

Short Communication

Evaluating the Effectiveness of Electrocoagulation (EC) Process in Reducing the COD of Simulated urban Runoffs

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A large scale of precipitation amounts in urban areas becomes runoff because of increasing proportion of impermeable surfaces relative to natural permeable surfaces. The runoff leads to numerous concerns in urban areas by carrying pollutants from urban surfaces, creating a major source of contaminants. Further concerns were raised on the quality of runoffs by observing their negative impacts on the river and lakes in the downstream and surrounding areas of the city, especially in the early rainfalls that occur after a period of no rainfall. The increased runoff contamination can cause organic shock in conventional wastewater treatment systems, such as activated sludge. The electrocoagulation (an electrochemical process) is an effective and rapid method for treating water-soluble salts. The present study was conducted to investigate the effects of reaction time, voltage and pH on reducing the COD levels of runoff collected from surfaces of highly trafficked urban streets in Torbat-e Heydarieh, Iran. The results of this study showed that this process was able to remove the COD, BOD and turbidity with maximum efficiency of 80, 82 and 92% in optimal conditions (reaction time of 60 min, pH of 10.4, voltage of 30 volts). However, this maximum removal was different for varied samples collected before humps, areas near to squares and the border of main streets. Investigating the reaction time variable determined that a large part of COD was removed in the first 15 minutes at 10 volts. In addition, the highest level of metal consumption was reported to be 0.9 g/cm at 20 volts. This study showed that the electrocoagulation can be used as a pre-treatment method for urban runoffs before delivery to conventional wastewater treatment processes or receiving waters.

Keywords: runoff, electrocoagulation, COD

1. INTRODUCTION

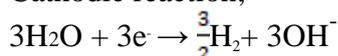
As the United Nations reported in 2017, the world population is projected to grow from 5.3 billion to 8.6 billion by 2030 [1]. Nowadays, the growth of urbanization has dramatically changed the

developmental process and population of the cities. A large scale of precipitation amounts in urban areas becomes runoff because of increasing proportion of impermeable surfaces relative to natural permeable surfaces. The runoff leads to numerous concerns in urban areas by carrying pollutants from urban surfaces. However, unfortunately, this issue still remained unmanaged so far. Problems related to runoff management arise from two main phenomena; the first is an increase in the volume of runoff on impermeable surfaces, and the second is high concentration of pollutants in the runoff [2]. The runoffs are often considered as an opportunity in urban management systems, and could potentially be reused based on their quality[3].

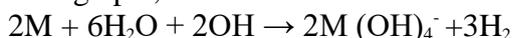
Estimation of the pollutant load of surface runoffs requires a detailed understanding of the nature of pollutants and the distribution of rain in different regions of runoff[4]. Several studies introduced the urban runoff as a major source of contaminants. So far, more than 6000 pollutants, including sediments, heavy metals, chlorides, hydrocarbons, nutrients, nitrogen, phosphorus, pesticides and pathogens have been detected in runoff, and the list is still expanding [5]. The results of Taebi and Droste (2003) by comparing the urban runoff and sanitary sewage in Isfahan, Iran, indicated that the amount of organic matter and suspended solids was higher than the permitted limits for discharge in the environment [6]. Gromaire-Mertz¹ et al. conducted a study on the characteristics of urban runoff contaminants in Paris. Shaver² et al. investigated the principles of urban runoff management; the most important pollutants identified in these studies included various metals, organic chemicals (Pesticides, petrols, oils, etc.), pathogens (viruses, bacteria and protozoa), nutrients (nitrogen and phosphorus), oxygen demanding organic matters (grass, leaves, hydrocarbons, human and animal wastes), sediments (soil, gravel, sand, silt), salts (sodium chloride and calcium chloride). The quantity and quality of urban runoff depends on varied factors, including volume and intensity of the rainfall, the number of previous dry days, geographical and geological characteristics of the region, land use, traffic volume, repair and maintenance, and the status of the drainage system [7]. Another study by Characklis and Wiesner determined the water quality, metal concentrations and the range of particle size, as well as the correlation of some metals such as zinc and iron with organic carbon and macrocolloids in urban runoff [8]. The most contaminants of the urban runoffs studied by previous investigations primarily are metals, while there are few studies evaluating organic matters in these water resources [9-11]. Studies have also suggested that reducing contaminants, as much as possible, or providing some permeable surfaces for runoffs are better solutions than attempting to prevent flooding in urban areas [12]. Negative influence of these runoffs on downstream receiving waters and the surrounding regions had raised concerns about the quality of runoffs [6, 13]. The weather and climate could affect the quality and quantity of surface runoff; for instance, the regions with low rainfall could experience highly concentrated runoffs. Since reuse and treatment of sewage before releasing in the environment is inevitable, the control and removal of environmental pollution are always accompanied by huge costs[14]. The organic matters usually consist of carbon (C), hydrogen (H) and oxygen (O) and nitrogen (N) in some cases, Other elements, such as sulfur, phosphorus and iron could also be found in the organic matters. The measurement of chemical oxygen demand (COD) is one of the laboratory methods to calculate high concentrations of organic matters in the sewage [15, 16]. Given the population growth and consequent increase in the urban impermeable surface area (ISA), resulting in higher loads of pollution caused by the runoffs, it is necessary for policy makers to take steps to

properly manage these water resources. The treatment and reduction of pollution load in such water resources are of the appropriate solutions in this regard. Marla Maniquiz et al. attempted to accumulate and remove the suspended solids and multiple heavy metals using filtration and sedimentation mechanisms [17]. Surya Pandey et al. reduced and removed contaminants with the help of layers of selective filters [18]. Over the last few years, advanced electrochemical oxidation processes (AEOPs) have been used as alternatives for the physical and chemical approaches of sewage treatment and removal of organic pollutants [19, 20]. The electrocoagulation (EC) is one of the electrochemical oxidation methods, which is a combination of physical and chemical treatment processes, involving reactions such as coagulation, flocculation, deposition and flotation. In addition, the EC is an effective and rapid method for treating water-soluble solids, in which the coagulation performed through an ion-exchange mechanism by the help of an anode plate [21-24]. On the other hand, no application of EC on the treatment or pretreatment of urban runoff has been investigated so far, while this method has been widely used in removal of the COD and BOD from the wastewaters as well as turbidity and dye from industrial wastewaters [19, 25-27]. The electrochemical process is an environment-friendly method, whose low cost makes it competitable than other treatment methods. The cost-effectiveness of equipment and operation, lower sludge production with low water content and high capacity of water absorption, and the need for little space for installation are of the advantages of this method. If the iron and aluminum electrodes are utilized in the EC process, Al^{3+} and Fe^{2+} ions (from the anode) could penetrate into the electrolyte [28-30]. These metal ions are electrically charged and possess high absorption properties, resulting in the flocculation due to the combination of metal hydroxides and contaminants that allow them to easily settle or float. The electrochemical reaction involved in the reactor can be written as following [31-36].

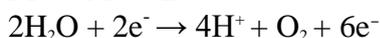
Cathodic reaction;



At high pH,



Anodic reaction;



M is the metallic anode and n is the number of electrons exchanged during the oxidation-reduction reaction [28]. The city of Torbat-e Heydarieh in Khorasan Razavi province, Iran, was selected as the study area, where has a homogeneous population structure as the residential and commercial sections of this city are not specifically separated. On the other hand, the industrial context of this city is outside of the urban area; as a result, the runoff generated in this city during rainfall has relatively similar characteristics in most urban areas.

2. EXPERIMENTAL

2.1 Material

All of the required chemicals were Merck (Germany), and the sampling method was the same for all three sampling sites. Sampling was carried out at intervals of at least 15 days from the last rainfall.

2.2 preparing artificial samples

The current pilot experiment was conducted on the surface runoffs simulated from the highly trafficked streets of Torbat-e-Heidarieh in a batch system during May to July 2017. Compositesampling was performed in three main points, including before humps (Sample 1), near the squares (Sample 2), and the border of main streets and parking lots (Sample 3).

2.3 Experimental procedure

At the beginning of the sampling, the samples were collected after 5 minutes flowing of runoff on the asphalt surfaces to avoid the effect of the first flush. The raw samples were analyzed to measure COD in accordance with the method 5220 presented in the Standard Methods for the Examination of Water and Wastewater book following the determination of parameters such as pH, turbidity, EC, and temperature. To assess the effect of electrolysis, the samples were poured into a 2-liter batch reactor equipped with 7 iron electrodes with a 3-cm electrode spacing, and electrode effective surface of 104 cm². A direct current power with ATTEN APS30056 model was used to supply electrical current. The electrodes were plugged into the power supply by a dipole connection. After adjusting the voltage of the device to 10, 20, and 30 volts, and settling time (5 min), the samples were taken after 15, 30, 45 and 60 min from the middle of the reactor. In order to evaluate the COD reduction efficiency of simulated runoff, the samples were collected at these times and voltages and analyzed to measure pH, turbidity, EC, temperature and COD in accordance with the Standard Methods for the Examination of Water and Wastewater book. The COD level of the specimens was determined by the spectrophotometer (DR 5000) at a wavelength of 600 nm. Table 1 represents the baseline analysis of runoff samples before processing by the electrocoagulation.

Table 1. Baseline chemical analysis of runoff samples before processing by electrocoagulation, sampling area

initial Total suspended solid (mg/lit) (Mean± Std. Deviation)	initial BOD5 (mg/lit) (Mean± Std. Deviation)	initial turbidity(23) (Mean± Std. Deviation)	initial Ec (µs/cm) (Mean± Std. Deviation)	initial pH (Mean± Std. Deviation)	Initial COD (mg/lit) (Mean± Std. Deviation)	sampling sites
274 ± 4.37	364 ± 12.52	159 ± 4.58	2120 ± 19.97	7.6 ± 0.11	639.9 ± 21.15	before humps(sample1)
312 ± 5.56	579 ± 13.07	248 ± 4.58	1380 ± 14.79	7.15 ± 0.32	968.64 ± 11.16	areas near to squares(sample2)
411 ± 6.24	516 ± 8.1	209 ± 4	2600 ± 16.37	7.3 ± 0.18	981.26 ± 13.98	border of main streets(sample3)

2.4 Energy and Electrode consumption

The amount of energy consumption (*kWh/m³*) in this study was obtained according to equation of $IVct/VR$; where, t is the reaction time (hours), Vc is the input voltage to the electrochemical cell (volts), I is current intensity (ampere), and VR is the electrolyte volume (*m³*) [37,

38]. The amount of laboratory consumption of metal used (electrode) was obtained from the difference in electrode weight before and after processing by electrocoagulation. Longitudinal analysis, marginal model, and analysis of variance with a significance level of 0.05 were used in SPSS20 software for analyzing the data.

3. RESULTS AND DISCUSSION

3.1 Simultaneous evaluation of time variations and potential differences in COD removal efficiency

The findings showed that the COD removal efficiency was improved by an average of 25% for an increase of 10 units of voltage. The initial COD level of the samples indicated that the pollution of the street surfaces near the central square of the city and the border of main and highly trafficked streets as the parking lots is far higher than the areas near to the humps. Evidence demonstrated that the COD removal percentage in the Sample 1 (before the hump) is less than the other two. The results of longitudinal analysis of data reveal that the effect of time and voltage variations on the mean COD index was statistically significant ($p < 0.05$), so that increased input voltage and prolonged reaction time improved the removal efficiency. This study is consistent with the results of studies by Mostafapour et al. and Wang et al [27, 39, 40]. According to Figure 1, the most effective electrocoagulation (80%) process to remove COD in this study occurred at the voltage of 30 volts and the reaction time of 60 minutes, meaning at the highest reaction time and voltage (Table 2), which is in line with the results of a study by Janjani concerning the removal of ion sulfite [39]. The reason for the further reduction of COD using the voltage of 30 volts compared to 10 and 20 volts can be attributed to the greater dissolution of the iron electrode and the further increase in the production of iron hydroxides in the process. Table 2 shows the results of the analysis of runoff samples after electrocoagulation at maximum efficiency.

Table 2. Chemical analysis of runoff samples after applying electrocoagulation method at maximum efficiency (reaction time of 60 min and potential difference of 30 volts and pH between 9.5-10.5)

final Total Suspended Solid (mg/lit) (Mean± Std. Deviation)	final BOD5 (mg/lit) (Mean± Std. Deviation)	final turbidity(23) (Mean± Std. Deviation)	final Ec (µs/cm) (Mean± Std. Deviation)	final pH (Mean± Std. Deviation)	final COD (mg/lit) (Mean± Std. Deviation)	sampling sites
30 ± 3.6	84 ± 5.2	20.6 ± 2.55	3009 ± 18.6	10.6 ± 0.14	174 ± 12.09	before humps(sample1)
20 ± 3.6	102 ± 6.5	19.8 ± 1.32	3146 ± 11.37	9.8 ± 0.11	215.3 ± 3.78	areas near to squares(sample2)
69 ± 4.99	95 ± 6.5	16.7 ± 1.55	3788 ± 32.57	10.4 ± 0.17	196 ± 6.24	border of main streets(sample3)

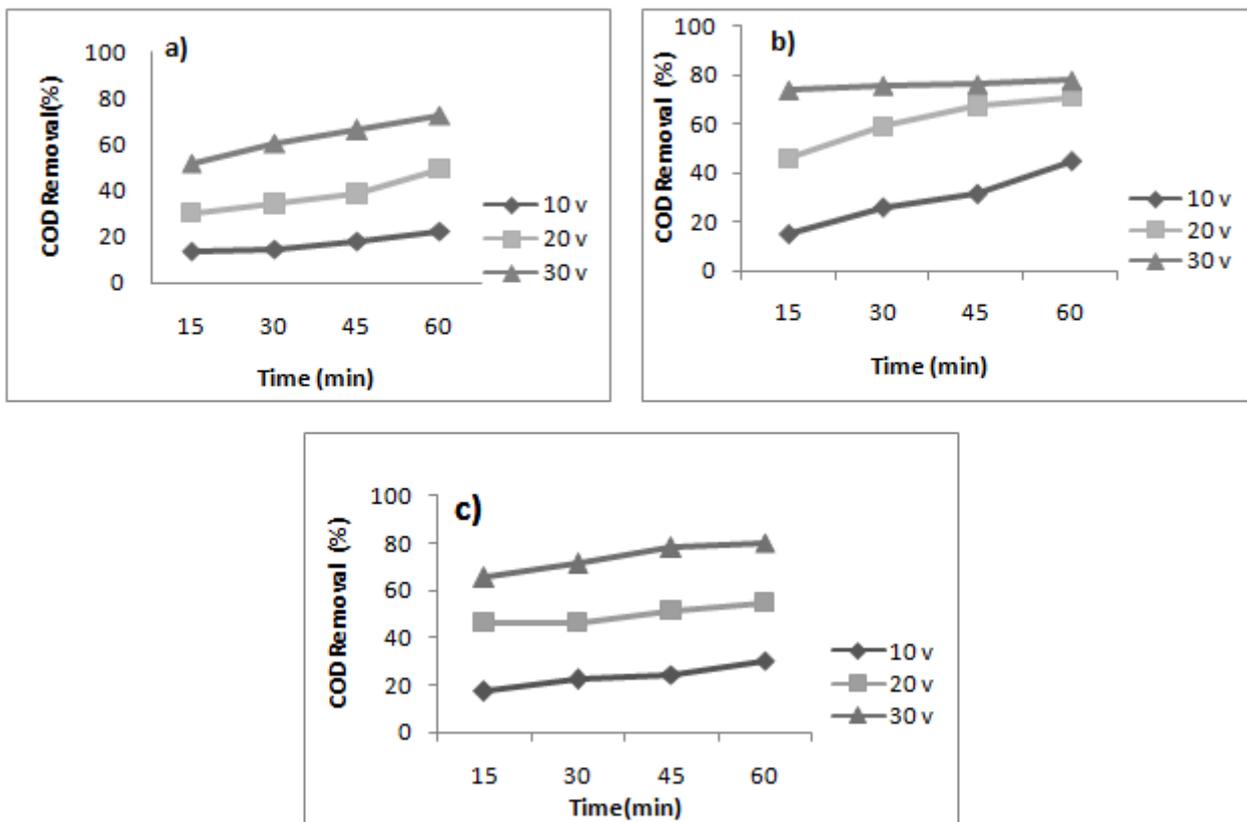


Figure 1. COD removal efficiency in the electrocoagulation process (%); a) before hump, b) areas near to squares, c) street border (10, 20 and 30 volts)

3.2 Assessing the effect of time variations on COD reduction

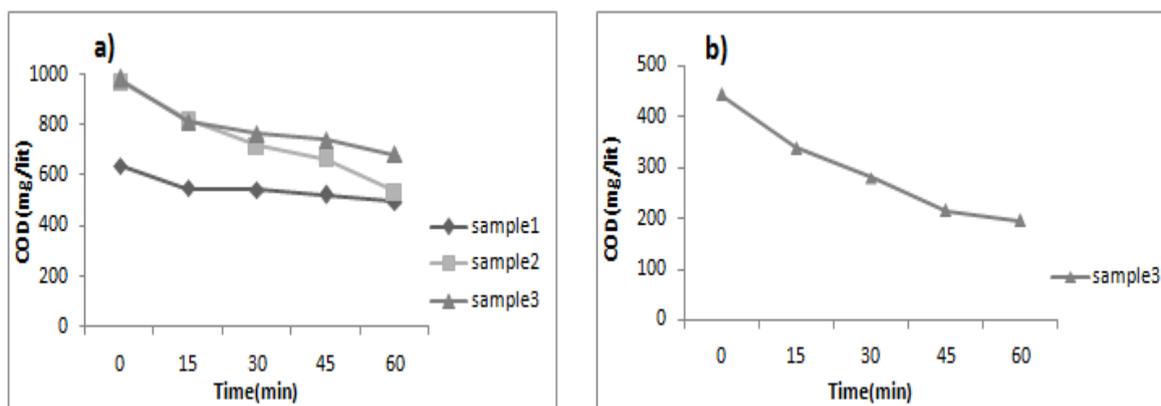


Figure 2. COD reduction process relative to time; a) at the voltage of 10 volts for each sample, and b) at the voltage of 30 volts for Sample 3

Investigating the reaction time variable determined that a large part of COD was removed in the first 15 minutes at 10 volts(Figure 2a). whereas this trend is not very noticeable at voltages of 20

and 30 volts. A similar result was achieved by Rahmani et al. (2016) who investigated the treatment of wastewater from the egg processing industry [33]. The removal efficiency was not significant at the voltage of 10 volts at the time between 15 and 45 minutes, and the COD reduction rate in the Sample 1 (before hump) is slower than the other two. Moreover, at the voltage of 30 volts that had the highest removal efficiency, it was found that the trend of COD reduction was more favorable in the Sample 3 than the other two, and given that its initial COD value was much higher than the initial COD of the Sample 1. However, the final COD level of these two samples is close to each other (Figure B2).

3.2.1 Effect of variable parameters

3.2.1.1 Evaluation of changes in pH and temperature in COD reduction

Regarding the fact that the iron electrodes are in the alkaline range known as coagulants in the coagulation process, this issue was also observed in this study [14, 41]. On the other hand, a higher amount of iron ion in the electrolyte was dissolved by increasing the voltage and the intensity of the current, thereby causing more coagulation in the solution. The results of pH increase at different reaction times and also the effect of pH on the COD removal efficiency at the voltage of 30 volts are shown in Figure 3. Considering the effect of initial pH value of the effluent on the efficiency of the electrochemical process in reducing COD, it is proven that the pH values in the conditions of maximum efficiency (the voltage of 30 volts and the reaction time of 60 min) for all three samples were 10.6, 9.8 and 10.4, respectively, which indicates that this process is less efficient in the acidic and neutral ranges. This corresponded to the results of the study by Afsharina et al. who used iron and copper electrodes to reduce the count of coliform bacteria in the leachate [14]. Rahmani et al. (2013) examined the effectiveness of Electro-Fenton and electrocoagulation in removal of acid black 1 dye from aqueous solutions and found that the electrocoagulation process was more efficient with increasing pH by 7 to 9 [42]. At all stages of the reaction in this study, the temperature of the samples was measured continuously, as the lowest temperature at 10 volts and the reaction time of 15 minutes was reported. It was also found that the temperature also was elevated with prolonging the reaction time; for example, the temperature was changing from 14.5°C to 15.1°C in the Sample 3 with the conditions of maximum removal efficiency (the voltage of 30 volts and 60 minutes), and the maximum removal efficiency occurred at 15.1°C (Figure 3c). According to the results, the COD removal efficiency was also improved with increasing temperature; the results of research by Hosseini et al. also confirm the present research [43]. It should be said that the formed flocculated cells are broken down by increasing the current intensity and, consequently, over-temperature rise, which should be considered as a fundamental point [33].

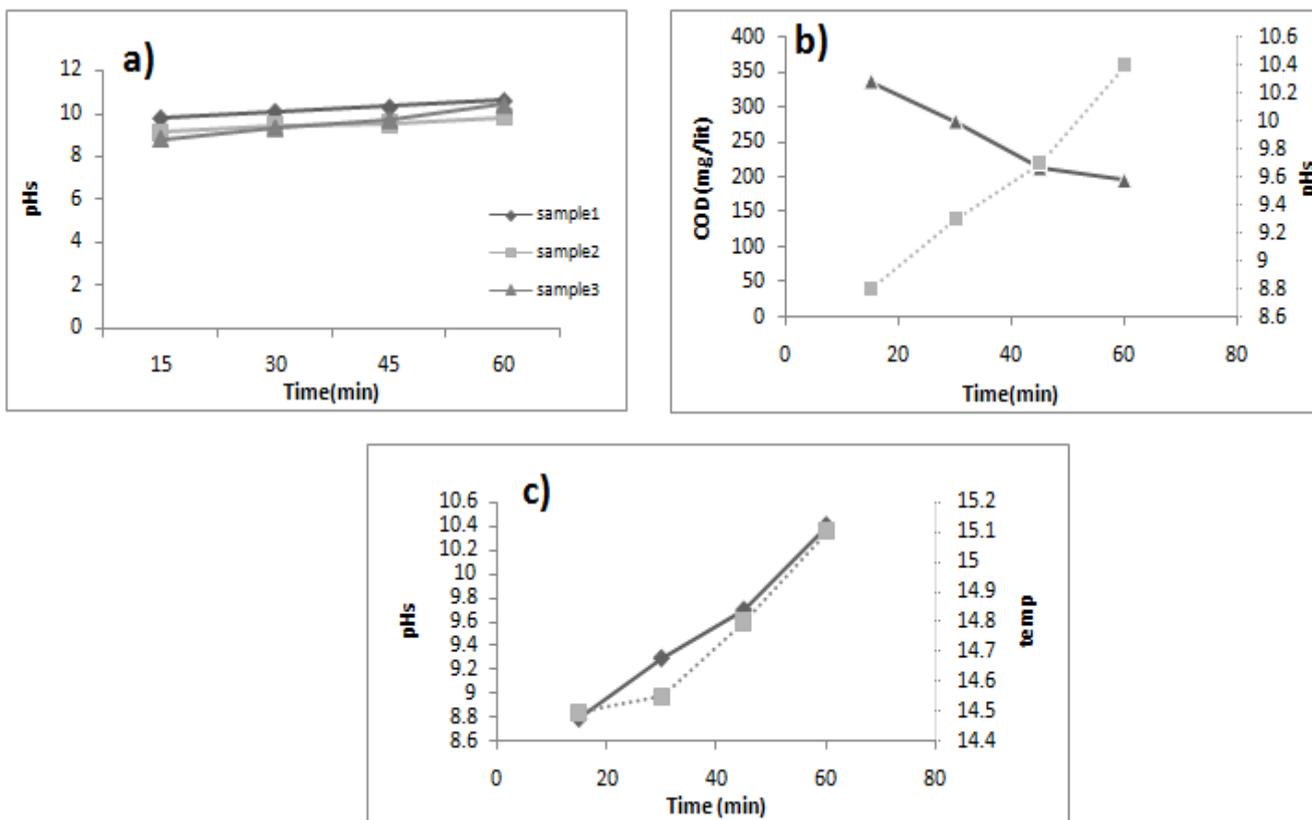


Figure 3. pH and temperature variations at 30 volts; a) pH variation relative to time in each sample, b) COD and pH variations at 30 volts with time for Sample 3, c) Temperature and pH variations with time for Sample 3

3.3 Energy and Electrode consumption:

As the current density increases, degradation of anode plates increases and consequently the removal level of organic pollution (decreased COD) increases. In addition, elevated current density increases the production level of the bubbles and decreases their diameter. The coincidence of both of these factors is beneficial in increasing the separation of contamination by hydrogen gas bubbles [44]. Saeedi and Khalvati showed that the rate of separation and reduction of COD would increase with enhancing ampere consumption and hence current density [45]. In this study, the prolonged time and further COD reduction increased the amount of energy consumption, so that the highest energy consumption was obtained at the voltage of 30 volts and the reaction time of 60 minutes (Figure 4). These results were consistent with the findings of Hosseini et al. regarding the removal of Acid Red 37 dye [43]. At the end of the 60-minute period (as shown in Figure 5), the electrical conductivity increases with enhancing voltage (higher energy consumption), resulting in more metal being released from the electrode plates to the solution (Figure 5), followed by increased COD removal efficiency. The maximum amount of released metal from the Sample 2 was reported to be 0.9 g/cm (Figure 5). According to Figure 5, the metal release rate from the electrode surface is on the rise from the voltage of 10 to 20 volts, but this trend is decreasing from the voltage of 20 to 30 volts, resulting in the highest

consumption of the electrode at the voltage of 20 volts. This suggests a higher cost of iron consumption at this voltage than the other two voltages.

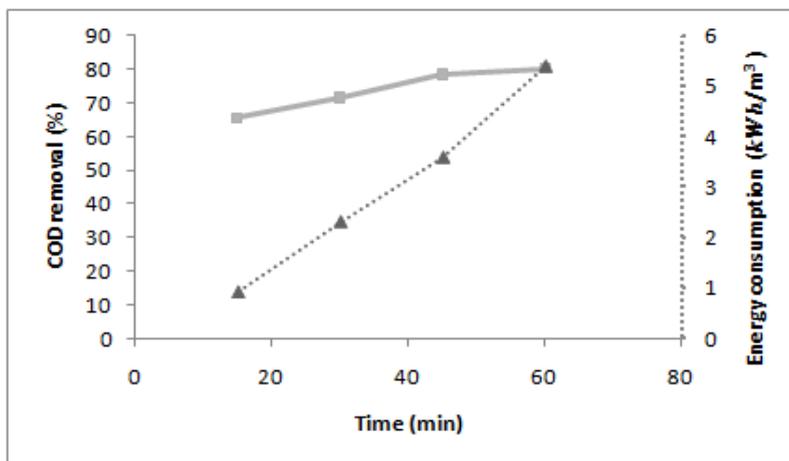


Figure 4. Change in electrical energy consumption and COD removal efficiency over time at the voltage of 30 volts in Sample 3

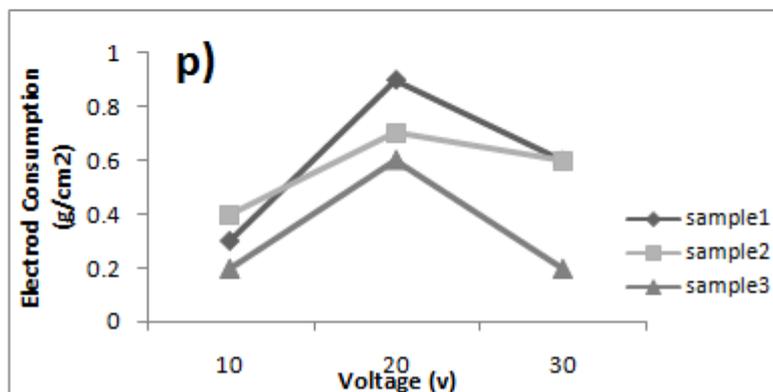
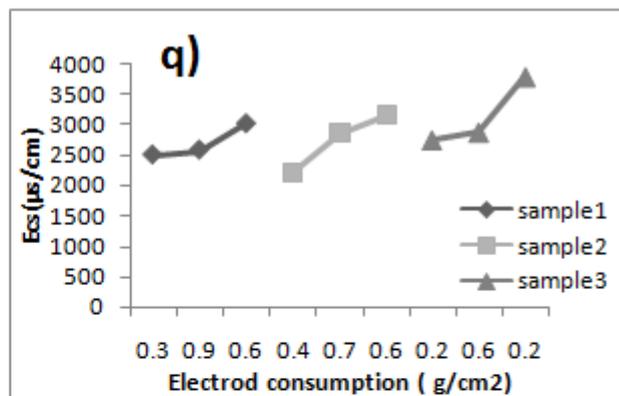
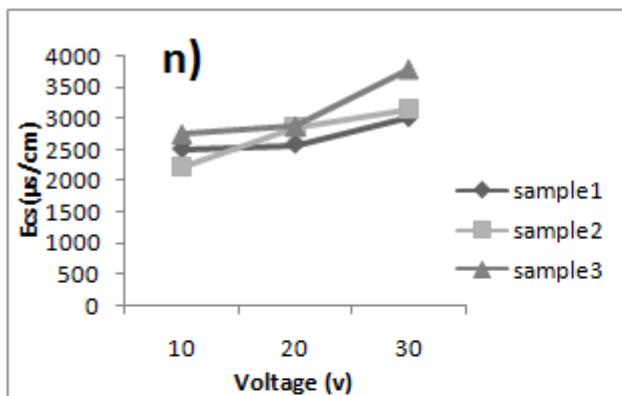


Figure 5. Energy and Electrode changes at the end of 60 minutes; n) electrical conductivity variations at different voltages, q) Change in the amount of electrode consumption in different electrical conductivities, p) Change in the electrode consumption in different voltages (10, 20 and 30 volts)

4. CONCLUSION

The aim of this study was to investigate the COD removal efficiency by electrocoagulation process using the iron electrode. In the present study, the use of electrocoagulation was investigated as a suitable method for reducing the amount of COD of runoff in Torbat-e-Heidarieh city. Regarding the climatic conditions of the region and the poor status of the water reservoirs in the city, the treatment or pre-treatment of urban runoff in this region can be an effective and interesting option for various uses, including agriculture, groundwater utilization, recreational and even in some industrial cases. The findings showed that the COD removal efficiency was approximately 80% by using this method. The optimal operating conditions for the highest removal efficiency were achieved at pH of 9.5-10.5, the potential difference of 30 volts and the reaction time of 60 minutes. The output COD at maximum efficiency was measured at 216, 215 and 196 mg/l in each of the 3 samples, in which the efficiency varied from 72 to 80% for all three samples. The findings indicate that the energy consumption was increased with prolonging contact time and voltage, and consequently, increased removal efficiency. On the other hand, due to the final pH of the wastewater, which was in the alkaline range, the discharge of this type of wastewater into the environment will require special attention. Considering that the wastewater treatment in Torbat-e Heydarieh is carried out using activated sludge method and microorganisms are very important in this treatment method, any shock from organic matter loads and toxic substances such as heavy metals or even hydrocarbons will make trouble in the operation of this treatment plant. The comparison of the results of baseline analysis of surface runoff samples before any pre-treatment (Table1) with the sanitary wastewater indicates that the discharge of these runoffs into the urban wastewater treatment plant, especially in dry and low rainfall season, can impose a shock to the treatment process in terms of organic matter loads (COD) and hence chemical pollution. The results show that, except for the final pH of the effluent from the electrochemical method in this study, other parameters are in a suitable range for discharging and directing this wastewater to the urban wastewater treatment plant.

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AUTHOR CONTRIBUTION

Mehdi Saeidi performed analysis on all samples, interpreted data, wrote manuscript and acted as corresponding author. Mojtaba Afsharnia was consultant and director of experimental.

ETHICAL ISSUE

Ethical issues entirely have been considered by the authors and desperately tried to avoid plagiarism.

CONFLICT OF INTEREST

Authors declare that there is no conflict of interests.

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