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Evaluation of treatment and recovery of leachate by bipolar membrane electrodialysis process

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ABSTRACT

Recovery studies are frequently carried out for electrodialysis (ED) processes. In this study, beyond examining the recovery of leachate components in an electrodialysis process, the use of that process to treat leachate-containing wastewater was simultaneously tested. Leachates were initially pre-treated (ultra filtration + cation exchange) to prevent clogging and harmful effect to the bipolar electrodialysis membranes. Optimum operating conditions were determined at the end of the experimental studies. Online observations during the electrodialysis process included the temperature-dependent reaction time, conductivity and changes of molar concentrations of H^+ and OH^- ions in both the anolyte and catholyte compartments in which removed ions were collected. The most important contaminants in leachates are organic substances and nitrogen compounds. For this reason, representations of organic substances, such as the chemical oxygen demand (COD), and nitrogenous compounds, such as total Kheldahl nitrogen (TKN) and ammonia-nitrogen (NH_3-N), were also monitored in the electrodialysis effluent. Under the optimum operating conditions, removal of NH_3-N, TKN and COD were determined in the effluent at 96.2%, 92.8% and 86.7%, respectively. The conductivity value was determined to be 1.97 mS/cm at the end of the study.

1. Introduction

Electrodialysis (ED) is a process for separating ions based on their size under the influence of an electrical current [1]. The most important difference of ED compared to other membrane processes is the use of an electrical current, rather than pressure, to induce the ions to pass through the membrane; the use of pressure is a major cost factor for other membrane processes. Low levels of electrical current can be used, which helps to reduce the cost of ED. ED is used extensively in recycling systems, as well as in drinking water treatment, because of the high quality of the effluent [2–6]. ED is also used in wastewater treatment [7,8].

ED process is favored among the other treatment processes for its numerous advantages. Also, it is important to know the advantages and disadvantages of a process to predict and prevent potential problems. The ED process is competitive with other separation processes, such as reverse osmosis (RO), ion exchange and dialysis, because of its advantages in many applications [9,10]. The advantages of ED include low-pressure operation, no anti-scalant requirements, longer membrane life expectancy (8–10 years compared with 1–2 years for reverse osmosis (RO)), lower maintenance requirements than RO, good effectiveness against various ionic species, applicability for moderate strength (10,000 mg/L TDS) wastewater, and the gathering of concentrated ions in two different compartments, which aids ion recycling. The advantages are that ED provides high selectivity, has a high product recovery ratio and can destroy at least some of the components in the raw water (drinking or waste), and it can run continuously. ED also does not require treatment chemicals, and it has relatively low energy and capital cost requirements [11]. Given all of these advantages, the use of ED is expected to continue to increase in the future [12].

The disadvantages of ED are that it requires stream pretreatment, the need for electrical safety, low experience to date with the process for wastewater, and that it is not effective for removing microorganisms and many anthropogenic organic contaminants.

The ED process includes several configurations, including electrodialysis, electro-deionization (EDI) and bipolar membrane electrodialysis (BMED). Electrodes and ion-selective membranes are used in all three systems. While ion-exchangers are used in the EDI process, bipolar membranes are included in BMED systems [13]. These bipolar membranes are formed by combining high-strength, low-voltage anion and cation exchange membranes [14].

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Table 1
The characterization of leachate from Odayeri Landfill.

Analysis	Units	Raw leachate	Pretreated leachate
pH	–	8.1	8.5
COD	mg/L	14,400	7200
BOD_5	mg/L	7350	3740
BOD_5/COD	–	0.51	0.52
TKN-N	mg/L	3110	2185
Ammonia-N	mg/L	2910	1790
Sodium	mg/L	3620	9430
Chloride	mg/L	4010	4370
Calcium	mg/L	195	12.50
Magnesium	mg/L	134	0.70
Conductivity (at 20 °C)	mS/cm	39.80	14.61

The bipolar membrane electrodialysis (BMED) process has an important advantage compared with a conventional ED process because anions and cations are separately removed from the sample and combined with H+ and OH− ions by means of a bipolar membrane so that separate acidic and alkaline solutions are obtained [15]. Thus, ions in the sample are removed from the leachate and are used simultaneously to produce acidic and alkaline solutions [16,17]. These solutions can then be used as by-products or for resource recovery rather than requiring disposal. This production of acids and bases increases interest in the process [18,19]. In particular, it is possible to find literature studies where BMED is used to produce acetic acid, propionic acid, citric acid and lactic acid [20–24]. Some biotechnological research has also been carried out on reducing the ED process cost [25]. Reactions in electrodes during the BMED process are as follows:

$$\text{Anode}: \quad OH^- \rightarrow 0.5H_2O + 0.25O_2 + e^- \qquad (1)$$

$$\text{Cathode}: \quad H^+ + e^- \rightarrow 0.5H_2 \qquad (2)$$

For one study, a leachate known to have high ionic species content was chosen to be treated with a BMED process [26,27]. The studied leachate was considered to be middle-aged, which is the most frequently encountered age.

The purpose of this study is to evaluate the treatability of leachate by BMEDs and simultaneously investigate the availability and use of anolyte and catholyte liquids as acidic and alkaline solutions.

2. Material and methods

2.1. Leachate

In the experimental study, leachate from the Odayeri Landfill in Istanbul was used. The Odayeri Landfill was established in 1995 and has served a 75-ha area for 8000 tons of municipal solid waste (MSW) per day since that date. The properties of the leachate were analyzed, and average values are given in Table 1. Next, optimum operating conditions were determined at the end of the experimental studies.

The six parameters, namely, pH, COD, biochemical oxygen demand BOD, BOD/COD, TKN and NH_3-N, given in Table 1 show that this leachate can be defined as "middle-aged". The concentrations after pretreatment for the four ionic parameters (sodium, chloride, calcium and magnesium) that can cause membrane clogging during the ED process are also shown in the table. The conductivity is another important parameter for ED process that is related to the ionic types and concentrations the leachate.

2.2. Pretreatment

The leachate was pretreated prior to ED to prevent clogging on membrane surface by particulate matter. An ultrafiltration system was used to remove particulate matter. A cationic ion exchange was subsequently used to remove higher ionic types, particularly Ca^{++} and Mg^{++} that are the main cause of calcification on a membrane surface. Calcification mechanism of Ca^{++} was shown as follows:

$$CO_3^{-2} + Ca^{+2} \leftrightarrow CaCO_3 \downarrow \qquad (3)$$

$$2HCO_3^- + Ca^{+2} \leftrightarrow CaCO_3 \downarrow + CO_2 + H_2O \qquad (4)$$

$$CO_2 + 2OH^- + Ca^{+2} \leftrightarrow CaCO_3 \downarrow + H_2O \qquad (5)$$

The main advantage of ion exchange is to exchange the higher-valence cations for the single valence cation Na^+. As shown in Table 1, the chloride value is also high in the leachate, but chloride does not cause any clogging problems on the membrane. On the contrary, the chloride actually contributes to producing an acidic liquid in the anolyte chamber as HCl, which aids material recovery with ED.

2.3. Bipolar membrane electrodialysis (BMED) process

In this study, a PCCell ED 64-4 unit and BMED 1-3 bench ED pump unit obtained from PCCell GmbH were used. The specifications and operation parameters of the cell and bench ED pump unit are given in Table 2.

PCCell PC-SA and PC-SK brand ion exchange membranes were used to ensure deionization process. The characteristics of ion-exchange membranes used in ED process were shown in Table 3.

Experimental setup of ED process was shown with three different ways in Fig. 1a–c for better understanding of the basic mechanism of ED process.

PCCell ED 64-4 unit and BMED 1-3 bench ED pump unit's overall layout were seen in Fig. 1a. PCCell ED 64-4 unit which is the point of the reaction take place was shown in detail in Fig. 1b.

As seen in Fig. 1b, deionization is provided with electrolytic reactions that are occurred between membranes and electrodes. An example reaction scheme was shown in Fig. 1c to comprehend clearly.

Deionization was provided with anion and cation selective membranes in basic ED process. In this way, anolyte solution contains intense anions and catholyte solution contains intense cations. Bipolar membrane ED process can be created with adding bipolar membrane to the basic ED process. In this way water molecules decompose to H^+ and OH^- ions by electrolysis. In this way, the conversion of anolyte and catholyte solutions to acidic and alkaline solutions can be provided. As a result, the mixed anolyte and catholyte solutions which have acidic/alkaline properties are obtained. Acidic and alkaline solutions can be recovered from this mixed solution. In this way, production of waste is prevented and economical contribution can be obtained.

Prior studies showed that conductivity decreased to 0.05 mS/cm with the BMED process; however, a decrease to 2 mS/cm is considered sufficient to stop treatment because the removal efficiency rate decreases as the conductivity is reduced below 2 mS/cm. Therefore, a 2 mS/cm conductivity was generally considered to be the treatment target in this study. In certain cases where the electrical voltage was low or there were multiple membrane sets so that reaching 2 mS/cm was not realistic, the treatment time was used as the criterion for ending an ED test. As a result of prior studies, 360 min was considered sufficient for ED processes with low electrical voltages.

Batch studies were conducted to understand the effects of the operation conditions. At the final stage of the study, process was conducted with continuous flow and similar results were obtained like batch studies. In addition to parallel results with batch studies, also lower resistors were obtained in continuous studies. This