The impact of electrode material in the treatment of landfill leachate by electrocoagulation process

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Abstract. This paper presents the results of the electrocoagulation treatment of leachate by the application of iron and aluminum as an electrode material. It was examined the influence of the current density and electrolysis duration on the removal efficiency of total nitrogen (N). Duration of electrolysis was 30 minutes, and based on the experimental results it was found that the optimal current density is 25 mA/cm². The highest removal efficiency of total nitrogen, achieved under these conditions, amounts 29.58% by the application Fe-Fe electrode pair, while removal of 7.70% is achieved by application Al-Al electrode pair.

1. Introduction

In addition to the municipal and industrial wastewater generated in various technological processes, landfill leachate require special attention and treatment because of the enormous impact on the quality of the environment. Disposal of waste in the form of landfill is still the cheapest and most widely used waste treatment in the world [1].

Sanitary landfill can be defined as a method of disposing of refuse on land without creating nuisances or hazards to public health or safety, by utilizing the principles of engineering to confine the refuse to the smallest practical area, to reduce it to the smallest practical volume, and to cover it with a layer of earth at the conclusion of each day’s operation or at such more frequent intervals as may be necessary [2]. Leachate is formed by draining rainwater through the landfill body, in which occurs extraction of soluble colloidal and suspended matter from the waste. While sanitary landfills are often economically and environmentally friendly methods for the disposal of solid waste, the leachate water that emerges poses a threat to the environment, as it can carry toxic pollutants into groundwater and surrounding land [3].

Leachate from landfills are one of the most toxic type of wastewaters, as well as in terms of the selection of appropriate techniques for their purification. It is particularly difficult to process due to its complex and variable composition. All waters generated during landfill operations according to the EU Directive 1999/31/EC on Waste Disposal [4], as well as the Waste Directive 2006/12/EC, should be collected and treated before any discharge into the final recipient [5].

Research has shown that each landfill is a separate system, and in this sense, the composition and the amount of leachate depend only on the characteristics of the landfill [6]. For the treatment of landfill leachate there are a variety of technical and technological solutions. The treatment strategy generally depends on the characteristics of the leachate. Young landfill leachates are generally easier to treat compared with old ones. Leachate from the old landfills contain less organic biodegradable...
matter and more toxic components, so the purification of the water should be used a combination of several different processes. The quality and quantity of the filtrate, the size of the landfill and the lifetime, the required quality of the effluent with economic analysis are the basic elements that define the selection of the method for treatment of these waters [7].

1.1. Electrochemical methods (literature review)
Electrochemical processes, such as electrocoagulation (EC), electrooxidation (EO), electroreduction and electro-photo-oxidation, are often used for treatment of wastewaters from various industries such as textile, leather, oil industry, and recently have been used in the treatment of leachate.

The EC process involves the in situ generation of coagulants through the electro-dissolution of a sacrificial anode, which is usually made of iron or aluminium. EC is one of the electrochemical methods used for treatment many types of wastewater. Numerous studies describe the application of this method for treatment of wastewater, including leachate.

Mussa et al. performed decolorization of leachate by researching the efficiency of EC. Several different parameters were tested, such as electrode material (Ni, Ag, Cu, Pd, Pt, Stainless Steel, C), experiment duration, voltage and chloride concentration. Good color removal (70%) was achieved using carbon in 120 minutes of treatment. It has been found that the electrochemical method of EC can be used as a pre-treatment for the leachate treatment [8]. Norma et al. applied a combination of electrochemical techniques, EC and EO. They found that these techniques are cost-effective for treatment of leachate from sanitary landfills [9]. Electrochemical methods have long been used to remove not only suspended solids, but also the chemical oxygen demand (COD) and ammonia nitrate (NH$_3$-N), and can provide good color removal [8]. Ilhan et al. researched the application of EC for the treatment of leachate using aluminum and iron electrodes. It has been shown that EC has higher purification effects from the chemical coagulation process and can be successfully used [10]. Xiangdong et al. examined the EC efficacy to remove NH$_3$-N and COD from leachate. The treatment was preform in a batch electrochemical reactor. The experimental conditions were as follows: current density 4.96 mA/cm$^2$, chloride concentration 2319 mg/L, electrolysis duration of 90 minutes. Treatment was carried out with iron electrodes, where the highest removal efficiency of COD and NH$_3$-N was achieved, 49.8% and 38.6% respectively [11]. In their research, Bouhezila et al. used EC to remove COD, total nitrogen (N$_{tot}$), color, turbidity and bacteria from leachate, characterized by high COD, high nitrogen concentration and dark color. It was found that increasing the current density (125-500 A/m$^2$) significantly accelerates the process, with a distance between the electrodes of 2.8 cm and a stirring speed of 150 rpm. The removal efficiencies of COD, N$_{tot}$, color and turbidity was 68%, 15%, 28% and 16% respectively, when iron electrodes are used. The consumption of electricity was 0.0196 kWh/L [12]. Similar to the research of Ilhan et al., in the research of Yadav et al. also was used iron and aluminum electrodes for EC treatment of leachate. The aim of the paper was to optimize parameters such as current density, electrolysis duration and pH value to achieve maximum COD removal. By using iron electrodes, COD removal efficiency was 56% at a current density of 466 A/m$^2$. Optimum electrolysis time was 90 minutes and a pH 6. The iron electrode improved higher COD removal efficiency in comparance with an aluminum electrode (47%). EC was assessed as a good pre-treatment [13].

2. Material and methods
The experimental part of the research consists in the application of EC treatment for purification of leachate. Sample of leachate was taken from the regional sanitary landfill - "JP Regional Landfill DEP-OT," Ramici, Banja Luka. Leachate drainage into pipes leading into the tank (Figure 1), and then in the reverse osmosis plant for treatment of landfill leachate.
Experimental setup is shown on the Figure 2. The batch reactor for EC of 500 cm³ capacity made from polypropylene with constant mixing (400 rpm) was used, combined with two electrodes of the same area surface. The dimensions of the electrodes are 40x40x1 mm. The total useful surface was 95.76 cm², and the distance between electrodes 3 cm. Electrodes were connected to digital power source (Atten, APS3005SI; 30V, 5A).

All the experiments were performed at an ambient temperature, with the leachate initial volume of 400 cm³ and with concentration of supporting electrolyte $\gamma_{NaCl} = 5$ g/L. As an electrode material was used iron, Fe (steel - EN 10130-91; max. 0.08% C, max. 0.12% Cr, max. 0.45% Mn, max. 0.60% Si) and aluminum, Al (aluminum - Al 99.5/ENAW-1050 A; max. 0.25% Si, max. 0.40% Fe, max. 0.05% Cu, max. 0.05% Mn, max. 0.05% Mg, max. 0.05% Ti, max. 0.07% Zn, min. 99.50% Al), which meet the required standards. After each EC treatment leachate was collected and filtered. The samples of leachate before and after treatment were analyzed on the content of $N_{tot}$ in accordance with standard method [14]. The determination of total nitrogen was carried by the persulphate method in standard cuvettes (Lovibond 53 55 50, Germany).

3. Results and discussion
In this paper has shown a part of results of case study for the Ramici regional landfill in Banja Luka, Bosnia and Herzegovina [15]. Results of the electrochemical treatment of landfill leachate are shown through removal efficiency, $E$ (%), whose ratio could be described by following equation:
\[ E = \frac{\gamma_i - \gamma_f}{\gamma_i} \times 100\% \]  

(1)

where \( \gamma_i \) and \( \gamma_f \) the initial and the final value of parameter which efficiency is determined.

The following figures show the impact of current density on removal efficiencies of \( N_{\text{tot}} \) for Fe-Fe electrode pair (Figure 3) and Al-Al electrode pair (Figure 4). In this experiment was used current densities of 5, 10, 25 mA/cm².

Figure 3 shows that the highest removal efficiency is achieved at a current density of \( j=25 \) mA/cm² \( (E=29.58\%) \). Although it was expected that the increase in the current density would lead to a constant increase in efficiency, the figure shows a decrease in efficiency. The efficiency achieved at a current density of 10 mA/cm² \( (E=10.28\%) \) is lower than when the current density is 5 mA/cm² \( (E=16.40\%) \).

For case of aluminium electrodes the highest removal efficiency was achieved at the lowest current density, \( j=5 \) mA/cm² and amounts 22.41\%, and at \( j=25 \) mA/cm² amounts 7.70\% (Figure 4). It can be seen that the iron electrode has a slightly greater removal efficiency compared to the aluminium electrode for 30 minutes of treatment. In further experiments it was applied current density of 25 mA/cm².

![Figure 3](image1.png)

**Figure 3.** Removal efficiency in relation to the current density by using iron electrodes \( (\gamma_{\text{NaCl}}=5 \text{ g/L}, t=30 \text{ min, stirring 400 rpm}) \)

![Figure 4](image2.png)

**Figure 4.** Removal efficiency in relation to the current density by using aluminum electrodes \( (\gamma_{\text{NaCl}}=5 \text{ g/L}, t=30 \text{ min, stirring 400 rpm}) \)
Figures 5 and 6 shows the impact of electrolysis duration (10; 20; 30 min) on the N\textsubscript{tot} removal efficiency. The highest removal efficiency for iron electrodes was achieved at the electrolysis duration of 20 minutes and amounts 40.59% (Figure 5). For aluminium electrode by extending of electrolysis duration, the removal efficiency decreases. The highest removal efficiency for was achieved at 10 minutes of electrolysis and amounts 16.39% (Figure 6).

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{iron.png}
\caption{Removal efficiency in relation to the electrolysis duration by using iron electrodes (γ\textsubscript{NaCl}=5 g/L, j=25mA/cm\textsuperscript{2}, stirring 400 rpm)}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{aluminum.png}
\caption{Removal efficiency in relation to the electrolysis duration by using aluminum electrodes (γ\textsubscript{NaCl}=5 g/L, j=25mA/cm\textsuperscript{2}, stirring 400 rpm)}
\end{figure}

4. Conclusions
It can be concluded that at lower current densities removal efficiency of N\textsubscript{tot} was higher for aluminum electrodes, while at higher current density and longer electrolysis duration the removal efficiency is greater for the iron electrode. From the results we can also see that with relatively low current density and short treatment time, good purification results have been achieved. Therefore, electrocoagulation can be successfully applied as a pre-treatment for landfill leachate. In further studies it is necessary to consider the application of post treatment, such as electro-oxidation.
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References
[1] Ismail S and Manaf L A 2013 The challenge of future landfill: A case study of Malaysia, Journal of Toxicology and Environmental Health Sciences 5(6) 86-96
[2] Ragh M S, El Meguid A M and Hegazi H A 2013 Treatment of leachate from municipal solid waste landfill, HBRC journal 9 187-192
[3] Llabanowski J, Pallier V and Feuillade-Cathaliufad G 2010 Study of organic matter during coagulation and electrocoagulation processes: Application to a stabilized landfill leachate, Journal of Hazardous Materials 179 166-172
[4] ***Council Directive 1999/31/EC European Union (1999)
[5] ***Directive on waste 2016/12/EC European Union (2006)
[6] Ehrig H and Robinson H 2010 Landfilling: Leachate treatment, in Solid Waste technology & Menagement, Volume 1 & 2 (ed T H Christensen), Chichester, UK: John Wiley & Sons, Ltd.
[7] Fudala-Ksiazek S, Luczkiewicz A, Quant B and Olanczuk-Neyman K 2011 The effectiveness of nitrification and denitrification with CO-treatment wastewarer with landfill leachate, Thirteenth International waste management and landfill symposium, Cagliary, Italy: IWWG-International waste working group, pp 321-329
[8] Mussa Z H, Othman M R, Abdullah P and Nordin N 2013 Decolorization of landfill leachate using electrochemical technique, International Journal of Chemical Science 11(4) 1636-1646
[9] Norma D, Fernandes A, Pacheco M J, Ciriaco L and Lopes A 2012 Electrocoagulation and Anodic Oxidation Intregated Process to Treat Leachate from Portuguese Sanitary Landfill, Portigalie Electrochimica Acta 30(3) 221-234
[10] Ilhan F, Kurt U, Apaydin O and Talha Gonullu M 2008 Treatment of leachate by electrocoagulation using aluminum and iron electrodes, Journal of Hazardous Materials 154 381-389
[11] Xiangdong L, Junke S, Jiandong G, Zhichao W and Qiyan F 2011 Landfill leachate treatment using electrocoagulation, Procedia Environmental Sciences 10 1159-1164
[12] Bouhezila F, Hariti M, Lounici H and Mameri N 2011 Treatment of the OUED SMAR town landfill leachate by an electrochemical reactor Desalination 280(1) 347-353
[13] Yadav J S and Dikshit A K 2017 Stabilized Old Landfill Leachate Treatment Using Electrocoagulation, Environment Asia 10(1) 25-33
[14] ***APHA. Standard Methods for the Examination of Water and Wastewater, New York: American public healt association, 1999
[15] Malinovic B N, Pavlovic M G, Djuricic T, Bjelic D and Neskovic Markie D 2018 Electrochemical treatmaent of leachate: case study for sanitary landfill “Ramici”, Banja Luka, XX International Conference YuCorr, Tara Mountain, Serbia, May 21-24, pp 16-27