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Kinetic and thermodynamic investigations on the electrocoagulation of methyl orange from aqueous solution using aluminum electrodes

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The removal of color from solutions containing methyl orange (MO) was experimentally investigated using direct current electrocoagulation (EC) on aluminum electrodes. The experimental results suggested that electrocoagulation was an effective method in MO removal. Metal hydroxides generated during EC were used to remove MO from aqueous solution. The influence of operational parameters such as electrolysis time, applied constant potential, initial dye concentration, temperature and solution conductivity on color removal efficiency was explored. Thermodynamic parameters, including Gibbs free energy, enthalpy, and entropy were calculated for adsorption of MO on metal hydroxides. Thermodynamic parameters indicated that MO adsorption on metal hydroxides was feasible, spontaneous and endothermic in the temperature range from 298 K to 358 K. The adsorption of MO preferably fitted the Langmuir adsorption isotherm suggesting monolayer coverage of adsorbed molecules. In addition, the adsorption kinetic studies showed that the first-order rate equation provided the best correlation for the removal rate of MO.

Keywords: Electrocoagulation; Methyl orange; Aluminum; Langmuir, Adsorption isotherm.

INTRODUCTION

Textile and printing industry are regarded as some of the most polluting sectors due to their high discharge volume of dye-containing wastewater. Among these dyes, azo dyes are some of the most important kinds since they constitute about 50–70% of the world dye production [1,2]. Such kinds of dye effluents are characterized by intense color, high organic content and stable chemical structure due to the existence of azo groups. Moreover, they are toxic and recalcitrant to biodegradation. Therefore, they have caused serious risks for environmental pollution [3,4]. Hence, the removal of dyes during processing of waste effluents becomes environmentally important. Methyl orange (MO) is an azo compound, which is commonly used as a coloring agent in several applications, such as textile, paint, ink, plastic, and cosmetic industries. The release of this coloring agent into wastewater definitely causes a severe environmental problem. Several techniques have been developed to treat wastewaters containing dye pollutants [5-9].

There are many processes to remove dyes from colored effluents such as adsorption, precipitation, chemical degradation, photodegradation, biodegradation, chemical coagulation and electrocoagulation. When chemical coagulation is used to treat dyed wastewater, pollution may be caused by the chemical substances added at a high concentration [10,11]. The adsorption process is one of the effective methods used to remove dyes from aqueous solutions. Activated carbon is the most widely used adsorbent for dye removal, but it is too expensive [12]. Photooxidation by UV/H_2O_2 or UV/TiO_2 and Fenton oxidation needs additional chemicals [13], and therefore cause secondary pollution. Biological methods are generally cheap and simple to apply and are currently used to remove organics and color from dyed and textile wastewater. However, biological methods are less effective because of the toxicity of dyes that has an inhibiting effect on the bacterial development [14]. This dyed wastewater cannot be readily degraded by conventional biological processes, because the structures of most commercial dye compounds are generally very complex and many dyes are non-biodegradable due to their chemical nature, molecular size, which results in sludge bulking [15].

Electrochemical process is more effective for dye removal [16]. Electrocoagulation (EC) as an electrochemical method was developed to overcome the drawbacks of conventional decolorization technologies. EC is an attractive alternative for the treatment of textile dyes [17-18]. EC process provides a simple, reliable and cost-effective method for the treatment of wastewater without any need for additional chemicals, and thus secondary pollution. It also decreases the amount of sludge, which needs to be disposed. EC is a process consisting of creating a floc of metallic hydroxides within the effluent to be cleaned by electrodissolution of soluble anodes. Iron and

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aluminum are generally used as sacrificial anodes. The dye in dye-containing wastewater is coagulated by iron (II) and aluminum hydrates or hydroxides produced from the sacrificial anode [19-20].

For aluminum electrodes, the electrolytic dissolution of the aluminum anode produces Al^{3+}ions, which are initially transformed into $Al(OH)_3$ and finally polymerized to $Al_n(OH)_{3n}$. The main reactions occurring in the anodic compartment are as follows [19,20].

$$Al \rightarrow Al^{3+} + 3e \ , \qquad (1)$$

$$Al^{3+} + H_2O \rightarrow Al(OH)_3 + 3H^+ , \qquad (2)$$

$$nAl(OH)_3 \rightarrow Al_n(OH)_{3n} \ , \qquad (3)$$

The main reaction in the cathodic compartment is:

$$2H_2O + 2e \rightarrow H_2 + OH^- \ , \qquad (4)$$

In recent years, different methods have been developed to treat wastewaters containing methyl orange. Saldan˜a-Robles *et al.* investigated the decolorization of methyl orange using the Fenton oxidation process and adsorption on vegetal carbon. and removal efficiency lower than 85% was obtained. Ling et al. [21] studied repeated oxidative degradation of methyl orange through bio-electro-Fenton in bioelectrochemical system and decolorization efficiency lower than 86% was obtained. In addition, EC has been successfully tested to treat decolorization of methyl orange from waste water [23-25]. Zhang *et al.* [24] investigated the decolorization of methyl orange by electrocoagulation with Fe electrodes and the effect of different operation parameters was studied. Pi *et al.* [25] studied the electrocoagulation of methyl orange from synthetic waste water with periodic reversal of the electrodes technique. Different experimental conditions such as cell voltage, current density, and initial concentration were investigated. Under optimum conditions, a color removal efficiency of 97 % was obtained [25].

The purpose of this investigation was to study the high-efficiency decolorization of methyl orange from aqueous solution in a very short time by an electrocoagulation process. In this method, aluminum electrodes were used as a simple instrument under applied direct constant current and voltage. The effects of constant applied potential, initial MO concentration, and solution temperature on the color removal were investigated. In addition, the adsorption thermodynamics, isotherms, and kinetic characteristics involved in the MO removal from aqueous solution were studied in order to lay a theoretical foundation for the adsorption of MO from aqueous solutions onto aluminum hydroxides.

EXPERIMENTAL

All chemicals were of analytical grade from Merck and were used without further purification. The schematic structure of MO is shown in Fig. 1. Working solutions were prepared by suitable dilution of the stock solutions. All solutions were prepared in doubly distilled water.

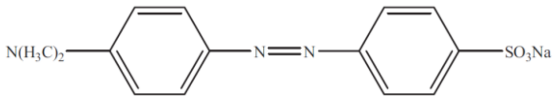

Fig. 1. Chemical structure of methyl orange.

In order to increase the conductivity of the solution, NaCl was added before injecting the solution into the apparatus. The chloride salt added to the solution prevents the formation of an oxide layer on the anode and decreases in this way the passivation problem of the electrodes.

The schematic diagram of the experimental apparatus is shown in Fig 2. The EC unit consists of an electrochemical cell, a D.C. power supply and aluminum electrodes. The aluminum cathode and aluminum anode consist of pieces of aluminum sheet separated by a space of 1 cm dipped in the wastewater. The electrodes were placed in aqueous dye solutions in a cylindrical glass cell. There were two electrodes connected in a monopolar mode in the electrochemical cell, each one with dimensions of 15×5.5×0.2 cm^3.

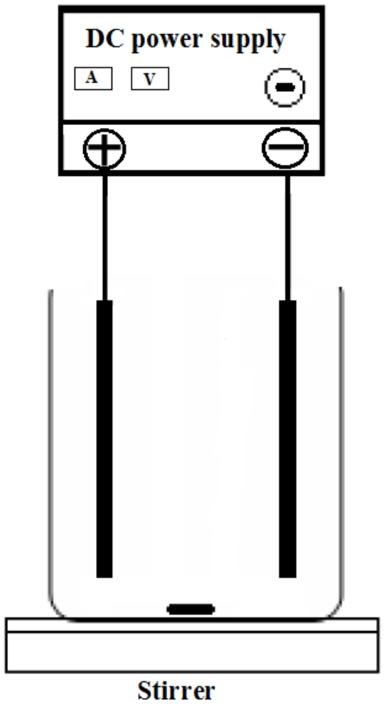

Fig. 2. Schematic diagram of the electrocoagulation equipment.

The stirrer was used in the electrochemical cell to maintain an unchanged composition and avoid the association of the flocs in the solution. The D.C. source was used through a power supply system with 0-24 V and 0-10 A. Electrodes were washed with dilute HCl between the experiments.

The dye concentration was measured using a UV/Vis spectrophotometer model Shimadzu UV1600, Japan at a wave-length corresponding to the maximum absorbance of the dye (λ_{max}=463 nm). The color removal efficiency, CR (%), was calculated using:

$$CR\% = \frac{A_0 - A_i}{A_0} \times 100 \quad (5)$$

where A_0 and A_i are the absorbance values of the dye solution before and after treatment.

RESULTS AND DISCUSSION

Effect of applied potential

It is well-known that electrical potential and current not only determine the coagulant dosage rate but also the bubble production rate and size and the floc growth, [26,27] which can influence the treatment efficiency of the electrocoagulation. Therefore, the effect of applied potential on the MO removal efficiency was investigated. Fig. 3 illustrates the comparative removal efficiency at different applied potentials with an initial MO concentration of 4 mg l^{-1}.

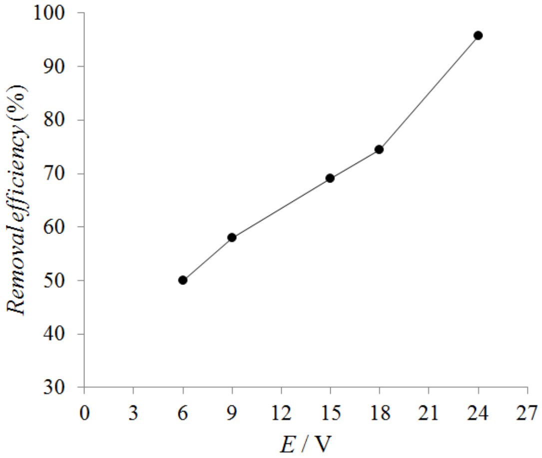

Fig. 3. Effect of applied potential on the removal efficiency of MO(C_0=4 mg l-1, t=30 min; C_{NaCl}=0.5 gl^{-1}).

For a given time, the removal efficiency increases significantly with increasing in current density. This is ascribed to the fact that at higher applied potential, the amount of aluminum oxidized increases, which results in a greater amount of precipitate for the removal of pollutants. In addition, the bubbles density increases and their size decreases with increasing current density [28-30] leads to a greater upwards flux and a faster removal of pollutants and sludge flotation. As the current decreases, the time needed to achieve similar efficiencies increases. However, the cost of the process is determined by the consumption of the sacrificial electrode and the electrical energy which increases in higher applied potentials. The removal efficiency reaches to 96% by applying constant potential 24 V in 30 min.

To observe the effect of initial dye concentration on the dye removal efficiency by EC, the experiments were carried out in different dye concentrations with constant applied potential of 18 V for 30 min. Fig. 4 shows the percentage removal for different initial dye concentrations. As the results indicated, the dye removal efficiency decreases when the dye concentration increases from 7 to 15 mg L^{-1}. It is due to the fact that at constant applied potential, current density and time, the same amount of aluminum hydroxide complexes generates in all the dye solutions. Consequently, the same amount of flocs would be produced in the solutions. As a result, the flocs produced at high dye concentration are insufficient to adsorb all of the dye molecules of the solution. In very low concentration of dye, below 7 mg L^{-1}, access to the MO molecule and adsorption decreases which lead to decreasing removal efficiencies.

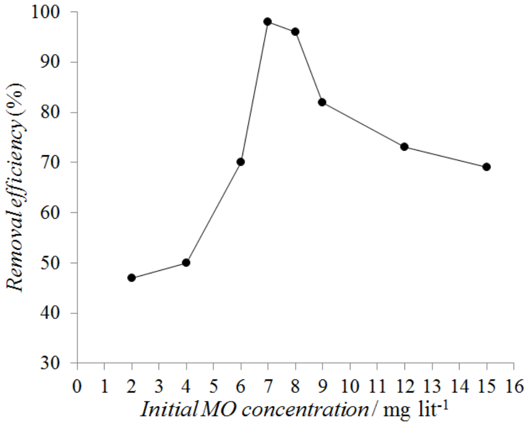

Fig. 4. Effect of initial concentration of MO on the removal efficiency (applied potential= 18 V, t=30 min, C_{NaCl}=0.5 g l^{-1}).

Effect of NaCl concentration

Solution conductivity affects the current efficiency, cell potential and consumption of electrical energy in electrolytic cells. The conductivity of the wastewater was adjusted to the desired levels by adding appropriate amounts of NaCl. The effect of NaCl concentration on the removal efficiency is shown in Fig. 5. Both the conductivity of the solution and the current density increase when the salt concentration in solution increases.

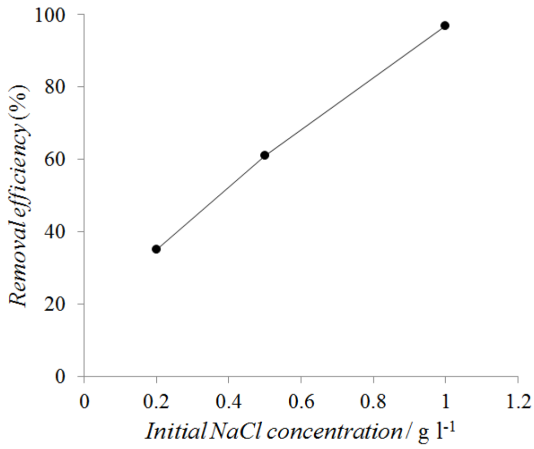

Fig. 5. Effect of NaCl concentration on the removal efficiency of MO (applied potential = 18 V, t=15 min, C_0 =8 mg l^{-1}).

The higher ionic strength generally causes an increase in the current density at the same cell potential or, equivalently, the cell potential decreases with increasing wastewater conductivity at a constant current density. Consequently, the necessary potential to attain a certain current density will be diminished and the consumed electrical energy will be decreased [31-33]. From Fig. 5, it was found that raising the conductivity of dye solutions has a considerable effect on color removal efficiency. At NaCl concentrations higher than 4 g l^{-1}, the removal efficiency approaches a constant value due to the constant electric conductivity of the solution.

Effect of temperature on MO removal

The effect of temperature on the MO removal efficiency obtained at 298, 318, 338, and 358 K is shown in Fig. 6. After 15 min of electrolysis, the MO removal efficiencies increase with increasing temperature. The electrochemical reaction rate increases with increasing solution temperature. The temperature influence can be attributed to an improved destruction of the aluminum oxide film on the anode surface and an increase in the rate of all reactions involved in the process according to the Arrhenius equation [33]. Also, the higher temperature promotes the generation of aluminum hydroxides formed in the EC process, which leads to a higher concentration and thus more frequent collisions of MO and aluminum hydroxides, resulting in an increase in the reaction rate of pollutant adsorption [34].

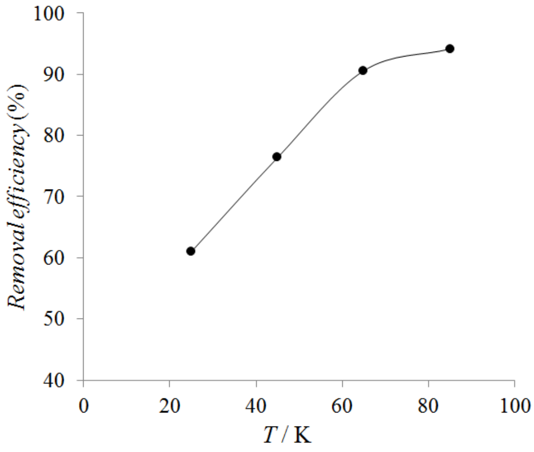

Fig. 6. Effect of temperature on the removal of MO (C_0=8 mg l^{-1}, applied current=0.9 A, C_{NaCl}=0.5 g/L).

Effect of temperature on thermodynamic adsorption parameters

Thermodynamic parameters, including Gibbs free energy ΔG^0, enthalpy ΔH^0, and entropy change ΔS^0, were calculated and used to determine the adsorption behavior of MO. The ΔG^0 was calculated by the following equation:

$$\Delta G^0 = -RT \ln K_d \quad (6)$$

where R is the universal gas constant (8.314 J mol^{-1} K^{-1}), T is the temperature (K), and K_d is the distribution coefficient (l g^{-1}). The K_d value is calculated using the following equation [35]:

$$K_d = \frac{q_e}{C_e}, \quad (7)$$

where q_e and C_e are the equilibrium concentrations of MO for the adsorbent (mg g^{-1}) and the solution (mg l^{-1}), respectively. The relationship between ΔG^0, ΔH^0, and ΔS^0 can be expressed by the following equation:

$$\Delta G^0 = \Delta H^0 - T\Delta S^0, \quad (8)$$

The combination of Eqs. (6) and (8) leads to:

$$\ln K_d = \frac{\Delta S^0}{R} - \frac{\Delta H^0}{RT}, \quad (9)$$

The effect of temperature on the adsorption efficiency of MO from aqueous solution was evaluated at different temperatures ranging from 298 K to 358 K. At each temperature, the corresponding C_e values at different fixed values of q_e were calculated. The thermodynamic parameters of ΔH^0 and ΔS^0 were obtained from the slope and intercept of the $\ln K_d$ *versus* T^{-1} plot. The Gibbs free energies at different temperatures were calculated from Eq. (8). The values of ΔG^0, ΔH^0 and ΔS^0 for MO adsorption onto aluminum hydroxides at different temperatures are given in Table 1. At all temperatures, ΔG^0 is negative which indicates the feasibility of the process and the spontaneous nature of the adsorption of MO on aluminum hydroxides. The increase in the absolute magnitude of ΔG^0 with increasing temperature indicates that these processes are favored at high temperatures. The positive value of ΔH^0 indicates that the adsorption process is endothermic. In addition, the positive value of ΔS^0 suggests that randomness at the solid–solution interface increases during MO adsorption onto aluminum hydroxides. While the adsorption process is endothermic under these conditions, it is spontaneous due to the positive entropy change.

Adsorption isotherms

The pollutant is generally adsorbed on the surface of the metal hydroxides generated during the EC process. Thus the removal of pollutant is similar to conventional adsorption, except for coagulants generated. In order to identify the mechanism of the adsorption process, it is important to establish the most appropriate correlation for the equilibrium curves. Different adsorption isotherms were applied to establish the relationship between the amounts of MO adsorbed onto the aluminum hydroxides and its equilibrium concentration in the aqueous solution. Among them, Langmuir and Freundlich adsorption isotherms show higher correlation coefficients for MO adsorption onto the aluminum hydroxides.

The Langmuir model is originally developed for a set of well-defined localized adsorption sites with the same adsorption energy, independent of the surface coverage, and with no interaction between adsorbed molecules. This model assumes a monolayer deposition on a surface with a finite number of identical sites. It is well known that the Langmuir equation is valid for a homogeneous surface. The mathematical expression for the Langmuir isotherm is [36]:

$$q_e = \frac{\alpha_L K_L C_e}{1+K_L C_e}\ , \qquad (10)$$

where α_L (mg g^{-1}) is a constant related to the area occupied by a monolayer of adsorbate, reflecting the maximum adsorption capacity, C_e (mg l^{-1}) is the equilibrium liquid-phase concentration, K_L (l mg^{-1}) is a direct measure of the intensity of adsorption, and q_e (mg g^{-1}) is the amount adsorbed at equilibrium. K_L and α_L can be determined from the slope and intercept of the straight line in the plot of q_e^{-1} versus Ce^{-1}. Fig. 7 shows the linearized Langmuir absorption isotherm plot with experimental data.

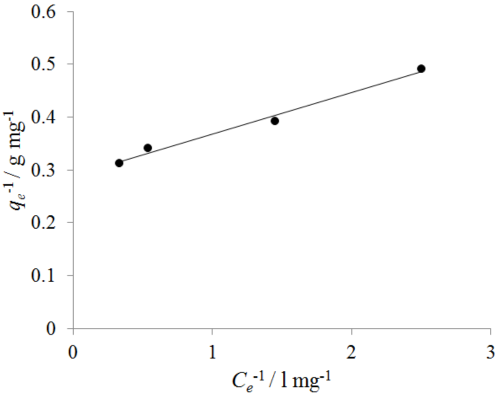

Fig. 7. Linearized Langmuir adsorption isotherms MO (C_0=8 mg l^{-1}, applied current=0.9 A, C_{NaCl}=0.5 g/L).

The Freundlich adsorption isotherm typically fits experimental data over a wide range of concentrations. This empirical model includes considerations of surface heterogeneity and exponential distribution of the active sites and their energies. The isotherm is adopted to describe reversible adsorption and is not restricted to monolayer formation. The mathematical expression for the Freundlich mode is [36]:

$$q_e = K_f C_e^{1/n}\ , \qquad (11)$$

where K_f ((mg g^{-1}) (l mg^{-1})$^{1/n}$) and n (dimensionless) are constants that account for all factors affecting the adsorption process, such as adsorption capacity and intensity.

Table 1. Thermodynamic parameters of MO adsorption onto aluminum hydroxides at different temperatures.

T / K	Thermodynamic equilibrium constant (K_d)	ΔG^0 / kJ mol^{-1}	ΔS^0 / J mol^{-1} K^{-1}	ΔH^0 / kJ mol^{-1}
298	1.075	-0.18	81.8	24.389
318	1.569	-1.19		
338	3.706	-3.681		
358	5.075	-4.834		

Table 2. Langmuir and Freundlich constants for the adsorption of salicylic acid onto aluminum hydroxides.

Langmuir			Freundlich		
K_L /l mg^{-1}	α_L / mg g^{-1}	R^2	K_f / (mg g^{-1}) (l mg^{-1})$^{1/n}$	n	R^2
3.68	3.46	0.98	2.58	4.78	0.93

The Freundlich constants K_f and 1/n are determined from the intercept and slope, respectively, of the linear plot of log q_e vs. log C_e. Fig. 8 shows the linearized Freundlich adsorption isotherm plot with the experimental data.

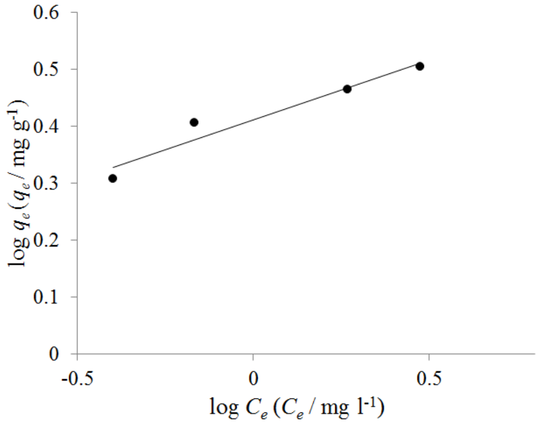

Fig. 8. Linearized Freundlich adsorption isotherms MO (C_0=8 mg l^{-1}, applied current=0.9 A, C_{NaCl}=0.5 g/L).

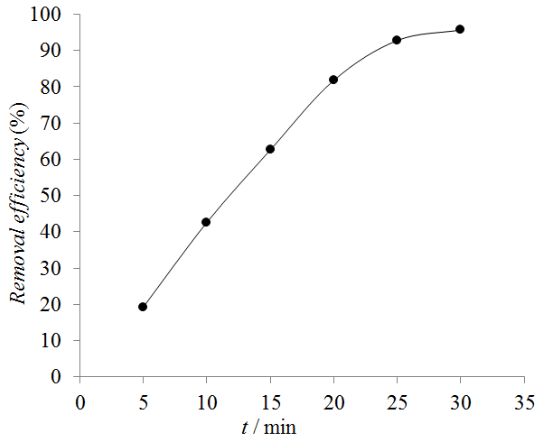

Fig. 9. Effect of electrocoagulation time on the removal efficiency (C_0 =4 mg l^{-1}, applied potential = 24 V, C_{NaCl}=0.5 g l^{-1}).

The applicability of the two isotherm equations is compared using the correlation coefficient R^2. The Langmuir and Freundlich constants for the adsorption of MO onto aluminum hydroxides are presented in Table 2. The value of the correlation coefficient R^2 is found to be 0.98 for Langmuir isotherm and 0.93 for Freundlich isotherm. However, the Langmuir isotherm model provides a better fit compared to Freundlich isotherm model. This can be due to the monolayer adsorption of MO onto aluminum hydroxides and the low interaction between the adsorbed MO molecules on the surface. Methyl orange has functional groups for adsorption which lead to strong interaction with aluminum hydroxides and increase the tendency for monolayer formation.

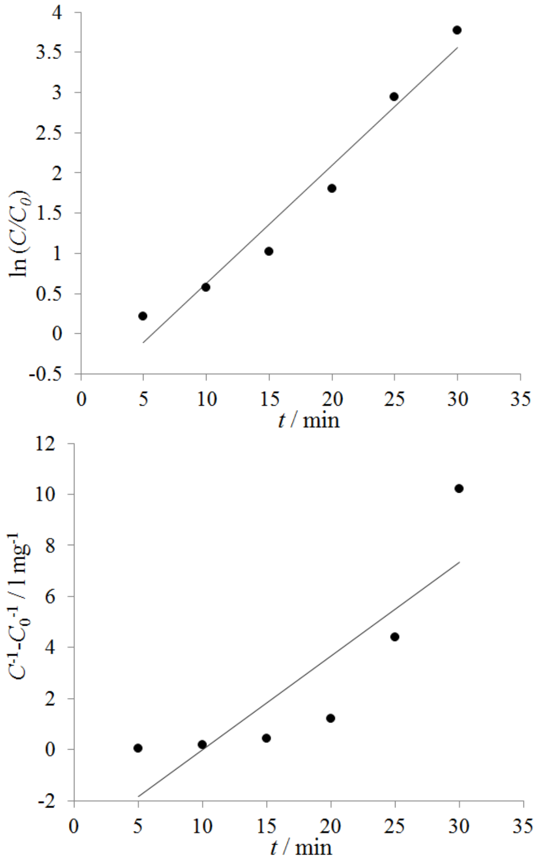

Fig. 10. Plot of (a) first-order and (b) second-order equation for the removal kinetics of MO. (C_0 =4 mg l^{-1}, applied potential = 24 V, C_{NaCl}=0.5 g l^{-1}).

Effect of electrocoagulation time

Fig. 9 shows the effect of electrocoagulation time on the removal efficiency at a constant applied potential. As can be seen, with increasing electrocoagulation time, comparable changes in the removal efficiency of MO are observed. The color removal efficiency directly depends on the concentration of hydroxyl and metal ions produced on the electrodes. The color of the dye solution decreases as a function of elapsed time. After 30 min of electrolysis, the color removal efficiency reaches a maximum value at 24 V applied potential. The rate of removal of MO can be presented by the following first-order reaction kinetics:

$$\frac{dC}{dt} = -k_1 C^m , \qquad (12)$$

where C represents the MO concentration, m the order of reaction, k the reaction rate coefficient and t is the time. For a first-order reaction, the above equation becomes

$$\ln\left(\frac{C}{C_0}\right) = -k_1 t , \qquad (13)$$

where C_0 is the initial MO concentration.

According to the above equation, a plot of ln (C_0 C^{-1}) against t yields a straight line with a slope of k_1.

For a second-order reaction, the above equation becomes:

$$\frac{1}{C}-\frac{1}{C_0}=-k_2t \quad , \qquad (14)$$

The slope of the plot of C^{-1}-C_0^{-1} versus time gives the value of the rate constant k_2.

Fig. 10 reveals the reasonably good fit of first-order kinetic model to the observed data. The conformity between experimental data and the model values was evaluated by the values of the correlation coefficient (R^2). The correlation coefficient R^2 for the first-order model is 0.96 which is relatively higher than that for the second-order model (0.73). The rate coefficient for a first-order removal rate of MO was obtained as 0.1468 min^{-1}.

CONCLUSION

Electrocoagulation was used to remove color from dye solutions containing MO. The effect of various operational parameters on color removal efficiency was investigated. The results indicated that EC was a promising treatment for the removal of MO from aqueous solutions. The experiments showed the feasibility of removing MO by adsorption and co-precipitation with aluminum hydroxides. The removal efficiencies increased in the presence of higher supporting electrolyte concentration. The effect of different applied constant potentials was checked and the removal efficiencies increased with increasing applied potential.

In addition, with increasing temperature, the MO removal efficiencies increased and the thermodynamic parameters were determined in different temperatures. Their values indicated that the adsorption process was favorable and spontaneous. As the temperature increased from 298 K to 358 K, ΔG^0 became further negative, indicating a stronger driving force, resulting in a greater adsorption capacity at higher temperatures. The positive value of ΔH^0 confirmed that the process was endothermic, meaning that the reaction consumed energy. The positive value of ΔS^0 suggested that the higher randomness at the solid–solution interface occurred during the MO adsorption from the aqueous solution onto aluminum hydroxides. The metal hydroxides generated by EC can efficiently remove MO by adsorption, and the EC process was modeled using adsorption isotherm models. The MO adsorption was best fitted by the Langmuir adsorption isotherm, and the results were in good agreement with the experimental data. Decolorization of MO wastewater was described well by a first-order reaction equation and the rate constant was obtained to be 0.1468 min^{-1}.

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КИНЕТИКА И ТЕРМОДИНАМИЧНО ИЗСЛЕДВАНЕ НА ЕЛЕКТРО-КОАГУЛАЦИЯТА НА МЕТИЛОРАНЖ ОТ ВОДЕН РАЗТВОР С АЛУМИНИЕВИ ЕЛЕКТРОДИ

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(Резюме)

Опитно е изследвано отстраняването на оцветяването на разтвори, съдържащи метилоранж (МО) с помощта на правотокова електро-коагулация с алуминиеви електроди. Опитните резултати показват, че това е ефективен метод за отстраняването на МО, което става поради образуването на метални хидроксиди . изследването на влиянието на работните параметри, като времето за електролизата, прилаганият потенциал, началната концентрация на багрилото, температурата и проводимостта на разтвора върху ефективността на обезцветяването. Термодинамичните параметри, включително свободната енергия, енталпията и ентропията са изчислени за адсорбцията на МО върху металните хидроксиди и показват, че тя е спонтанна и ендотермична за температури в интервала от 298 до 358 K. Адсорбцията се описва добре с изотермата на Лангмюир, което предполага мономолекулен слой на покритието. Освен това, кинетиката на адсорбция на МО е на реакция от първи порядък.