Raw landfill leachate treatment using an electrocoagulation process with a novel rotating electrode reactor

Ahmed Samir Naje, Mohammed A. Ajeel, Isam Mohamad Ali, Hussein A. M. Al-Zubaidi and Peter Adeniyi Alaba

ABSTRACT

In this work, landfill leachate treatment by electrocoagulation process with a novel rotating anode reactor was studied. The influence of rotating anode speed on the removal efficiency of chemical oxygen demand (COD), total dissolved solids (TDS), and total suspended solids (TSS) of raw landfill leachate was investigated. The influence of operating parameters like leachate pH, leachate temperature, current, and inter-distance between the cathode rings and anode impellers on the electrocoagulation performance were also investigated. The results revealed the optimum rotating speed is 150 rpm and increasing the rotating speed above this value led to reducing process performance. The leachate electrocoagulation treatment process favors the neutral medium and the treatment performance increases with increasing current intensity. Furthermore, the electrocoagulation treatment performance improves with increasing leachate temperature. However, the performance reduces with increasing inter-electrode distance.

Key words | electrocoagulation, impeller, landfill leachate, rotating electrode

INTRODUCTION

The continuous increase in the human population and the changes in lifestyle have led to a huge amount of solid waste pollutant, which causes an unpleasant environmental impact and resources depletion (Azni 2009). Even though many techniques have been adopted to overcome the solid waste pollution problem such as recycling and reuse, the landfilling process is the main method adopted for the disposal of solid waste. However, the production of leachate from landfilling is the main drawback of this method and it has become a serious threat to the environment. Landfill leachate is a complex combination pollutant consisting of heavy metals, and organic and inorganic substrate. Some of these materials are toxic and refractory pollutants (Van der Bruggen et al. 2003).

It is well known that biological treatment is the cheapest technique for wastewater treatment. However, the presence of the toxic refractory pollutants (non-biodegradable organic pollutants) in landfill leachate reduces the performance of this technique (Naje et al. 2018). Selection of a suitable method for landfill leachate treatment basically depends on the composition of the leachate. Various techniques have been attempted for treatment of landfill leachate, including adsorption, electro-oxidation, biological, and advanced oxidation. Most of these techniques suffer certain drawbacks. For instance, the adsorption method is a very slow process with low efficiency (Hassani et al. 2016); the electrochemical oxidation process is selective and suffers from the electrode passivation phenomenon; and the
advanced oxidation process is costly and requires specific storage tanks for oxidant reagents (Ajeel et al. 2018). The conventional chemical coagulation technique is an economical method for treatment of landfill leachate. Chemicals used for this method include alum with other substrates. The main drawback of this method is the use of other chemicals, which produce a high amount of solid pollutants.

Electrocoagulation has gained immense attention over the last few decades for many reasons like the ease of implantation and automation, it requires no chemical addition, and has simplicity of equipment, short retention time and low sludge generation (Emamjomeh & Sivakumar 2009). Furthermore, leachate with high salinity boosts the electrical conductivity of the electrocoagulation process, which leads to a reduction in the electrical energy consumed during the treatment process (Pérez et al. 2010). The electrocoagulation technique has been used successfully for treatment of various wastewater effluents like textile effluent (Naje et al. 2016; Mook et al. 2017), heavy metal wastewater (Nidheesh & Singh 2017), pharmaceutical pollutants (Farhadi et al. 2012), and industrial effluents (García-García et al. 2013). Several researchers have used the electrocoagulation technique for landfill leachate treatment using various electrodes like the aluminium electrode (Li et al. 2011), and the iron electrode (Huda et al. 2017). However, most of the electrocoagulation researches have used a conventional reactor design with a static electrode and agitated the solution using a magnetic stirrer. Inefficient solution mixing reduces the mass transfer during the process due to poor diffusion of molecules, which leads to formation of a passive film on the electrode surface. Electrode fouling is one of the most frequent drawbacks in electrochemical wastewater treatment, which decreases the treatment performance. Moreover, electrode passivation increases the resistance of the electrocoagulation cell, which increases the required operation potential, thereby increasing the overall energy consumption.

An electrocoagulation treatment with a novel rotating anode reactor was used for the first time in our laboratory for the treatment of industrial textile dye effluent. The report showed that the treatment of color and chemical oxygen demand (COD) of the wastewater effluent is economical and very efficient (Naje et al. 2017, 2018). In this work, the performance of the electrocoagulation reactor with a rotating electrode was investigated for the treatment of landfill leachate. This is the first study to report the influence of rotating electrode speed on the electrocoagulation treatment performance of landfill leachate. The effects of current, pH, temperature, and inter-electrode distance on the removal efficiency of COD, total suspended solids (TSS), and total dissolved solids (TDS) of landfill leachate are also investigated.

EXPERIMENTAL

Reactor constriction

The electrocoagulation reactor with a rotating anode that was used during this work is depicted in Figure 1. The reactor size is 10 L, constructed from Perspex and consisting of a cylindrical stirred tank with a 174 mm inner diameter, 180 mm external diameter, and 500 mm length. The rotating anode consists of 40 rods of aluminum substrate, each rod having dimensions of 50 mm length and 12 mm diameter. These 40 rods were divided into 10 impellers, with each impeller comprising four rods. The 10 impellers were assembled with a rotating shaft of 32 mm diameter, which was connected to a variable speed motor to hold the impeller structure and maintain the electrode rotation. The rotating anode, with 10 impellers, was assembled with a cathode of 10 rings, each ring encapsulating one impeller. The rings were arranged in such a way that the distance between two rings is 30 mm and the dimensions of each ring were 172 mm external diameter, 134 mm internal diameter, and 12 mm thickness. An AC electrical motor was used to supply different steady state speeds (0 to 500 rpm, 220 V, 0 to 4 A). The total geometric anode surface area was 500 cm² and the reactor had three symmetrical zone baffles to prevent movement and mass fluid’s tangential flow arrangements and to position the cathode rings.

Materials

Ultra-pure deionized Milli-Q water was used to formulate all solutions. H₂SO₄ (97% Pro Analysis, Merck) and NaOH (95%, Fisher Scientific) were used to adjust the pH of the solution during the treatment process. HCl solution (35% purity, AR Grade, Merck) was used to clean release the scales from the electrodes’ surface.

Sample collection and characterization

The landfill leachate sample has been collected without pre-treatment from Air Hitam Sanitary Kuala Lumpur landfill (HSK) location, Kuala Lumpur, Malaysia. HSK is one of the major landfills in Kuala Lumpur city, which daily receives more than 2,000 tons of solid waste. The collected samples were characterized and
stored at 3 °C to be used throughout the research work. Dichromate technique was used to measure the COD with Cell Test (300–3,500 mg/l, Spectroquant-Merck). The TSS and TDS were determined using the Gravimetric method. HANNA HI-99301 and HACH 2100P technologies were used to investigate the conductivity and turbidity of the landfill leachate, respectively. The DC current source is supplied by an ISO-TECH programmable Power supply.

Electrocoagulation process

For electrocoagulation of landfill experiments, a 10 L electrochemical cell arranged as a stirred tank equipped with aluminium electrodes was used. The effect of anode rotating speed at 0, 75, 100, 150, 200, and 250 rpm was investigated. The experiments were conducted with 1, 2, 3, 4, and 5A applied current intensities. The effect of initial pH of 5, 6, 7, 7.8, 8, 9, and 10 was investigated at 25 °C. Furthermore,
the effect of solution temperature and the inter-distance between cathode rings and anode impellers on treatment process performance were also investigated. The effect of temperature was investigated in the range of 25–45°C using water circulation (WiseCircu Model WCR-P6) to maintain the temperature during the EC process. In every iteration, the samples were allowed to settle for 30 min and then filtered. The samples were filtered using a Whatman 934 AH filter. Approximately 100 ml of supernatant sample was gathered for analysis and investigation in replicate. Similar parameters were estimated in every replicate sample.

RESULT AND DISCUSSION

Landfill leachate characterization

Table 1 presents the landfill leachate specification, revealing that the leachate has a pH 7.82, which is closed to a neutral solution, and exhibits a very limited value of alkylation. The discharged effluent with this value of pH is within the acceptable limit of Malaysian Environment Department standards. This value of pH reveals the leachate is old and inert, whereas early life leachate usually has a low pH of approximately less than 4 due to the presence of volatile organic compounds (Bakraouy et al. 2017). However, landfill leachate pH increases gradually to above 7 towards alkaline as a result of methanogens’ activities (Bhalla et al. 2022). It is obvious from Table 1 that the value of COD is relatively high (about 2,130 mg/L). The high COD value of the stabilized leachate may be attributed to the presence of humic-acid substrates, which the microorganism could not stabilize. However, the leachate has humic-like substrates, which are the most difficult to treat because they comprise mainly aromatic rings and aliphatic chains (Dia et al. 2017). The TDS of the landfill leachate is 4,397 mg/L, which is relatively high and hence improves the leachate conductivity.

Table 1 | Characteristics of the raw landfill leachate

<table>
<thead>
<tr>
<th>Properties</th>
<th>Values</th>
</tr>
</thead>
<tbody>
<tr>
<td>pH</td>
<td>7.82 (+0.1)</td>
</tr>
<tr>
<td>COD (mg/l)</td>
<td>2,130 (+8)</td>
</tr>
<tr>
<td>BOD (mg/l)</td>
<td>850 (+3)</td>
</tr>
<tr>
<td>TSS (mg/l)</td>
<td>270 (+1.5)</td>
</tr>
<tr>
<td>TDS (mg/l)</td>
<td>4,397 (+10)</td>
</tr>
<tr>
<td>DO (mg/l)</td>
<td>0.82 (+0.01)</td>
</tr>
<tr>
<td>EC (µs/cm)</td>
<td>8,650 (+10)</td>
</tr>
</tbody>
</table>

Effects of rotation speed

Anode rotation speed is considered a significant factor that influences anode dissolution. It enhances the anode dissolution as a result of reducing the anode passivity, also by moving the generated coagulate and releasing it to the bulk solution from the anode surface (Naje et al. 2016). Furthermore, a suitable rotation speed helps to homogenize the pollutant and coagulant particles in the solution (Naje et al. 2017). In contrast, the high electrode rotating speed may lead to fracture of the flocs and produce small particle flocs that are difficult to collect after the treatment process. Figure 2 presents the removal efficiency of COD, TSS, and TDS of raw landfill leachate on the rotating anode reactor with 2 A current, 25°C temperature, and pH 7. The effect of electrode rotating speed was investigated using different electrode velocities including 0, 75, 100, 150, 200, and 250 rpm. The highest removal performance was observed at 150 rpm electrode rotating speed. After 10 min of the treatment process the COD, TSS, and TDS removal efficiencies were 85%, 90%, and 83% respectively. Contrarily, the observed removal efficiency of COD, TSS, and TDS on the static electrode were 50%, 55%, and 55%, respectively, which represents almost half of those observed on the electrode with 150 rpm rotating speed. The COD, TSS, and TDS removal is enhanced with increasing electrode rotating speed, and the maximum is attained at 150 rpm. However, further increasing the electrode rotational speed to 200 and 250 led to a gradual reduction in removal efficiency, as depicted in Figure 2. This may be because increasing anode rotating speed led to incorporation of the flocs produced with Al(OH)3, hence the coagulant precipitation becomes faster. However, increasing the electrode rotating speed above 150 rpm to 200 rpm and then 250 rpm destroys the flocs, thereby desorbing the adsorbed pollutants (Naje et al. 2018). This leads to a reduction in the removal efficiency of COD, TSS, and TDS. This finding is in agreement with the previous researches that studied the effect of stirring speed on the electrocoagulation performance (Gürses et al. 2002; Can et al. 2003; Bayar et al. 2011).

Effects of current intensity

Current intensity is considered as a significant factor during electrocoagulation treatment since it affects the dosage rate of the coagulant particles into the bulk of the electrolyte, size and growth of the flocs, and bubble production rate, thereby affecting the efficiency of the electrocoagulation process. Increasing the current boosts the flocculation of
Al(OH)₃ due to increasing anode dissolution rate. Therefore, improved flocculation of Al(OH)₃ in solution promotes the efficiency of the electrocoagulation process. However, increasing the current intensity above the optimum level does not significantly improve the removal efficiency of the pollutant as sufficient flocs are available for the sedimentation of the pollutant. The effect of current intensity on the performance of the treatment process was investigated by varying from 1, 2, 3, 4 to 5 A, and the results were observed after 10 min using 150 rpm electrode rotation speed. Figure 3 illustrates the influence of current intensity on electrocoagulation treatment performance on landfill leachate. It is obvious that the first increment in current increasing from 1 A to 2 A had a significant impact on the treatment process, whereas, the COD, TSS, and TDS removal increased from 70%, 75%, and 68% to be 85%, 84%, 90%, and 83% due to increasing current intensity for the first time. In contrast, a further rise in the current had no significant influence on COD, TSS, and TDS removal efficiency, as depicted in Figure 3. This phenomenon may attribute to the decreasing pollutant concentration; after a short time, the electrocoagulation process commenced due to the high rate of flocculation associated with high current intensity. Therefore, increasing the rate of flocculation in a system containing a low pollutant concentration solution is not necessary to improve the removal efficiency, although a high current could reduce the required time for the electrocoagulation process to be completed. Furthermore, an increase in the operating current increases the energy consumption and reduces the current efficiency (Ajeel et al. 2015a, 2015b, 2017).

**Effect of pH**

The pH of the electrocoagulation process is a significant operating factor. The optimal performance of the
The electrocoagulation process is achieved in a solution with suitable pH value. The optimum pH for electrocoagulation process greatly depends on the nature of the pollutant and the electrode material. The influence of the initial pH of the solution on the performance of the treatment process has been investigated by changing the nature of the solution from acidic, to neutral, to basic using an initial pH of 5, 6, 7, 7.8, 8, 9, and 10. Figure 4 presents the influence of pH on the electrocoagulation process of landfill leachate. The optimum pH solution was found to be 7, giving the maximum removal of COD, TSS, and TDS. Meanwhile, the electrocoagulation process performance declined when the pH tended toward acidic or basic values. Increasing the initial pH value a little from 7 to 7.8 led to a significant decline in the removal efficiency of COD, and TDS but an insignificant reduction in the value of TSS removal efficiency. This result is in agreement with the previous works of Li et al. 2011 and Hassani et al. 2016. During the electrocoagulation treatment process for landfill leachate, the effect of a change in pH value is observed for different initial pH values (5, 6, 7, 7.8, 8, 9, and 10) that were investigated. The results show the pH values at the end of the treatment process are increased for the different values investigated.

Effect of temperature

Prior to the treatment process, the solution temperature was modified to specific levels and fixed throughout the electrocoagulation treatment, sedimentation, and filtration processes. The effect of temperature on the treatment process was investigated using 5 different temperature levels including 25, 30, 35, 40, and 45 ºC under optimal operation conditions. Figure 5 demonstrates the impact of increasing temperature on COD, TSS, and TDS removal efficiency. Obviously, an increase in the solution temperature improves the performance of the electrocoagulation treatment of landfill leachate. The improvement could be ascribed to several reasons. The first reason is enhanced hydrolysis of Al³⁺ to Al(OH)₃, resulting from improved diffusivity of Al³⁺ from the anode surface to the bulk of solution according to Einstein’s standard equation (El-Ashtoukhy et al. 2013).

Effect of the inter-distance between anode impellers and cathode rings

Many researchers have studied the effect of the electrode distance parameter on the performance of the electrocoagulation process for different pollutant natures. Among these researches, the effect of the inter-electrode distance parameter on the performance of the electrocoagulation process depends on many factors such as electrode structure, hydrodynamic conditions, and pollutant nature (Daneshvar et al. 2004; Modirshahla et al. 2007). Figure 6 shows the effect of inter-electrode distance between the impellers of the anode and cathode rings at different distances of 10, 15 and 20 mm with optimum operation conditions of 150 rpm electrode rotating speed, 2 A current, pH 7, and 10 min. The results reveal declines in the values of

Figure 4 | The effect of pH on the removal efficiency of COD, BOD, TSS, and TDS of raw landfill leachate at 150 rpm impeller velocity, 2 A current intensity, 10 min treatment time, 1 cm electrode gap, and 25 ºC.

Figure 5 | The effect of temperature on the removal efficiency of COD, BOD, TSS, and TDS of raw landfill leachate at 150 rpm impeller velocity, 2 A current intensity, 10 min treatment time, 1 cm electrode gap and pH 7.
The electrocoagulation technique using a rotating anode reactor has been used for treatment of raw landfill leachate. The optimum performance of the electrocoagulation process was achieved at anode rotation speed of 50 rpm and a further increase in the rotating speed will reduce the performance of the treatment process. The leachate treatment using the electrocoagulation technique with a novel reactor was very efficient. The removal efficiency of COD, TSS, and TDS after 40 min was 94.5%, 95.5%, and 93% respectively. Electrocoagulation of landfill leachate was favored at neutral pH and the performance of the treatment reduced significantly when the pH was altered toward the acidic or alkaline region. Increasing applied current and leachate temperature led to improved treatment performance. Furthermore, increasing the inter-distance between the impeller’s anode and cathode rings reduces the process performance.

Figure 6 | The effect of inter-electrode distance on the removal efficiency of COD, BOD, TSS, and TDS of raw landfill leachate at 150 rpm impeller velocity, 2 A current intensity, pH 7, 10 min treatment time, and 25 °C.

COD, TSS, and TDS with increasing inter-electrode distance from 10 mm to 15 and 20 mm. The observed reduction in the performance of the electrocoagulation process is due to the decline in the electrical conductivity of the process with an increase in the distance between electrodes. Reducing electrical conductivity also leads to a decrease in the ion metal dissolution, thereby minimizing the number of flocs in the bulk solution. Furthermore, a treatment bath with a large distance between electrodes requires a high voltage over the electrodes to operate the electrocoagulation process as due to an increase in the resistance of the solution. The use of high voltage throughout the process leads to an increase in the operational cost of the treatment. However, increasing inter-electrode distance could reduce the interaction of the pollutants with the ionic hydroxyl. Therefore, the electrostatic attraction and local concentration declined, thereby reducing the performance of the treatment process (Abdel-Gawad et al. 2012).

CONCLUSION

The electrocoagulation technique using a rotating anode reactor has been used for treatment of raw landfill leachate. The optimum performance of the electrocoagulation process was achieved at anode rotation speed of 50 rpm and a further increase in the rotating speed will reduce the performance of the treatment process. The leachate treatment using the electrocoagulation technique with a novel reactor was very efficient. The removal efficiency of COD, TSS, and TDS after 40 min was 94.5%, 95.5%, and 93% respectively. Electrocoagulation of landfill leachate was favored at neutral pH and the performance of the treatment reduced significantly when the pH was altered toward the acidic or alkaline region. Increasing applied current and leachate temperature led to improved treatment performance. Furthermore, increasing the inter-distance between the impeller’s anode and cathode rings reduces the process performance.

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