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Donnan dialysis and electrodialysis as viable options for removing bromates from natural water

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ABSTRACT

Donnan dialysis with an anion-exchange membrane and electrodialysis were used for removing bromate ions from natural water. Donnan dialysis with the Selemion AMV membrane provided total bromate removal from water containing 50 μg BrO_3^-/L, at a salt concentration in the receiver of 100 mM NaCl. The exchange of bromates for chloride ions was concomitant with the exchange of associated anions: sulfates (93% efficiency) and bicarbonates (73% efficiency). Donnan dialysis with the Neosepta ACS membrane (which is characterized by a compact surface structure) also provided total removal of bromates, but the efficiency with which SO_4^{2-} and HCO_3^- were exchanged for chloride ions was far below that achieved with Selemion AMV (3% and 47%, respectively). Electrodialysis with standard ion-exchange membranes, Neosepta AMX/CMX, was less efficient at removing bromates (83%), but was able to reduce their concentrations in the treated water (8.8 μg/L) to a level lower than the maximum admissible value for potable water (10 μg/L). The efficiency of bromate removal by electrodialysis can be enhanced using the mono-anion-selective membrane Neosepta ACS, which augments the extent of removal to 94%, the concentrations of bromates in the treated water being as low as 3.3 μg/L.

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1. Introduction

Bromide ions are ubiquitous in natural water, and their concentration may reach 2 mg/L [1]. Although bromides are not reported to be detrimental to public health, their presence in the water being disinfected is a contributing factor in the formation of harmful disinfection by-products (DBP). During water disinfection with chlorine, bromides oxidize to hypobromous acid (HOBr), which reacts with natural organic matter (NOM) to form deleterious brominated trihalomethanes (THM) [1]. When water disinfection entails ozone, the bromides that are present in the water oxidize to hypobromite ions (OBr^-) and thereafter to bromates (BrO_3^-) [2]. Bromates are ions exerting a carcinogenic effect on human organisms. According to the data published by the US Environmental Protection Agency, the lifetime risk of cancer disease amounts to 10^{-4} (at a concentration of 5 μg BrO_3^-/L in drinking water) and 10^{-5} (at a concentration of 0.5 μg BrO_3^-/L) [3]. In the USA, as well as in the member states of the European Union, the admissible concentration of bromates in potable water is now 10 μg/L [3]; the same value being recommended by the World Health Organization [4].

These data offer clear evidence that bromide ions (as precursors of detrimental DBPs) or bromate ions need to be removed from water. Aluminum chloride coagulation provides the highest bromide removal efficiency which amounts to 87% at the coagulant dose of 15 mg/L [5]. The extent of bromide removal obtained with other methods is low: up to 10% with nanofiltration [6] and 17% with the process involving the anion-exchange resin MIEX® [7].

Among the group of methods used for the removal of bromates from drinking water, adsorption onto granular activated carbon deserves special attention [8,9]. In this process, on the surface of the activated carbon, bromate ion (BrO_3^-) is reduced to bromide ion (Br^-). However, after a certain time of adsorption, a noticeable rise in the concentration of bromates was observed in the water being treated. It is attributable to the gradual transition of new granular activated carbon to biological activated carbon [8] and to the adsorption of natural organic matter onto the carbon surface [9].

Very promising efficiency of bromate removal from water (up to 96%) was achieved in an adsorption process involving granular ferric hydroxide [10]. High removal of bromates has also been reported for the hybrid coagulation–nanofiltration process [11]. Ferrous sulfate provided complete reduction of bromates to bromides that, however, persist in the water, because the nanofiltration process was able to produce only a low extent of their removal (up to 17%).

A high extent of bromate ion separation, which totaled 96%, was reported for the process of reverse osmosis. Electrodialysis reversal (EDR) provided lower removal efficiencies, which failed to exceed 64% [12]. UV irradiation and alum coagulation were found to produce poor bromate removal, amounting to 19% [13] and 26% [14], respectively.

One of the attractive methods for removing bromate ions from water is Donnan dialysis. The process entails anion exchange between

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two solutions separated by an anion-exchange membrane; the transport of the driving anions (*e.g.* chlorides) from the receiver to the feeding solution induces an equivalent, oppositely directed anion flow to the receiver [15]. In this way the troublesome anions that occur in the solution being treated (*e.g.* bromates) are replaced with neutral ions from the receiver (*i.e.* chlorides).

Donnan dialysis has been successfully applied to removing two deleterious anions from natural water — fluoride [16] and nitrate [17]. As a consequence of anion exchange, the concentrations of these harmful ions were reduced to levels below those recommended for potable water (1.5 mg F^-/L and 50 mg NO_3^-/L). Velizarov and co-workers [18] used an ion-exchange membrane bioreactor, which enabled simultaneous removal of perchlorate, bromate and nitrate ions and, thereafter, biological reduction of the anions to harmless products (chlorides, bromides and gaseous nitrogen, respectively). The extent of anion removal attained *via* this route was very high, amounting to 91% and 99% for bromates and nitrates, respectively.

Our previous studies involving a multi-component model solution have shown that Donnan dialysis with the anion-exchange membrane Selemion AMV produces a high efficiency of bromate ion removal [19,20]. For a solution of a concentration of 50 μg BrO_3^-/L, the extent of bromate ion removal totaled 85% at a concentration of 300 mM NaCl in the receiver [20]. This paper here presents the results of our research onto bromate ion removal from natural water by two ion-exchange membrane processes — Donnan dialysis and electrodialysis.

2. Methods

Donnan dialysis was performed in a laboratory dialytic set-up containing 20 cell pairs with anion-exchange membranes, Selemion AMV (Asahi Glass) or Neosepta ACS (Tokuyama Corp.). Table 1 compiles some major parameters of the membranes.

The working area of the membranes totaled 0.140 m^2. The process was conducted with recirculation of the feed and the receiver until the minimum concentration of bromate ions in the feed was attained. The volume ratio of the feed to the receiver was 4:1 (10 L:2.5 L). The feed was natural water (its composition is shown in Table 2) with a total anion concentration of 3.7 mM. The natural water was enriched with bromate salt (50 μg BrO_3^-/L). The receiver was a solution containing NaCl with concentrations of 50, 100, 200 or 300 mM.

Electrodialysis was conducted in the laboratory set-up Goemasep 136, which contained 15 cell pairs with Neosepta AMX/CMX or Neosepta ACS/CMX membranes (Table 3). The working surface area of the ion-exchange membranes was 0.112 m^2, and this included the working surface area of the anion-exchange membranes, which amounted to 0.058 m^2. The distance between the membranes in the stack was 0.5 mm and linear velocity of the solutions in the electro-dialyser cells amounted to 10 cm/s. The process was performed at a constant current density (14 A/m^2), with recirculation of diluate and concentrate, until bromate ion concentration in the diluate reached a minimal value (duration of the process was 1.5 h). The diluate and concentrate loops consisted of natural water (of the composition specified above), which contained bromate ions. The diluate-to-concentrate volume ratio was 10 L:1.8 L.

Anion concentrations in the water being treated were measured in the course of the processes. Concentrations of bicarbonates and chlorides were determined by titration with color indicators [23,24]. Bromate concentration was measured photometrically with 3,3′-dimethylnaftidin and iodine [25], using a UV mini 1240 spectrophotometer (Shimadzu) and 50 mm glass cuvettes. Sample absorbance was established at the wavelength of 550 nm. Each anion concentration was measured twice. The mean measurement error did not exceed 10%.

Based on the measured results, the efficiency of anion removal and the average anion flux from the water being treated to the receiver (concentrate) were determined. Average anion flux was calculated taking into account the working area of the anion-exchange membrane (0.140 m^2 for Donnan dialysis and 0.058 m^2 for electrodialysis).

Table 1
Parameters of the anion-exchange membranes used in the Donnan dialysis process [21,22].

Parameter	Membrane	
	AMV	ACS
Electric resistance, Ω·cm^2	1.5–3.0	2.0–2.5
Transport number		
✓ Cl^-	>0.94	>0.98
✓ SO_4^{2-}		<0.005
Exchange capacity, mmol/g	1.85	1.4–2.0
Water content,%	19.9	20–30
Thickness, mm	0.11	0.15–0.20
Membrane type	Anion-exchange	Mono-anion-exchange

Table 2
Major parameters of natural water.

Parameter	Unit	Value
Conductivity	μS/cm	105.6
pH		5.6
Alkalinity	mg/L	150
Chlorides	mg/L	13.6
Sulfates	mg/L	29
Nitrates	mg/L	0.9
Total hardness	°dH	8.9
Sodium	mg/L	7.6
Potassium	mg/L	1.3

3. Results

3.1. Donnan dialysis

Anion exchange in natural water during Donnan dialysis with Selemion AMV, at NaCl concentration of 100 mM in the receiver, is depicted in Fig. 1. As shown by these data, both bromates and sulfates are exchanged for chlorides with high efficiency. In consequence of anion exchange, BrO_3^- ions were removed completely, and SO_4^{2-} ions with 93% efficiency. The exchange of bicarbonates for chlorides, however, was noticeably less efficient (73%).

The diverse efficiency of exchanging anions for chloride ions is attributable to the size of the anions being removed. In terms of the size of the hydrated radius, HCO_3^- is the largest anion (0.394 nm), SO_4^{2-} and BrO_3^- ranking second and third (0.379 nm and 0.351 nm, respectively) [26]. It is essential to note that the proportion of the bicarbonate ions is the largest in the flux of the anions being removed from the water. The average flux of bicarbonate ions (until their minimal concentration in the water being treated has been attained) amounts to 0.079 mol/m^2·h, the average fluxes of sulfate ions and bromate ions being equal to 0.020 mol/m^2·h and 0.011 · 10^{-3} mol/m^2·h, respectively. The comparatively

Table 3
Parameters of the anion-exchange membranes used in the electrodialysis process [22].

Parameter	Membrane		
	AMX	ACS	CMX
Electric resistance, Ω·cm^2	2.5–3.5	2.0–2.5	2.5–3.5
Transport number			
✓ Total anion or cation	>0.98	>0.98	>0.98
✓ SO_4^{2-}		<0.005	
Exchange capacity, mmol/g	1.4–1.7	1.4–2.0	1.5–1.8
Water content,%	25–30	20–30	25–30
Thickness, mm	0.16–0.18	0.15–0.20	0.17–0.19
Membrane type	Anion-exchange	Mono-anion-exchange	Cation-exchange

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