



The removal of fluoride from water based on applied current and membrane types in electrodialysis



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ABSTRACT

In many places over the world, the fluoride concentration in ground water as raw water is higher than the threshold standard for healthy drinking water. This paper discusses a fluoride ion removal from sodium fluoride solution by using batch-system electrodialysis (ED) process. The effect of type of anion exchange membranes on the deionization rate of fluoride ion is investigated. The removal of fluoride ion from water by electrodialysis using CMX/AMX and CMX/ACS cation/anion exchange membranes combinations has been investigated on various applied currents, such as 3.64, 5.45, 9.09, 12.7, and 18.2 (A/m²). It was found that the increase of current densities made an operational time of ED shorter to achieve maximal ion removal. The effect of other coexistent ions such as chloride ion in water on the profile of ion removal and the ED cell resistance was discussed. It was also found that chloride ion passed more easily through anion exchange membranes than through fluoride ion.

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1. Introduction

One of the most precious and important resources in human life is clean water with sufficient quantity and quality. This is the reason why the drinking water shortage problem becomes a serious issue throughout the world. Ground water contains high concentration of fluoride in many parts of the world, particularly in parts of India, China, Central Africa and South America [1–3]. In Indonesia, high fluoride concentration in drinking water has been found in coastal areas of Asembagus, East Java [4]. The concentration of fluoride in well water based on the observation was up to 4.2 mg/L. Furthermore, fluoride concentration in river water was found between 5.5 and 14.2 mg/L. Based on World Health Organization (WHO) and the Indonesian Drinking Water Standard, 1.5 mg/L is the maximum level of fluoride ion contained in drinking water [5,6]. High concentration of fluoride in drinking water results in fluorosis (dental/skeletal abnormalities), cognitive impairment problem, anxiety, depression, and several neurological damages [7–9].

Because of permanent effect for body health, it is necessary to control the concentration of fluoride in drinking water. Recently, an adsorption has been considered as the most advantageous method for reducing excess fluoride in raw water. This is because the adsorption process offers a simple technology, and easy operation and maintenance [10,11]. Nevertheless, a low purity of treated water is a crucial drawback of the adsorption technique. Adsorbents are commonly fabricated in the form of fine powder or particles. Consequently, such particles could penetrate to treated water, which may cause safety issues for human consumption [12]. Beside adsorption, membrane separation through ultrafiltration (UF), nanofiltration (NF), reverse osmosis (RO), and electrodialysis (ED) has been introduced as superior technologies for the removal of specific ion in raw water [13–16]. Among them, ED process has been reported as an interesting method to remove unique contaminant such as excess fluoride ion in drinking water. Because of high pressure operation, energy consumption is still a challenge for NF and RO membranes [17].

In electrodialysis, a direct current (DC) is used as a driving force to transport ions through ion exchange membranes. Electrodialysis process is used for the concentration of electrolyte solutions, and for the dilution or desalination of solutions [18,19]. One of the advantages of ED process is the capability to produce safe drinking water with low energy consumption [20,21]. In addition, ED process is a simple operation without applied pressure for ion

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removal like NF and RO processes [22,23]. The present study discusses a detail investigation from the basic concept of electro dialysis for fluoride ion removal from ground water sources. ED process with the constant voltage system was reported by other researchers [24], while the constant current system is used in this work. The principle of desalination of ED process is the Faraday's law. The desalinated amount of ions is proportional to the amount of electric current flowed through the system. Thus, the constant current system is the best to discuss the desalination phenomenon, including the effect of coexisting ions.

2. Experimental

2.1. Materials

ED stack consisted of pairs of cation exchange membrane (CEM) and anion exchange membrane (AEM). CMX (Astom Corp. Tokyo, Japan) was used as CEM. AMX and ACS (Astom Corp.) were used as AEM. AMX is a standard AEM, and ACS is a monovalent anion selective AEM. The ion exchange capacity (IEC) of CMX was 2–3 meq/dry-g and those of AMX and ACS were 1.5–2.5 meq/dry-g.

NaF and NaCl (WAKO Pure Chem. Ind., Osaka, Japan) were used as a source of fluoride and chloride, respectively. NaNO₃ (WAKO Pure Chem. Ind.) was used in electrode solution.

2.2. ED setup

The laboratory scale of electro dialysis system Micro Acilyzer S3 (Astom Corp. Tokyo, Japan) described elsewhere by search group of Takagi [25] was used to investigate the permeation of fluoride and chloride ions. The detail design of cation and anion exchange membranes in ED stack is indicated in Fig. 1. The ED system was composed of a stack and electrode compartments. In Fig. 1, the compartments where a diluted solution flew through are called a diluted compartment and the compartments where a concentrated solution flew through are termed a concentrated compartment. Initial composition and concentration of dilute and concentrate compartments are the same. The stack consisted of 10 pairs of anion and cation exchange membranes with the effective surface area of 55 cm². In this setup, the volume of diluted solution, concentrated solution and electrode solution were 500 mL each. Each solution was circulated from a reservoir through the compartments at the flow rate of 1.2 L/min. The research was carried out at several applied current densities, i.e. 3.64, 5.45, 9.09, 12.72, and 18.18 (A/m²). ED operation was continued until the

applied voltage reached 18V which is the maximum available voltage of equipment.

2.3. Sample preparation and analysis

The solutions of NaF and NaCl were used as models of fluoride and chloride contaminated ions in groundwater sources. In dilute and concentrated compartments, initial concentrations of fluoride and chloride are 380 mg/L (20 meq/L) and 710 mg/L (20 meq/L), respectively. Concentrations of fluoride and chloride ions in the dilute and concentrated solution were analyzed by the conductivity measurement for single solution and by the ion analyzer (IA-300, DKK-TOA Corp., Japan) for mixed solution. For ion analysis by conductivity meter, a standard solution of fluoride or chloride is needed to predict calibration curves. At least seven bottles of fluoride or chloride solutions are prepared with various concentrations from 2.00 meq/L to 50.00 meq/L. Then, the conductivity of each solution is checked. Furthermore, data of concentration are plotted versus conductivity to obtain linear regression *c* as formula to calculate solution concentration in other sample.

The amount of fluoride or chloride ions passing through AEM per unit time per unit membrane surface area was calculated as a flux. The ion flux (eq/(m²s)) through the membrane is given by Eq. (1)

$$J(\text{eq}/\text{m}^2\text{s}) = \left(\frac{V_{\text{cell}}}{A}\right) \left(\frac{dC}{dt}\right) \quad (1)$$

Here, *V*_{cell} denotes the volume of concentrated compartment (m³), *A* is the effective surface area of anion-exchange membrane (m²), *C* is the ion concentration in concentrated compartment (eq/m³), and *t* is the time (s).

3. Results and discussion

3.1. Fluoride removal

In electro dialysis, one of the performances that should be evaluated is ion removal from feed to concentrated compartment. In this section, ED performance is evaluated by analyzing the time course of fluoride ion concentration in diluted and concentrated compartments every 3 min. Fig. 2 shows the concentration change of fluoride ion during electro dialysis using membrane set of CMX-AMX at various conditions of applied current density.

In Fig. 2, the feed solution contained only NaF and the initial concentration was 20.0 meq/L. It can be seen that fluoride concentration in the diluted compartment declined linearly to zero after an 84-min operation with the current density of 3.64 A/m² (Fig. 2a). Conversely, fluoride ion concentration in the concentrated compartment linearly increased up to 36.8 meq/L after 84 min. On the other hand, the operation time needed for fluoride ion removal became shorter with the increase of current density (Figs. 2b–d). When ED was run by applied current density of 18.18 A/m² (Fig. 2d), fluoride ion concentration in the diluted compartment went down to 0.53 meq/L after an 18-min operation. Meanwhile, fluoride ion concentration went up to 35.3 meq/L in the concentrated compartment. Based on those results, it was found that current density gave significant effect on migration of fluoride ion in an electro dialysis system. This is because the amount of deionized ions is determined by the amount of current flowed through IEMs that adopts the Faraday's law [18].

Fig. 3 shows the effect of AEM type on the amount of removed fluoride ions in a diluted compartment. For membrane set of CMX-AMX, it appears that the amount of removed fluoride ion achieved maximum capacity of 10 meq after an 18-min operation with the applied current density of 18.18 A/m² (Fig. 3a). Likewise, when ED

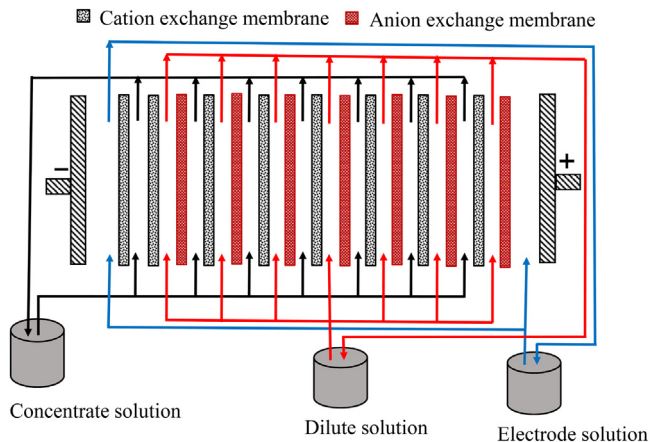


Fig. 1. Schematic setup of ED system for removal of fluoride and coexisting ions. Effective membrane surface area of each IEM is 55 cm² and the volume of each solution is 500 mL.

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