A systematic review and statistical analysis of nutrient recovery from municipal wastewater by electrodialysis

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GRAPHICAL ABSTRACT
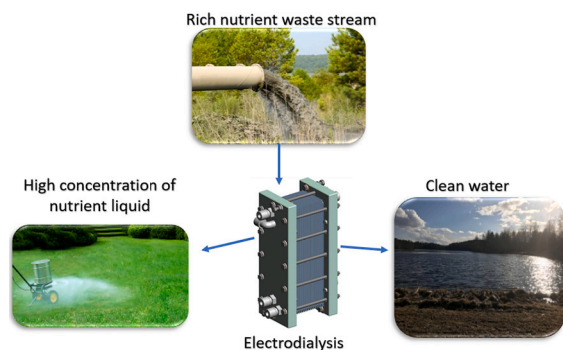
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ABSTRACT

These days electrodialysis is recognized as a promising process for the removal and recovery of nutrients from wastewater. However, the electrodialysis process for nutrient recovery is different from traditional desalination electrodialysis, and limited knowledge exists regarding its new application. Therefore, a comprehensive review and critical comparison of the different parameters and optimal operation conditions for nutrient recovery by electrodialysis in various studies are discussed in this manuscript. The relationship between the nutrient recovery rate and critical operating parameters such as voltage, the total effective area of the membrane, number of cell pairs, and feed composition in various wastewater sources is statistically analyzed. Regression results showed that increasing voltages, membrane, and cell numbers improved the recovery rate, respectively. However, it should be mentioned that different mobility behavior of ions are influenced by an electric double layer, selectivity, repulsion of the ion-exchange membrane, and operation conditions including voltage, current density, operation time, and ion concentrations. Hybrid processes enhance the electrodialysis efficiency in wastewater by increasing the feed concentration. The capital and operating costs of the electrodialysis process can be significantly affected by the pump, electrode, and ion-exchange membrane characteristics. Fouling and scaling as a challenge in electrodialysis are investigated in our study.

1. Introduction

1.1. Background

The growing global population has increased stress on the food and water supply, and consequently on the fertilizer industry. The limited resources of natural phosphorus fertilizer are leading to concerns regarding their long-term availability [1]. On the one hand, a considerable percentage of the reactive nitrogen in soil fertilizer leaches into surface water or penetrated into the groundwater. This in turn can cause eutrophication in surface water resources and other health issues [2]. On the other hand, a strict limitation has recently been set for wastewater treatment plants regarding the removal of nutrients to acceptable limits before discharge and reuse. However, most conventional wastewater treatment technologies are costly and not highly effective for this purpose [3].

These days, wastewater streams are perceived as a promising resource of energy, water, and even nutrient recovery [1]. Consequently, the supply of phosphorus and nitrogen can be increased through nutrient recovery, while the nutrient loading into water resources can be reduced simultaneously. Nevertheless, the design of conventional sewage treatment systems is based on evacuating nutrients as waste sludge or releasing them as a gas into the atmosphere, which is hard to retrieve for later use. For instance, in wastewater, nitrogen is mostly in the form of ammonium (NH_4^+) and nitrate (NO_3^-), which traditional wastewater treatment plants convert to N_2 gas through the nitrification-denitrification process which in turn creates secondary air pollution [2,4,5].

Different nutrient recovery technologies from wastewater have been proposed and assessed, such as chemical precipitation [6,7], membrane processes [8–10], enhanced biological phosphorus removal (EBPR) [2], adsorption processes [11], gas permeable membranes, membrane, adsorption, and the Haber–Bosch process [5,12]. However, there is no clearly definitive nutrient recovery technology [13]. Besides most of the technologies mentioned are expensive, and the destructive environmental impacts of produced fertilizers are also significant. Among nitrogen recovery technologies from the aqueous phase, electrodialysis (ED), which produces a desalinated stream and a concentrated stream simultaneously, has received affair degree of attention. This method has several advantages for nitrogen recovery, such as lower energy consumption, less required pretreatment, high water recovery rates, and low chemical consumption without producing sludge [2,14].

1.2. The basic principle of ED

ED is an electromechanical separation method using ion-exchange membranes as a dynamic force (within an electric field) to support ionic separation and is considered mainly for the extraction of ionic species, as well as extracting hardness and organics from electrolytes [15]. An ED unit typically consists of two electrode compartments at the end side contacting with electrodes and a series of anion and cation exchange membranes (AEMs, CEMs) located between electrodes, which creates alternating concentrate and dilute solution compartments. A power source charges the electrodes, and an electrical current flows through the ED stack. The electrically charged anions of the feed solution migrate towards the anode (positively charged electrode) while the cations migrate towards the cathode (negatively charged electrode). Cations moving through the CEM are blocked by the AEM, and contrariwise anions move through the AEM are blocked by the CEM, which leads to the depletion of the salt content in the dilute compartments and the enrichment of the concentrate compartment. The charged groups on the membranes covalently bonded to the polymer backbone of the membrane are responsible for this phenomenon. The CEM contains negatively charged groups (e.g., sulfonic acid ($-SO_3^-$), carboxylic acid ($-COO^-$), phosphoryl($-PO_3^{2-}$), and phosphonic acid ($-PO_3H^-$)) while the AEMs have positively charged groups (e.g., ammonium ($-NH_3^+$), secondary amine ($-NRH_2^+$), tertiary amine ($-NR_2H^+$), quaternary amine ($-NR_3^+$)) that selectively transport anions but exclude cations [16].

One AEM and CEM are defined as cell pairs, and an ED stack contains a few cell pairs in laboratory-scale units up to several hundreds of cell pairs in pilot-scale units [17–19]. Fig. 1 shows a schematic of an electrodialysis stack.

ED is often operated in a batch mode; however, in large-scale plants, continuous processes are preferred in order to maintain a constant stream and quality of feed water [20]. In some studies, batch mode operation is used for concentrate compartments while the continuous mode is used for dilute compartments so they can retrieve highly concentrated solutions and to reduce the polarization phenomenon due to lower conductivity in the dilute compartments [21].

1.3. Development of ED application

ED was proposed for the first time in 1890 by Maigret and Sabates. They built a new concept unit to demineralize sugar syrup. In their prototype, they used carbon for the electrodes and permanganate paper for the membrane. Then, the first synthetic ion-exchange mem-branes were produced by W. Juda and W.A. McRay in 1950. In 1974, electrodialysis reversal concept (EDR) was developed Several "ED-derived" alternatives, applications, and processes have been developed and presented in the literature, providing a further boost to the development of electro membrane technologies in general [18]. Fig. 2
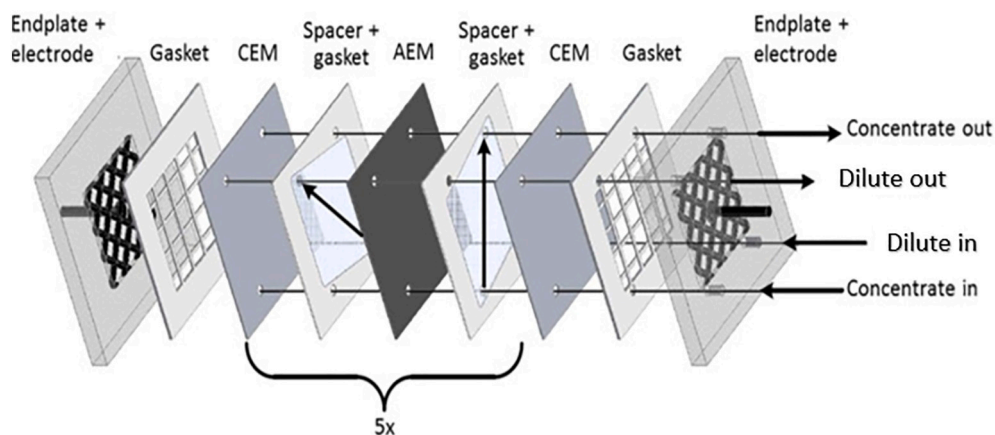

Fig. 1. Scheme of an electrodialysis stack with five cell pairs (AEM and CEM). Reproduced from Sosa-Fernandez et al. [22], with permission from Elsevier.
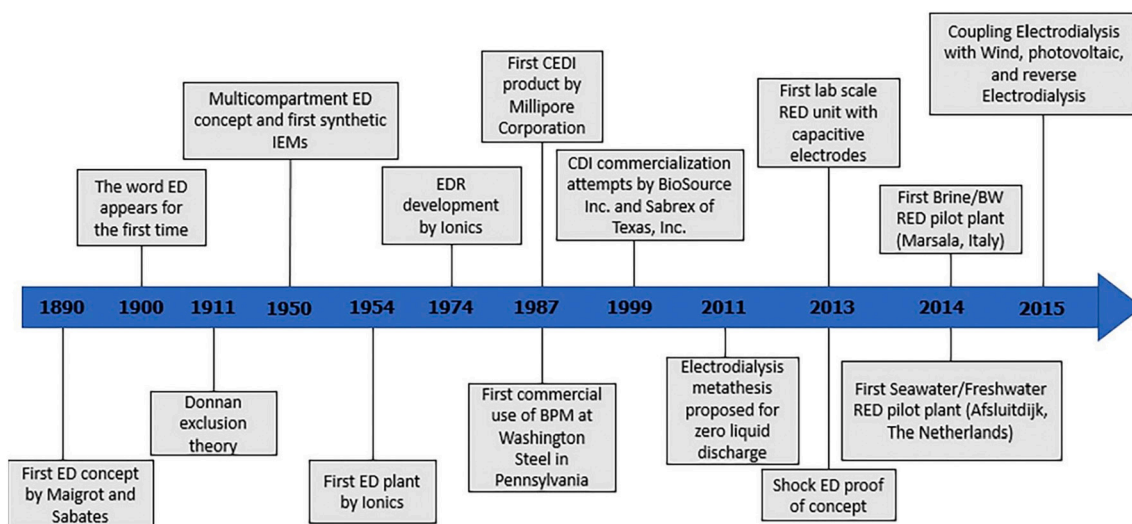
Fig. 2. Historical development of the ED process.
Reproduced from Al-Amshawee et al. [30] and Campione et al. [18], with permission from Elsevier.

represents a synthetic timeline of the most critical development steps in ED and ED-related technologies. The application of ED has been considered as a mature desalination technology for over 50 years [23]. The treatment potential of ED and all procedures founded on ED differs from below $100\,m^3\,d^{-1}$ to over $20,000\,m^3\,d^{-1}$ [24]. Besides desalination, the other significant function of ED was the retrieval of nutrients from water and wastewater. For instance, Khaoua et al. [25] removed ammonium by ED from polluted water. Ippersiel et al. [26] obtained $21.35\,g\,L^{-1}\,NH_4^+$-N from swine manure in a batch ED system. Chon et al. [27] tested various forms of ED membranes by regulating the solution's pH while the ED operating period was anticipated in order to escalate the dissolved inorganic nitrogen's selectivity by ED. In another study, Takagi et al. [28] proposed a technique for predicting the ED system's monovalent anion selectivity. In 2013, Wang et al. [29] examined the potential of conventional ED to extract phosphate from surplus sludge solutions, and the results indicated a nearly complete recovery ratio at a $10\,mL\,min^{-1}$ feed rate.

Besides the separation of the ions in ionic nutrients, other researchers have examined the separation of organic nitrogen from inorganic nitrogen [27], as well as ionic nutrients from organic compounds [23].

However, the ED process is not competitive for very dilute solutions; a high resistance, high energy consumption, and polarization are the main drawbacks for ED operations. Electrodialysis-based electrodeionization (EDI) is a hybrid system process which eliminates these problems. As seen in Fig. 3, the incorporation of ion exchange resins in the dilute compartments of EDI enhances the conductivity of the compartments by providing a path for enhanced ion migration. The ion exchange resins increase the conductivity across the process channel and enable ion transport towards the ion exchange membranes in a low conductivity process and in a manner that prevents concentration polarization [31]. EDI's energy consumption is equally minimized, making it appropriate for solutions that have a deficient electrolyte concentration. Unlike ED, which is usually operated below the limiting current density due to the concentration polarization effects, EDI commonly operates slightly above the limiting current density and is thus characterized by water dissociation and concentration polarization [32]. Both hydroxyl and protons ions generated in water dissociation may function to reproduce resins incessantly to some degree and consequently prevent additional chemical usage for resin regeneration. Therefore, EDI saves a lot of energy and is and more ecological [33]. As mentioned earlier, in most cases, ED is used for the desalination of brackish water and water demineralization. However, there are questions such as the rate of efficiency, critical operating parameters, cost-effectiveness, and likely challenges regarding the application of this technology for nutrient recovery from the wastewater.
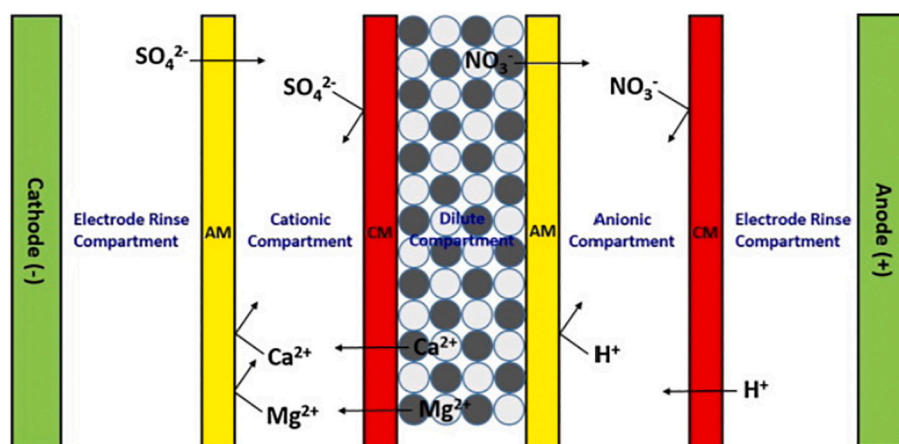

Fig. 3. Schematic diagram of an electrodeionization (EDI) cell for NO_3^-, Ca^{2+}, and Mg^{2+} separation and recovery.
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Therefore, this review paper provides a systematic overview, as well as a classification of the essential factors and their effect on the electrodialysis efficiency regarding nutrient recovery, as well as the operating challenges, and finally the costs of electrodialysis. This study, in turn, fosters the development and implementation of nutrient recovery by electrodialysis.

2. Database structure and statistical methods

The main stages for conducting the present study are described in the following sections and a summary of the methodology is shown in Fig. S1.

2.1. Searching for studies

A systematic review was conducted for all articles published in PubMed, Elsevier, American Chemical Society between 2006 and 2019, with an emphasis on the nutrient recovery from wastewater by electrodialysis system. The search phrases involved the following keywords: 'nutrient' OR 'nitrate' OR 'phosphate' OR 'ammonium' AND 'recovery' OR 'fractionation' OR 'concentration' AND 'electrodialysis' AND 'municipal waste water' OR 'wastewater' OR 'effluent' OR 'sewage.'

2.2. Selecting the studies

Full texts of the retrieved 30 articles were downloaded and reviewed on the basis of screening the titles and the abstracts. Studies with lack of access to the full article, or which were on inappropriate subjects, or which lacked treatment information were removed. 18 final articles with similar specifications were included (Table 1) for data analysis.

2.3. Data extraction

In order to extract information, all of the articles were evaluated independently, and the information extracted from the articles was included in a checklist. Data included in the checklist was based on the study characteristics (e.g., recovery rate, operating voltage, current, time, effective membrane area, energy consumption). Some data was taken directly from the selected studies, and the other data which was not available directly was extracted from graphs using the Web Plot Digitizer software.

Further, the recovery rate of the ED process of the selected studies was calculated using Eq. (1).

$$\text{Recovery rate} = C_P(t)/C_f(0) \qquad (1)$$

where Cp is the concentration of the concentrated solution or product, and C_f is the concentration of a feed solution, in mg L^{-1}, or mol L^{-1}.

The recovery efficiency (presented in Table 4) calculation was based on Eq. (2):

$$\text{Recovery efficiency} = \frac{C_P(t) - C_P(0)}{C_f(0) - C_f(t)} \times 100\% \qquad (2)$$

where $C_P(t)$ and $C_p(0)$ are the concentrations of nutrient ions at time t and 0 in the product streams, respectively, and C_f is the concentration of nutrient ions in the feed solution [35].

2.4. Data synthesis and analysis

Once the data was extracted, it was combined and analyzed. Regression coefficients were calculated for the nutrient recovery concerning the voltage, the number of cell pairs, effective membrane area, and their relationship. The SPSS software package was used for the regression analysis.

Since the data was obtained for different systems with various specifications, research data was mostly selected from the batch mood

studies of Table 1 for statistical analysis. The uncertainty of the regression equations was calculated using a Monte Carlo simulation on the Gold Sim website. "10,000" random data implemented at each Monte Carlo simulation.

There was other various data, which was shown in a case study. This included aspects such as the influence of the operation time at different voltages, and the influence of different competing ions on nutrient recovery in wastewater.

3. Results and discussion

3.1. A comparison of ED recovery efficiency under different wastewater and operational conditions

Several studies have highlighted the applicability of nutrient recovery from waste streams using ED (summarized in Table 1). From the table it can be seen that the design and properties of the electrodialysis desalination process are based on a set of fixed and variable parameters such as the stack construction, feed and product concentration, membrane properties, flow velocities, current density, applied voltage, number of cell pairs, the effective area of the membrane, and the pH. These parameters are interrelated and somewhat different for different applications. The results from the research are discussed in the following sections to determine the strength of the relationship between controllable factors and efficiency.

The parameters of cell pairs and voltage were selected as independent variables, and the recovery rate was chosen as the dependent variable. We did not consider the total effective area of the membrane as a variable since, in theory, the higher the number of cell pairs, the higher the total effective area of the membrane, and thus the results might overlap. Therefore, this aspect is discussed separately.

Table 2 shows the Pearson correlation, which is used to determine a linear relationship between variables and the strength of the relationship. As can be seen, there is no overlap concerning the effect of the variables on ED efficiency between any 2 variables as far as the correlations are < 0.7.

If the Pearson value is near ± 1, then it indicates a perfect correlation, and if the coefficient value lies between ± 0.50 and ± 1, it means there is a strong relationship. A value between ± 0.30 and ± 0.49 represents a medium correlation, and when the value is below ± 0.29, then a slight correlation is demonstrated [36]. The Pearson correlation was 0.880, and the p-value was 0.00 between the voltage and the recovery rate, which shows a strong linear relationship. Additionally, the Pearson correlation of 0.508 and p-value of 0.046 between the number of cell pairs and the recovery rate shows a substantial degree of interdependence as well. A positive value means that as one variable increases in value, the second variable also increases in value. The Pearson correlation of 0.309 and P-value of 0.146 between voltage and cell pairs numbers represent a moderate to weak dependency between these two variables. The scatter plots in Figs. 5 and 10 summarize our results for the relation between the voltage and number of cell pairs with the recovery rate, respectively.

Based on Table 3, the general form of the equation to forecast the rate of nutrient recovery from electrodialysis is:

The predicted nutrient recovery rate

$$= 1.6 + (\text{voltage} \times 0.129) + (\text{number of cell pairs} \times 0.329).$$

Unstandardized coefficients represent how much the recovery rate varies with independent variables when all other independent variables are kept constant.

Table S1 in the Supplementary material shows a summary and parameter estimates of multiple regression analyses of the recovery rate concerning the voltage and number of cell pairs for wastewater. The correlation coefficient, R^2, measures the quality of the prediction of the recovery rate as a dependent variable. The R^2 for this model is 0.836,

Table 1
Summary of different studies on nutrient recovery using different electrodialysis processes.

Types of wastewater	Methods	Number of cell pairs, type of membrane.	Operation conditions	Operation time (min)	Initial concentration	product concentration	Energy consumption	Voltage (V) or current (A)	Current density (mA/cm²)	Effective area per membrane (cm²)	Ref.
Swine manure	Hybrid ED coupled + air stripping.	10 cell pairs; AR204SZRA anionic (AM) and CR67HMR cationic (CM) membranes.	10 batch mode; dilute flow rate: 36 cm/s.		3200 mg L^{-1} NH$_4$$^+$-N	21,352 mg L^{-1} NH$_4$$^+$-N	18.05 kWh kg^{-1} NH$_4$$^+$-N	17.5 V	40	220	[26]
Swine manure	Hybrid ED + RO.	3 cell pairs; a combination of CM/AM Tokuyama Japan and CM from Ionics Inc., Watertown, USA.	10 batches: 30 cell pairs.	600	3.71 ± 2.45–5.54 ± 0.40 g L^{-1} NH$_3$-N	16 g L^{-1} NH$_3$-N		3 V	40	100	[37]
Synthetic and real pig manure	Two-stage bipolar membrane ED.	5 cell pairs; CM, AM, bipolar membrane (BM) from NEOSEPTA Inc.	4.1 L d^{-1} of dilute. 0.7 L d^{-1} of concentrate; flow rate: 300 mL min^{-1}.	330	187 mg L^{-1} PO$_4$$^{-3}$	13,757 mg L^{-1} NH$_4$$^+$-N	0.40 kWh^{-1}	Current: 3 A. Voltage: 60 V	15	200	[38]
Diluted human urine	Hybrid precipitation + nitrification + ED.	10 cell pairs; AM, CM.	1.8–2.1 L dilute; 0.5–2 L concentrate; electrode material: titanium coated with a mixed metal oxide platinum/iridium.	2880–4320 min for concentrate	340 ± 56 g L^{-1} K 23 ± 5 g P L^{-1}, 832 ± 98 NO$_3$$^-$-N, 25 ± 35 mg L^{-1} NH$_4$$^+$-N	1527 ± 349 g L^{-1} K, 59 ± 18 g P L^{-1}, 3528 ± 625 NO$_3$$^-$-N, 158 ± 168 mg L^{-1} NH$_4$$^+$-N		3.9 V 5 mA	61	64	[39]
Synthetic excess sludge	Conventional ED.	6 cell pairs; CM, AM.		480	100 mg L^{-1} P	1500 mg L^{-1} P	5.3 kWh kg^{-1} H$_3$PO$_4$	62.3 V		99	[29]
Synthetic excess sludge	ED with bipolar membranes (EDBM).	4 cell pairs, BM, From NEOSEPTA Inc.		300	100 mg L^{-1} P	1550 mg L^{-1} P	29.3 kWh kg^{-1} H$_3$PO$_4$	62.3 V	50	7.07	[29]
Domestic anaerobic digester supernatant	Pilot-scale ED.	30 cell pairs; pilot reactor.	Flow rate of 1250 mL min^{-1} (75 L h^{-1}). 200 L recirculated concentrated and electrode. Single-pass feed.	4320	835 ± 267 mg L^{-1} NH$_4$$^{+1}$-N, 232 ± 41 mg L^{-1} K	7100 ± 300 mg L^{-1} NH$_4$$^+$-N, 2490 ± 40 mg L^{-1} K	4.9 ± 1.5 kWh kg^{-1} NH$_4$$^+$-N	30 V		72,000	[21]
The effluent of an up-flow anaerobic	Struvite + selective ED.	3 cell pairs; CM, AM and monovalent		4000	2.5 mmol L^{-1} PO$_4$$^{-3}$	7 mmol L^{-1} PO$_4$$^{-3}$	16.6	0.2 A	3.125	64	[40]

Table 1 (*continued*)

Types of wastewater	Methods	Number of cell pairs, type of membrane.	Operation conditions	Operation time (min)	Initial concentration	product concentration	Energy consumption	Voltage (V) or current (A)	Current density (mA/cm²)	Effective area per membrane (cm²)	Ref.
sludge blanket reactor (USAB)		selective anion exchange membranes (MVAs).									
Secondary effluent	Selective electrodialysis.	Batch mode, 3 cell pair, CM, AM, MVA.	The electrodes were titanium coated ruthenium, with flow rate of 8 mL min⁻¹.	960	2 mg L⁻¹ N, P	22 mg L⁻¹ N, 40 mg L⁻¹ P	1.85 kWh m⁻³	5 V		25	[41]
Urine solution containing micropollutants	Conventional electrodialysis.	2 cell pairs; batch operated, Ralex AM, CM.	Flow rate: 0.96 L h⁻¹.		230 mg L⁻¹ PO₄⁻³-P, 4850 mg L⁻¹ NH₄⁺-N	610 mg L⁻¹ PO₄⁻³-P, 14,150 mg L⁻¹ NH₄⁺-N		3.6 V	22.5	49	[6]
Urine solution containing micro pollutants	Conventional ED.	Continuous, 5 cell pairs. CM.			230 mg L⁻¹ PO₄⁻³-P, 4850 mg L⁻¹ NH₄⁺-N			9 V			[6]
urine	ED + ozonation.	20 cell pairs, pilot.	960 L h⁻¹ continuously operated.		2.9 g L⁻¹ NH₄⁺-N, 0.18 g L⁻¹ PO₄⁻³-P	11.89 g L⁻¹ NH₄⁺-N, 0.738 g L⁻¹ PO₄⁻³-P		36 V		960	[42]
Anaerobic digesting excess sludge	Hybrid ED + struvite.	11 cell pairs.	Titanium coated with ruthenium.		7.5 mmol L⁻¹ PO₄⁻³-P, 50 mmol L⁻¹ NH₄⁺¹-N	80 mmol L⁻¹ PO₄⁻³-P, 600 mmol L⁻¹ NH₄⁺¹-N		62.0 V 1.7 A	21.25	80	[43]
Aqueous solutions	ED.	110 cell pairs; 30 L feed tank.		40	11 mg L⁻¹ NO₃⁻-N	22 mg L⁻¹ NO₃⁻-N		60		390	[44]
Synthetic wastewater	Selective ED.	3 cell pairs: CM, AM, MVA.	Volumes 3 L, batch mode,		0.32 mmol L⁻¹ NO₃⁻, 0.43 mmol L⁻¹ PO₄⁻³-P	1.6 mmol L⁻¹, 0.68 mmol L⁻¹ PO₄⁻³-P			2.8	180	[45]
Effluent from an equalization tank	Electrodialytic, continuous-flow reactor.	1 cell pair; CMX1AMX, 2 stages. Membranes International Inc.	34 l of wastewater.	600	3.52 mg L⁻¹ PO₄⁻³-P, 5,821 mg L⁻¹ NH₄⁺-N	12.52 mg L⁻¹ PO₄⁻³-P, 11.03 mg L⁻¹ NH₄⁺¹-N		0.75 A 24 V	5	200	[46]
Synthetic wastewater	Selective-ED + struvite	5 cell pairs, 5 repeating units, consisting of 5 MVK, 5 PC-SA, 4 PC-SK membranes, and 2 SC end membranes.	2:1:1:1 volume, 10.62 cm/s flow, electrodes made of titanium coated with ruthenium oxide.		40 mg L⁻¹ P, 500 mg L⁻¹ NH₄Cl-N	100 mg L⁻¹ P, 1000 mg L⁻¹ NH₄Cl-N		7.8 V	4.53	16	[35]
Synthetic wastewater	Conventional ED.	10 cell pairs, 2 PCA SC, cation exchange end	19 L h⁻¹; 10 batch system; Pt/Ir coated titanium anode	250	1.5 g L⁻¹ NH₄-N	10 g L⁻¹ NH₄-N		1.69	1.6	16	[47]

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Table 1 (*continued*)

Types of wastewater	Methods	Number of cell pairs, type of membrane.	Operation conditions	Operation time (min)	Initial concentration	product concentration	Energy consumption	Voltage (V) or current (A)	Current density (mA/cm²)	Effective area per membrane (cm²)	Ref.
Synthetic municipal wastewater	Selective ED	(CEEM), 10 PCA SA standard anion exchange (AEM), 9 PCA SK standard cation exchange membranes. 3 cell pairs; 4 AM, MVA, CM from PCCELL company	and a V4A steel cathode. a flow rate of 20 L h⁻¹, volume of feed to product 10/2 L	300	1.00 ± 0.06 mM H₃PO₄	2.00 ± 0.06 mM H₃PO₄		1.6	6.25	64	[48]

Table 2
Correlation between independent variables.

		Recovery rate	Voltage	Cell pair
Pearson correlation	Recovery rate	1.000	0.880	0.508
	Voltage	0.880	1.000	0.309
	Cell pair	0.508	0.309	1.000
(p-Value)	Recovery rate	.	0.000	0.046
	Voltage	0.000	.	0.164
	Cell pair	0.046	0.164	.

Table 3
Estimated model coefficients.

Model	Unstandardized coefficients		Standardized coefficients		t	p-Value
	B		Std. Error		Beta	
1	(Constant)	1.607		1.054		
	Voltage	0.129		0.023	0.800	
	Cell pair	0.329		0.179	0.261	

which means that the linear regression explains 83.36% of the variance in the data; in other words, the regression model is significant.

Table S2 in the Supplementary material indicates whether the overall regression model is a good fit for the data. The significance associated with this value is minimal (0.0000). These values confirm that the independent variables reliably predict the nutrient recovery rate by ED.

In the following, we will discuss each of the research studies presented in Table 1. However, some of these have already been discussed in the introduction and so they will not be discussed again in this section.

The ED process selectively recovers nutrients from a wide range of wastewater effluent containing various ions. Although, nutrient recovery efficiency and product purity were significantly improved when a bipolar membrane was employed in the ED process. An ED process with a bipolar membrane (BM) provided H^+ and OH^- in situ without the introduction of salts. The combination of H^+ and anions in individual chambers leads to the production of acid, while the combination of OH^- ions and cations in other chambers leads to the formation of the corresponding base. As a result, an ED process with the bipolar membrane concept could diversify the final products and enhance purity for nutrient recovery [49]. For example, Shi et al. [38] demonstrated the feasibility of nutrient and volatile fatty acid recovery from pig manure by employing an ED system using bipolar membranes (Fig. 4). In their study, 78% of the ammonium and 75% of phosphate ions were recovered in a lab-scale system. Similarly, Wang et al. [29] employed an ED process with a bipolar membrane to convert the phosphate in sludge supernatant into purified phosphoric acid.

In general, most membranes have no means of selecting different anions and cations. Some membranes are selective of monovalent ions compared to multivalent ions and fractionation effect can be achieved in this way [45]. In the study by Ward et al. [21] and Liu et al. [41] application of mono selective anion exchange membranes (MVA) showed the capability to separate monovalent anions from the solution containing multivalent anions (one concentrate compartment with nitrate and another concentrate compartment with phosphate) resulted in more manageable application as fertilizers devoid of the requirement of extra activities to enable the suitability of the concentrate for certain soil conditions.

Using this approach, Zhang et al. [40] used selective electrodialysis (SED) and a hybrid system (ED + struvite) to enhance the phosphate recovery of the effluent's phosphate in an up-flow anaerobic sludge blanket reactor. The effluent was shifted to the recycled effluent of the struvite reactor and then electrodialysis was applied. The primary
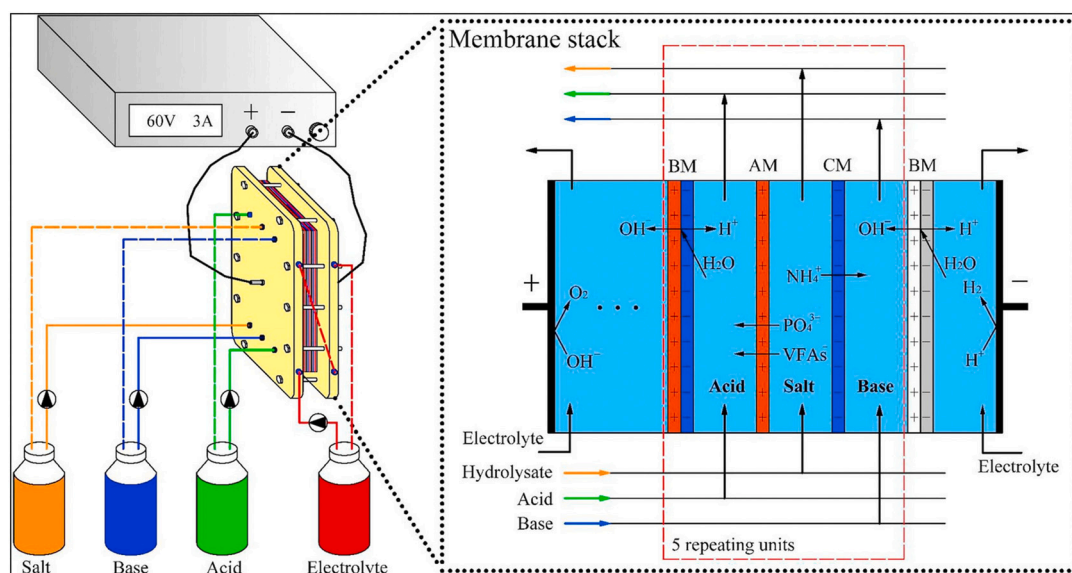
Fig. 4. A schematic of the BMED apparatus. The BMED membrane stack is composed of five units that contain CEM, AEM, and BM. The membrane compartments are connected to the salt (5 L of pig manure hydrolysate), base, acid (1 L of deionized water), and electrolyte containers (1 L of 0.1 mol L^{-1} Na$_2$SO$_4$ solution). Reproduced from Shi et al. [38] with permission from Elsevier.

current efficiency attained a level of 72%, with a satisfactory phosphate concentration of 9 mmol L^{-1}. From the experiment's cost evaluation results, it was evident that 1 kWh of electricity was able to generate 60 g of phosphate in a full-scale stack, at a 95% desalination level for the feed wastewater. In another study, the separation of sulfate from a NaCl/Na$_2$SO$_4$ mixture was investigated; the results showed that the sulfate purity could reach over 85%, and the selective electrodialysis process could be predicted under optimized conditions [45]. In a study carried out by Tran et al. [48] nutrient selectivity in a selective ED process further enhanced by either adjusting the pH range of feed stream or increasing current density.

As it was discussed in Section 1, it is more convenient to run electrodialysis with higher conductivity and fewer fouling compounds in the feed wastewater. Hybrid systems have been used in some studies to improve the efficiency by removing organics or other nonionic pollutants and also by increasing the concentration of the ED feed (waste) water. For instance, De Paepe et al. [39] operated a pilot-scale process combining precipitation, nitrification, and electrodialysis (ED), for concentrating urine. In other research, Mondor et al. [37] produced a concentrated nitrogen fertilizer from liquid swine manure using electrodialysis (ED) and reverse osmosis (RO) that offered an alternative to chemical nitrogen fertilizer production. The maximum achievable total NH$_3$–N concentration was measured experimentally for both processes, and similar values were obtained at approximately 13 g L^{-1} [37]. In like this manner, Ippersiel et al. [26] applied direct aeration or a vacuum system to isolate the transmitted ammonia volatilization from a concentrated solution produced by ED in an acidic trap.

3.2. The effect of voltage and operating time

There are two standard power operation modes for the electrodialysis: constant current and constant voltage [35]. Both electrodialysis operation modes achieved similar performance in a study on simultaneous nutrient ion fractionation by Ye et al. [35], which was also confirmed by comparing the recovery efficiency, energy consumption, and current efficiency ratio, as presented in Table 4. Under a constant current, more ions were transported from the feed stream into the product and brine chambers, and consequently the electrical resistance of the feed stream increased significantly [35]. It has also been shown that the current decreased in constant voltage mode, whereas a

drastic increase in the voltage (15.7 V) at the end stage was detected in a constant current mode. It was found that such a high noncontrollable voltage in constant current mode would damage the membrane near the electrode during lengthy operation [35].

Increasing the voltage in ED can enhance the transfer of nutrients across the ion exchange membrane, which resulted in faster desalination and recovery rates [49]. However, when ED operations at such a high voltage may also reduce efficiency. The applied current in this condition is referred to as the limiting current, and its density is called the limiting current density (LCD) [51]. The main reason for this is that the number of ions in this voltage area is not sufficient to carry the current and this increases the cell resistance. Therefore, the dissociation of water occurs and generates H$^+$ and OH$^-$ ions, which consume energy and this leads to less salt removal [49,50]. It was also shown by Rottiers et al. [52] that when the concentration ratio between the dilute and concentrate stream becomes too high, back diffusion occurs.

On the other hand, also shown by Banasiak et al. [53] that a very low voltage does not generate a sufficient current between the cathode and anode and cannot overcome the resistance of the membranes and ion transfer; therefore, the removal of ions at lower voltage values is slow. Large ED systems with numbers of stacks in series operate with different current densities in each stack based on the changes in the feed water salt concentration and conductivity in series methods. Fig. 5 shows a scatter plot and the linear regression of the nutrient recovery rate with the voltage/cell pair based on our studies represented in Table 1. This figure shows that the increases in the voltage in the examined studies were correlated with increases in nutrient recovery rate by equation of:

Table 4
Fractionation ratio, energy consumption, and current efficiency under different operational modes according to data by Ye et al. [35].

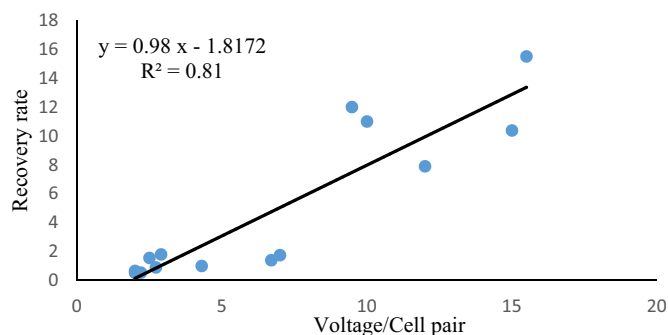|	Item	Constant current	Constant voltage
Recovery efficiency (%)	NH$_4^+$	56.2%	63.2%
	PO$_4^{-3}$	87.1%	89.6%
Energy efficiency	kWh kg^{-1} NH$_4^+$	0.949	0.783
	kWh kg^{-1} PO$_4^{-3}$	33.717	28.380
Current efficiency	NH$_4^+$	25.27%	30.23%
	PO$_4^{-3}$	2.54%	4.16%

Fig. 5. Scatter plot and regression line of the nutrient recovery rate with respect to voltage/cell pair with uncertainties. The chart is drawn from the data summarized in Table 1.
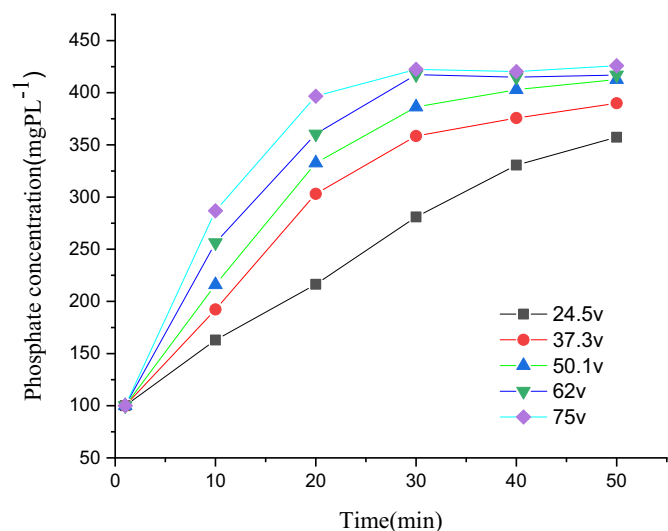

Fig. 6. Effect of voltage on phosphate concentration over operating time. (Data from Wang et al. [29].)

Nutrient recovery rate $= (0.98 \times \text{Voltage/cell pair}) - 1.87$

where R^2 is 0.81.

Fig. 6 shows the change of phosphate concentration as a nutrient in the sludge permeate under different operating voltages over the operating time by Wang et al. [29] as a case study. Based on Fig. 6, it can be noted that increasing the voltage improves the recovery rate and shortens the working time. It was found that the concentration trend-line changed over time from a linear regression at a lower voltage (Fig. 7) to a non-linear at a higher voltage (Fig. 8); the final phosphate concentration was steady or even dropped at a higher voltage [29]. This phenomenon can be explained by both the concentration polarization in the membrane boundary layer, as well as the depletion of electron carriers in the dilute part [29]. In this case, the model is an association type of exponential model. Based on the figure, a maximum phosphate concentration of $417.4 \, \text{mg L}^{-1}$ can be achieved in 30 min with an optimum voltage of 62 V. Fig. 8 shows the exponential regression between phosphate concentration in respect to the operating time for different voltages in sludge permeate. The general form of the equation for Wang et al. [29] study can be obtained from Table 5 as follows based on average data:

$$Y = y_0 + A * \exp(R_0 * x)$$

where: Y is phosphate concentration as the dependent variable (mg L^{-1} P), and X is time (min) as an independent variable, y_0 is the phosphate concentration value when X (time) is zero. The convergence between the experimental and theoretical results confirms the reliability of the model.

Similarly, larger voltages have shown improved ion transport in other studies [45,53,54]. In a further study by Wang et al. [41] the recovery rate increased as the processing capacity of the IEM stack increased with the increasing applied voltage from 3 to 5 V. However, water dissociation occurred when the voltage was increased to 7 V, which reduced the current efficiency and was also confirmed by a mathematic fitting analysis. Balster et al. [55] realized that calcium transport through a cation exchange membrane (CEM) was low at lower current densities, but that it increased sharply at higher current densities [55]. The results by Hanrahan et al. [56] showed that at low voltages, the repulsion force of CEM hinders calcium transport, but at higher applied voltages the electrical driving force overcomes this repulsion, significantly reducing the influence of the rejection on calcium ion transport.

3.3. The effect of the number of cell pairs and the total effective area of the membranes

One of the most significant parts of the capital cost of ED is associated with the value of the ion exchange membrane [57]. How the ion exchange membrane stack is arranged in the ED process is called staging. The staging provides a sufficient membrane area and a sufficiently long residence time to remove more salt from the dilute stream [58]. The capacity can be enhanced with a greater number of cell-pairs within a stage or by adding another stack in parallel.

However, the design of dilute and concentrate cells should as thin as possible and the stress on the membranes should be minimized by keeping a low-pressure difference between the dilute and concentrate cells [58]. Brauns [59] demonstrates that the development of new, thinner membranes can significantly enhance the process performance. Generally, the total effective area and number of cell pairs are cross-related. However, we analyzed their effect on efficiency separately to determine which parameter is more important for design purposes.

Fig. 9 shows a scatter plot and linear regression between the nutrient recovery rate and total effective area for various wastewater based on the data of our study in Table 1. The equation can be obtained as follows:

$$Y = 1.91 + (0.01 \times \text{total effective area})$$

where $R^2 = 0.55$.

The scatter plot shown in Fig. 10 summarizes the results for the relation between the nutrient recovery and numbers of cell pair for
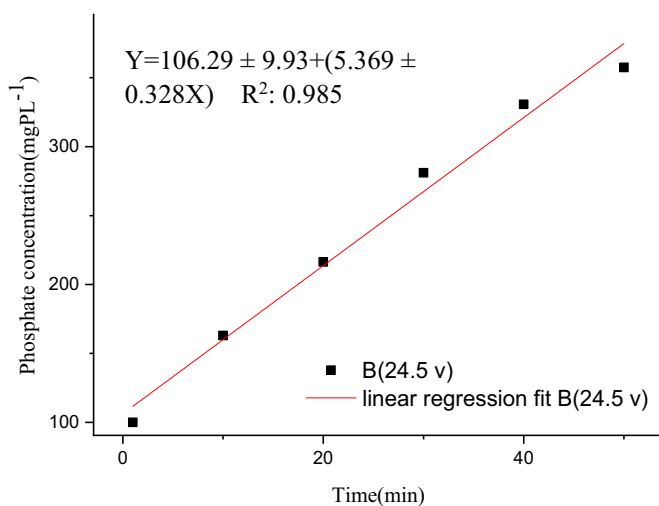

Fig. 7. Linear regression between phosphate concentration with respect to the operating time in 24.5 v (data from Wang et al. [29]). The markers represent experimental values, while the solid lines represent the fitted equation.
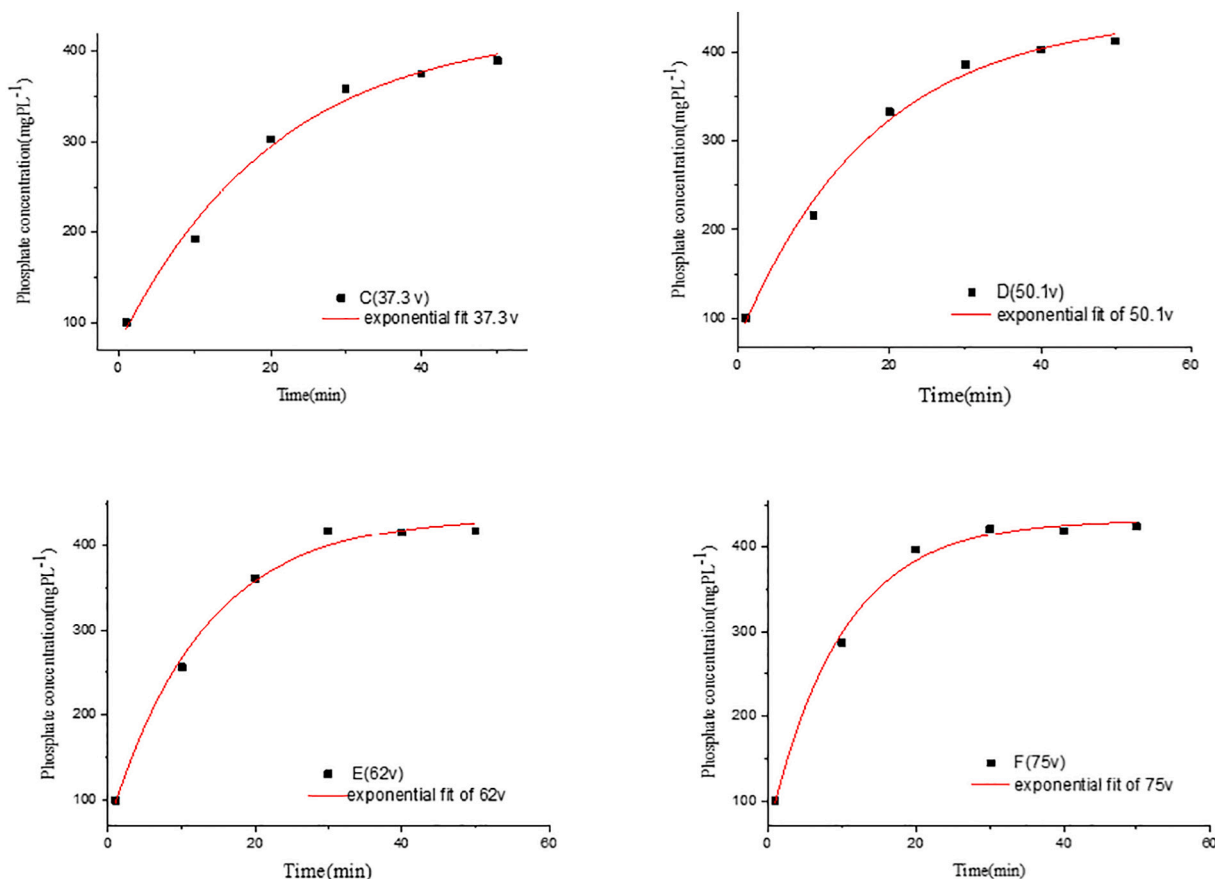
Fig. 8. Regression between phosphate concentration in respect to the operating time for different voltages (data from Wang et al. [29]). The markers represent experimental values while the solid lines represent the fitted equations.

Table 5
Equation formula and data for a non-linear regression with uncertainties between the phosphate concentration in respect to the operating time at different voltages (data from Wang et al. [29]).

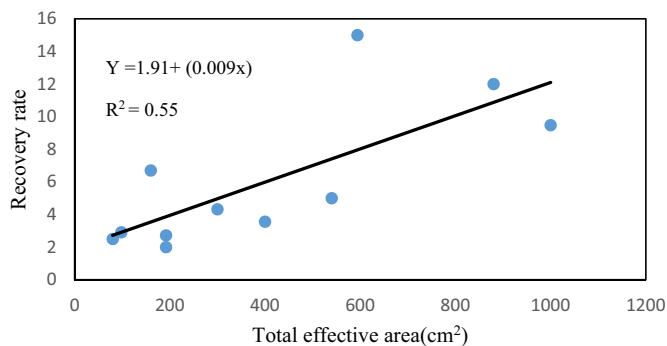| Model	Exponential			
Equation	$y = y_0 + A * \exp(R_0 * x)$			
Plot	C (37.3 V)	D (50.1 V)	E (62 V)	F (75 V)
y_0	428.139 ± 27.50	441.974 ± 20.07	434.019 ± 13.01587	433.18 ± 8.68
A	−351.62 ± 26.108	−368.049 ± 20.80	−364.87 ± 17.84	−371.204 ± 15.03
R0	−0.048 ± 0.010	−0.056 ± 0.009	−0.0783 ± 0.01	−0.10 ± 0.01
Reduced Chi-sqr	233.498	201.574	192.10	136.32
R-square (COD)	0.989	0.992	0.992	0.995
Adj. R-square	0.982	0.987	0.988	0.99

Fig. 9. Scatter plots and regression line of the nutrient recovery rate with respect to the total effective area of the membranes. The chart is drawn from the data summarized in Table 1.
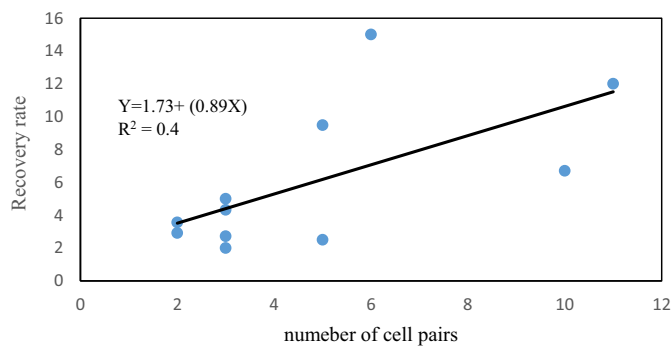
Fig. 10. Scatter plots and regression of the nutrient recovery rate with respect to the number of cell pairs. The chart is drawn from the data summarized in Table 1.
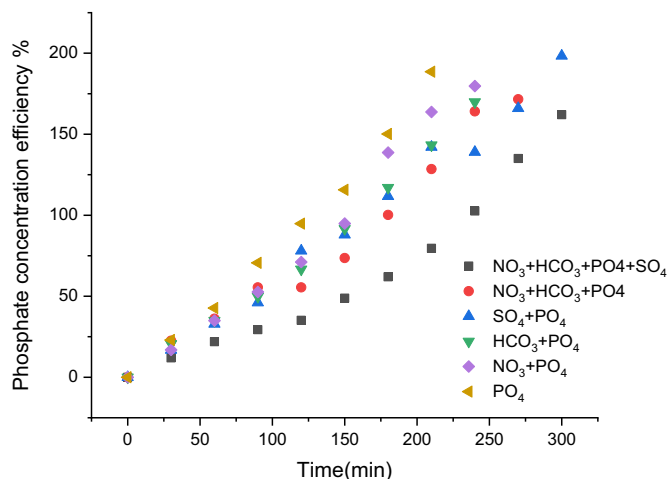
Fig. 11. Effect of different ions on the phosphate concentration efficiency over operating time.
(Data from Tran et al. [48].)

various types of wastewater from our studies in Table 1. The predicted recovery rate is:

$$Y = 1.73 + (0.89 \times \text{cell pairs numbers})$$

where $R^2 = 0.4$.

The above regression analysis shows that the nutrient recovery rate in different wastewater systems depends mostly on the total effective area of the membrane in the selected studies. Furthermore, there is a correlation between the number of cell pairs and the total effective area, as by increasing the number of cell pairs, the total effective area also increases.

The required membrane area can be calculated from the current density and the feed and product solution concentrations base on the following equation:

$$A = \frac{Q_{st}^d F (C^f - C^d)}{i\xi}$$

where A is the membrane area, Q is the flow, i is the electric current density for a cell pair, C is the concentration, F is the Faraday constant, and ξ is the current utilization. The subscript st refers to the stack, and the superscripts d and f refer to the dilute and feed solution [24].

3.4. The effect of the feed composition

Since wastewater contains a variety of ionic species, a key consideration is the competitive impact of these ions on the nutrient concentration efficiency [23]. Tran et al. [48] studied the nutrient recovery efficiency in terms of phosphate from a single and multi phosphate feed solution for 2 mM, bicarbonate, nitrate, and sulfate either alone or in combination under a current density of 62.5 A m^{-2} (Fig. 11). Fig. S2 in the Supplementary material displays the linear regression and correlation equation when only phosphate is present in wastewater and Fig. S3 in the Supplementary material shows the regression and equation for phosphate concentrating efficiency when all major competing anions are present in feed wastewater. It can be seen from Fig. 11 that the phosphate concentrating efficiency in the product slightly decreased when adding nitrate, bicarbonate, and sulfate to the feed solution. The concentration efficiency after 210 min was 188% without an ionic effect and was reduced to the range of 142.5–162.5% with the addition of competing ions. In this case study when three anions were added at the same time, the efficiency of the phosphate concentration process was reduced from 188% to 78.6% in 120 min. However, this effect became insignificant if the system was run for a more extended time, and the final concentrating efficiency was about 170%–198% for a single phosphate feed, while it was about 160% with the simultaneous effects
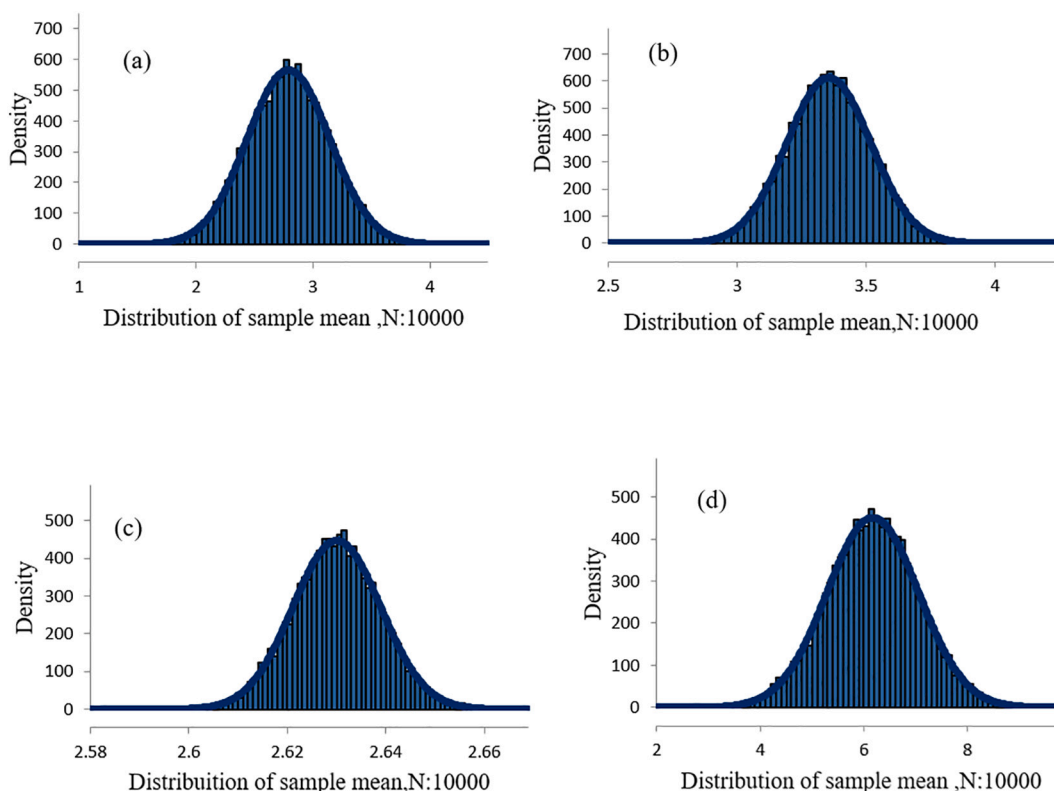

Fig. 12. Histogram of the Monte Carlo simulation for: a) the total regression equation represented, b) the regression equation for the effect of voltage/cell pairs, c) the regression equation for the effect of the effective area, and d) the regression equation for the effect of cell pairs.

Table 6
The statistical parameters corresponding to the histogram of regression equations.

Process outputs	Total equation	Effect of voltage	Effect of effective area	Effect of cell pair numbers
Equation formula	1.6 + (voltage × 0.129) + (number of cell pairs × 0.329)	(0.98 × voltage / cell pair) − 1.87	1.91 + (0.01 × total effective area)	1.73 + (0.89 × number of cell pairs)
Number of simulations	10,000	10,000	10,000	10,000
Mean	2.790	3.354	2.630	6.170
Std dev.	0.353	0.162	0.008	0.885
Median	2.792	3.354	2.630	6.175
Normal distro statistics				
KS test p-value (normal)	> 0,15	> 0,15	> 0,15	> 0,15
dpm	0	0	0	0

of three ions [48]. The electromigration of ions in the ED process is influenced by both the ion mobility and the ion exchange selectivity. The mobility of the ions is dependent on ion characteristics such as the diffusion coefficient, ion charge, and the applied electrical field [60]. Therefore, since different ions have varied diffusion coefficients and may react differently to the fixed and surface charges of the IEM, diverse levels of voltage have different effects on different ions [61]. Kabay et al. [62] report that a voltage change from 5 to 10 V affects the removal of monovalent ions more than the removal of divalent ions [62]. Similarly, enhancing the voltage accelerated the migration of monovalent anion in the study by Ye et al. [35] and Shi et al. [38] due to their small hydrated radius size. The hydrated size can control the selectivity of ion-exchange membranes for a specific ion in an electrical field. According to a study by Awual et al. [63], Ions with smaller intrinsic crystal radii have a higher hydration number, larger hydrated radii and hold their hydration shells more strongly, and this is more attractive for the ion exchange membrane. The general anion selectivity anion exchange and Hofmeister series were reported by Elmidaoui et al. [64] was in the order of: $I^- > (\sim Br^-) > NO_2^- > Cl^- > OH^- > SO_4^{-2} > F^-$ As to the cations, the permeation sequence was: $NH^{+4} \approx K^+ > Ca^{+2} > Mg^{+2} \approx Na^+$ in Ye et al. [35] study.

The other parameters that control the selectivity of ion-exchange membranes are the rejection of ions with the same charge, as well as the thin boundary layer on the membrane [23]. For example, calcium is more favored by CEM due to the higher ionic charge than potassium. In contrast, the boundary layer is preferential for potassium due to its 3 times greater diffusivity than calcium. In an ohmic regime under a limited current, where the concentration polarization is negligible in the boundary layer, the CEM selectivity for calcium is dominant compared to the concentration polarization-driven transport of potassium. On the other hand, in a non-ohmic regime with a higher voltage, the concentration polarization in the boundary layer is increased, and the potassium transport is enhanced due to a steep concentration gradient [65]. Further works will be devoted to investigating the effect of co-existing cations/anions on ion exchange membrane and electrodialysis performance.

3.5. Uncertainty of regression equations

A Monte Carlo simulation was set to run N: 10,000 trials of the proposed models, using the first described input sources. Fig. 12a, b, c & d shows the final histogram representing the possible values for the real nutrient recovery of the data reported in Table 3 (the total equation), Fig. 5 (the effect of the voltage), Fig. 9 (the effect of the total effective area) and Fig. 10 (the effect of cell pairs), respectively.

Table 6 shows the statistical parameters obtained corresponding to the histogram. Additionally, the Monte Carlo simulation and statistical parameters of data reported in Figs. 7 and 8 are shown in the Supplementary data in Fig. S4 and Table S3. Fig. S5 and Table S4 in the Supplementary data are shown in the histogram of the Monte Carlo simulation and statistical parameters for the effect of different ions on the phosphate concentration efficiency over the operating time for the data reported in Figs. S2, and S3 in the Supplementary data.

3.6. Costs of ED

For nutrient recovery to become applicable in industry and for it to be a viable option, the process must be cost-effective, simple to operate and maintain, and it should have similar treatment efficiency as conventional treatment. Moreover, there must be a market for recovered nutrient products. The development of low-cost technology to recover nitrogen from wastewater is essential to achieve economic and environmental benefits. Table S5 compares the energy and water production costs of ED with other membrane technologies. These technologies consist of reverse osmosis (RO), forward osmosis (FO),
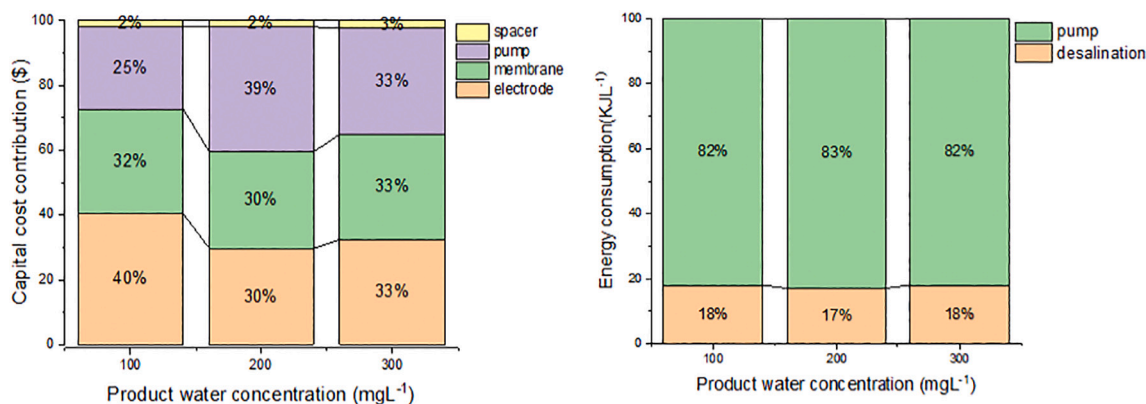
Fig. 13. The capital cost and the average energy comparison of optimal systems of a batch electrodialysis system for 3 different product water concentrations. (Data from Shah et al. [66].)

electrodialysis (ED) and nanofiltration (NF). As can be seen from the table, ED is an economical process with low energy consumption compared to the other membrane technologies.

Generally, an ED plant involves both capital and operational costs. The operational costs are associated with electric energy and pumping for daily desalination and recovery [66], as well as labor and maintenance. The labor and maintenance costs are directly related to the plant size, and usually, a specific ratio of the investment-related costs is considered for that purpose [24].

The desalination energy is expressed as the amount of energy spent on the electromigration of ions through the membranes and the solution and the pumping energy used for the flow of the solution through the piping and the stacks [67,68].

Fig. 13 shows the capital cost of a batch electrodialysis system for 3 different product water concentrations in Shah et al. [66] study. Pumps accounted for a significant fraction, particularly at 200 and 300 mg L^{-1}, and the rest of the costs are related to the electrodes and membranes. Fig. 13 also illustrates the percentage of different operational costs for desalination and pumping when increasing the product concentration. Energy for pumping accounted for 83% of the total energy consumption.

The development of low-cost and energy-efficient pumps that are suitable for flow-rates ranging between 200 and 300 L h^{-1}, while sustaining pressures up to 2 bar, will assist the commercialization of domestic ED systems [66]. The most significant component of the capital cost is to provide the membrane area required for the desired production capacity. This segment of the capital cost is impacted by the prices of the membranes plus the membranes' lifetime; this life is much dependent on the operating conditions and the quality of the feed water [69]. The main reasons for the replacement of membranes are scaling and fouling and increased membrane resistance due to the sorption of iron ions, degradation of the anion-exchange membranes in the alkaline solution, and depreciation of the membranes near the anode chamber due to oxidation caused by electrolysis reaction [67,68].

3.7. The challenge of fouling and scaling in ED process

Fouling is a significant drawback affecting membrane applications in nutrient recovery from wastewater by electrodialysis over longer operation times. It should be mentioned that fouling and scaling are dependent on the feeding water quality and components. This phenomenon causes an increase in the electrical resistance of the membrane, which raises the energy costs of electrodialysis [70]. An anion exchange membrane is more vulnerable to organic fouling since most of the colloids and organic foulants are negatively charged, which means they are strongly attracted by the positive charges of the anion exchange membranes. On the other hand, CEM is more prone to inorganic fouling or scaling [71,72].

Spacers also might have an essential role in fouling formation in water compartments. A recent study reports that spacers were more susceptible to biofouling than membranes [16]. It might be due to their surface charge, in comparison to uncharged spacers [70], as well as the low speed and turbulence on some parts of the spacers.

Inverting the polarity or applying an electrodialysis reversal (EDR) system could be the best option because no extra chemicals are required. In EDR, when the polarity of the electrodes is reversed, foulants attaching to the membrane surface break apart and move in the opposite direction [73]. Some researchers have been investigating how to improve the antifouling potential of an anion exchange membrane through surface modification [74–77]; however, their studies have only been tested on the laboratory scale. There is also some disagreement between researchers regarding the effectiveness of pulsed electric fields (PEF) on fouling mitigation [30,78].

4. Conclusion

The present research on ED applications for nutrient recovery from different types of wastewater was technically analyzed in order to gain in-depth knowledge. Regression equations between the nutrient recovery rate in electrodialysis and different operational variables were developed for different types of wastewater. The variables for the voltage and number of cell pairs showed an 83% integrated effect on nutrient recovery. The regression calculation of the total effective membrane area represents a 55% contribution towards improving the recovery rate of ED in different studies. The results of the case study showed the downside effect of other components present in feed wastewater on nutrient recovery.

The operating and capital costs of ED technologies for nutrient recovery were discussed in this research review and showed that pumping accounted for 80% of the energy consumption. Regarding the capital costs, the pump, membrane, and electrode play an essential role. ED applications for nutrient recovery still need more research to promote and optimize their application for processing municipal wastewater.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.desal.2020.114626.

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