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POLLUTANT REMOVAL FROM LEACHATE BY ELECTROCHEMICAL TREATMENT: A CASE STUDY FOR SANITARY LANDFILL BALIKESIR

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ABSTRACT

This paper presents a comparison of alternative sacrificial electrodes for the electrocoagulation (EC) process that can be applied in the treating of leachate of Balikesir Landfill. The purification performance of the process was determined by monitoring COD (Chemical Oxygen Demand) values remaining in solution as a result of electrocoagulation process of leachate containing many different pollutants in a full batch reactor. Test samples were collected from municipal solid waste landfill in Balikesir. Three values for current density ($CD=196, 392$ and 588 A/m^2), three electrode materials (aluminum, iron and zinc) and treatment time were taken into consideration in the scope of this study. Optimum current density was 196 A/m^2 and the most suitable electrode material was aluminum (53% COD removal efficiency). Besides, cost analysis of the process was assessed according to energy consumption. Experimental findings implied that electrocoagulation process could also be applied employing alternative electrode materials like zinc and iron even though the most economical electrode type was aluminum. Considering all results, it was concluded that the electrocoagulation process is highly applicable to treat the leachate originated from Balikesir landfill.

KEYWORDS:

Electrocoagulation, leachate treatment, iron/aluminum/zinc electrode, COD removal.

INTRODUCTION

Wastes resulting from the rapid increase in population and consumption in urban areas are accumulated in landfills. Landfill in many parts of the world stands out as a cheap and simple-to-operate waste disposal method compared to other methods [1]. Leachate is an end product that must be reconsidered at landfills. Leachate, which has a complex composition, is formed by percolating the rain water through the wastes in the landfill [1,2]. Leachates originated from landfill may contain large amount of both biodegradable and resistant organic matter with

predominance of humic substances [3], heavy metals, chlorine-containing organic compounds and inorganic salts [4]. Parameters such as Biochemical Oxygen Demand (BOD), heavy metals, Chemical Oxygen Demand (COD), pH, Total Organic Carbon (TOC) and ammonical nitrogen (NH_3-N) can be traced to inspect the leachate generated from the landfill [5,6]. In view of its complex composition, landfill leachate has been considered as a potential contamination source for the surrounding area, if it is not properly collected, treated and safely disposed [7]. Therefore by today, various wastewater pretreatment and combined treatment methods have been documented to treat the leachate such as biological treatment, physicochemical treatment, advanced oxidation treatment, and leachate recirculation. These treatment methods are substantially useful, but have one or more limitations. Leachate must be young, not old in order to implement the biological treatment efficiently [8,9]. Advanced oxidation methods have high economic costs and points of chlorine oxidation [10]. Although there are limitations such as saturation and slowing of metabolic activities, recirculation of leachate is one of the cheapest methods [11]. There are also other leachate treatment methods like ozonation [12], anaerobic filters [13], coagulation-flocculation [14] and membrane processes [12,15-17]. However, problems such as high cost, experimental difficulty and post-process contamination limit the industrial applications of these processes [18]. Briefly, both economic and domestic treatment methods should be developed to clean contaminated water. Among these methods, electrochemical treatment has become more preferred day by day. Electrocoagulation, one of the electrochemical treatment techniques, has emerged as a simple, economical and efficient water treatment process [19]. In addition, electrocoagulation is an easy, low equipment and maintenance-repair cost and chemical-free treatment process. Over and above, it reduces the secondary pollution load by producing less sludge than the traditional chemical coagulation/flocculation process [20-22]. In the electrocoagulation system, employed anodes dissolve and metal ions are released into the liquid medium. Synchronously, the water contained in the treatment medium is hydrolyzed in the cathode resulting in hydroxyl ions, and hydroxyl ions and metal ions form metal hydroxide. The main factors

affecting the solubility of complexes in the treatment environment are pH and ionic strength. Metal components that react with negative charged species form flocs, and these flocs produced as a result of flocculation destabilise and aggregate the suspended particles in order to precipitate or adsorb dissolved contaminants [23].

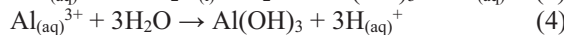
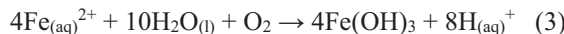
A number of authors presented the theoretical aspects of electrocoagulation process [24,25]. In accordance with the authors, electrocoagulation process involves three sequential stages: (a) coagulant generation by electrolytic oxidation of the metal electrode; (b) contaminant destabilization, granular suspension, and emulsions' breaking; (c) aggregation of the destabilized phases to form flocs [20]. By monitoring the solution electrochemistry and electrical conductivity, it may be possible to provide an explanation of the mechanisms of reactions during the electrocoagulation process. The production mechanism of ions, which will initiate pollution removal in the treatment process, is provided by metal electrodes such as iron and aluminum. In the electrochemical purification cell, iron hydroxide is produced as a result of the chemical reactions initiated by the electrical energy conducted to the iron plates used as electrodes. From this point of view, the formation of metal hydroxide has been described in detail by the evaluation of two alternative mechanisms [20,26]. The reactions are given in Eqs. 1-11.

Mechanism 1

Anode



Chemical



Cathode

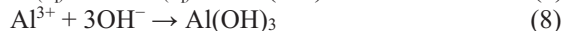


Mechanism 2

Anode



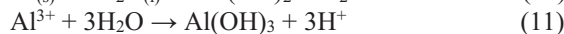
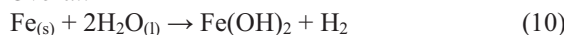
Chemical



Cathode

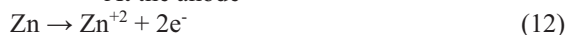


Overall



The possible reactions that may occur when zinc is used as electrode are presented in Eqs. 12-15 [27].

At the anode



At the cathode



In the solution



Global reaction



Based on the existing literature, several electrocoagulation studies on the treatment of leachate have been frequently performed by using Al or Fe electrodes. However, the formation of more sludge and cost of electrodes are disadvantages of the use of these sacrificial electrodes [28], therefore the search for new electrode materials to promote the electrochemical formation of coagulants is encouraged.

This study focuses on the use of three different electrode materials (Fe, Al, and Zn) to remove pollutants in the leachate of Balıkesir landfill and comparing the electrochemical treatment performance of employed electrodes. In order to evaluate the treatment performance, effects of various operating conditions such treatment time, electrode material and current density were discussed, and the best operating conditions were determined to verify the possible real application of such technology. Besides COD as major pollution index, turbidity, conductivity and electrical energy consumption values were also assessed.

MATERIALS AND METHODS

Fresh landfill leachate used in the experimental study was collected from Balıkesir Landfill (Balıkesir, Turkey) serving for 500 tonnes on an average municipal solid waste in a day. This is still an active waste storage area with about 70 m³ of leachate per day. Leachate samples were collected from the stabilization lagoon without any pretreatment and kept in the refrigerator at 4 °C in dark polyethylene containers to maintain initial properties until the experiment was carried out. Some average properties of the raw leachate sample are given in Table 1. Standard Method procedures were used as reference in the COD and turbidity analysis of the experiments [29]. COD analyses were carried out in thermoreactor Spectroquant Pharo 300 by carrying out the closed reflux titrimetric method. The pH and conductivity of the samples were measured with a WTW 315i apparatus. Turbidity was monitored with WTW S12. Merck analytical grade chemicals were used in the preparation of reagents.

A schematic representation is shown in Figure 1. A cylindrical plexiglass (0.8 L capacity) was used as batch EC reactor. One pair parallel bounded electrode (made of aluminum, iron or zinc) with dimension of 20 cm x 6 cm x 0.2 cm was applied as anode and cathode and, the total effective area of each electrode is calculated to be (8.3 cm x 6 cm) 50 cm². The distance between cathode and anode influencing the electrical energy requirement during the test was 5.5 cm. The electrodes were immersed in the electrocoagulation reactor with a leachate volume of 0.5

L and connected to a DC power supply (TTT-ECHNI-C RXN-3010D) providing a controlled voltage up to 30 V and regular electricity current up to 3 A. Because the electrical conductivity of the leachate sample was sufficient for electrocoagulation, no salt addition was used to support of the electrolytic medium. All electrochemical COD abatement tests were followed at 15, 30, 45 and 60 minute-intervals. The solution was mixed via a Yellowline MST Basic magnetic stirrer by the speed of 250 rpm which was optimum stirring rate in the literature [30]. The electrolysis was conducted at room temperature under different current densities and natural pH = 8.9. The effect of electrode type was studied with three conventional electrodes (Al, Zn and Fe). The metallic electrodes and EC cell were thoroughly irrigated with aqueous HCl solution and deionized water prior the next operation in order to get passivation under control. After electrolysis, the suspension withdrawn from the supernatant was filtered before being analyzed.

TABLE 1
The properties of collected leachate from Balıkesir Landfill

Parameter	Value
pH	8.9
COD (mg/L)	3668
BOD ₅ (mg/L)	4700
BOD ₅ /COD	1.28
Turbidity (NTU)	106
Sulfate (mg/L)	1079
Chloride (mg/L)	2172
Phosphate (mg/L)	23
Nitrate (mg/L)	32
Electrical Conductivity (mS/cm)	14.02 (at 22 °C)

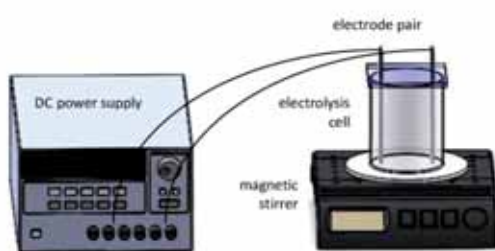


FIGURE 1
Schematic figure of experimental set-up

In order to evaluate the effect of the parameters selected in the study on the EC process, the removal efficiency (RE) of the electrochemical treatment applied to the landfill leachate was taken into account: it is the difference between C_0 and C concentrations of the chemical oxygen demand before and after electrocoagulation divided by C_0 .

$$RE\% = \frac{[C_0 - C]}{C_0} \times 100 \quad (16)$$

Additionally unit electrical energy consumption was also calculated in terms of kWh/m³ to present economic and environmental impacts of the process according to Eq. (17) where E is the electrical energy consumption, U is the potential (V), I is the electrical current (A), t is the time (h), V is the volume of the solution treated (m³), respectively [27].

$$E = \frac{UIt}{V} \quad (17)$$

RESULTS AND DISCUSSION

The BOD₅/COD ratio, which is one of the parameters of physicochemical characterization of the leachate depurated with electrochemical process, presented in Table 1 assigns a meaning of the rate of biodegradable organic matter in leachate employed in the process. Young leachate usually has a higher BOD₅/COD (> 0.5) ratio compared to the leachate from an older or stable landfill (< 0.2). As indicated in the table, the BOD₅/COD value expressing the degree of biodegradability of the leachate studied is quite high (BOD₅/COD=1.28). The characteristics of the leachate formed as a result of decomposition of municipal wastes in the landfill vary according to the age of the storage and affect fairly the treatment performance. The most important operating parameters in electrochemical processes are electrode material type, cell current and electrolysis time. Accordingly, the effects of these parameters on the pollution removal performance of the electrocoagulation process were examined in terms of final COD and percent COD removal efficiency within the scope of the study. To summarize the results, a significant reduction in COD concentration was achieved with aluminum electrodes and current density of 196 A/m².

Influence of treatment time. In the process of electrocoagulation, the critical parameter that determines the financial and environmental needs of the treatment plants is the time spent on the operation. Processing time is a major factor that guides the determination of the number of metal ions transferred to the wastewater to be treated. In short, it is the time to be processed to ensure the production of various metal hydroxide structured-polymeric phases that incorporate contaminants contained in the aqueous solution phase. In this study, samples were taken from the bulk solution at 15-minute intervals and COD values were analyzed to determine the effect of electrolysis time on process efficiency. Most of the graphs are based on the time of treatment in the x-axis, so the impact of the treatment time can be inspected on most figures. The most effective removal capacity was achieved for 45 min operation with the

efficiency of 53% employing Al electrodes at current density of 196 A/m² and, 34% for Zn electrodes at current density of 588 A/m² and 24% for Fe electrodes at current density of 588 A/m². The findings confirm the Faraday's law which says that the amount of metal released into the solution linearly increase with reaction time. The deceleration in COD removal rate after 45 minutes of electrocoagulation is due to the reduction of the pollutant concentration in the leachate. As can be seen in Figure 2a, an increase in time from 15 to 45 minutes yield an increase in the efficiency of COD removal from %39 to %53 for Al electrodes, from %28 to %34 for Zn electrodes and from %2 to %24 for Fe electrodes. It is striking to note that electrocoagulation system accomplished a remarkably high removal efficiency of COD for this optimum reaction time. As the time spent for removal in the electrocoagulation process increases as the amount of electricity energy usage and cost increases, so the most efficient and economical working time can be selected as 45 minutes.

Compared to the literature, 45 min contact time with Al electrode yields quite high COD removal efficiency [31,32]. Besides Al, electrocoagulation with Zn and Fe electrode could be carried out as alternative electrode material.

When the electrolysis time gets along, the concentration of iron ions and iron hydroxide flocks increases and also the formation of bubbles is accelerated [31]. Similarly, other pure metal ions produced from aluminum and zinc electrodes are transferred from the electrodes forming the hydroxide flocks to the aqueous phase in the EC cell. Thus leachate impurities are eliminated by the effect of coagulation and flotation of the electrocoagulation process.

Influence of electrode materials. Materials of electrodes formed from different metals affect the performance of the electrocoagulation process [33]. Aluminum and iron are easy materials to obtain and often preferred, as well as low-priced, high-efficient and accessible electrode options [34]. In addition to these common electrode types, Zn metal electrode was also used in this study. Figures 2a-c shows the COD removal efficiency versus electrocoagulation time for Fe, Al and Zn electrodes at pH=8.9. As seen, Al electrodes exhibited better COD removal efficiency than Fe and Zn electrodes. This can be regarded as a natural consequence of the increased release of Al ions from the Al cathodes to the solution, and hence more of the flocks produced in the bulk. Aluminum ions are cathodically polarized because of a phenomenon called "chemical dissolution" or "cathodic corrosion" [35,36].

Since the cost of all three electrodes is almost the same, it is more convenient to employ aluminum

metal as electrodes to achieve higher pollutant removal efficiency in the electrocoagulation process. [32]. Figures 4a-c illustrate the change in time versus COD removal efficiency for three types of electrodes at current densities of CD=196 A/m², CD=392 A/m² and CD=588 A/m², respectively. As seen, Al electrode showed better performance compared to Fe and Zn electrode at minimum current density value (196 A/m²). Surprisingly, Zn electrode was more effective in the removal of COD than Al and Fe at higher current density values.

Influence of current density. In order to determine the current density, the current applied in the electrochemical process is proportional to the unit surface area of the electrode used, and the unit of this ratio is A/m². Current density stands out as a major factor that determines the required coagulant amount in the electrocoagulation process. As the electrode material dissolves, the amount of coagulant increases, that is, the removal performance and electricity consumption increases.

Figures 3a-c demonstrates the performance of EC process in terms of decrease in COD values for different current density values by applying aluminum, zinc and iron electrodes, respectively. As the treatment period progresses, COD values of the leachate decreased. This change in all three electrodes is clear evidence that electrocoagulation is a feasible process for pollutant removal.

Figures 4a-c shows COD removal (%) from landfill leachate by EC process with aluminum, zinc and iron electrodes for different current densities at pH = 8.9. As shown, the current density and the amount of pollutant removal increased in direct proportion during the process. This is consistent with the results of other researchers [31,32,37]. However, there may be no improvement in the pollutant removal after a certain current density value [38]. Also it is expected that current density exerts a major effect on reaction kinetics and energy consumption of an EC process [39,40].

In the EC process, current is applied between the metallic electrodes immersed in the polluted water. When electric current is applied to the electrodes, the electrode first dissolves, and then the coagulant species, which destabilise and collect the pollutants in the wastewater, begin to form. Additionally, it determines the rate of hydrogen bubbles formation and the growth of flocs that may influence the efficiency of the process [41]. The performed current densities in this study were 196, 392 and 588 A/m². Because low current density means low electrical energy consumption, 196 A/m² current density could be recognized as optimum value for this process.

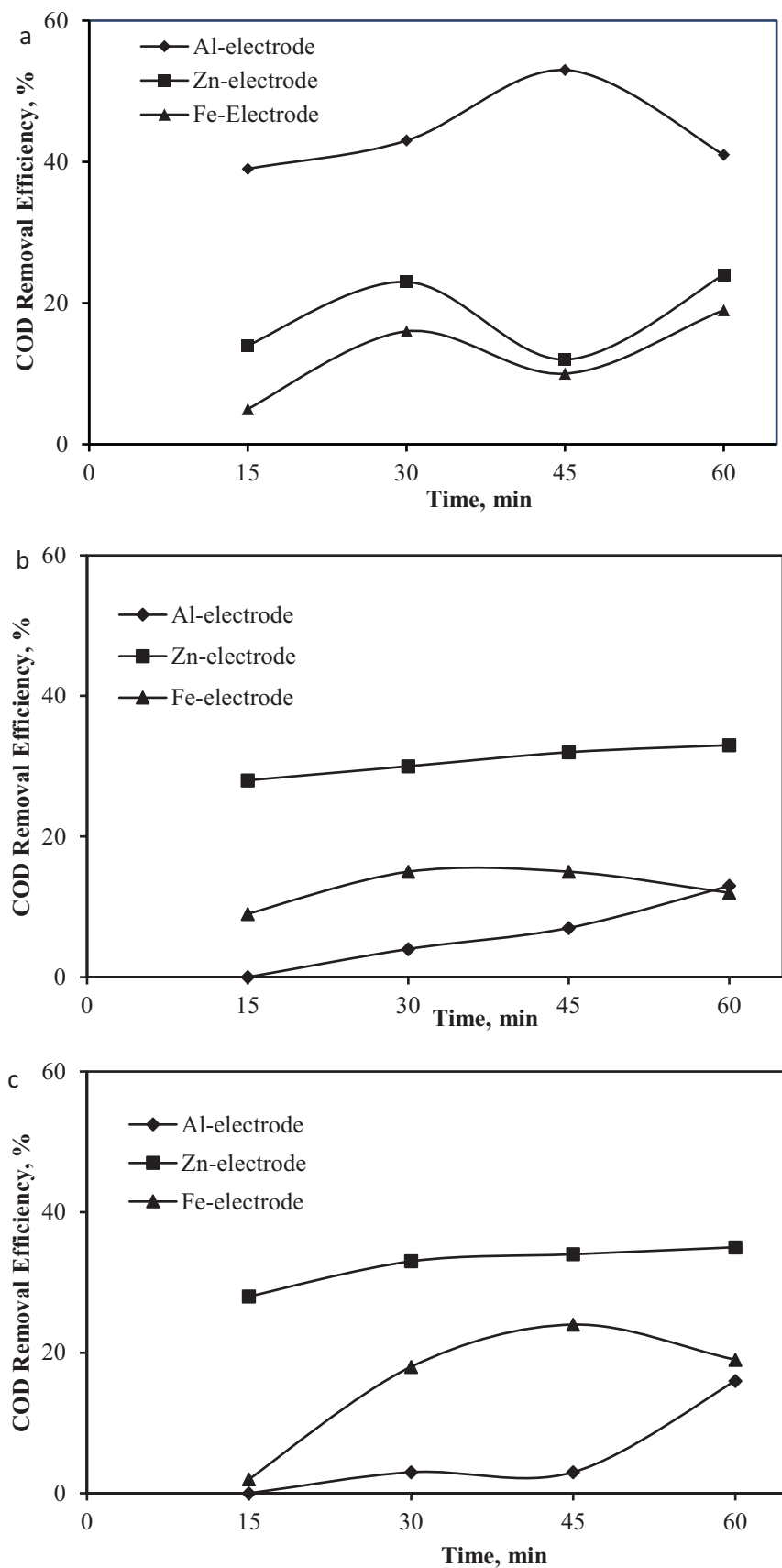


FIGURE 2

The effect of electrode material type on COD Removal Efficiency % for (a) CD=196 A/m², (b) CD=392 A/m² and (c) CD=588 A/m² (pH=8.9).

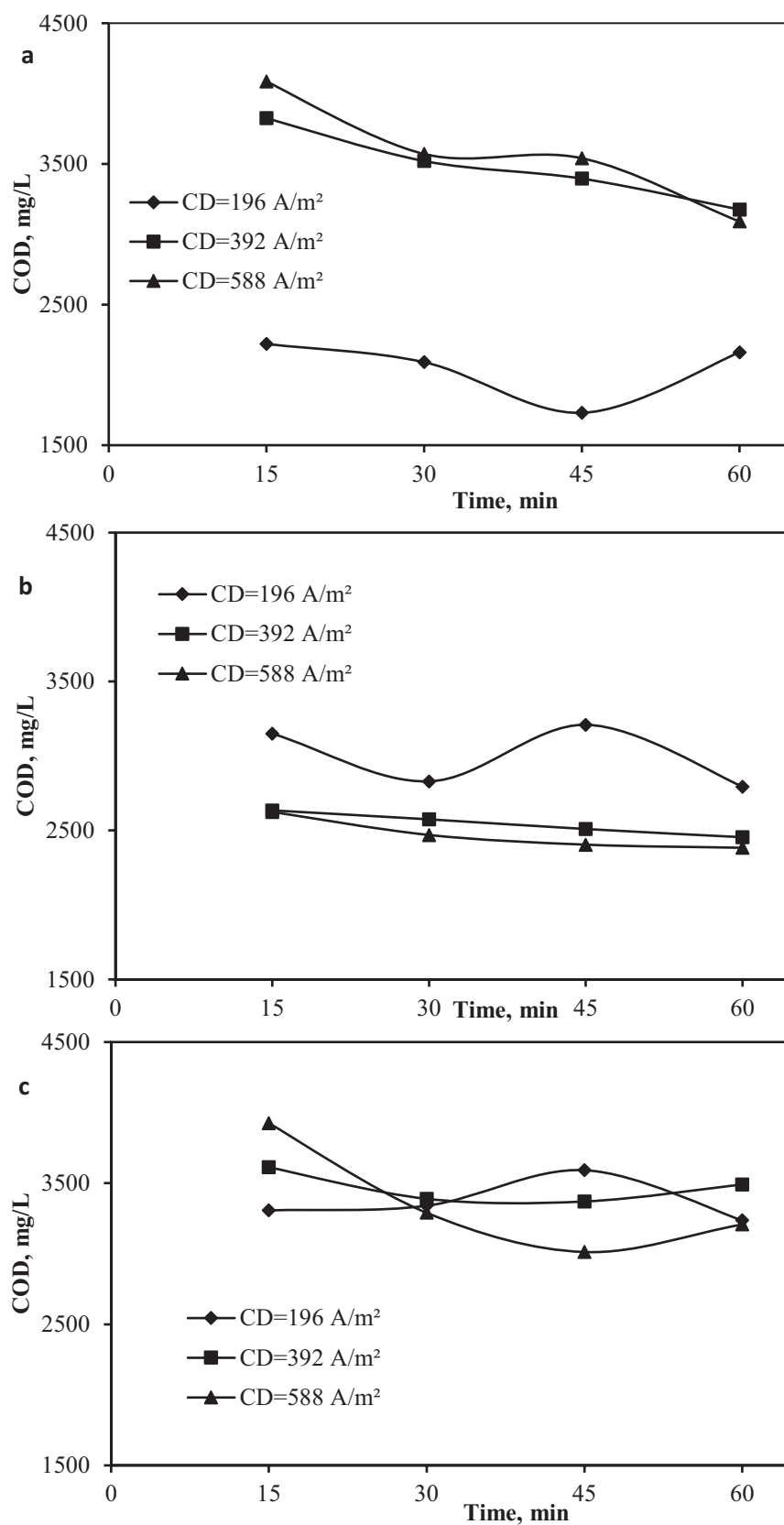


FIGURE 3

Effect of the current density on final COD concentration of type of (a) Al-electrode, (b) Zn-electrode and (c) Fe-electrode (pH=8.9).

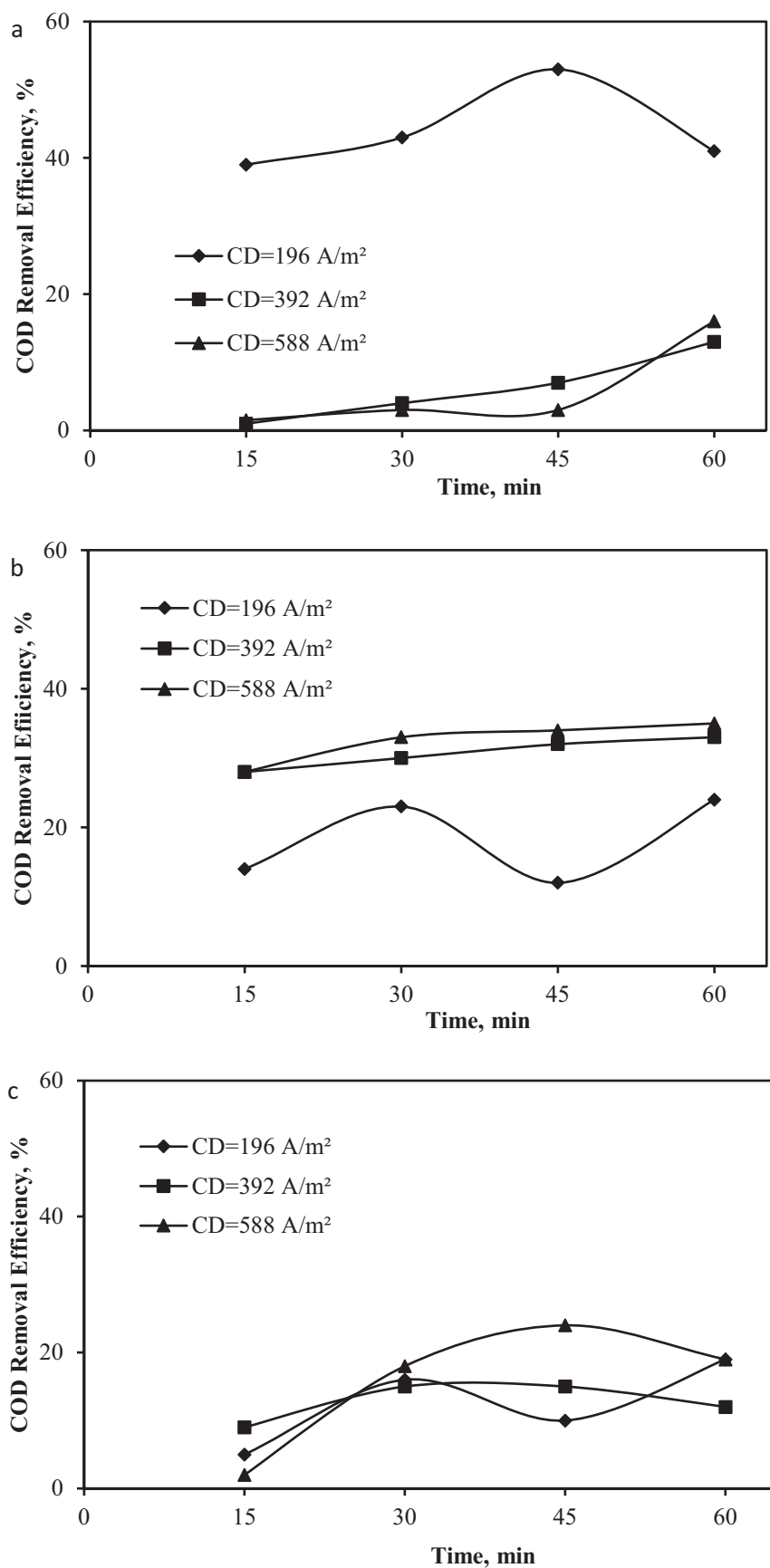


FIGURE 4

Effect of the current density on COD removal efficiency of type of (a) Al-electrode, (b) Zn-electrode and (c) Fe-electrode (pH=8.9).

Figures 5a-5d show the changes in electrical conductivity, pH, turbidity and temperature over time for aluminum, iron and zinc electrodes at the current density of 196 A/m^2 , respectively. As can be observed from the Figures 5a and b that the electrical conductivity and turbidity of the samples tend to decrease during the operation. Significantly observed reduction in turbidity and electrical conductivity of samples can be considered as evidence of removal of COD from the leachate and consequently removal of contaminants.

On the other hand, the temperature and pH of the medium (Figures 5c and 5d) increased during the time period due to the electrolytic reactions of the electrocoagulation process [32]. While the electrocoagulation process continues, the polymeric iron

hydroxides formed in the electrolysis cell carried the pH of the bulk phase to basic values.

Cost analysis of electrocoagulation process.

In order to investigate the influence of the process time, applied current density and electrode type on the leachate treatment energetic costs, the unit electrical energy consumption, E , in kWh/m^3 , for different experiments practised on leachate samples were calculated, by means of Eq.(17), and energy requirements per kilogram COD are presented in Figure 6. Additionally, energy requirements for pollutant removal per cubic meter of leachate and energy consumption per kilogram COD removed for all electrode material types are shown in Figures.7 and 8, respectively.

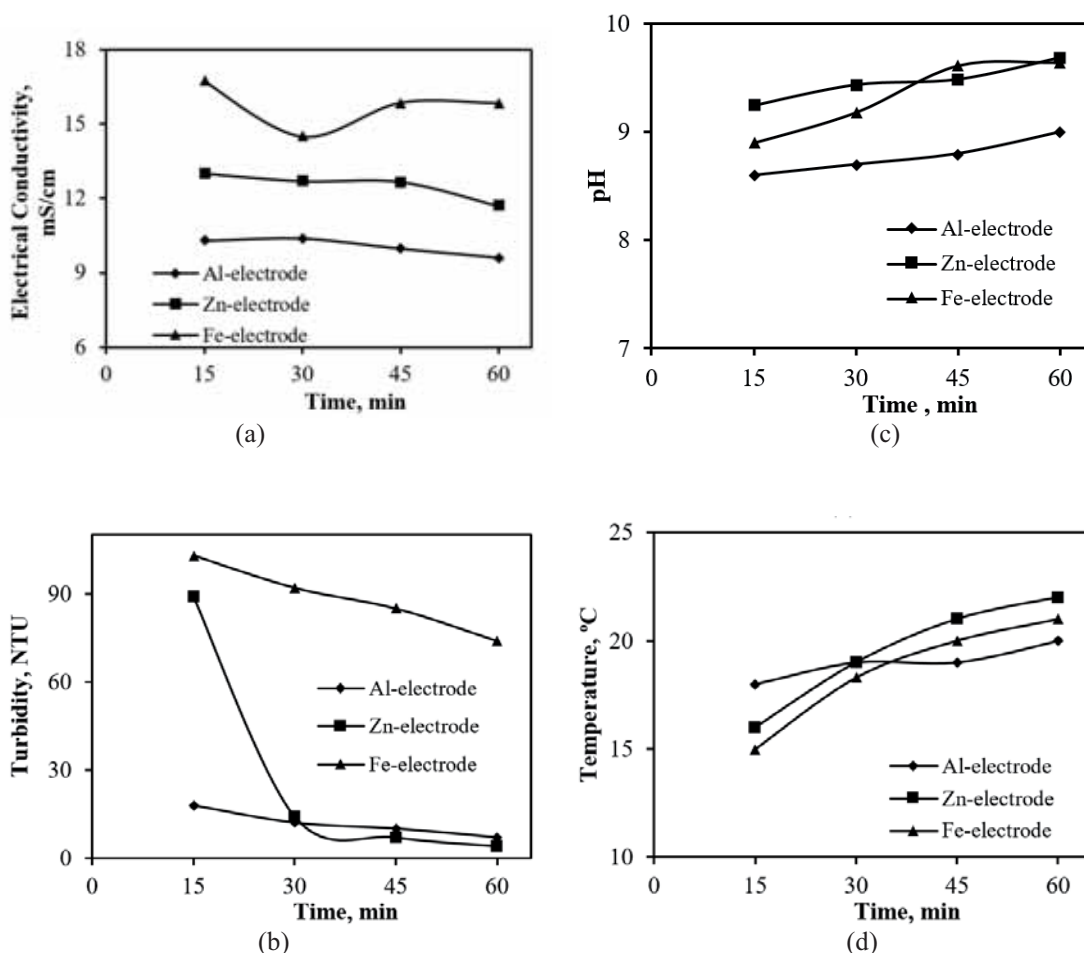


FIGURE 5

Effect of the electrode material type on (a) Electrical conductivity, (b) Turbidity, (c) pH, (d) Temperature ($\text{CD}=196 \text{ A/m}^2$).

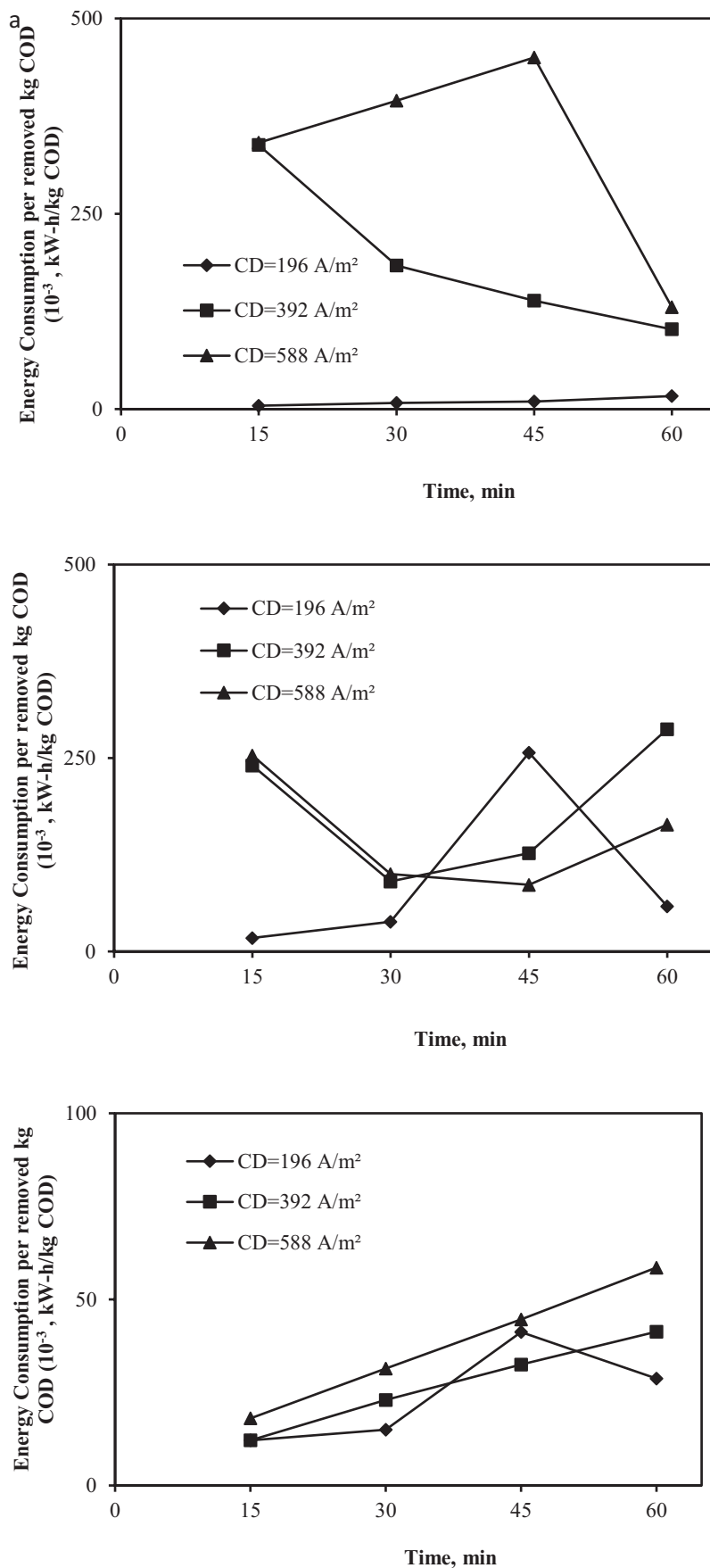


FIGURE 6

Effect of the current density on energy requirement per removed COD for (a) Al-electrode , (b) Fe-electrode and (c) Zn-electrode.

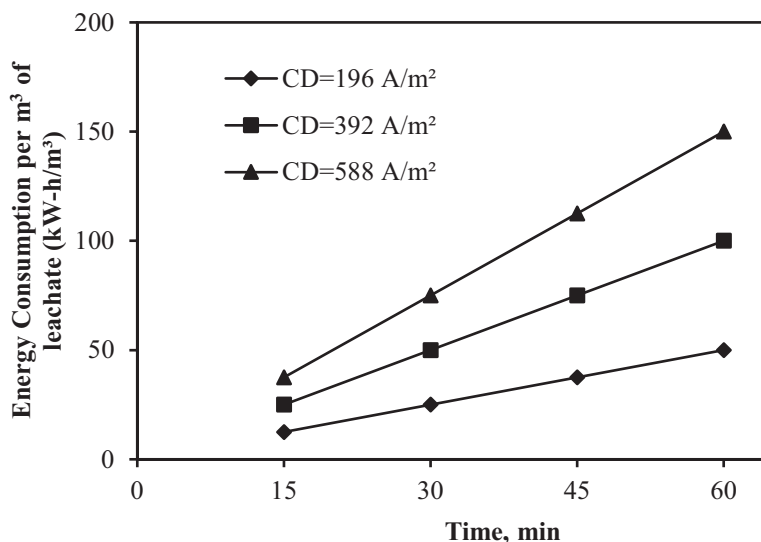


FIGURE 7

Energy consumptions per m³ of leachate for different current density values.

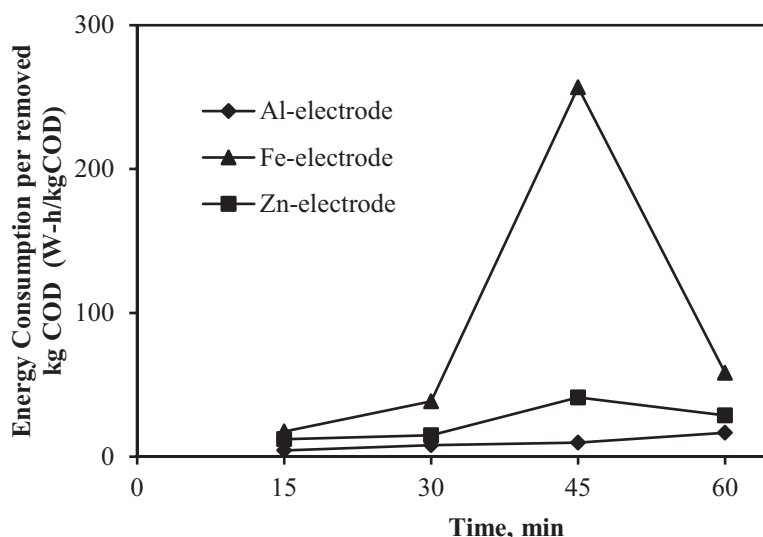


FIGURE 8

Energy consumption per removed kg COD of leachate versus time for different electrode materials.

Operational costs of electrical current are based on time given for the electrocoagulation. So short contact time is preferred and current density with optimum pollutant removal [42]. On this bases, low current density values are preferred for minimal electrical energy consumptions corresponding with operational conditions of the process. It is concluded from the figures that the most economical treatment of the leachate is determined under low current density values.

Figure 8 states that since energy consumption for removed COD values of aluminum electrode are lowest, the most suitable electrode is aluminum to provide a sufficiently good COD removal efficiency and also to reduce the electrical energy payments. Iron electrode tests were concluded with highest energy consumption costs compared to the other types of electrode material.

As known, landfill leachate is a heavily contaminated industrial wastewater that can vary in its physical-chemical parameters depending on disposal time and location, so special attention should be paid to reach desirable discharge quality. Integration the electrocoagulation with other conventional or advanced treatment methods could be a rational solution.

Another issue that is being studied today is the secondary pollution emerges from decontamination processes. Some researchers focused on the reuse of resulting sludge of electrocoagulation like pigment production in ceramic industry or adsorbent material for pollution removal by adsorption [43,44]. Studies on reuse of sludge, which is formed as a secondary pollution as a result of cleaning the leachate by electrocoagulation, will be the subject of a new article.

CONCLUSIONS

This paper studies the factors affecting the removal efficiencies of leachate employed the electrocoagulation process. Eventually outcomes of tests enlighten that;

- COD removal efficiency of leachate could be managed with a satisfactory result.
- 53% removal of COD could be achieved in 45 min contact time for 196 A/m² current density and using aluminum electrode at room temperature and natural pH conditions.
- Aluminum electrode is the best electrode material with quite high removal efficiency and low energy consumption in treatment by electrocoagulation.
- Electrocoagulation could be carried out with alternative electrode materials like Zn and Fe as well as Al.
- Owing to low energy charge, low current density values are more desirable than high ones.
- Electrocoagulation process could be recommended for treatment of leachate integrating with other treatment processes.
- As a continuation of this study, another research can be conducted on reuse of electrocoagulation sludge.

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