Electrocoagulation of Landfill Leachate with Monopolar Aluminum Electrodes

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Abstract—In this paper, the removal of COD from landfill leachate using aluminum electrode by the electrocoagulation method was investigated. The studies were run with the parallel plate monopolar aluminum electrodes and the effect of pH and current density on removal efficiency of COD and energy consumption were determined. The initial COD concentration of 4100 mg/L was reduced to 1763 mg/L with the removal efficiency of 57% at the current density of 75 mA/cm² and pH 5.

Index Terms—Aluminium electrode, landfill leachate, treatment.

I. INTRODUCTION

The most common and desirable integral indispensable solid waste management strategy is sanitary landfilling because it is simple and has low exploitation and capital costs. Up to 95% of the total municipal solid waste collected in the world is disposed of in landfills [1]. Physico-chemical and biological conversions are occurred during the landfilling process and a result of conversions degenerated organic compounds that soluble in rainwater are formed. Any liquid material that drains from land material is called “leachate” and it is highly toxic liquid with dissolving organic compounds, heavy metals and different soluble materials derived from the material that it has passed through. The composition of leachate from a landfill depends on the age of the landfill and the type of waste that it contains. Leachate can potentially contaminate nearby surface and ground water if left untreated. Therefore landfills require close environmental monitoring during their design, operation, and long-term post-closure period.

Leachate must be treated before discharging to environment. Many different physico-chemical techniques such as adsorption [2], chemical precipitation [3], coagulation/flocculation [4], chemical oxidation [5] and biological methods [6] have been being applied for the treatment of landfill leachate. Electrocoagulation, one of the most useful electrochemical methods, is a comparatively new technique for wastewater treatment. Also, electrocoagulation has some advantages such as, short retention time, easy operation, not needed the chemical addition, simple equipment, high sedimentation velocity and possibility of complete automation [7].

Metal complexes further react with negatively charged particles in the solution to form flocs. The pH and ionic strength of solution determines the type and solubility of metal hydroxide complexes formed. Insoluble flocs of Al(OH)₃ are generated at pH range between 6.0 and 7.0. However, depending on the pH of the aqueous medium other ionic species, such as Al(OH)₃⁵⁺, Al₅(OH)₄⁴⁺ and Al(OH)₆ can also be present in the system [10], [11].

The aim of this study was to treat the landfill leachate by electrocoagulation (EC). In this purpose the parallel plate aluminum electrodes were used. This study elucidates the effects of parameters such as: initial pH and current density on the COD removal efficiency for a batch EC reactor. Information regarding the electrical energy consumption (EEC) is also included to provide an estimation of the cost of COD removal by an EC system.

II. ELECTROCOAGULATION

Electrocoagulation is based on the electric charge stability of colloids, emulsions and suspension. It is known that, particles are charged neutral in the wastewater. However, the surface charge of the particles tends to be neutralized, opposite charged particles combine and make larger floccules, when the additional electric charges are provided to the particles with appropriate electrodes [8].

In electrocoagulation process aluminum or iron electrodes as sacrificial anodes are dissolved to produce their cations upon the application of a direct current. The metal ions generated are hydrolyzed in the electrochemical cell to produce metal hydroxide ions according to reactions (1)-(3) [9].

\[
\begin{align*}
\text{Al} & \rightarrow \text{Al}^{3+} + 3e^- \quad (1) \\
\text{Al}^{3+} + 3\text{H}_2\text{O} & \rightarrow \text{Al(OH)}_3 + 3\text{H}^+ \quad (2) \\
n\text{Al(OH)}_3 & \rightarrow \text{Al}_n\text{(OH)}_{3n} \quad (3)
\end{align*}
\]

A. Leachate

The leachate was collected from landfill in İzmit/Turkey. In this landfill area 3.106.028 tons of domestic and household waste, 297.485 tons of domestic industrial waste were received between 1997-2011. The leachate wastewater taken from this area was stored at refrigerator and used without any pre-treatment. It has strong odor and dark brown/black color with a pH of 9 and COD concentration of 4100 mg/L.
B. Bench Scale Batch EC Apparatus

Experimental set-up was consisted of DC power supply (Statron 2257), magnetic stirrer (IsoLab MS-010) and EC reactor. In the electrochemical cell, six aluminum plates (dimension 4 cm × 4.5 cm) with the anode surface area of 108 mA/cm² were used as electrodes. The electrodes were connected using a monopolar configuration which three aluminum plate operated as anode in the electrocoagulation reactor. The detail of reactor is shown in Fig. 1. The reactor was dipped in wastewater in glass beaker. Also, beaker was dipped into water-bath to stabilize temperature increase.

C. Experimental Procedure

In this study, 0.4L wastewater was poured into the electrochemical cell and electrocoagulation experiments were performed for 60 min for each run. Samples were taken every 15 min interval from the electrocoagulator, filtered and analyzed to determine COD concentrations using Close-Reflux Methods.

All the samples were analyzed in duplicate to ensure data reproducibility, and an additional measurement was carried out, if necessary. The calculation of removal efficiency (RE%) after EC was calculated using the equation

\[ RE\% = \left(\frac{C_0 - C}{C_0}\right) \times 100 \]  

where \( C_0 \) and \( C \) are the concentrations of COD before and after EC, respectively, in mg/L.

Additionally, energy consumption (\( E_c \); kWh/m³) was calculated using following equation;

\[ E_c = V \times I \times T / \nu_w \]  

where \( V \) is Voltage (V), \( I \) is Current (A), \( T \) is operation time (h),and \( \nu_w \) is the volume of the wastewater (m³).

IV. RESULTS AND DISCUSSION

A. Effect of Initial pH

PH has an important role in the EC process. The effect of initial pH was studied in the ranges of 3-9 at the current density of 30mA/cm². The initial pH of the wastewater was adjusted using 1N H₂SO₄. In all experiments, pH was not controlled but monitored during operation. Although pH increases during EC process due to hydroxyl ions release.

The effect of pH on removal efficiency is presented in Fig. 2. After 60 minutes EC removal efficiencies were 46%, 48%, 42%, and 40% at the pH of 3, 5, 7, and 9, respectively. The best removal efficiency was obtained at pH 5 because the hydrolysis of Al³⁺ depends on pH. At higher pH, the dominant compound is Al(OH)₄⁻, which is not able to coagulate with pollutants [12], [13].

The effect of pH on energy consumption is shown in Fig. 3. Energy consumption is expressed with kWh per m³. Electrical conduction is a phenomenon in which is able to carry electricity. The decreasing pH of solution causes to increase in conductivity. So, when the conductivity increases, ability to carry electricity is also increased and low energy needed. That can explain the relation between pH and energy consumption [14]. The energy consumptions were 42, 52, 49 and 125 kWh/m³ at pH of 3, 5, 7, and 9, respectively after 60 minutes EC operation.

B. Effect of Current Density

Current density is defined as current applied per unit surface area of the electrode. Current density is the operational parameter which is the easiest to control and determines coagulant dosage and bubble generation rates. According to Faraday’s law (Eq. 6), the amount of ions released from anode increases with increasing current (I).

\[ m = I \times t \times M / (z \times F) \]  

where \( m \) is the mass of generated metal ions (gram); \( I \) is the current(Ampere), \( t \) is the operation time (min,h); \( M \) is the atomic weight of metal (g/mol), \( z \) is the number of electrons transferred in the anodic dissolution (z=3 for Aluminum), and \( F \) is Faraday’s constant(96 486 C eq⁻¹).

The effect of current density was determined at the pH of 5 and results can be seen from Fig. 4. As seen from Fig. 4 the removal efficiencies were not change significantly after 15 min. of EC. The removal efficiencies of 49%, 47%, 50%, 51%, 50% were obtained after 15 minutes EC at the current
densities of 30, 45, 55, 65, 75 mA/cm², respectively. Further electrocoagulation to 45 min the removal efficiencies of 48%, 47%, 48%, 52% and 57% were obtained at the current densities of 30, 45, 55, 65, 75 mA/cm², respectively.

The energy consumption depends on time as well as the current and potential that are applied (Eq. 5). As the reaction time increased, the system energy consumption also increased. As it can be seen from Fig. 5 increasing current resulted with increasing energy consumption. After 15 minutes EC the electrical energy consumptions of 5, 10, 13, 16, 26 kWh/m³ were obtained current densities of 30, 45, 55, 65, 75 mA/cm², respectively. As the time proceeds the final COD concentration did not change significantly beyond 15 minutes. Therefore the long reaction time was not preferred from an economical viewpoint.

**Fig. 4. Variation of removal efficiency with time at different current densities.**

**Fig. 5. Variation of electrical energy consumptions with current densities.**

V. CONCLUSION

Based on the results achieved from the experiments, the following conclusions may be outlined:

1) Electrocoagulation with aluminum electrode could be applicable for the treatment of landfill leachate.

2) The treatment of landfill leachate by electrocoagulation was found to be pH dependent. The highest removal efficiency was achieved at pH 5.

3) The removal efficiencies increased by increasing the current density while increasing the current density caused the energy consumption to increase. The energy consumption also depends on time. The removal efficiency of 51% was obtained with the energy consumption of 16 kWh/m³ after 15 min electrocoagulation.

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