Proceedings of the 4th World Congress on Civil, Structural, and Environmental Engineering (CSEE'19)
Rome, Italy – April 7-9, 2019
Paper No. ICEPTP 152
DOI: 10.11159/iceptp19.152

Wastewater Treatment by Electrocoagulation: A comparative study using different anode materials

Khaled Zaher[1], Abdelsalam Elawwad[1], Rana Nadeem[2]
[1]Environmental Engineering Dept., Faculty of Engineering, Cairo University
El-Gamaa St., 12613 Giza, Egypt
elawwad@cu.edu.eg, dr.khaledzaher@gmail.com
[2]Sanitary and Environmental Engineering Institute, Housing and Building Research Center
87 Tahir St., Dokki, 11511 Giza, Egypt
rona.rose@hotmail.com

Abstract **-** This study investigated COD removal by electrocoagulation in both synthetic and real wastewater using different anode material. Experiments were conducted in a batch-scale EC cell using aluminium, iron and copper electrodes as anode and stainless-steel electrode as cathode. Effects of different parameters including pH (4, 7, and 10), time (10 to 60 min), voltage (3 to 12 V) were studied in order to evaluate the efficiency of electrocoagulation process. The initial synthetic COD concentration was 1000 mg/l. The highest COD removal efficiency of 97.7% was observed by aluminium electrode under the following conditions: pH = 7, time of 60 min, and voltage of 12 V. Moreover, it was observed that most of COD removal happened during the first 10 min (about 86%) then the removal rate decreased. For real wastewater, the highest COD removal efficiency was 91.9% which achieved by using aluminium electrode, pH =7.4, time of 60min, and voltage of 12 V. The results showed the feasibility of electrocoagulation process for the treatment of high-strength municipal wastewaters.

Keywords**:** Electrocoagulation, Wastewater treatment, Chemical Oxygen Demand, Anode material.

1. Introduction

Wastewater contains high organic and nutrient concentrations. So, direct discharge of raw untreated sewage into the water body is one of the main sources of pollution. The main objectives of wastewater treatment are protecting the environment, conserving fresh water resources and wastewater reuse [1]. Nowadays, more effective methods are required to treat a wide range of pollutants. Electrocoagulation (EC) is a promising process in which a sacrificial metal anode produces electrically active coagulants. EC has many advantages as it is fast, simple and cost-effective method. Moreover, it requires lower maintenance cost and needs less labour [2].

EC involves many chemical and physical phenomena that use consumable metal electrodes to supply ions into the water stream. Electrodes are classified into cathode and anode based on the type of reaction occurred on its surface where, reduction occurs at the cathode, while oxidation occurs at the anode [3]. The amount of electricity that passes through the cell governed the amount of substance that undergoes oxidation or reduction at each electrode [4]. Metal anodes continuously scarifies metal ions, which immediately hydrolyse to polymeric metal hydroxide. Pollutants present in wastewater are treated either by chemical coagulation or by attachment to coagulated particles. They are then removed by electro-flotation, sedimentation, or filtration.

The literature indicates that EC can be used to treat water containing pollutants such as: heavy metals, textile wastewater, oily wastewater, food industry, potable water, urban wastewater, oil wastes, Paper Mill Wastewater, and many other pollutants [5-9].

In this study, the authors want to investigate the efficiency of EC process in treating municipal wastewater. EC is a simple technology, needs small footprint and isn't limited by the existence of toxic compounds or pH. So, it can be used in rural areas which suffer in many countries from lack of appropriate sanitation services [10]. The aim of this study is to compare between different anode electrode materials including Aluminum (Al), Iron (Fe), and Copper (Cu) in EC process used for the removal of chemical oxygen demand (COD) in wastewater.

2. Methodology

The bench-scale EC cell was constructed for the experimental works as shown in figure1. EC reactor consists of one litre Pyrex glass beaker and two electrodes (15 cm high, 5 cm wide and, 0.5 cm thickness) which were fixed vertically and parallel to each other and they are connected to a variable power supply (0 to 12V). The configuration of the cell is a monopolar configuration and the internal spacing between the two electrodes was 4 cm. The cathode was stainless steel while the anode different (Al, Fe or Cu) according to the experiment. The solution was mixed using a magnetic stirrer adjusted to 250 rpm. After each experiment, the electrodes were washed using an acid bath (4% HCl) and they were carefully cleaned using steel wool to remove any oxides that may have been formed on the surface. All experiments were conducted at room temperature of 24±2 °C.

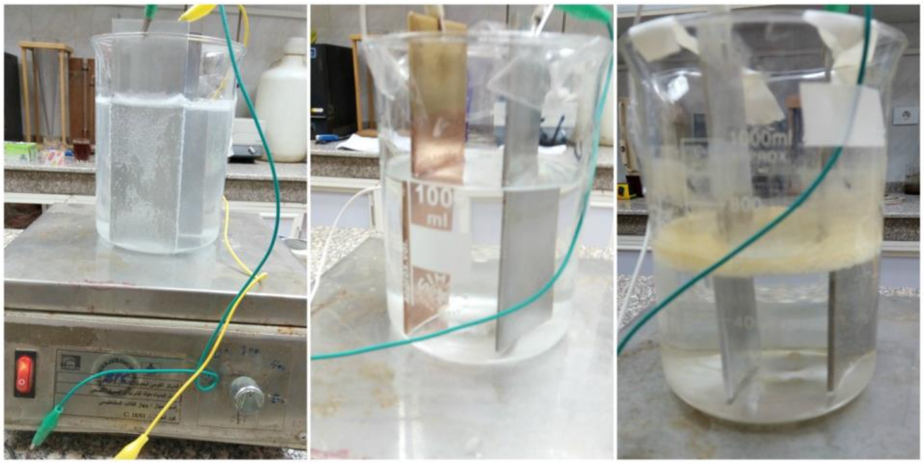

Fig.1: The batch-scale EC cell.

Table 1: Characterizations of synthetic and real wastewater.

Parameter	Synthetic Wastewater	Real Wastewater
pH	7	7.22
COD mg/l	1000	397
BOD mg/l	NA	217
Ammonia mg NH_4/l	40	10.5
Nitrates mg NO_3/l	10	0.41
Phosphorus mg/l	10	4.5
Total solids mg/l	1580	710
TSS mg/l	580	198
TDS mg/l	500	512
Conductivity µs/m	1110	987
Fe mg/l	Zero	0.1
Al mg/l	Zero	Zero
Cu mg/l	Zero	Zero
Total Alkalinity mg/l	NA	264

Synthetic wastewater and domestic wastewater were used in the experiments. First experiments were carried out to study the removal of COD in model wastewater. The synthetic wastewater used was prepared using distilled water contains phosphate, nitrates, 1.5 g/L soluble starch. Sodium chloride (NaCl) was added with 0.5 g/L concentration as a supporting electrolyte to increase conductivity so the power consumption decreases, and the removal efficiency increases electrolytes. Although there are different electrolytes that can be used, NaCl is the best choice due to its low cost and its availability. All chemicals used in the study were analytical grade. pH values were adjusted using 1N sodium hydroxide (NaOH) and 1N hydrochloric acid (HCl). All experiments were performed at Temperature 24°C ± 2 and to adjust neutral pH= 7.0 except when studying the effect of initial pH. The characteristics of synthetic wastewater were shown in Table 1.

Further experiments were carried out using real wastewater to study the efficiency of COD removal with optimum conditions obtained during synthetic wastewater experiments. The real wastewater used in the study was obtained from the influent wastewater flow of Zenin wastewater treatment plant, Egypt with the characteristics shown in Table 1.

All samples were collected using sterilized pipettes at regular 10 minutes interval of time (from 10 to 60 minutes). Each time a sample with volume of 5 ml was taken then filtered with Whatman filter paper before analysis. COD was measured for all samples, while, total dissolved solids (TDS), total suspended solids (TSS), conductivity, and pH were measured at the start and end of the experiment. COD was measured in terms of mg/l using UV/VIS Spectrometer (PG Instruments Ltd). The analysis model was the closed reflux method at λ 600 nm according to Standard methods for examination of water and wastewater [11]. Moreover, Al, Fe, and Cu concentrations were measured according to the electrode used at the end of each experiment. The removal of pollutants was calculated from the COD values by using Equation (1), where COD_o and COD_t are COD at start and at time t, respectively.

$$\text{Removal \%} = (1 - \frac{COD_o}{COD_t}) \text{ x } 100 \quad (1)$$

3. Results and discussion

3.1. Effect of Initial pH of the solution on COD removal

The effect of solution pH on COD removal efficiency of EC process was investigated with initial pH ranged from 4 to 10. The results showed that Al electrode has higher efficiency near to neutral pH. At pH = 7, COD removal efficiency of 97.7% was obtained after 60 min, while at pH = 10 the removal efficiency was 95.4% and at pH = 4 was 92.2%. Also, the same For Fe electrode, the best performance was also at pH =7 with COD removal efficiency 96.8%, while at pH = 10 the removal efficiency was 93.6% and at pH= 4.0 was 92.2%. The lower performance at low pH values for Fe electrode might be due to the less amount of iron insoluble precipitates which are responsible for coagulation. While lower performance for Al electrode at low pH values, Al species will be in the form of Al^{3+} and $Al(OH)^{2+}$. Therefore, effective electrocoagulation did not happen in liquids with low pH [3, 12, 13]. In contrast, the best performance for Cu electrode was at Alkaline media (pH = 10). At pH = 10, COD removal efficiency was 95.5 %, while it was 94.6% and at pH = 7 and only 91.9% at pH= 4.0. Different values of COD removal at different pH might be due to the quality and quantity of copper hydroxide ions formed.

Values of pH of the effluent wastewater at the end of each experiment are shown in Table 2 at voltage of 12V. The value of pH changed due to different Al/OH- ratios produced by EC cell. When initial pH was 4, and 7, the pH increased and this due to OH ions generated by electrolysis followed faraday`s law. When initial pH was 10, the value of pH decreased due to the H_2 (hydrogen) production leading to a decrease in pH.

Table 2: pH values after 60 minutes.

Electrode	Initial pH = 10	Initial pH = 7	Initial pH = 4
Al	8.6	7.6	5
Fe	8.7	7.4	5.2
Cu	8	7.5	5.1

3.2. Effect of Time on COD removal

In this study, the effect of Time was studied in the range of 0 to 60 min. Under operating conditions: 1000 mg/l initial concentration of COD, voltage of (3, 6, 9 and 12 V), and pH = 7, using different electrodes as anode (Al, Fe, and Cu). Figures 2, 3, and 4 shows the relation between COD removal and time (in minutes) with different volts and electrodes. The figures show that COD removal increases with increase in time. By increasing EC time, more metal hydroxides generate from anode, and thus the generation of flocs increases so the coagulation process become better and removal efficiency become higher. Also, the change in the removal with further increase in time for more than 10 min is observed to be small. COD removal efficiency after 10 min using Al electrode as anode at pH = 7 and voltage =12V was 86.3% and after 60 min was 97.7%. By using Fe electrode under the same operating conditions, COD removal efficiency after 10 min was 86.3% and after 60 min 96.8%. Finally, by using Cu electrode, COD removal efficiency after 10 min was 84% and after 60 min was 96%. For this reason, the value of 10 min could be taken as the optimum value for EC process time for the three electrodes. The pollutant removal efficiency does not increase after a certain time as enough flocs are available for the removal of the pollutant.

3.3. Effect of voltage on COD removal

As shown in figures 2, 3, and 4, the voltage and current have a main important role in increasing the removal efficiency at the same pH. By using Al electrode as anode with constant pH = 7, time 60 min, COD removal efficiencies were 92.4, 94.6, 95.9, and 97.7% at voltages of 3, 6, 9, and 12V, respectively. By using Fe electrode as anode under the same operating conditions, COD removal efficiencies were 91.1, 93.5, 95, and 96.8 respectively finally by using Cu electrode as anode the removal efficiency of COD was 90.3%, 92.9%, 94%, and 96% at voltages of 3, 6, 9, and 12V, respectively. The current induced at 3, 6, 9 and 12V were observed as 0.1, 0.3, 0.6 and 0.9 A, respectively.

Current density determines both coagulant dosage and bubble generation rates and influences both solutions mixing and mass transfer at the electrodes [14]. COD removal efficiency increased with increase in voltage due to the increase in metal ions dissolute from anode. According to Faraday's law, as metal hydroxide increases, it adsorbs more COD. Also, it is well known that voltage and current density determines the production rate of coagulant, adjusts also bubble production, and hence affects the growth of flocs [5].

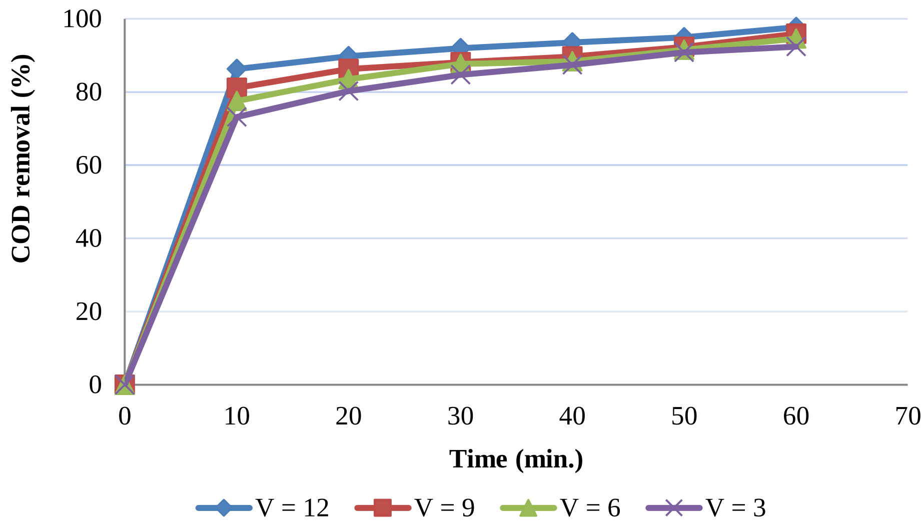

Fig. 2: COD removal over time using Al electrode as anode at different voltage, initial pH = 7, and initial COD =1000mg/l.

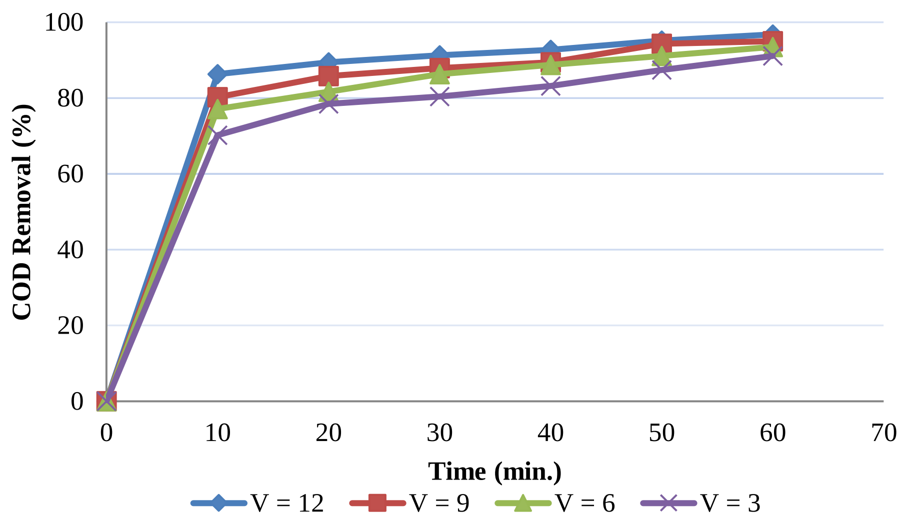

Fig. 3: COD removal over time using Fe electrode as anode at different voltage, initial pH = 7, and initial COD =1000mg/l.

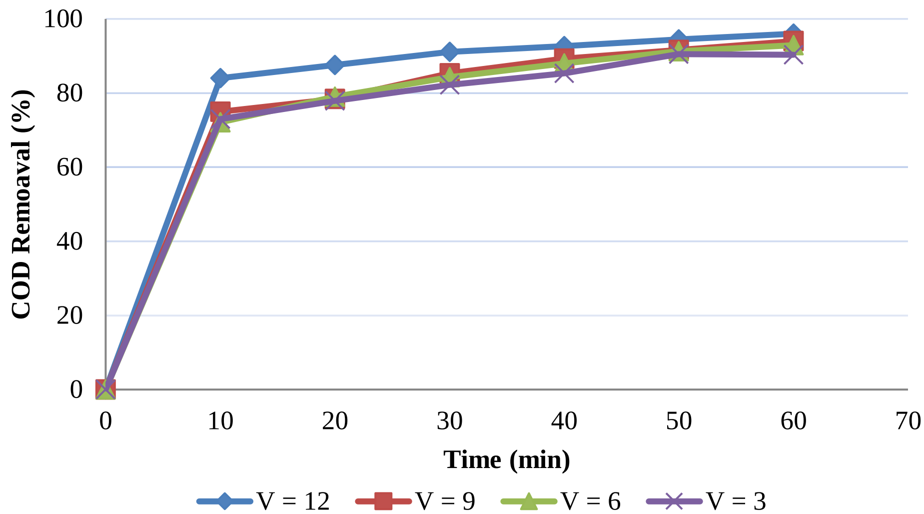

Fig. 4: COD removal over time using Cu electrode as anode at different voltage, initial pH = 7, and initial COD =1000mg/l.

Al, Fe, and Cu electrodes were used because they are cheap, so it can be used in large-scale plants. The maximum COD removal efficiency was 97.7, 96.8, and 96% for Al, Fe, and Cu, respectively. The result showed that the Al electrode is better than Fe and Cu electrodes. For economical point of view, Fe is cheaper than Al, however, Fe give yellow colour in water before filtration which mean the existence of dissolved iron salt in the solution.

3.4. Case Study with Real Raw Wastewater

The system was operated for 60 minutes to treat real raw wastewater with the characteristics shown in Table 1. Different electrodes of Al, Fe, Cu where examined at the optimum voltage 12V. The maximum COD removal efficiency was 91.9 % and obtained using the Al electrode while the removal efficiency was 90.4% for Fe electrode and 90.7% Cu electrode. Also, the values of pH of the treated real wastewater at the end of each experiment were increased. This increase in pH value might be due to H_2 evolution at cathodes and formation of metal hydroxides at the anode which increase the concentration OH ions in water. pH increased from 7.2 to 7.8, 7.6, and 8.2 for Al, Fe, Cu electrodes, respectively.

Overall the performance of three electrodes on treating real wastewater is shown in Table 3. The table shows the removal efficiency of COD, TDS, and TSS. The three electrode materials have almost the same removal efficiency for TSS and BOD. Ammonia removal was poor where it was reduced from 10.5 mg/l to 9.1, 9.2, 8.5 mg/l for Al, Fe, and Cu electrodes. The concentration of Aluminium, Fe, and Cu in the treated effluent water was determined. Al concentration in the effluent water was 0.14 mg/l when using Al electrode, while Fe reduced from 0.1 mg/l to 0.05 mg/l when using Fe electrode. By using Cu electrode as the anode, Cu in the effluent water was 0.95 mg/l. From these results, use of copper is not recommended for Electrocoagulation as elevated copper concentration could be restricted by environmental regulations.

Raw data presented in this paper will be used for a further kinetic study. Further data collection is required to determine exactly the sludge quantity produced by EC when treating municipal wastewater as the sludge handling represents a high portion of the operational costs. Disposal of sludge without treatment could cause negative environmental and public health problems [15].

Table 3: Comparison between the three electrodes when treating real wastewater.

Electrode/ Parameter	Cu	Fe	Al
	Removal efficiency %		
COD	90.7%	90.42%	91.9 %
BOD	92.2%	88.5%	88.5%
Ammonia	13.3%.	12.4%	18%
TDS	54.9%	43.2%	48.6%
TSS	90.4%	89.9%	90.4%

4. Conclusion

EC is an efficient treatment process which needs less footprint compared to conventional chemical coagulation. Moreover, unlike biological treatment that can be inhibited by the existence of toxic compounds or pH [16], EC can be used for the treatment of industrial wastewaters or in WWTPs that receive mixed domestic and industrial wastewaters. The present study investigated the efficiency of a batch-scale EC cell as a simple process to treat synthetic and real wastewater using with different anode materials (Al, Fe, and Cu). The experimental results showed that COD removal efficiencies increased by increasing the Time, conductivity and the voltage. The highest COD removal efficiency achieved at pH= 7. It was found that the difference electrodes have different effectiveness in COD removal and an Al electrode was the most effective electrode in the treatment of synthetic and real wastewater with removal efficiency up to 97.7% and 91.1% for synthetic and real wastewater, respectively.

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