Treatment of municipal solid wastes leachate by means of chemical- and electro-coagulation

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Abstract

In this study, treatment of municipal solid waste leachate (L1, L2) by means of chemical- and electro-coagulation (EC) methods was investigated. The removal of chemical oxygen demand (COD), total organic carbons (TOC) and color from leachate was experimentally investigated by using chemical- and electro-coagulation. Aluminum and iron were used simultaneously in the reactor as materials for electrodes. Percent of COD, TOC and color removal versus the electro-chemically generated Fe2+ and Al3+ doses are examined in this study. The best removal has been achieved in the COD parameter. Thus, in electro-coagulation method, the maximal removal is 87% for the L1 solution, while for L2 it is 90% for Fe2+ and 77% for L1 and 88% for L2 for Al3+ ion solutions. High color removal for Fe2+ is observed for L1 (86%), while for L2 it is approximately 99%. The removal indices for the leachate treatment by chemical coagulants (FeSO4·7H2O and Al2(SO4)3·18H2O) appeared to be lower than for the electrochemically generated Fe2+ and Al3+. The results show that electro-coagulation could be considered as an effective alternative solution for the treatment of leachate.

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Keywords: Leachate; Chemical-coagulation; Electro-coagulation; Al/Fe electrode; Wastewater treatment

1. Introduction

Solid waste removal has become a serious environmental problem due to the increase in the world population and changes in the consumption habits. In the solid waste management, landfill is one of the most prevalent methods used by many countries in the world [1]. With approximate calculations, it is found that 450–500 million tonnes of domestic solid waste is produced annually in the world [2]. Three hundred and twenty to three hundred and fifty million tonnes of this, that is equivalent to 70%, is removed by landfill [3]. During or after landfill operation, leachate occurs due to the humidity rate of the waste on site, chemical and physical disposal reactions of wastes, rain water and the increase in the level of underground water. With the characteristics of high chemical oxygen demand (COD), total organic carbon (TOC), color and potential toxicity, the wastewater has become another aspect in the solid waste problem.

The composition of leachate and the concentration of contaminants highly depend on the composition of the waste and the age of the landfill. Generally landfills are classified as young for <5 years, middle aged for 5–10 years, and old for >10 [4].

The infiltration of untreated leachate may contaminate both soil and eventually ground water. Therefore, a proper treatment of leachate is an important issue. There are various methods in leachate treatment. Different aerobic and anaerobic treatment techniques are the most widely used treatment methods. These systems provide high efficiency, in spite of the difficulties resulting from the basic composition of the leachate, which are related to the age of the landfill site. The efficiency of these treatment processes is highly affected by the flow and the composition of the leachate. Nevertheless, biological treatment processes are insufficient in the removal of persistent organics. That is why for the treatment of leachate, we need different treatment processes, the most widely used of which are the physicochemical processes [5]. The characteristics of leachate play an important role in the determination of the treatment method. To treat these wastewaters, advanced oxidation processes [6], air stripping [7], ion exchange, and membrane processes [8] are other treatment methods used for the removal of nitrogen and organic
and spring are rainy, heavy rain occurs in winter. Annual
mate characteristics are dominant in this region. While autumn
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Solid Waste Landfill, located in the village of Solaklar, 15 km
2.1. Landfill site description
was determined.
2. Materials and methods
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The study was carried out in Izmit Domestic and Industrial
Solid Waste Landfill, located in the village of Solaklar, 15 km
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climate characteristics are dominant in this region. While autumn
and spring are rainy, heavy rain occurs in winter. Annual
average temperature is 14.7 °C, and the rainfall is 876.6 mm

The landfill is constructed on a total area of 800,000 m² in
Kocaeli. 31,841 m² of this area are used as Clinical and Haz-
ardous Wastes Incineration Plant. Approximately 362,557 m² of
this area is separated for the storage of domestic and industrial
solid waste. The landfill is designed to close in 25–30 years. The
storage site has seven lots of different size with the total capacity
of 4,132,919 million m³. The area and the volume of each lot are
shown in Table 1.

One out of seven lots is used to deposit clinic and haz-
ardous wastes. The others having the area of 264,392 m² are
used for domestic solid wastes. The layer permeability of the
lots is designed with respect to the related directory. The stor-
age activity is completed in one lot that is the 7th lot, among the
six lots. Now, the storage activity is still going on in the
5th lot and the leachate used in this study is collected from this
lot. The leachate formed within the lots is collected with
high-density polyethylene (HDPE) pipes and transferred to the
collector ventilator.

Domestic solid wastes, domestic treatment sludge, and the
industrial wastes that can be stored with domestic wastes are
accepted in the domestic solid waste site. The daily solid waste
entering the plant is approximately 500 tonnes [21]. The calorific
value of the solid wastes in this region changes between 950 and
1300 kcal kg⁻¹. The composition, amount and percentages of solid waste stored are given in Table 2, and the amount of the
solid waste stored in the plant is presented in Table 3.

In this study, the treatment efficiency of the leachate collected
from the domestic waste lot of “Izmit Domestic and Industrial
Solid Waste Landfill Plant” by means of chemical- and electro-coagulation processes is dis-
cussed. In the plant, six lots are separated for domestic solids
and only one of them had been full. The study was carried out
for the leachate collected from the lots where the storage activity
still goes on.

First of all, the general characteristics of the leachate col-
lected from a younger domestic solid waste lot were analyzed,
and the treatment efficiency of a pilot electro-coagulation reactor
with respect to TOC, COD and color parameters was searched.
The results, which were obtained from chemical-coagulation
method have been compared with the results, which were
obtained by the electro-coagulation method. By comparing the
results obtained and the results of chemical-coagulation process,
the place of the new process, i.e. electro-coagulation process,
was determined.

2. Materials and methods

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<table>
<thead>
<tr>
<th>Nomenclature</th>
</tr>
</thead>
<tbody>
<tr>
<td>$i$</td>
</tr>
<tr>
<td>$\Delta m$</td>
</tr>
</tbody>
</table>

Some researchers tried to find appropriate physicochem-
ic methods (coagulation/flocculation and ozonation) for the
efficient treatment of leachates [9,10]. As well as efficient usage
of these methods for leachate treatment, in the case of in situ
treatment and discharge, unit processes having few steps are
used [11].

In addition, the leachate treatment efficiency of the electro-
coagulation (EC) method being a kind of electrochemical
process, which has a continuously increasing interest, is exam-
inied in this study. Electro-chemical technology became an
inventive step for the resistant contaminants in wastewater [12]. Especially, it is used efficiently in the treat-
ment of textile [13], food [14,15], metal and galvanization [16],
and petrochemistry industry [17,18] effluents.

Electro-chemical processes are supposed to have a wide
perspective. It is considered that this technology will have a
widespread usage in water and wastewater treatment due to such
its characteristics as less equipment requirement, shorter treat-
ment period, no chemical matter need and less sludge formation
as a result of all this [19]. Besides all these advantages, method
has some disadvantages such as exchange of anodes as elec-
trodes consume and high operation costs where electricity is
expensive.

In this study, the treatment of the leachate generated from
‘Izmit Domestic and Industrial Solid Waste Landfill Plant’ by
means of chemical- and electro-coagulation processes is dis-
cussed. In the plant, six lots are separated for domestic solids
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was determined.

Table 1
Areas and capacities of the Izmit municipal and industrial solid waste landfill lots

<table>
<thead>
<tr>
<th>Lot no.</th>
<th>Area (m²)</th>
<th>Capacity (m³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>76,841</td>
<td>1,580,000</td>
</tr>
<tr>
<td>2</td>
<td>28,586</td>
<td>68,000</td>
</tr>
<tr>
<td>3</td>
<td>32,645</td>
<td>95,000</td>
</tr>
<tr>
<td>4</td>
<td>11,845</td>
<td>101,000</td>
</tr>
<tr>
<td>5</td>
<td>63,980</td>
<td>1,029,000</td>
</tr>
<tr>
<td>6</td>
<td>98,165</td>
<td>969,919</td>
</tr>
<tr>
<td>7</td>
<td>50,495</td>
<td>290,000</td>
</tr>
<tr>
<td>Total</td>
<td>362,557</td>
<td>4,132,919</td>
</tr>
</tbody>
</table>

Table 2
Composition of solid wastes collected in Kocaeli city

<table>
<thead>
<tr>
<th>Composition</th>
<th>Amount (tonnes)</th>
<th>Percentage (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Organics</td>
<td>232</td>
<td>68</td>
</tr>
<tr>
<td>Paper and wooden</td>
<td>27</td>
<td>8</td>
</tr>
<tr>
<td>Textile</td>
<td>27</td>
<td>8</td>
</tr>
<tr>
<td>Plastics</td>
<td>27</td>
<td>8</td>
</tr>
<tr>
<td>Glass</td>
<td>14</td>
<td>4</td>
</tr>
<tr>
<td>Metals</td>
<td>14</td>
<td>4</td>
</tr>
</tbody>
</table>
## Table 3
Annual waste deposition at the landfill

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Waste (tonnes)</td>
<td>51/152</td>
<td>163/188</td>
<td>183/184</td>
<td>187/190</td>
</tr>
</tbody>
</table>

## Table 4
Chemical characteristics of leachate at active landfill

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Concentration</th>
</tr>
</thead>
<tbody>
<tr>
<td>Chemical oxygen demand (mg L(^{-1}))</td>
<td>4022.5</td>
</tr>
<tr>
<td>Total organic carbon (mg L(^{-1}))</td>
<td>1295</td>
</tr>
<tr>
<td>Conductivity (mS cm(^{-1}))</td>
<td>25.11</td>
</tr>
<tr>
<td>Color (Hz)</td>
<td>2650</td>
</tr>
<tr>
<td>pH</td>
<td>7.71</td>
</tr>
</tbody>
</table>

## 2.1.1. Analysis
COD analyses were carried out in thermoreactor (VELP-ECO16) by using 5220C Closed Reflux Titrimetric Method [22]. Spectrophotometric method was used for determining color. Color analyses were made in fotometer (MERCK-SQ118). Also, pH-meter (SOLOMAT-520C) was used for pH measurements. pH adjustment was done by using 0.1N H\(_2\)SO\(_4\) and 0.1N NaOH.

## 2.2. Scheme and characterization of EC reactor

The laboratory-scale reactor (16 cm × 15 cm × 20 cm, 2 L) was used during experimental studies (Fig. 1). Two groups of alternating electrodes being cathodes and anodes (by seven of each type) were arranged vertically. Distance between an anode and a neighboring cathode is 3 mm.

Iron/aluminum were used in the EC process as materials for anodes/cathodes or vice versa. Before the experiments, the electrodes were immersed in 1% HCl for 8 h.

As seen from Fig. 1, the process of water treatment was divided into two steps. At the first one, the coagulant preparation takes place in the Reactor 1. After this, the solution obtained was mixed with wastewater in the Reactor 2 for the sedimentation.

The electrolyte duration in the EC cell was taken as 5, 10, and 15 min under different values of current density (CD).

To get ions of Fe\(^{2+}\) and Al\(^{3+}\) in the Reactor 1, stable pH was adjusted (pH 3). Low pH for the electrolyte promotes additional conditions for the metal chemical dissolving. This also prevents electrodes from the premature passivity. In the Reactor 2, the necessary for the solution pH levels have been held up for the hydroxide Fe\(^{2+}\) (pH ≈ 8.5–9.0) and Al\(^{3+}\) (pH ≈ 6.5–7.0) sediment. Temperature (21–22 °C) was stable during the experiments.

## 3. Results and discussion

Metal dissolving is a complex reaction related to the electrochemical process. In the Reactor 1, a chemical (anodic and cathodic) dissolution occurs under acidic conditions. A preliminary study of the Reactor 1 capabilities, as to getting Fe\(^{2+}\) and Al\(^{3+}\) ions, was carried out.

Dependence between the quantities of the metal dissolved (\(\Delta m, g L^{-1}\)) and current density values (\(i, mA cm^{-2}\)) under different values of duration (from 5 to 15 min) of the electrolyte staying in the reactor cell (RC) is a linear one. Thus, for different values of CD, one can calculate the quantity of the dissolved Fe and Al from the experimental data (Table 5).

## Table 5
The linear equation [\(\Delta m (g L^{-1}) = a + b \times i (mA cm^{-2})\)] parameters for different values of CD (\(i\)) and duration (\(t\)) of the electrolyte staying in the EC cell (Reactor 1)

<table>
<thead>
<tr>
<th>(t) (min)</th>
<th>(a)</th>
<th>(b)</th>
<th>(R^2)</th>
<th>S.E.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fe anode</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>-0.083368</td>
<td>0.030804</td>
<td>0.96</td>
<td>0.062</td>
</tr>
<tr>
<td>10</td>
<td>0.000355</td>
<td>0.044263</td>
<td>0.99</td>
<td>0.001</td>
</tr>
<tr>
<td>15</td>
<td>-0.023679</td>
<td>0.071700</td>
<td>0.99</td>
<td>0.026</td>
</tr>
<tr>
<td>Al anode</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>-0.016062</td>
<td>0.009271</td>
<td>0.98</td>
<td>0.012</td>
</tr>
<tr>
<td>10</td>
<td>-0.005217</td>
<td>0.015457</td>
<td>0.99</td>
<td>0.006</td>
</tr>
<tr>
<td>15</td>
<td>-0.001639</td>
<td>0.021411</td>
<td>0.99</td>
<td>0.002</td>
</tr>
</tbody>
</table>

\(R\): Correlation coefficient; S.E.: standard error.
These calculations were used for the preliminary estimation of the metal ions quantity in the solution and for comparing electro-coagulation process with the process of chemical-coagulation. The optimal anodic CD for Al is in the range of 2–15 mA cm$^{-2}$, and for Fe anodes it is 1–10 mA cm$^{-2}$. The CD $(i)$ values greater than 12 mA cm$^{-2}$ cause the passivity of the Fe anode.

As seen from Fig. 1, wastewater comes to the EC unit (it passes among aluminum/iron electrodes) and, being enriched by ions of Al$^{3+}$/Fe$^{2+}$, enters the Reactor 2, where the coagulation of Al(OH)$_3$/Fe(OH)$_2$ takes place (pH 6.5–7.0 for Al$^{3+}$ ions, and pH 8.5–9.0 for Fe$^{2+}$ ions). In the process of the EC, the absorption volumes of the formed metal hydroxides are very high. Coagulated particles attract and absorb micro-colloidal particles and ions from the wastewater.

The mechanisms of reactions that happen in the electrochemical units are studied extensively. When direct current passes through the electrodes, metal ions dissolve and react with OH$^-$ ions in water. Metal hydroxyls that can dissolve in water partially are being formed under appropriate pH level. This stage ends with the formation of colloidal particles. Hydroxyls form the nucleus of the colloidal particle, and the adsorption layer of cations and anions is being formed around the nucleus. So, the nucleus and the adsorption layer form the positively charged colloidal granule. Diffusion layer then occurs around the granule, and the particle becomes neutral. The metal hydroxyls that are formed in the EC process have a high adsorption capacity. Some chemical reactions, which occur on electrodes and in the bulk wastewater, are shown below.

Reactions on the Fe-anode:

\[ \text{Fe}^0 - 2e^- \rightarrow \text{Fe}^{2+} \]

\[ \text{Fe}^{2+} + \text{OH}^- \rightarrow \text{FeOH}^+ \]

\[ \text{FeOH}^+ + \text{OH}^- \rightarrow \text{Fe(OH)}_2 \]

\[ \text{Fe(OH)}_2(\text{solution}) \rightarrow \text{FeOH}^+ + \text{OH}^- \]

\[ \text{FeOH}^+ \rightarrow \text{Fe}^{2+} + \text{OH}^- \]

Reactions on the Al-anode:

\[ \text{Al}^0 - 3e^- \rightarrow \text{Al}^{3+} \]

\[ \text{Al}^{3+} + \text{H}_2\text{O} \leftrightarrow \text{Al(OH)}_2^+ + \text{H}^+ \]

\[ \text{Al(OH)}_2^+ + \text{H}_2\text{O} \leftrightarrow \text{Al(OH)}_3 + \text{H}^+ \]

\[ \text{Al(OH)}_3 + \text{H}_2\text{O} \leftrightarrow \text{Al(OH)}_4^- + \text{H}^+ \]

In anode:

\[ \text{M} \rightarrow \text{M}^{n+} + ne^- \]

\[ 2\text{H}_2\text{O} \rightarrow 4\text{H}^+ + \text{O}_2 + 4e^- \]

In cathode:

\[ \text{M}^{n+} + ne^- \rightarrow \text{M} \]

\[ 2\text{H}_2\text{O} + 2e^- \rightarrow \text{H}_2 + 2\text{OH}^- \]

In the process of the study, leachate of two different concentrations was taken. The first one corresponds to the plant leachate concentration, and the second one has been diluted as 1:2. Natural dilution of leachate takes place under raining or snowing.

3.1. Effect of Fe$^{2+}$ concentration

Fig. 2 shows the plot of removal of COD, TOC and color versus the Fe$^{2+}$ dose. It is apparent that with the increase in the

\[ \text{Al(OH)}_2^+ + \text{H}_2\text{O} \leftrightarrow \text{Al(OH)}_3 + \text{H}^+ \]

\[ \text{Al(OH)}_3 + \text{H}_2\text{O} \leftrightarrow \text{Al(OH)}_4^- + \text{H}^+ \]

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Fe²⁺ dose up to a certain level, the removal of COD, TOC and color increases.

As seen from Fig. 2, the best removal efficiency was seen in the COD parameter. Thus, the maximal removal was 87% for the 1:2 diluted solution (L1), while for the original solution (L2) it was 90%. High color removal is observed for L1 (86%), while for L2 it is approximately 99%. For both solutions, TOC indices are in the range of 58–68%.

The removal indices for the leachate treatment by chemical coagulants (FeSO₄·7H₂O) appeared to be lower than for the electrochemically generated Fe²⁺ (Fig. 3). Thus, the indices of the COD removal are 78 and 56% for L1 and L2, respectively. As to the rest two parameters (TOC and color), they are also lower than their corresponding values for the EC (Fig. 3).

The result tells in the favor of the higher adsorption ability of the electrochemically generated Fe²⁺ than of Fe(OH)₂, which is obtained from the FeSO₄ by hydrolysis. As the experience shows, the growth of CD to a definite level promotes the increase of the hydroxides sorption activity.

3.2. Effect of Al³⁺ concentration

As in the case of Fe electrodes, Al³⁺ that had been generated in the electrolytic cell was added to the reactor with coagulant to be blended with leachate later on. Similar to the experiments with Fe²⁺ two solutions were studied, namely L1 as 1:2 diluted one and L2 as the solution of original concentration. For the same removal leachate parameters (COD, TOC and color), the results on their dependence on the Al³⁺ dose are presented in Figs. 4 and 5.

As seen from Fig. 4, at the first stage (dose Al³⁺ is 0.12 g L⁻¹) the percentage of COD removal is 55% for L1 and 58% for L2. The further increase of concentration (the Al³⁺ dose is 0.3 g L⁻¹) causes the maximal treatment effect, as to the COD, which reaches 75% for L1 and 86% for L2. After this, the removal vol-

![Fig. 3. Percent of COD, TOC and color removal versus chemical generated Fe²⁺ (FeSO₄) dose (a) 50% solution (L1), (b) the original solution (L2).](image1)

![Fig. 4. Percent of COD, TOC and color removal versus electrochemically generated Al³⁺ (Al₂(SO₄)₃) dose (a) 50% solution (L1), (b) the original solution (L2).](image2)
ume changes incomensurably with the increase of concentration (the Al$^{3+}$ dose increases to 0.34 g L$^{-1}$).

Highly effective results are observed for color removal. Thus, by the dose of Al$^{3+}$ equal to 0.26 g L$^{-1}$, the color removal reaches the value of 98% for L2. On the other hand, the results showed that the Fe$^{2+}$ is not effective in TOC removal. The maximal value of the TOC removal is 78% for L1 and 65% for L2 by the maximal Al$^{3+}$ dose being 0.34 g L$^{-1}$.

To compare the effectiveness of the processes of electrical and chemical-coagulation (by the analogous concentrations of Al$^{3+}$), the solutions of Al$_2$(SO$_4$)$_3$:18H$_2$O were prepared and the process of the leachate purification was studied. Fig. 5 presents the resulting graph, where the common wastewater treatment effect is expressed in percents and shown in dependence on the values of Al$^{3+}$ dose.

In this study, investigating the treatment of municipal leachate by electro- and chemical-coagulation methods, it is observed that the sludge formed in chemical-coagulation is twice formed in electro-coagulation.

As seen from Fig. 5, the maximum of COD, TOC and color removal is reached when the Al$^{3+}$ concentration is 0.34 g L$^{-1}$. For L1, the removal is 73% for COD, 49% for TOC, and 76% for color parameters. For L2, these parameters have lower values and are correspondingly, 56% for COD, 46% for TOC, and 69% for color removal.

4. Conclusions

The efficiency of the chemical- and electro-coagulation processes applied for the leachate purification was studied in this work. By using these methods, COD, TOC, color parameters were investigated. The results have shown that EC processes provide higher efficiency than chemical-coagulation. When removal efficiencies are investigated, it is seen that electro-coagulation is an efficient pre-treatment method for the studied leachate.

By the electro-coagulation method, the maximal COD removal is 87% for the L1 solution, while for L2 it is 90% for Fe$^{2+}$ and 77% for L1 and 88% for L2 for Al$^{3+}$ ion solutions. High color removal is observed for L1 (86%), while for L2 it is approximately 99% for Fe$^{2+}$.

During electro-coagulation process electro-floatation and electro-oxidation take place simultaneously. In the process pollutants are oxidated and at the same time dissolved and suspended pollutants are carried to water surface. Existing of three mechanisms at the same time directly effects the removal efficiency.

The EC method does not use any chemical reagents and makes the process of leachate treatment easy for regulation and automation. A constructional optimization of the EC apparatus for the leachate treatment is offered. In our opinion, further studies can promote successive passing from laboratory-scale models to the industrial specimens possessing the same level of technological characteristics. The results show that the EC processes provide high and stable effects as to the contaminants removal.

Composition of leachates forming in landfills is effected by many factors such as seasonal changes, landfill operation type, waste composition, moisture content of waste and amount of leachate. As a result, different leachates form in each landfill so each study about different landfills can be evaluated as a sample case. At the same time variations in leachates cause diversity in treatment methods. So determination of appropriate method for the studied leachate is righter than generalization of efficiencies derived by methods used for leachate treatment.

References


