Chemical Engineering Journal 395 (2020) 125143

Contents lists available at ScienceDirect

Chemical Engineering Journal

journal homepage: www.elsevier.com/locate/cej

Exploiting Donnan Dialysis to enhance ammonia recovery in an electrochemical system

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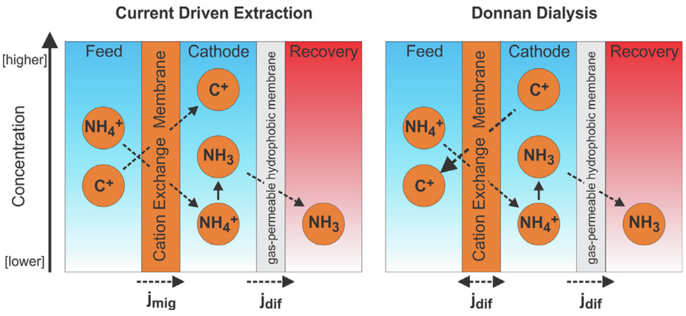
HIGHLIGHTS

- Hydrogen recycling electrochemical system was used for ammonium removal.
- Exploiting Donnan Dialysis for ammonia recovery via cation exchange with ammonium.
- In continuous operation, Donnan Dialysis did not clearly affect ammonium removal.
- In batch operation, Donnan Dialysis increased the ammonium removal by 10%.
- The system was more energy efficient compared to previous studies.

GRAPHICAL ABSTRACT



ARTICLE INFO

Keywords:
Donnan dialysis
Ammonia recovery
Electrochemical system

ABSTRACT

A hydrogen recycling electrochemical system (HRES) can be used for energy efficient removal of TAN (Total ammonia nitrogen, ammonium and ammonia) from wastewater. When a current is applied, a concentration gradient of cations builds up between catholyte and feed solution. When no current is applied, cations (Na^+ and K^+) diffuse back to the feed solution from the catholyte as a result of the concentration difference. These cations will be exchanged for other cations (NH_4^+ and H^+) to maintain electroneutrality: a phenomenon known as Donnan Dialysis. In this study, Donnan Dialysis was explored as a strategy to enhance the TAN removal efficiency in an HRES. In continuous operation, Donnan Dialysis did not clearly affect TAN removal efficiency. In batch operation, Donnan Dialysis resulted in (10 ± 2) % higher removal efficiency compared to operation without Donnan Dialysis. By analyzing transport numbers of the different cations, we show that in batch mode, Donnan Dialysis indeed exchanges mostly NH_4^+ with Na^+ and K^+. In continuous mode, however, more protons were transported from anode to cathode. Batch operation with Donnan Dialysis achieved similar removal to continuous operation but consumed less energy (between 7.8 kJ g_N^{-1} and 10.1 kJ g_N^{-1}) than continuous operation. Donnan Dialysis can be a good strategy to enhance TAN recovery in batch operation mode since additional ammonium was removed at a lower energy input.

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https://doi.org/10.1016/j.cej.2020.125143
Received 19 September 2019; Received in revised form 17 March 2020; Accepted 18 April 2020

1. Introduction

Recovery and re-use of ammonia from aqueous waste streams is gaining more and more attention, both for environmental and economic reasons [1]. The conversion of N_2 to synthetic fertilizer through the energy-intensive Haber–Bosch process is responsible for 1% of the primary global energy consumption. Just in Europe, the removal of the nitrogen compounds from water streams costs up to 320 billion euros per year [2]. Besides these conventional processes, several ammonia/ammonium recovery methods exists and have been studied at different scales, including NH_3-stripping, struvite precipitation, and use of zeolites. However, they do require high energy and/or chemicals input [3].

Electrochemical recovery of total ammonia nitrogen (TAN; i.e. $NH_4{}^+$ and NH_3) from concentrated streams like urine is an interesting option, considering that TAN can be recovered using less energy compared to other recovery technologies [4]. The TAN recovered from the electrochemical process is directly suitable for re-use because it is in its reactive form [5–7].

Electrochemical TAN removal relies on an electrical current to drive the positively charged ammonium ion through a cation selective membrane [8,9]. Water can be oxidized at the anode to produce oxygen and reduced at the cathode to produce hydrogen gas. Electrodialysis systems have been shown to recover ammonia from wastewater at high energy input [4,8,10,11]. Recently, it was shown that the energy input for TAN removal can be reduced when the produced hydrogen gas at the cathode is recycled to the anode to be oxidized again [12]. This hydrogen recycling electrochemical system (HRES) achieved a TAN removal efficiency of 73% while consuming 26 kJ $g_N{}^{-1}$, when operating at 20 A m^{-2}. These results were a considerable improvement compared to a previous electrochemical system without H_2 recycling treating digestate achieving 58% TAN removal and consuming 60.1 kJ $g_N{}^{-1}$, when operated at the same current density [8]. Rodriguez Arredondo et al., 2017 achieved a TAN removal efficiency of 63% in an electrochemical system using synthetic urine as influent. This system was operated at 50 A m^{-2} and used 49 kJ $g_N{}^{-1}$ [10].

A successful HRES design provides high TAN removal at low energy input and is dependent on several parameters, the main ones being: the ratio between applied current density and the TAN loading (L_N, load ratio), the catholyte pH and the internal resistance of the system [7].

One major challenge in achieving a high TAN removal efficiency is reducing the co-transport of cations other than ammonium (e.g., Na^+, K^+, Ca^{2+}, Mg^{2+}) towards the cathode (as a result of the applied current, see Fig. 1). Most of these cations are present in urine at high concentrations [13,14]. The flux of ions through a cation exchange membrane (CEM) can be described by two driving forces, an electric field and a concentration gradient in the membrane. The Nernst-Planck equation describes these two forces:

$$J_i = D_i\left(\frac{\partial c_i}{\partial x} + \frac{z_i F}{RT} c_i \frac{\partial \varphi}{\partial x}\right) \tag{1}$$

where J_i is the flux of the ion (mol m^{-2} s^{-1}), D_i is the diffusion coefficient of the ion (m^2 s^{-1}), c_i is the concentration of ion (mol m^{-3}), x is the position inside the membrane (m), z_i is the valence of the ion, F is the Faraday constant (C mol^{-1}), R is the universal gas constant (J K^{-1} mol^{-1}), T is the temperature (K), and φ is the electric field potential (V). In general, we assume for simplification that: (i) CEM behaves as an ideal membrane and no counter ions (anions) are being transported, (ii) electron neutrality is maintained, (iii) the diffusion coefficients of the main cations are almost identical, and (iv) hydroxide ions concentrations are negligible (pH range 4–10). Therefore, when operating an HRES, the concentration of cations in the catholyte will initially increase, until a stationary equilibrium is reached. At equilibrium conditions, only ammonium and protons are effectively transported through the CEM [15]. The accumulation of cations other than ammonium in the catholyte leads to an additional energy loss, due to the formation of a concentration gradient induced membrane potential [16]. Therefore, to maintain the same TAN removal more energy is required to force the ammonium transport against the concentration gradient, increasing the cost of operation.

In the absence of an applied electrical potential, the cations accumulated in the cathode compartment will diffuse back towards the anode (feed) compartment as a result of the concentration difference. For reasons of electro neutrality, this can only happen when either anions also diffuse from cathode to anode, or when other cations diffuse from anode to cathode. Based on the above mentioned assumption the flux of ammonium $J_{NH_4^+}$can be described as following:

$$J_{NH_4^+} = -\sum_{CEM} z_i J_i \tag{2}$$

This exchange of ions of the same charge through a selective membrane is known as Donnan Dialysis. As an example, Donnan Dialysis has been explored in desalination processes Dialysis via DC Electrodialysis [17], in the removal of metal ions such as Cu^{2+} from refinery wastewater or to remove divalent cations present in the

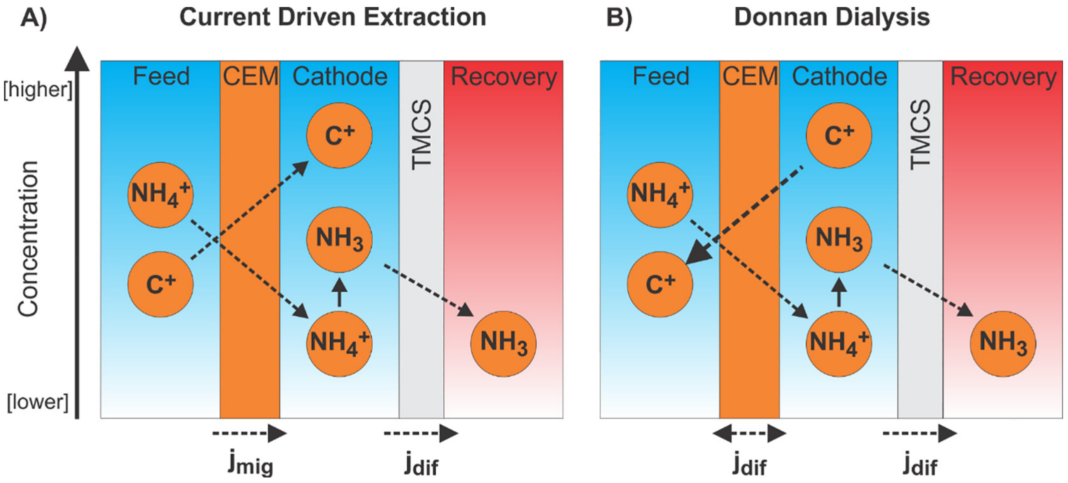

Fig. 1. Donnan Dialysis principle for the removal of ammonium. (A) Under the influence of the applied current (j_{mig}), ammonium and other cations (i.e. K^+ and Na^+; here represented as C^+) are transported over the cation exchange membrane (CEM) to the cathode. Since ammonium is continuously removed from the cathode solution, only a concentration gradient of C^+ builds up across the CEM. (B) When the current is switched off (j_{dif}), this concentration gradient can be used to extract the remaining ammonium from the feed compartment. C^+ diffuse to the feed and because of electroneutrality, ammonium is forced to diffuse towards the cathode compartment.

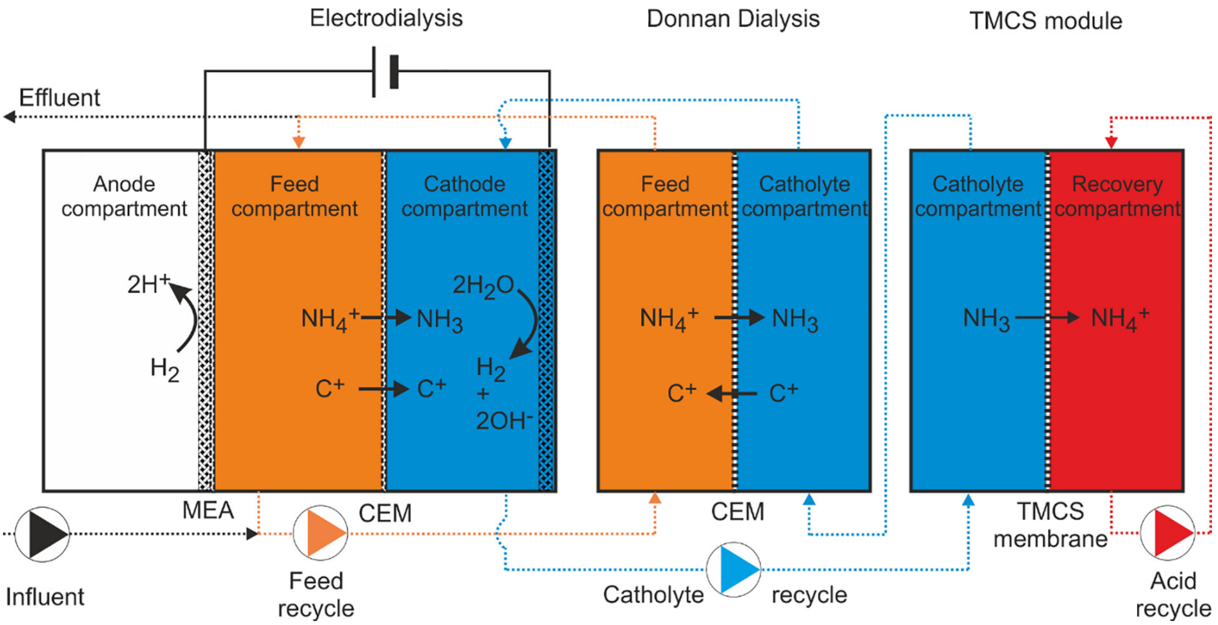

Fig. 2. Continuous setup including the HRES, the Donnan Dialysis cell and the TMCS module. Ammonium was continuously extracted from the feed stream in the HRES via electrodialysis, due to the applied current. Additionally, ammonium was removed in the Donnan Dialysis unit through exchange with C^+. In continuous operation the Donnan Dialysis unit was placed after the TMCS module. First ammonium was extracted from the feed stream through the current and recovered from the catholyte compartment via the TMCS module. Afterwards, remaining ammonium in the feed was extracted through Donnan Dialysis.

influent of reverse electrodialysis [18,19]. Donnan Dialysis has been previously observed for current driven ammonium transport in Microbial Fuel Cells (MFCs) [20]. It was shown that the diffusion (j_{dif}) of potassium from cathode to anode increased the expected transport of ammonium from anode to cathode. Donnan Dialysis has not yet been investigated as a strategy to enhance ammonium recovery [20].

The combination of HRES and Donnan Dialysis can increase the recovery efficiency for ammonium by (i) improving the TAN removal efficiency, as the diffusion of cations from cathode to anode can be used to transport additional TAN, (ii) reducing energy consumption, since the Donnan Dialysis operates without electrical energy input.

In this work, we study how Donnan Dialysis can be exploited to increase the removal efficiency of TAN from synthetic urine using an HRES. We show how TAN transport is influenced by Donnan Dialysis in batch and continuous operation of the system.

2. Materials and methods

2.1. System design

The HRES cell, or electrodialysis cell, consisted of two PMMA (poly methyl methacrylate) panels (21 cm × 21 cm) as frame, with three compartments in between: (i) anode, (ii) feed, and (iii) cathode. The inner dimensions of the anode and the cathode compartments were 10 cm × 10 cm × 0.2 cm. Both compartments were separated from the feed compartment by a cation exchange membrane (CEM; 15 cm × 15 cm Nafion 117, Ion Power GmbH, Germany) with a projected surface of 100 cm^2. The CEM between feed and anode was coated on one side with a 10 cm × 10 cm of Platinum - Vulcan (carbon) catalyst (0.5 mg Pt cm^{-2}), which was connected through a current collector (Pt coated titanium mesh electrode (9.8 cm × 9.8 cm, 5 mg Pt cm^{-2} Magneto Special Anodes BV, The Netherlands)), with the electrical circuit. This combination of the CEM and Pt/C catalyst formed a gas diffusion electrode. At this anode, hydrogen was oxidized to electrons and protons. A Pt coated titanium mesh electrode (9.8 cm × 9.8 cm, 5 mg Pt cm^{-2} Magneto Special Anodes BV, The Netherlands) was used as a cathode for water reduction to hydrogen gas.

The total volume of the feed and catholyte loops were 1L and 0.5L, respectively. A peristaltic pump was used to circulate the feed and catholyte at 80 mL min^{-1}. The catholyte was recirculated over a gas-permeable hydrophobic membrane (TransMembrane ChemiSorption, TMCS) module where the ammonia was extracted from the catholyte into a 1 M sulphuric acid solution. The TMCS consisted of a custom-made polypropylene (PP) housing and commercially available membranes (1.5 m, 0.2 µm pore size, V8/v Type TP, MICRODYN-NADIR GmbH, Wiesbaden, Germany). The initial solution was 2L sulphuric acid (1 M) and it was replaced regularly before saturation occurred, to avoid limitations in the extraction of ammonium from the catholyte solution.

For continuous operation, a separate cell was used for the Donnan Dialysis process. This Donnan Dialysis cell consisted of two PMMA (poly methyl methacrylate) panels (21 cm × 21 cm) as frame, including two compartments for: (i) feed and (ii) catholyte. The feed and cathode compartment were separated by a cation exchange membrane (CEM; 15 cm × 15 cm Nafion 117, Ion Power GmbH, Germany) with a projected surface of 100 cm^2 and dimensions of 10 cm × 10 cm × 0.2 cm.

2.2. Experimental strategy

The experiments were performed in continuous and batch mode of operation. The electro dialysis cell was always operated at constant applied current density, 20 A m^{-2}. The TAN loading to feed compartment was previously optimized in relation to the applied current resulting using the load ratio model [7,10]. For a L_N lower than 1, the applied current is lower than the TAN loaded to the system, meaning that TAN removal over the CEM is limited by the available current, and a limited removal efficiency. A L_N equal to 1 means the current is equal to the TAN loading, meaning that in theory, 100% of TAN can be removed. However, it was previously determined both experimentally and theoretically that a LN of approximately 1.3 is ideal to achieve high removal efficiency [10]. For higher LN values, the TAN removal efficiency only slightly increases, as the energy input increases drastically. In the experiments presented, three different load ratios were tested for both modes of operation to characterize the system under ideal and close to the optimum conditions, an excess (L_N = 0.8), a sufficient

(L_N = 1) and an insufficient (L_N = 1.3) TAN loading with respect to the applied current density of 20 A m^{-2}.

2.3. Continuous experiments

The continuous Donnan Dialysis experiments were performed in the system illustrated in Fig. 2. Fresh influent was fed continuously to the feed compartment. Both feed and catholyte solutions were replaced with new synthetic urine at the start of each experiment. The system was operated for 3 to 4 days to reach steady-state (constant TAN removal, cathode pH and cell voltage). The average and standard deviation presented here were from the 5 days during which the system was in steady-state. The catholyte was recirculated in the electro dialysis cell, to the Donnan Dialysis and TMCS module as presented in Fig. 2. When operating continuously without the Donnan Dialysis cell, the catholyte was recirculated over the electrodialysis cell and the TMCS module only.

2.4. Batch experiments

The batch experiments consisted of a 24 h period where current was applied (Electrodialysis), followed by a 72 h period where no current was applied (Donnan Dialysis); in the same cell (Fig. 3). The batch experiments were performed with a predefined amount of TAN (volume of synthetic wastewater) in the feed compartment. The desired load ratio was obtained by using a certain volume of synthetic urine and at a current density of 20 A m^{-2} during the first 24 h.

2.5. Feed

The synthetic urine consisted of 13.7 g L^{-1} $(NH_4)_2CO_3$, 4.45 g L^{-1} NaCl, 1.04 g L^{-1} K_2SO_4 and 1.9 g L^{-1} KCl. It had a conductivity of 28.8 mS cm^{-1} and a pH of 9.1. This composition was previously described in Kuntke et al., 2018 and it has similar concentrations of TAN and other cations to urine after a phosphate recovery step. Only monovalent cations were added as it allows to simplify the study of ammonium transport [21,22]. For continuous operation, the synthetic urine was supplied to the feed recycle vessel by a peristaltic pump (Masterflex L/S, Metrohm Applikon BV, Schiedam, The Netherlands) at a rate of 0.39 mL min^{-1}, 0.51 mL min^{-1} and 0.63 mL min^{-1} corresponding to load ratio of 1.3, 1.0 and 0.8 respectively. During batch experiments, the feed compartment contained 0.93 L, 0.74 L, and 0.62 L of synthetic urine corresponding to a load ratio of 0.8, 1.0 and 1.3, respectively. The acid solution in the TMCS consisted of 1 M H_2SO_4 with a total volume of 2 L.

2.6. Measurements

The following parameters were recorded every minute: the current and cell voltage applied to the electrodialysis cell, the pH and conductivity of feed and catholyte, and the potentials of the anode and cathode. The data was stored in a Memograph M RSG40 datalogger (Endress + Hauser BV, Naarden, The Netherlands). Ag/AgCl reference electrodes (+0.2 V vs NHE, QM711X, QiS-Prosence BV, Oosterhout, The Netherlands) were placed in the catholyte and the feed compartment. The pH was measured using an Orbisint CPS112D sensors connected to a Liquiline CM444 transmitter (Endress + Hauser BV). The conductivity of the feed compartment and the catholyte compartment was measured using QC205X EC electrodes and P915-85 – Controller (QiS-Prosence BV, Oosterhout, The Netherlands).

2.7. Chemical analysis

For the continuous experiment, samples were taken daily from influent and effluent stream of the feed compartment and from the catholyte. For the batch experiment, samples were taken every 24 h. All samples were analyzed in duplicate for cations (Na^+, K^+, NH_4^+) and anions (SO_4^{2-}, Cl^-, NO_3^-, NO_2^-) with a Metrohm Compact IC Flex 930 with a cation column (Netrosep C 4–150/4.0) and a Metohm Compact IC 761 with an anion column (Metrosep A Supp 5–150/4.0) each equipped with a conductivity detector (Metrohm Nederland BV, Schiedam, The Netherlands).

2.8. Calculations

All calculations (L_N, removal/recovery, energy use, ion transport number) were based on the methodology reported earlier [12]. L_N was calculated for continuous (a) and for batch operation mode (b).

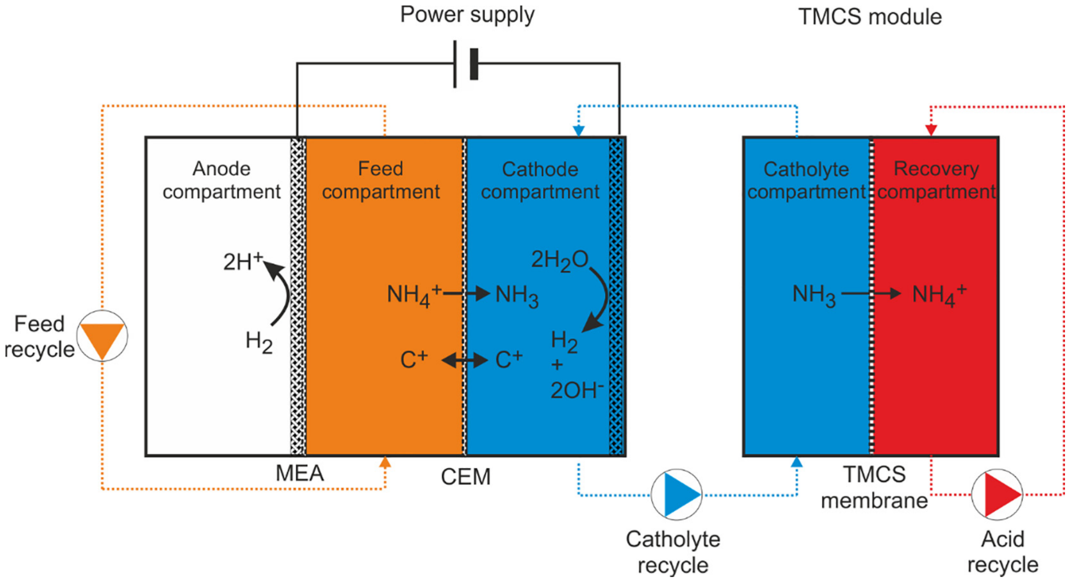

Fig. 3. Batch setup including the HRES and the TMCS module. During the first 24 h, current was applied to the HRES and cation including ammonium were extracted from the feed compartment via electrodialysis. After 24 h, current was no longer supplied and the HRES acted as Donnan Dialysis cell. Cations (Na^+ and K^+) in the cathode compartment diffused from cathode to the feed compartment and an equal amount of ammonium ions from feed to cathode compartment. The TMCS module was used to continuously extract ammonia from the cathode compartment.

$$L_{N,c} = \frac{JA}{[NH_4^+]QF} \tag{3}$$

$$L_{N,b} = \frac{JAt}{[NH_4^+]VF} \tag{4}$$

where J is the current density (A m^{-2}), t is the time (s), A is the area of the cell (m^2), [$NH_4{}^+$] is the TAN concentration (mol m^{-3}), V is the volume of the feed (m^{-3}), Q is the inflow rate (m^3 s^{-1}) and F is the Faraday constant (96485 C mol^{-1}).

The TAN removal efficiency was calculated as the difference between the initial concentration (C_{0TAN}; mol L^{-1}) and the final concentration of ammonium (C_{xTAN}; mol L^{-1}) related to the initial concentration during the time intervals (0, 24, 48, 72, 96 h)

$$Removal_{NH_4} = \frac{C_{0TAN} - C_{xTAN}}{C_{0TAN}} \tag{5}$$

The energy required for TAN recovery was calculated based on the average cell voltage (E, V), the applied current density (J, A m^{-2}), the time interval (t, h), the amount to TAN removed (m_{TAN}, g_N) and the CEM surface area (A, m^2).

$$P_{TAN} = \frac{EJt}{m_{NH4}A} \tag{6}$$

The transport number was calculated based on the difference between the initial ion concentration of the influent and effluent (feed compartment for batch experiments) related to the produced current. The transport number (t_i) was calculated for all ions in solution.

$$t_i = \frac{\Delta C_i z_i FQAt}{Jt} \tag{7a}$$

$$t_i = \frac{\Delta C_i z_i FVA}{Jt} \tag{7b}$$

where ΔC_i is the difference of concentration between two time points for component i (mol L^{-1}), z_i is the net charge of that cation (–).

3. Results and discussion

The effect of Donnan Dialysis on the ammonium removal efficiency was studied in continuous and batch mode at different load ratios. For both operation modes, we compared the HRES performance with and without Donnan Dialysis.

3.1. Donnan Dialysis led to an increase in TAN removal efficiency only for batch operation

Fig. 4 shows the removal efficiency for TAN in the continuous system with and without Donnan Dialysis at different load ratios. The TAN removal efficiency without Donnan Dialysis was 71% for a load ratio of 0.8, 80% for a load ratio of 1, and 85% for a load ratio of 1.3. Unexpectedly, the TAN removal efficiency with Donnan Dialysis was similar: 68%, 83% and 87%. This increase in TAN removal efficiency with an increase in load ratio is in accordance with previous studies [10].

To understand why Donnan Dialysis did not clearly enhance TAN removal efficiency in the continuous system, batch experiments were performed. In these batch experiments, the Donnan Dialysis process can be distinguished more clearly since it does not occur at the same time as current driven TAN transport.

Fig. 5 shows the removal efficiency obtained after the first 24 h (without Donnan Dialysis) and after 72 h, with Donnan Dialysis. The removal efficiency without Donnan Dialysis resulted from current driven migration of cations through the CEM to the cathode. During this period, the removal of TAN was 61% for a load ratio of 0.8, 68% for a load ratio of 1 and 77% for a load ratio of 1.3.

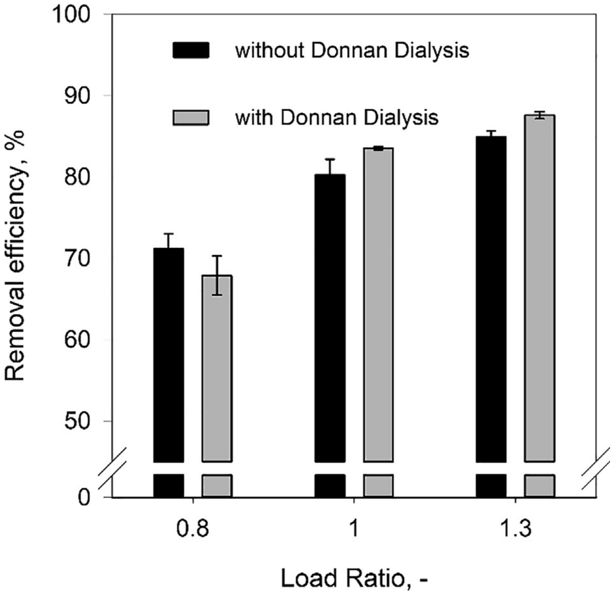

Fig. 4. Average removal efficiency obtained during 5 days of continuous operation. The removal was determined with and without Donnan Dialysis at load ratios of 0.8, 1 and 1.3. The total removal was similar with and without Donnan Dialysis. Moreover, the relation of removal with the load ratio was confirmed.

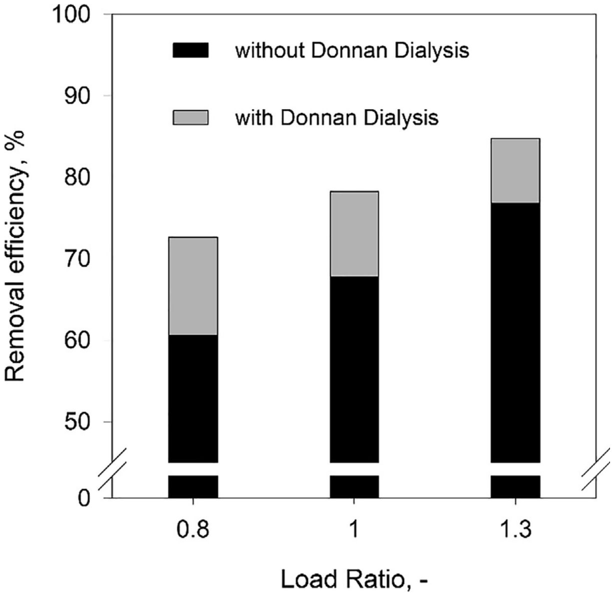

Fig. 5. TAN removal efficiency obtained during batch operation. The TAN removal was determined with and without Donnan Dialysis. Donnan dialysis increased the total removal about 10%. The effect of load ratio was also observed as the removal efficiency increased at the higher Load Ratio.

In the period with Donnan Dialysis, the TAN removal efficiency increased around 10% (Fig. 5) to 73%, 78% and 85%. Overall, the TAN removal efficiencies for the batch experiments with Donnan Dialysis were similar to those in the continuous experiments and values previously reported [10].

3.2. In batch operation, the Donnan Dialysis cell successfully exchanged sodium and potassium from the catholyte with ammonium from the feed.

To have a closer look at the individual influence of Donnan Dialysis and electrodialysis, the transport number of each ionic species and its contribution to the total charge transport through the CEM were calculated for the batch and continuous experiments (Fig. 6). The (ion)

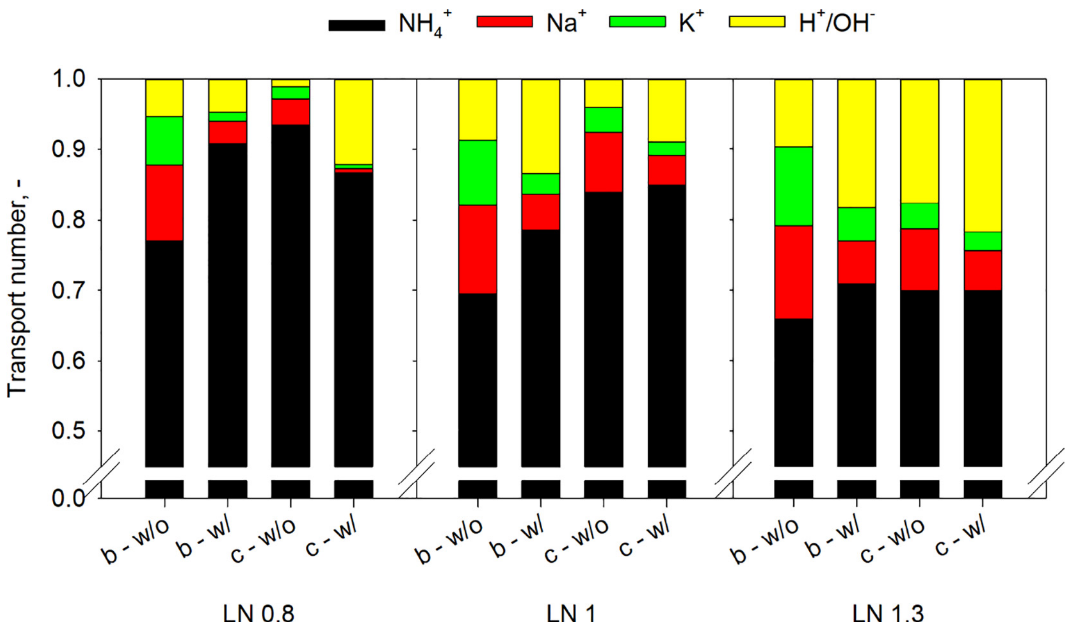

Fig. 6. Ion transport numbers calculated per Load Ratio (0.8, 1 and 1.3) for continuous (c -) and batch experiments (b -). The influence of batch and continuous operation as well as the operation with (w/) and without (w/o) Donnan dialisys on the transport number were examined.

transport number describes the fraction of current used to transport the different ions present in solution over the total charge supplied to the system [7]. The main transported charge was in the form of ammonium, representing over 60% of the charge transport through the CEM. The remainder of the charge was carried by Na^+, K^+ and protons/hydronium ions.

For batch operation mode (b -), all load ratios presented the same trend in the transport number. During batch operation mode, a decrease of the transport number of sodium and potassium (b – w/o to b – w/), shows that most of the sodium and potassium returned to the feed due to Donnan Dialysis. The sodium and potassium ions were successfully exchanged with ammonium ions from the feed, as observed by an increase of the ammonium transport number and consequently an additional 10% TAN removal, in b – w/. Moreover, the transport number of ammonium in batch with Donnan Dialysis and continuous without Donnan Dialysis were similar for each Load Ratio. Hence, the TAN removal efficiency achieved was similar.

When operating continuously with Donnan Dialysis, for Load Ratio 0.8, the transport number of sodium, potassium and ammonium ions were lower compared to the situation without Donnan Dialysis. Hence, the exchange of sodium and potassium ions from the catholyte occurred mostly with protons from the feed. Additionally, the exchange of sodium and potassium with protons resulted in catholyte pH of 9.3 that led to an inefficient TAN recovery. A lower cathode pH decreases the amount of nitrogen present as ammonia (NH_3), which leads to a lower TAN transport over the TMCS. As reported earlier [7], the pH influences the TAN stripping efficiency from the catholyte into the acid. At a pH of 9.24 (pKa for ammonium/ammonia), the TAN in solution is 50% ammonia (NH_3) and 50% ammonium (NH_4^+). In the TMCS module, the ammonia (NH_3) concentration gradient across the gas permeable membrane drives the transport of ammonia. At lower pH, the concentration of ammonia is lower, hence the transport of ammonia to the acid is lower. Whereas, for Load Ratio 1 and 1.3 in continuous operation with Donnan Dialysis (c – w/) the ammonium transport number was equal to operation without Donnan Dialysis. The exchange of Na^+ and K^+ from catholyte also occurred mostly with protons from the feed, but this did not impair the ammonia extraction over the TMCS since the pH of the catholyte was sufficiently high (LN 1: pH 10.2 and LN1.3: pH 10.1). Donnan dialysis is not selective for the exchange of a specific ion. The exchange of Na^+ and K^+ from the cathode with ions from the feed will occur based on the concentration of the specific ions in the feed. In continuous operation, protons were exchanged with Na^+ and K^+, since TAN concentration was mostly removed from the feed. As the same amount of ammonium is transported through the membranes, the Donnan Dialysis did not contribute to increase the overall TAN removal.

3.3. The energy input was lower than reported in previous studies

Ideally, a balance between NH_4^+ removal and energy input should be achieved when operating a (HR)ES [7]. The energy consumption for TAN removal in all Electrochemical Systems is mostly influenced by three factors: cell voltage, applied current density, and the TAN transport. The energy input for batch operation was between 9.3 kJ g_N^{-1} and 11.1 kJ g_N^{-1}, increasing with the load ratio (Table 1). The energy input for continuous operation was between 9.7 kJ g_N^{-1} and 14.2 kJ g_N^{-1} (Table 1). The energy input was lower for batch operation with Donnan Dialysis compared to without Donnan Dialysis, since additional ammonium was removed from the feed without additional energy input (Fig. 6). For continuous operation, there was no clear impact of Donnan Dialysis on the energy consumption. The higher energy input required in continuous compared to batch operation mode was a result of a higher internal resistance (due to ionic and membrane losses) built up during the longer operation with applied current in continuous mode. The higher concentration of positively charged ions in solution means more energy was spent to force ions with the same charge in this direction. The required specific energy input for ammonium removal was lower than Rodriguez Arredondo et al. (2017) [10] and Kuntke et al. [12]. The studied hydrogen recycling electrochemical system (HRES)

Table 1
Energy input per gN removed for batch and continuous operation.

	Continuous		Batch	
LN	E w/o Donnan (kJ g_N^{-1})	E w/ Donnan (kJ g_N^{-1})	E w/o Donnan (kJ g_N^{-1})	E w/ Donnan (kJ g_N^{-1})
0.8	9.7	10.1	9.3	7.8
1	11.5	11.7	10.3	9.0
1.3	13.4	14.2	11.1	10.1

achieved a TAN removal efficiency of 87% at Load Ratio 1.3 while consuming 13.4 kJ $g_N{}^{-1}$, when operating constinuously at 20 A m^{-2}. Rodriguez Arredondo et al., 2017 [10] achieved 83% TAN recovery while consuming 21.5 kJ $g_N{}^{-1}$ at 10 m^{-2} and the same load Ratio. Kuntke et al., 2017 [12] achieved 73% TAN recovery with a similar HRES while consuming 26.1 kJ $g_N{}^{-1}$ when operated at the same current density in Load Ratio.[8] Tarpeh et al., 2018 achieved a 93% efficiency in batch experiments with real urine, which required 30.6 MJ $kg_N{}^{-1}$ [4]. The "CapAmm cell" from Zhang et al., 2018 exhibited up to 90% ammonia removal and 80% recovery efficiencies. The energy consumption for synthetic urine was 28.1 MJ $kg_N{}^{-1}$ [11].

Donnan Dialysis is most suitable to enhance TAN recovery during batch operation, as it increases the removal efficiency and decreases the energy input of the process. Moreover, by operating an HRES system in batch mode, a higher removal can be achieved without additional membrane surface area and the need of a new cell.

4. Conclusion

In continuous operation, the benefits of including a dedicated Donnan Dialysis cell on the overall ammonium recovery were limited. Donnan Dialysis can be exploited to enhance TAN recovery in batch operation. It increased the transport of ammonium from feed to cathode and consequently the TAN removal. Additionally, it resulted in lower energy consumption for TAN removal.

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.

Acknowledgements

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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