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THE REMOVAL OF INORGANIC TRACE CONTAMINANTS BY ELECTRODIALYSIS IN REMOTE AUSTRALIAN COMMUNITIES

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Abstract: Water provision for developing countries is a critical issue as a vast number of lives are lost annually due to lack of access to safe drinking water. The presence and fate of inorganic trace contaminants is of particular concern. Trace inorganic contaminants have been found in elevated concentrations in drinking waters supplied directly from brackish groundwaters in developing countries. Desalination and the removal of trace inorganic contaminants from bore water sources from a remote community in Australia using electrodialysis (ED) were investigated. The influence of applied voltage on the removal of the trace contaminants was evaluated. The results from this study demonstrated that electrodialysis is an effectual method for the removal of total dissolved solids and a number of trace inorganic contaminants from brackish groundwaters to below drinking water guideline levels. The deposition of trace contaminants on the membranes influenced the electrodialysis process in relation to ionic flux and the effectiveness of trace contaminant removal.

Keywords: Electrodialysis, brackish groundwater, inorganic trace contaminant removal, desalination.

Introduction

Water provision in remote communities is a serious problem globally, as a vast number of lives are lost annually due to lack of access to potable drinking water [1]. In Australia, approximately 67% of Indigenous communities with a population less than 100 people use bore water for their supply [2]. In these remote communities, drinking water is generally sourced from groundwater bores of varying quality, with salinity and hardness being widespread issues. Health issues related to salinity range from dehydration, as a result of reduced water consumption, to kidney dysfunction and hypertension [3, 4]. While there are no drinking water guideline values for calcium (Ca^{2+}) and magnesium (Mg^{2+}), high levels of hardness can reduce the palatability of water and also cause water distribution pipes to block. High salt concentrations and hardness are however not the only issues facing remote communities.

The occurrence and fate of inorganic trace contaminants in surface, brackish and groundwaters is also of considerable concern with regards to public health and the environment, especially in developing countries and remote communities. While a number of compounds occur natural within groundwaters, the concentration of these compounds at elevated levels and the consumption of drinking water supplied from contaminated groundwater can lead to significant health issues. Bromide (Br^-) is naturally present in raw water, especially in groundwater and surface water in coastal regions, as bromide salts or as organic bromine substances which are mostly associated with seawater evaporates and salt lakes [5]. Bromide is generally considered non-toxic at concentrations found in most drinking water sources; however, when water containing high levels of bromide are treated by ozonation, bromate species are formed [6], which are highly toxic for human health. For example, in developing countries such as Morocco, fluoride (F^-) concentrations up to 20 mg/L have been found in groundwaters [7, 8], while the maximum acceptable concentration level is 1.5 mg/L [1]. The detrimental effects of excess concentrations of fluoride on teeth and the skeletal system (dental and skeletal fluorosis) is prevalent [9]. Strontium (Sr^{2+}) minerals are widely distributed throughout the earth and are released to the groundwater by the weathering of rocks and soils. High concentrations of strontium in groundwater of up to 53 mg/L have been reported in certain parts of Denmark. [10].

Strontium is relatively low toxic but continued exposure may have adverse health effects. Studies have however have shown that the ingestion of higher levels of strontium in drinking water prenatally may have a protective effect against caries [11]. There is also no guideline value established for strontium. Zinc (Zn^{2+}) concentrations are usually low in drinking water, compared to the WHO guideline value of 3 mg/l [1]. Significant anthropogenic influences have led to increases in the bioavailability of zinc in the environment. Ingestion of enhanced zinc can cause memory impairments and copper deficiencies [12]. Elevated concentrations of zinc have also been linked to Alzheimer's disease [13].

The importance of membrane processes, such as electrodialysis (ED), for treatment of drinking water is a continuously growing field. ED has been used for a wide range of applications including desalination, the production of potable water from brackish water [14, 15], seawater [16, 17] and industrial wastewater [18, 19] and numerous other applications. The fouling of ion-exchange membranes is the main problem of electromembrane technologies. High levels of calcium and magnesium hardness and the formation of divalent and trivalent ion hydroxides [20] can lead to membrane fouling. The build-up of a scalant layer on the ion-exchange membranes leads to an increase in cell resistance, which in turn reduces the effectiveness of the ED process and leads to a decrease in membrane integrity [21].

The aim of this study is the evaluation of ED for desalination and the removal of inorganic trace contaminants from a brackish groundwater sourced from a remote Australian community. The focus of this study was the influence of applied voltage and solution pH (results not presented for solution pH).

Materials and Methods

Electrodialysis System Characteristics

The ED cell used was a BEL-500 unit (Berghof, Germany) with seven Neosepta CMX-SB cation-exchange membranes (CEMs) and six Neosepta AMX-SB anion-exchange membranes (AEMs) (supplied by Eurodia, Germany; manufactured by Tokuyama Soda Ltd., Tokyo, Japan) providing for each an available membrane area of 58 cm². The ED cell was connected to a DC electric potential (GW Instek DC Power supply Model GPR-1810HD, Taiwan) through TiO₂-coated titanium electrodes. The layout of the ED system used in the experiments is shown in Figure 1. The setup consisted of three separate solution tanks for the diluate (4L), for the concentrate (4L) and for the electrode rinse (2L), which were recirculated by three peristaltic pumps.

Figure 1. Electrodes dialysis system layout with pump control, power supply, peristaltic pumps, ED stack, and three containers for concentrate, diluate and electrode rinse

Bore Water Location and Characteristics

The ED experiments were performed on a bore water (bore number RN13693) sourced from Pine Hill Station, a cattle farm approximately ~140 km northwest of Alice Springs, Northern Territory, Australia. This bore was chosen for this study due to its poor water quality in regards to salinity, hardness and trace contaminant content. Chemical analysis results of the bore water are shown in Table 1.

Table 1. Water quality at Pine Hill Station in comparison with World health Organization (WHO) 2006 and Australian Drinking Water Quality Guidelines (ADWG) 2004

PARAMETER	UNIT	SAMPLE VALUE	ADWG [22]	WHO [1]
Bromide	mg/L	10.584	-	-
Calcium	mg/L	104.56	-	-
Chloride	mg/L	2000	250 ^a	-
Copper	mg/L	0.017	2 (1 ^a)	2
Fluoride	mg/L	5.82	1.5	1.5
Magnesium	mg/L	172	-	-
Potassium	mg/L	35.36	-	-
Sodium	mg/L	1650	180 ^a	-
Strontium	mg/L	2.281	-	-
Sulphate	mg/L	717	500 ^a	-
Uranium	mg/L	0.261	0.02	-
Zinc	mg/L	0.222	3 ^a	-
TDS	mg/L	5300 ^{b,c}	500 ^a	-
pH	-	8.4	6.5-8.5 ^a	-
Electrical conductivity (EC)	μS/cm	8290 ^c	-	-

^a Aesthetic guideline value. ^b Calculated using TDS = 0.064 EC [23]. ^c Average of feed conductivity throughout field testing at Pine Hill Station [24].

Experimental Protocol

To determine the influence of ED on the removal of trace contaminants from the Pine Hill bore water and the influence of applied voltage, experiments were conducted at two voltages: 12 and 18V. The ED system was operated in continuous operation mode during each experiment, where the feed water (initially filled in both diluate and concentrate solution tanks) was recirculated throughout the concentrate and diluate until the desired product was achieved. As a result of this, the ED experiments were not all run for the same time period. An applied electric potential of 10V was fixed for all experiments. Both the diluate and concentrate were fed into the ED stack at a flow rate of 3 L/min. An electrode rinse solution of 0.5 mol/L Na₂SO₄ was fed through the ED stack at a flow rate of 2.5 L/min, in order to prevent the generation of chlorine or hypochlorite which could be hazardous for the electrodes. After each experiment, the diluate and concentrate stream were mixed and returned to the system as feed (diluate and concentrate) for the next experiment. EC was converted into total

dissolved solids (TDS, mg/L) using a conversion factor $k = 0.64$ for Australian groundwater with high sodium content [23]. It is important to note however that this conversion is dependent on feed water characteristics.

Analytical Methods

Sampling was undertaken periodically from the recirculation line in the diluate and concentrate solution tanks during each experiment. pH, electrical conductivity and temperature of samples taken from the diluate and concentrate were measured using a pH/Conductivity meter (Multiline P4 epoxy gel combination pH electrode, automatic temperature compensation immersion probe, WTW). Anion analysis was undertaken using a DX-500 ion chromatograph (DIONEX, Sunnyvale, CA, USA). Separation was achieved using an IonPac AS4A-5C analytical column and a mobile phase composed of 1.7 mM sodium bicarbonate/1.8 mM sodium carbonate delivered at a flow rate of 2 mL/min. Post-column eluent suppression was achieved using an anion self-regenerating suppressor (ASRS-1) operated at a current of 50 mA. Detection was via an ED40 electrochemical detector set at an output range of 30 μ S. Instrument control and data collection were accomplished using PeakNet software. Cation analysis was performed with Inductively-coupled Plasma Optical Emission Spectroscopy (ICP-OES) (Perkin Elmer Optima 5300 DV) and Inductively-coupled Plasma Atomic Mass Spectroscopy (ICP-MS) (Agilent 7500ce).

The cell resistance across the ED stack was calculated using Ohms law ($I = E/R$, whereby R is the resistance (Ohm), E is the electrical potential (Volt) and I is the electrical current (Amperes)), from the voltage and current read from the DC power supply. Ion concentration removal (%) from the diluate was calculated from using $(C_{Dt=0} - C_{Dt=n})/C_{Dt=0} \times 100\%$ ($C_{Dt=0}$ is the initial ion concentration, $C_{Dt=n}$ is the ion concentration in sample taken at time t (min)). Flux of ions from the diluate into the concentrate were calculated using $(C_{Ct=n} - C_{Ct=n-10})/(A_m \times t)$ ($(C_{Ct=n} - C_{Ct=n-10})$ is the concentration increase within the concentrate between sample measurements, A_m is the membrane area used in ion transport). A mass balance of all contaminants, including NOM, within the ED system was calculated using $M_F = M_D + M_C + M_{ads/dep}$ (M_F feed mass, M_D diluate mass, M_C concentrate mass, $M_{ads/dep}$ adsorbed/deposited mass).

Results and Discussion

Electrodialytic parameters and ED performance

The cell resistance ($I = E/R$) across the ED stack as a function of time and voltage is shown in Figure 2A. The cell resistance is relatively stable in the beginning of the experiment followed by a significant increase towards the final stages of the experiment. This is especially noticeable in the experiment with an applied voltage of 12V. This increase as occurs as a result of the depletion of ions within the diluate solution circulating through the ED cell and the resultant lowering of the current (amperes). An increase in cell resistance can also be attributed to fouling of the ion-exchange membranes, which will be discussed later. The final cell resistance achieved in experiments with an applied voltage of 18V is lower than that achieved in experiments with an applied voltage of 12V, as a result of deeper demineralization of the diluate stream. This can be seen in Figure 2B, where the concentration of total dissolved solids (TDS, mg/L) within both the diluate and concentrate are shown. While the experiments with an applied voltage of 12V was run longer than the 18V experiment and a removal of 97.3% (110 min) TDS was achieved in comparison to 94.9% (70 min), when considering the percentage of TDS removal at the same final value the TDS removal in the 18V experiment was greater than that achieved in the 12V experiment (80.5%). This is due to the higher applied voltage and the resultant increase in the rate of TDS removal. In both experiments the ADWG value of 500 mg/L TDS was reached. In regards to the opportunity for reuse of the ED concentrate stream for other applications, for example household reuse and/or for stock water, the possibility depends on the

quality of the feed water to be treated and the resultant TDS concentration. Both the diluate and concentrate could be used to maintain condition of sheep. Only the diluate would be suitable to maintain condition for beef cattle. When considering the tolerance for healthy growth of the sheep (TDS 6400 mg/L) only the diluate would be suitable for this purpose [25]. In regards to healthy growth for beef cattle (TDS 4288 mg/L), the diluate would be suitable after approximately 10 minutes.

A mass balance of TDS (g) in the ED system is illustrated in Figure 2C. As can be seen the amount of TDS deposited on the membrane was higher in the 12V experiments (9881mg) than in the 18V experiment (6968mg). This is only due to the longer duration of the 12V experiment. If the 18V experiment had been allowed to run for the same time period as the 12V experiment, the final mass loss of TDS would have been greater. Generally, at each time period, the TDS mass deposited is greater in the 18V experiment. While some of this loss can be attributed to transport into the electrode rinse, the majority of this would be due to deposition on the ion-exchange membranes. The negatively-charged trace contaminants within the diluate solution, for example F^- , would be deposited on the anion-exchange membranes and the positively-charged contaminants, for example Ca^{2+} , deposited on the cation-exchange membranes. Figure 2D shows the flux of TDS (mg/m^2h) from the diluate into the concentrate as a function of time. TDS flux (mg/m^2h) was greater in the 18V experiment in the beginning stages of the ED process in comparison to the TDS flux in the 12V experiment. After 40 min the TDS flux in the 18V experiment is seen to be lower than in the 12V experiment. This is a result of the deposition of contaminants on the ion-exchange membranes and the subsequent reduction in the transport of ions through the membranes. Further results on the flux of individual trace contaminants will be discussed later.

Figure 2. ED cell resistance, Total dissolved solids (mg/L), Total dissolved solids loss/membrane deposit (mg) and Total dissolved solids flux (mg/m^2h) as a function of time and applied voltage. Also plotted is the ADWG (B bottom: 500 mg/L) and the maximum TDS for using the diluate and/or concentrate for beef cattle stock (B middle: 5632 mg/L, 8800 $\mu S/cm$) and sheep (B top: 13 952 mg/L, 21 800 $\mu S/cm$) in maintaining condition [25]

Trace contaminant removal

The removal of trace contaminants (anions and cations) from the Pine Hill bore water is shown in Figure 3. Complete removal (100%) of the anions Br^- , F^- and NO_3^- was achieved in the 12V experiment (Figure 3A) compared to 55.8%, 68.1% and 62.3% in the 18V experiment (Figure 3B). This difference can be explained by the longer duration of the 12V experiment. However, when considering the removal of these anions at 70 min in the 12V experiment (the same time duration as the 18V experiment), the percentage removed is still greater with 86.9-100%, 62.1-88%, 80.6-81.5% removal achieved for Br^- , F^- and NO_3^- , respectively. The transport of ions in ED is influenced by the concentration of the competitive ions within solution, whereby transport is increased if the concentration of competitive ions within solution is reduced. While the TDS concentration in the 12V experiment was 1108 mg/L at 70 min, the concentration in the 18V experiment was 291 mg/L. Therefore, theoretically the transport of ions in the 18V experiment should have been enhanced in comparison with the 12V experiment. However, the amount of TDS deposited on the ion-exchange membranes during the 18V experiment was significantly higher, possibly hampering the transport of the anions through the anion-exchange membranes and into the concentrate. Results on the mass of trace contaminants deposited on the ion-exchange membranes and the flux of the trace contaminants through the ion-exchange membranes is outlined in the following section. The percentage removal achieved for Cl^- and SO_4^{2-} ions in the 12V experiment was 98.2% (95.8% at 70 min) and 92.0% (52.7% at 70 min) respectively compared with 66.8% and 55.% in the 18V experiment. Again, the reduced percentage removal of Cl^- ions in the 18V experiment could be related to greater mass of contaminants deposited on the membranes. The removal of SO_4^{2-} ions was relatively similar in both experiments. Results from this study showed that ED was successful in removing Cl^- , F^- and SO_4^{2-} ions to concentrations below the ADWG values [22]. As mentioned previously, complete removal of F^- ions was achieved in the 12V experiment compared to the guideline value of 1.5 mg/L. The final concentration of Cl^- and SO_4^{2-} ions in the diluate were 46.6 mg/L and 74.6 mg/L, respectively, compared to the aesthetic guideline value of 250 mg/L and 500 mg/L. The final concentration of SO_4^{2-} ions in the 18V experiment (365.2%) was also below the guideline value.

Figure 3. Removal of trace contaminants from the Pine Hill Bore water as a function of time and voltage

The removal of cations from the Pine Hill bore water is shown in Figure 3C (12V) and Figure 3D (18V). These results showed similar trends in removal as with the anions. Complete removal (100%) of Mg^{2+} and Sr^- ions was achieved in the 12V experiment compared with 66.9% and 81.5% in the 18V experiment, respectively. The ED process was successful in the removal of Sr^- ions as a result of the relatively low initial concentration in the bore water (2.281 mg/L). Complete removal of Sr^- ions in the 12V experiment was reached after 80 min. The removal of Ca^{2+} (75.6%) and K^+ (63.3%) were also lower in the 18V experiment than in the 12V experiment (Ca^{2+} 98.4%, K^+ 89.4%). The removal of Ca^{2+} and K^+ at 70 min in the 12V experiment was 90.6-95.9% and 76.4-94.7%, correspondingly. While no guideline values have been reported for Ca^{2+} and K^+ , the final concentrations in the diluate in the 12V experiment were greatly reduced from their initial concentration in the bore water (Table 1) with 1.9 mg/L for Ca^{2+} and 2.9 mg/L for K^+ . In the 18V experiment the final concentrations in the diluate were 24.2 mg/L and 15.9 mg/L, respectively.

Overall, Figure 3 shows different kinetics for the anions and cations, with the removal of cations being significantly greater in the beginning stages of the ED process compared with a more steady increase in the removal of anions as a function of time. This difference in kinetics may be the result of different ionic characteristics of which further work on explaining these differences in removal trends is currently underway. Extensive results on the removal of trace contaminants as a function of solution pH and the performance of the ED system are also in preparation for publication.

Trace contaminant loss/membrane deposition

Figure 4 shows that the mass of trace contaminants deposited on the ion-exchange membranes increases as a function of time. Understandably, the mass of ions deposited on the membranes is directly related to both the initial concentration within the bore water and also the applied voltage. The mass of ions deposited on the membranes was higher in the 18V experiment than in the 12V experiment due to the higher initial flux of ions through the ion-exchange membranes. The mass of K^+ ions deposited on the membrane in the 18V experiment (154 mg) was considerably higher than in the 12V experiment (7.1-8.5 mg). The mass of Cl^- (12V-70 min: 1868 mg, 18V: 4339 mg) and SO_4^{2-} (12V-70 min: 679 mg; 18V: 878 mg) ions deposited on the membrane is considerably greater than the other contaminants due to the high initial concentration within the Pine Hill bore water. This is also the case for Ca^{2+} (12V-70 min 332-514 mg; 18V 406 mg) and Mg^{2+} (12V-70 min: 294-375 mg; 18V: 406 mg) ions. This membrane deposit would form a fouling layer (scaling) at the membrane surface. Scaling is the precipitation of crystalline divalent and trivalent ion hydroxides such as CaCO_3 , MgCO_3 , $\text{Mg}(\text{OH})_2$ and CaSO_4 on the concentrate of the cation-exchange, and to a lesser extent on the anion-exchange membranes [26]. In ED this scaling manifests itself as an increase in the electrical resistance, a decrease in the selectivity of the ion-exchange membrane and a decline in the flux of ions [21]; which was demonstrated in the results from this study.

Figure 4. Ion loss/membrane deposit as a function of time and applied voltage

Conclusions

The results from this study demonstrated the performance of an electrodialysis system in the treatment of groundwater of high salinity and trace contaminant concentration from an isolated community in Central Australia. The treated water can be used to provide the community with potable drinking water as well as the provision of water for stock purposes. In regards to the removal of total dissolved solids, electrodialysis was successful in treating the bore water to below drinking water quality guideline levels. The applied voltage had an influence on the removal of the trace contaminants from the bore water, with 12V overall being the most effective. Results also demonstrated that electrodialysis was effective in reducing the concentration of the contaminants chloride, fluoride, sodium and sulphate to below guideline levels. The deposition of trace contaminants on the ion-exchange membranes had an influence on the overall cell resistance throughout the experiments with a significant increase occurring in the later stages of the electrodialysis process. This increase in cell resistance was due to the formation of a fouling layer on the membranes. This fouling layer in turn influenced the flux of total dissolved solids and trace contaminants from the diluate into the concentrate. This would have an influence on the practical applicability of electrodialysis in the treatment of waters from remote communities in developing regions.

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