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Electrochemical removal of acid red 18 dye from synthetic wastewater using a three-dimensional electrochemical reactor

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ABSTRACT

Electro-oxidation degradation of acid red18 (AR18) dye from synthetic wastewater was investigated using a three-dimensional (3D) electrochemical reactor with granular activated carbon (GAC) as a particle electrode. The effect of operating parameters such as pH, current density, particle electrode dose, dye concentration and electrolysis time was studied and the optimization was performed using Taguchi fractional factorial design. The results showed that maximum removal of AR18 (96.5%) and COD (84.8%) were obtained under pH of 3, current density of 20 mA/cm^2, initial concentration of dye of 100 mg/L, GAC dose of 250 mg/L and time of 45 min. High R^2 and P value < 0.05 showed the excellent fitting of the selected model with experimental data. In addition, the experimental results showed that current density was the most important factor in the removal of the AR18 and COD. The result of comparative tests showed higher electrocatalytic activity, higher H_2O_2 production and lower energy consumption of 3D electrochemical reactor compared to the electrolysis process alone. The reusability and stability tests demonstrated that GAC, as a particle electrode, has a high potential for catalytic degradation of AR18 from aqueous solution at six consecutive runs. Finally, the 3D electrochemical process can be considered as a good alternative to treat wastewater containing AR18 dye.

Keywords: Three-dimensional electrochemical; Acid red 18; Granular activated carbon; Chemical oxygen demand; Synthetic wastewater

1. Introduction

Discharged effluents from the textile industry, leather manufacturing, paper and food industry contain high concentrations of color and COD, which have a negative effect on environmental ecology [1]. Among all kinds of dye materials, azo dyes such as acid red 18 (AR18) have a great application due to properties such as high solubility, the simplicity of dyeing, cheap and easy production. These colors, due to the chemical structure containing the azo band (–N=N–) and aromatic rings, are non-degradable and resis-

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tant against the light and chemical oxidizing agent [2–5]. According to studies, the presence of azo dyes in the environment leads to environmental and health problems such as skin irritation, allergy, cancer and mutation [6]. Therefore, the removal of dye from aqueous solutions before discharging to the environment is necessary and crucial.

In recent years, various biological and physicochemical methods such as activated sludge, coagulation, filtration, membrane filters have been used to remove the dyes from aqueous solutions [7,8]. However, these methods for removing the pollutants with a complex and stable structure have disadvantages such as relatively expensive, secondary pollutants production, low efficiency, and transmission of contamination from one phase to another [6,7]. To overcome these problems, the advanced oxidation process (AOPs) has recently been considered due to unique features such as high efficiency, high mineralization, and non-production of hazardous products [9]. In this process, radical hydroxyl (•OH, a strong oxidizing agent with an oxidation potential of 2.8 V) produced by chemical and photochemical methods can react with organic compounds and their intermediates until their complete mineralization into mineral ions, water and CO_2 [10,11]. Among the AOPs, most researchers proposed electrochemical advanced oxidation processes (EAOPs) as a promising process for treating aqueous solutions containing high concentrations of persistent organic pollutants [10,12]. The two-dimensional (2D) electrochemical reactor, as an EAOP, has the features such as producing high levels of various oxidants, easy operation, simple configuration and easy maintenance in the effective degradation and even complete mineralization of pollutants [12,13]. In spite of these advantages, there are still some drawbacks limiting the practical application of 2D electrochemical, e.g., low mass transfer, low energy efficiency, short lifetime of electrodes and temperature increase [14,15]. To improve these disadvantages, particles such as granular activated carbon were added between anode and cathode electrodes to form a three-dimensional electrochemical reactor [12]. During the electro-oxidation process, the particles are easily polarized into the form of a large number of bipolar micro electrodes, which not only reduces the distance between the electrode and the reactant, but also increases mass transfer, the development of electrochemical reactions, the contact surface and the efficiency of the electrolysis process alone [14,16]. In addition, particles as the electrode can increase some of the catalytic reactions, such as reducing the electrochemical oxygen to H_2O_2 and then decomposing it into •OH [17]. Recently, 3D reactor technology has been successfully used to remove various organic pollutants. Sowmiya et al. [18] investigated the degradation of reactive B dye by a 3D electrochemical process and found that the 3D process, due to the presence of a particle electrode, provides high removal efficiency and low energy consumption compared to the 2D process. Li et al. [19] reported that the removal efficiency of pyrimidine and COD in the 3D process is higher than the 2D process due to an increase in the direct and indirect oxidation process. In addition, Zhang et al. [20] and Wei et al. [21] investigated the use of 3D electrochemical reactors for treatment of pollutants and found that the presence of the particle electrode is led to facilitate the electro-oxidation reactions and mass transfer in the degradation process.

The purpose of the present study was to investigate the effectiveness of the 3D electrochemical process coupled with activated carbon as a particle electrode in the removal of AR18 dye from aqueous solutions. Taguchi design model was used to optimize the operational parameters (such as pH, GAC dose, initial dye concentration, current density, and electrolysis time) and to reduce the number experiments. Comparative experiments were performed to better understanding the effect of particle electrodes in the electrochemical process. The stability and re-usability of the GAC catalyst were evaluated by repeating the experiments under the same conditions.

2. Materials and methods

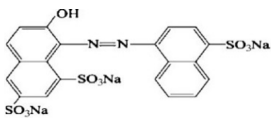
2.1. Materials

Acid red18 dye, with the characteristics shown in Table 1, was purchased from Sigma Aldrich Company. NaCl and HCl were obtained from the Merck Co., Germany. At all stages of this study, the distilled water was used and the chemicals were of analytical grade. Granular activated carbon (GAC) with a purity of 99.99% was provided from the Sigma Aldrich Company and was used as a particle electrode for the electrochemical treatment of dye.

2.2. Experimental procedures

Electro-oxidation of AR18 in a synthetic solution was performed in a 0.5 L reactor (Fig. 1). Titanium and graphite felt with dimensions of 5×10×2 cm were used as anode and cathode. Granular activated carbon was used as a particle electrode in a fluidized bed between electrodes. The distance between the anode and cathode electrodes was adjusted according to the requirement. To enhance the ionic strength and to increase the electrical conductivity of the solution, the NaCl electrolyte was used at a constant value of 0.1 M. The initial pH of the solution was adjusted with 0.01 M HCl and NaOH. In order to prepare a uniform solution during the electrolysis process, the mixture was stirred by a magnetic magnet at a constant speed of 300 rpm. The oxygen required for electrochemical production of the H_2O_2 with a constant flow rate of 1 L/min was provided by the HEILEA air compressor (model of ACO-5505, China). To adjust the current, a power supply (micro, PW-4053R model, Iran) was used. The Wattman filter was used to separate the granular activated carbon from the solution. Finally, the process of oxidation was also carried out on real wastewater samples.

Table 1
Chemical properties of AR 18

Parameters	
Molecular formula	$C_{20}H_{11}N_2Na_3O_{10}S_3$
Molecular weight	604.5 (g/moL)
COD of 1 g-AR18/L	597 ± 17 (mg/L)
λ_{max}	507 (nm)
Chemical structure	
CAS-NO.	2611-82-7

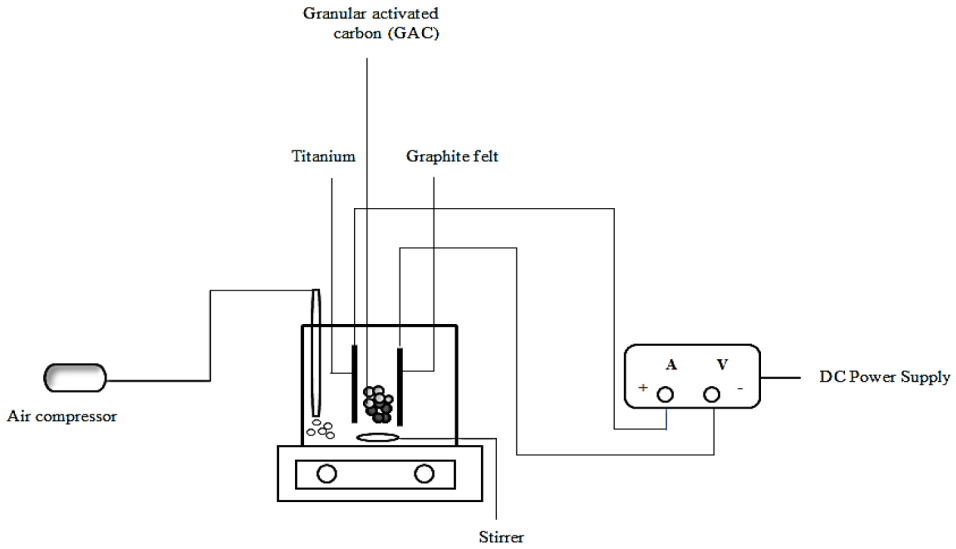


Fig. 1. Schematic diagram of the 3D electrochemical reactor.

2.3. Analytical method

To evaluate the individual effect of pH, particle electrode dose, initial dye concentration, current density and reaction time on the removal of AR18 and COD, data was analyzed using the Design Expert software (Design Expert 10 Stat-Ease, Inc., USA). Taguchi orthogonal design model with 5 factors in 4 levels (Table 2) was used to optimize the electro-oxidation degradation process. The chemical oxygen demand was determined by the standard method (D5220). The concentration of H_2O_2 was determined by titration with potassium permanganate. In this method, 100 mL of the working solution containing H_2O_2 and 3 mL of concentrated HCl was titrated by 0.01 M potassium permanganate until the appearance of purple color. Then, the determination of the concentration of H_2O_2 produced was calculated by Eqs. (1) and (2). The concentration of AR18 was measured by UV-vis spectrophotometer at the wavelength of 507 nm. The removal efficiency (RE) and energy consumption (EC, kWh/m^3) were calculated using Eqs. (3) and (4).

$$5H_2O_2(aq) + 2MnO_4^-(aq) + 6H^+(aq) \rightarrow 5O_2(g) + 2Mn^{2+}(aq) + 8H_2O \quad (1)$$

Table 2
Experimental design parameters

Factors	Level 1	Level 2	Level 3	Level 4
pH	3	5	7	9
Current density (mA/cm^2)	10	15	20	25
AR18 concentration (mg/L)	25	50	100	150
Particle electrode dose (mg/L)	50	100	250	500
Electrolysis time (min)	15	30	45	60

$$\frac{2}{5} = \frac{V_{MnO_2} \times M_{MnO_2}}{V_{H_2O_2} \times M_{H_2O_2}} \quad (2)$$

$$\mathrm{RE}(\%) = \frac{C_0 - C_f}{C_0} \times 100 \quad (3)$$

$$\mathrm{EC}\left(\frac{\mathrm{kWh}}{\mathrm{m}^3}\right) = \frac{\mathrm{IV_c t}}{\mathrm{V_s}} \quad (4)$$

where V is the solution containing H_2O_2 (L), M is molar (moL/L), C_0 is AR18 initial concentration (mg/L), C_f is the final concentration of AR18 (mg/L), I is the current (A), V_c is the cell voltage (V), t is the treatment time (h), and V_s is the treated volume (m^3).

The stability and re-usability of the particle electrode was investigated through the consecutive reaction cycle. During these reactions, the particles used were washed with distilled water, dried in an oven at 60°C and then added to the system.

3. Results and discussion

3.1. Analysis of variance

To understand the effect of each parameter on the removal efficiency of dye and COD, the analysis of variance (ANOVA) using the Design Expert software and Taguchi design model was studied and the results were shown in Table 3. In the ANOVA, P value indicates the importance of a parameter in the model and the F value represents the level of significance of the parameter in the model. Considering the results of Table 3, it can be seen that all factors have a significant effect on the removal efficiency of dye and COD due to high F values and low P value. The F-value of the model for removing the dye and COD was 704.7 and 27.1 respectively, which indicates that the model is significant and the probability of error for F-value is only 0.01%. The P values of less than 0.0001 show that the model is sig-

Table 3
ANOVA table for AR18 and COD removal

Source	DF	Sum of squares		Mean squares		F value		prob > F
		AR18	COD	AR18	COD	AR18	COD	
Model	15	14183.3	13475.6	945.5	898.3	704.7	27.1	<0.0001
A-pH	3	1930.4	1721.6	643.4	573.8	479.5	17.3	<0.0001
B-current density	3	4211.4	4553.2	1403.8	1517.7	1046.2	45.8	<0.0001
C-initial dye concentration	3	3411.8	3267.3	1137.2	1089.1	847.6	32.8	<0.0001
D-GAC dose	3	929.9	941.2	309.9	313.7	231	9.4	<0.0001
E-time	3	3699.6	2992.2	1233.2	997.4	919.1	30.1	<0.0001
R^2		0.927	0.997					
R^2_{adj}		0.892	0.995					
R^2_{pre}		0.835	0.993					
C.V		8.45	2.08					

nificant. In addition, the results showed that the regression model is significant, based on the high coefficient of determination for the removal efficiency of dye (R^2 = 0.927) and COD (R^2 = 0997). Typically, the ratio of the coefficient of variation (CV) less than 10% can be considered as a logical model in terms of repeatability. As shown in Table 3, the CV values for removal of both dye and COD were relatively small and were 8.45% and 2.08%, respectively. In addition, the correlation coefficient of the proposed model (R^2_{Pred}) for the removal efficiencies of dye and COD is logically consistent with the correlation coefficient of the adjusted model (R^2_{Adj}). The results represented in Table 3 also showed that the current density, due to high F value and the sum of squares, is the most important factor among all operational parameters. Zhang et al. [14] reported that current density is one of the important parameters in the 3D process due to the effect on electrochemical oxidation and polarization behavior of the particle electrode. At a suitable current, the particle electrode is polarized in the form of different microelectrodes, which not only increases the electro-sorption, but also increases the electro-oxidation efficiency by creating cathode and anode electrodes on both sides of the particles.

Figs. 2 and 3 show diagnostic plots such as predicted versus actual and normal probability plots for experimental data. As shown in Fig. 2, there is a good relationship between predicted and experimental values, which indicates the adequacy of the selected model to assume the response variable for experimental data. In addition, Fig. 3 shows that the residuals are close to the straight line, which confirms the normal distribution of observed data. The

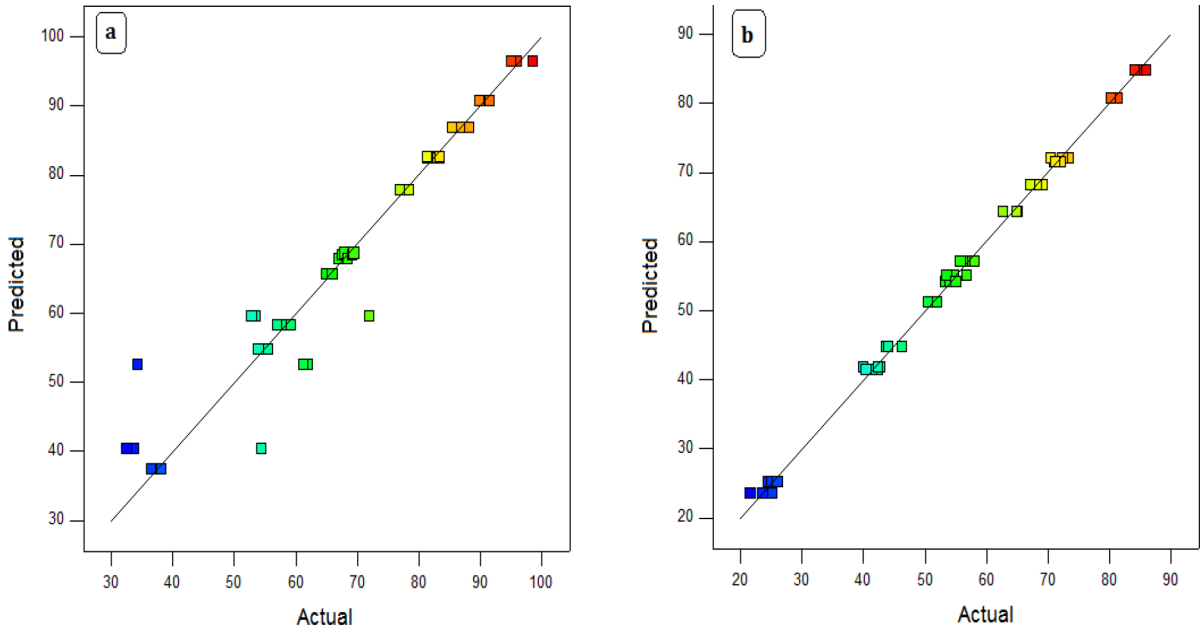


Fig. 2. Predicted vs. actual values plots for removal of AR18 (a) and COD (b).

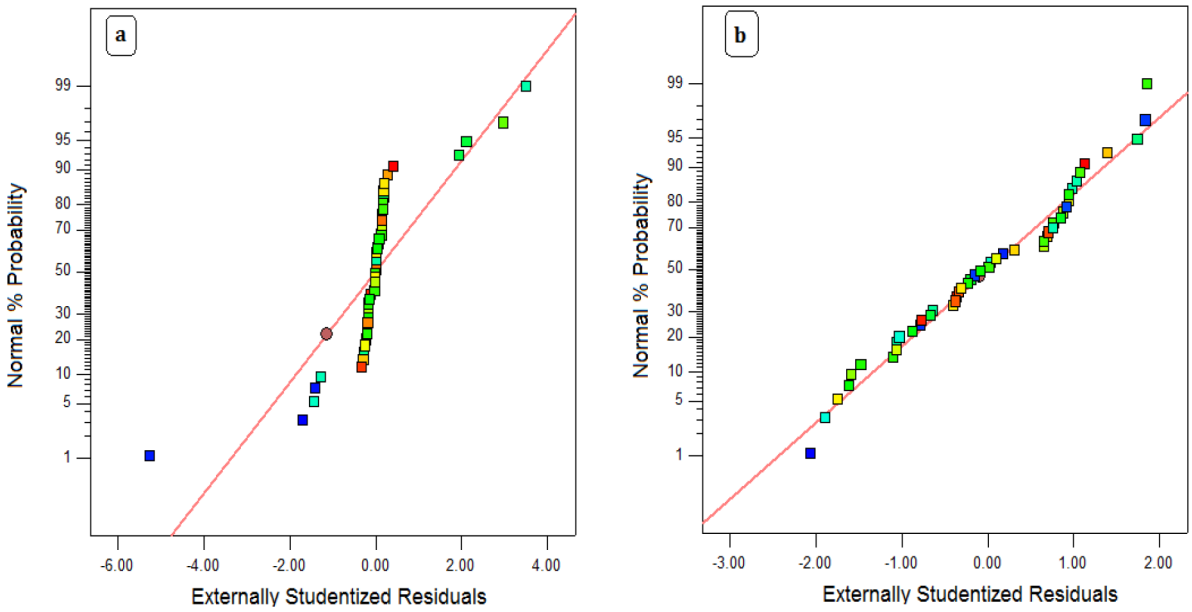


Fig. 3. Normal probability plot of residual for removal of AR18 (a) and COD (b).

above results explain the accuracy and applicability of the Taguchi design model for optimizing the removal of dye and COD.

3.2. The effect of parameters

The effects of different parameters such as initial pH, current density, initial concentration of dye, particle electrode dose and reaction time on the removal of dye and COD are shown in Fig. 4 at the average level of each factor. As shown in Fig. 4 (a), increasing the initial pH from 3 to 9 has led to decreasing the removal efficiencies of the dye and COD from 78.25% and 65.57% to 63.75% and 49.85%, respectively. This may be due to the instability of H_2O_2 and the reduction of $^{\bullet}OH$ concentration due to the presence of carbonates and bicarbonates as scavengers [21]. Kong et al. [22] performed the electrochemical treatment of anionic surfactants using 3D electrodes and reported that by increasing the initial pH from 3 to 10, the pollutant removal efficiency decreases due to the accumulation of hydroxyl on the anode surface and the competition with anodic material for oxygen production. Wei et al. [21] showed that the process efficiency increases by reducing the initial pH due to the easy control of oxygen evolution and the overall current improvement for the direct and indirect oxidation mechanism. In addition, the results of this study were consistent with the studies conducted by Qin et al. [23] and Xiong et al. [24]. The effect of the current density on the removal efficiency of dye and COD is shown in Fig. 4 (b). According to this figure, the removal efficiencies of the dye and the COD were initially increased and then were decreased by more increase in the current density. For example, by increasing the current density from 10 mA/cm^2 to 20 mA/cm^2, the COD removal efficiency rises from 43.62% to 66.03%. However, a further increase in the current density from 20 mA/cm^2 to 25 mA/cm^2 reduces the efficiency from 66.03% to 63.72%. The initial increase in efficiency can be explained by the fact that, at a proper current density, the particles are polarized in the form of multiple micro electrodes, which can increase the electro sorption and oxidation [14]. In addition, increasing the current density is led to an increase in the electrochemical production of H_2O_2 and $^{\bullet}OH$ on the surface of the main electrodes and GAC. Similar results were obtained by Iranpour et al. [6] in the degradation of reactive black 5 from aqueous solutions. Pourzamani et al. [25] examined the use of carbon nanotubes to developing the degradation of diclofenac in a 3D electrochemical reactor, and found that the diclofenac removal efficiency increases by increasing the current density due to the production of the high amount of $^{\bullet}OH$ at the surface of Ti/TiO_2-RuO_2 and the carbon nanotube. Reducing the efficacy at high current densities may be due to the presence of parasitic electrochemical reactions such as reducing O_2 to H_2O instead of H_2O_2. In addition, an increase in current density leads to the evolution of oxygen and hydrogen and the lack of their participation in the main reactions for the production of the reactive species [26]. Eshaghzadeh et al .[27] obtained the same results and reported that, by increasing the current density, the removal efficiency of the dye was decreased due to the occurrence of unwanted reactions and production of hydrogen and oxygen gases [Eqs. (5) and (6)]. In addition, Li et al. [19] reported similar results for the electrochemical degradation of 2-diethylamino-6-methyl-4-hydroxypyrimidine.

$$2H^+ + 2e^- \rightarrow H_2 \quad (5)$$

$$2H_2O \rightarrow O_2 + 4H^+ + 4e^- \quad (6)$$

The results of Fig. 4c show that, by increasing the initial concentration of dye, the efficiency of removal of dye and COD significantly decreases. This reduction amount for dye

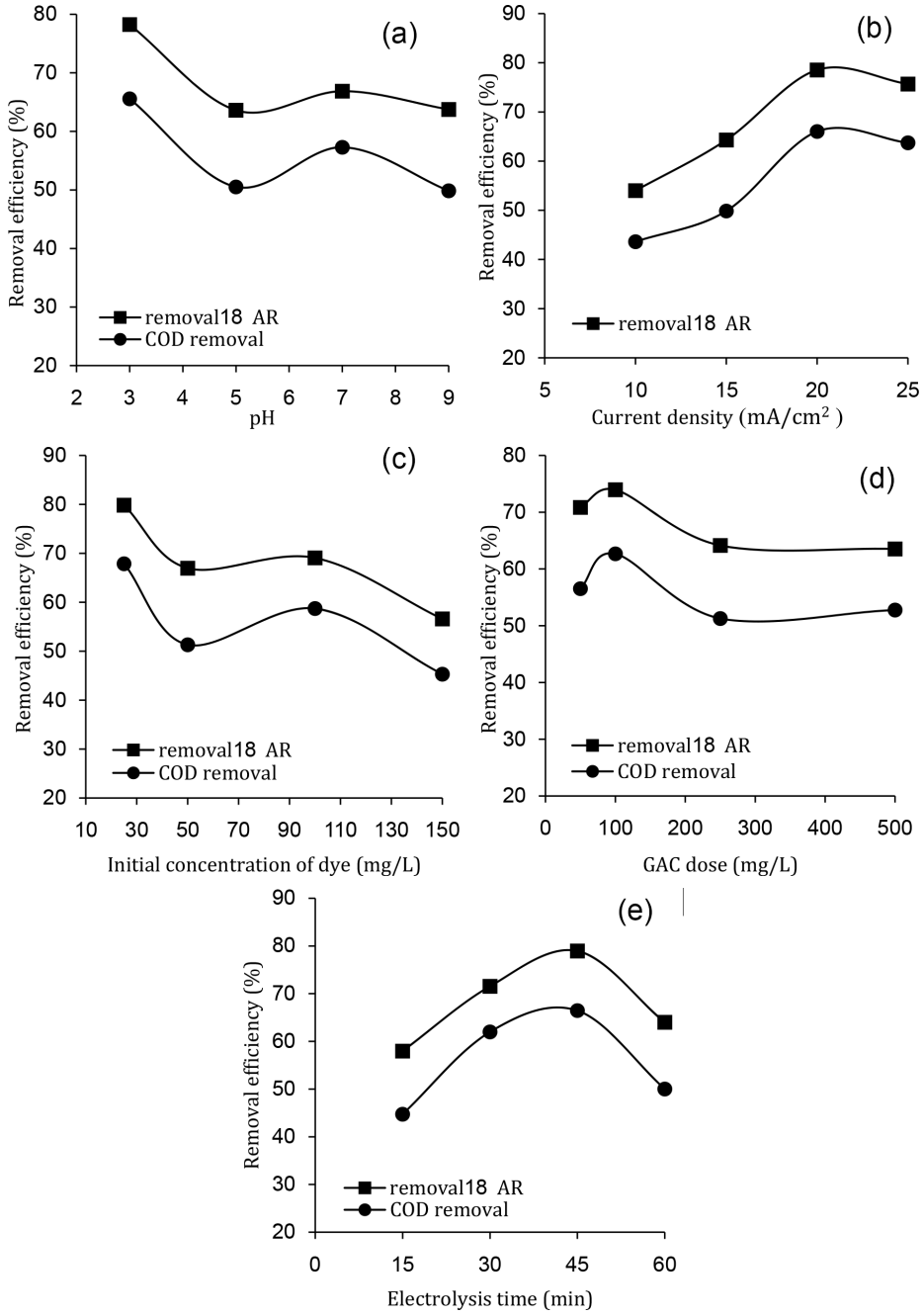


Fig. 4. The effect of the parameters on the efficiency of removal of AR18 and COD under the average level of each factor.

was from 79.84% to 60.66% and for COD was observed to be from 67.87% to 45.31%. This reduction in the removal efficiency in high concentrations of the dye can be interpreted as follows: when all factors are constant, a certain amount of reactive species such as H_2O_2, $^{\bullet}OH$, and active chlorine are produced. This amount of reactive species produced can degrade the specific amount of pollutant. Therefore, if the initial concentration of the pollutant increases, the amount of reactive species present in the solution is not sufficient for removal of excess molecules of the pollutant [28]. Similar results were observed on the removal of ibuprofen and naproxen by the 3D electro-Fenton process [29]. The results of Fig. 4c also show that the removal efficiency increases between 50 and 100 mg/L. This may be due to the more availability of reactive species to the pollutant. Similar results were observed by Manu [30] and Pourzamani et al. [17] for the degradation of diclofenac by oxidation processes.

The effect of the initial concentration of the particle electrode on the removal efficiency of dye and COD by the 3D process was studied. The results in Fig. 4d show that with increasing the initial concentration of GAC from 50 mg/L to 100 mg/L increased the removal efficiency of the dye and COD from 70.85% and 53.56% to 73.97% and 67.62%, respectively. The results of Fig. 5d also show that

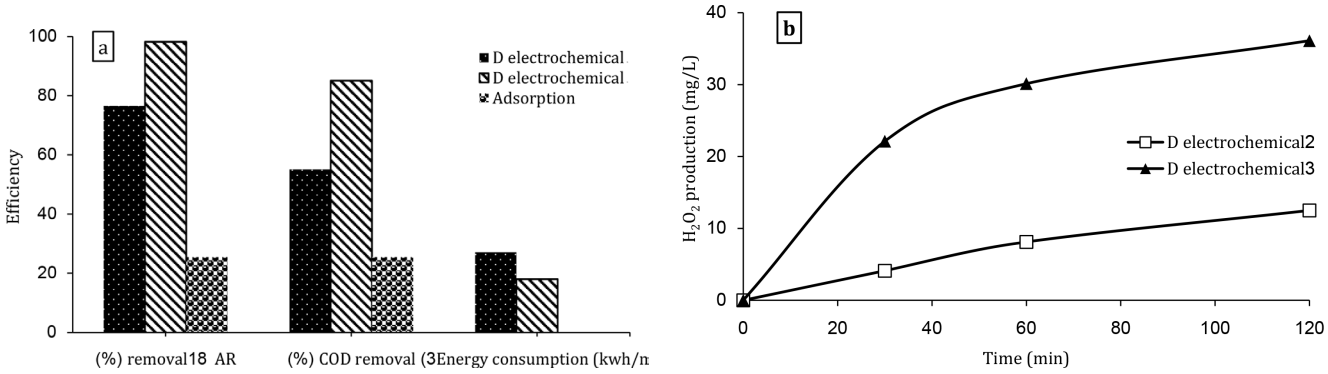


Fig. 5. Comparison of 2D and 3D electrochemical processes under optimal conditions.

by a further increase in the concentration of the particle electrode, the removal efficiency of both variables was decreased. The initial increase in the efficiency can be explained by the fact that the addition of GAC to the reaction solution is led to develop the efficiency of the direct and indirect electrolytic oxidation process due to the formation of multiple micro electrodes in the electric field. The activated carbon in the reaction medium, in addition to electrochemical reduction of the oxygen to H_2O_2, acts as a catalyst for the decomposition of H_2O_2 into the $^{\bullet}OH$ [Eqs. (7) and (8)]. However, with further increase in the particle electrode concentration, the removal efficiency of dye and COD is reduced due to the short circuit current. Sowmiya et al. [18] obtained the same results and reported that the dye removal efficiency is reduced due to increase the amount of electro sorption and, subsequently, the occurrence of short circuit current in the presence of additional particle electrode. Chu et al. [31] studied the effects of particle electrode dosage on the removal efficiency of COD and dye and founded that the removal of dye and COD increased by increasing the particle electrode. Wu et al. [32] also reported that the amount of decolorization was increased by increasing the particle electrodes due to increasing the surface area of the electrodes. In another similar study, Hou et al. reported that the addition of particle electrodes, in addition to increasing the surface area of the electrode, accelerates and promotes the electro-Fenton reactions [33].

$$O_2 + 2H^+ + 2e^- \rightarrow H_2O_2 \quad (7)$$

$$H_2O_2 + e^- \rightarrow OH^- + {}^{\bullet}OH \quad .(8)$$

The results of Fig. 4e show that by increasing reaction time, the removal efficiency is initially increased and then decreased. For example, with an increase in reaction time from 15 to 45 min, the removal efficiency of AR18 dye was increased from 57.97% to 78.97% however, it decreased to 64.01% by increasing the reaction time to 60 min. The initial increase can be due to the production of more reactive species by main electrodes and particles. The decrease in the efficiency at the long reaction time can be related to the occupation of active sites of GAC by intermediates produced by degradation of AR18 [34]. Similar results were observed by Liu et al. [35] for the removal of COD from aqueous solution. Xiong et al. [36] reported that with an increase in the reaction time, the removal efficiency was initially increased and then decreased due to parasitic and scavenging processes.

3.3. Process optimization

One of the main purposes of this study was to find the optimum conditions of the parameters to obtain the maximum removal efficiencies of dye and COD by the 3D electrochemical process. Based on this, the numerical optimization was performed using the Taguchi design model and the highest removal efficiency of dye (96.5%) and COD (84.8%) was achieved at pH of 3, current density of 20 mA/cm^2, initial dye concentration of 100 mg/L, GAC dose of 250 mg/L, and time of 45 min. In order to confirm the adequacy of the predicted optimal conditions, the verification experiments were carried out in triplicate according to above conditions and the average removal efficiency of the dye and COD was observed to be 98% and 85.9%, respectively. The good correlation between actual and predicted results showed the desirability of the Taguchi model and it could be effectively used to optimize the 3D electrochemical process parameters for textile industry effluent.

3.4. Comparison of 3D and 2D electrochemical systems

To find the electro catalytic performance of the GAC-based 3D process, the removal efficiencies of dye and COD were investigated in various processes and the results are shown in Fig. 5a. As can be seen, in the adsorption process by GAC, the dye removal efficiency was only 25.5%, which indicates that the adsorption of AR18 by GAC was inadequate. This may be due to the low amount of catalyst and lack of its activation before use. In addition, the dye and COD removal efficiencies in the electrochemical process without catalyst were 78% and 58%, respectively; while, in the electrochemical process containing GAC, the removal efficiencies of dye and COD, at 45 min, were 98% and 85.9%, respectively. These results were confirmed by the TOC removal efficiency shown in Fig. 5a. This increase in the 3D system can be due to the synergistic effect of adsorption and 2D electrochemical processes. In other words, increasing the removal efficiency can be related to the occurrence of adsorption mechanisms, direct oxidation (direct transfer of electrons to the anode) and indirect oxidation (electrochemical production of reactive species such as

•OH, H_2O_2, Cl_2, etc.). In indirect oxidation, •OH is produced by electrolysis of water on the anode surface. In addition, most of the researchers have proven that the activated carbon, in addition to adsorbing the contaminants, can act as a catalyst for the decomposition of H_2O_2 into •OH due to having the functional groups and polyaromatic moieties. Activated carbon is also useful in 3D systems as multiple micro electrodes for direct and indirect electrochemical oxidation [37]. Similar results were observed by Shen et al. [38] for the degradation of estriol by a 3D electrochemical reactor. They reported that, by adding the particle electrode and selecting the appropriate current density, the granular particles are converted to cathode and anode micro electrodes, which is led to developing the reaction surface, facilitating the mass transfer, and producing more •OH.

Along with the removal of dye and COD, the energy consumption of different systems was examined and the results are shown in Fig. 5a. It can be seen that the energy consumption in the 3D process is 1.5 times lower than the 2D process. This can be attributed to the further pollutants removal efficiency by the production of more reactive species in the GAC-based 3D system compared to other systems [39]. Similar results were reported by Xiao et al. [40] for the degradation of phenacetin and Pourzamani et al. [13] for the degradation of ciprofloxacin. In this study, the H_2O_2 production rate was also investigated in 2D and 3D processes and the results (Fig. 5b) show that H_2O_2 production in the 3D process is much higher than the 2D process. This increase can be due to the higher production of H_2O_2 by activating molecular oxygen and its electrochemical reduction by GAC [14]. Xu et al. [41] studied the electro catalytic degradation of acid orange 7 using a 3D reactor and found that carbon-based particles have a high ability in the electrochemical reduction of oxygen to H_2O_2. Given these results, the GAC-based 3D process is a promising system due to high removal efficiency, high H_2O_2 production and low power consumption.

In addition to the above findings, the AR18 removal efficiency by the present process was compared with other advanced oxidation processes used in other studies. From the results of Table 4 it can be seen that the GAC-based 3D process have higher efficiency, smaller reaction time and the lower dose required compared to other oxidation processes. Hence, the 3D process-coupled with particle electrodes can act as a good oxidation process for the degradation of the dye.

3.5. Stability of particle electrode

In practical applications, the stability of the particle electrode of the electro-oxidation process is very important for long-term use and cost reduction. Therefore, the stability and re-usability of the GAC particle electrode were evaluated in 10 consecutive cycles at pH of 3, the current density of 20 mA/cm^2, dye concentration of 100 mg/L, catalyst dose of 250 mg/L, and the electrolysis time of 45 min. As shown in Fig. 6, the dye and COD removal efficiency after 6 consecutive cycles have not a significant change, while, by the prolonging the cycles run, the efficiencies were decreased from 98% and 85.9% to 80% and 63.5% on the 10th run. This can be due to adsorption of intermediates produced by oxidation of AR18 on the GAC surface and the effect of oxidants produced on catalyst functional groups [17]. According to these results, GAC, as a particle electrode, has good stability and can be used for long-term degradation without the loss of electro-catalytic activity.

3.6. Electro catalytic treatment of real textile wastewater

In order to investigate the applicability of the activated carbon-based 3D process for the treatment of real wastewater, the experiments of the electro-oxidation degradation were carried out in optimal conditions obtained by Taguchi test. As shown in Fig. 7, the COD removal efficiency in real wastewater is 25.5% less than synthetic wastewater at 45 min. This may be related to the presence of organic

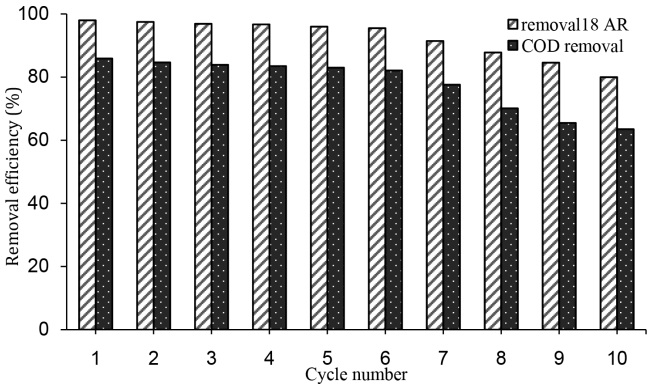


Fig. 6. The stability of the GAC electrode particle for the removal of AR18 and COD.

Table 4
Comparison with other oxidation processes for the removal of AR18

AOPs type	Condition	Acid red dye removal (%)	Reference
Photocatalytic	pH: 7, catalyst: 1500 mg/L, Time: 60 min	85.1	[42]
Fenton	pH: 11, catalyst: 4000 mg/L, C_{AR18} = 0.004 M, Time: 120 min	79.6 for COD removal	[43]
Electrofenton	pH: 3, Voltage: 30 V	99.9	[44]
Sonocatalytic	pH: 6, C_{dye}: 15 mg/L, Time: 20 min, ultrasonic power: 75 W, catalyst: 1000 mg/L	75	[45]
3D electrochemical	pH: 3, C_{dye}: 100 mg/L, Time: 45 min, current denisity: 20 mA/cm^2, catalyst: 250 mg/L	96.5	Present study

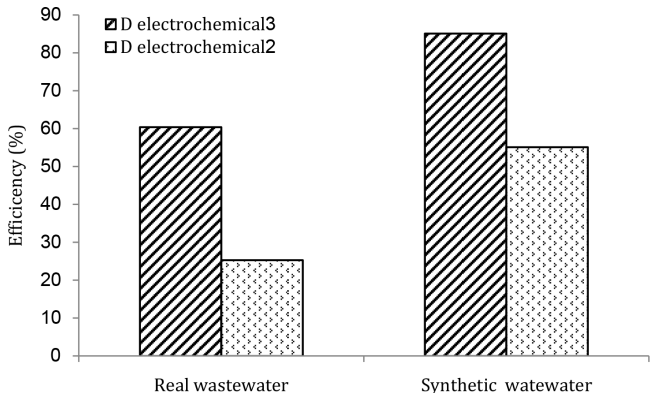


Fig. 7. The COD removal efficiency in real and synthetic wastewater sample.

compounds in the matrix to compete in the reactive species and active sites of particle electrode. The results of Fig. 7 also show that the efficiency of the 3D process is 2.38 times higher than the 2D process. This increase in efficiency can be due to the presence of activated carbon as a particle electrode to produce more reactive species and to absorb pollutants. Similar results were observed by Iranpour et al. [6] and Mengelizadeh et al. [46] for the degradation of reactive dye.

4. Conclusion

The present study explained the use of a 3D electrochemical process based on GAC to remove the AR18 dye and COD from the synthetic solution. Taguchi design was used to investigate the effect of five factors (pH, current density, particle electrode concentration, dye concentration and electrolysis time) on the removal of dye and COD. The results of the analysis performed by Design Expert software showed that the current density is the most important factor in removing the dye and COD. The highest removal efficiency of dye and COD at optimum condition predicted using numerical software was 96.5% and 84.8%, respectively. The results of the effect of the factors showed that the degradation efficiency was increased by increasing the catalyst concentration, current density and electrolysis time, while it was decreased by increasing the initial pH and dye concentration. The 3D electrochemical system had high catalytic activity, high H_2O_2 production and low energy consumption compared to other processes due to the formation of multiple micro electrodes under appropriate current density. 10 consecutive cycle tests performed under the same conditions for the study of electro-catalytic activity showed high stability and re-usability of GAC catalyst. According to these results, the 3D electrochemical reactor can be considered as an effective alternative for the degradation of AR18 dye from aqueous solutions.

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