

Minimal Bipolar Membrane Cell Configuration for Scaling Up Ammonium Recovery

Mariana Rodrigues, Thiago T. de Mattos, Tom Sleutels, Annemiek ter Heijne, Hubertus V.M. Hamelers, Cees J.N. Buisman, and Philipp Kuntke*

Cite This: *ACS Sustainable Chem. Eng.* 2020, 8, 17359–17367

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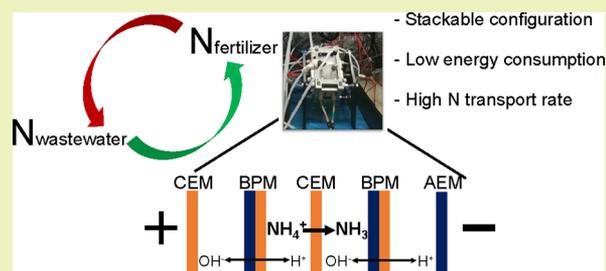
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ABSTRACT: Electrochemical systems for total ammonium nitrogen (TAN) recovery are a promising alternative compared with conventional nitrogen-removal technologies. To make them competitive, we propose a new minimal stackable configuration using cell pairs with only bipolar membranes and cation-exchange membranes. The tested bipolar electro dialysis (BP-ED) stack included six cell pairs of feed and concentrate compartments. Critical operational parameters, such as current density and the ratio between applied current to nitrogen loading (load ratio), were varied to investigate the performance of the system using synthetic wastewater with a high nitrogen content as an influent ($\text{NH}_4^+ \approx 1.75 \text{ g L}^{-1}$). High TAN removal ($>70\%$) was achieved for a load ratio higher than 1. At current densities of 150 A m^{-2} and a load ratio of 1.2, a TAN transport rate of $1145.1 \pm 14.1 \text{ g}_\text{N} \text{ m}^{-2} \text{ d}^{-1}$ and a TAN-removal efficiency of 80% were observed. As the TAN removal was almost constant at different current densities, the BP-ED stack performed at a high TAN transport rate ($819.1 \text{ g}_\text{N} \text{ m}^{-2} \text{ d}^{-1}$) while consuming the lowest energy ($18.3 \text{ kJ g}_\text{N}^{-1}$) at a load ratio of 1.2 and 100 A m^{-2} . The TAN transport rate, TAN removal, and energy input achieved by the minimal BP-ED stack demonstrated a promising new cell configuration for upscaling.

KEYWORDS: ammonium recovery, upscaling bipolar electro dialysis, stackable configuration, cell pairs



INTRODUCTION

Currently, we rely on energy-intensive processes for ammonia production (Haber–Bosch process, $37 \text{ kJ g}_\text{N}^{-1}$) and subsequent nitrogen removal (nitrification/denitrification process, $46 \text{ kJ g}_\text{N}^{-1}$) in wastewater treatment plants (WWTP).^{1,2} For example, the sludge produced during the wastewater treatment is frequently processed by anaerobic digestion for methane production.^{3,4} After digestion, the sludge dewatering process results in a nitrogen-rich stream known as reject water or a centrate. The centrate contains between 400 and 2400 $\text{mg}_\text{N} \text{ L}^{-1}$.^{5–7} Therefore, the centrate requires further treatment before discharge, for example, by the anammox process (requiring an energy input of $16 \text{ kJ g}_\text{N}^{-1}$).^{7–12}

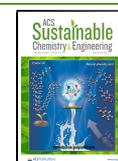
Recently, electro dialysis (ED) has been proposed as an alternative to conventional TAN (total ammonium nitrogen) removal processes (nitrification/denitrification) as it allows subsequent nitrogen recovery instead of merely removing it.^{10,13–17} ED has already been demonstrated as one of the most energy efficient systems to remove nitrate from drinking water and wastewater.^{16,18,19} ED is a separation process based on ion-exchange membranes where positive ions are transported through a cation exchange membrane (CEM), and negative ions are transported through an anion exchange membrane (AEM) due to an applied electric field.²⁰ In recent years, the application of bipolar membranes (BPM) in ED

technology has gained increasing attention, considering their unique behavior and properties such as high permselectivity and a high rate of water splitting.^{21,22} BPMs dissociate water in protons and hydroxide, which move through the cation and anion layers of the BPM, respectively, and thereby produce acidic and alkaline streams.^{23,24} This allows for stacking of compartments with fewer electrodes while still supplying the protons/hydroxide also provided by the electrode oxidation/reduction reactions. BPMs have a lower energy consumption compared to water electrolysis that produces hydrogen and oxygen gases at the electrodes (79.9 vs $198.5 \text{ kJ mol}^{-1}$).²³ Other advantages of replacing the electrodes include the absence of reduction/oxidation species that could lead to unwanted by-products such as chlorine gas or halogenated organic compounds. In established bipolar ED (BP-ED) configuration, cell triplets (acidic concentrate, diluate, and alkaline concentrate) are used following the order: anode|CEM|BPM|AEM|...|CEM|BPM|AEM|cathode.^{25,26} Ions are re-

Received: July 9, 2020

Revised: November 2, 2020

Published: November 18, 2020



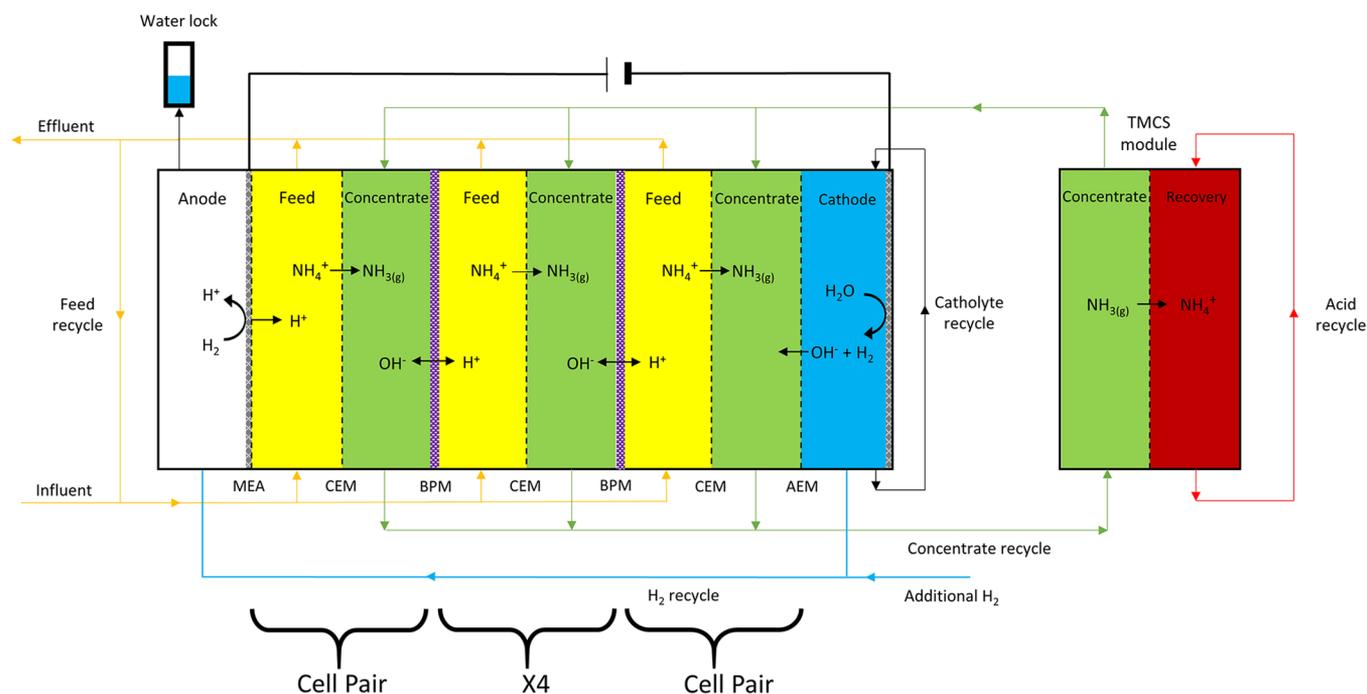


Figure 1. Scheme of the minimal cell configuration of the electrodiolysis cell with a bipolar membrane (BPM) coupled with a cation exchange membrane (CEM) as a cell pair. The minimal bipolar electrodiolysis cell configuration includes six cell pairs of feed and concentrate compartments between the anode and the cathode compartments. The BPM dissociates water into protons and hydroxyl ions. The cation side of the BPM supplies protons to the feed compartment. The protons protonate ammonia into ammonium, which can cross over the CEM to the concentrate compartment. On the other side, the anion side of the BPM, the hydroxyl ions pass to the concentrate compartment. Here, the hydroxide reacts with the NH_4^+ , forming NH_3 . NH_3 can then be recovered in acid of the TMCS module (transmembrane chemisorption unit, membrane stripping unit). The membrane electrode assembly supplies H^+ to the first feed compartment next to the anode. The anion exchange membrane works as a shielding membrane for the cathode (only OH^- was transported to the concentrate). The hydrogen gas formed at the cathode is then reused at the anode, decreasing the energy requirement.

moved from the diluate stream due to an applied electric field. Cations move from the diluate through the CEM toward the alkaline concentrate, while anions move from the diluate through an AEM toward the acidic concentrate. This configuration of the membranes (BPM, CEM, and AEM) has previously been used in BP-ED processes such as desalination or reverse electrodiolysis (RED).^{21,27,28} Pronk et al., in 2006, were first to report BP-ED for TAN recovery from urine in combination with a mass transfer unit (i.e., bubble columns or a gas-filled (hydrophobic) membrane) to obtain a product containing ammonium and phosphate.²⁵ Their results showed that ammonia was removed from urine, but also, undesired carbonate was recovered in the basic concentrate, limiting the purity of the product.²⁵ More recently, van Linden et al., in 2020, operated a BP-ED system in batch containing 10 cell triplets (acid, diluate, and base) and achieved up to a 90% ammonium removal from a synthetic medium (NH_4HCO_3 solution) at an energy input of $19 \text{ kJ g}_\text{N}^{-1}$.²⁶

To make the established BP-ED a more competitive technology for TAN recovery, we propose a new minimal cell configuration (Figure 1). The most important difference with previously reported BP-ED is that each cell pair in the stack has only two compartments instead of three, and no AEMs are employed except one facing the cathode. Therefore, no concentrated acid stream is formed. As a result, fewer membranes and compartments are needed, and therefore, the costs of the stack are reduced, and less energy is required.

The anode supplies protons generated in the MEA to the adjacent feed compartment and at the cathode hydroxide ions cross through an AEM to the adjacent concentrate compart-

ment. Inside the BP-ED stack, the BPMs supply protons to the feed and hydroxide ions to the concentrate compartment, thereby taking over the function of the electrodes (Figure 1). This leads to the formation of a pH gradient. The protons in the feed compartment, where the influent is supplied, protonate ammonia to ammonium, which can be transported through the CEM toward the concentrate in the direction of the cathode. The ammonium ions are then retained in the concentrate as they are blocked by the AEM side of the BPM or by the AEM next to the cathode. Simultaneously, ammonium is deprotonated to ammonia in the concentrate as a result of hydroxide ions transported through the anion side of the bipolar membrane or the AEM from the cathode. The ammonia can then be recovered from the concentrate stream in the TMCS module (transmembrane chemisorption, membrane stripping unit).²⁹ During membrane stripping, the ammonia is separated from the other cations in the concentrate and can be recovered in an acid solution as a fertilizer [$(\text{NH}_4)_2\text{SO}_4$].^{1,30}

In this BP-ED stack, the hydrogen produced at the cathode was recycled to the anode and oxidized to provide energy to operate this system at low energy input. Hydrogen recycling has been shown to provide half of the energy required for TAN recovery.²⁹

The main goal of this study is to assess the performance of this new minimal BP-ED stack configuration, consisting of six-stacked cell pairs, for TAN recovery from synthetic wastewater. In this work, we demonstrate that stacking ED cell pairs enhances the TAN transport rate, TAN removal and recovery

efficiencies, at low energy consumption in a continuous operation mode.

EXPERIMENTAL SECTION

Setup Configuration. The minimal BP-ED stack configuration comprised one anode, six feed compartments and six concentrate compartments (alternated), and one cathode. Figure 1 shows a schematic representation of the setup.

The ED stack materials were supplied by REDstack BV (Sneek, The Netherlands). The stack included a gas compartment for the anode and a cathode chamber. Both in the anode and cathode compartments, a Pt-coated titanium mesh electrode (9.8 cm × 9.8 cm × 0.2 cm, 5 mg of Pt cm⁻² MAGNETO special anodes B.V., Schiedam, The Netherlands) was used. Twelve polypropylene spacers (14 cm × 14 cm × 0.05 cm, 53% open) with a silicon gasket layer (2 cm × 14 cm) at two opposing sides for sealing were used as a feed and concentrate compartments (DEUKUM GmbH, Frickenhausen, Germany).

The anode was separated from the feed compartment by a membrane electrode assembly (MEA). The MEA was composed of an electrode for gas oxidation, facing the anode, coated on a CEM, facing the feed side. The MEA was a commercial 14 cm × 14 cm Nafion N117 CEM coated with a 10 cm × 10 cm platinum Vulcan (carbon) catalyst (0.5 mg Pt cm⁻²) and an integrated gas diffusion layer (GDL) purchased from FuelCellsETC (College Station, TX, USA).

The feed compartments were separated from the concentrate compartments on one side by CEMs. The CEMs used were fumasep FKB-PK-130 (14 cm × 14 cm, Fumatech BWT GmbH, Bietigheim-Bissingen, Germany). On the other side, BPMs (fumasep FBM-PK, 14 cm × 14 cm, Fumatech BWT GmbH) were used to separate the concentrate compartment from the feed compartment.

The last concentrate compartment was separated from the cathode by an AEM (14 cm × 14 cm, fumasep FAB-PK-130, Fumatech BWT GmbH). Thus, in total, one MEA, six CEMs, five BPMs, and one AEM were used in the following order from the anode to cathode: MEA|CEM|BPM|CEM|BPM|CEM|BPM|CEM|BPM|CEM|BPM|CEM|AEM. Each membrane had a projected surface area in contact with the electrolyte of 100 cm².

A TMCS module (Liquicell membrane contactor, EXF 8 × 2.5" model, 3M, Germany) with a membrane surface area of 1.4 m² was used for the recovery step, where the concentrate was recirculated on the lumen side and an acidic solution on the shell side of the membrane.

The H₂ was carried from the cathode to anode by a 15 mL min⁻¹ flow of nitrogen gas. An external source (support cell) supplied 10% extra hydrogen gas to this recycling stream to compensate for any leakages in the main setup, as described in Kuntke et al., in 2018.¹

During operation, a Memograph M RSG40 data logger (Endress+Hauser BV) recorded every minute the applied current and cell voltage of BP-ED and the support cell, the pH and temperature in the feed, the concentrate, the catholyte and acid compartment, and the conductivity in the feed compartment. Each compartment (feed, concentrate, cathode, and acid) had its own pH sensor (Orbisint CPS11D) connected to a Liquiline CM444 transmitter (Endress+Hauser BV, Naarden, The Netherlands). A conductivity sensor (QC205X EC electrodes and P915-85, Controller (Qis-Prosence BV, Oosterhout, The Netherlands)) was placed in the feed

compartment. The applied current was controlled by a power supply (ES 030-5, Delta Elektronika BV, Zierikzee, The Netherlands).

Operational Conditions. Synthetic wastewater was used as an influent, to study a solution with a defined and constant composition and better quantify the transport over the membrane and avoid undesired phenomena such as scaling. The synthetic wastewater had the following composition: 12.8 mM KCl, 10.9 mM NaCl, 0.26 mM K₂SO₄, 18.7 mM NH₄Cl, and 46.2 mM (NH₄)₂CO₃. This composition was based on ion concentrations measured from the real centrate collected from Rioolwaterzuiverings installatie (a WWTP) in the city of Heerenveen, Netherlands (see Appendix A, Table S1).

All liquids were replaced at the beginning of each experiment. Initially, the concentrate was filled with synthetic influent solution and the cathode was filled with 0.1 M NaOH to increase the conductivity. The OH⁻ originated from the salt dissociation was negligible compared to the water dissociation at the BPMs and at the cathode electrode. A 3M H₂SO₄ solution was dosed continuously to the acid side of the TMCS to guarantee a concentration gradient between concentrate and acid, while maintaining a final product with pH 3.

All liquid compartments were recirculated at 4 cm s⁻¹ (720 mL min⁻¹) to avoid mass transfer limitations³¹ (see Appendix B, Table S2).

Different current densities per electrode area (25–150 A m⁻²) were tested to quantify the transport rate over the CEMs. The current densities were selected within the membrane operation range recommended by the manufacturer, below the limiting current density. Additionally, different ratios between current and TAN loading (load ratio, LN) described by eq 1 were tested (0.4–1.7).¹³ These load ratios were selected to demonstrate the performance of the system under an insufficient current (0.4), close to an optimum (0.9–1.2), and an excess of current (1.4–1.7) compared to TAN loading (Table 1). The stack was operated with a continuous inflow of

Table 1. Load Ratio and Current Densities Applied to the Stack

current density (A m ⁻²)	load ratio				
	0.4	0.9	1.2	1.4	1.7
25			X		
50	X	X	X	X	X
100		X	X	X	X
150			X		

feed for each parameter tested for a minimum of 5 days, with the exception of 150 A m⁻². For this condition, the system was operated for 3 days due to the impracticality of the large influent volume needed for operation (approx. 74 L/d). All results show the average and standard deviation over 5 days of constant operation and of duplicate samples, unless stated otherwise.

Calculations. The load ratio was previously studied as a crucial parameter to characterize electrochemical systems for ammonium recovery and can be described by the following equation¹³

$$LN = \frac{j A_e}{C_{\text{TAN}} Q F} \times 6 \quad (1)$$

where j is the current density (A m^{-2}), A_e is the projected surface area of the electrode (anode) (m^2), 6 is the factor to account for 6 cell pairs, C_{TAN} is the concentration of TAN in the feed inflow (mol L^{-1}), Q the inflow rate (L min^{-1}), and F is the Faraday constant ($96\,485 \text{ C mol}^{-1}$).

The TAN transport rate per total CEM area in $\text{g}_\text{N} \text{ m}^{-2} \text{ d}^{-1}$ for the ED system was calculated as

$$\text{TAN transport rate} = \frac{(C_{\text{inf,TAN}} Q_{\text{inf}} - C_{\text{eff,TAN}} Q_{\text{eff}})}{6 \times A_m} \quad (2)$$

where $C_{\text{inf,TAN}} Q_{\text{inf}}$ is the influent TAN concentration ($\text{g}_\text{N} \text{ L}^{-1}$), Q_{inf} is the influent flow rate (L d^{-1}), $C_{\text{eff,TAN}} Q_{\text{eff}}$ is the effluent TAN concentration ($\text{g}_\text{N} \text{ L}^{-1}$), Q_{eff} is the effluent flow rate (L d^{-1}), 6 is the number of CEMs used in the stack, and A_m the area of one CEM (100 cm^2). Water transport toward the concentrate was observed and quantified, meaning that the effluent volume was smaller than the influent supplied therefore requiring the introduction of Q_{eff} .

TAN – removal efficiency in % for the BP-ED system is given by

$$\text{TAN removal} = \frac{C_{\text{inf,TAN}} Q_{\text{inf}} - C_{\text{eff,TAN}} Q_{\text{eff}}}{C_{\text{inf,TAN}} Q_{\text{inf}}} \quad (3)$$

TAN recovery efficiency in % for the TMCS module

$$\text{TAN recovery} = \frac{C_{\text{acid,TAN}} Q_{\text{acid}}}{C_{\text{inf,TAN}} Q_{\text{inf}} - C_{\text{eff,TAN}} Q_{\text{eff}}} \quad (4)$$

where $C_{\text{acid,TAN}}$ is the TAN concentration in the final product ($\text{g}_\text{N} \text{ L}^{-1}$), and Q_{acid} is the volume of final product formed per day (L d^{-1}).

Energy consumption in $\text{kJ g}_\text{N}^{-1}$ for the BP-ED system

$$\text{Energy consumption} = \frac{(E_{\text{cell}} I_{\text{cell}} + E_{\text{sup,cell}} I_{\text{sup,cell}}) t}{\text{TAN transport rate} \times 6 \times A_m} \quad (5)$$

where E_{cell} is the cell voltage (V), I_{cell} is the applied current (A), $E_{\text{sup,cell}}$ is the cell voltage of the support cell supplying the 10% additional hydrogen (V), $I_{\text{sup,cell}}$ is the applied current of the support cell supplying the additional hydrogen (A), and t is the number of seconds per day (86400 s d^{-1}). The TMCS energy consumption was not accounted as no caustic addition or temperature increase was used.

Analysis and Measurements. IC cations analysis (Na^+ , K^+ , NH_4^+) and IC anions (SO_4^{2-} , Cl^- , NO_3^- , NO_2^-) were done daily for the influent, feed, concentrate, cathode, and acid streams, using a Metrohm Compact IC Flex 930 instrument with a cation column (Metrosep C 4-150/4.0) and a Metrohm Compact IC 761 instrument with an anion column (Metrosep A Supp 5-150/4.0), respectively, each equipped with a conductivity detector (Metrohm Nederland BV, Schiedam, The Netherlands). Inorganic carbon was measured using a TOC-L CPH, (Shimadzu BENELUX, 's-Hertogenbosch, The Netherlands) for all samples. Last, pH and conductivity of all samples were also measured daily with a handheld meter (Seven Excellence S470, Mettler Toledo, Tiel, The Netherlands).

RESULTS AND DISCUSSIONS

The performance of the minimal BP-ED stack will be described when operated at different current densities and load ratios

regarding TAN-removal efficiency, TAN transport rate per total CEM area, energy consumption, and TAN-recovery efficiency. All results show the average and standard deviation over 5 days of constant operation and, of duplicate samples, unless stated otherwise.

High TAN-Removal Efficiency from Synthetic Wastewater Using the Minimal BP-ED Can Be Achieved for a Load Ratio above 1. Figure 2 shows the effect of the load ratio (a ratio between current applied and the TAN loading) on the TAN-removal efficiency, at current densities 50 and 100 A m^{-2} .

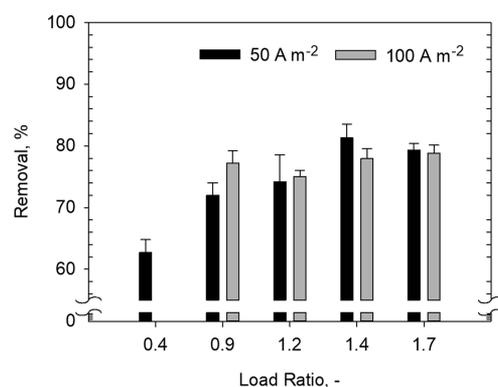


Figure 2. TAN-removal efficiency obtained for different load ratios at 50–100 A m^{-2} . The TAN-removal efficiency increases with a load ratio. Approximately, an 80% removal is achieved for a load ratio higher than 1, at both current densities.

For a load ratio of 0.4 at 50 A m^{-2} , 63% of the TAN was removed from the influent (feed compartment) (Figure 2). For a load ratio above 0.9, TAN-removal efficiencies of >70% were obtained, independent of the current density. When compared to previously described studies (Table 2), this BP-ED stack achieved a comparable or higher TAN-removal efficiency.^{10,16,32}

Based on previous work of Rodríguez-Arredondo et al., in 2017, a lower TAN removal of around 40% was expected at a load ratio of 0.4.¹³ In addition, the ion migration caused by the applied current diffusion can play a significant role, as it has been demonstrated in other electrochemical ammonium treatment systems.²⁶ A possible explanation for the additional removal observed here is ammonium diffusion from the feed toward the concentrate in exchange for other ions that have accumulated in the concentrate solution, an exchange previously described as Donnan dialysis (see Appendix D, Figure S1).^{33,34} After ammonium is removed from the feed, it is converted to ammonia (NH_3) in the concentrate compartment. NH_3 is transported through the TMCS module where a sulfuric acid solution is constantly recirculated, converting NH_3 back to ammonium and producing an ammonium sulfate solution. Ideally, all the ammonium transported over the CEM should be recovered in the acid, meaning that the recovery should match the removal efficiency. However, TAN recovery was slightly lower than the TAN removal for lower current densities. The difference between TAN recovery and TAN removal can be due to losses in the system such as ammonia losses (volatilization) from the compartments.²⁶ It was observed that the difference between TAN-removal efficiency and TAN recovery can be avoided by operating at higher current densities as a high concentrate pH was achieved (pH

Table 2. TAN-Removal Efficiency and Other Key Operation Parameters of ED-like Systems Reported in Literature^{43–45}

	LN	<i>j</i>	Influent/ amount N	Mode ^(b)	Cell pairs	<i>A_m</i>	TAN transport rate	TAN Removal	Energy input	H ₂
		<i>A m⁻²</i>	<i>g L⁻¹</i>			<i>cm²</i>	<i>g_N m⁻² d⁻¹</i>	%	<i>kJ g_N⁻¹</i>	recycling
Pronk et al., 2006 ²⁵	-	-	Urine ≈ 6.3	B	3 ^(c)	49	-	100	-	-
Desloover et al., 2012 ¹⁴	1.6	20	Digestate ^(a) ≈ 5	C	1	64	90	58	16.7	✓
Luther et al., 2015 ³²	-	50	Urine ^(a) ≈ 5	-	1	64	384	86.7	12.4	✓
Kuntke et al., 2017 ²⁹	1.3	50	Urine ≈ 5.1	C	1	100	342.1	73	15.6	✓
Kuntke et al., 2018 ¹	1.3	100	Urine ^(a) ≈ 5.1	C	1	400	598	58	23.4	✓
Tarpeh et al., 2018 ¹⁶	-	100	Urine ≈ 4	C	1	64	-	60.6	30.6	-
Ward et al., 2018 ⁴³	-	20	Centrate ≈ 0.7	C	30 ^(c)	3.6x10 ⁴	129.6	78	18	-
Yan et al., 2018 ⁴⁴	-	2.9	Wastewater ^(a) ≈ 0.2	C	2 ^(c)	189	-	65	4.5 ^(d)	-
Yan et al., 2020 ⁴⁵	-	200	Wastewater ^(a) ≈ 5	B	4 ^(c)	189	-	99	7.9 ^(d)	-
van Linden et al., 2020 ²⁶	-	-	Diluate ^(a) NH ₄ HCO ₃ ≈ 1.5	B	10 ^(c)	640	-	85	18	-
This study	1.2	25	Wastewater ^(a) ≈ 2	C	6 ^(c)	600	202.1	81.3	12.2	✓
This study	1.2	50	Wastewater ^(a) ≈ 2	C	6 ^(c)	600	363.5	74.2	17	✓
This study	1.2	100	Wastewater ^(a) ≈ 2	C	6 ^(c)	600	819.1	78	18.3	✓

^aSynthetic influent. ^bOperation mode: B, batch; C, continuous. ^cSystem includes BPMs. ^dRecalculated to kJ g_N^{-1}

>9.5 was achieved when operating at 100 and 150 A m^{-2}). The pH of the concentrate compartment increased with an increasing current density since cations (other than ammonium and protons) accumulate in the concentrate compartment and more OH^- ions are produced by the BPMs and the cathode.³⁵ The high pH in the concentrate favors the conversion of ammonium to ammonia in the concentrate. Guštin et al., in 2011, have shown that a minimum pH of 10 must be achieved to avoid a limitation on the ammonia-recovery efficiency and consequently improve the amount of stripped ammonia-nitrogen and the TMCS performance.³⁶

At a Load Ratio of 1.2, the BP-ED Stack Maintains a High TAN Transport Rate While Consuming the Least Energy. Electrodialysis is an electricity-driven system, which means that energy consumption is an important parameter that affects the system economic feasibility. The energy consumption of an ED stack is determined by the system internal resistance and efficiency of ion transport across the membrane. Figure 3 presents the energy consumption for the complete stack and the TAN transport rate per total membrane area at 50 A m^{-2} at different load ratios.

An increase in energy consumption was observed between load ratios of 0.4 and 0.9 as the TAN transport rate decreased (Figure 3). At the load ratio of 0.9, the energy consumption was $18.0 \pm 1.1 \text{ kJ g}_N^{-1}$, slightly decreasing to $17.0 \pm 0.4 \text{ kJ g}_N^{-1}$ when operating at a load ratio of 1.2 ($p = 0.04$, t -test). Additionally, the TAN transport rate slightly increased for a load ratio of 1.2 compared to 0.9. The same trend in the TAN transport rate as a function of load ratio was previously observed by Kuntke et al., in 2018, for a hydrogen recycling electrochemical system using only a single-cell pair.¹ In our study, it was observed that for a load ratio of 1.2, the system consumed the least energy (Figure 3) while reaching a high TAN-removal efficiency of 74.2% (Figure 2).

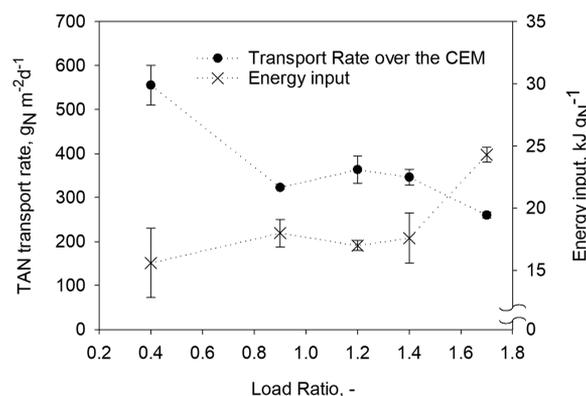


Figure 3. Energy input and TAN transport rate for different load ratios at 50 A m^{-2} . A dashed line was added between points to guide the reader. The minimum energy input was observed for a load ratio of 1.2, for a similar TAN transport rate obtained at LN 0.9 and 1.4 and without compromising the TAN removal previously described.

Again, the energy input increased from a load ratio of 1.4 to 1.7 as the TAN transport rate decreased. Rodríguez-Arredondo et al., in 2017, previously showed that TAN could be removed from urine at LN 1.2 using an electrochemical system.¹³ Rodríguez-Arredondo et al., in 2017, demonstrated that operation at a load ratio higher than 1.2 increases significantly the energy consumption of the system without the further benefit on the TAN-removal and TAN-transport rates.¹³ In our study, we also found that for a load ratio of 1.7, the energy consumption of the system increased without the extra benefit on the TAN-removal rate and TAN-removal efficiency, and for this reason, no higher load ratios were tested.^{10,13} When the current is much higher than the TAN loading supplied to an electrochemical system ($\text{LN} \gg 1.3$), the load ratio model predicts that the extra charge supplied is used to transport mostly other cations (i.e., H^+ , Na^+ , K^+) instead of NH_4^+ .¹³

Based on the ion transport numbers (see Appendix D, Figure S1), the charge transported by H^+ increased with the load ratio in the BP-ED system. Hence, the removal efficiency no longer increases, and consequently, the TAN transport rate decreases, while the energy input increases.

The energy input can be also further analyzed using the ion transport numbers through the CEM, (see Appendix D, Figure S1). Ammonium ions are the main-transported charge over the CEM for all load ratios. However, the percentage of current used to transport NH_4^+ decreases with increasing load ratio. With increasing load ratio, the applied current is higher than the available amount of ammonium in the feed that can be carried with the applied current. Therefore, protons will be transported, while the transport of potassium and sodium remains constant. This was previously observed in other electrodialysis studies, including other electrochemical systems for TAN recovery.^{1,33,37} The potential transport of anions over the CEM was measured but not detected.

Operation at High Current Density Increased the TAN Transport Rate, Without Compromising the Removal Efficiency for the Same Load Ratio. Figure 4

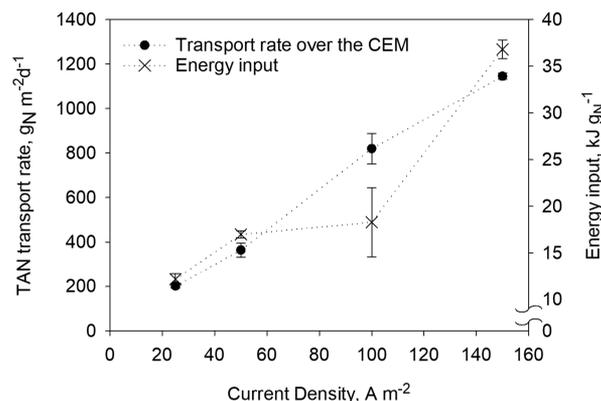


Figure 4. Transport rate per total installed CEM area and energy input at a load ratio of 1.2 for different current densities. A dashed line was added between points to guide the reader. The TAN transport rate increased with current density. Also, the energy consumption gradually increases with current density, except at 100 A m⁻².

shows the TAN transport rate per total CEM area and energy consumption at different current densities for a load ratio of 1.2 as the previous results indicated a more successful operation at this condition.

The TAN-removal rate increased almost proportionally with current density (Figure 4). The TAN transport rate was 202.1 g_N m⁻² d⁻¹ at 25 A m⁻² and increased to 1145.1 g_N m⁻² d⁻¹ at 150 A m⁻². For the different current densities tested, the TAN-removal efficiency was between 74.2 and 81.3% and did not significantly change with the applied current density ($p = 0.93$, t -test), (Table S3, Appendix C). Although the TAN-removal efficiency was almost constant, the energy cost to achieve this result increased with increasing current density from 12.2 to 38.8 kJ g_N⁻¹.

The ammonium transport number was constant and independent of the current density, which also reflects in the TAN-removal efficiency, (see Figure S2, Appendix D). Ammonium accounted for around 60% of the charge transported over the CEM. The internal resistance consisted of the anode and cathode overpotentials, ionic and pH losses, and membrane potential.^{24,38} Our hypothesis is that the

increase in current density amplifies the membrane potential loss. As earlier reported, the membrane potential increases as the transport of ions occurred against a higher concentration gradient between the feed and concentrate (due to the accumulation of Na⁺ and K⁺ in the concentrate).^{37,39,40} At 25 A m⁻², the concentrations of Na⁺ and K⁺ in the concentrate solution reached 87 and 107 mM, respectively. At 100 A m⁻², the concentrations of Na⁺ and K⁺ in the concentrate solution were 314 and 440 mM, respectively. Additionally, a water flux from the feed to the concentrate was observed for all the tested conditions. Water transport in ED was previously described by Tedesco et al., in 2017, as a result of (1) hydrostatic pressure difference across the membrane, (2) osmotic pressure difference across the membrane, or (3) ion-water friction (electro-osmosis).^{22,41} When operating the BP-ED stack at 150 A m⁻², the water transport was 12% higher than when operating at 50 A m⁻².

The BP-ED achieved 80% TAN removal while consuming approx. 18 kJ g_N⁻¹ at 50 and 100 A m⁻². The anammox process alone consumes 16 kJ g_N⁻¹, and when combined with Haber-Bosch, it consumes 53 kJ g_N⁻¹.^{10,42} Therefore, the system developed during this work represents a much more energy-efficient alternative technology to remove and recover nitrogen. Compared to similar electrochemical systems for TAN recovery, the BP-ED stack consumed either lower or comparable energy (see Table 2).^{10,16,26} Interestingly, in this minimal BP-ED stack, we observed an increase of the TAN transport rate with current density without compromising removal efficiency, while previous studies observed that a high TAN transport rate is only achieved in combination with lower TAN-removal efficiency.^{1,29} This result is supported by the constant NH_4^+ transport number over the CEM (60%) and a positive consequence of the proposed BP-ED design improvement over other ED systems for TAN recovery.

Minimal Bipolar Electrodialysis Stack Including a CEM and BPM per Cell Pair Is a Feasible Option for TAN Recovery. Table 2 includes some of the most recent developments on ED for TAN recovery, which achieved high TAN removal at low energy consumption.

The minimal BP-ED cell configuration stack achieved a much higher TAN transport rate per membrane area compared to Kuntke et al., in 2018, namely, 819.1 g_N m⁻² d⁻¹ at 100 A m⁻² using synthetic wastewater, at a lower energy input.¹ The minimal BP-ED cell configuration stack consumed slightly more energy per gram of nitrogen removed than Luther et al., in 2015 and Kuntke et al., in 2017, while transporting a higher amount of nitrogen per membrane area.^{29,32} The average TAN-removal efficiency was 80%, which is comparable to the aforementioned studies. However, it is worth mentioning that these studies achieved similar energy input and TAN-removal efficiency using a system with only 1 CEM.

Compared to previous BP-ED systems, the energy input and TAN removal were comparable.^{25,26} The use of a minimalistic approach for BP-ED using only 2 membranes per cell pair instead of three membranes is beneficial, as less membrane area and fewer compartments are used. As an AEM is more prone to biological fouling of negatively charge compounds, a longer performance is expected for the tested BP-ED configuration in our study.⁴⁶ However, our minimal BP-ED configuration does not remove the anions presents in the wastewater.

During BP-ED operation, the pH increased in the concentrate compartment, allowing our minimal BP-ED

combined with membrane stripping (transmembrane chemisorption, TMCS) to recover nitrogen by only requiring an acid solution and applied current. Unlike conventional ammonia (membrane) stripping processes for TAN recovery, no caustic addition and temperature increase were required BP-ED.¹⁶ Additionally, although BP-ED still requires an acid to capture the ammonia, from the end-user point of view, the fertilizer formed is pure, and consequently, the product can be introduced in the market.^{22,25}

Overall, the minimal BP-ED operated in continuous feed flow at a higher current density, treating a larger volume of (synthetic) wastewater. The minimal BP-ED cell configuration stack treated up to 73 L d⁻¹ and removed around 100 g of nitrogen per day at a load ratio of 1.2 and a current density of 150 A m⁻². As previously described, the energy consumption per gram of nitrogen was similar to a conventional 1 cell pair ED system while improving the amount of nitrogen transported per membrane area. The use of BPMs guarantee a comparable energy input and together with hydrogen recycling, no harmful compounds are produced in this system, such as chlorine gas and halogenated organic compounds. As this BP-ED system operated at both high TAN transport rate and removal efficiency with low energy consumption, it is a promising solution toward upscaling of ammonia recovery from wastewater. Additionally, this more compact system with high treatment capacity can become an interesting alternative to technologies such as conventional stripping or forward osmosis.^{47–51} The influence of bivalent ions found in centrate was not included in this study. Several studies previously demonstrated the scaling effects on a CEM when calcium and magnesium are present in the influent, even at low concentrations.^{52–54} Therefore, future research should focus on real wastewater to investigate the effects of scaling and (bio)fouling in this BP-ED stack for ammonia recovery and further scaling up with more cell pairs. Thompson-Brewster et al., have shown that a suitable pretreatment (i.e., precipitation) can reduce the amount of scaling in anion and cation exchange membranes.⁵³ Tran et al., described that scaling of the bipolar membrane occurs at the anion exchange layer and can be reduced by a precipitation pretreatment step.⁵⁵ Wastewaters with a high TAN concentration are the main focus for this minimal BP-ED configuration, and further research might be needed when treating wastewater with a lower TAN concentration (less than 0.5 g L⁻¹).

CONCLUSIONS

In this work, we investigated a new cell configuration design for TAN recovery. A minimal BP-ED stack was built with cell pairs made of CEM and BPM instead of cell triplets with AEM, CEM, and BPM. The minimal cell configuration was demonstrated to be energy efficient in TAN removal and recovery from synthetic wastewater. The minimal BP-ED cell configuration was stacked with 6 cell pairs and removed up to 80% of the supplied TAN during continuous operation. At a load ratio of 1.2 and a current density of 100 A m⁻², the BP-ED stack treated 49 L d⁻¹ at a TAN transport rate of 819 g_N m⁻² d⁻¹ while consuming 18.3 kJ g_N⁻¹. The TAN transport rate and treatment capacity were improved by increasing the current density without compromising the TAN-removal efficiency. We demonstrated that this minimal BP-ED stack is a promising option for TAN recovery.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acssuschemeng.0c05043>.

Centrate composition, recirculation speed effect on the performance of the system, experiments performed in this study, and ion transport number (PDF)

AUTHOR INFORMATION

Corresponding Author

Philipp Kuntke – Wetsus, European Centre of Excellence for Sustainable Water Technology, Leeuwarden 8900CC, The Netherlands; Environmental Technology, Wageningen University, Wageningen 6700 AA, The Netherlands; orcid.org/0000-0002-2342-8662; Email: philipp.kuntke@wur.nl

Authors

Mariana Rodrigues – Wetsus, European Centre of Excellence for Sustainable Water Technology, Leeuwarden 8900CC, The Netherlands; Environmental Technology, Wageningen University, Wageningen 6700 AA, The Netherlands; orcid.org/0000-0003-0534-4817

Thiago T. de Mattos – Wetsus, European Centre of Excellence for Sustainable Water Technology, Leeuwarden 8900CC, The Netherlands

Tom Sleutels – Wetsus, European Centre of Excellence for Sustainable Water Technology, Leeuwarden 8900CC, The Netherlands; orcid.org/0000-0001-8251-7879

Annemiek ter Heijne – Wetsus, European Centre of Excellence for Sustainable Water Technology, Leeuwarden 8900CC, The Netherlands; Environmental Technology, Wageningen University, Wageningen 6700 AA, The Netherlands; orcid.org/0000-0002-6882-8395

Hubertus V.M. Hamelers – Wetsus, European Centre of Excellence for Sustainable Water Technology, Leeuwarden 8900CC, The Netherlands; orcid.org/0000-0002-0990-4773

Cees J.N. Buisman – Wetsus, European Centre of Excellence for Sustainable Water Technology, Leeuwarden 8900CC, The Netherlands; Environmental Technology, Wageningen University, Wageningen 6700 AA, The Netherlands

Complete contact information is available at: <https://pubs.acs.org/doi/10.1021/acssuschemeng.0c05043>

Notes

The authors declare no competing financial interest.

ACKNOWLEDGMENTS

This work was performed in the cooperation framework of Wetsus, European Centre of Excellence for Sustainable Water Technology (www.wetsus.eu). Wetsus is co-funded by the Dutch Ministry of Economic Affairs and Ministry of Infrastructure and Environment, the European Union Regional Development Fund, the Province of Fryslân, and the Northern Netherlands Provinces. This work was also supported by the LIFE-NEWBIES project. The LIFE-NEWBIES project (LIFE17 ENV/NL/000408) has received funding from the LIFE Programme of the European Union. The authors like to thank the participants of the research theme “Resource Recovery” for the fruitful discussions and their financial support.

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