Investigation of the performance determinants in the treatment of arsenic-contaminated water by continuous electrodeionization

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ABSTRACT

Arsenic is one of the most toxic elements which cause serious human health problems with even a little amount in water. This study investigated the pentavalent arsenic anion ($HAsO_4^{2-}$) removal from water using continuous electrodeionization (CEDI). CEDI is a hybrid separation process of electrodialysis and ion exchange process that removes ionized species from liquids using electrical potential. The arsenics removal behavior with the change in the system condition was investigated under the programmed cell potential. Potentials from 0.8 to 5.2 V by 0.2 V step change were introduced with three-hour retention time to investigate the cell saturation level. At each potential step, conductivity, ion concentration, and pH of each compartment of CEDI stack were analyzed together to develop an ion transport and ion exchange model. Under the fixed feed condition, three major parameters, the resin saturation level, the cell voltage, and the age of a cell, affected the removal efficiency of the CEDI operation. The relationship among the factors are entangled, and they have a complex impact on ion removal efficiency. This study proposes ion drain rate (IDR) as an important indicator of the electroregeneration state. The system response to the voltage change was classified into five types by observing the IDR value in the concentrate stream and the conductivity in the diluate stream. While the system showed the highest ion removal efficiency under a high voltage, a low cell age, and a low saturation level condition, the three factors are complementary to one another.

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1. Introduction

Arsenic-contaminated water is threatening more than 100 million people's life in South and Southeast Asia by its carcinogenic, acute and chronic poisoning properties in the human body [1,2]. Especially, arsenic is harmful due to the inhibition on repairing process of damaged DNA. Long-term exposure of arsenic intake causes skin, bladder, lung, renal, and liver cancers as well as skin thickening, neurological disorders, muscular weakness, and nausea [3–5]. Many countries and the World Health Organization (WHO) have stipulated that the maximum level of arsenic in drinking water should be 10 μg L^{-1} [6,7]. Many of researches have been conducted for the remediation of arsenic contamination of river water, groundwater, and industrial wastewater by various methods of oxidation, phytoremediation, coagulation-flocculation, adsorption, ion exchange, electrocoagulation, electrodialysis (ED), reverse osmosis (RO), capacitive deionization (CDI) and electrodialysis reversal [4,2,8–12].

This study suggests an electrochemical method of continuous electrodeionization (CEDI) as an efficient treatment for arsenic-contaminated water. CEDI is a hybrid separation process involving ED and conventional ion exchange technology which appropriates removing ions from water. Various CEDI applications have been reported, such as the recovery of copper, nickel, chromium, and other heavy metals from the primary coolant of nuclear power plants, water softening, and ultrapure water production [13–21].

Membrane filtration, electrodialysis, and capacitive deionization are the comparable technologies. Reverse osmosis and nanofiltration provide As(V) removal efficiencies above 90% and 85%, respectively [22,23], electrodialysis can remove As(V) up to 80% [12]. Capacitive deionization is an emerging technology for removal of charged ions by electrosorption onto the electrode. CDI enables high energy efficiency in salt removal with small cell voltage. The removal efficiency of CDI is above 95% with a feed concentration below 1 mg L^{-1}, but the residual arsenic amount increases steeply when the initial concentration increases [24,25]. Moreover, a CDI cell can be operate for a finite time until the electrode is fully charged, and then the process must cease for a desorption process. This intermittent operation

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Nomenclature

C_c	concentrated water concentration, $\frac{mg}{L}$		J_{total}	total ion flux of the cross sectional EDI cell, $\frac{mol}{s\,cm^2}$
$C_{c,MB}$	mass balance concentration for concentrate compartment, $\frac{mg}{L}$		q	flow rate, $\frac{cm^3}{s}$
C_f	feed water concentration, $\frac{mg}{L}$		u	velocity of fluid across the EDI cell, $\frac{cm}{s}$
C_i	concentration of species i, $\frac{mol}{cm^3}$		V_c	concentrate flow rate, $\frac{mL}{min}$
C_p	product water concentration, $\frac{mg}{L}$		V_f	feed flow rate, $\frac{mL}{min}$
D_i	diffusion coefficient of species i, $\frac{cm^2}{s}$		V_p	product flow rate $= V_f$, $\frac{mL}{min}$
F	Faraday constant, 96,485 $\frac{C}{mol}$		x	thickness of the EDI cell, cm
I_a	current carried by anionic species, A		z_i	valence of species i
I_{cell}	total current across the EDI cell, A		φ	electrical potential across the EDI cell, V
J_i	ion flux of species i, $\frac{mol}{s\,cm^2}$			

requires complicated fluidic handling [26]. CEDI can provide a higher ion removal efficiency compared to other electrochemical methods, also it can maintain constant water quality with continuous flow.

The forms of an arsenic present are dependent on the type and amounts of sorbents, pH, redox potential (Eh), and microbial activity [27]. Two forms are common in natural waters: As(III) and As (V). Pentavalent (+5) species are predominant and stable under aerobic conditions like surface water. Trivalent (+3) arsenites predominate in moderately reducing anaerobic conditions such as groundwater [4]. Removal of arsenic is hindered by the fact that arsenic has various valence states. A number of researchers have reported that arsenic is most effectively removed or stabilized when it is present in the As(V) form. Moreover, As(III) is not a proper form to treat with an electrochemical method such as CEDI. The As(III) generally exists as a neutral form (H_3AsO_3), which is not affected by an electric field. CEDI system uses ion exchange resin to adsorb ions and electric field to discharge ions. Therefore, As(V) which exists in the form of the anion ($HAsO_4^{2-}$) is the most appropriate state of arsenic to treat.

In the configuration of CEDI stack, ion exchange membranes are placed between two electrodes to separate the compartments of diluate and concentrate, which is similar to ED. The membranes in the system cause the depletion and concentration of ions in the diluate and concentrate compartments, respectively [28]. Ionized species are removed from liquids under an electrical potential through electrically active media filled in the diluate compartments. In CEDI devices, electrically active media play an important role in the diluate compartment by reducing the electrical resistance of the compartment. It enables CEDI to have a much lower cell resistance and a higher removal efficiency compared to ED. Unlike the conventional ion exchange process, CEDI does not require an additional chemical process to regenerate the ion exchange resin. The ion exchange resin is continuously regenerated by hydrogen and hydroxide produced by water dissociation under the applied direct current (DC) [29]. Fig. 1 shows the basic concept of electrodeionization.

Although the principle of CEDI is rather simple, the actual operation is quite complicated. Among the several operating parameters which should be under surveillance, the status of resin regeneration should be frequently monitored for continuous operation with maintained system performance. However, it is difficult to evaluate the regeneration status of the resin directly; it can be only indirectly demonstrated by the mass balance of the two compartments. This present study focused on analyzing the system characteristics of a CEDI process. During a test operation, a variety of factors related to the system performance were monitored. The ion transport phenomenon in a diluate chamber and the performance deciding factors of CEDI operation were investigated.

2. Materials and methods

2.1. Materials

A cation exchange membrane (CMX, ASTOM Co., Japan) and an anion exchange membrane (AMX, ASTOM Co., Japan) were used as the ion exchange membranes. For the diluate compartment, strongly acidic cation exchange resin (AMBERLITE™ IR120 H⁺, Rohm and Haas Company, USA) and strongly basic anion exchange resin (AMBERLITE™ IRA 402 OH⁻) were used as the ion exchange media. Synthesized arsenic-contaminated water, which contained $Na_2HAsO_4 \cdot 7H_2O$ (A6756, Sigma-Aldrich Co., USA), was used as the feed water. As the solutions for the concentrate and the electrode rinse, Na_2SO_4 (238,597, Sigma-Aldrich Co., USA) was used.

2.2. CEDI cell preparation

Concentrate compartments were placed on both sides of the diluate compartment which also used as the electrode rinse compartments. The volume of the diluate compartment was 7.2 mL (24 cm² × 0.3 cm). Ion exchange resins were packed into the diluate compartment. Each compartment was separated by an array of membranes between the electrodes. Each CEDI cell had an effective membrane area of 24 cm² (16 cm × 1.5 cm). Gaskets not only supported the membranes but also distributed the process fluids in the compartments. The components of the CEDI stack are shown in Fig. 2.

A mixed bed cell was used for the CEDI analysis. The mixing ratio of the cation exchange resin and anion exchange resin was 1:1 as the total exchange capacity. To compare the system with different resin states, an aged cell and a fresh cell were prepared. To investigate the system characteristics under normal operation condition, an aged cell was prepared with the ion exchange resin used for 200 h under various voltage conditions. The fresh cell was examined to observe the characteristics of fresh ion exchange resin that has especially not been through the regeneration process. The ion exchange resin itself is a strong adsorbent. Unsaturated fresh ion exchange resin could adsorb the inflow ions even without electric field. Therefore, to evaluate the effect of As(V) removal under an electric field, a fresh cell was prepared with the saturation of arsenate ions by immersing it in 8 g L⁻¹ sodium arsenate solution for 12 h. Consequently, the aged cell with an intermediate saturation degree and the fresh cell with fully saturated resin were prepared.

2.3. CEDI operation with increasing potential steps

The various potentials with long retention time were used to observe the overall CEDI system response and the ion transport