The removal of chromium ions from water in Donnan dialysis process

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ABSTRACT

The paper presents results of the studies on the removal of chromium ions ($Cr_2O_7^{2-}$) from multicomponent water solutions in the process of Donnan dialysis with anion-exchange membrane. The feeding solution contained the following salts: $NaNO_3$, $NaHCO_3$, NaCl (with a concentration of 3 mM for each component), and $Na_2Cr_2O_7$ (with concentrations of 100, 300, or 500 µg Cr(VI)/L). The Donnan dialysis process was conducted using a laboratory dialytic set-up equipped with 10 cell pairs with anion-exchange membranes, Selemion AMV (Asahi Glass, Japan) or Neosepta ACS (ASTOM Corp., Japan). The total working area of the membranes was 0.0756 m². NaCl solutions with concentrations of 100, 200, or 300 mM were used as receivers. The best effects of the anion exchange were obtained in the process with Selemion AMV membrane. Dichromate ions were efficiently removed from solutions with initial concentrations of 100 and 300 µg Cr(VI)/L, at a relatively low salt concentration in the receiver equal to 100 mM NaCl. As a result of the anion exchange, chromium(VI) concentration was reduced to the value of 14 and 45 µg Cr(VI)/L, respectively, that is, below the admissible value for potable water (50 µg Cr(VI)/L). Mono-anion-selective membrane Neosepta ACS allowed to achieve satisfactory results of dichromate ion removal from the solution with a concentration of 100 µg Cr(VI)/L – chromium(VI) concentration was reduced to the value of 13 µg Cr(VI)/L (at the salt concentration of 100 mM NaCl). It should be underlined that for Neosepta ACS dichromate ions flux was three times lower than it was for Selemion AMV (0.030 × 10⁻³ and 0.089 × 10⁻³ mol/m² h, respectively). It standed for an adequately longer time of the anion exchange process with Neosepta ACS membrane.

Keywords: Donnan dialysis; Dichromate ion; Membrane

1. Introduction

Chromium occurs in an aquatic environment most commonly in two states: trivalent chromium: Cr(III) and hexavalent chromium: Cr(VI). Chromium(III) is a microelement indispensable to life: it takes part in the metabolism of carbohydrates [1], lowers the cholesterol level and thus slows down the progress of atherosclerosis [2]. The concentration of chromium(III) in natural waters is relatively low due to the low solubility of chromium(III) compounds. Unlike chromium(III), chromium(VI) poses a threat to living organisms. It is toxic, carcinogenic, and mutagenic. Due to its mobility, it can permeate into the microbial cell and cause damage to DNA [2]. Additionally, it affects human skin, respiratory system, and internal organs (liver and kidneys) [3]. As chromium(VI) is extremely harmful, world organizations (World Health Organization, US Environmental Protection Agency) recommend that the maximum concentration of chromium(VI) in drinking water should not exceed 50 µg/L, and in industrial wastewater which enters the surface water – it should not exceed 100 µg/L [1]. According to UE guidelines, the maximum concentration of total chromium in drinking water should not exceed 50 µg/L [4].

Chromium drains into water from natural and anthropogenic sources. The concentration of chromium in surface waters is approximately 0.5 µg/L, and it can rise up to

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100 μg/L in polluted waters [2]. When water comes into contact with rocks like serpentinite, dunite, and ophiolite, the concentration of chromium(VI) in underground water may increase – the phenomenon, which was observed, for example, in Greece, Italy, Mexico, the United States, Brazil, and Japan [5,6].

However, the main source of chromium in water is anthropogenic emission. Chromium is emitted mainly as chromium(VI), and its mobility in the environment exceeds that of chromium(III) due to its superior solubility in water [2]. One of the biggest sources of chromium emissions are chromite smelters, that is, plants, at which chromium ore is processed. Another important source of chromium(VI) emissions is electroplating plants, where steel components are coated with a layer of chromium to prevent corrosion [2]. Chromium(VI) is also present in industrial wastewater generated by tanneries and dye and pigment industries [2,5].

Numerous methods of chromium removal from water have been described in literature. Activated carbon adsorption [7,8] and the use of carbon nanotubes [9,10] have been found to be very effective as well as using natural zeolites [11,12]. Very good results, especially at high concentrations of chromium, have been achieved in the precipitation process using calcium hydroxide followed by reduction of chromium(VI) to chromium(III) [11]. Another highly effective method of chromium ions removal from aqueous solutions is the ion exchange [11,13,14], which in optimal conditions achieves the rate of over 95% [13].

Membrane filtration as a method of removal of chromium from water has also attracted a lot of attention. Micellar enhanced ultrafiltration using polysulfone membranes of 10 kDa molecular weight cut-off and rhamnolipid surfactants have enabled an effective removal of 24.3%–98.7% of chromium(VI) and 99.4% of chromium(III) [15]. Nanofiltration and reverse osmosis, used for metallurgy wastewater treatment, have each proved 97.0% and 99.5% efficiency [16]. The effective removal of chromium from aqueous solutions is achieved with electromembrane processes: electrodialysis and electrodeionization. Chromium removal rate of 99% from the solution with the initial concentration of 10 mg/L [17] and 99.8% from the solution with the concentration of 100 mg/L have been reported [18].

An interesting membrane technology is Donnan dialysis with an ion exchange membrane. The process is driven by the chemical potential gradient of the solutions on both sides of the membrane. The ion exchange membrane separates two solutions of different content and concentration of particular components: a feeding solution (with a low concentration of ions) and a receiving solution (which contains an electrolyte – typically NaCl – with a high concentration of ions) (Fig. 1).

As a result of significant difference in chemical potentials, anions (here: Cl⁻) present in the receiving solution are transported through the anion exchange membrane to the feeding solution. As cations cannot flow in this same direction (they are rejected by positively charged ionic groups of the membrane), thus, to preserve the electroneutrality of both solutions, a flow of anions is forced in the opposite direction – from the feeding solution to the receiving solution [19]. Hence, noxious anions present in the treated water are replaced by neutral ions (i.e., chlorides).
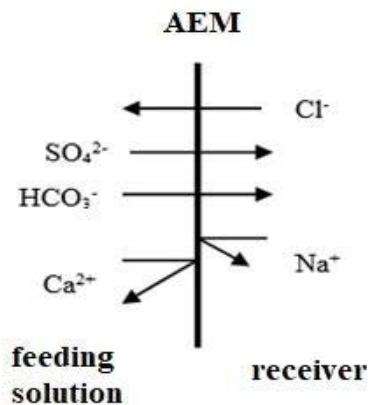

Fig. 1. The exchange of anions in Donnan dialysis process with anion-exchange membrane (AEM).

Donnan dialysis with anion exchange membrane was successfully applied to remove fluorides [20,21], nitrates [22,23], and bromates [24,25] from water so that the concentration of these ions did not exceed values permitted for drinking water. This process has also been proven useful for removing objectionable anions (sulfates and bicarbonates) from water before desalination by electrodialysis [26].

This paper presents the results of a study on chromium(VI) removal from aqueous multicomponent solutions using Donnan dialysis with anion exchange membranes. The impact of salt (NaCl) concentration in the receiving solution and the type of an anion exchange membrane on the efficiency of the anion exchange were analyzed.

2. Methods

Donnan dialysis was conducted using dialysis laboratory equipment containing 10 pairs of cells with anion exchange membranes of Selemion AMV (Asahi Glass) type or Neosepta ACS (ASTOM Corp.) type. The total working area of the membranes was 0.0756 m². Selected parameters of applied membranes are listed in Table 1. The process

Table 1
Parameters of the anion-exchange membranes used in the Donnan dialysis process [27,28]

Parameter	Membrane	
	AMV	ACS
Electric resistance (Ω cm²)	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

was conducted with the recirculation of solutions (feeding and receiving), until the concentration of chromium ions in the feeding solution reached the lowest value. Each solution flow rate was 80 L/h, and the solutions volume ratio was 10:2.5 L, respectively. The feeding solution contained $NaNO_3$, $NaHCO_3$, NaCl (with concentration of each component equal to 3 mM), and $Na_2Cr_2O_7$ (100, 300, or 500 µg Cr(VI)/L). The receiving solution was a 100, 200, or 300 mM solution of NaCl.

The concentration of anions in the feeding solution was measured during the process. The concentration of nitrate ions was determined with the spectrophotometric method using spectrophotometer DREL 2000 by HACH and standard NitraVer 5 Reagent Powder Pillows [29]. The concentration of bicarbonate and chloride ions was determined by titration using HCl and $AgNO_3$ solutions [30,31]. The concentration of chromium(VI) was determined colorimetrically with diphenylcarbazide [32]. The measurement results allowed for determining the effectiveness of chromium anion removal and the mean anions flux from the feed to the receiving solution, which was calculated with respect to the total working area of anion exchange membranes (0.0756 m²).

3. Results

3.1. Chromium(VI) removal by Donnan dialysis using Selemion AMV membrane

Fig. 2 shows a decrease in anion concentration in the feeding solution and the removal effectiveness for the initial chromium concentration equal to 100 µg Cr(VI)/L and NaCl concentration in the receiving solution equal to 100 mM. It was observed that an effective dichromate ion $(Cr_2O_7^{2-})$ to chloride ion exchange (85.7%) occurred even with the lowest NaCl concentration in the receiving solution (100 mM NaCl). As a result, the chromium(VI) concentration in the aqueous solution was lowered to 14 µg Cr(VI)/L. It is worth noting that the final concentration of chromium(VI) was much lower than actually accepted for drinking water (50 µ/L). Along with dichromate ions, 64.7% of nitrate ions and 39.3% of bicarbonate ions were also removed. The reason for a relatively low effectiveness of

HCO_3^- ion exchange to Cl⁻ ion (and hence the low effectiveness of their removal from the aqueous solution) was the size of the anion; the radius of the hydrated hydrocarbon ion is 0.394 nm and it is significantly bigger than the radius of NO_3^- ion (0.335 nm) [33].

The discussed data show that the presence of typical anions in the treated water (even at the concentration three orders of magnitude higher than that of dichromate ions) did not significantly affect the effectiveness of $Cr_2O_7^{2-}$ ion exchange to Cl⁻ ion.

The increase in salt concentration in the receiving solution caused only a slight rise in the effectiveness of the exchange of dichromate ions. With the salt concentration equal to 200 mM NaCl, chromium(VI) concentration was lowered to 12 µg Cr(VI)/L, and with salt concentration equal to 300 mM NaCl – the minimum chromium(VI) concentration was 11 µg Cr(VI)/L. It corresponded to the effectiveness of ion removal with rates of 88.1% and 89.0%, respectively (Fig. 3).

It can be noted (Fig. 3) that with a relatively low increase in the effectiveness of dichromate ions removal from water, the exchange of nitrates and bicarbonates for chloride ions was effectively risen. It resulted of the boosted driving force of Donnan dialysis: the rise in NaCl concentration caused the driving ion (i.e., chloride) concentration gradient increased and their flow into the feeding solution was enhanced. In the counterbalance, an equivalent flux of anions in the opposite direction (from the feed to the receiving solution) was formed. As the $Cr_2O_7^{2-}$ ions ratio in the total anion charge in the solution was insignificant (0.02%), the flow of the main ion components of the treated water: nitrates and bicarbonates to the receiving solution increased.

A similar relationship was found for other studied concentrations of chromium(VI) (300 and 500 µg Cr(VI)/L). Figs. 4 and 5 show the course of anion exchange with the salt concentration in the receiving solution equal to 100 mM NaCl. The anion exchange resulted in lowering the chromium(VI) concentration to 45 and 66 µg Cr(VI)/L, respectively.

It should be noted that in case of high chromium(VI) concentration (300 and 500 µg Cr(VI)/L), the rise in salt concentration in the receiving solution did not really improve the effectiveness of dichromate ion exchange to chloride
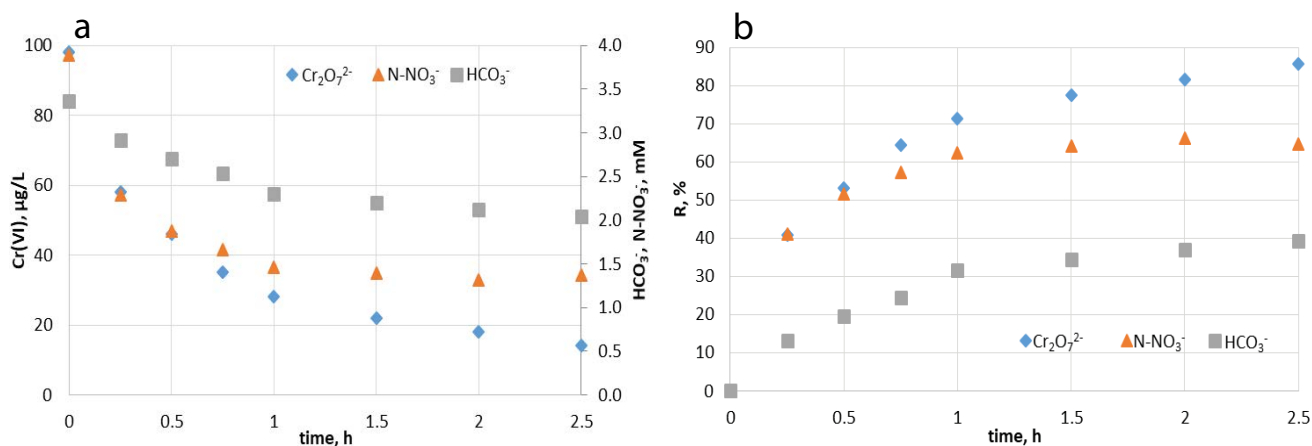

Fig. 2. The decrease in concentration of anions (a) and the effectiveness of anions removal (b) by Donnan dialysis (Selemion AMV membrane, C_{Cr} = 100 µg Cr(VI)/L, C_{NaCl} = 100 mM).
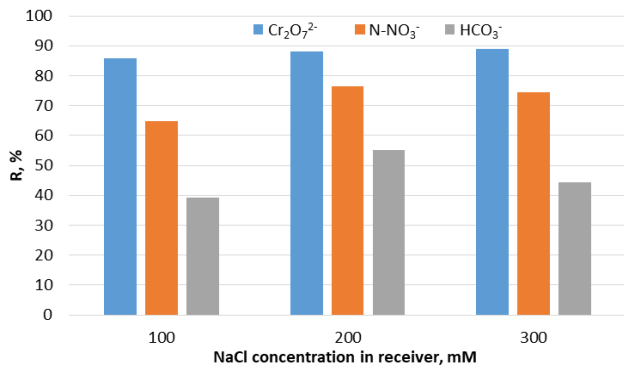
Fig. 3. The effect of salt concentration in the receiving solution on the effectiveness of anions removal from water (Selemion AMV membrane, C_{Cr} = 100 μg Cr(VI)/L).

ion; however, the effectiveness of accompanying anions (nitrates and bicarbonates) exchange to chlorides markedly rose (Figs. 6 and 7). It means that in case of high chromium(VI) concentration (here: 500 μg Cr(VI)/L), the

attainable effectiveness of dichromate removal did not allow to reduce its level to an acceptable one for drinking water.

3.2. Chromium(VI) removal by Donnan dialysis using Neosepta ACS membrane

The objective of this stage of study was to test the transport properties of the mono-anion-selective Neosepta ACS membrane toward dichromate ions in the presence of nitrates and bicarbonates. The specific structure of the membrane (due to the thin, cross-linked surface layer of the membrane) makes the flow of big anions (bicarbonates and sulfates) difficult. Simultaneously, the limitation of the flux of bigger anions from the feed to the receiving solution may cause the rise in the flux of other anions which are smaller in size [24]. Fig. 8 shows the decrease in the anion concentration in the feeding solution and the effectiveness of their removal at the initial chromium(VI) concentration equal to 100 μg Cr(VI)/L.

The cross-linked surface structure of the Neosepta ACS membrane markedly slowed down the transport of anions. As a result, the obtained values of the flux of anions removed from the feeding solution were relatively low. The mean
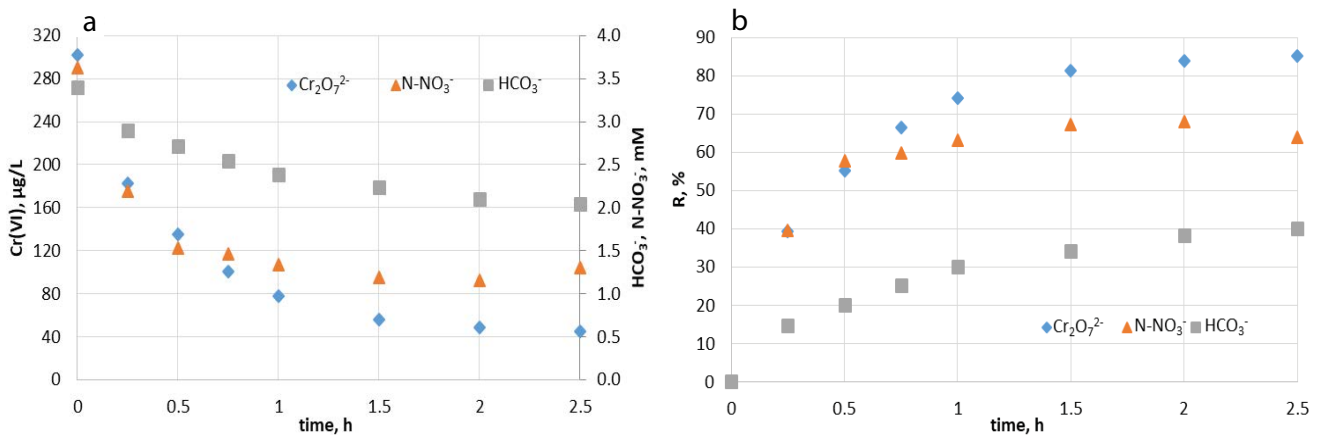
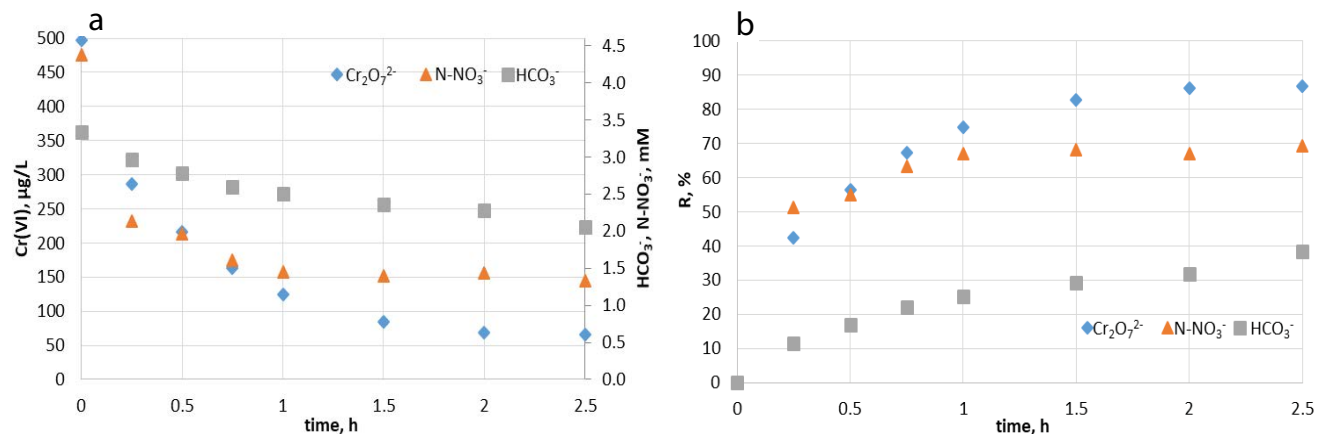

Fig. 4. The decrease in concentration of anions (a) and the effectiveness of anions removal (b) by Donnan dialysis (Selemion AMV membrane, C_{Cr} = 300 μg Cr(VI)/L, C_{NaCl} = 100 mM).

Fig. 5. The decrease in concentration of anions (a) and the effectiveness of anions removal (b) by Donnan dialysis (Selemion AMV membrane, C_{Cr} = 500 μg Cr(VI)/L, C_{NaCl} = 100 mM).
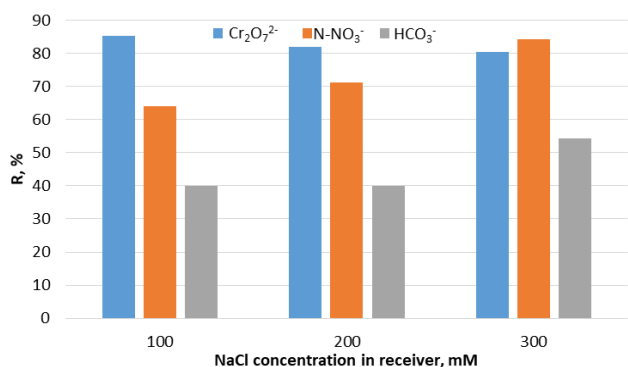
Fig. 6. The effect of the salt concentration in the receiving solution on the effectiveness of anions removal from water (Selemion AMV membrane, C_{Cr} = 300 µg Cr(VI)/L).
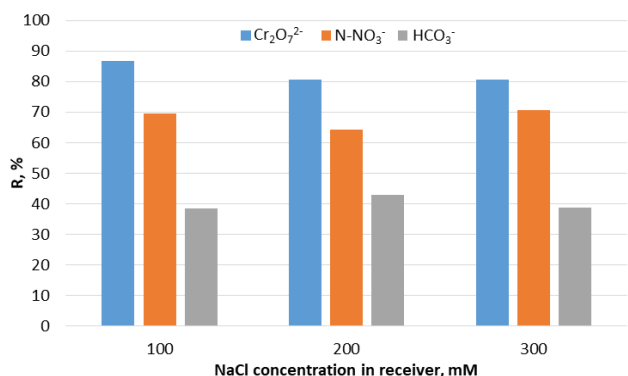

Fig. 7. The effect of the salt concentration in the receiving solution on the effectiveness of anions removal from water (Selemion AMV membrane, C_{Cr} = 500 µg Cr(VI)/L).

flux of nitrates was 0.046 mol/m² h and the effectiveness of their exchange to chloride ions was 60.8%. The mean flux of bicarbonates was 0.032 mol/m² h and the effectiveness of their removal was 30.5%. It can be noted that the mean values of nitrates flux and bicarbonates flux for Neosepta

ACS membrane were three times lower as compared with Selemion AMV membrane (Table 2). It corresponded to a longer time of anion exchange with Neosepta ACS membrane.

The cross-linked surface structure of the Neosepta ACS membrane also limited the removal of dichromate ions from the solution. The mean flux of these ions was $0.030 × 10^{-3}$ mol/m² h, and the removal rate was equal to 87.4%. However, it should be stressed that despite the slow passage of anions through the Neosepta ACS membrane (as compared with anion passage through the Selemion AMV membrane), the concentration of chromium(VI) in the feeding solution decreased to 13 µg Cr(VI)/L (i.e., much below the value acceptable in drinking water) at the salt concentration in the receiving solution equal to 100 mM NaCl.

A similar course of anion exchange was seen for other chromium(VI) concentrations in the feeding solutions (300 and 500 µg Cr(VI)/L). Figs. 9 and 10 show the decrease in concentration of anions and the effectiveness of their removal at the salt concentration in the receiving solution equal to 100 mM NaCl. As a result of the anion exchange, chromium(VI) concentration dropped to 81 and 124 µg Cr(VI)/L, respectively. These values, however, exceeded the acceptable level of chromium concentration in drinking water.

It was observed that with the Neosepta ACS membrane, the salt concentration in the receiving solution did not markedly improve the effectiveness of dichromate and accompanying (nitrate and bicarbonate) ions removal from the feeding solution (Fig. 11). It resulted of the significant resistance to anions flow through the cross-linked surface layer of the Neosepta ACS membrane.

Table 2
Mean ions fluxes through Selemion AMV and Neosepta ACS membranes (C_{Cr} = 100 µg Cr(VI)/L, C_{NaCl} = 100 mM)

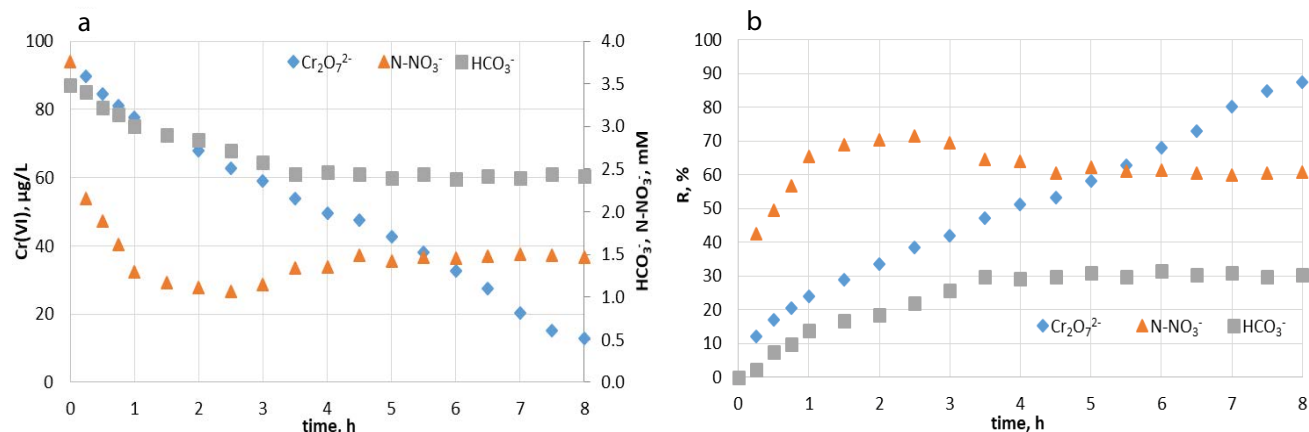| Membrane	Mean ion flux (mol/m² h)		
	$Cr_2O_7^{2-}$	NO_3^-	HCO_3^-
Selemion AMV	$0.089 × 10^{-3}$	0.143	0.085
Neosepta ACS	$0.030 × 10^{-3}$	0.046	0.032

Fig. 8. The decrease in concentration of anions (a) and the effectiveness of anions removal (b) by Donnan dialysis using Neosepta ACS membrane, C_{Cr} = 100 µg Cr(VI)/L, C_{NaCl} = 100 mM.
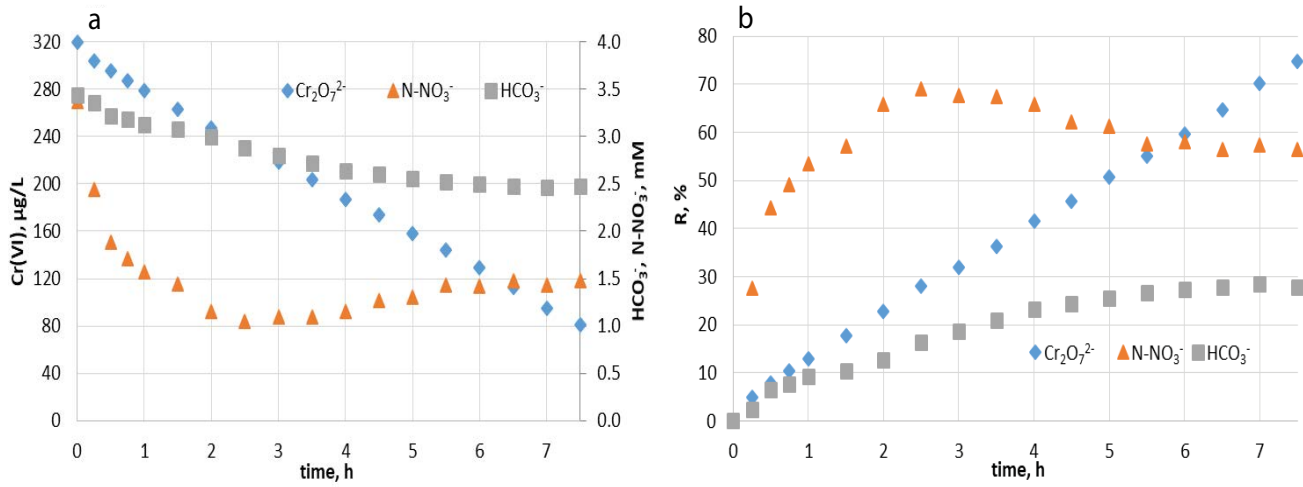
Fig. 9. The decrease in concentration of anions (a) and the effectiveness of anions removal (b) by Donnan dialysis (Neosepta ACS membrane, C_{Cr} = 300 µg Cr(VI)/L, C_{NaCl} = 100 mM).
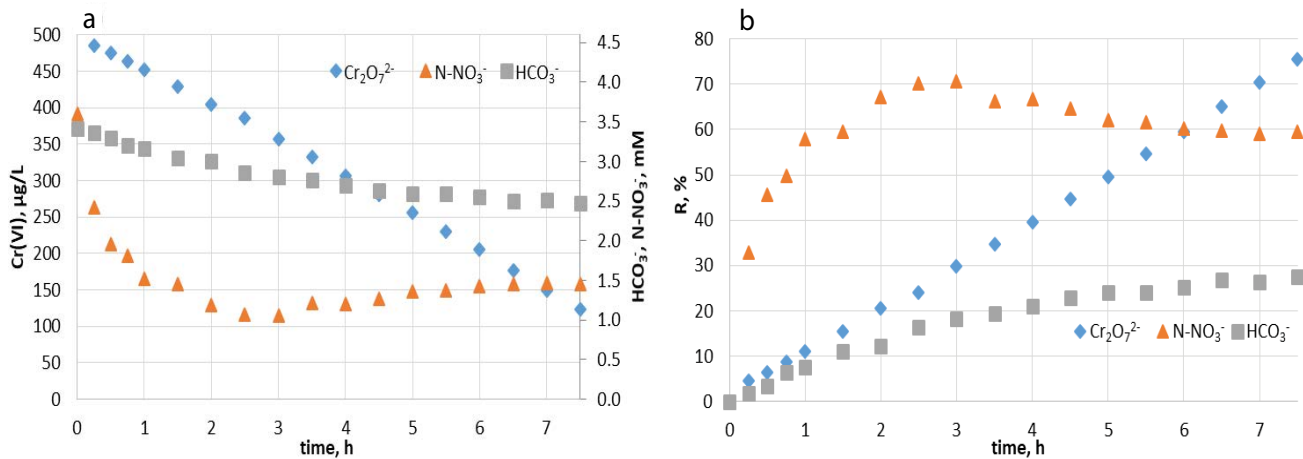

Fig. 10. The decrease in concentration of anions (a) and the effectiveness of anions removal (b) by Donnan dialysis (Neosepta ACS membrane, C_{Cr} = 500 µg Cr(VI)/L, C_{NaCl} = 100 mM).
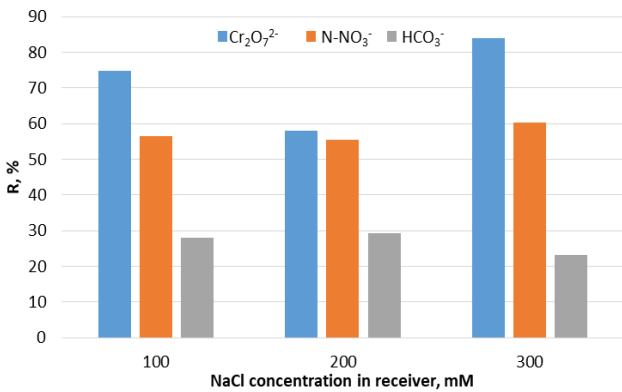

Fig. 11. The influence of the salt concentration in the receiving solution on the effectiveness of anions removal from water (Neosepta ACS membrane, C_{Cr} = 300 µg Cr(VI)/L).

Fig. 12 represents the comparison of Selemion AMV and Neosepta ACS membranes in terms of the pace and effectiveness of dichromate ion removal from aqueous solution containing 100 µg Cr(VI)/L. The data show that both membranes enabled almost an equally effective $Cr_2O_7^{2-}$ ion removal (85.7% and 87.4% for Selemion AMV membrane and Neosepta ACS membrane, respectively). It corresponds to a possibility of the decreasing of the concentration of those ions below the value acceptable in drinking water (14 and 13 µg Cr(VI)/L, respectively). A clear difference, though, could be seen in how quickly dichromate ions were removed from the treated water: $Cr_2O_7^{2-}$ ion flux through the AMV membrane was three times higher than through the ACS membrane, which corresponded to a respectively shorter time of the process (2.5 h against 8 h). It unambiguously pointed the superior transport properties of Selemion AMV membrane in regard to dichromate ion removal from water.
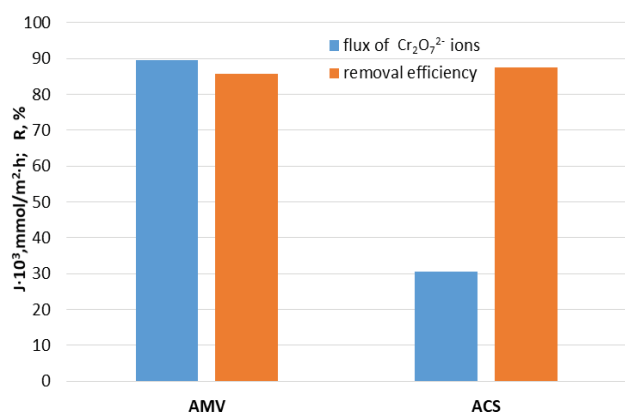
Fig. 12. The comparison of the effectiveness of dichromate ion removal from water using different anion-exchange membranes (C_{Cr} = 100 µg Cr(VI)/L, C_{NaCl} = 100 mM).

4. Conclusions

- Donnan dialysis using anion-exchange membrane allowed for an effective removal of chromium(VI) ($Cr_2O_7^{2-}$ ions) from water through the exchange of dichromate ions to nonobjectionable chloride ions.
- The best results of anion exchange were obtained using the Selemion AMV membrane. The effectiveness of dichromate ion removal from solutions of 100 and 300 µg Cr(VI)/L concentration at the salt concentration in the receiving solution equal to 100 mM NaCl was satisfactory. As a result of the anion exchange, chromium(VI) concentration dropped below the value acceptable for drinking water (i.e., below 50 µg Cr(VI)/L).
- In Donnan dialysis using Selemion AMV membrane, the rise in salt concentration in the receiving solution (up to 200 and 300 mM NaCl) merely improved the effectiveness of dichromate ion removal from water; however, it increased the effectiveness of accompanying anions (i.e., nitrates and bicarbonates) removal.
- Mono-anion-selective Neosepta ACS membrane enabled a satisfactory removal of dichromate ions only from the feeding solution with concentration of 100 µg Cr(VI)/L. Moreover, due to the increased resistance to anion transport through the membrane, the $Cr_2O_7^{2-}$ ion flux was three times lower than the flux through the Selemion AMV membrane at the same conditions; it corresponds to a longer time of anion exchange required for Neosepta ACS membrane.

Acknowledgment

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