

# USE OF THE ELECTRODIALYSIS PROCESS FOR FLUORIDE ION AND SALT REMOVAL FROM MULTI-CONSTITUENT AQUEOUS SOLUTIONS

Martyna GRZEGORZEK <sup>a\*</sup>, Katarzyna MAJEWSKA-NOWAK <sup>b</sup>

<sup>a</sup> MSc Eng.; Wrocław University of Science and Technology, Faculty of Environmental Engineering, Wybrzeże Wyspiańskiego 27, 50-370 Wrocław

\*E-mail address: [martyna.grzegorzek@pwr.edu.pl](mailto:martyna.grzegorzek@pwr.edu.pl)

<sup>b</sup> Prof.; Wrocław University of Science and Technology, Faculty of Environmental Engineering, Wybrzeże Wyspiańskiego 27, 50-370 Wrocław

Received: 1.04.2016; Revised: 29.04.2016; Accepted: 5.07.2016

## Abstract

Fluorine is a common chemical element. According to WHO guidelines, the  $F^-$  ion content in drinking water cannot be higher than  $1.5 \text{ mg/dm}^3$ . Excess of fluorine leads to many health problems: Alzheimer's disease, neurological disorders or fluorosis (dental or skeletal). Fluoride can be removed from aqueous solutions by means of various methods (adsorption, precipitation, ion-exchange or membrane techniques). The aim of this paper was to evaluate the efficiency of electrodialysis in fluoride removal under the presence of organic substances. During experiments solutions containing fluorides ( $5, 10, 100$  and  $200 \text{ mg F}^-/\text{dm}^3$ ), mineral salt ( $0.5 \text{ g NaCl}/\text{dm}^3$ ) and organic matter ( $5, 10$  and  $15 \text{ mg/dm}^3$  of humic acids) were used. The research was conducted with the use of the PC Cell BED-1 System. The current density was equal to  $1.72 \text{ mA/cm}^2$ . It has been shown that electrodialysis is an efficient technique of fluoride removal provided that the initial concentration of  $F^-$  ions is not higher than  $10 \text{ mg F}^-/\text{dm}^3$ . The impact of organic matter on the process run and efficiency was dependent on the fluoride content in the treated solution.

## Streszczenie

Fluor jest powszechnie występującym pierwiastkiem chemicznym. Zgodnie z wytycznymi WHO zawartość fluorków w wodzie do picia nie może być większa niż  $1.5 \text{ mg/dm}^3$ . Nadmiar fluoru prowadzi do licznych problemów zdrowotnych (Alzheimer, problemy neurologiczne, fluoroza zębów lub szkieletu). Fluorki mogą być usunięte z roztworów wodnych z wykorzystaniem różnych metod (adsorpcja, strącanie, wymiana jonowa czy procesy membranowe). Celem pracy była ocena efektywności elektrodializy w usuwaniu jonów fluorkowych w obecności substancji organicznych. W trakcie doświadczeń zostały wykorzystane roztwory zawierające fluorki ( $5, 10, 100$  i  $200 \text{ mg F}^-/\text{dm}^3$ ), sól mineralną ( $0.5 \text{ g NaCl}/\text{dm}^3$ ) oraz substancje organiczne ( $5, 10$  i  $15 \text{ mg/dm}^3$  kwasów humusowych). Doświadczenia zostały przeprowadzone z wykorzystaniem instalacji PCCell BED-1 System. Gęstość prądu wynosiła  $1.72 \text{ mA/cm}^2$ . Wykazano, że w procesie elektrodializy fluorki są skutecznie usuwane pod warunkiem, że ich początkowe stężenie nie przekracza  $10 \text{ mg F}^-/\text{dm}^3$ . Wpływ substancji organicznych na przebieg i efektywność procesu zależał od stężenia fluorków w oczyszczanym roztworze.

Keywords: Water Treatment; Electrodialysis; Fluoride.

## 1. INTRODUCTION

Fluorine is a chemical element with the symbol F. Many fluorine compounds are very water-soluble. In aqueous environments it only occurs as fluoride. The

content of  $F^-$  ions in natural surface water is usually not higher than  $0.3 \text{ mg/dm}^3$  [1, 2]. According to WHO guidelines the permissible concentration in drinking water cannot be higher than  $1.5 \text{ mg/dm}^3$ . There are two sources of fluoride in the environment – natural

and anthropogenic. Fluorine is used in glass, fertilizers, semiconductors, phosphates, ceramics, the production of cosmetics and in the food industry. The production of pesticides also generates waste containing a large amount of fluorine in organic and inorganic form. Fluorine commonly occurs in the Earth's crust (625 mg/kg). Fluorine is present in minerals like fluorspar, cryolite, sellaite, basalt, topaz, granite, syenite and fluoroapatite or hydroxyapatite. As the result of mineral dissolution, fluoride is released to the environment. The dissolution process can be affected by many factors such as residence time, groundwater age, well depth or rock chemistry. Fluorine also migrates to the natural environment as a result of volcanic activity [3-7]. In small amounts fluoride has a beneficial influence on health – it protects teeth against decay and also has a positive effect on bone mineralization [6]. It is important to maintain the permissible level of fluoride content in drinking water. Excessive concentrations lead to neurological disorders, cancer, gastrointestinal problems, Alzheimer's disease and fluorosis (skeleton or dental). Fluorine also causes lesions of the thyroid, liver and endocrine glands [2, 4, 8, 9]. Fish, water plants and algae are also very vulnerable to fluoride toxic action [8]. Excessive concentrations of F<sup>-</sup> ions in natural water have been observed in China, India, Brazil, Argentina, Chile, Pakistan and in northeastern regions of Africa. For example, the fluoride content in lakes in the East African Rift Valley is about 2800 mg/dm<sup>3</sup>. In Kenya, F<sup>-</sup> concentration is around 180 mg/dm<sup>3</sup>, in China – 10 mg/dm<sup>3</sup> and in Tanzania – 250 mg/dm<sup>3</sup> [4]. F<sup>-</sup> content in wastewater from phosphoric acid production can achieve a value of 3000 mg/dm<sup>3</sup> [10].

There are many methods that allow fluoride to be removed from aqueous solutions: chemical precipitation, ion exchange, adsorption and membrane techniques (reverse osmosis, nanofiltration) [2, 4, 8, 11]. Fluoride removal efficiency is affected by many factors: pH, type of adsorbent, membrane characteristics, temperature or coexisting ions. Most of the conventional methods have disadvantages such as the generation of toxic sludge, the requirement of pretreatment, high costs of waste disposal. For example, after the ion-exchange process reagents used for resin regeneration will be released to the environment [6, 12].

Dev Brahman et al. [13] applied biosorption for simultaneous fluoride and arsenic removal. As an adsorbent, sawdust of the plant *Tecomella undulate* was used. Tests were conducted on the real water samples from different sites in Mithi (Pakistan).

Fluoride concentration decreased from 42.5 to 12 mg F<sup>-</sup>/dm<sup>3</sup>, which is unacceptable for drinking water.

Nasr et al. [14] also used adsorption for fluoride removal from water solutions. Calcite was applied as an adsorbent under the presence of acetic acid. It has been proved that a dosing of 0.1 mol/dm<sup>3</sup> of acetic acid brought about a significant improvement of fluoride removal (from 17.4 to 30.4% without and with acetic acid respectively, with an initial fluoride content of 5 mg F<sup>-</sup>/dm<sup>3</sup>). The observed beneficial effect of acetic acid on process efficiency was attributed to the increase of the surface area available for adsorption on calcite particles. However, a further increase of acetic acid concentration (to 0.4 mol/dm<sup>3</sup>) only resulted in a 38.2% reduction of fluoride content, which did not comply with WHO standards (< 1.5 mg F<sup>-</sup>/dm<sup>3</sup>).

Membrane processes, including dialytic techniques, are characterized by high separation efficiency and can be potentially attractive methods for removing undesirable ionic pollutants or concentrating valuable compounds [9, 11, 15]. The driving force in these techniques involves concentration gradient (dialysis, DD) or electrical potential gradient (electrodialysis, ED). Due to the rather slow kinetics of the DD process, electrodialysis seems to be more beneficial for natural water treatment. During the ED process ions migrate in the constant electric field. They are transported through the ion-exchange membranes. Cations migrate to the cathode and are able to pass through the cation-exchange membranes but they cannot be transported through the anion-exchange membranes. The opposite situation occurs in the case of anions. As a result, two streams are formed – diluate and concentrate (brine) [15, 16]. ED is known as a simple and environmentally friendly technique. A lack of waste solids and a low demand for chemicals are crucial advantages [17].

Boubakri et al. [11] conducted experiments with the use of Donnan dialysis (DD) for fluoride removal. During the research, strongly basic anion-exchange AM3 membranes (Tokuyama Soda) were applied. The initial fluoride concentration was equal to 5, 10 and 15 mg F<sup>-</sup>/dm<sup>3</sup>. The lowest removal efficiency (34.14%) was observed when F<sup>-</sup> concentration amounted to 15 mg F<sup>-</sup>/dm<sup>3</sup> and therefore the final water quality after the DD process was above the standard for drinking water. In the case of lower fluoride content (10 mg F<sup>-</sup>/dm<sup>3</sup>), the removal efficiency reached 56.48% and the final water quality also exceeded WHO guidelines.

Although there are undeniable advantages of the ED method in the treatment of aqueous solutions containing ionic compounds, only some research has been performed on fluoride ion removal. Moreover, most of the reported studies deal with the impact of mineral species on fluoride separation [9,18,19]. Ergun et al. [9] used electrodialysis (ED) for fluoride removal from natural water containing  $20.6 \text{ mg F}^-/\text{dm}^3$ . The strong-basic anion-exchange membranes (SB-6407, Gelman Science) were used during the ED experiments. By applying a current density of  $8.48 \text{ mA}/\text{cm}^2$ , it was possible to diminish  $\text{F}^-$  concentration to  $0.8 \text{ mg F}^-/\text{dm}^3$  (a value significantly lower than WHO guidelines –  $1.5 \text{ mg F}^-/\text{dm}^3$ ). The authors also reported that chloride and sulphate ions significantly deteriorated the rate of fluoride transport through anion-exchange membranes. On the contrary, Kabay et al. [18] proved that sulphates had no impact on fluoride removal in the course of the ED process and only chlorides influenced  $\text{F}^-$  separation efficiency. A study [19] conducted in a pilot ED installation revealed a satisfactory removal of fluoride ions from solutions containing less than  $10 \text{ mg F}^-/\text{dm}^3$  – the reduction of  $\text{F}^-$  ions reached 99.6%. The increased initial concentration of fluorides, i.e.  $20\text{-}50 \text{ mg F}^-/\text{dm}^3$  arisen to moderate  $\text{F}^-$  ion removal (79.22-95.95%) and diluates did not meet WHO standards.

Taking into account the above reported results on fluoride removal by ED it can be concluded that there are still some gaps in recognizing the mechanism of  $\text{F}^-$  ion separation, especially from multi-component systems involving not only mineral species, but organic substances as well.

Organic matter (OM) is a common constituent of natural water. Humic substances are the main constituents of natural organic matter, which are typically present in surface water in an amount of  $0.1\text{-}20 \text{ mg}/\text{dm}^3$  [20]. Dissolved organic substances (e.g. humic substances) can interact with water contaminants (also with fluoride ions) and form soluble or insoluble complexes [21]. On the other hand, humic substances (especially humic acid) are known as a serious foulant in electromembrane processes [22]. Consequently, humic acid can affect both inorganic ion transport through the ion exchange membranes and also membrane resistance. Due to the health concern of fluoride content in drinking water, it is reasonable to evaluate the impact of OM on fluoride removal by ED.

The aim of this paper was to evaluate the influence of organic matter on fluoride removal efficiency by batch electrodialysis. Aqueous salt solutions contain-

ing variable amounts of fluoride and humic acid were subjected to treatment by electrodialysis. The energy needed for ion transport through ion-exchange membranes was also estimated.

## 2. MATERIALS AND METHODS

In the course of the ED experiments the PC-Cell BED-1 System (PCCell GmbH, Germany) equipped with classic ion-exchange membranes (PC-SA and PC-SK, PCA GmbH, Germany) was used. The membrane stack contained 11 cation-exchange membranes and 10 anion-exchange membranes. The active surface area of each membrane was equal to  $64 \text{ cm}^2$ . The volume of the diluate and concentrate chambers amounted to  $2 \text{ dm}^3$  (each). Sodium chloride ( $0.1 \text{ mol}/\text{dm}^3$ ) was used as the electrode solution.

The ED experiments were conducted at a constant electrical current (0.11 A) and the current density was equal to  $1.72 \text{ mA}/\text{cm}^2$ . The applied current density was chosen in such a way as not to exceed the limiting current density. The limiting current density was evaluated theoretically on the basis of the Rauntenbach et al. [16] approach, with an assumption that the final salt concentration in the diluate cells should not exceed 15% of the initial salt concentration. The calculated limiting current density (for the initial salt concentration  $0.5 \text{ g}/\text{dm}^3$ ) was equal to  $2.46 \text{ mA}/\text{cm}^2$  at 85% desalination efficiency for the ED process with standard PC-SA and PC-SK membranes. The process was terminated when the voltage value was equal to 24 V.

Feed solutions containing 5, 10, 100 and  $200 \text{ mg F}^-/\text{dm}^3$  were prepared with the use of sodium fluoride (NaF) and distilled water. To increase the salinity, sodium chloride ( $\text{NaCl}$ ,  $0.5 \text{ g}/\text{dm}^3$ ) was added to the feed solutions. Organic matter in the form of humic acid (HA) was dosed to the fluoride solutions in amounts of 5, 10 and  $15 \text{ mg HA}/\text{dm}^3$ . The solutions of HA were made of pulverised humic acid (Aldrich) and distilled water. Humic acid (HA) was used to simulate humic substances (HSs) in surface water and the composition of feed solutions was characteristic for natural water.

At the beginning of the ED process the concentrate chamber was filled with a solution consisting of fluoride and mineral salt, whereas the diluate tank contained a fluoride and salt solution supplemented with organic matter. In the course of the ED process, samples of diluate and concentrate were taken at the same time intervals. The fluoride content was measured by the colorimetric method with SPADNS

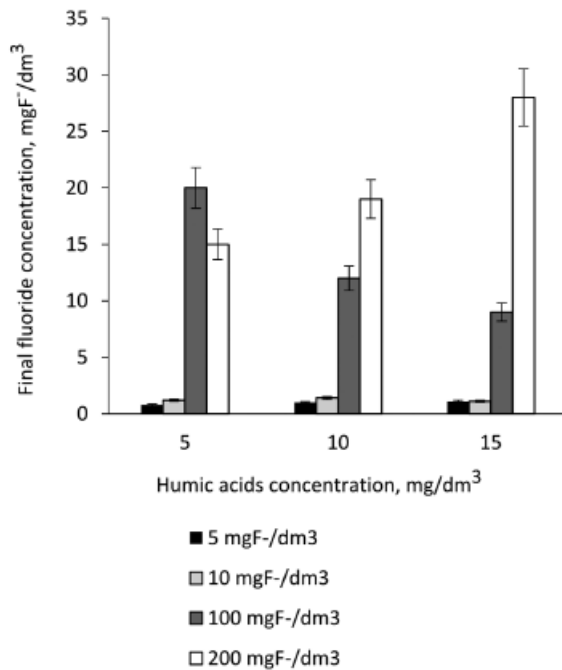


Figure 1. Final fluoride concentration in diluate versus humic acid concentration and initial fluoride content (5, 10, 100 and 200 mg F<sup>-</sup>/dm<sup>3</sup>) (error bars ±9%)

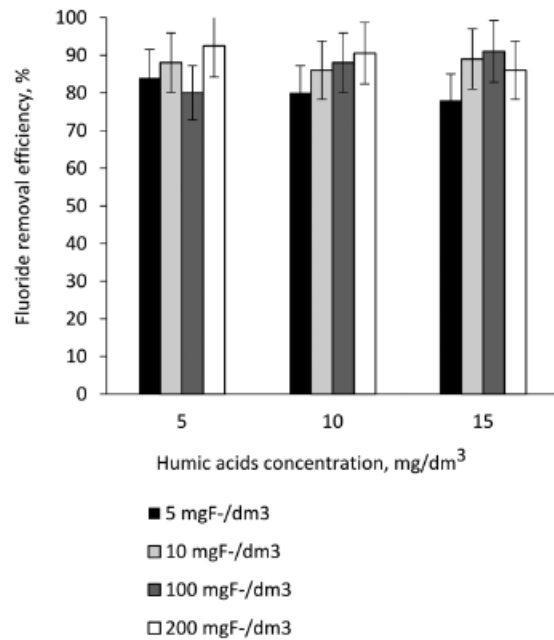


Figure 2. Fluoride removal efficiency versus concentration of humic acids and initial fluoride content (5, 10, 100 and 200 mg F<sup>-</sup>/dm<sup>3</sup>) (error bars ±9%)

reagent. This reagent contains zirconium, which creates complexes with fluoride ions. The intensity of sample color decreases when the fluoride content increases. A spectrophotometer DR 2000 (HACH) was used for fluoride analysis. The measurement error was estimated to ±9%.

The fate of salts (including NaCl) in the ED runs was monitored indirectly by electrical conductivity measurements in both the diluate and concentrate. A conductometer Elmetron CC-411 was used for this analysis. The initial conductivity of the feed solutions varied from approximately 1100 to 2300 µS/cm depending on the composition of the solution. The measurement error was estimated to ± 0.25%.

During the ED tests voltage was also monitored at equal time intervals. Voltage data allowed for the calculation of the energy consumption needed for ion transport through ion-exchange membranes according to the equation [15]:

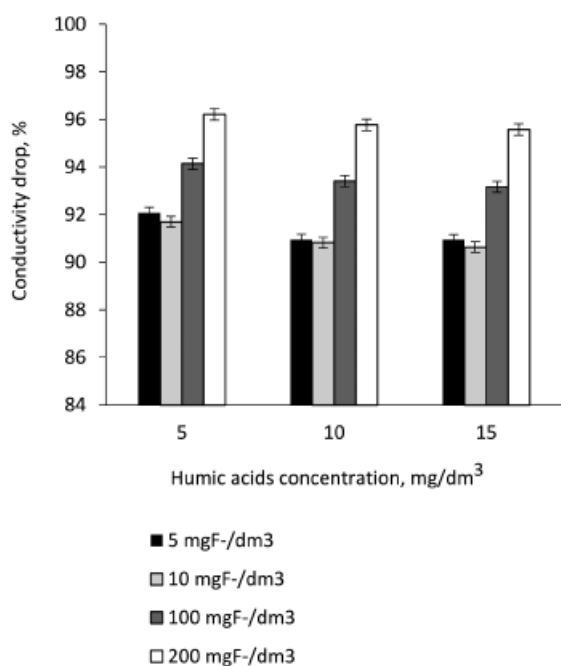
$$EC = \frac{I \sum U \Delta t}{V}, kWh/m^3 \quad (1)$$

where: I – current (A), U – voltage (V), V – volume of the diluate (m<sup>3</sup>), t – duration of the process.

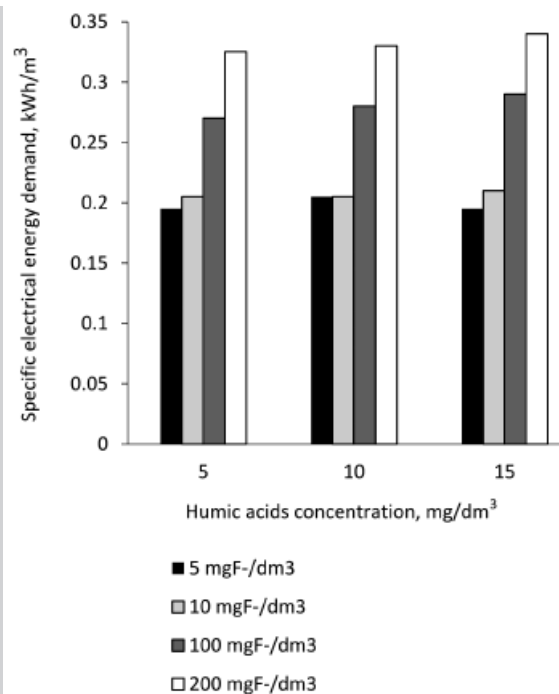
### 3. RESULTS AND DISCUSSION

The impact of humic acid presence in the feed solution on the final fluoride content in the diluate is presented in Fig. 1.

In the case of low initial fluoride content (5 and 10 mg F<sup>-</sup>/dm<sup>3</sup>), the final concentration was below the permissible limit (1.5 mg F<sup>-</sup>/dm<sup>3</sup>). When the fluoride concentration in the feed solution was high (100 and 200 mg F<sup>-</sup>/dm<sup>3</sup>), the final F<sup>-</sup> content in the diluate significantly exceeded WHO guidelines. It can be generally concluded that the presence of humic acids has no significant influence on fluoride removal efficiency for low initial F<sup>-</sup> concentration only. Some fluctuations in the final F<sup>-</sup> concentration were observed in the case of an elevated initial fluoride content – for solutions containing 100 mg F<sup>-</sup>/dm<sup>3</sup> an increasing HA concentration improved fluoride removal, whereas for solutions with 200 mg F<sup>-</sup>/dm<sup>3</sup> the opposite effect was observed. It seems that two mechanisms can govern fluoride removal under the presence of humic acids – the attachment of fluoride ions to humic acid macroparticles or the hindering of the rate of F<sup>-</sup> ions transport in the ED cells by an excessive amount of HA. Besides, dosing of HA into fluoride solutions did not have an impact on the duration of the process. The duration of the process increased with an



**Figure 3.** Conductivity drop versus humic acids concentration and initial fluoride concentration (5, 10, 100 and 200 mg F<sup>-</sup>/dm<sup>3</sup>), (error bars ± 0.25%)



**Figure 4.** Desalination energy consumption versus concentration of humic acids and initial fluoride concentration (5, 10, 100 and 200 mg F<sup>-</sup>/dm<sup>3</sup>)

increasing initial fluoride content. For 5, 10, 100 and 200 mg F<sup>-</sup>/dm<sup>3</sup>, the process was terminated after approximately 22, 22, 33.5 and 47 minutes, respectively. The lowest fluoride content (0.8 mg/dm<sup>3</sup>) was obtained for the solution containing 5 mg F<sup>-</sup>/dm<sup>3</sup> and 5 mg HA/dm<sup>3</sup>. The highest final F<sup>-</sup> content occurred in the case of 200 mg F<sup>-</sup>/dm<sup>3</sup> and 15 mg HA/dm<sup>3</sup> and was equal to 28 mg F<sup>-</sup>/dm<sup>3</sup>.

The percentage removal of fluoride for solutions varying in initial composition is presented in Fig. 2.

The lowest removal, equal to 78%, was obtained for solutions containing 5 mg F<sup>-</sup>/dm<sup>3</sup> and 15 mg HA/dm<sup>3</sup>. The highest removal efficiency (92.5%) occurred when the fluoride content was equal to 200 mg F<sup>-</sup>/dm<sup>3</sup> and humic acid concentration amounted to 5 mg HA/dm<sup>3</sup>. It was noticed that in almost all cases elimination of F<sup>-</sup> ions was above 80%, which is a rather satisfying result.

Overall, the results obtained on fluoride removal from HA solution were in agreement with data reported by other researches on F<sup>-</sup> ion separation without HA [9, 19] – by applying electrodialysis it is possible without any difficulty to reach an F<sup>-</sup> ion concentration below 1.5 mg F<sup>-</sup>/dm<sup>3</sup>, on condition that fluoride concentration in raw water is not higher than 10 mg F<sup>-</sup>/dm<sup>3</sup>. However, the impact of HA on fluo-

ride separation efficiency wasn't as noticeable as it was expected.

It is obvious that sodium chloride will be removed from diluate to concentrate cell. As it was the study was focused on fluoride transport, the salinity was monitored indirectly by conductivity measurements. The desalination efficiency is given in Fig. 3. As was expected, the percentage of mineral salt removal was very high (in the range of approximately 92-98%). It should be pointed out that the final diluate conductivity for all tested solutions was equal to approximately 110-130 μS/cm. The diluate quality (in view of salt content) was limited by the voltage increase in the ED stack (maximal 24 V).

The values of calculated energy consumption are presented in Fig. 4. Although the total energy consumption in electromembrane processes comprises of both energy needed for desalination (ion transport through ion-exchange membranes) and energy for the pumping of the circulated solution, only the desalination energy demand was involved in this study (according to equation 1). The aim of the research was to evaluate the effect of organic matter on fluoride removal and the electrodialysis run. It was assumed that the calculated desalination energy for the ED process with solutions containing various

contents of HA could be an indication of whether membrane fouling occurred or not.

Nevertheless, the energy needed to pump the concentrate and diluate flow streams through the ED stack was estimated based on the pressure drops (maximal 50 kPa) and volume flow rates (80 dm<sup>3</sup>/h) [23]. The pumping power amounted to approximately 2.0 W (which corresponds to energy consumption from about 0.3 to even 1.0 kWh/m<sup>3</sup>).

It has been shown that energy demand increased with an increasing fluoride content. The lowest EC (0.195 kWh/m<sup>3</sup>) was noticed for solutions containing 5 mg F<sup>-</sup>/dm<sup>3</sup> and 5 mg HA/dm<sup>3</sup>, as well as 15 mg HA/dm<sup>3</sup>. When fluoride content amounted to 10 mg F<sup>-</sup>/dm<sup>3</sup>, the energy demand varied from 0.205 to 0.21 kWh/m<sup>3</sup>. In the case of a raised F<sup>-</sup> concentration (100 mg F<sup>-</sup>/dm<sup>3</sup>) the EC was equal to 0.27-0.29 kWh/m<sup>3</sup>. The highest value of EC (0.34 kWh/m<sup>3</sup>) was obtained for the solution with 200 mg F<sup>-</sup>/dm<sup>3</sup> and 15 mg HA/dm<sup>3</sup>. It was noticed that the presence of organic matter influenced the desalination energy consumption insignificantly. The results obtained for different contents of HA were comparable.

Based on the obtained results of energy consumption it can be presumed that humic acid did not foul anion-exchange membranes significantly. The increase in desalination energy consumption with an increasing organic matter content was insignificant. Probably, due to a neutral charge of HA under experimental conditions (pH<7 for all feed solutions) [20], the interaction between HA macroparticles and ion-exchange membranes was weakened. This finding is a contradiction of the general view that humic substances can cause serious fouling in membrane processes [22]. It seems that fouling phenomena due to HSs depends on the chemical composition of treated water, but this supposition needs further study.

#### 4. SUMMARY

The obtained results showed that electro dialysis can be used as an effective method of fluoride removal from aqueous solutions. The content of fluoride ions in product water in the case of a moderate initial concentration of F<sup>-</sup> ions (5 and 10 mg F<sup>-</sup>/dm<sup>3</sup>) was reduced below the permissible limit (1.5 mg F<sup>-</sup>/dm<sup>3</sup>) irrespective of the concentration of organic matter. For solutions with an elevated F<sup>-</sup> ion concentration (100 and 200 mg F<sup>-</sup>/dm<sup>3</sup>) the final fluoride content was above WHO guidelines and the process efficiency was influenced by humic acid concentration. The fluoride removal efficiency varied from 78-80% for an initial concentration of 5 mg F<sup>-</sup>/dm<sup>3</sup> to 86-89% in the case of solutions containing 10 mg F<sup>-</sup>/dm<sup>3</sup>. Raised fluoride concentrations, 100 and 200 mg F<sup>-</sup>/dm<sup>3</sup>, brought about an F<sup>-</sup> ion elimination in the range of 80-91% and 86-92.5%, respectively. In the case of a moderate fluoride content (5 and 10 mg F<sup>-</sup>/dm<sup>3</sup>), removal efficiency of 80-90% was recognized as sufficient to meet drinking water standards. It was found that the energy needed for ion transport (desalination energy) increased with an increasing initial F<sup>-</sup> content in the treated solutions. The lowest energy consumption (0.195 kWh/m<sup>3</sup>) was determined for solutions containing 5 mg F<sup>-</sup>/dm<sup>3</sup> under the presence of 5 and 15 mg HA/dm<sup>3</sup>. The highest EC was noted when fluoride concentration was equal to 200 mg F<sup>-</sup>/dm<sup>3</sup> and humic acids amounted to 15 mg HA/dm<sup>3</sup>. However, the presence of organic matter in the treated solutions had only a minor impact on EC, which indicated impaired membrane fouling due to HA. The obtained results allow the conclusion that the initial fluoride concentration in treated water will have an influence on fluoride removal efficiency as well as on process duration. Electro dialysis can be recommended as a final treatment of drinking water after appropriate pretreatment of raw water.

#### ACKNOWLEDGEMENTS

The financial support of the statutory activity (Faculty of Environmental Engineering, Wrocław University of Science and Technology, grant no. S50-518) is greatly appreciated.

This article was presented in 11<sup>th</sup> Scientific Conference Membranes And Membrane Processes In Environmental Protection, 15<sup>th</sup>-18<sup>th</sup> of June 2016 Zakopane, Poland, and is printed here with their full permission.

## REFERENCES

- [1] *Agalakova N.I., Gusev G.P.*; Molecular mechanism of cytotoxicity and apoptosis induced by inorganic fluoride, *Cell Biology*, 2012, Vol.2012, p.1-16
- [2] *Arda M., Orhan E.*; Removal of fluoride from geothermal water by electrodialysis (ED), *Separation Science and Technology*, 2009, Vol.44, p.841-853
- [3] *Mohapatra M., S. Anand., Mishra B.K, Dion E. G., Singh P.*; Review of fluoride removal from drinking water, *Journal of Environmental Management*, 2009, Vol.91, p.67-77
- [4] *Shen J., Schäfer A.*; Removal of fluoride and uranium by nanofiltration and reverse osmosis: A review, *Chemosphere*, 2014, Vol.117, p.679-691
- [5] *Bhatnagar A., E. Kumar, M. Silanpaa.*; Fluoride removal from water by adsorption – A review, *Chemical Engineering Journal*, 2011, Vol.171, p.811-840
- [6] *Boubakri A., Bouchrit R., Hafiane A., Bouguecha S. A.-T.*; Fluoride removal from aqueous solution by direct contact membrane distillation: theoretical and experimental studies, *Environ. Sci. Pollut. Res.*, 2014, Vol.21, p.10493-10501
- [7] *Li Y., Zhang H. Zhang Z., Shao L., He P.*; Treatment and resource recovery from inorganic fluoride-containing waste produced by the pesticide industry, *Journal of Environmental Sciences*, 2015, Vol.31, p.21-29
- [8] *Bejaoui I., Mnif A., Hamrouni B.*; Performance of reverse osmosis and nanofiltration in the removal of fluoride from model water and metal packaging industrial effluent, *Separation Science and Technology*, 2014, Vol.49, p.1135-1145
- [9] *Ergun E., Tor A., Cengeloglu Y., Kocak I.*; Electrodialytic removal of fluoride from water: Effects of process parameters and accompanying anions, *Separation and Purification Technology*, 2008, Vol.64, p.147-153
- [10] *Ndiaye P.I., Moulin P., Dominguez L., Millet J.C., Charbit F.*; Removal of fluoride from electronic industrial effluent by RO membrane separation, *Desalination*, 2005, Vol.173, p.25-32
- [11] *Boubakri A., N. Helali, M. Tlili, M.B. Amor.*; Fluoride removal from diluted solutions by Donnan dialysis using full factorial design, *Korean J. Chem. Eng.*, 2014, Vol.31(3), p.461-466
- [12] *Hou D., Wang J., Zhao Ch., Wang B., Luan Z., Sun X.*; Fluoride removal from brackish groundwater by direct membrane distillation, *ScienceDirect*, 2010, Vol.22(12), p.1860-1867
- [13] *Dev Brahman K., Kazi T. G., Baig J. A., Afridi H. I., Arain S. S., Saraj S., Arain M. B., Arain S. A.*; Biosorptive removal of inorganic arsenic species and fluoride from aqueous medium by the stem of *Tecomella undulate*, *Chemosphere*, 2016, Vol.150, p. 320-328
- [14] *Nasr A.B., Walha K., Puel F, Mangin D., Amar R.B., Charcosset C.*; Precipitation and adsorption during fluoride removal from water by calcite in the presence of acetic acid, *Desalination and Water Treatment* 2014, Vol.52, p.2231-2240
- [15] *Kabsch-Korbutowicz M., Majewska-Nowak K.*; Membrane separation processes in environmental protection, *Wrocław University of Technology, Wrocław* 2011
- [16] *Rautenbach R., Albrecht R.*; Membrane processes, *John Wiley&Sons, UK* 1989
- [17] *Scott K.*; Handbook of Industrial Membranes, *Elsevier Advanced Technology, Kidlington* 1995
- [18] *Kabay N., Arar O., Samatya S., Yuksel U., Yuksel M.*; Separation of fluoride from aqueous solution by electrodialysis, Effect of process parameters and other ionic species, *Journal of Hazardous Materials*, 2008, Vol.153, p.107-113
- [19] *Keri R.S., Hosamani K.M., Seetharama Reddy H.R., Nataraj S.K., Aminabhavi T.M.*; Application of the electrodialytic pilot plant for fluoride removal, *Journal of Water Chemistry and Technology*, 2011, Vol.33, p.293-300
- [20] *Rodrigues A., Brito A., Janknecht P., Proenca F.M., Nogueira R.*; Quantification of humic acids in surface water: effects of divalent cations, pH, and filtration, *Journal of Environmental Monitoring*, 2009, Vol.11, p.377-382
- [21] *Rajca M.*; Impact of the water composition on the degradation kinetics of natural organic matter in photocatalytic membrane reactors, *Environment Protection Engineering*, 2015, Vol.41, p.29-39
- [22] *Banasiak L.J., Schafer A. I.*; Sorption of steroidal hormones by electrodialysis membranes, *Journal of Membrane Science*, 2010, Vol.365, p.198-205
- [23] *Zhu X., He W., Logan B.E.*; Reducing pumping energy by using different flow rates of high and low concentration solutions in reverse electrodialysis cells, *Journal of Membrane Science*, 2015, Vol.486, p.215-22