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Optimization of Landfill Leachate Treatment Process by Electrocoagulation, Electroflotation and Sedimentation Sequential Method

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This work is dealing with the performance of a sequential Electrocoagulation (EC), Electroflotation (EF), and sedimentation method for the treatment of landfill leachate of Gachsaran city. Effective parameters on the EC process such as electrode type, electrode distance, voltage, process time, and effect of pH have been investigated. Various water treatment criteria such as Chemical Oxygen Demand (COD), Total Suspended Solid (TSS), Oil and Grease (O&G) and turbidity were used to assess process efficiency. Also removal of Total Kjehldahl Nitrogen (TKN) and phosphorus was surveyed as nutrient pollutant criteria. Finally removal of Pb and Cr as typical heavy metals was assessed. Generally, using iron electrode as anode at the optimum condition of pH=8, Voltage = 40 V, Electrode Distance = 2 cm, and contact time of 90 min, the removal efficiency of COD, TSS, O&G, Turbidity, TKN, total phosphorus, Cr, and Pb were obtained 86.9%, 88.7%, 90.2, 93.7%, 81.8%, 90.3%, 70%, and 66% respectively. The results show that this technique is a promising method for treatment of the landfill leachate.

Keywords: landfill leachate treatment; Electrocoagulation; Electroflotation; Sedimentation; nutrient removal; heavy metals removal.

1. INTRODUCTION

Increasing world population and industrial growth are enhanced municipal and industrial solid wastes, and the waste volume is growing faster than the world's population. The aqueous effluent generated from solid wastes are defined as leachates, which is created from consequence of rainwater percolation through wastes and the inherent water of the wastes [1]. Landfill leachates may contain organic and inorganic compounds, ammonia-nitrogen, and heavy metals. Due to its different and toxic composition, the landfill leachate has a detrimental effect to the aquatic life and its treatment is one of the main environmental problems [2]. Also because of high deal and high solubility of landfill leachate components and its inadequate management, it will infect water resources. Up to now many literatures have been published about collection, storage, and landfill leachates [1]. In order to treatment of landfill leachate, biological [3, 4], electro-chemical oxidation [5], adsorption [6], and oxidation by Fenton and persulfate [7, 8] processes have been used by researchers. Landfill leachates are rich in humic acids and their pollutants are toxic which are not easily removed by the biological treatment processes [9]. As mentioned, the landfill leachate has different pollutant with high deal, so its treatment by adsorption process will be time-consuming and costly with low efficiency. Also advanced oxidation process for treatment of pollutants with high value of organic matters will be time-consuming and costly. On the other hand the landfill leachate is rich in salts and in AOPs more cations and anions are radical scavenger [10], so in this case AOPs will be low efficiency. One of the effective methods to treat pollutants with different analysis is coagulation process. It has been widely used to treat pollutants with various analysis such as municipal wastewater, anions and heavy metals, dyes, and landfill leachate [11-15]. In the chemical coagulation process, the chemicals such as $Al_2(SO_4)_3$ and $FeCl_3$ as a coagulant are directly poured to the media. In this method a high value of sludge is produced. Electro-Coagulation (EC) process is a consistent and coast effective method with low amount of sludge and without any sensitivity to toxic materials [16, 17]. In the EC process coagulant is produced by electrolytic oxidation of an appropriate sacrificial anode by suing a direct current [15]. The EC mechanism for iron and aluminum anode could be represented as follow [15, 18]:

Iron:

$$Fe_{(S)} \rightarrow Fe^{2+}_{(aq)} + 2e^- \quad (1)$$

$$2H_2O + 2e^- \rightarrow H_{2(g)} + 2OH^-_{(aq)} \quad (2)$$

$$Fe^{2+}_{(aq)} + 2OH^-_{(aq)} \rightarrow Fe(OH)_{2(s)} \quad (3)$$

Aluminum:

$$Al_{(S)} \rightarrow Al^{3+}_{(aq)} + 3e^- \quad (4)$$

$$3H_2O + 3e^- \rightarrow 3/2H_{2(g)} + 3OH^-_{(aq)} \quad (5)$$

$$Al^{3+}_{(aq)} + 3OH^-_{(aq)} \rightarrow Al(OH)_{3(s)} \quad (6)$$

One of the effective ways to separate coagulated pollutant is its flotation by gas bubbles. The separation yield is increased with decreasing bubble size because the collision between particles and bubbles will be enhanced by using fine bubbles. If the bubbles produce electrically, the size and production rate of bubbles will be controllable [19]. Flotation of pollutant by bubbles which was produced electrically is called Electro-Flotation (EF). Operational parameters such as the electrode genus, position and structure of electrode, voltage density, and pH are effective parameters in the size of bubbles in EF process [20]. In EF process corrosion-resistant electrode should be used. In EF process hydrogen and oxygen gas are produced at cathode (Eq. 2) and anode (Eq. 7) respectively, which are floated the pollutant [21].

$$2H_2O_{(l)} \rightarrow O_{2(g)} \uparrow + 4H^+_{(aq)} + 4e^- \quad (7)$$

In the present study treatment of landfill leachate of Gachsaran city in Iran was performed by EC process and separation of coagulants was consecutively done by EF and precipitation methods. Effective parameters on the EC process such as electrode type, electrode distance, voltage, process time, and effect of pH have been investigated. Because landfill leachate has different type of pollutants, various water treatment criteria such as Chemical Oxygen Demand (COD), Total Suspended Solid (TSS), Oil and Grease (O&G) and turbidity were used to investigate process efficiency. To survey nutrients removal by the process, removal of Total Kjehldahl Nitrogen (TKN) and phosphorus were investigated. Finally removal of Pb and Cr as typical pollutant of heavy metals were surveyed in the optimum condition.

2. EXPERIMENTAL

2.1. Reagents and instruments

The landfill leachate of Gachsaran city of Kohgiluyeh and Boyer-Ahmad province in Iran was selected as a typical landfill leachate. Chemicals (e.g. potassium dichromate, sulfuric acid, ferrous ammonium sulfate, sodium hydroxide, etc.) were supplied from Merck and Fluka Company to measure pollutants quantity. Distillated water was used for preparing the chemical solutions. Adjusting of the solution pH was performed by sulfuric acid and sodium hydroxide. NaCl and Na_2SO_4 were used as electrolyte. All the chemicals were products of Merck Company with analytical purity. pH measurements were done by a portable pH meter (HACH, USA). Digital DC power supply (Shenzen-Masteck, V = 0-50 volt, I = 0-5 A, China) was used as an electrical source. Measurement of Pb and Cr concentrations were performed by atomic absorption spectrometer (Varian techtron MODEL 1200, Australia) at wavelength of 217 and 357 nm for Pb and Cr respectively. To analyze turbidity of the samples a spectrophotometer (HACH DR500, USA) was used.

2.2. The electrochemical reactor

The landfill leachate treatment process was investigated in an electrochemical reactor which includes a preliminary electrocoagulation step, then electro-flotation step and finally sedimentation step. In the first step EC unit with 1 liter volume (10cm*10cm*10cm) equipped by 8 electrodes with dipole arrange was used. Iron and aluminum plates were separately used as electrode with dimension of 9 cm × 7 cm × 0.08 cm. It is notable that 8 cm of the electrodes drown into the wastewater. In the second step EF unit with 1 liter volume was used which equipped with three titanium plates covered by ruthenium oxide as anode and three stales steel plates as cathode. The dimension of the electrodes is 1 cm*0.2 cm* 16 cm which were placed vertically in the floor of the tank. The third step was designed as a settling chamber to settle solids. The figure 1 shows schematic view of the electrochemical reactor.

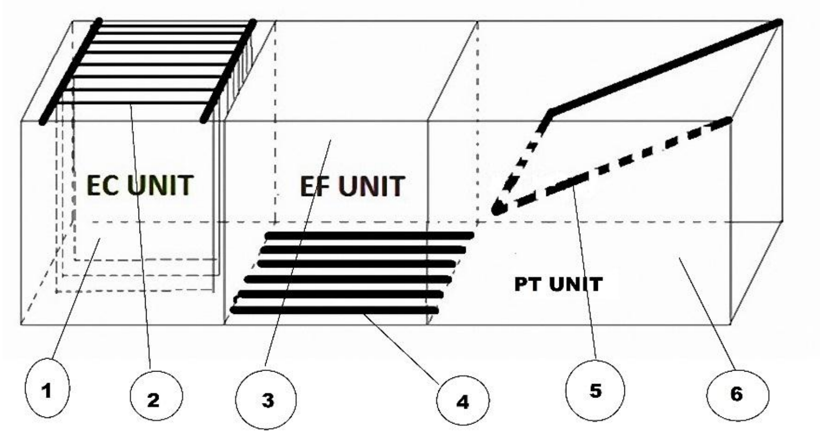

Figure 1. the schematic view of the electrochemical reactor. 1: EC unit (Electrocoagulation Unit), 2: Electrocoagulation Electrode, 3: EF Unit (Electroflotation Unit), 4: Electroflotation Electrodes, 5: precipitation adjustable sheet and 6: PT Unit (Precipitation Tank)

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2.3. Procedure

Raw landfill leachate samples were collected from the gachsaran city. The sampling was performed at different time and after assessment of their pH and temperature, they are transferred to laboratory for subsequent experiments. The samples were stored in the refrigerator at 4 °C and before doing the experiments their temperature were regulated at room temperature. After adjustment of pH the samples were transferred into the EC vessel and then the process was started. The sampling was done after specific time and the effect of operational parameters on the EC process were studied. After completion of EC process the produced clods and foam were separated and the treated landfill leachate in the EC step was transferred into the EF vessel. In the EF section flotation of the remained clods were performed and after their separation the treated landfill leachate was transferred into the settling chamber. In the third section the water was settled to sediment the solid pollutants. Finally sampling was performed from treated landfill leachate and the different analyses were performed to assess efficiency of the process. It is notable that the experimental analyses were repeated three times and they were performed according to standard methods as follow:

COD of the samples was determined by the open reflux method (method number of 5220). TSS measurement was carried out by filtering, drying and weighting the samples (method number of 2540). TDS analysis was performed by total filterable solids (method number of 5907). TKN was carried out by the Kjehldahl methods (method number of 4500). To measure O&G hexane extractable gravimetric method was used (method number of 5520). Ascorbic acid method was used to measure total phosphorous (method number of 4500) [22]. To avoid interferences the samples were filtered through a whatman filter.

3. RESULTS AND DISCUSSION

3.1. Characteristics of the landfill leachate

Several parameters such as the people living standards of the study area, landfill age, and climatic conditions influence on the composition and characteristic of the landfill leachate [1]. In this work the landfill leachate of gachsaran city of Kohgiluyeh and Boyer-Ahmad province in Iran was studied. To this aim at different time the sampling was performed and various water treatment criteria such as pH, COD, TSS, TDS, O&G, TKN, total phosphorus, turbidity, and Pb and Cr concentration were used to assess characteristics of the landfill leachate which are presented in the t able 1. It can be seen that the pH of the leachate is in the range 7.1 to 8.2 that in this pH biological activity could be done. The average amount of COD of the leachate is 6317 mg per liter that it is in the high level. The conventional biological methods are ineffective to treatment of the leachate because it has high ammonia nitrogen and low phosphorus content [11]. According to literatures to treat a wastewater with mentioned characteristics, coagulation, flotation, and sedimentation sequential method is an effective method [1, 11, 21, 23].

Table 1. Characteristics of the landfill leachate of gachsaran city.

Criteria	Sampling Date						Average Value
	March, 1, 2015	March, 16, 2015	April, 10, 2015	May, 8, 2015	June, 5, 2015	June, 19, 2015	
pH	8.2	7.1	7.6	8.3	8.4	8.3	8
COD (ppm)	5670	6715	7105	6475	4020	7920	6317
TDS (ppm)	1960	2481	2413	1853	2540	2286	2255
TSS (ppm)	298	327	284	211	315	305	290
Turbidity (NTU)	338	390	311	243	372	345	333
TKN (ppm)	990	765	976	830	653	1030	874
Total phosphorus (ppm)	48	65	49	37	35	52	48
Oil&Grease (ppm)	2.6	1.7	1.8	1.5	3.2	4.4	2.5
Pb (ppm)	0.22	0.15	0.2	0.18	0.15	0.12	0.17
Cr (ppm)	0.45	0.45	0.65	0.5	0.5	0.55	0.52

3.2. Electrode type on the EC process

As mentioned aluminum and iron ions are the most coagulant agent and they are widely used by researchers [15, 24]. In this work iron and aluminum were selected as the anodes which were separately applied in the EC step. In order to survey the effect of electrode type on the EC process COD, TSS, O&G and turbidity were investigated that the results illustrated in Figure 2. As shown in the figure in the all examined criteria the iron electrode has more efficiency rather than aluminum. According to the literatures the iso-electric point of iron oxide/hydroxide is 7.7 and aluminum oxide/hydroxide is 8.8 [13]. Since the experiments were performed at pH 8 (natural pH of the landfill leachate), in this condition the iron electrode will have more efficiency because in the iso-electric point metal hydroxide is better coagulant. So iron was selected as EC electrode for doing subsequent experiments.

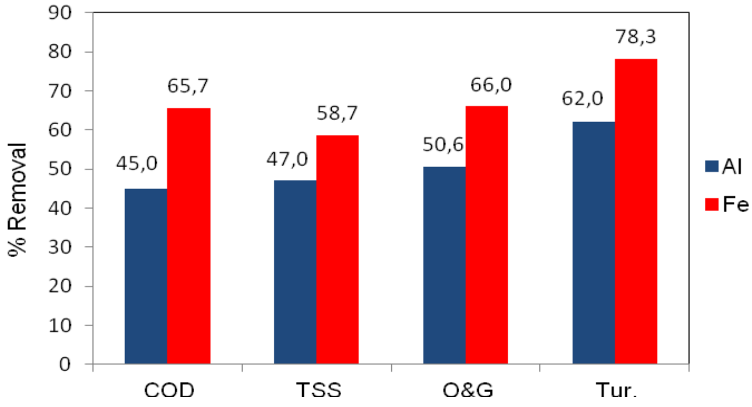


Figure 2. Effect of anode type on the EC process (pH: 8, time: 60 min and Voltage: 30 V)

3.3. Electrode distance

In order to study electrod distance effect on the EC process, a sreies of experiments were performed and the results were demonstrated in Figure 3. As seen the figure the removal efficiency of COD, O&G, TSS and turbidity were increased with increasing electrod distance from 0.5 to 2 cm, and then it is decreased with increasing electrod distance from 2 to 3. The results can be explined by this subject that, when the electrod distance is low, passivation of the electrode could be done faster specially in this case that the pollutant is rich in contaminant especially COD. Based on the results, in low electrode distances the efficiency is low. On the other hand, when the electrod distance is extremely increased the solution resistance will be increased. If the solution resistance increase the dissolution rate of metal will decrease, for this reason after 2 cm of electrode distance the efficiency has been decreased. So electrode distance of 2 cm was selected as optimum electrode distance for doing subsequent experiments.

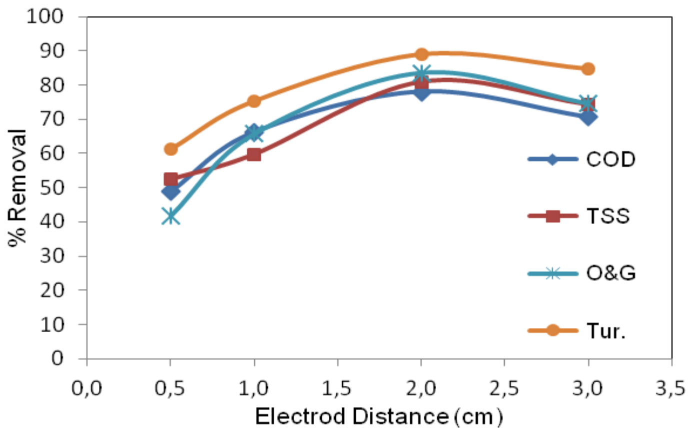


Figure 2. Effect of electrode distance on the EC process (Electrode: Iron, pH: 8, time: 60 min and Voltage: 30 V)

3.4. Effect of pH

The EC process is heavily dependent on the solution pH, because it affects on stability of coagulant in the solution. Figure 3 shows the effect of pH on the EC process at the optimum condition obtained from previous sections. The figure shows that COD removal efficiency is in the maximum value at pH = 5. However the COD removal variation versus pH is low and COD removal efficiency is in acceptable amount at acidic and alkaline pHs. On the other hand the removal efficiency of TSS, O&G, and turbidity versus pH is intensively varied such that maximum efficiency is observed at the pH = 7-8. As mentioned in section (3.1) the iso-electric point of iron oxide/hydroxide is 7.7 [13]. So the coagulant will have positive and negative charge at the below and abow pH of 7.7 respectively. The coagulant with surface charge will be soluble in the solution and dose not precipitate [15], and therefore can not perform coagulation process. As regards at the pH 8 O&G removal efficiency is in the acceptable level (83%) which is only 10% lower than pH 5 efficiency, and considering that the natural pH of the lechate is 8 (section 3.1) the pH = 8 was selected as optimum pH of the EC process.

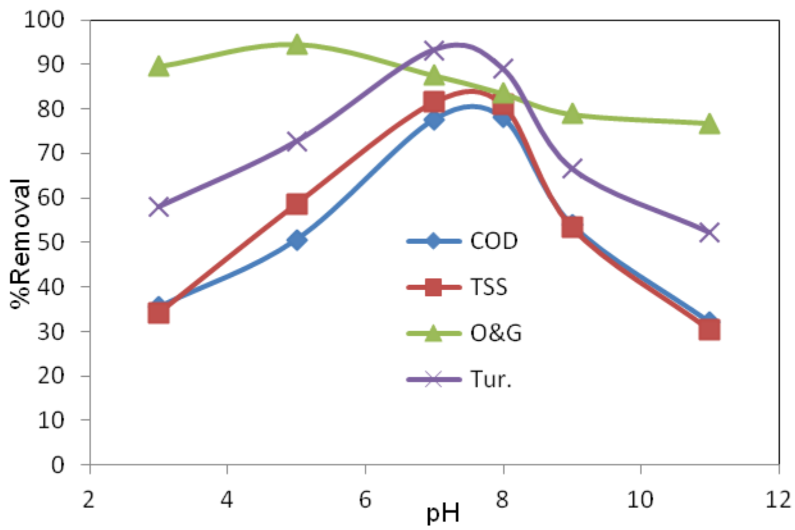


Figure 3. Effect of pH on the EC process (Electrode: Iron, distance: 2cm, time: 60 min, and Voltage: 40 V)

3.5. Effect of voltage on the EC process

Voltage is an important parameter to control the electrocoagulation reaction rate. In the EC process, with inducing current into the sacrificial electrode the metal ions release into the solution which are precursor of coagulant. To investigate the effect of voltage on the process, the experiments were carried out at obtained optimum condition from previous sections, with the voltage in the range of 10-40 V. Figure 4 demonstrate the effect of voltage on the efficiency of COD, TSS, O&G, and turbidity in the EC process. As seen the figure, when the voltage increased from 10 to 40 A in the EC process, the removal efficiency of COD, TSS, O&G, and turbidity increased. Energy consumption as an important criterion in industry can be calculated as follow:

$$E = VIt \tag{8}$$

Where E is the electrical energy consumption (Wh), V is the potential (volt), I is the current (A) and t is the time of the EC process (h).

High current raises the formation of higher concentration of coagulant because metal ion production at the anode depends on the quantity of electricity which passes through the solution [2, 25]. In accordance equation 8 energy consumption is increased with increasing voltage. Since at the voltage of 40 V, the efficiency of the process is at high level to prevent excessive consumption of electrical energy the voltage of 40 V was selected as optimum voltage and subsequent experiments were performed using this voltage.

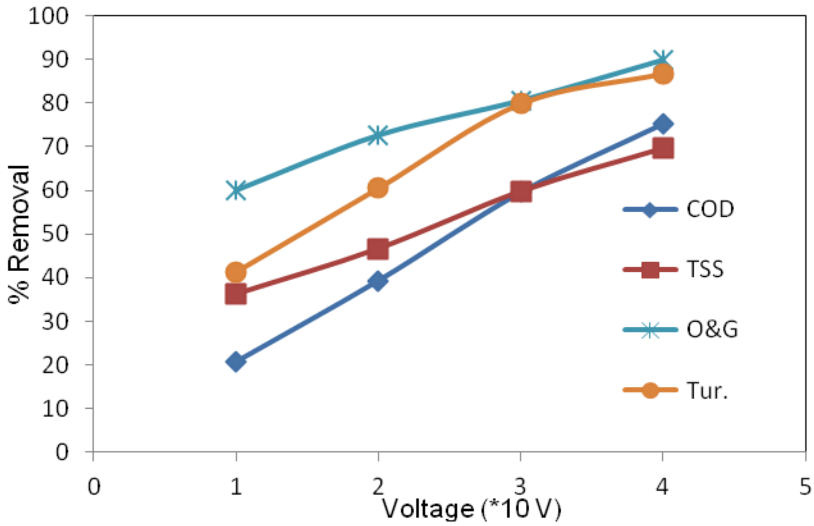


Figure 4. Effect of voltage on the removal efficiency the EC process (Electrode: Iron, distance: 2cm, pH: 8, and time: 60 min)

3.6. Process time

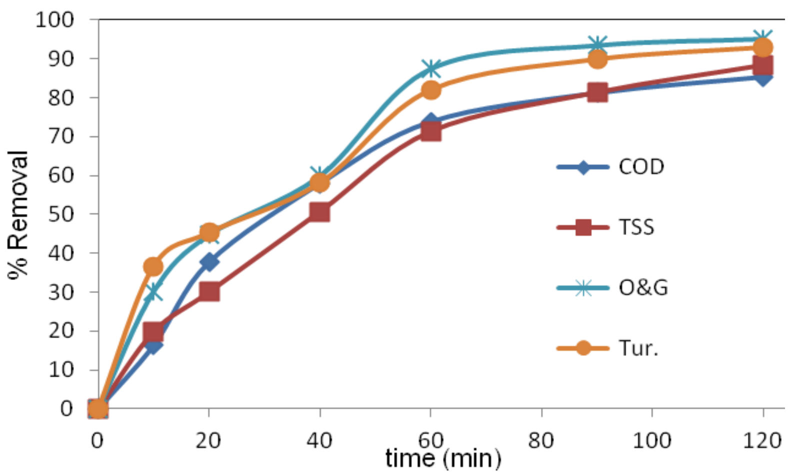


Figure 5. Effect of the EC process time on the efficiency (Electrode: Iron, distance: 2cm, pH: 8 and V: 40)

In order to prevent the use of extra electrical energy, the enough contact time for the completion of the EC process was studied at optimum conditions obtained from previous sections, that the result was demonstrated in Figure 5. As shown the figure the removal efficiency of COD, O&G, TSS and turbidity were increased by time passing with a good slope up to 90 min. The removal efficiency of COD, O&G, TSS and turbidity were reached to 81%, 93%, 81%, and 93% respectively after 90 min of the EC process time which is an appropriate efiiciency. So the contact time of 90 min was elected as an enough time for the EC process. In comparison the other methods such as biological and oxidation process this method has a good results in a shorter time [4, 26-28].

3.7. Optimum condition

As mentioned in the previous sections, optimization of the EC process was assessed and an experiment was performed in the obtained optimum condition, and process efficiency was investigated. The results demonestrated in Figure 6. As can be seen from the figure, after 90 min of the process using iron as anode at pH = 8, Electrod Distance = 2 cm, Voltage = 40 V, temperature of 25 °C, removal efficiency of COD, TSS, O&G, and Turbidity were obtained 86.9%, 88.7%, 90.2, and 93.7% respectively. The obtained removal efficiency for the mentioned criteria are at high level.

In comparison the other methods in landfill leachate tratment this work has a good operation. For example Ilhan et al. investigate treatment of leachate by electrocoagulation using aluminum and iron electrodes and they reported 56% and 35% COD removal for aluminum and iron respectively after 30 min operating time [18]. Also Chiemchaisri et al. used biological method to treat landfill leachate and they reported that the organic removal efficiencies, i.e. COD, BOD and DOC, were relatively constant with an overall average of roughly 97 percent after 150 day-period [4]. Advanced oxidation process were also used to trate landfill leachate such that, Electrochemical oxidation, and O_3/PDS process resulted COD removal of 56.9% (160 min), 73% (180 min), and 72% (210 min) respectively [26-28]. So the results shows that, the EC process is a effective method for traeting landfill leachate.

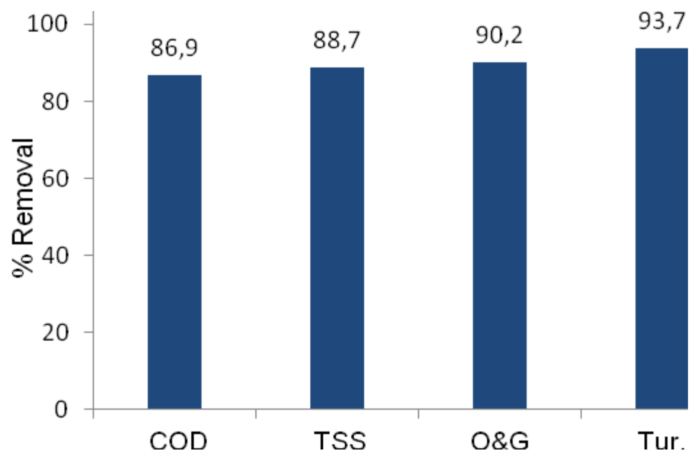
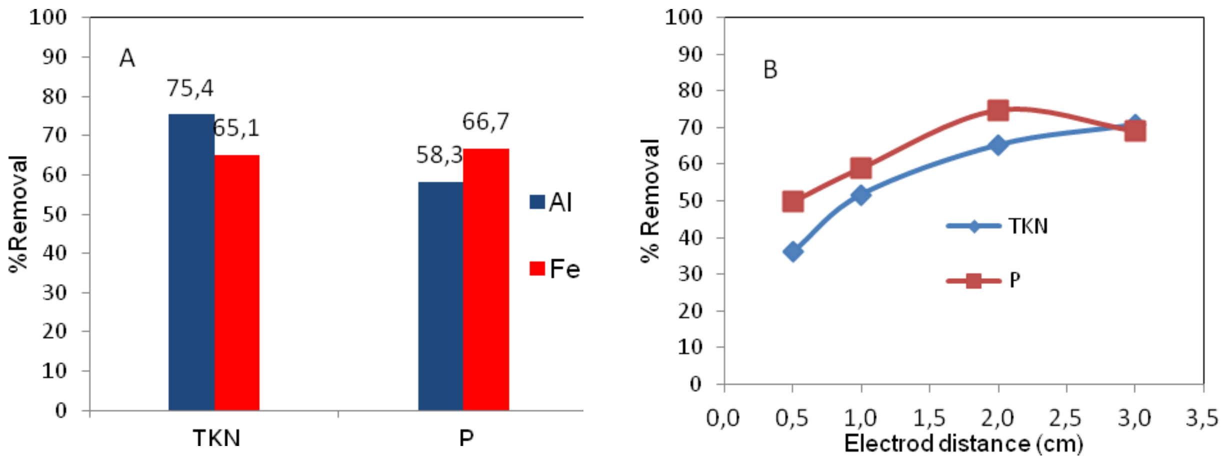


Figure 6. Removal efficiency of EC process (Electrode: Iron, distance: 2cm, time = 90 min, pH = 8 and V = 40 V)

3.8. Nutrients Removal

One of the effective methods to treat wastewater contain high level of organic carbon, nitrogen (N), and phosphorus (P) compounds is biological method [29, 30]. But in many cases there is an improper ratio between carbon, nitrogen, and phosphorus which causes ineffectiveness of the conventional biological methods. In these cases to balance nutrients ratio some of them are added manually to the media which will be costly. In the landfill leachate studied in this work, because of high ammonia nitrogen and low phosphorus content the conventional biological methods are ineffective [11]. In this work to remove TKN and phosphorus content subsequence methods of EC, EF, and sedimentation were used. Similar to the previous sections to assess TKN and phosphorus removal the effective parameters such as electrode type, electrode distance, voltage, process time, and effect of pH have been investigated. The results were demonstrated in Figure 7. In electrode type (Figure 7. A), it can be seen that when aluminum used as anode TKN removal was performed better than iron and when iron used as anode phosphorus removal was performed better than aluminum. As regards iron is cheaper than aluminum and as mentioned in the section (3.2) using iron as anode got better removal efficiency about other criteria (COD, TSS, Tur., and O&G), the iron was selected as anode. The effect of electrode distance, voltage, and time on the TKN and phosphorus removal was demonstrated in the Figure 7 B, C, and D respectively. In these cases optimum amounts are 2 cm, 40 V, and 90 min for electrode distance, voltage, and time of process respectively. The reasons of selection of these optimums are mentioned at pervious sections. Figure. 7 E shows the effect of pH on the removal of TKN and phosphorus content. As seen the figure removal of TKN was increased with increasing pH. As mentioned in table 1, more content of nitrogen of the leachate is in ammonia form. Ammonia contains a lone pair electron upon the neutral molecule which causes ammonia adsorbed on the Fe $(OH)_3$ coagulants, but at acidic pH the lone pair electron gets a proton and the ammonia is in the form of ammonium that its adsorption will be weaker than ammonia. In the case of phosphorus, unlike ammonia, with increasing pH its removal is decreased. Depending on the pH, phosphorus is in the form of phosphate ion (H_3PO_4, $H_2PO_4^-$, HPO_4^{2-}, PO_4^{3-}) at the wastewaters.



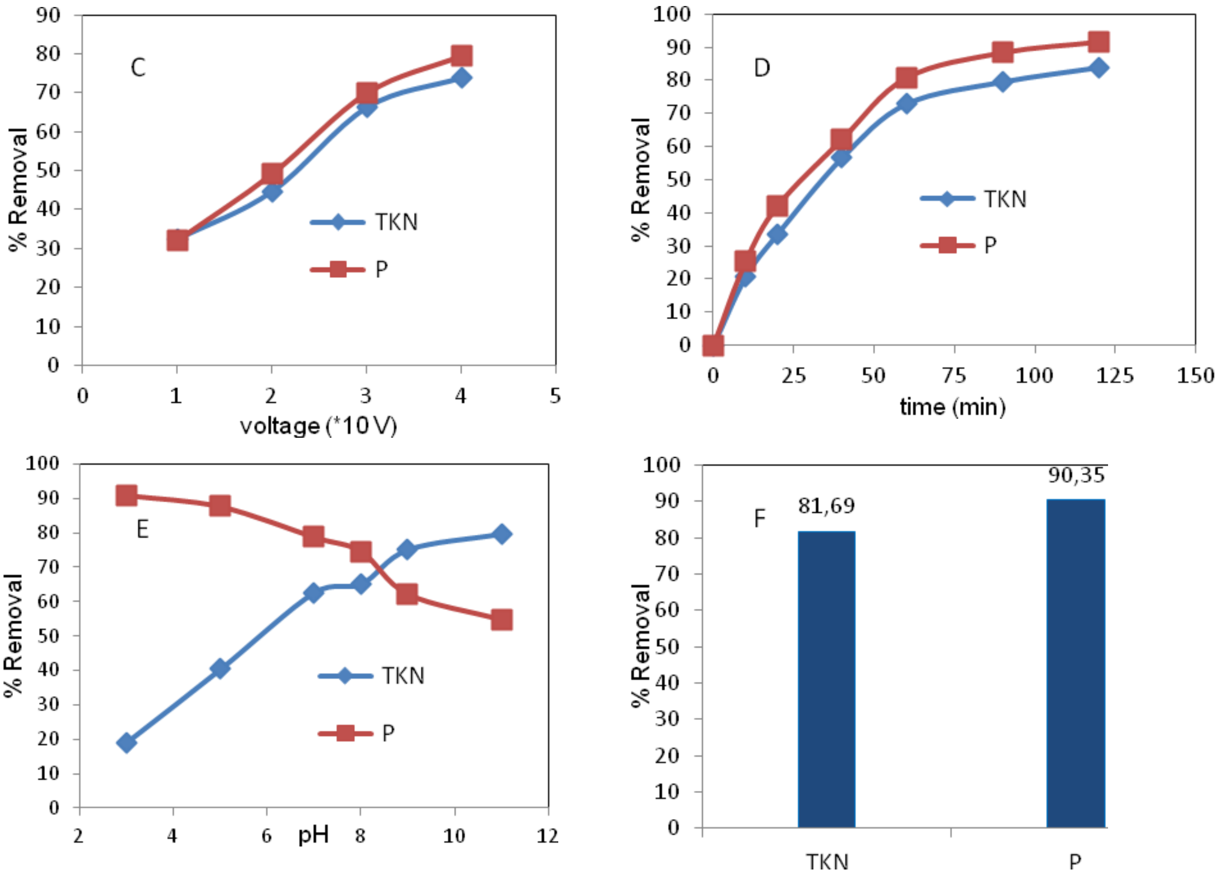


Figure 7. Removal of TKN and phosphorus, A: Effect of electrode type (pH:8.2, Time: 60min, Voltage: 30V, Electrode distance: 1cm), B: Effect of electrode distance (Electrode: Iron, Voltage: 30V, Time: 60min, pH: 7.1), C: Effect of voltage (Electrode: Iron, pH: 8.3, Time: 60min, Electrode distance: 2cm), D: Effect of EC process time (Electrode: Iron, Distance: 2cm, pH: 8.4, Voltage: 40V), E: Effect of pH (Electrode: Iron, Distance: 2cm, Voltage: 30V, Time: 60min), F: efficiency in Optimum condition.

With increasing pH the charge of phosphate ion is increased and its solubility will be raised and its adsorption on the coagulants will be diminished. Generally the removal of TKN and phosphorus in the pH of 8 are in appropriate level, so the pH = 8 was selected as optimum pH. Figure. 7 F shows the removal of TKN and phosphorus in the optimum condition such that removal of TKN and phosphorus are 81.8% and 90.3% respectively.

3.9. Removal of heavy metals

Heavy metals are well known for their toxicity, and have infinite lifetime and can be accumulated in food chains. Toxicity and persistence of heavy metals in environment has a detrimental effect on the aquatic organisms and humans [31]. Because of non-degradability of metal ions electrocoagulation method as an effective method was used to remove heavy metals from aqueous media [32]. In this work to remove heavy metals of landfill leachate by EC, Cr and Pb were selected as a typical heavy metals and their removal were surveyed. To assess removal efficiency of Cr and Pb, experiments were performed under optimum conditions obtained from previous sections. The results are demonstrated in Figure 8. As can be seen from the figure Cr and Pb were removed 70% and 66% respectively. The results show that the EC process is a promising method to remove heavy metals from landfill leachates.

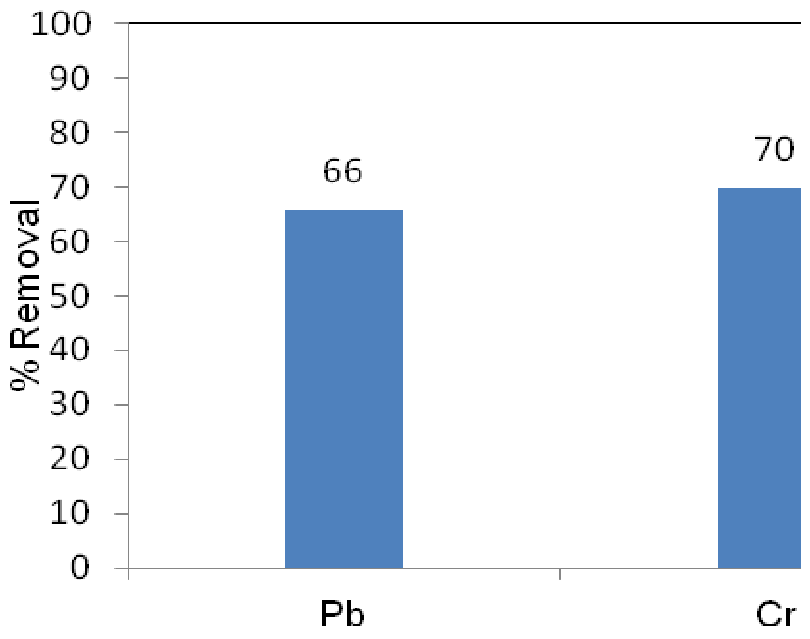


Figure 8. Removal efficiency of the EC process (Electrode: Iron, distance: 2cm, time = 90 min, pH = 8 and Voltage = 40 V)

4. CONCLUSION

In the present study, sequential EC/EF/sedimentation method was applied to treat landfill leachate of gachsaran city. The results of this study approve that the method is a promising technique for treatment of the landfill leachate. Concisely, the follow information was obtained from this study:

- For treatment of the landfill leachate, iron is better than aluminum as anode in the EC process.
- In the optimum conditions of pH 8, Voltage = 40 V, Electrode Distance = 2 cm, and contact time of 90 min, the removal efficiency of COD, TSS, O&G, and Turbidity were obtained 86.9%, 88.7%, 90.2%, and 93.7% respectively.
- In removal of nutrient removal, the iron electrode was proper than aluminum and in the optimum condition the removal of TKN and phosphorus was obtained 81.8% and 90.3% respectively.
- In removal of Cr and Pb as typical heavy metals, 70% and 66% removal efficiency were obtained for Cr and Pb respectively.

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