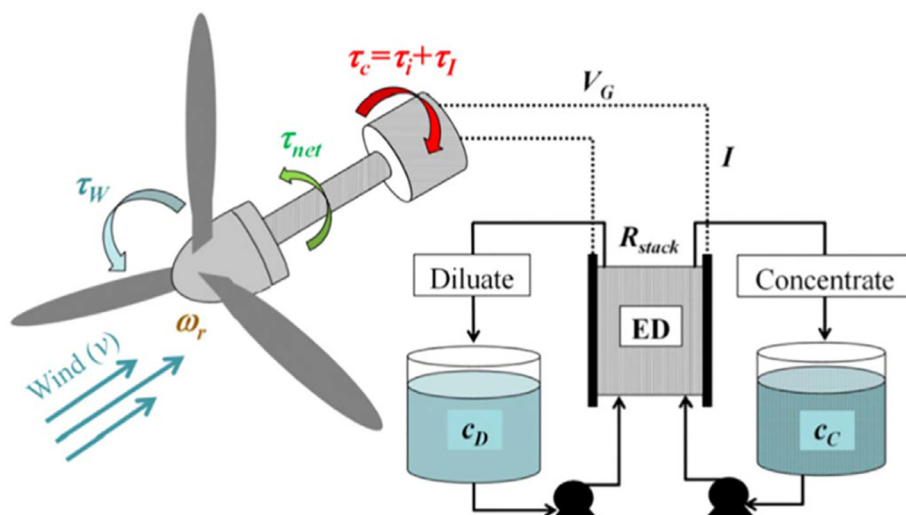


Fig. 7. A wind-ED system for brackish groundwater desalination (adapted from ref. [181]).

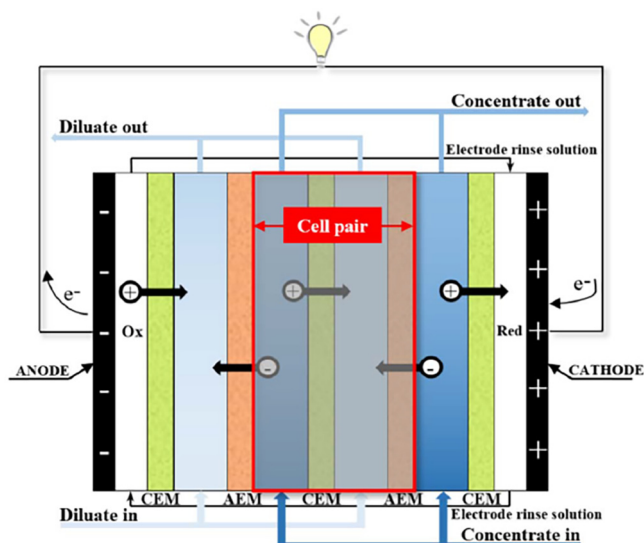


Fig. 8. Scheme of reverse electro dialysis process (adapted from ref. [40]).

generates energy, while ED uses energy to create salinity difference, i.e. dilute and concentrate. Therefore, RED has received broad interest from water and energy scientists. Fundamentally, ions driven by salinity difference start to move from the concentrate compartment to the dilute compartment passing through selective IEMs once the external circuit is closed. Negative ions move to the anodic side, and positive ions go to the cathodic side, resulting in ionic current through the RED stack. Consequently, the ionic charges generate an electrochemical potential and turn into electric charges recorded as a voltage across electrodes and passing through the external circuit. Researchers reported the usage of the generated electronic current in their literature [188–191]. Fig. 8 shows the structure of RED with the ionic movement in the stack.

Campione et al. (2018) reviewed RED energy generation concept, and the maximum power density was found obtainable when the external load resistance is low as the internal resistance of the stack [40]. Besides, only 50% of the available Gibbs free energy can be theoretically harvested, and only a portion of this energy can be converted into electric energy. In the recent years, RED technology has been improved by Veermaas et al. (2009 and 2011) and becomes able to generate $1\text{--}2\text{ W m}^{-2}$, while it was generating 0.05 W m^{-2} in the early '50s [192,193]. Recently, researchers obtained the highest power density $\sim 6.70\text{ W m}^{-2}$ from the salinity difference of brackish/fresh water

and concentrated brines at $40\text{--}60\text{ }^{\circ}\text{C}$ [111,194]. In 2014, REAPower pilot plant was installed in Marsala (Sicily, Italy) having 3 different RED stacks generates about 1 kW from concentrated brines, brackish water and seawater. Moreover, the pilot plant operation was investigated for several months and found no technical issues nor performance reduction [195].

A breakthrough concept was introduced to couple ED with RED to utilise the generated energy by RED to power ED desalination, known as A power free electro dialysis (PFED). In fact, ED is considered as a load while RED is considered as a battery, i.e. constant voltage. Yet, there is a little number of studies on this concept. In 2015, Chen et al. designed a membrane stack involving RED and ED for energy self-sufficient desalination on small islands [196]. In 2017, Wang et al. (2017) investigated RED-ED system for treating high salinity brine containing phenols [197]. Luo et al. (2017) proposed to supply RED with the brine of ED and RED dilute can be supplied from wastewater, resulting in a sustainable and economic treatment with power generation too [183]. Also, any increase in RED energy generation would improve ED desalination performance. Moreover, RED-ED combination helps to utilise high salinity wastewater as brine instead of discharging it. Luo et al. (2017) successfully integrated RED and ED with simulated wastewater and seawater [183]. It was found that ED stack must contain much less membrane area than RED to guarantee the generated power is sufficient to power ED desalination. Also, the resistance of ED stack should be equal or less than RED stack to receive a maximum power output. Then, Luo et al. (2017) concluded that RED-ED is economically attractive and technically feasible to produce clean water from two concentration gradient water without consuming external power. In 2014, the installed capacity of RED-ED systems around the world was 2.59 million gallons per day [198]. The contracted RED-ED plants in 2015–2016 covered 1–2% of the total desalination constructed capacity with processing reaches few tens to $10,000\text{ m}^3\text{ day}^{-1}$ of brackish water [199]. Fig. 9 shows the coupling possibilities between the two technologies.

In 2002, Cifuentes et al. found ED desalination having low selectivity in a solution containing a wide range of various metallic ions, which makes it useless [200]. Richard Baker (2004) and Peng et al. (2014) found ED desalination incapable of multi-component aqueous solutions [201,202], which confirms Cifuentes's findings. Some researchers have called to integrate ultrafiltration or nanofiltration with ED desalination [203,204]. Also, some additives (e.g., complexing agents, EDTA, ligands) have been introduced to deliver weak, medium or strong affinity toward metal ions and enhance IEMs selectivity [205]. Other researchers have suggested increasing the active membrane area, but it results in a decrement in the electric current, weak

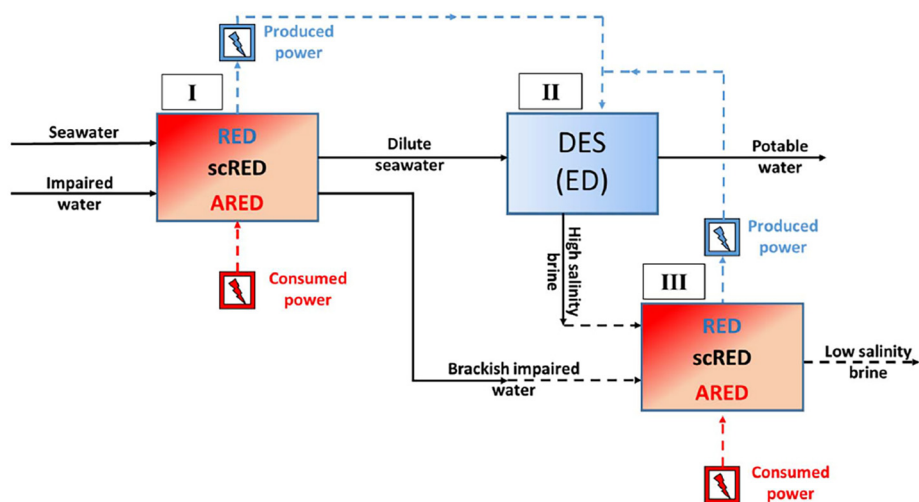


Fig. 9. Possibilities of RED-ED integration (adapted from ref. [40]). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

electromigration and then concentration polarisation [206]. Luo et al. (2017) and Campione et al. (2018) found little and limited studies on RED-ED and called for more research [40,183], and the reported self-desalination [207–209] by RED-ED system have poor efficiency. Besides, RED-ED demonstrated an overall reduction of energy consumption of about 30% compared to the stand-alone ED case, though such figures cannot be compared with RO, due to the much higher energy consumption of the base case (above 20 kWh m^{-3}), and higher cost of IEMs compared to RO membranes. In conclusion, RED-ED desalination is an excellent system, but it has some limitations including selectivity, sufficient power generation, fouling and scaling. Table 2 shows operating conditions, advantages, and challenges of the latest ED studies.

Securing a sustainable supply of water and energy is still ongoing research to combatting climate change, saving the environment, and securing welfare for the present and future generations. The International Renewable Energy Agency (IRENA) has estimated that water availability needs to be increased by 55% and energy generation by 80% to meet global demand in 2050 [223]. Therefore, novel water production systems from renewables (e.g., PV-ED system, RED) are still requiring enhancements. Also, research on energy conversion and storage devices (e.g., PFED) has received great attention due to increasing concerns over climate change and high energy costs [224]. In this section, this article had reviewed the latest ED advancements that promote sustainability and it was found greatly developed. Accordingly, this systematic review raises a question, why would a flexible and highly advanced ED desalination continuously fail to be a leading water and wastewater treatment? In fact, the bottlenecks of the ED desalination are presented as matching the PV energy with ED energy demand, generation of concentrated brines, poor efficiency of RED-ED system, and only a portion of the available Gibbs free energy can be converted into electric energy have widely limited scaling ED technology into industrial scale. Moreover, the competition with RO has limited ED applications in water and wastewater. In terms of studies number, RO studies are more than three times than ED studies. Also, there are only 25 original research papers investigated PFED system. In conclusion, the slow evolution of ED desalination has largely failed in solving its drawbacks.

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3. Conclusion and future outlook

Great studies have developed water desalination aiming to achieve a high loading rate, high selective separation, less fouling, and high-water recovery. It decreased the required energy 20 kWh in 1970 for 1 m^3 of fresh water production to 2.5 kWh in 2010 and reduced desalination cost to less than USD 0.75 per m^3 in 2008. However, many desalination technologies still require amounts of chemicals for saline water pre-treatment and post-treatment. Electrodialysis (ED) desalination delivers high water recovery without the use of chemicals. ED's capability in removing ionic and non-ionic components has qualified it to process municipal wastewater, brackish water, industrial wastewater, and has even been used in chemical and food industries. In fact, the evolution of ion exchange membrane (IEM) is the reason behind the diverse applications of ED technology. Moreover, a large number of studies developed and refined mathematical models as a powerful tool to develop ED system to examine and optimise its performance.

The rapid developments have qualified ED to be scaled up and process brackish water having salinity above than $15,000 \text{ mg L}^{-1}$. Despite that, this study reviewed ED evolution and technical flaws (e.g., IEMs fouling, high energy consumption, poor perm-selectivity, proton leakage of AEM, defects in the concentration diffusion and electromigration) were found limiting ED desalination. Among the very little number of studies that reported ED drawbacks, this review study confirms ED drawbacks as major technical limitations. Moreover, researchers had debated about supplying ED system with a lower or higher voltage than the limiting current density. At present, researchers had agreed to supply higher and close voltage to the limiting current density due to the internal resistance (i.e., electric resistance, solution conductivity) and to receive the wanted performance without damaging the membrane films. Although, other researchers critiqued the recent ED studies because it neglected the major technical problems and focused on the limiting current density.

Moreover, approaches like coupling ED technology with reverse ED (RED) and photovoltaic (PV) have been presented as possible solutions to sustain ED desalination and promote a cleaner production. At the end, this review found ED sustainability is important and could be the only way to minimise ED limitations. For instance, electrodialysis

Table 2
Electrodialysis technologies for water and wastewater treatment, and chemicals recovery.

Author (s)	Electrodialysis Technology	Operating conditions	Findings & Advantages of ED process	Drawbacks/challenges
Luo et al. [183]	PFED for fresh water production	Conductivity ($72 \mu\text{S cm}^{-1}$); Flow rate ratio of concentrate to dilute (1:1); Flow rates ($40\text{--}700 \text{ mL min}^{-1}$); Power consumption (0.35 W m^{-2}); Resistance (5 Ohm)	(1) Technically feasible; (2) Economically attractive; (3) Doesn't require batteries; (4) The concentrate of ED can be fed as concentrate for RED to generate energy; (5) Voltage generated by RED is constant	(1) Concentration polarization; (2) Lower membrane perm selectivity at high concentrations; (3) ED resistance is a slightly higher than RED internal resistance
Xu et al. [112]	A selective ED involving mono and divalent membranes for brackish groundwater desalination	Pilot-scale ED: Conductivity ($4626 \mu\text{S cm}^{-1}$); 186 cell pairs of CEMs and AEMs; Effective membrane area 3200 cm^2 ; Voltage ($28\text{--}37 \text{ V}$); Energy consumption ($1.3 \text{ kWh kgal}^{-1}$); Salt removal (0.42 and $0.74 \text{ kg salt m}^{-2} \text{ kWh}^{-1}$ at current density of 3.3 or 5.2 mA cm^{-2} , respectively); Water recovery 55%	(1) Selective membranes with increasing current density results in higher salt removal at higher flow rate; (2) Selective membranes showed stable ion selectivity over a wide range of operating conditions; (3) Pulsed electric field can be used to control concentration polarization at the membrane interface [160]; (4) A higher applied voltage enhanced removal of inorganic elements [210]	(1) Salt removal efficiency decreased with the flow rate increasing from; (2) The monovalent selective membranes achieved the same desalting efficiency as the normal grade membranes during bench and pilot scale ED; (3) Stack resistance and membrane deposition [210]
Luiz et al. [80]	Upgrading bio-refinery streams by a bench scale batch ED	Bench-scale ED: 10 cell pairs; Effective membrane area (220 cm^2); Voltage (2 V cell^{-1}); Energy consumption ($1.9 \text{ kWh kgal}^{-1}$); Salt removal (5.17 and $4.97 \text{ kg salt m}^{-2} \text{ kWh}^{-1}$ at current density of 0.15 mA cm^{-2} and 0.3 mA cm^{-2} , respectively); Water recovery 50% Conductivities (72.4 mS cm^{-1}); Current ($110\text{--}650 \text{ mA}$); Current efficiencies ($69\text{--}104\%$); Specific power consumption ($0.44\text{--}1.59 \text{ kWh kg}^{-1}$); Salt removal (96%) Duration (30 h); PV efficiency (14.2%); Energy consumption ($7.3 \text{ kWh kg}^{-1} \text{ HCl}$); Produced 1 mol L^{-1} of HCl; Salt removal ($95.6\text{--}98.2\%$) [55]	(1) If the desired conductivity cut is not high, the ED would be recommended to operate at higher flow rate to increase water production; (2) Bench-scale testing results can be used to predict the ion selectivity of pilot and potentially full-scale ED (1) Cost-effective potential; (2) Increasing salt content amplified the ionic transfer rate (1) PV led to a noticeable drop from 7.3 to $4.4 \text{ kWh kg}^{-1} \text{ HCl}$; (2) Combining ED process with external forces/renewable energy/treatments reduces desalination time and minimises energy consumption [55]	(1) Organic losses ($0.3\text{--}6.3\%$) across the ED membrane
Herrero-Gonzalez et al. [177]	ED with bipolar membranes (EDBM)-PV to produce HCl and NaOH from seawater		(1) Cleaning procedures restored the membrane properties; (2) Feasible for recovering water and inputs (e.g., chemicals), and reducing waste generation (1) Cost-effective separation; (2) The choice of anion exchange membrane determines power consumption and desalination degree; (3) Feed with low anions causes high permeability (1) Optimal conditions resulted in high concentration of AKG, high current efficiency and low energy consumption (1) Using AEMs removed over 60% of Fe, Mg, and Ca.	(1) The divalent cations transport rate was reduced due to electrostatic repulsion
Scarrazato et al. [43]	Chemicals recovery from cyanide-free copper plating bath using ED	Flow rate (3.5 L h^{-1}); Produced a solution 5–6 times more concentrated than the initial one; Demineralization (90%); Extraction ($> 80\%$)	(1) Effective cleaning procedures led to operational sustainability (2) ED is an economically promising technology for the nutrients recovery; (3) Investigating the correlation between CE and the changes of electric current and of potential drop leads to find the moment when CE declines and the energy consumption increases [126]; (4) Activated carbon reduced nitrate selectivity [214]; (5) Semi-continuous operation mitigated particle and chemical deposition [215]	(1) Backward diffusion; (2) Interactions between organic acid and membrane fixed groups
Luiz et al. [211]	Desalination of bio-refinery effluents using ED	10 pairs of CEMs and AEMs in batch mode; Membrane area 0.0121 m^2 ; Flow rate 30 L h^{-1} ; Current of 440 mA ; Duration (3 h); Maximum voltage (30 V)	(1) Concentration difference and potential gradient transported undesired protons; (2) Simulation and empirical results were in good agreement	(1) Organics crossing the membrane increased stack resistance
Szczygiełda et al. [79]	Alpha-ketoglutaric (AKG) acid production using EDBM	Two-chamber EDBM stack; Anion exchange membrane-bipolar membrane (AM-BM) configuration; Current efficiency (71.8%); Energy consumption (3.72 kWh kg^{-1}); Duration (180 min); Flow rate of 5.6 L h^{-1} ; AKG production (4.83 g L^{-1}) Experiment time (360 min); Current efficiency (68.13%); Membrane price ($50 \text{ \$ m}^{-2}$); Membrane lifetime (3 year); Energy consumption (2.73 kWh kg^{-1}); Industrial electricity ($0.15 \text{ \$ kWh}^{-1}$)	(1) Further investigations are required to bring EDBM to industrialization	(1) Further investigations are required to bring EDBM to industrialization
Duan et al. [212]	phosphoric acid purification by electro dialysis having polysulfone-based anion exchange membrane	30-cell pair pilot reactor (31 CEM membranes and 30 AEM membranes); Effective membrane area (7.2 m^2); Duration (72 h); Flow rate (75 L h^{-1}); Current efficiency (76%); Power consumption (4.9 kWh kgN^{-1}); 5400 L of feed in total was treated		(1) Depletion of ions in the membrane boundary layer; (2) Back diffusion of ions from the concentrate to dilute compartment due to a large concentration gradient, osmotic and electro-osmotic fluxes; (3) $\text{NH}_4\text{-N}$ total loss of 28% ; (4) Water dissociation [216]; (5) Membrane crystallinity [214]; (6) Fouling caused by chemical and particle aggregation [215]
Ward et al. [213]	Nutrient recovery through pilot scale ED	Membrane effective area (0.785 cm^2); Two compartments of equal volume (about 50 mL)		(1) Acid leakage
Jia et al. [100]	ED for inorganic acid recovery			
Faucher et al. [217]				

(continued on next page)

Table 2 (continued)

Author (s)	Electrodialysis Technology	Operating conditions	Findings & Advantages of ED process	Drawbacks/challenges
	Cranberry juice deacidification by semi-industrial EDBM	batch mode; Duration (95 h); Total surface area (0.834 m ²); Conductivity (2.2–2.3 mS cm ⁻¹); Migration rates (61.23–38.13 mg L ⁻¹ min ⁻¹)	(1) Eco-friendly technology; (2) Reusing the recovery solution increases conductivity and leads to a 42.9% energy savings; (3) It is suggested to run Cranberry juice deacidification by EDBM for 1000 h before scaling it up	(1) H ⁺ leakage was observed and it is possibly led to plateau; (2) Leakage of potassium through the anion-exchange membrane after 3 h of treatment
Lu et al. [218]	Penicillin G and sulfate ions separation using ED ultrafiltration configuration	Effective membrane area (214.7 cm ²); Flow rate (250 mL min ⁻¹); Current intensities 0.2–0.4 A; Demineralization (69–84%); Laboratory scale process costs 10.7 \$ for recovering 1 kg of penicillin G	(1) No significant fouling; (2) Recovering 203 tons of penicillin G saves 203,000 m ³ freshwater; (3) Recovering 1000 tons/year penicillin G leads to annual profit of 6.85 million \$	(1) Stack resistance; (2) Water splitting at the membrane surface changes the pH level
Selvaraj et al. [219]	Mono selective electro dialysis for sodium formate separation	Four compartments; Current density (30 mA cm ⁻²); Limiting current density (21.6–75 mA cm ⁻²); Flow rate (240–840 mL h ⁻¹); Current efficiency (70%); Sodium formate purity (87%); Formate recovery (69%); Energy consumption (96 kWh m ⁻³)	(1) No significant change in pH; (2) Diffusion of formic acid and formate leakages didn't influence the current efficiency	(1) OH ⁻ leakage through CEMs decreases the current efficiency
Andreeva et al. [220]	Membrane scaling in ED by applying a pulsed electric field, enhance electroconvection, and adjust pH level	Current (1.5–2 times higher than the limiting current); Flow rate (30 mL min ⁻¹); Experimental run (5 h); Current density (18 mA cm ⁻²)	(1) No precipitation on all the membrane sides; (2) An enhanced electroconvection increases mass transfer; (3) Pulsed electric field delivers a relaxation of the concentration profile	(1) Low mass transfer due to funnel effect
Zhao et al. [11]	EDR for industrial RO brine treatment	Three cell pairs; current (0.89 A); Membrane active area (180 cm ²); Conductivity (10.5 ms cm ⁻¹); Current (0.75 A); Voltage (0–20 V); Batch operation (8 h); Removal efficiency (7.1%); Water recovery (85%); Reduction of RO brine (6.5 times); Conductivity reduction (50%)	(1) Able to produce highly concentrated brine; (2) Organic fouling wasn't detected even after 6 days of continuous operation	(1) EDR resistance increased during 8 h of process; (2) Inflection point due to ion depletion; (3) Polarization occurred when using a current above the limiting current; (4) CaCO ₃ precipitation
Nam et al. [221]	Pilot-scale RED fed with seawater and municipal effluent	1000 cell pairs; total membrane area (250 m ²); Velocity (1.5 cm s ⁻¹); Energy efficiency (11.4–13.3%); Power production (95.8 W)	(1) Sustainable renewable energy source	(1) Spacers clogging; (2) Fouling due to inorganics precipitation
Hosseini, et al. [222]	Magnetic CEM incorporated with cobalt ferrite nanoparticles for chromium ions removal via ED	–	(1) CoFe ₂ O ₄ nanoparticles increased the membrane hydrophilicity and conductivity and enhanced permselectivity, transport number, ion exchange capacity and water content in the membrane	–

reversal (EDR) can successfully reduce the tendency of IEMs fouling and reduce pre-treatments, cleaning procedures, and avoid pH controllers and presence of acids tanks.

Future works will point to advance ED coupling with other technologies to increase its sustainability, performance, and minimise its drawbacks. Additional pilot-scale investigations are wanted to validate the feasibility of ED applications and play an important role in bridging lab-scale ED application to full-scale implementation. Other future papers will present detailed studies to simulate ED applications and quantify the process economics. Finally, development of new types of ionic, stable, selective, and conductive AEMs can be useful for solving numerous industrial serious issues, enhancing membrane lifespan, and projecting a new horizon of applications and new markets.

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