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Removal of organic materials and hexavalent chromium from landfill leachate using a combination of electrochemical and photocatalytic processes

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ABSTRACT

Leachate is one of the major problems of municipal waste landfills. Landfills produce a dark black colored liquid with high levels of chemical oxygen demand (COD) and heavy metals and low biochemical oxygen demand (BOD)/COD ratio. The objectives of this study were to evaluate the effectiveness of an electrochemical process using graphite and platinum electrodes and photocatalytic properties of titanium dioxide nanoparticles stabilized on bentonite at different reaction times for removal of chromium and organic materials from leachate. Equipment used in the electrochemical reactor included an anode electrode plate, a commercial platinum electrode and a graphite cathode. We also evaluated the impact of electrical current density (1–4 A/m^2), reaction time (1–8 h), concentration of catalyst (1–4 g/L) and UV radiation intensity (3–8 UV lamps). The results showed that the removal efficiency increased with increasing reaction time, current density, intensity of UV radiation and dose of catalyst. Moreover, biodegradability (BOD/COD ratio) was improved. Based on the results, this electrochemical pretreatment process can remove organics materials, heavy metals, reduce organic load and increase wastewater biodegradability. Thus, it can be used as an efficient option for treating sewage and preventing environment pollution.

Keywords: Leachate; Organic matter; Chromium; Electrochemical; Photocatalytic processes

1. Introduction

Sanitary landfill is the main method used for disposal and management of industrial and municipal solid waste (MSW) [1]. Up to 95% of total MSW collected worldwide is disposed of in landfills [2]. Decomposition of organic waste and rainfall generate leachate at the bottom of landfills [1], which is a major problem for municipal waste landfills [3]. The highly contaminated landfill leachate may cause groundwater contamination in addition to several other environmental effects such as surface water pollution, change in aquifer, hydraulic conductivity values and mineral resources pollution. These adverse outcomes may affect human health and aquatic environment [2,4].

Leachates in classical and conventional wastewater treatment plants are rarely treated due to its nature and the presence of high levels of pollutants such as organic matter, heavy metals, pathogens and hazardous materials [4]. Therefore, advanced and special facilities are required before leachate is discharged to the environment or to the wastewater collection system [4].

High chemical oxygen demand (COD) and the presence of toxic matter (include heavy metals, organic materials, persistent pollutants, etc.) in the landfill leachates, are the most

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important problems in leachate management. The high COD levels could be due to high concentration of biodegradable and non-biodegradable materials [5]. Heavy metals are also important pollutants in leachates. Among heavy metals, chromium(VI) is a toxic and carcinogenic contaminant of leachates that can be released into the environment through electroplating, metal finishing, tannery and fertilizer industries [6].

Leachate's quality and characteristics affect operating and capital costs, regulations and discharge limits. Various advanced treatment techniques can be used for the treatment of leachate such as advanced oxidation processes, membrane methods, coagulation, flocculation, electrochemical processes, biological processes, combination of advanced oxidation and biological processes, leachate recirculation through the landfill, etc. [4,7,8]. Karimi et al. [9] used three advanced oxidation processes based on wet air oxidation for leachate treatment and found that wet air oxidation with granular activated carbon and wet peroxide oxidation is much more efficient for removing resistant organic matter. Zazouli et al. [10] used Fenton, photo-Fenton and Fenton-like processes for leachate treatment and reported that the efficiency of the Fenton-like process was less than that of Fenton and photo-Fenton processes. Meanwhile the Fenton-like process was carried out at higher pH and did not show any problems.

In recent years, various biological, physical and chemical processes have been used for the treatment of landfill leachate. Electrochemical process is one of the methods used for water purification and wastewater treatment, which has recently received much attention for the treatment of environmental pollutants, specially leachate and wastewater. These methods have been successfully applied for purification of refractory organic pollutants, toxic compounds, phenolic substances and other pollutants [7].

Electrocoagulation is a simple and efficient electrochemical method for water and wastewater treatment [11]. It requires less coagulation, simple equipment, easy operation, less amount of sludge, less number of wastewater treatment plants and less land area for treatment plant [11].

Many researchers have investigated the electrochemical oxidation of leachate and different wastewaters containing various types of pollutants including paint, textile and tannery wastewater [12–14]. Electrochemical processes have also been used for the treatment of landfill leachate in various studies. These studies have used a flow electrochemical reactor using titanium dioxide (TiO_2) and RuO_2 anode and Ti cathode [1]. Various electrodes such as iron, aluminum, titanium dioxide and graphite have been used in the electrochemical processes [1,5,15]. This study focuses on the photocatalytic (UV/TiO_2) and electrochemical treatment of leachate generated from Kahrizak landfill site in Tehran, Iran. The objectives of this study were as follows:

- To develop a relationship between current density, reaction time and influent COD and biochemical oxygen demand (BOD) in the electrochemical reactor with palatine (Pt, anode) and graphite (cathode) electrodes. Pt was chosen as the working electrode in this study because of its relatively high over potential [16]. Cathode materials should have high overvoltages for hydrogen evolution, while anode materials should have overvoltages for oxygen evolution [17].
- To develop a relationship between reaction time, catalyst dosage, number of UV lamps and influent COD, BOD and chromium(VI) in the photocatalytic reactor.
- To evaluate application of both reactors and removal efficiency of COD and chromium.
- To determine optimum operational conditions (influent COD, current density, reaction time, catalyst dosage and number of UV lamps) for leachate treatment.

2. Materials and methods

Experiments on electrochemical and photocatalytic degradation of landfill leachate were conducted at the laboratory or pilot plant scale. In this study, use of photocatalytic properties of titanium dioxide nanoparticles stabilized on bentonite and electrochemical methods using platinum and graphite electrodes were used to remove the chromium and COD from leachate.

2.1. Leachate sampling

Leachate samples (1,000 cc) were collected from the Kahrizak landfill site in Tehran, Iran. This 55-year-old landfill produces a dark black colored liquid with high levels of COD and heavy metals with low BOD/COD ratio. The Kahrizak landfill site has an area of 1,400 ha that receives 7,000 tons of MSW daily. The samples were injected into the reactor under different retention times, electrical density, UV radiation intensity and concentration of catalyst. The characteristics of raw leachate samples are shown in Table 1.

2.2. Pilot system and used apparatus

In this study, a direct power supply was prepared with an output current of 5 A. Positive pole and negative pole were connected to a platinum and graphite electrode, respectively. To use ultraviolet light (UV), eight 6-watt lamps were placed in a quartz cylindrical chamber. The reactor was made with a working volume of 2 L, and a water circulation system was exploited to adjust the reactor temperature. Supernatant (1,000 mL) was collected and subjected to electrochemical degradation. During the examination of different variables, 5 mL

Table 1
The characteristics of raw leachate

Parameter	Unit	Value
COD	mg/L	49,610
BOD	mg/L	5,500
TSS	mg/L	30,249
pH	–	7.8
BOD/COD	–	0.11
Temperature	°C	22.4
Chromium	mg/L	0.05
Landfill age	Year	55
Phosphate	mg/L	92.6
Ammonia nitrogen	mg/L	2,700
Sulfate	mg/L	1,800
Chloride	mg/L	4,100

of the reaction suspension was transferred into a conical centrifuge tubes every 10 min and centrifuged (4,000 rpm for 10 min) to remove nanoparticles. The supernatant was then taken using a sampler. In this study, 0.05 M sodium sulfate was used as the electrolyte in the catalytic oxidation reactor. It has been reported that the use of alkaline electrolyte will increase the production efficiency of hydroxyl radicals through hydroxide ions. The reactor was also equipped with a pH meter and the solution pH was maintained at about 7.2 throughout the study. The impact of factors including electrical density (1–4 A/m^2), reaction time (1–8 h), catalyst concentration (1–4 g/L) and the intensity of UV radiation (3–8) were investigated. It is worth noting here that 6 W UV lamps were used in our study. The solution was continuously mixed in order to ensure the correct homogenization of suspension and the sufficient contact between leachate with catalysts and electrodes.

The electrochemical reactor included an anode electrode plate (dimensions of 3 × 6 cm^2), a commercial platinum electrode and a graphite cathode (dimensions of 3 × 8 cm^2). The electrical current was directly supplied by a digital power supply (Central Stores PS-305D model). Fig. 1 shows the schematic diagram of the reactor.

The TiO$_2$ nanoparticles were fixed on bentonite using a standard method [18]. Fig. 2 shows the fixed nanoparticles.

2.3. Experimental procedures and analytical methods

The samples were digested with concentrated nitric acid. Chromium content of the samples was measured by atomic absorption [19,20]. Chromium and organic matter (BOD and COD) were determined according to standard methods for examination of water and wastewater [21]. Results were analyzed using Microsoft Excel and SPSS (version 20).

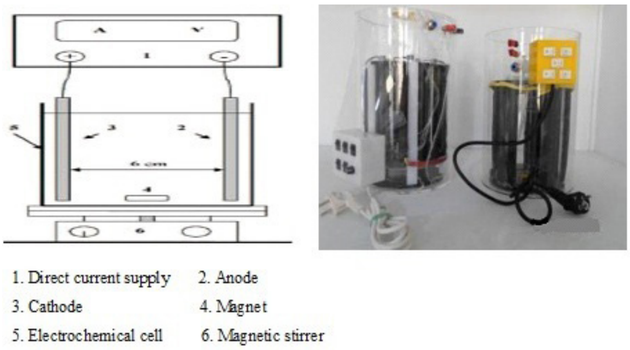

Fig. 1. Schematic diagram of the electrochemical reactor.

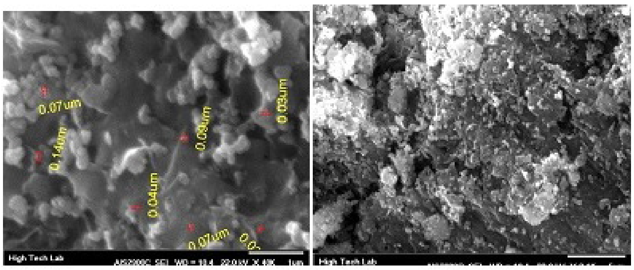

Fig. 2. TiO$_2$ nanoparticles fixed on bentonite.

3. Results and discussion

This study investigated the effectiveness of the electrochemical process using graphite and platinum electrodes and photocatalytic properties of TiO$_2$ nanoparticles stabilized on bentonite at different reaction times for removal of chromium and organic materials from leachate. The pH of leachate samples in all tests was adjusted within the neutral range. The concentration of COD was extremely high in this type of leachate, which can be classified as severe pollution. The removal efficiency of organic matter and chromium from leachate flow at the optimum current density and different reaction times are presented in Table 2. Maximum removal of BOD and COD was observed after 8 h. Maximum removal of Cr(VI) was observed after 4 h.

The removal efficiency of studied parameters at the various electrical current densities is presented in Table 3. Maximum removal of BOD, COD and Cr was observed with 4A/m^2 of electrical current density. The removal efficiency of the studied parameters with different number of UV lamps is presented in Table 4. Maximum removal of BOD, COD and Cr(VI) was observed with eight UV lamps.

Based on the results, maximum removal of BOD, COD and Cr(VI) was observed when using 4 g/L of the catalyst (Table 5).

Fig. 3 shows the removal efficiency of BOD, COD and Cr(VI) in the electrocoagulation and nanophotocatalytic processes with respect to the reaction time. Maximum removal of BOD and COD was observed after 8 h. Maximum removal of Cr(VI) was observed after 4 h. Figs. 4 and 5 show the removal efficiency of BOD, COD and Cr(VI) in the electrochemical and nanophotocatalytic processes with respect to the intensity of UV radiation and concentration of the catalyst.

3.1. Effect of reaction time

As shown in Table 2 and Fig. 3, rate of decomposition increased with increasing the reaction time. In the electrochemical and UV/TiO$_2$ process, significant amounts of Cr(VI) were removed after 4 h. Ku and Jung studied the photocatalytic reduction of Cr(VI) in aqueous solutions by UV irradiation in the presence of TiO$_2$ and found that the removal of Cr(VI) increased with increasing the reaction time [22]. Maximum removal of BOD and COD was observed after 8 h. The increase in reaction time increases removal of COD and BOD and Cr(VI) and the degradation efficiency [23,24].

During the electrochemical process, two active oxygen species can be electrochemically generated on oxide anode (MO$_x$). One species is the chemisorbed (oxygen in oxide lattice, MO$_{x+1}$) responsible for electrochemical conversion through Eq. (1), the other is physisorbed (absorbed hydroxyl radicals, °OH) responsible for electrochemical combustion through Eq. (2) [17].

$$\mathrm{R} + \mathrm{MO}_{x+1} \rightarrow \mathrm{RO} + \mathrm{MO}_x \qquad (1)$$

$$\mathrm{R} + \mathrm{MO}_x(^{\circ}\mathrm{OH})_z \rightarrow \mathrm{CO}_2 + z\mathrm{H}^+ + z\mathrm{e} + \mathrm{MO}_x \qquad (2)$$

where R is the organic compounds and *z* is the number of absorbed °OH on anode.

Table 2
The removal efficiency of studied parameters at the various reaction time (catalyst dose = 4 g/L, number of UV lamps = 8, electrical current density = 4 A/m^2)

Parameter								
Time (h)	1	2	3	4	5	6	7	8
BOD	5,700	6,100	6,300	6,500	6,800	6,900	5,800	4,600
COD	39,800	36,500	32,250	28,200	26,700	25,800	21,900	20,500
BOD/COD	0.14	0.17	0.2	0.23	0.25	0.27	0.26	0.22
Cr	0.017	0.019	0.012	0.006	0.015	0.012	0.011	0.009

Table 3
The removal efficiency of studied parameters at the various electrical current densities (catalyst dose = 4 g/L, number of UV lamps = 8, reaction time = 8 h)

Parameter	Electrical current density (A/m^2)			
	1	2	3	4
BOD	7,100	6,150	5,850	4,700
COD	30,450	22,500	20,900	12,500
BOD/COD	0.23	0.27	0.28	0.38
%COD	38.6	54.6	57.8	74.8
Cr	0.012	0.011	0.01	0.004
%Cr	76	78	80	92

Table 4
The removal efficiency of studied parameters at the various radiation lamps (catalyst dose = 4 g/L, electrical current density = 4 A/m^2, reaction time = 8 h)

Parameter	Number of UV lamps					
	3	4	5	6	7	8
BOD	6,200	6,380	6,260	6,150	6,050	4,200
COD	23,300	16,300	13,800	12,400	10,800	10,100
BOD/COD	0.27	0.39	0.4	0.5	0.56	0.42
Cr	0.017	0.013	0.011	0.009	0.006	0.003

Table 5
The removal efficiency of studied parameters at the various dosage of catalyst (electrical current density = 4 A/m^2, number of UV lamps = 8, reaction time = 8 h)

Parameter	Catalyst dosage (g/L)			
	1	2	3	4
BOD	7,000	5,200	4,400	3,800
COD	23,700	10,900	5,400	4,380
BOD/COD	0.3	0.48	0.81	0.87
Cr	0.015	0.007	0.006	0.001

During the electrochemical process, organic compounds are only partially oxidized, so that a subsequent biological treatment may be required [17]. The results of this study show that the electrochemical process increases the biodegradability. Due to their higher cost, electrochemical processes can be used as a pretreatment before the biological process.

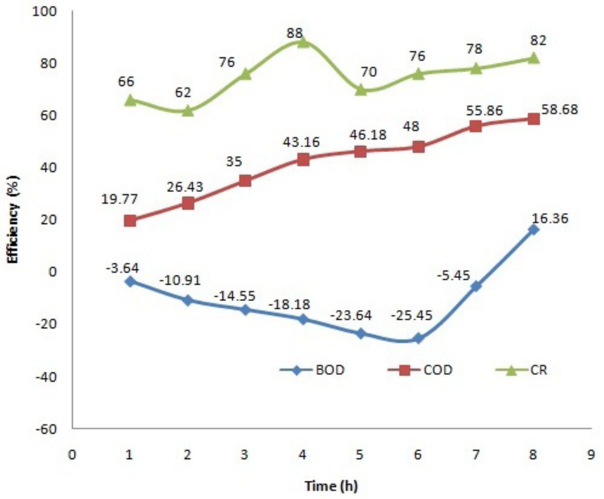

Fig. 3. Removal efficiency of BOD, COD and Cr(VI) at various reaction times (catalyst dose = 4 g/L, number of UV lamps = 8, electrical current density = 4 A/m^2).

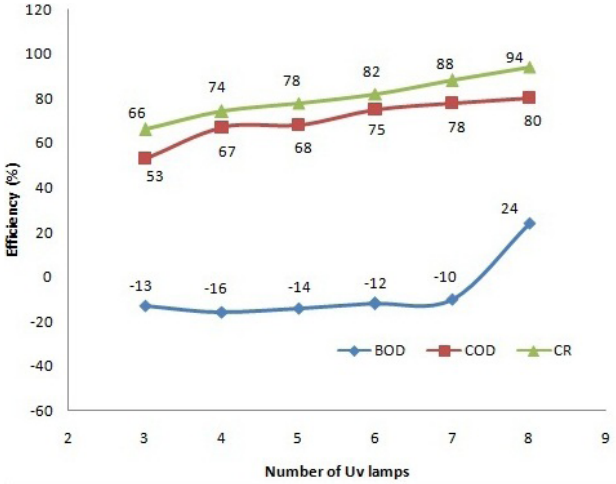

Fig. 4. Removal efficiency of BOD, COD and Cr(VI) at various intensity of UV radiation (catalyst dose = 4 g/L, electrical current density = 4 A/m^2, reaction time = 8 h).

According to Faraday's law for electrochemical processes, electrolysis time increases the amount of metal ion

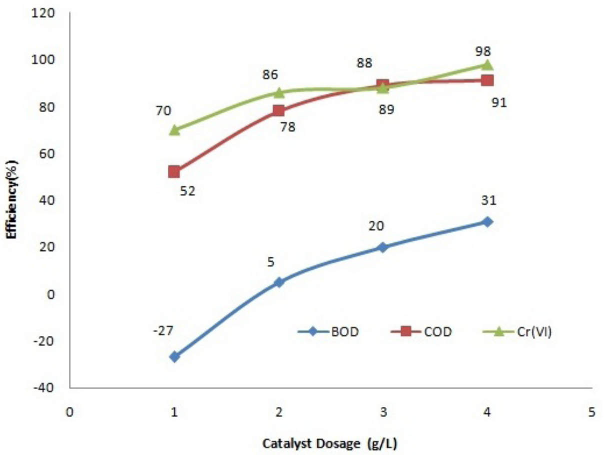

Fig. 5. Removal efficiency of BOD, COD and Cr(VI) with various doses of catalyst (electrical current density = 4 A/m^2, number of UV lamps = 8, reaction time = 8 h).

release into the system. The hydroxide ions produced by the electrochemical reaction in the cell are dependent on the current density and will follow the Faraday's law. Electrode consumption rate can be calculated using the following equation:

$$\Delta m = I\,t\,M/Z\,F \quad (3)$$

where I is the electrical current density, t is the reaction time, Z is the number of electrons moved in the reaction, F is the Faraday coefficient 96,486 c/mol and M is the molecular weight.

Reduction of electrical current density increases the time required for achieving the same efficiency. According to Chen et al. [25], this could be due to the effect of loading rate on the treatment efficiency [26].

$$Q = I \times t \quad (4)$$

The ions produced in the higher reaction time needed to higher load, but this parameter due to the economic costs is kept low.

3.2. Effect of electrical current density

When applying current density of 4 A/m^2, BOD$_5$/COD ratio increased from 0.23 to 0.38, as a result this process could be considered as a pretreatment for the biological treatment. Increasing the electrical current reduces the time required for achieving the same removal efficiency. This is because the increased electrical current increases ion release from the electrode, and thus elevates the precipitation and removal of the contaminants. In addition, it increases the number of bubbles produced and reduces the bubble sizes [26,27]. Therefore, it can be concluded that the electrical current affects the efficiency of the process and breakdown of organic matter and non-biodegradable materials. In this study, increasing the electrical current led to the removal of color and turbidity from the leachate, and increased efficiency. It is, however, worth noting here that corrosion rates are increased by raising current densities, which may eventually be profitless. Thus, the electrode exploited in this study could be affordably used under considerably higher current densities, which is believed as a positive and innovative idea of this research.

3.3. Effect of the number of UV lamps

The highest COD removal efficiency (80%) was observed after 8 h in the presence of eight UV lamps, while the lowest COD removal efficiency (53%) was observed in the presence of three UV lamps. Increase in the number of UV lamps increases BOD$_5$/COD ratio from 0.27 to 0.56, which is suitable for biological treatment.

Increased UV radiation intensity caused increases in the number of excited electrons, hydroxyl radical production and COD removal rate. The highest rate of Cr(VI) removal occurred in the presence of eight tubes (0.003 concentration in mg/L) and three bulbs (0.017 concentration in mg/L).

Titanium dioxide exhibits photocatalytic activity under ultraviolet irradiation. With increasing intensity of ultraviolet radiation, the number of electrons is further stimulated. Increasing in the excitation of electrons was due to the increased intensity of the radiation received on a bed of titanium oxide nanoparticles stabilized on the bentonite [28].

The photocatalytic oxidation reaction often described for irradiated titanium dioxide is summarized as follow:

$$\mathrm{TiO_2} + \mathrm{hv} \rightarrow \mathrm{TiO_2}.(\mathrm{e^-_{cb}} + \mathrm{hole^+_{vb}}) \quad (5)$$

Recombination of electrons and holes on the surface of the catalyst is a common reaction, reducing the quantum efficiency of the process.

$$\mathrm{TiO_2}.\left(\mathrm{e^-_{cb}} + \mathrm{hole^+_{vb}}\right) \xrightarrow{\mathrm{recomb}} \mathrm{TiO_2} + \mathrm{heat} \quad (6)$$

Oxidizing species such as oxygen molecules may also react with electrons. Positives holes can oxidize these species, which subsequently result in reducing the possibility of recombination.

Reactions that yield additional hydroxyl radicals may also continue along with other reactions. Hydroxyl radicals are highly reactive and consequently can oxidize organic pollutants, directly or through their intermediate compounds [28].

The increased removal efficiency was due to increased UV radiation to catalyst. In addition, the electrons and hydroxyl radicals increased. The hydroxyl radical production subsequently increases the photocatalytic oxidation rate [29,30].

3.4. Effect of concentration of the catalyst

In the photocatalytic UV/TiO$_2$ process, pH remained neutral after 8 h. The highest rate of COD removal (86.3%) was observed in the presence of seven UV lamps when using 4 g/L of catalyst. However, the lowest percentage of COD removal (39.3%) was observed when using 1 g/L of catalyst. In other words, increasing the dose of catalyst from 1 to 4 g/L in the leachate solution increased the removal of COD and BOD from 30,100 to 6,780 and from 8,400 to 5,200, respectively.

Moreover, BOD_5/COD ratio increased from 0.27 to 0.76 when using 4 g/L of catalyst. The highest Cr(VI) removal efficiency (84%) was observed when using 4 g/L of catalyst, while the lowest Cr(VI) removal efficiency (72%) was observed when using 1 g/L of catalyst. In other words, increasing the concentration of the catalyst from 1 to 4 g/L in the leachate solution increased the removal of chromium from 0.014 to 0.008 mg/L.

Meshram et al. [31] evaluated removal of phenol using ZnO–bentonite nanocomposite (photocatalyst) with UV irradiations under continuous stirred tank reactor. They found that the degradation efficiency increases by increasing the concentration of nanocomposite [31]. Meeroff et al. [32] compared photochemical iron-mediated aeration and TiO_2 photocatalysis for the treatment of landfill leachate at laboratory scale. Bench scale testing of real landfill leachate samples using the two potential on-site pretreatment technologies showed up to 86% conversion of refractory COD to complete mineralization, up to 91% removal of lead, up to 71% removal of ammonia without pH adjustment and up to 90% effective color removal with detention time of 4–6 h using 4 g/L of catalyst [32].

4. Conclusions

This study was designed to evaluate the use of photocatalytic and electrochemical processes for landfill leachate treatment. Using graphite and platinum electrodes causes no corrosion in the treatment system. Based on the results of this study it can be concluded that:

- UV radiation alone in the photocatalytic process is unable to breakdown resistant pollutants in the leachate. This is due to high turbidity of leachate and inability of UV radiation to produce enough active hydroxyl radicals.
- Electrical current density affects the efficiency of the process and could breakdown organic material with low biodegradability. Thus, increasing electrical current density leads to removal of color and turbidity from leachate and improves the photocatalytic effect and efficiency. At 4 A/m^2 electrical current, the COD and chromium removal efficiencies are 74.8% and 95%, respectively. This current also increases the BOD_5/COD ratio and provides the conditions suitable for biological treatment.
- The number of UV lamps also affects the efficiency of the process in a way that increasing the number of lamps increases COD removal rate up to 80% and increases BOD_5/COD ratio to 56%. The highest chromium removal efficiency (94%) is achieved when using eight UV lamps.
- Concentration of the catalyst also affects the efficiency of COD removal. Increasing the dose of catalyst increases COD removal efficiency and BOD_5/COD ratio (to 87%), providing suitable conditions for biological treatment of leachate.
- The bentonite used in this study is suitable for color reduction and removal of organic matter.
- The electrochemical pretreatment process can remove organics matter, heavy metals, reduce organic load and increase wastewater biodegradability. Thus, it can be used as an efficient option for treating sewage, preventing environment pollution and protecting water resources. However, more studies should be conducted to evaluate the economic aspect of using this process.
- The photocatalytic process combined with UV/TiO_2 is more efficient for removal of organic matter and chromium compared with the electrochemical or photocatalytic process alone.
- The hydroxyl radical is a very powerful oxidizing agent which can oxidize organic pollutants, directly or through intermediate compounds.

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Competing interests

Not applicable.

Authors' contributions

The article is derived from a Master's thesis by Elahe Rezaie. Elahe Rezaie proposed the study design and hypothesis and conducted data collection. Mahdi Sadeghi and Ghodratollah Shams Khoramabadi were advisers of the thesis. Data analysis and manuscript preparation were done by Mahdi Sadeghi. All authors read and approved the final manuscript.

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