Desalination and Water Treatment
www.deswater.com

doi: 10.5004/dwt.2021.26530

214 (2021) 8–15
February

Removal of nitrate and bromate ions from water in processes with ion-exchange membranes

Jacek A. Wiśniewski, Małgorzata Kabsch-Korbutowicz*

Department of Environment Protection Engineering, Wroclaw University of Science and Technology, Wybrzeze Wyspianskiego 27, 50-370 Wrocław, Poland, emails: malgorzata.kabsch-korbutowicz@pwr.edu.pl (M. Kabsch-Korbutowicz), jacek.wisniewski@pwr.edu.pl ((J.A. Wiśniewski)

Received 25 February 2020; Accepted 26 May 2020

ABSTRACT

Natural waters often contain ions, such as nitrate ions (NO_3^-), that are harmful to human health. Moreover, in the ozonation process of water containing bromide ions (commonly found in surface waters), bromate ions (BrO_3^-), which are harmful to health, are produced. The effectiveness of removing these ions from water in conventional water purification processes is low and insufficient. Studies were carried out on the removal of nitrates and bromates from aqueous solutions using processes with ion exchange membranes: Donnan dialysis and electrodialysis. In the Donnan dialysis process with anion exchange membranes (Selemion AMV), harmful anions are exchanged for neutral chloride ions. Satisfactory effects of bromate and nitrate removal were obtained at a NaCl concentration in the receiver equal to 300 mM: the bromate ions were completely removed from the purified solution and the concentration of nitrates was reduced by up to 22.9 mg/L. Two types of anion-exchange membranes were used in the electrodialysis process: typical Neosepta AMX membranes and Neosepta ACS mono-anion-selective membranes. The process with ACS/CMX membranes resulted in higher efficiency of bromate removal and similar results of nitrate removal when compared to the process with AMX/CMX membranes. At a current density of 20 A/m^2, the concentration of bromates was reduced to 0.97 μg/L, and the concentration of nitrates to 10.5 mg/L. These values are significantly lower than the limit values for drinking water: 10 μg/L (for bromates) and 50 mg/L (for nitrates).

Keywords: Bromates; Nitrates; Membrane; Donnan dialysis; Electrodialysis

1. Introduction

Surface water or groundwater that provides a source of drinking water for residents may contain ions which are harmful to human health. These include nitrates (NO_3^-). In waters containing bromide ions, harmful bromate ions are formed during the process of water ozonation. The effectiveness of conventional water treatment processes in removing these components from water is generally low and insufficient.

Nitrogen is essential for all living organisms, but its excessive consumption may lead to serious health problems. The intake of excessive nitrates via drinking water may lead to "blue baby syndrome" and could be responsible for an increased incidence of cancer in adults and children [1].

Taking into account the adverse effects of nitrates on consumer health, the maximum concentration of nitrates in drinking water has been set at 10 mg/L as N–NO_3^- (44.3 mg/L as NO_3^-) in the US and Canada [2]. A comparable limit value

* Corresponding author.

of nitrate ions concentration (50 mg/L) was established by the WHO [3], while the European Community standards allow a maximum admissible nitrate concentration of 50 mg/L and a guide level of 25 mg/L [4].

The average nitrate ions level in groundwater across Europe equals 18.8 mg/L, while in rivers the level is 1.72 mg/L [5]. Although a downward trend in nitrate concentrations has been observed over the last 20 y, thanks, among other things, to the implementation of the nitrates directive [6], concentrations of nitrate compounds, and above all nitrates in groundwater, exceed the recommended and acceptable values in the groundwater of many European countries. The reason for this phenomenon (the so-called nitrate problem) was the intensive application of mineral and organic fertilizers in cultivated areas, which, as a result of infiltration and surface run-off, caused the pollution of ground- and surface-water.

Increased nitrates concentrations in natural waters means that when used for food purposes they must be treated before being fed into the water supply system. The group of effective methods of removing ions from water includes ion exchange, reverse osmosis and electrodialysis, biological denitrification, and processes using membrane bioreactors [7]. The above methods allow for the effective removal of nitrate ions from water, but the application of some (reverse osmosis and electrodialysis) causes a simultaneous desalination of the water, which is not advisable in the case of waters with low salt content.

Bromide ions are a natural component of all ground- and surface-waters. Their concentration in surface water and groundwater ranges from a few to about 800 μg/L [8]. However, in some cases, the concentration of these ions reaches even 2 mg/L (Lake Galilee, Israel), and in Crete, in periods of drought, their concentration reaches 4 mg/L [9,10]. Natural sources of bromides occur when water is in contact with a substrate material (e.g., water contact with soil, where the typical bromide concentration is about 1 mg/kg), as well as in cases of intrusion of sea and ocean waters. Anthropogenic sources are also important: run-off from salted roads in winter, run-off from agricultural areas where brominated pesticides are used, and oil and gas produced waters [11].

To date, no harmful effects of the bromides present in drinking water on human health have been observed [12]. However, in the process of ozonation of water containing bromide ions, bromate ions (BrO_3^-) are formed. These ions are formed in the process of multistage bromide oxidation by molecular ozone and hydroxyl radicals [13]. The final concentration of bromates in water after ozonation depends, among others, on the concentration of bromides in the raw water and on the dose of ozone used for disinfection. The data presented in the paper [14] indicates that as a result of ozonation of water with a relatively low bromide concentration (160 μg/L), the content of bromates in the water after ozonation reaches 30 μg/L (assuming the ozone dose and contact time necessary for 99% inactivation of *Cryptosporidium oocysts*).

Bromates are ions with a carcinogenic effect on the human body. According to USEPA data, the lifetime risk of cancer is 10^{-4} (with a concentration of bromates in drinking water of 5 μg/L) and 10^{-5} (with a concentration of bromates of 0.5 μg/L) [15]. For this reason, the permitted concentration of bromates in drinking water is currently 10 μg/L [4].

In the group of methods used to remove bromates from water, adsorption on granulated activated carbon should first be mentioned [16,17]. In this process, bromate ions are reduced on the surface of activated carbon to bromide ions. However, the effectiveness of bromate adsorption is significantly reduced with time as a result of the gradual development of the biological membrane on the surface of activated carbon, and also due to adsorption of natural organic compounds on the surface of carbon. Very good effects of removing bromates (up to 96%) were obtained by adsorption on granular ferric hydroxide [18]. Membrane techniques are a very effective group of processes that allow the removal of bromates from water, either alone or in combination with other processes [19]. A high efficiency of bromate removal was also achieved in the hybrid process of coagulation – nanofiltration [20]. The ferrous sulfate used in this process completely reduces bromates to bromides, while the ferric hydroxide, arising from a chemical reaction, effectively removes humic acids and bromates remaining in the solution. Good effects of removing bromate ions from water were also obtained in pressure-driven membrane processes. In the process of reverse osmosis, 96% of bromate removal was achieved [21], and in the process of nanofiltration – 89% [22].

The paper presents the effects of removing nitrates and bromates from water in the processes of Donnan dialysis (DD) and electrodialysis (ED). The factors influencing the effectiveness of removing anions were analyzed, and both processes were compared in terms of removal efficiency.

2. Experimental methods

The Donnan dialysis process was carried out in a laboratory installation equipped with 20 cell pairs with the anion-exchange membranes Selemion AMV (Asahi Glass, Japan) (Table 1). The active area of the membranes was 0.140 m^2. The feeding solution contained the following components: $NaHCO_3$, $NaNO_3$, NaCl (the concentration of each component was 3 mM) and $NaBrO_3$ (100 μg/L = 0.782 μM of BrO_3^-). NaCl solution of 100, 200, or 300 mM concentration was used as the receiver. The process was conducted with recirculation of the feed and receiver until the lowest concentration of bromate ions in the feeding solution was reached. The solution (feed to receiver) volume ratio was 10:2.5 L.

The electrodialysis process was carried out in a laboratory installation containing 15 cell pairs with Neosepta AMX/CMX or Neosepta ACS/CMX membranes (ASTOM Corp., Japan). The total active area of the membranes was 0.104 m^2. The electrodialysis process was conducted at a constant current, as a batch-type operation. The electrodialysis process was conducted with recirculation of diluate and concentrate solutions (with the same initial composition as the feeding solution in the Donnan dialysis process) until the lowest concentration of bromate ions in the diluate was reached. The volume ratio of the solutions (dilute and concentrate) was 10:1.8 L.

The principle of Donnan dialysis and electrodialysis process for bromate and nitrate ions removal is presented in Fig. 1.

Table 1
Parameters of ion exchange membranes for Donnan dialysis and electrodialysis [23,24]

Parameter	Membrane			
	Selemion AMV	Neosepta ACS	Neosepta AMX	Neosepta CMX
Membrane type	Anion-exchange	Mono-anion-exchange	Anion-exchange	Cation-exchange
Electric resistance, Ω cm^2	2.8	2.0–2.5	2.5–3.5	2.5–3.5
Transport number: anions/cations	>0.96	>0.98	>0.98	>0.98
Transport number: (SO_4^{2-})		<0.005		
Exchange capacity, mmol/g	1.85	1.4–2.0	1.4–1.7	1.5–1.8
Water content, %	19.9	20–30	25–30	25–30
Thickness, mm	0.13	0.15–0.20	0.16–0.18	0.17–0.19

During the Donnan dialysis and electrodialysis processes, the concentration of anions in the circulating solutions was measured. The concentration of chloride and bicarbonate ions was determined by titration using, respectively, the solution of $AgNO_3$ and HCl [25]. The concentration of nitrate ions was determined spectrophotometrically using the DREL 2000 spectrophotometer from HACH (US) and NitraVer 5 reagent powder pillows reagent [26]. The concentration of bromate ions was measured spectrophotometrically using the UV mini 1240 spectrophotometer (Shimadzu, Japan) and reagents: 3,3′-dimethylnaphthidine and iodine [27]. In the case of the analytical methods used, the minimum detection limit for nitrates and bromates equaled 0.02 mM and 0.01 µM, respectively. Absorbance of the sample was measured at 550 nm. The mean measurement error did not exceed 10%.

For electrodialysis process, the energy consumption (W_e) was calculated according to the following equation:

$$W_e = \frac{I \cdot \int_0^t U\,dt}{V_d} \quad (1)$$

where W_e is the electrical energy demand per 1 L of treated solution, Wh/L, I is the current, A, U is the voltage, V, V_d is the volume of the diluate, L, and t is the duration of the process, h.

3. Results and discussion

3.1. Removal of anions from aqueous solutions using the Donnan dialysis process

In the Donnan dialysis process with an anion-exchange membrane, the anions present in the feeding solution (bromates, nitrates, and bicarbonates) are exchanged for neutral chloride ions. The driving force of the process is determined by the electrochemical potential gradient (expressed by the activity difference) of the Cl^- ions between the solutions separated by the membrane: the higher its value, the greater should be the stream of driving ions (here: Cl^- ions) from the receiving solution. The Cl^- ions transport to the feed results in an electrical potential difference between the solutions (referred to as Donnan potential [28]) which in turn causes an equivalent, opposite directed stream of ions from the feed to the receiver. This indicates the advisability of conducting tests at different salt concentrations in the receiver.

Fig. 2 shows the changes in the concentration of anions in the feeding solution in the DD process at a salt concentration in the receiver equal to 100 and 300 mM NaCl.

It can be seen that as a result of ion exchange, the concentration of harmful bromate and nitrate ions in the feeding solution decreases. At a salt concentration in the receiving solution of 100 mM NaCl, the concentration of BrO_3^- and NO_3^- ions were reduced to 0.27 µM (35.0 µg/L) and 0.92 mM (57.0 mg/L), respectively. These values were obtained in the time required to reach the minimum concentration of bromate ions in the purified solution. Together with harmful anions, bicarbonate ions are also removed from the solution – their concentration was reduced to 1.20 mM. It should be pointed out that the above anions are removed from the feeding solution at different rates, depending on the size of the ion and its concentration. The ions removed at the highest rate (measured by the ion flux through the anion-exchange membrane) are nitrates – the average flux of NO_3^- ions is 0.080 mol/m^2 h, the average flux of HCO_3^- ions is 0.055 mol/m^2 h, while the average flux of BrO_3^- ions is only 0.016 × 10^{-3} mol/m^2 h.

However, fully satisfactory effects of removing harmful anions (bromates and nitrates) were only obtained when

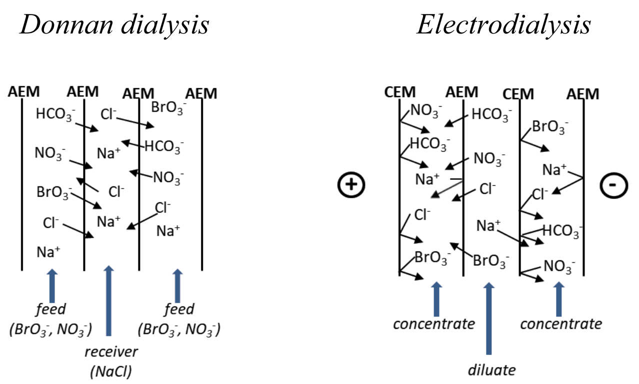


Fig. 1. Idea of bromate and nitrate ions separation in Donnan dialysis and electrodialysis processes (AEM: anion-exchange membrane, CEM: cation-exchange membrane).

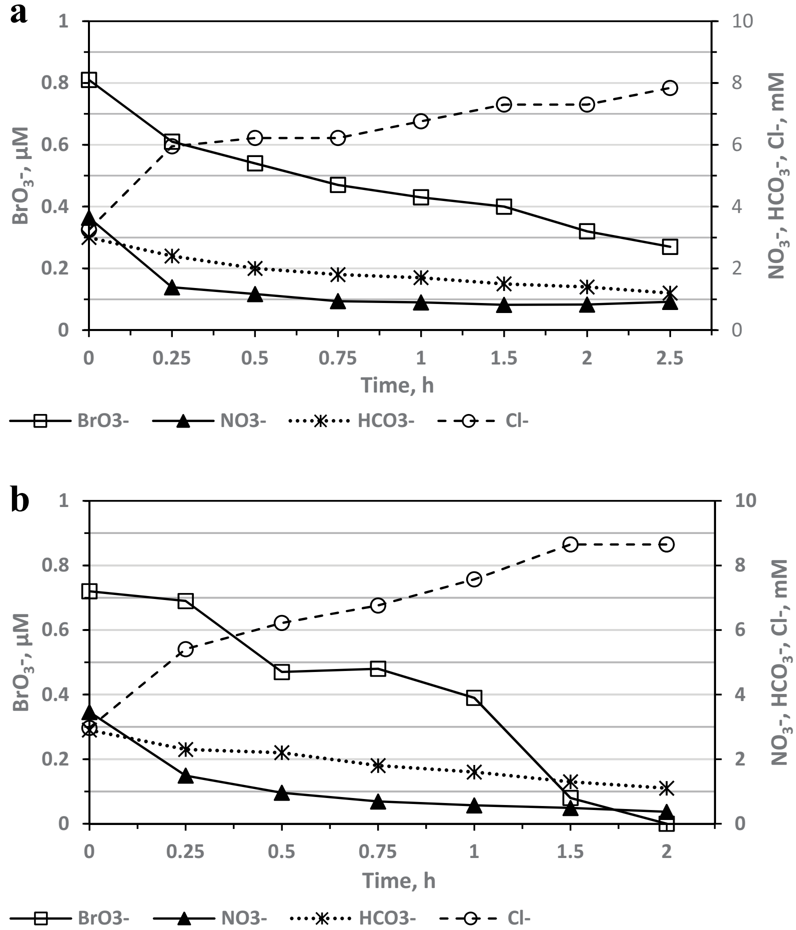


Fig. 2. Course of anion exchange in the Donnan dialysis process at a NaCl concentration in the receiver equal to 100 mM (a) and 300 mM (b).

the concentration of salts in the receiving solution was equal to 300 mM NaCl:BrO_3 ions were completely exchanged for Cl^- ions, whereas in the case of NO_3^- ions, their concentration was reduced to 0.37 mM (22.9 mg/L) (Fig. 2b). The concentration of bicarbonate ions was reduced to 1.10 mM (within the time required to obtain the minimum concentration of bromate ions in the treated solution). An increased salt concentration in the receiver also has a positive effect on the rate of anion exchange: as a result of an increase in the stream of driving ions (i.e., chloride ions), an appropriately high stream of anions is formed from the feeding solution to the receiving solution. Under such conditions (300 mM NaCl), the average stream of NO_3^- ions is 0.107 mol/m^2 h, the average stream of HCO_3^- ions is 0.061 mol/m^2 h, while the average stream of BrO_3^- ions reaches 0.023×10^{-3} mol/m^2 h.

Fig. 3 shows the dependence of the effectiveness of removing bromates, nitrates, and bicarbonates (as a result of exchange of these ions for chloride ions) on the concentration of NaCl in the receiving solution. The presented data show that in order to obtain a sufficiently low concentration of harmful bromate and nitrate ions in the treated water, a salt concentration in the receiver equal to 300 mM NaCl is required. This allows the total removal of BrO_3^- ions and 89.3% of NO_3^- ions to be removed (the concentration of nitrate ions after ion exchange is 22.9 mg/L and is much lower than the permissible value for drinking water [4]).

3.2. Removal of anions from aqueous solutions using electrodialysis

The process of electrodialysis of aqueous solutions was carried out at three current densities (i): 20, 25, and 30 A/m^2. These values were assumed on the basis of the calculated limiting current density ($i_{\lim}$) [29]. The value $i_{\lim}$ was computed on the following assumptions: mass transfer coefficient, k = 0.1 L/(m^2 s); final salt concentration in the diluate (assuming 85% salt removal), $C_d = 1.35 \times 10^{-3}$ M; transport number in the membrane and in the solution, T_m = 0.95 and T = 0.45, respectively. For the above values, the calculated value of $i_{\lim}$ amounts to 26 A/m^2.

Fig. 4 shows the changes in the concentration of ions in the diluate in the process of electrodialysis with AMX/CMX membranes at a current density of 20 and 30 A/m^2, respectively.

It can be seen that the electrodialysis process effectively removes typical anions (chlorides, bicarbonates) from the diluate, as well as anions harmful to human health (nitrates, bromates). Due to its small size (the radius of the hydrated ion is 0.335 nm [30]) and significant molar content (about 1/3 of all anions), the NO_3^- ion is removed from the diluate at the highest rate (Table 2). This reduces its concentration below the drinking water limit at a current density of 20 A/m^2 (to 0.69 mM, i.e., 42.8 mg/L) after 1.5 h. However, the BrO_3^- ion, which is characterized by a similar size (the radius of the ion is 0.351 nm [30]), but has a molar share three orders of magnitude smaller, is removed from the diluate at an adequate low velocity (Table 2). For this reason, in order to achieve satisfactory results in the removal of both harmful anions (bromates and nitrates), an increased current density of 30 A/m^2 is required – the BrO_3^- ion concentration is then reduced to 0.07 μM (9.5 μg/L), whereas the NO_3^- ion concentration is reduced to 0.16 mM (9.9 mg/L).

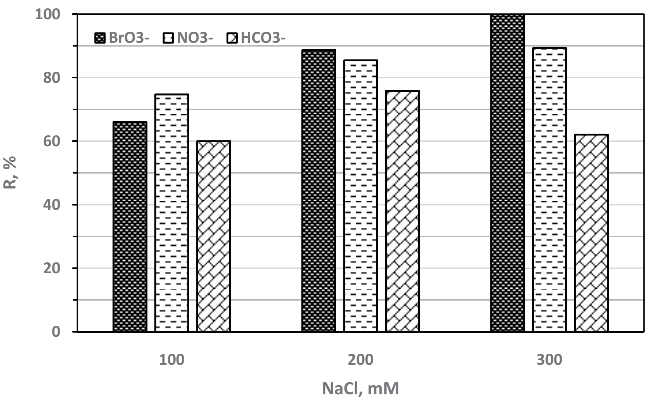


Fig. 3. Effects of anion removal in the Donnan dialysis process at different NaCl concentrations in the receiver.

Fig. 5 shows the influence of current density on the effectiveness of anion removal from water in the electrodialysis process with AMX/CMX membranes. A positive effect of current density increase on the effectiveness of removing harmful anions can be observed. In the studied range of current density (from 20 to 30 A/m^2), the removal of bromates increases from 86% to 90%, whereas for nitrates the removal is from 93% to 95%. The favorable current density in this process should be 30 A/m^2, which allows water with a bromate and nitrate ions content not exceeding the acceptable values in drinking water to be obtained. At a current density of 30 A/m^2, water containing 9.5 μg/L of BrO_3^- ions and 9.9 mg/L of NO_3^- ions was obtained.

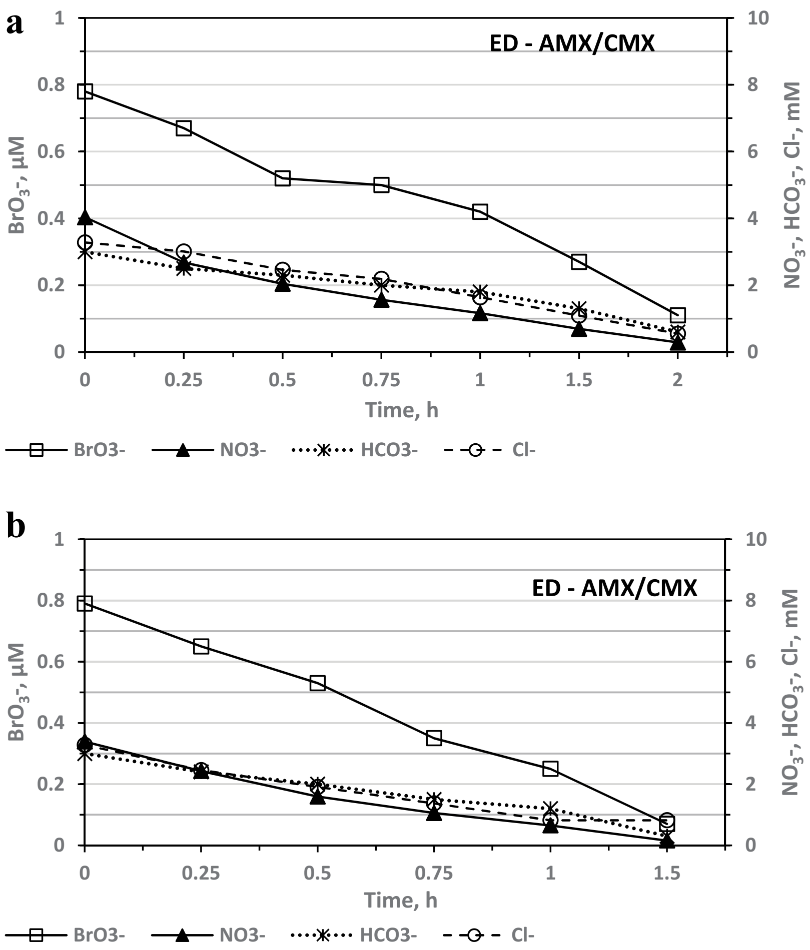


Fig. 4. Course of anion removal in the electrodialysis process with AMX/CMX membranes at a current density of 20 A/m^2 (a) and 30 A/m^2 (b).

Table 2
Comparison of ion fluxes for anions removed from the diluate using electrodialysis with AMX/CMX membranes at different current densities (t = 1.5 h)

i, A/m^2	Average ion flux			
	BrO_3^-, mol/m^2 h	Cl^-, mol/m^2 h	NO_3^-, mol/m^2 h	HCO_3^-, mol/m^2 h
20	0.033 × 10^{-3}	0.141	0.210	0.111
25	0.037 × 10^{-3}	0.171	0.201	0.151
30	0.045 × 10^{-3}	0.198	0.200	0.168

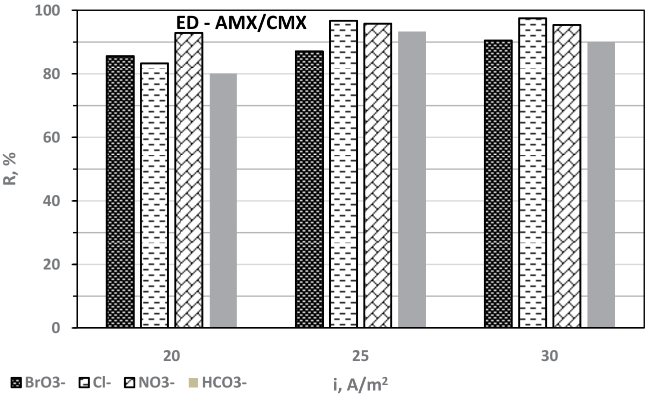


Fig. 5. Effectiveness of anion removal at different current densities in electrodialysis with AMX/CMX membranes.

Fig. 6 shows the changes in the concentration of ions in the diluate in the process of electrodialysis with ACS/CMX membranes, where the ACS membrane has mono-anion-selective properties. It can be observed that as a result of the application of the mono-anion-selective membrane (Neosepta ACS), the transport of large bicarbonate anions is slowed down (the radius of the hydrated HCO_3^- ion is 0.394 nm [30]). This is accompanied by intensification of the transport of the remaining anions, including bromates. As a result of this phenomenon, satisfactory effects of removing both harmful anions were obtained at the current density of 20 A/m^2: the concentration of BrO_3^- ions was reduced to 0.01 μM (0.97 μg/L), and the concentration of NO_3^- ions to 0.17 mM (10.5 mg/L).

The limited transport of large anions through the Neosepta ACS membrane is the result of the characteristic structure of this membrane, which has a thin, strongly cross-linked layer on its surface that hinders the flow of multivalent anions and large monovalent anions [28]. It is only at high a current density (here: 30 A/m^2) that HCO_3^- ions are removed at an increased rate (Table 3).

Comparing the efficiency of the electrodialysis process with AMX/CMX membranes and ACS/CMX membranes, it can be stated that the use of mono-anion-selective membranes (Neosepta ACS) allows a higher efficiency of bromate removal to be achieved and a similar efficiency of nitrate removal from water to be obtained – even at a low current density (here: 20 A/m^2). This is accompanied by an increased retention of bicarbonates, which (due to their large size) are retained to a greater extent by the Neosepta ACS membrane than by the standard Neosepta AMX membrane. The latter effect is particularly important when water has a low HCO_3^- ion concentration.

3.3. Comparison of the efficiency of Donnan dialysis and electrodialysis in the removal of bromate and nitrate ions from aqueous solutions

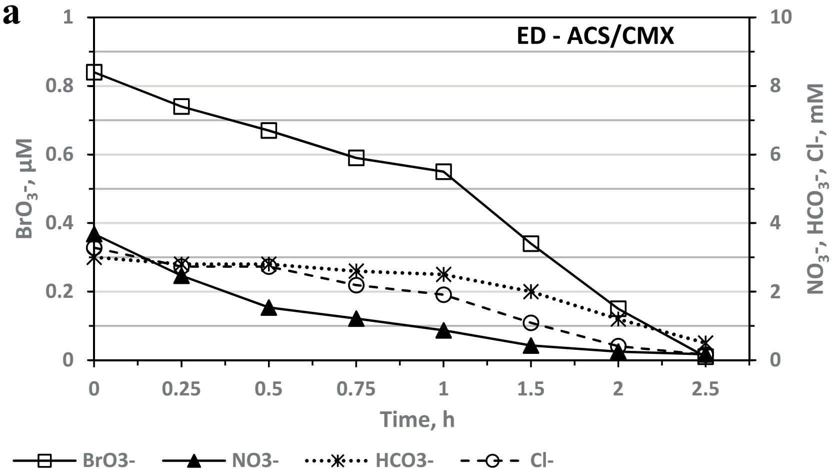
The effectiveness of the membrane processes (DD and ED) was compared on the basis of the efficiency and rate of removing bromate and nitrate ions. The conditions of the above-mentioned processes, which allow the reduction of the concentration of these ions below the acceptable values for drinking water, were taken into account [4]. This means that it is necessary to carry out Donnan dialysis when the concentration of salt in the receiver is equal to 300 mM NaCl. On the other hand, in the electrodialysis process with the AMX/CMX membranes, the required current density is 30 A/m^2, and with the ACS/CMX membranes, satisfactory effects of removing harmful anions can be achieved at a lower current density of 20 A/m^2.

Table 4 presents selected parameters characterizing the effectiveness of Donnan dialysis and electrodialysis in removing harmful anions from aqueous solutions.

It can be seen that Donnan dialysis and electrodialysis with mono-anion-selective membranes provide the highest

Table 3
Comparison of ion fluxes for anions removed from the diluate using electrodialysis with ACS/CMX membranes at different current densities (t = 1.5 h)

i, A/m^2	Average ion flux			
	BrO_3^-, mol/m^2 h	Cl^-, mol/m^2 h	NO_3^-, mol/m^2 h	HCO_3^-, mol/m^2 h
20	0.032 × 10^{-3}	0.141	0.203	0.072
25	0.038 × 10^{-3}	0.179	0.208	0.117
30	0.044 × 10^{-3}	0.192	0.204	0.151



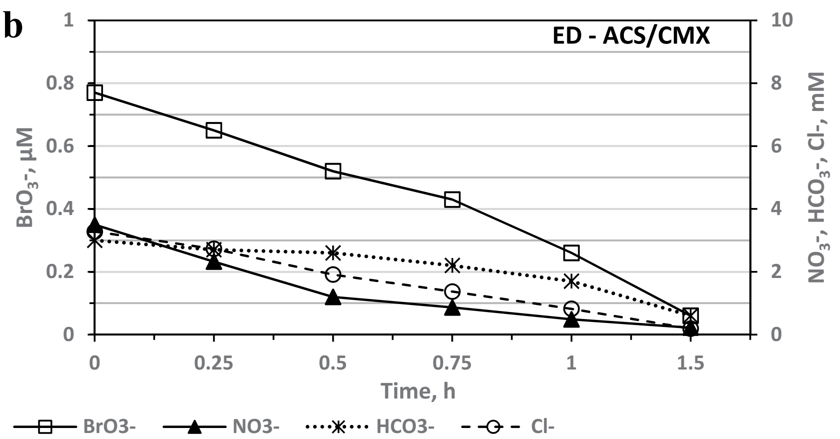


Fig. 6. Course of anion removal in the electrodialysis process with ACS/CMX membranes at a current density of 20 A/m² (a) and 30 A/m² (b).

Table 4
Comparison of the efficiency of removing bromates and nitrates from aqueous solutions in the Donnan dialysis and electrodialysis processes (C_e: final concentration, R: removal efficiency, J: average ion flux, T: process time, W_e: energy consumption for ion transport)

Process	BrO_3^-			NO_3^-			T, h	W_e, Wh/L
	C_e, µg/L	R, %	J, mol/m² h	C_e, mg/L	R, %	J, mol/m² h		
Donnan dialysis (AMV, 300 mM NaCl)	0.0	100.0	0.026×10^{-3}	22.9	89.3	0.107	2.0	–
Electrodialysis (AMX/CMX, 30 A/m²)	9.5	90.5	0.044×10^{-3}	9.9	95.4	0.201	1.5	0.41
Electrodialysis (ACS/CMX, 20 A/m²)	0.97	99.1	0.031×10^{-3}	10.5	95.3	0.131	2.5	0.34

efficiency in removing bromate ions. The rate of removing these ions from aqueous solutions is also similar, which makes the duration of both processes (to obtain a minimum concentration of bromates in the solution) similar. In the case of nitrate ions, electrodialysis (regardless of the type of anion-exchange membranes) allows a greater efficiency to be achieved in removing these ions than Donnan dialysis.

Therefore, it can be assumed that electrodialysis with Neosepta ACS/CMX membranes enables the best results to be obtained in terms of removing harmful BrO_3^- and NO_3^- ions from water. It should also be noted that in the electrodialysis process there is additional electricity consumption for ion transport, while in the Donnan dialysis process the ion transport is caused by a gradient of the concentrations of driving ions (i.e., chlorides).

4. Conclusions

- The processes with ion exchange membranes, that is, Donnan dialysis and electrodialysis, effectively remove harmful anions (bromates and nitrates) from water, reducing their concentration below the limit value for drinking water.

- Donnan Dialysis with the anion-exchange membrane Selemion AMV causes the exchange of anions in the purified solution (bromates, nitrates, and bicarbonates) for neutral chloride ions. In order to obtain a final concentration of harmful anions below the drinking water limit, a relatively high concentration of salt in the receiver of 300 mM NaCl is required.
- As a result of anion exchange in the Donnan dialysis process (at a 300 mM NaCl salt concentration in the receiver), the bromate ions are completely removed from the purified solution and the concentration of nitrate ions is reduced to 22.9 mg/L (89.3% removal). Bicarbonate ions are also removed in this process – the concentration of these ions is reduced to 1.10 mM (62.1% removal).
- In the electrodialysis process with typical Neosepta AMX/CMX ion-exchange membranes, satisfactory results in the removal of bromates and nitrates from aqueous solutions can be obtained at a current density of 30 A/m^2: the concentration of bromates is reduced to 9.5 μg/L (90.5% removal) and the concentration of nitrates to 9.9 mg/L (95.4% removal). In this process, the concentration of bicarbonates is reduced to 0.3 mM (90.0% removal).
- In the process of electrodialysis with Neosepta ACS/CMX membranes (Neosepta ACS membrane is a mono-anion-selective membrane), better bromate removal effects and similar, when compared to the process with Neosepta AMX/CMX membranes, nitrate removal effects can be achieved. At a current density of 20 A/m^2, the concentration of bromates is reduced to 0.97 μg/L (99.1% removal) and the concentration of nitrates to 10.5 mg/L (95.3% removal), whereas the concentration of bicarbonates is reduced to 0.5 mM (83.3% removal).
- Comparing both processes with ion exchange membranes in terms of the effectiveness of removing bromate and nitrate ions from water, it can be assumed that electrodialysis with ACS/CMX membranes achieves the best results – with a relatively low energy consumption for ion transport (0.34 Wh/L).

References

[1] M.H. Ward, R.R. Jones, J.D. Brender, T.M. de Kok, P.J. Weyer, B.T. Nolan, C.M. Villanueva, S.G. van Breda, Drinking water nitrate and human health: an updated review, Int. J. Environ. Res. Public Health, 15 (2018) 1–31.

[2] USEPA, National Primary Drinking Water Regulations. Available at: https://www.epa.gov/ground-water-and-drinking-water/table-regulated-drinking-water-contaminants (accessed April 30, 2020).

[3] WHO, Guidelines for Drinking-Water Quality, 4th ed., World Health Organization, Geneva, 2017.

[4] Council Directive 98/83/EC of 3 November 1998 on the Quality of Water Intended for Human Consumption.

[5] Nutrients in Freshwater in Europe, 2019. Available at: www.eea.europa.eu

[6] Council Directive of 12 December 1991 Concerning the Protection of Waters against Pollution Caused by Nitrates from Agricultural Sources (91/676/EEC).

[7] P.M. Ayyasamy, S. Rajakumar, M. Sathishkumar, K. Swaminathan, K. Shanthi, P. Lakshmanaperumalsamy, S. Lee, Nitrate removal from synthetic medium and groundwater with aquatic macrophytes, Desalination, 242 (2009) 286–296.

[8] M. Soyluoglu, M.S. Ersan, M. Ateia, T Karanfil, Removal of bromide from natural waters: bromide-selective vs. conventional ion exchange resins, Chemosphere, 238 (2020) 1–9.

[9] R.S. Magazinovic, B.C. Nicholson, D.E. Mulcahy, D.E. Davey, Bromide levels in natural waters: its relationship to levels of both chloride and total dissolved solids and the implications for water treatment, Chemosphere, 57 (2004) 329–335.

[10] A.A. Kampioti, E.G. Stephanou, The impact of bromide on the formation of neutral and acidic disinfection by-products (DBPs) in Mediterranean chlorinated drinking water, Water Res., 36 (2002) 2596–2606.

[11] F. Soltermann, C. Abegglen, C. Götz, U. von Gunten, Bromide sources and loads in Swiss surface waters and their relevance for bromate formation during wastewater ozonation, Environ. Sci. Technol., 50 (2016) 9825–9834.

[12] T. Myllykangas, T.K. Nissinen, A. Hirvonen, P. Rantakokko, T. Vartiainen, The evaluation of ozonation and chlorination on disinfection by-product formation for a high-bromide water, Ozone Sci. Eng., 27 (2005) 19–26.

[13] U. Pinkernell, U. von Gunten, Bromate minimization during ozonation: mechanistic considerations, Environ. Sci. Technol., 35 (2001) 2525–2531.

[14] K. Tyrovola, E. Diamadopoulos, Bromate formation during ozonation of groundwater in coastal areas in Greece, Desalination, 176 (2005) 201–209.

[15] B.M. De Borba, J.S. Rohrer, C.A. Pohl, C. Saini, Determination of trace concentrations of bromate in municipal and bottled drinking waters using a hydroxide-selective column with ion chromatography, J. Chromatogr. A, 1085 (2005) 23–32.

[16] D. Barlokova, J. Ilavsky, I. Marko, J. Tkacova, Removal of bromates from water, IOP Conf. Ser.: Earth Environ. Sci., 92 (2017) 1–5.

[17] Y.Q. Zhang, Q.P. Wu, J.M. Zhang, X.H. Yang, Removal of bromide and bromate from drinking water using granular activated carbon, J. Water Health, 13 (2015) 73–78.

[18] A. Bhatnagar, Y.H. Choi, Y.J. Yoon, Y. Shin, B.H. Jeon, J.W. Kang, Bromate removal from water by granular ferric hydroxide (GFH), J. Hazard. Mater., 170 (2009) 134–140.

[19] M.A. Zazouli, L.R. Kalankesh, Removal of precursors and disinfection by-products (DBPs) by membrane filtration from water; a review, J. Environ. Health Sci. Eng., 12 (2017) 15–25.

[20] K. Listiarini, J.T. Tor, D.D. Sun, J.O. Leckie, Hybrid coagulation–nanofiltration membrane for removal of bromate and humic acid in water, J. Membr. Sci., 365 (2010) 154–159.

[21] J.P. van der Hoek, D.O. Rijnbende, C.J.A. Lokin, P.A.C. Bonne, M.T. Loonen, J.A.M.H. Hofman, Electrodialysis as an alternative for reverse osmosis in an integrated membrane system, Desalination, 117 (1998) 159–172.

[22] T.F. Marhaba, K. Bengraine, Review of strategies for minimizing bromate formation resulting from drinking water ozonation, Clean Technol. Environ. Policy, 5 (2003) 101–112.

[23] Asahi Glass Company, Selemion Ion Exchange Membranes. Available at: https://www.amp-ionex.com/products/selemion/pdf/selemion.pdf (accessed May 19, 2020).

[24] ASTOM Corp. (Former Tokuyama Corp.), Neosepta, Ion Exchange Membranes, Japan.

[25] E.W. Rice, R.B. Baird, A.D. Eaton (Eds.), Standard Methods for the Examination of Water and Wastewater, 23rd ed., American Public Health Association, American Water Works Association, Water Environment Federation, 2017.

[26] Hach Water Analysis Handbook, Photometric Procedures, Hach Company, US, 1997.

[27] Merck Applications, Bromate in Water and Drinking Water, Photometric Determination with 3,3'-Dimethylnaftidin and Iodine, US.

[28] H. Strathmann, Ion-Exchange Membrane Separation Processes, Elsevier, Amsterdam, 2004.

[29] R. Rautenbach, Procesy Membranowe, WNT, Warszawa, 1996 (Polish ed.).

[30] E.R. Nighitingale, Phenomenological theory of ion solvation. Effective radii of hydrated ions, J. Phys. Chem., 63 (1959) 1381–1387.