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Life cycle assessment of environmental effects and nitrate removal for membrane capacitive deionization technology

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Abstract Nitrate is among the important types of pollutant sources in drinking water worldwide, and there are a number of methods to remove it from water. New treatment methods are being developed as an alternative to traditional treatment methods. One of them is membrane capacitive deionization (MCDI). In this study, the removal of nitrate ions with MCDI system was investigated. Nitrate solutions were treated via MCDI at different operating conditions. The obtained nitrate removal efficiency reached 83.07% at a flow rate of 2.5 L/min and a potential of 0.8 V was applied. In addition, during the nitrate removal of the MCDI system, the environmental effects were evaluated by life cycle analysis. As a result of these analyses, MCDI system offered advantages of low energy demand and low-energy environmental effects during operation. The results showed that the improved MCDI technology holds a great potential to be an energy-efficient process for nitrate removal. Life cycle assessment was applied to the experimental study. According to the assessment, water consumption had the highest effect in all damage assessment categories.

Keywords Nitrate · MCDI system · Life cycle assessment

Introduction

Rapid increasing population and improved living standards cause climate change, energy, and fresh water shortages (Hanjra and Qureshi 2010). The rapid increase in total water demand on a global scale and pollution of fresh water resources seriously threaten ecological balance (Malmqvist and Rundle 2002). Today, one third of the world population has difficulties in accessing fresh water (Damkjaer and Taylor 2017; Britto et al. 2019). Despite the increasing demand, technological developments do not show a sufficient development in terms of both water saving and widespread treatment systems, which will leave humanity to face water scarce in the next few decades (Robins 2019).

Nitrate ions are found naturally due to the transformations in the nitrogen cycle, and in addition, nitrate enters the water bodies through the sewage disposal system and livestock facilities. Nitrate is an important component for the growth of organisms and otherwise, at high concentrations, is known to have toxic effects on humans. Considering to the World Health Organization (WHO), the safe limit of nitrate concentration in drinking water is 10 mg/L (Uzun and Debik 2019). High concentrations pose health hazards for babies and pregnant women.

Membrane capacitive deionization (MCDI) has become a promising alternative among treatment system due to its low capital cost and energy demand (Choi et al. 2019a, b). MCDI is gaining more attention in recent years as a low-cost, energy-efficient, and environmentally friendly electrochemical technology (Tan

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et al. 2018; Dorji et al. 2018). Typical MCDI uses anion and cation exchange membranes, respectively, on the anode and cathode compartments (Choi et al. 2019a, b; Jain et al. 2019). The MCDI system has several uses, including water softening and removal of heavy metals and nutrients (Cetinkaya 2019). MCDI is a new electromembrane process that uses electrostatic adsorption to remove ions from a feed stream (Tan et al. 2020). The depletion of natural resources and environmental degradation has reached an alarming level, and the industry is exploring solutions to minimize these negative effects.

Life cycle assessment (LCA) is an environmental impact assessment method, the foundations of which were laid in the 1970s and systematically standardized and developed since the 1990s. The main feature that distinguishes LCA from other conventional methods is the whole life cycle (raw material extraction, manufacturing, use, disposal/recycling) of a product or service (i.e., product system), and all other sub-processes (transport, maintenance/repair, etc.) can be used to calculate all environmental impacts. LCA is also from one of the most important methods determined to reach the sustainability goals of the United Nations (Life Cycle Assessment-Theory and Practice 2018; Çetinkaya et al. 2018).

This study examined the removal of a nitrate solution via MCDI system, and during this removal, LCA analysis investigated the environmental impact. LCA is a valuable tool to evaluate environmental impacts associated with a technology and quickly find solutions to problems facing developing technologies (Bilgili et al. 2019). Nitrate removal was analyzed at different applied potentials and flow rates. Optimization of the applied voltage and flow rates was also performed for the definition of the optimum operating conditions. These results are expected to supply knowledge for the future development and promotion of MCDI for nitrate removal.

Materials and methods

MCDI system

The whole MCDI system was comprised of a feed vessel and peristaltic pump (Watson Marlow 350, USA), and applied voltage was supplied using a potentiostat (Gamry Interface 1000, USA).

The MCDI system was constructed by screwing Plexiglass plates to both ends, and the system consisted of cation exchange membrane Ultrex CMI-7000 (Membranes International Inc., USA), a spacer, and an anion exchange membrane (Neosepta AMX, Astom Co., Japan). The detailed structure of the MCDI system was thoroughly explained in our previous paper (Cetinkaya and Bilgili 2019).

Experimental methods

Nitrate concentration in water sample was 250 mg L^{-1}. The nitrate concentration in the water samples was determined using an ICS-3000 ion chromatography (Dionex, USA).

Nitrate removal (%) was calculated according to Eq. 1. input (J_0) and output (J) using the following equation:

$$\text{Percentage of nitrate removal} = \frac{J_0 - J_i}{J_0} \times 100 \qquad (1)$$

where J_0 and J are input and output for nitrate, respectively.

The LCA

The amount of materials used in the manufacturing of the filter system and the amount of electricity are presented in Table 1.

For LCA calculations, 8.0.2 version of SimaPro package program was used, which was developed by PRé Sustainability. ReCiPe 2008 Version 1.09 was preferred among the various methods embedded in this

Table 1 Material and energy amounts

Material	Amount
Carbon	37.97 g
Glass	66.47 g
Acrylic	89.14 g
Steel	18.01 g
Polyethylene	9.03 g
Rubber	0.25 g
Silicone	15 g
Water	0.75 l
Energy	
Electricity	0.275 Wh

program. ReCiPe 2008 was developed by Leiden University in the Netherlands, enabling comprehensive environmental LCA calculations in a wide range of fields. ReCiPe 2008 consists of eighteen midpoint categories that are linked to three endpoint categories and are used to calculate the environmental impacts of a product system and their final effects.

The human health category is used to examine the effects of emissions and wastes, which are generated during the life cycle stages of products or processes, directly on human health. Disability-adjusted loss of life years (DALY) is used as the unit. DALY refers to the loss caused by products or processes in a person's life expectancy. The ecosystem quality category is used to measure the impact of products or processes on the ecosystem. Its unit is species.yr which refers to the number of species lost within a year. Resources represent the economic loss caused by products or processes in the extraction of raw materials and the availability of resources, and US dollar is used as a unit (Goedkoop et al. 2008; Huijbregts et al. 2016).

In addition to these final values, various wastes and emissions known to be harmful to the environment are also taken into consideration. These emissions and wastes are presented in Table 2.

In addition, a sensitivity analysis was performed by evaluating the values obtained in all damage assessment and characterization categories in the short, medium, and long term. Sensitivity analysis allows evaluating

Table 2 Emission inventory

Compartment	Substance
Air	Carbon dioxide
	Carbon monoxide
	Dinitrogen monoxide
	Heat, waste
	Methane
	Nitrogen oxides
	Non-methane volatile organic compounds
	Particulates$_{2.5}$
	Sulfur dioxide
	Water vapor
Water	Arsenic
	Cadmium
	Chromium
	Lead
	Mercury
	Oils
	Solids, inorganic
	Suspended solids, unspecified
	Water
Soil	Arsenic
	Cadmium
	Chromium
	Lead
	Mercury
	Oils
Raw	Natural gas
	Forest occupation
	Permanent crop occupation
	Oil, crude

Fig. 1 Removal efficiency of nitrate at different potentials and flow rates

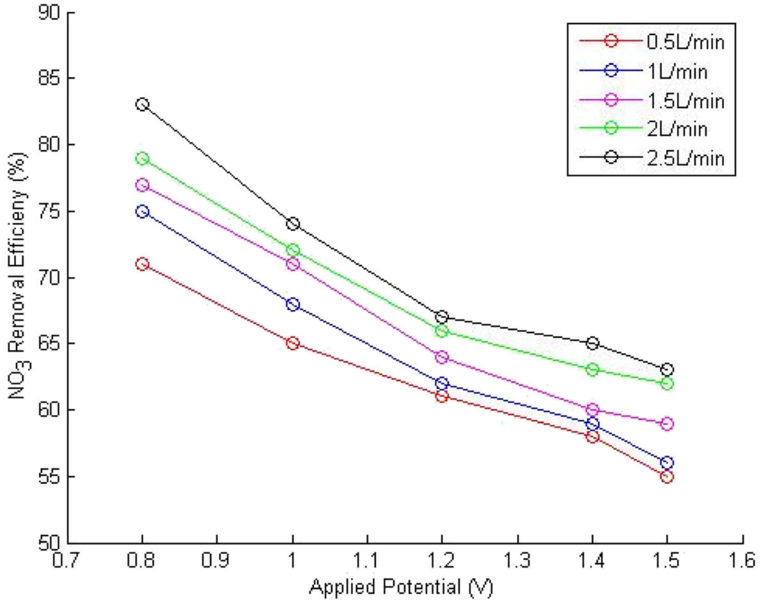


Table 3 Damage assessment values for three perspectives

Damage assessment category	*E*	*H*	*I*	Unit
Human health	0.0000068	0.000001	0.000001	DALY
Ecosystem quality	0.000000014	0.0000000045	0.0000000048	species.yr
Resources	0.056	0.056	0.031	US $

the environmental impacts in the short, medium, and long term and provides information on which process has more impact among the periods. Accordingly, situations that require precautionary measures can be determined in a short time and it can be decided which process should be preferred in the long term.

Sensitivity analysis is examined in three perspectives (Huijbregts et al. 2016):

(1) Individualistic (I): It is a 20-year short-term approach and is based on an optimistic basis in which future problems are assumed to be eliminated. Only strictly proven effects are examined in this perspective.

(2) Hierarchist (H): It is a 100-year medium-term approach that covers a period that the average person can experience and all known factors are evaluated. This perspective is also considered by default.

(3) Egalitarian (E): It is a long-term approach of 1000 years (500 years to infinity in some sources) and includes the most pessimistic scenarios. All known factors are evaluated, as in the H perspective.

Results and discussion

Optimization of operating conditions

The sampled water passing through the MCDI system was collected at different flow rates and applied potentials. The operating conditions were

Table 4 Characterization values for three perspectives

Characterization	*E*	*H*	*I*	Unit
Climate change	0.44	0.47	0.55	kg CO_2 eq
Ozone depletion	0.000000071	0.000000071	0.000000071	kg CFC-11 eq
Terrestrial acidification	0.0026	0.0024	0.0023	kg SO_2 eq
Freshwater eutrophication	0.00011	0.00011	0.00011	kg P eq
Marine eutrophication	0.000081	0.000081	0.000081	kg N eq
Human toxicity	7.02	0.16	0.018	kg 1,4-DB eq
Photochemical oxidant formation	0.0019	0.0019	0.0019	kg NMVOC
Particulate matter formation	0.0011	0.0011	0.0011	kg PM_{10} eq
Terrestrial ecotoxicity	0.00052	0.000053	0.000053	kg 1,4-DB eq
Freshwater ecotoxicity	0.010	0.010	0.010	kg 1,4-DB eq
Marine ecotoxicity	10.93	0.010	0.0079	kg 1,4-DB eq
Ionizing radiation	0.051	0.051	0.028	kBq U235 eq
Agricultural land occupation	0.042	0.042	0.042	m^2a
Urban land occupation	0.0053	0.005	0.0053	m^2a
Natural land transformation	0.00013	0.00013	0.00013	m^2
Water depletion	2.59	2.59	2.59	m^3
Metal depletion	0.27	0.27	0.27	kg Fe eq
Fossil depletion	0.22	0.22	0.22	kg oil eq

Table 5 Emission inventory

Compartment	Substance	Amount	Unit
Air	Carbon dioxide	428.88	g
	Carbon monoxide	903.52	mg
	Dinitrogen monoxide	11.43	mg
	Heat, waste	3.46	kJ
	Methane	1.54	g
	Nitrogen oxides	1.17	g
	NMVOC	416.89	mg
	PM2.5	356.61	mg
	Sulfur dioxide	1.70	g
	Water vapor	6.91	l
Water	Arsenic	1.08	mg
	Cadmium	495.02	μg
	Chromium	71.02	μg
	Lead	5.09	mg
	Mercury	23.65	μg
	Oils	87.46	mg
	Solids, inorganic	239.34	mg
	Suspended solids	1.46	g
	Water	15.82	mm^3
Soil	Arsenic	4.23	μg
	Cadmium	1.78	μg
	Chromium	23.21	μg
	Lead	25.26	μg
	Mercury	7.47	ng
	Oils	53.26	mg
Raw	Natural gas	94.61	l
	Forest occupation	412.45	cm^2
	Permanent crop occupation	7.47	mm^2
	Oil, crude	110.52	g

optimized. Especially, the examination of the applied potential and flow rates was carried out. As shown in Fig. 1, a lower applied voltage and a lower flow rate are shown to increase nitrate removal in sampled water.

Nitrate removal efficiency reached 83.08% at a flow rate of 2.5 L/min and an applied potential of 0.8 V. The removal efficiency of nitrate decreased by increasing the working voltages from 0.8 to 1.5 V. This study corroborated the results obtained in the previous study of Yeo and Choi (2013) and Kim et al. (2013). They indicated high removal performance of nitrate at lower voltages.

LCA results

According to the findings, the values in the damage assessment categories are as given in Table 3.

According to the values in Table 3, the short- and medium-term effects of the filter system on human health are the same and the system causes a loss on the average healthy life span of an average person by 0.000001 years. However, in the long term, this value increases by almost seven times to reach 0.0000068. Evaluating the results on the ecosystem presents that different effects occur in all three periods and the worst effect is seen in the long term with the value of 0.000000014. Accordingly, the filter system causes 0.000000014 species to disappear in 1 year. The medium- and long-term impact values of the filter system in terms of the negative effects it brings to resource use are $ 0.056 and the impact in the short term is seen to be less with the value of $ 0.031. Table 4 presents the characterization values.

Table 4 explains that the long-term effects are worse than the others, especially in the categories of human toxicity and marine ecotoxicity. Although other categories give approximately the same values, it was also observed that there are larger effects in the short term for the climate change category. The reason for the greater impact of climate change in the short term can be explained by the fact that the factors causing climate change lose their effects over time and are diluted in the atmosphere.

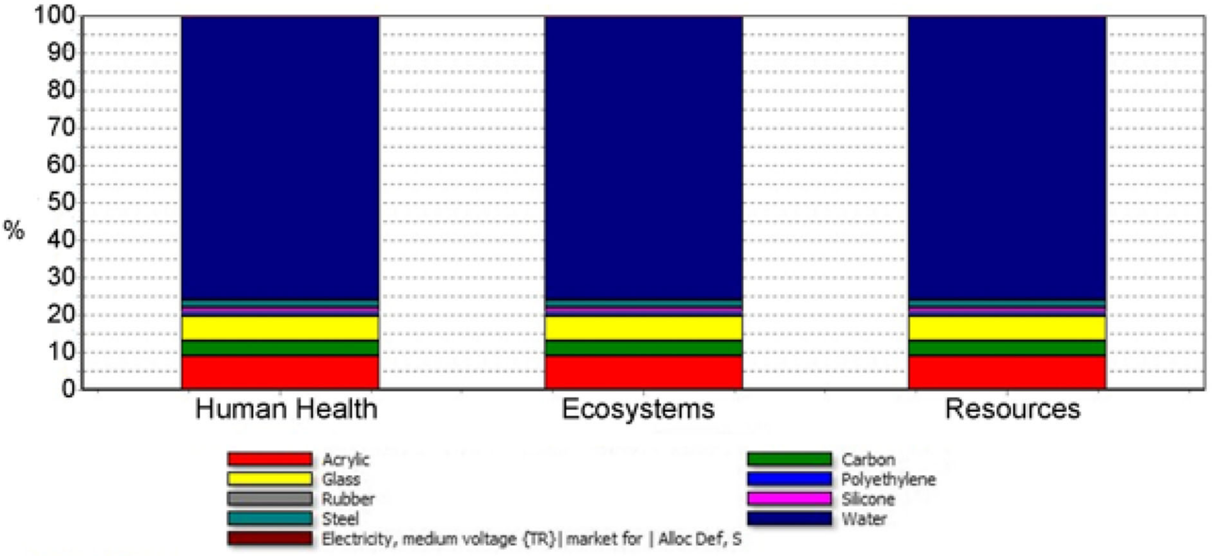
Table 5 presents the selected emission inventory. Accordingly, emissions to the air have a more dominant impact than the emissions to soil and water. Although heavy metals mixed with soil and water cause various environmental problems, these problems are expected to remain minor due to their small amount.

Figure 2 shows the share of the inputs shown in Table 1 for the total damage assessment effects.

According to Fig. 2, the consumed water has the greatest effect in all damage assessment categories. The water is followed by acrylic, glass, carbon, silicon, polyethylene, and steel, respectively. It was observed that electricity and rubber did not have much effect.

Similarly, Fig. 3 shows the shares of the inputs presented in Table 1 for the total characterization effects.

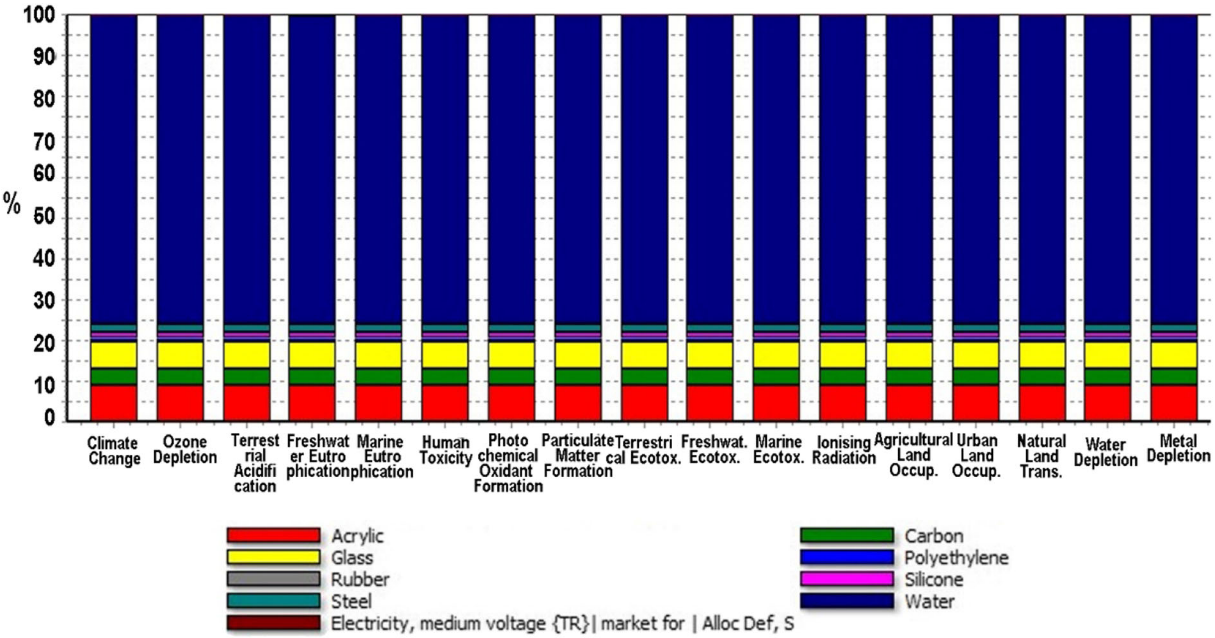
According to Fig. 3, just as in Fig. 2, the greatest environmental impact was due to water. The results were compared with the results of the study conducted by Yu et al. (2016), where LCA calculations of a filter



Analysing 1 p 'System1';
Method: ReCiPe Midpoint (H) V1.09 / Europe Recipe H / Characterisation

Fig. 2 Damage assessment

system were carried out. In this study, it was concluded that the majority of the total environmental impact originates from the materials used in manufacturing. Yu et al. (2016) calculated the values for climate change, human toxicity, freshwater aquatic ecotoxicity, marine aquatic ecotoxicity, terrestrial ecotoxicity, and photochemical oxidation as 0.808 kg CO_2 eq, 0.461 kg 1,4-DB eq, 0.17 kg 1,4-DB eq, 439 kg 1,4-DB eq, 0.00612 kg 1,4-DB eq, and 0.000224 kg 1,4-DB eq, respectively. It can be seen from Fig. 2 and Fig. 3 that the greatest environmental impact is due to consumed water. The effects of production and use processes of the



Analysing 1 p 'System1';
Method: ReCiPe Midpoint (H) V1.09 / Europe Recipe H / Characterisation

Fig. 3 Characterization effect

capacitive deionization system in Table 3, Table 4, and Table 5 are minor. The effects of the system on human health, ecosystem quality, and resources were calculated as 0.000001 DALY, 0.0000000045 species.yr, and $ 0.056, respectively. However, this calculation for only one system should not be misleading. Due to the large number of similar filters used worldwide, such systems can lead to significant environmental impacts in total. Similarly, the total emissions seen in Table 5 are also relatively low. In order to reduce these effects, as can be understood from Fig. 2 and Fig. 3, it is primarily aimed to develop innovative technologies that will reduce the water consumption. The amount of consumed electricity had lower effects in both studies compared with general environmental results. On the other hand, despite the conclusion of the current study that the water contributes to the overall environmental impact at a high rate, the effects of water were not included in the LCA calculations of the mentioned study. Thus, comparing the results with Table 4 presents quite different amounts due to the consumed water amount and the additional results of disposal process. Due to the lack of information, disposal process was not included in the current study. Besides, due to the different databases of the applied methods (CML 2000 and ReCiPe 2008), the differences in results are at expected levels.

Shiu et al. (2019) also conducted a study in which an LCA was implemented on membrane capacitive deionization (MCDI). The study also utilized CML 2000. The authors compared the LCA results of five scenarios in which 1 m^3 of desalinated product water was determined as the functional unit. The scenarios were S0, Original CDI-4; S1, Basic CDI-10; S2, Basic CDI-10 with inflows of secondary effluents; S3, Improved MCDI-10; and S4, Scale-up MCDI-40. According to the results, except ozone depletion and climate change, the material manufacturing phase had the greatest share on total life cycle inventory. Due to the differences between the scenarios, the LCA results also differ. S1 produced the greatest total impacts to the environment and S2 follows S1. The environmental impacts of S1 are greater because it uses more electricity and generates more waste. Because we did not add any waste disposal process in the current study, it is quite difficult to make a complete comparison; however, the impacts of the share of the materials remain greater compared with the energy processes followed by acrylic, glass, carbon, silicon, polyethylene, and steel, respectively.

Conclusion

In this study, the removal of nitrate with MCDI system and the factors affecting this removal efficiency were investigated. Nitrate removal can be decreased by increasing the applied current. However, nitrate removal rate also decreases at low flow rate. The MCDI system showed in this study demonstrated the benefits of low electricity consumption and low energy concerned with environmental impacts during its operational phases. Consequently, considering to experimental results, the MCDI process can be successfully used as a practical treatment in nitrate-contaminated waters. Considering that the use of water affects the marine ecotoxicity category in the long term, it can be considered that reducing the amount of water will reduce the overall environmental effects. It may be suggested as an alternative solution that the materials used in the filter system leave their places to more environmentally friendly ones. LCA study proposed that the majority of the total environmental impact is originated from the materials used in the manufacturing process. Also, the greatest environmental impact was due to water consumption. Moreover, emissions to the air have a more dominant impact than the emissions to soil and water.

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