

Review on Removal of Ionic Impurities (Arsenic, Iron, Fluorides and Nitrates) Using Electrodialysis

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Abstract—One of the major problems in the 21st Century is that of water scarcity. With only less than 2% of total water available on earth's surface as fresh water and the rate at which population is growing, the amount of available freshwater won't suffice for the demand of potable water in near future. Thus, it is becoming a challenge for scientists and engineers to develop technologies which can solve the problem of water scarcity. Electrodialysis has proved to be an effective technique to remove the ionic impurities from the water with more than 90% removal. The concentration of the ionic impurities can be brought down below the permissible limit as recommended by the WHO by adjusting the operating parameters like pH, flow rate, initial concentration, voltage, time and by using an appropriate selective membrane. Unlike other techniques, electrodialysis is simple to use, has fewer pretreatment costs, doesn't use any chemicals, has a high purity of the product, is robust in nature and doesn't form any sludge at the end of the process and hence is also environment-friendly. This short review focuses on how electrodialysis is emerging out as a solution to meet potable water demand in future and presents the experimental work of various researchers on the recovery of potable water from the contaminated water containing toxic elements such as iron, fluoride, arsenic and nitrates using electrodialysis.

Keywords: Electrodialysis, potable water, membranes, arsenic, iron, nitrates, fluoride.

1. INTRODUCTION

Electrodialysis is one of the separation techniques for removal of ionic impurities from an electrolytic solution. In electrodialysis, we have an array of Ion exchange membranes namely cation exchange membranes (CEM) and anion exchange membranes (AEM) placed in alternate fashion between two electrodes: cathode (negatively charged electrode) and anode (positively charged electrode). These Ion Exchange Membranes act as a barrier and allow only selective ions to pass. The driving force for migration of ions in electrodialysis is electric field or electric potential. When an electric field is applied between two electrodes, the positively charged ions (cations) start migrating towards cathode through cation exchange membranes and are blocked by anion exchange membranes. Similarly, negatively charged ions

(anions) migrate towards anode through anion exchange membranes and are blocked by cation exchange membranes. Consequently, ionic migration and selective permeability of membranes give rise to alternate ions rich and solvent rich compartments called concentrate and diluate, respectively. Schematic diagram has been shown in the Fig.1 [1].

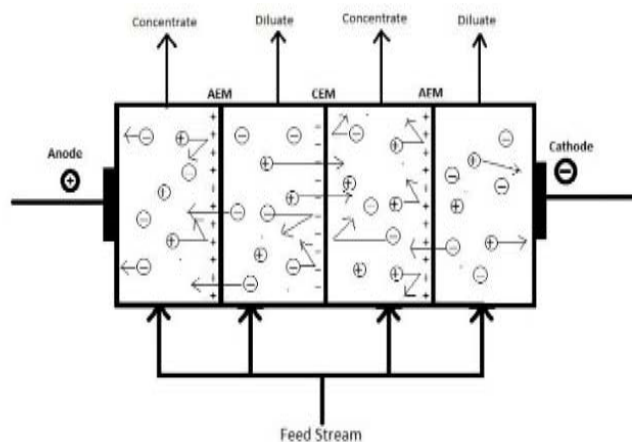


Figure 1: Schematic Diagram of electrodialysis Ref(1)

This technique is applied to remove various ionic impurities from the water to solve the problem of water scarcity which is one of the major problems that humankind is facing in the 21st century. Population explosion around the world has drastically increased the demand for drinking water which has resulted in water shortage in most parts of the world, especially in arid and semi-arid regions [2]. By the year 2030, it is projected that the global needs of water would increase to 6900 billion m³ from the current 4500 billion m³ [3]. So, about a 53% increase in the amount of drinking water, by the year 2030. To summarize, the present water resources will be unable to meet the drinking water demand in the future [4]. Thus there is a paramount need to harness the maximum possible potential of the membrane technologies to meet the ever-increasing drinking water demand. Various treatment methods have been used based on adsorption [5-7], ion-exchange [8-10], precipitation [11], electrocoagulation, oxidation filtration,

bioremediation[12] and membrane processes such as nanofiltration, reverse osmosis[13] and electrodialysis have been employed for the purpose of contaminants removal. High initial cost, lack of selectivity, low capacity, formation of slug and complicated or expensive regeneration are some of the drawbacks of most of these methods. Electrodialysis and reverse osmosis are the only two membrane technologies that have reached the practical application. These two processes are employed for the removal of salt from brackish water and hence are called desalination processes[14-15]. RO (Reverse Osmosis) and ED (Electrodialysis) are used in the integrated system to produce more water than in individual processes with a low amount in rejected water which is also low in salinity[4]. The main advantage of ED over other separation processes is that it doesn't use any chemicals for operation[16].

2. ELECTRODIALYSIS FOR THEREMOVAL OF ARSENIC CONTAMINANTS FROM DRINKING WATER

Arsenic is an abundant element found in earth's crust and in the sea water. It mainly occurs in organic and inorganic forms and as metalloid. In groundwater, it occurs in two oxidation states and these are As(III)[H_3AsO_3 , H_2AsO_3^- , HAsO_2^-] and As(V)[H_3AsO_4 , H_2AsO_4^- , HAsO_4^{2-} and AsO_4^{3-}][17]. Inorganic arsenic species are more toxic than organic ones[18] and arsenite As(III) is about 60 times more toxic than arsenate As(V)[19]. Arsenic, if present in drinking water, can cause stomach pain, diarrhoea, vomiting, partial paralysis and blindness [20]. It is also responsible for causing various types of cancers [21-22]. The maximum allowable arsenic concentration in the drinking water as recommended by the WHO is 0.01 ppm[23]. According to Singh et al. [24], the natural contamination of groundwater with Arsenic has reached up to 5 ppm and at different places in the world, it is 48 ppm by the human sources.

Electrodialysis is emerging as a major technique for the removal of arsenic from the groundwater. Ribeiro et al. [25] used electrodialysis for the removal of arsenic from sawdust in a batch set-up. They report 99% removal of arsenic contaminants from the water by this technique. The ED cell used contained three compartments having side compartments as anode and cathode, respectively, and the middle compartment was filled with sawdust. Sawdust and electrode compartments were separated by the ion exchange membranes having anion exchange membrane on the anode side and the cation exchange membrane on the cathode side, respectively. In the electrode compartments, 10^{-2} M solution NaNO_3 was used as the electrolyte. The electrodes used were platinized titanium bars. The current used was 0.2 mA/cm^2 and the time duration was 30 days. The authors[25] extracted 26.7, 98.7, 96.6 and 92.2% arsenic from sawdust by using distilled water, 2.5% oxalic acid, 5.0% oxalic acid and 7.5% oxalic acid, respectively. Another author [26] reports the removal of arsenic down to 0.01 ppm which is the WHO[23]

recommended concentration but uses resins in integration with the ionic membrane for the separation.

3. ELECTRODIALYSIS FOR THEREMOVAL OF NITRATES FROM DRINKING WATER

High nitrate concentration in the drinking water sources is a public health concern. The increasing concentration is caused by the industrial and agricultural wastes and due to the large utilization of artificial nitrogenous fertilizers. This contamination has caused the shutdown of wells and rendered many aquifers unusable. The permissible level of nitrates in the drinking water as suggested by the WHO[27] is 50 ppm. In some regions of the world, the contaminations have been reported as exceeding 250 ppm[28, 29]. High nitrate contamination can be fatal to the infants under 6 months of age. When the nitrate rich water is consumed by infants, NO_3^- is reduced to NO_2^- in their bodies which combines with haemoglobin in the blood to form methemoglobin, and leads to a condition commonly known as "blue baby syndrome"[27-30].

The electrodialysis process is more simple to conductive in comparison with the conventional denitrification processes such as biological denitration, chemical denitration and catalytic denitration [30]. Economically, the operating cost of ED is not high. El Midaoui et al. [31] have reported the removal of nitrates and other ions using electrodialysis. The electrodialysis stack was equipped with a Neosepta anion exchange membrane(ACS) and Neosepta cation exchange membrane(CMX) manufactured by Tokuyama Co and the batch set-up was employed. The electrodialysis was carried out with a nitrate selective membrane to avoid demineralisation which is not desired. The electrode compartments were separated from the others to prevent any change in the composition of the concentration sides by possible electrode reactions. The polarity and the flow were reversed, periodically, to prevent any fouling and scaling. The concentration of each ion was determined analytically. Increase in the removal rate was observed with increase in the temperature due to the increase in the ion mobility and dilation of membrane network which promotes the membrane swelling and diffusion of ions into the membrane. For all temperatures the different anions are removed in the following order $\text{NO}_3^- > \text{Cl}^- > \text{HCO}_3^- > \text{SO}_4^{2-}$. Sulphate removal kinetics were found to be very slow at lower temperatures. At 15°C and after 10 min only 20% of sulphate was removed which demonstrated the controlled transport of bivalent ions at lower temperatures. It was also observed that removal rates of NO_3^- and Cl^- increase rapidly with time at various voltages. Bicarbonate and sulphate removal rate was slow at lower voltage (5V) and increased rapidly at higher voltage (15V). Another research[32] selected the best anionic membrane for the removal of nitrate among five types of cation exchange membrane used. The current density used for the selection process was 10 mA/cm^2 and evolution of three parameters i.e., specific ion rejection, overall rejection and voltage on both

ends of the stack, was followed. The criteria for the selection of the best membrane corresponded to the minimum variation of the TDS, maximum nitrate removal and the minimum energy cost. The best results were obtained with the ACS membrane. Sulphate rejection and the overall rejection was found to be lower and the nitrate rejection was higher. The value of the various electrodialysis parameters were: temperature 20°C, voltage 15V, flow rate 180 l/h. After 10 min electrodialysis, it was found, 90.7% of NO_3^- , 88.5% of Cl^- , 48.3% of HCO_3^- and only 7.5% of SO_4^{2-} was removed. The nitrate content obtained in the output water was under the recommended level[23].

4. ELECTRODIALYSIS FOR THE REMOVAL OF IRON FROM DRINKING WATER

Iron is one of the most abundant metals on the earth. It occurs naturally in fresh water as well as groundwater in various forms such as Fe^{2+} , Fe^{3+} , along with the complex forms. Iron is an essential element in human nutrition and daily requirements of iron depend on sex, age, physiological status and iron bioavailability [23]. Excess of iron in drinking water gives rise to different aesthetic problems like taste, discolouration, turbidity, staining of laundry, odour, clogging etc., [12]. Iron also promotes the growth of various chlorine tolerant microorganisms which increases the cost of cleaning and sterilization and pose a health problem to humans[33]. As per WHO guidelines permissible concentration of iron in groundwater is 0.3 ppm and a concentration range of 1-3 ppm is acceptable for the people taking anaerobic well water [23]. High concentration of iron in the human body cause serious problems like damage to the liver, heart and endocrine glands[34]. Consequently, groundwater needs to be properly treated for the removal of iron from it before using it domestically. Various researches have been carried out on the removal of iron using electrodialysis. Chekioua, A. & Delimi, R[35] successfully separated iron from pickling bath of sulphuric acid with a percentage removal of 70.17% using electrodialysis. They treated the contaminated sulphuric acid solution in a four compartment ED cell using a Neosepta anion exchange membrane (AMX) and CMX membranes to separate iron(II) from solution. The test solution was introduced in compartment while anodic and cathodic compartments were filled with 0.1N H_2SO_4 and 0.1N HNO_3 solutions, respectively. Their studies showed that iron removal increased with increasing current density from 1 mA/cm^2 to 20 mA/cm^2 then decreased by further increase in the current density. Initial concentration has a positive effect on iron removal and 70.17% iron removal was observed at 52000 ppm of initial concentration. Among the different membranes tested namely Selemion cation exchange membrane (CMV), Nafion 117 and CMX, CMX proved to be the most effective. Mourad Ben Sik Ali [36] concluded that iron can be removed from brackish water using electrodialysis under various process parameters (pH, flow rate, initial concentration, ionic strength, applied voltage). They studied the removal of iron

from synthetic brackish water using six compartment electrodialysis cell consisting of Pt/Ir coated titanium electrodes and three cation exchange membranes (PCA-SK) and two anion exchange membranes (PCA-SA). Spacers were introduced between membranes to reduce boundary layer effects. A solution of Sodium chloride and iron chloride was used as a test solution and sodium sulphate solution was used as electrode solution to prevent chlorine effect on electrodes. Their study showed the effect of various parameters (applied voltage, pH, initial concentration, ionic strength, flow rate) on demineralization rate, iron removal rate, current efficiency and transport flux [36]. Cifuentes, L. et al. [37] showed the effectiveness of electrodialysis in the removal of iron from solutions analogous to those found in Copper electrowinning operations for water recovery ($\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$ and $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$). They conducted the experiment on five compartment electrodialysis cell consisting of 304 stainless steel cathode and lead anode. Two cation exchange membranes (MC3470) and two anion exchange membranes (MA3475) were used in the experiment. The test solution ($\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$ and $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$) was introduced in the middle compartment, cathode and anode compartments were filled with sulphuric acid of concentration 54000ppm. The effect of cell current density, recirculation flow rate and time were studied by means of 4hr and 24hr runs. Their results showed that iron removal increased with increasing current density and in going from laminar to turbulent flow regime. They also showed that 99.5% removal of iron was achievable for the 24hr run test with specific power consumption of 1 kW/kg of water. Thus, ED proved to be an effective way of water recovery from acidic electrolytes containing Cu and Fe.

5. ELECTRODIALYSIS FOR THE REMOVAL OF FLUORINE FROM DRINKING WATER

Fluorides in drinking water as a result of human activities and natural processes is a major health concern worldwide. Consequently, removal of fluorides from water is an urgent necessity. The permissible limit of fluoride ion concentration, according to WHO[23], in drinking water is 1.5 ppm. High concentration of fluorine in drinking water causes fluorosis and bone lesions. Concentration in the range of 1.5-4 ppm is responsible for cancer, liver deterioration and Alzheimer disease. Intake of 3-5 g sodium fluoride is fatal for adults [38-39]. On the other hand, shortage of fluorine may result in dental caries and osteoporosis[38].

Electrodialysis is a relatively simple technique with fewer disadvantages compared to conventional methods for the fluoride removal present in the water. Ergun et al. [40] used anion exchange membrane from Gelman Sciences (SB-6407) to study the fluoride removal via electrodialysis. They studied the effect of current density on the removal efficiency and found that the increase of current density increased the amount of fluoride separated. They had taken the sample with initial fluoride content equal to 20.6 ppm and were successful in bringing it down to 0.8 ppm which is lower than permissible

level as recommended by the WHO. Researchers[41] used two commercial membranes, ACS and Neosepta anion exchange membrane (AXE 01), and he found that ACS membranes furnished the better results. The fluoride content was brought down to 0.5 ppm from 1.8 ppm and TDS decreased from 1127 ppm to 656 ppm. Keri et al. [42] studied the effect of various parameters (pH, applied potential, initial fluoride concentration, and flow rate) on the fluoride removal by ED. Results obtained were sufficient for the fluoride concentration lower than 10 ppm with removal efficiency equal to 99.6% and the final fluoride concentration in the water lower than 1 ppm. For the experiments performed with fluoride concentrations higher than 10 ppm, the results were moderate with the removal of 95.95% fluorides for 20 ppm and 79.22% for 50 ppm. The final diluates, however, did not fulfil the WHO standards. Another author[43] performed the research on ED for fluoride removal using anion membranes from Asahi Glass Co. (AMP) and photopolymer membranes MZA. The AMP membranes removed 97% and 69% of fluoride ions by using the current density of 7mA/cm^2 and 1mA/cm^2 , respectively. MZA removed only 40% of fluoride ions when the current density had the same values.

6. CONCLUSION

Electrodialysis has proved to be an effective technique for the removal of iron, fluoride, arsenic, nitrates (with more than 90% removal) to make potable water. By using appropriate selective membranes and adjusting operating parameters (pH, flow rate, initial concentration, voltage, time) potable water can be recovered which has the concentration of above-mentioned elements under permissible limit as prescribed by WHO. Unlike other techniques, electrodialysis is simple to use, has fewer pretreatment costs, doesn't use any chemicals, has high purity of product and is robust in nature. Also, electrodialysis doesn't form any sludge material at the end of the process which may, in turn, harm the environment unlike other separation techniques such as bio-separation, nanofiltration, flocculation etc. However, electrodialysis has not been yet commercialized by industries for making potable water because of high initial costs & more energy consumption.

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