Treatment of Domestic Wastewater by Using Electrochemical Process Using Different Metal Electrodes

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Abstract

In recent years, operational and capital cost for the treatment facilities have become more important when sustainability is considered in relation to domestic wastewater treatment in developing countries. The electrochemical processes are one of the sustainable process. In this study, the removal of chemical oxygen demand (COD: 5760 mg/L), suspended solids (SS: 260 mg/L), total phosphorus (5.3 mg/L), nitrate nitrogen (1.1 mg/L) and nitrite nitrogen (0.24 mg/L) from domestic wastewater were experimentally investigated at 7.5 and 15V voltage and different processing times (15-30-60-120 min.) by using electrochemical processes. Electrical conductivity, temperature, and pH as experimental parameters were measured after the treatment. Aluminum, iron, titanium and graphite electrodes were used in the reactor. Samples were taken in certain time intervals and removal efficiencies were determined. The results showed that 80-99% COD removals were obtained by using iron electrode. Experimental results showed that the electrochemical process was also effective on SS and nutrients (total phosphorus, nitrate nitrogen and nitrite nitrogen) removal.

ABBREVIATIONS

COD: Chemical Oxygen Demand; SS: Suspended Solids; TP: Total Phosphorus; EC: Electrocoagulation Process

INTRODUCTION

Among of all wastewaters, Domestic wastewater is the most produced type of waste in the world. Although biological treatment is the conventional method for domestic wastewaters, treatability by using novel technologies have been studied cause of determinate effective variety of pollutants operating cost, operating difficulties and undesirably low efficiency.

In general mechanism of electrochemical treatment processes involve coagulation, adsorption, precipitation and flotation. The most significant difference is the form and structure of the process of electrochemical processes. Especially the electrode type is one of the factors that determines what the system trigger is electrocoagulation whether to electrooxidation [1].

Electrochemical treatment methods have been successfully used for the treatment of and leachate water [2], textile [3], slaughter [4], olive oil [5], yeast industry [6], tannery wastewater [7], bilge wastewater [8], pharmaceutical [9], dairy wastewater [10].

Electrocoagulation (EC) contains the production of coagulants in situ by electrically dissolving metal electrodes. The metal ion production occurs at the anode, while hydrogen gas from the electrolysis of water is produced at the cathode. The metal hydroxide ions absorb and trap contaminants while the hydrogen gas floats these particles [11]. Reactions that take place for the iron and aluminum in an electrocoagulation process are as follows:

$\text{Al(s)} \rightarrow \text{Al}^{3+} \text{(aq)} + 3e^{-}$  \hspace{1cm} (1)

$\text{Al}^{3+} + 3 \text{OH}^- \rightarrow \text{Al(OH)}_3$  \hspace{1cm} (2)

$\text{Fe(s)} \rightarrow \text{Fe}^{3+} \text{(aq)} + 3e^{-}$  \hspace{1cm} (3)

$\text{Fe}^{3+} + 3 \text{OH}^- \rightarrow \text{Fe(OH)}_3$  \hspace{1cm} (4)

Oils, suspended solids and organic compounds can be treated by electrofloation [11].

Electrofloation process does not occur alone, mostly observed with electrocoagulation process. Process is based on elimination of pollutant by the gas bubbles which were produced by electrodes. Reactions that take place on the electrode surface are shown below.

Anode: $2\text{H}_2\text{O} \rightarrow 2\text{H}_2\uparrow + 4\text{H}^+ + 4\text{e}^-$  \hspace{1cm} (5)

Cathode: $4\text{H}^+ + 4\text{e}^- \rightarrow 2\text{H}_2\uparrow$  \hspace{1cm} (6)
The electrooxidation is mainly used for degradation of: aromatic compounds [12], pharmaceuticals [13], paints [14,15], industrial pollutants [16], pesticides [17], waste [18] and other organics [19-21] by using inert electrodes. The advantage of this method is that finally products are mainly CO$_2$ and H$_2$O [22].

Electrochemical treatment methods are more favorable since the low volume of sludge is produced compared with the conventional chemical treatment methods. Electrochemical processes are usually applied at room temperature and the produced sludge is easily recoverable [23]. The electrocoagulation process is more applicable than the electrooxidation process because it affects various contaminants. Nevertheless electrooxidation is likely to be more preferable because the amount of sludge formed is almost negligible compared to the electrocoagulation as a result of conversion of the final product to CO$_2$ and H$_2$O.

In this study, the efficiency of electrochemical treatment process in removing of COD, SS and nutrient from domestic wastewater was examined. The effects of potential difference, types of used electrode and retention time on removal efficiency were investigated to determine the optimal operational conditions. At the same time, the effect of non-metallic graphite electrode on removal efficiency was investigated for comparison with metal electrodes.

**MATERIALS AND METHODS**

**Study area**

The wastewater sample which was taken from a domestic wastewater treatment plant in Cigli (Izmir, Turkey) was used in the experiments. Water sample only passed through a mechanical sieve that removed coarse material from water but concentrations of colloidal and dissolved material were not affected. The characteristics of the wastewater sample are given in Table 1.

**Electrochemical reactor**

Electrochemical treatments were studied in regular beherglass. Water was stirred with a standard magnetic stirrer during the process. Treatment by electrocoagulation/ electrofloation occurs while aluminum and iron materials used as electrodes in reactor and treatment by electrooxidation takes place while graphite and titanium electrodes used in reactor. Aluminum-iron electrodes were in the form of plate and scaled as 6 cm * 9 cm. Graphite-titanium electrodes were in the form of stick and scaled as 10 cm * 1 cm$^2$. All wastewater samples settled 20°C before electrochemical process. The inherent pH of the wastewater sample was measured as 7.7. The voltages in the experiments were set as 7.5V and 15V. Samples withdrawn during the study in different time intervals (15, 30, 60, 120 min) were subjected to analyses for determining COD, SS and nutrients. A laboratory scale reactor was used for the EC process as shown in Figure 1.

Distances between the electrodes are 6.5 cm and electrodes were placed as 80-90% of surface area stay in water. After electrochemical treatment, the electrodes were washed with 6% HCl solution and distilled water for the removal of the residual on the surface and weighed.

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Concentration</th>
</tr>
</thead>
<tbody>
<tr>
<td>Chemical oxygen demand (COD) (mg O$_2$/L)</td>
<td>5760</td>
</tr>
<tr>
<td>Suspended solids (SS) (mg/L)</td>
<td>260</td>
</tr>
<tr>
<td>Nitrate (NO$_3$-N) (mg/L)</td>
<td>1.1</td>
</tr>
<tr>
<td>Nitrite (NO$_2$-N) (mg/L)</td>
<td>0.24</td>
</tr>
<tr>
<td>Total phosphorus (TP) (mg/L)</td>
<td>5.3</td>
</tr>
<tr>
<td>pH</td>
<td>7.7</td>
</tr>
<tr>
<td>Electrical conductivity (mS / cm)</td>
<td>5</td>
</tr>
<tr>
<td>Salinity (ppt)</td>
<td>1</td>
</tr>
</tbody>
</table>

**Methods of analysis**

Water samples were stored after pH, conductivity and temperature were measured by using standard laboratory devices. The organic matter removal efficiency was determined by analyzing COD. 5220 D Method, (APHA, 2005) was used to determine chemical oxygen demand and it was repeated twice. Nutrient content of samples were analyzed using a Merck Nova60 spectrophotometer. Suspended solids were analyzed based on standard methods for both raw wastewater and treated water with EC processes.

**RESULTS AND DISCUSSION**

This study is mainly aimed to determine basic operation parameters of treatment of domestic wastewaters. Therefore, COD, SS and nutrient were investigated in terms of selection of electrode material, applied voltage and retention time in order to determine optimum operating conditions for maximum removal efficiency of COD, SS and nutrient.

Removal efficiency (RE) equation is shown below:

$$RE\% = \frac{A - B}{A} \times 100$$

Where A is initial COD and B is current result.

**Changes on pH, electrical conductivity, temperature**

The changes during process operation are also important as much as start up parameters. The changes in pH, temperature and electrical conductivity were measured in 15, 30, 60, 120 minutes.

The initial pH was not changed; all electrodes were studied.
at pH 7.7. But as a result of reactions taking place in the reactor, pH values were measured for purpose of control. pH values were change between 6.68 to 9.16 as shown in Figure 2. For both voltage values, pH get higher than initial pH when iron electrode was used. pH value was almost the same as initial pH when titanium electrode was used. pH values were get lower than initial pH where aluminum and graphite electrodes were used.

Although there is no negative effect of temperature, some studies about effect of temperature on electrochemical treatment methods show that amount of removal efficiency increase as the temperature increases [24].

Figure 3 show that temperature increases for four electrodes as time passes. Above all temperature was 57°C in reactor when voltage was 15V and aluminum electrode was used.

For the iron, aluminum, titanium and graphite electrodes, Figure (4(a) and (b)) shows the effect of electrochemical treatment processes on the changing of the wastewater conductivity.

As seen, operation of process caused an increase dissolved ions as from 15 minutes. And results indicate that changes in electrical conductivity is not related with pH.

**Chemical oxygen demand removal**

To optimize the electrocoagulation operating conditions, the COD was measured as a function of treatment time and different electrodes. The reduction of the COD as a function of treatment time under conditions of electrical conductivity 5 mS/cm pH 7.7 and reactor voltage 7.5V is shown in Figure 5 for four electrodes.

There is a rapid increase in the COD for the first 15 min of treatment then a limiting value is reached with no further improvement.

To determine effect of voltage, reactor was operated at 15V while other parameters were the same. The results of this operation were given in Figure 6.

When we examine the literature and check other studies [25,26], COD removal efficiencies are almost the same for domestic wastewater if we compare about the same conditions.
Figure 4 Changes in electrical conductivity on four electrodes for 7.5V (a) and 15V (b).

Figure 5 COD removal efficiency at 7.5V as a function of time for iron, aluminum, titanium and graphite electrodes.

Figure 6 COD removal efficiency at 15V as a function of time for iron, aluminum, titanium and graphite electrodes.
like time intervals and iron electrode. But in this study removal efficiencies are remarkable with aluminium, titanium, graphite electrodes and also results show us that removal efficiency gets higher enough with iron electrode when the reaction time is increased 60 to 120 minutes.

The effluent treatment of titanium electrodes was found to be more clear and stable than the others. The removal efficiency with iron, aluminium and graphite electrodes was highly desired values in 15 minutes whereas removal efficiency results changes due to inorganic pollution in 30, 60 and 120 minutes retention time.

The effect of applied voltage is an important parameter for organic matter removal in EC process and also causes undesirable decomposition of the electrode. As in Figure 5 and Figure 6 7.5 voltage is provide sufficient energy to ionize metal ions which has better removal capacity than 15 V.

### Suspended solids removal

The relationship between the SS removal efficiency and retention time for four electrodes used is depicted in Figure 7 and Figure 8 under 7.5V – 15 V operation voltage. The figure shows that as the contact time increased from 15 to 120 min, concentration of suspended solids varies from 40 mg/L to 2600 mg/L for both voltages of 7.5V and 15V as a function of time.

In 15 voltage operation conditions, aluminum ions were over dissolved in reactors and cause increase in dissolved solids amount after 60 min for iron and 30 min for aluminum. These results show that the applied voltage is high enough to cause over dissolved metal ions which effect suspended solid results. However, Titanium electrode was extremely efficient at removing suspended solids after 60 minutes reaction time as seen at Figure 7 and Figure 8. These suspended solid results were under limit of Turkish Domestic Wastewater Discharge Standards [27].

### Nutrient removal

<table>
<thead>
<tr>
<th>Sample</th>
<th>Total P (mg/L)</th>
<th>NO₃ (mg/L)</th>
<th>NO₂ (mg/L)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Raw Wastewater</td>
<td>5.3</td>
<td>1.1</td>
<td>0.24</td>
</tr>
<tr>
<td>Iron</td>
<td>1.65</td>
<td>0.3</td>
<td>0.87</td>
</tr>
<tr>
<td>Aluminium</td>
<td>0.8</td>
<td>1.5</td>
<td>1.84</td>
</tr>
<tr>
<td>Titanium</td>
<td>4.7</td>
<td>1.1</td>
<td>1.73</td>
</tr>
<tr>
<td>Graphite</td>
<td>1</td>
<td>5.8</td>
<td>2.15</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Sample</th>
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<th>NO₂ (mg/L)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Raw Wastewater</td>
<td>5.3</td>
<td>1.1</td>
<td>0.24</td>
</tr>
<tr>
<td>Aluminium</td>
<td>0.254</td>
<td>5.5</td>
<td>1.15</td>
</tr>
<tr>
<td>Titanium</td>
<td>2.8</td>
<td>4</td>
<td>1.68</td>
</tr>
<tr>
<td>Graphite</td>
<td>2.3</td>
<td>13.4</td>
<td>3.38</td>
</tr>
</tbody>
</table>

Abbreviations: COD: Chemical Oxygen Demand; SS: Suspended Solids; TP: Total Phosphorus

Nitrogen in domestic wastewater exists as ammonium or organic form, both may be solid or dissolved. Only 30% of total nitrogen is removable with conventional biological treatment methods [28].

%90 of total phosphorus is dissolved in wastewaters. Removal of dissolved phosphorus is quite difficult with the conventional biological treatment methods [29].

Due to this facts, nutrient removal efficiency was investigated during this research. Total nitrate, nitrite and phosphorus concentrations were analyzed after 120 minutes reaction time under 7.5V and 15V voltage in reactor as shown in Table 2 and Table 3.

In 15V process operation, iron electrodes over dissolved after 120 minutes reaction time and react with chemicals during analysis. These results show that 120 minutes reaction time is more than it should be. For the other electrodes, aluminium electrode was quite effective at removing nutrients from
domestic wastewaters.

CONCLUSION

This study aimed to determine optimum electrode type, applied voltage and reaction time for removal of COD, SS and nutrients in domestic wastewater. Domestic wastewater was successfully treated by electrochemical processes in a very short time (15 min.). These results show that aluminium was effective at removing COD and nutrients with 15 minutes reaction time however need more sedimentation time to provide desired values at suspended solids removal. Although graphite is more limited conductive quality, results show that graphite electrodes highly effective for COD, SS and nutrients removal and with enough reaction time (60 min.) can provide dischargeable wastewater.

When the results are compared with the domestic wastewater discharge standards in Turkey, it seems that COD, SS and pH are parameters which to be controlled. It is seen that the data obtained as a result of the study provide the desired conditions.

Based on these results, electrochemical treatment methods are effective method of treatability of domestic wastewater and also the results obtained from the graphite electrode seem to be promising in terms of realizing other studies.

REFERENCES


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Cite this article