Application of Electrodialysis Process for Reduction of Electrical Conductivity and COD of Water Contaminated By Composting Leachate

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Abstract

The presented paper describes an experimental study to reduce electrical conductivity (EC) of composting leachate-polluted water by using electrodialysis (ED) process. High efficiency, simple operation, low waste generation and selectivity are considered as major advantageous of applying ED process. Along with evaluation of ED method for desalination, the possibility of the process for COD (chemical oxygen demand) removal was also studied. The impact of applied voltage, feed concentration and process time on ED performances were investigated. Increasing of the applied voltage and decrease of feed concentration enhanced the reduction of EC and improved the COD removal from the sample. At optimal condition (Voltage=10 Volt, feed solution=Cf/4 and time operation=120 min), the reduction of EC and COD removal were 92.7%, and 83.8%, respectively. Applying higher voltage and using more feed solution concentrations resulted in more energy consumption. The obtained results showed that ED method can be considered as an acceptable method to reduce salt and organic content.

Keywords: Electrodialysis; Electrical Conductivity; COD; Membrane; Composting Leachate-Polluted Water.

1. Introduction

Along with considerable benefits of composting process, which is a technology for converting organic matter to organic fertilizer, there is always rise of high different types of biodegradable and non-biodegradable contaminants within the process [1]. Consequently, release of leachate without proper treatment pose serious threads to the environment and human health [1, 2].

Although there are several biological and physic-chemical methods to treat leachate, due to the complex composition of leachate, conventional treatment processes are not promising enough for the treatment of this kind of wastewater [3-5]. In recent years, advanced oxidation processes have been widely used as an alternative method for treatment of refractory wastewater [5-7]. In our previous experience, the performance of Electro-Fenton (EF)-which is an advanced oxidation method-for reduction of chemical oxygen demand (COD) from composting leachate-polluted water was investigated. Results illustrated a COD removal efficiency (up to 50%) in optimal condition which obtained at initial pH of 3, current of 3A, H_2O_2 concentration of 10 mL and process time of 60 min.

However, during EF method, the electrical conductivity of the water increased from 5 to 15 mS/cm. Since there is a

standard limitation for conductivity of wastewater to be discharged into the sewage system or surface water, the conductivity needs to be reduced before dumping [8].

In the current study, application of electro-dialysis which is a type of electrochemical membrane process on electrical conductivity reduction was evaluated. ED process is able to concentrate and separate ions from wastewater [8] and could be efficient on EC reduction. Also, because of not very high COD reduction efficiency (50%) obtained with electro-fenton method, along with EC, COD reduction during ED process was evaluated.

There are many applicable processes to reduce salinity such as ion exchange, electrodialysis (ED), reverse osmosis (RO), crystallization and nanofiltration (NF). RO and NF are known as pressure driven membrane processes and need a quite complex pre-treatment to reduce membrane fouling which leads to high capital cost of the process. Moreover, RO technology is ion uncontrollable and relying on size of molecules [9].

However, ED process has several advantages over other separation methods. It doesn't generate large amount of waste, and use hazardous solvent. In addition, ED doesn't suffer from short lifetime of absorbent [10]. ED also offers simple operation, low chemical consumption, appropriate selectively for ion separation, high product recovery [11-16]. Similar to other membrane technologies, membrane fouling is a main obstacle of ED technology [17]. Fouling in ED leads to an increase in cell resistance which consequently decreases ion migration and selectively [8]. However, this problem can be addressed by different methods such as applying struvite crystallization pre-treatment to ED [8], or by combination of ion exchange membrane (IEM) and ultrafiltration (UF) membrane [16].

 ED process is a combination of conventional ion exchange technology with membrane technology for removing ions (electrically charged particles) from one solution (known as diluted compartment) to another (concentrated compartment) through selective semi permeable ion exchange membranes (IEMs) by introducing an electrical field [18-20]. ED cell consists cation and anion exchange membranes (CEM and AEM) which are the main component of cell and placed alternatively between on pair cathode and anode. These membranes partition electrolyte into segments [15, 21]. By applying an electrical potential to the cell, charged particles (negative or positive) in the electrolyte move towards the electrodes with the opposite charge, consequently, ionic concentration will increase in some compartment and deplete in the adjacent ones [12, 21, 23].

ED has been successfully applied for different purposes such as: removal of hardness [12-14], separation of ionic species [21, 24], recovery of beneficial products (e.g. PO4-P and NH4-N) from effluents [8, 9, 22, 23, 25] and desalination [9, 15, 26, 27]. ED can also be implemented for the treatment of industrial wastewater [17, 28] such as reducing electrical conductivity from almond industry [29], chloride removal from wet flue gas desulfurization wastewater [28], or removal of harmful ions such as F^-, NO_3^-, CL^- and Br^- and heavy metals such as pb^{2+}, Ni^{2+}, Cu^{2+}, Cr^{3+}, Mn^{2+}, Fe^{3+}, etc. [31, 32].

ED has also been studied for other applications, such as removing chromium (VI) from aqueous solutions using a novel hybrid liquid membrane [33], processing a solution of lithium chloride containing organic solvents [34] and concentrating NaCl from seawater reverse osmosis brines for the chlor-alkali industries [35]. Additional functions include selective separation of mono and di-valent cations during brackish water desalination [36], purification of pentoses from hemicellulosic hydrolysates with sulfuric acid recovery [37], purification of H_2SO_4 of Pickling Bath contaminated by Fe(II) Ions [38], and Synthesis and transport of impurities and production of choline dihydrogen phosphate [39, 40].

According to the ED principles, by applying electric potential to the ED cell containing wastewater, cations and anions existing in the feed solution pass through the CEM and AEM toward the cathodic and anodic compartments, respectively. It is expected for feed compartment to be depleted from the ions as time goes on. By this mean, the hardness of feed solution is reduced. Additionally, the electric field makes a situation for transfer of acidic organic materials to ions which pass through CEM and AEM from diluted (feed) to the concentrated compartments. Thus, it leads to COD reduction of feed compartment [23].

2. Experimental Section

2.1. Materials and Methods

All samples were leachate-contaminated water, which was taken from Esfahan Composting Plant (ECP), Iran. Samples containing raw and pre-treated wastewater were analyzed to determine the EC, COD, BOD, and TDS (Table 1). EC was measured using water quality meter (Model 8603) and COD was measured by using "closed reflux method" according to standard methods. All samples were stored at 4°C until use. An ED set up containing three cylindrical compartments was made from plexiglass, owing to its proper resistance to acid, base and feed solutions and all compartments had equal thickness of 10 mm. The compartments were separated by a pair of CEM and AEM. Effective area of each membrane was 9.5×4.5 cm². The anion and cation exchange membranes used in this work were AR204SXR412 and CR67, MK111 (Ionics, MA, USA), both supplied by Arak Petrochemical Complex. Main properties

of the membranes have been listed in Table 2.

Table 1. The characteristics of raw and pre-treated wastewater

Parameters	Raw wastewater	Pre-treated wastewater
EC (mS/cm)	5-6	11-15
COD (mg/L)	12,000	4500
BOD (mg/L)	7500	1600
Turbidity (NTU)	142	112
Colour	Dark-brown	Light-brown
TDS (mg/L)	8875	-

Table 2. Properties of the anion and cation exchange membrane

Specification	Membrane	
	AR204SXR412	**CR67, MK111**
Matrix structure	Homogeneous polystyrene DVB copolymer	Homogeneous polystyrene DVB copolymer
Specific Weight (mg/cm^2)	13.7	13.7
Thickness (mm)	0.5	0.5
Burst Strength (kg/cm^2)	7.0	7.0
Water Content	46 % of wet resin only	46 % of wet resin only
Capacity (meq/dry gram resin)	2.8	2.4
Chemical stability, pH	1-10	1-10

2.2. Experimental Procedures

Before each experiment, per-treatment of membranes was performed to remove impurities and stabilize them. In this regard, AEM and CEM were dipped into the 1M NaOH and 1M HCl solutions, respectively, for two hours and washed with distilled water. As a result, the anion and cation exchange membranes were changed to hydroxide and hydrogen form. Platinum-coated titanium and nickel mesh (supplied by Niroocholer Co, Esfahan, Ira) which had the same effective area as membranes, were used as the anode and cathode, respectively. Adjustable DC power supply (Star 305, Iran) was utilized to apply electric potential difference to the cell. It could supply the desired voltage and direct current in the range of 0-40 Volt and 0-4 A, respectively. The experiments were performed at ambient temperature and batch mode. Firstly, the reservoir tanks of electrode rinse solutions were fed with 1 litter of 0.02 M H_2SO_4 and 1 litter of 0.02 M HCl solution. Then the feed tank was filled with 400 ml of wastewater and solutions were circulated by proper pumps through the compartments. For each experiment, fresh solutions were used. Afterward, appropriate voltage was applied, and experiment was started. All the experiments were conducted for 120 minutes at constant voltage and samples of feed solution were taken every 30 min. A schematic diagram of the experimental set up for ED is illustrated in Figure1.
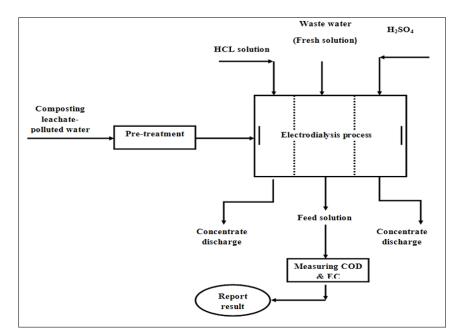
Figure 1(A). A schematic diagram of the experimental set up for ED method
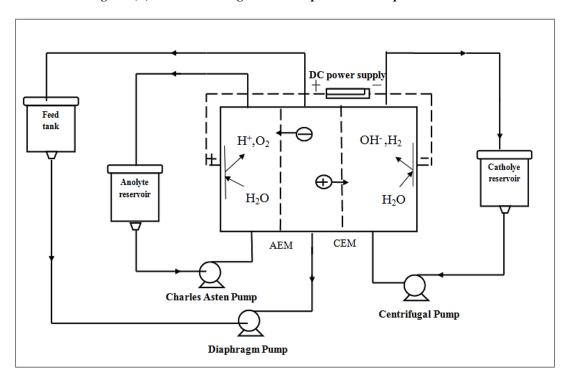

Figure 1(B). Batch mode experimental set-up of ED system, electrodialysis process (three-compartment cell) with the Anion exchange and Cation exchange membrane

2.3. Calculation

In this study, ED performances such as energy consumption, efficiency of COD reduction, and EC reduction in the term of some operating parameters (voltage, concentration, time) were evaluated. Removal efficiency of COD indicated the percentage of organic load removed from the feed solution and it is determined by using the following equation:

$$\eta = \frac{COD_0 - COD_t}{COD_0} \times 100 \tag{1}$$

Where COD_0 (mg/L) and COD_t (mg/L) are COD of feed solution at time zero and t, respectively. Energy consumption can be described as energy needed for electrolysis cell and was calculated via following equation:

$$E = \frac{\int_0^t VI\, dt}{V_D} \tag{2}$$

Where E, V, I, t and V_D are energy consumption (W.hr/L-1), applied voltage (Volt), cell current (A), operation time and initial volume of diluted compartment (L) respectively [18, 41, 43]. Further, the reduction of EC is defined as following equation:

$$EC\% = \frac{EC_0 - EC_t}{EC_t} \times 100 \tag{3}$$

Where EC_0 (ms/cm) and EC_t (ms/cm) are electro conductivity of feed solution at time zero and t, respectively.

3. Results and Discussions

Different operating parameters can affect the performance of an ED process. Operation parameters considered in this research were: applied voltage (5, 10, 15 and 20 Volt), concentration of feed solution, which was considered Cf when no dilution was done and Cf/2 and Cf/4 when the feed solution diluted twice and quadruplicate, respectively) and process time (30, 60, 90 and120 min).

3.1. Effect of Applied Voltage on EC and COD Removal

To investigate the effect of voltage, the experiments were conducted with 400 mL initial volume of feed solution at ambient temperature for 120 min. Anolyte and catholyte reservoirs consisted the 0.02 M H_2SO_4 and 0.02 M NaOH solutions, respectively. Limiting current (LC) is an important factor which determines current utilization and electrical resistance in an ED cell. Prior to doing any ED experiment, LC should be investigated to see its influence on the cell performance. In high levels of voltage, sometimes ions depletion occurs at the membrane interface of the feed compartment. Current density at this moment is called LC. This chemical phenomenon which is known as concentration polarization results in reduction of ions transfer through the membrane. Existence of a point of inflection in the plot of voltage against the current shows the occurrence of concentration polarization [44, 45]. Performing ED below the limiting current is desirable [18, 20]. Here, our preliminary experiments were attributed to study the concentration polarization in the proposed ED cell considering all levels of applied voltage. As Figure 2 shows, for all concentration levels, electrical current increased with increase of applied voltage and no inflection point was observed. This directly indicates that no concentration polarization occurs in the ED cell in the range of voltages that studied in this research (5-20 V).

As the first step to evaluate the ED performances, the ability of the proposed setup to reduce the conductivity of samples was studied. Figure 3 describes the variation of "conductivity reduction" as a function of applied voltage. The ED setup can decrease the sample conductivity up to 90%. As illustrated, when voltage increased, "conductivity reduction" efficiency of feed chamber enhanced. This can be explained by considering the fact that by applying more voltage to the ED cell, more electric field and consequently more potential difference (driving force) for transfer of ions through membranes is provided. This ultimately enhances the EC reduction ability of the device. Considering all voltage levels, as time goes on, the EC reduction rate will be less because with the passage of time more ions are migrated from feed compartment so concentration of ions in feed chamber decreased (high electrical resistance). Consequently, at the constant voltage, lead to lower electrical current, higher driving force would be needed to transfer remained ions. Furthermore, according to the above reason, for all voltage levels at the initial time, EC% was augmented sharply. Also, it was observed that at the beginning of the process, increasing applied voltage had no effect on conductivity of feed compartment. It can be explained by the fact that although the driving force increased, the number of ions can pass through membranes is limited.

The next step was to study the efficiency of ED process toward COD reduction. According to Figure 4.a, as applied voltage was increased, due to increment of driving force, the percentage of "COD reduction" increased from 37% in voltage of 5 to 84% in voltage of 20 V. In other word, dissociated acidic organic materials in feed solution are transformed to the ions under the generative electrical field. These ions are transferred through ion-exchange membranes from feed to the concentrated solution [46]. Besides, no significant change in COD was observed for different voltages after 120 min. Figure 4.b represents the variation of the current density with time for different values of applied voltage. Increasing the applied voltage provided more driving force for ions to transfer through the membranes. This ultimately increases the current density. As time goes on, more ions migrate toward the anolyte and catholyte chambers, the feed compartment is more depleted, so electrical resistance was augmented and as a consequence, overall cell current density decreased. Feed compartment has major contribution of current density in an ED cell.

The energy consumption should always be considered as a critical parameter in the ED process. Figure 5 indicates the variation of energy consumption with applied voltage for different feed concentrations. SPC increased as higher voltage applied to the ED setup. Energy dissipation in the form of heat generation which is directly affected by using higher values of voltage, ultimately increases the temperature. In this case more energy is needed to achieve reasonable EC/COD reduction efficiency. Besides, sometimes, higher temperature destroys the functionality of the membranes. Considering all figures here, since voltage can affect the EC/COD reduction ability of ED setup (positively) and energy consumption of the cell (negatively), there is a trade-off for choosing the optimum value for voltage to be applied to the ED cell. Both positive and negative impacts of voltage is needed to be considered to design an optimum ED process. The proposed ED method should meet the reduction efficiency requirements while considering energy limitations [47-49].
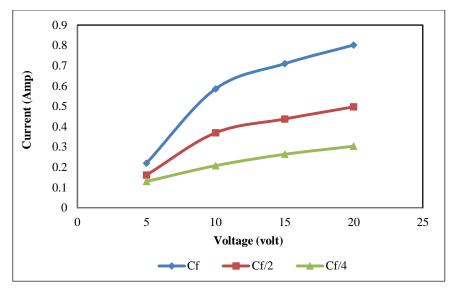
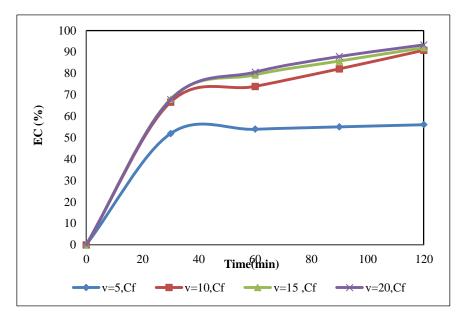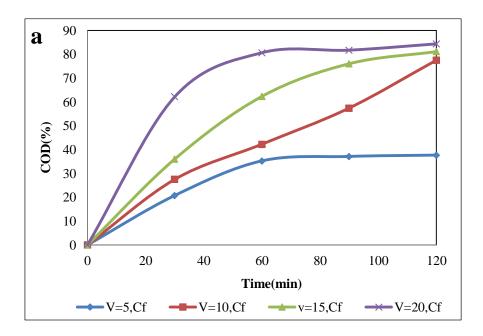**Figure 2. Variation of averaged current with applied voltage**

Figure 3. Effect of applied voltage on conductivity reduction efficiency
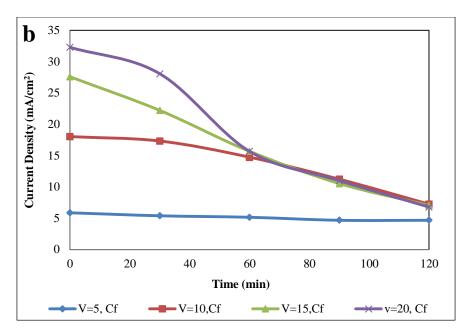
Figure 4. Effect of applied voltage on a) removal efficiency of COD and b) cell current density
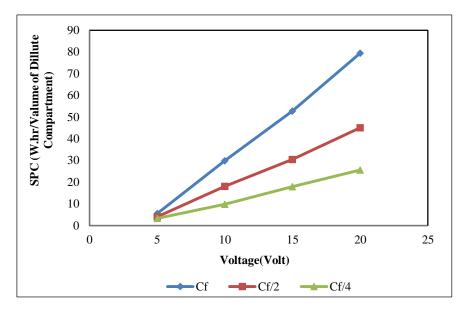

Figure 5. Effect of applied voltage on specific energy consumption

3.2. Effect of Feed Concentration on EC and COD Removal

In order to study the effect of feed concentration on EC/COD removal and energy consumption, experiments were carried out with three different concentrations of feed solution (Cf, Cf/2, Cf/4). These experiments were performed by using 1lit of 0.02M H_2SO_4 and NaOH as solutions of anode and cathode compartments, respectively for 120 min. Figure 6.a presents the variation of "EC reduction efficiency" and COD (as evaluating parameters) versus time. Here, applied voltage kept constant at 15 V. Maximum efficiency (~91%) was achieved in the lowest initial feed concentration (Cf/4). The result was in accordance with similar study was done by Masigol et al, [44]. They have shown that using lower feed concentrations leads to obtain more ions removal efficiency. This can be explained by the fact that at lowest initial feed concentrations, total number of ions in the feed chamber is reduced. As a result, the cell electrical resistance is augmented thus leading to lowest EC so conductivity reduction" efficiency of feed chamber enhanced.

The variation of "COD reduction efficiency" with time as function of feed concentration has also been shown in Figure 6.b. Here we observed that the impact of using lower feed concentration on COD reduction is more significant in initial stages of experiment (operation time less than 60 min). As time goes on, specifically higher than 60 min, the effect of feed concentration becomes less significant. As a summary, both EC and COD reduction efficiency reach to their 90% maximum when ED process is performed for about 80 min.

To complete the study, influence of feed concentration on energy consumption and cell current density was also evaluated. As can be seen in Figure 7 (a and b), increasing the feed concentration results in higher cell current density, which obviously is due to existence of more ions into the feed compartment. Besides, less energy dissipation occurs when Cf/4 concentration was applied to the feed chamber.
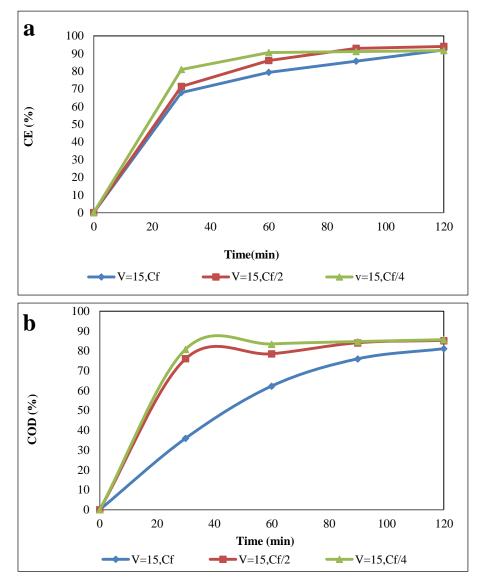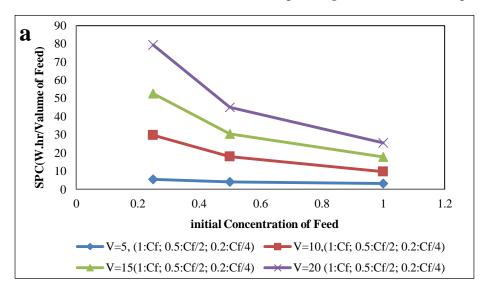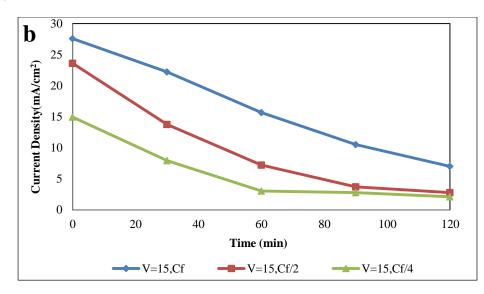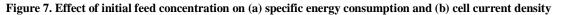

Figure 6. Effect of initial feed concentration on (a) EC reduction percentage and (b) COD removal percentage

Figure 7. Effect of initial feed concentration on (a) specific energy consumption and (b) cell current density

4. Conclusion

In this work ED process was utilized for reducing electrical conductivity and COD of pre-treated wastewater. Here the major hypothesis was that efficiency of the ED process is directly dependent on cell applied voltage and feed initial concentration. Therefore, impact of these operating parameters was investigated on ED performances including the electrical conductivity reduction and removal efficiency of COD. In order to have better evaluation, ED energy consumption, as a limiting and crucial factor, was also studied. The results showed that applying ED can be considered as an optimal approach for reduction of EC and COD of wastes samples. By increasing the applied voltage and decreasing initial concentration of feed solution, both EC and COD reduction efficiency improved. As voltage increased from 5 to 20 volt, EC and COD reduction efficiency reached to their maximum values of 92% and 81%, respectively in the Cf concentration. In addition, as the parameter of initial concentration decreased from Cf to Cf/4, EC and COD enhanced and reached to values of 92% and 85%, respectively in the voltage of 15. As an economical factor, although using higher voltages increase the efficiency of ED process, but there is a limitation for that since it simultaneously increases the total energy consumed by the device.

5. Nomenclature

Cf	lithium ions concentration in the feed solution (mol)
Cf/4	lithium ions concentration in the feed solution (mol)
$\eta\%$	COD removal efficiency percentage
COD_0	Chemical oxygen demand at time zero (mg/L)
COD_t	Chemical oxygen demand at time t (mg/L)
E	Energy consumption (W.hr/L^{-1})
V	Applied voltage (V)
I	Cell current (Amp)
t	Operation time (s)
V_D	Initial volume of diluted compartment (L)
EC_0	Electro conductivity of feed solution at time of zero (ms/cm)
EC_t	Electro conductivity of feed solution at time of t (ms/cm)
ED	Electrodialysis
EC	Electrical Conductivity
COD	Chemical Oxygen Demand
RO	Reverse Osmosis

NF	Nanofiltration
IEM	Ion Exchange Membranes
CEM	Cation Exchange Membrane
AEM	Anion Exchange Membranes
LC	Limiting Current

7. References

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