


Influence of alternating current on the adsorption of indigo carmine

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Abstract The main purpose of this work is to study the effect of a new process of accelerating which consist to couple the electrochemical process with the adsorption to remove an anionic dye, the indigo carmine. That is why, we investigated the effects of the new process of accelerating the adsorption process by using alternating current (AC) on the retention of an anionic dye, the indigo carmine. The adsorption capacity of dye (mg/g) was raised with the raise of current voltage in solution, temperature, and initial indigo carmine concentration and decreased with the increase of initial solution pH, current density, and mass of carbon. The results demonstrate that the removal efficiency of 97.0 % with the current voltage of 15 V is achieved at a current density of 0.014 A/cm², of pH 2 using zinc as electrodes and contact time of 210 min for adsorption in the presence of AC. Concerning the adsorption without AC, the results obtained showed that for an initial concentration equal to 20 mg/L, more than 95 % amount of adsorbed dye was retained after 405 min of contact in batch system. The comparison between adsorption in the presence and absence of an alternating current shows the importance of the alternating current in the acceleration of the adsorption method and

improve the performances of FILTRASORB 200. For both cases, the adsorption mechanism follows the fractal kinetics BSf(n,α) model and the Brouers–Sotolongo isotherm model provides a good fit of the experimental data for both adsorption with and without alternating current.

Keywords Adsorption · Alternating current · Activated carbon · Anionic dye · Indigo carmine · Modeling

Introduction

Colorants are largely employed in textile, paper, plastic, food, and cosmetic industries (Sumanjit et al. 2013; Ong et al. 2007; Vargas et al. 2011). Several dyes and their break-down products contribute to water toxicity and constitute a menace for the environment, human, and animals (García et al. 2014). In fact, dyes are recalcitrant and hardly biodegradable and are not taken off from wastewater by classic wastewater treatment method (Bello et al. 2013) due to their complex structure and synthetic origins (Khodaie et al. 2013; Wang et al. 2008).

Indigo carmine (5,5'-indigodisulfonic acid sodium salt or indigotine) is a dark blue dye used primarily in textile industries for the dyeing of polyester fibers and denim (blue jeans) (Stergiopoulos et al. 2014). In addition, indigo carmine is employed as dye in food and cosmetics industry (Stergiopoulos et al. 2014). The indigo carmine is classified as a very toxic dye (Mittal et al. 2006). The contact of this dye with the human body can provoke irritations of the skin and eye, permanent damage with the cornea and the conjunctive one, gastro-intestinal irritation with nausea, vomiting, and diarrhea (Maghri et al. 2012).

The treatment of colored water was the subject of several studies (Rodrigues et al. 2013; Căilean et al. 2009; Vijayageetha et al. 2013; Ncibi et al. 2008a, b; Abidin and

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Rahmat 2010) whose purpose is to reduce the intensity of the color and the organic matter contained in these waters. Different processes have been proposed for the elimination of dyes and in particular the indigo carmine from wastewater such as adsorption (Ramesh and Sreenivasa 2015; Aliabadi et al. 2006; Babel and Opiso 2007; Forgacs et al. 2004), photochemical (Kirupavasam and AllenGnana Raj 2012), electrochemical (Stergiopoulos et al. 2014; El-Ashtoukhy 2013; Hammami et al. 2012), electrocoagulation (Deo Mall et al. 2013), ultrafiltration (Căilean et al. 2009), and biological methods (Karthik et al. 2014; Abidin and Rahmat 2010).

However, several of these technologies are very expensive (Mckay et al. 1987; Low et al. 1995). The conventional biological process is not very efficient because most dyes are nonbiodegradable (Stergiopoulos et al. 2014). Physical process of dyes treatment are nondestructive. It transfer dye from water into the adsorbent which generates large amount of residue (Ramesh and Sreenivasa 2015). For these reasons, researchers are encouraged to search for new technologies (Kesraoui et al. 2015; Ncibi et al. 2006, 2007; Zoughuir et al. 1998; Elkassimi et al. 1998). In this context, the alternating current appears as a new alternative, original, and very interesting in accelerating the phenomenon of adsorption and the removal of heavy metals (Shul'gin et al. 1975).

The main purpose of this work is to study the effect of a new process of accelerating by coupling of electrochemical process with adsorption to remove dyes. In order to reach this objective, the adsorption of indigo carmine (Fig.1) by activated carbon (FILTRASORB 200 which is more used in textile industries in Tunisia) in the absence and presence of alternating current (AC) was investigated. This anionic dye is considered in this paper as a model representative of the pollution of textile wastewater which is the most important source of pollution in Tunisia. The influence of several parameters such as pH, contact time, initial IC concentration, temperature, and voltage on the adsorption capacity was studied. Furthermore, mathematical modeling of the kinetics and adsorption isotherms was investigated.

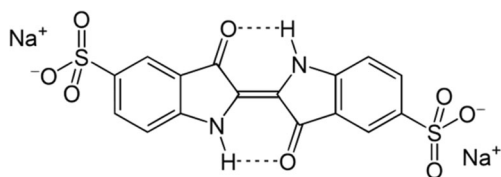


Fig. 1 Chemical structure of indigo carmine

Materials and methods

Materials

The FILTRASOB 200 is a commercial and granulated activated carbon, it was obtained from a textile industry (Chimitex, Tunisia). The origin of this adsorbent is bituminous whose characteristics are shown in Table 1.

Indigo carmine (or indigotin) is a blue dye (number E132) natural extract of the indigo plant. It is part of the family of indigoids. It was purchased from Sigma (purity >85 %). Stock solutions were prepared by dissolving the same quantity of IC in distilled water to give a concentration of 1 g/L. All studied solutions were prepared by diluting the stock solution with distilled water to reach the required concentrations.

Methods

The adsorption experiments have been performed in batch reactor by adding 0.5 g of activated carbon in 150 mL of IC solution with the desired concentration (20–100 mg/L), pH (1–4), and temperature (25–80 °C). All the experiments were carried out at 25 °C, except the tests made to study the effect of temperature on adsorption. The electrical part of the mounting comprises a current source which delivers an AC voltage of 2 to 15 V. It is connected to a voltmeter that allows us to read the voltage. In a 150-mL capacity cell, we plunged two zinc electrodes and the distance between two Zn electrodes has been 4 cm. The current supplied between the electrodes is controlled by an ammeter. Identical mounting to the previous but without the electric part has been used for the blank test. The initial pH was adjusted by adding dilute solutions of HCl (1 M) or NaOH (1 M). After adsorption, the residual concentrations of indigo carmine were determined by a double beam spectrophotometer (FT-IR) (Camspec M550) at a wavelength λ_{max} of 608 nm. The estimation of indigo carmine elimination, the adsorption capacity at equilibrium time (Q) or adsorption elimination efficiency will be calculated, respectively, in accordance with the Eqs. 1 and 2:

$$\% \text{indigo carmine removal} = \frac{(C_0 - C_i) \times 100}{C_0} \tag{1}$$

$$Q = \frac{(C_0 - C_i) \times V}{M} \tag{2}$$

where

- C_0 is the initial indigo carmine concentration (mg/L)
- C_i is the residual indigo carmine concentration at any time (mg/L)
- V is the volume of solution (L) and M is the mass of the sorbent (g)

At equilibrium, C_i is equal to C_e and Q is equal to Q_e .

Table 1 Commercial carbon characteristics

Filtrasob 200	Iodine index (mg/g)	Humidity (%)	Granulometry (mm)	Apparent density (g/cm ³)	BET surface (m ² /g)	pHzpc
Granulated	850	2	0.55–0.75	0.86	825	7.20

Results

Influence of the alternating current voltage

To study the effect of the AC voltage on the adsorption of indigo carmine by activated carbon, one series of experiments was performed on indigo carmine solutions with the concentration of 20 mg/L at pH 2. Figure 2 shows the percentage removal of the dye by activated carbon. When the AC voltage is augmented from 5 to 15 V, the elimination of indigo carmine by adsorption increases.

A remarkable increase in removal of indigo carmine by activated carbon was found by increasing AC voltage. This can be explained by the increases of the ions mobility of this solution especially the anionic dye which move to the surface of the activated carbon protonated by H⁺ ions in acidic medium. Hence, there is an increase in contact between the adsorbate and the adsorbent. As a consequence, there is an increase in the amount of adsorbed ions.

Influence of contact time and initial indigo carmine concentration

The influence of contact time at different initial concentrations on the adsorption of indigo carmine onto activated carbon is presented in Fig. 3. As shown, the quantity of adsorbed indigo carmine increases with time and it is left constant after an

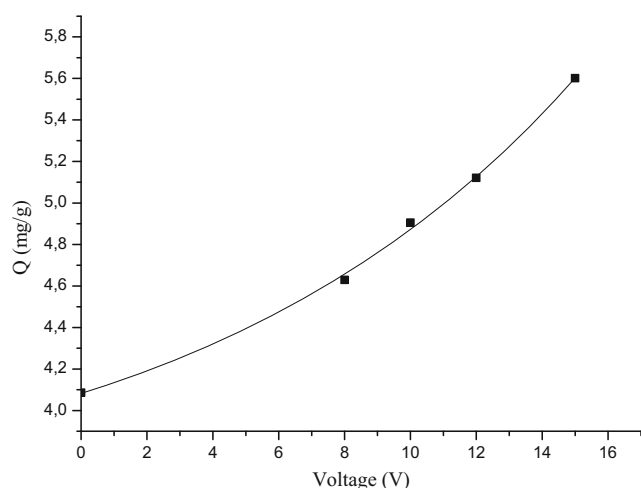


Fig. 2 Effect of AC voltage on the adsorption capacity. Initial concentration: 20 mg/L, temperature: 25 °C, current density: 0.017 A/cm², pH: 2, contact time: 2 h, mass of activated carbon: 0.5 g, volume: 150 mL

equilibrium contact time of about 2 and 4 h, respectively, with AC and without AC for the lower concentrations (i.e., 20 mg/L) and almost 10 and 30 h, respectively, with AC and without AC for the higher concentrations (100 mg/L). Moreover, raising the indigo carmine concentration from 10 to 100 mg/L allows the activated carbon to increase their adsorption capacities from 5.86 to 31.05 mg/g with AC and from 5.85 to 30.94 mg/g without AC, respectively, at pH 2 and a constant temperature of 25 °C. Furthermore, the adsorption capacity is similar for adsorption in the presence and absence of AC. On the other hand, in the absence of AC, the adsorption is very slow and reaches equilibrium after 30 h for concentration of 100 mg/L with adsorption capacities of 30.95 mg/g. Whereas in the presence of AC, the adsorption is very speed and equilibrium is reached after only 10 h for the same concentration with adsorption capacity of 31.5 mg/L.

In each case, it was noted that adsorption of indigo carmine was rapid during the first hours and then it became slower to finally attained saturation. The higher adsorption rate during the initial period may be due to the high number of sites availables for the sorption, and no less than 96 % of total dye was removed. After this phase, the number of sites accessible for adsorption decreased such that the colorant molecules needed longer periods for to access the most inaccessible sites (Kesraoui et al. 2015; Ben Hamissa et al. 2010).

When comparing between adsorption in the presence and absence of the AC, only the speed of absorption has changed, while the adsorption capacity is almost the same for both. This is probably due to the existence of the electric field which has

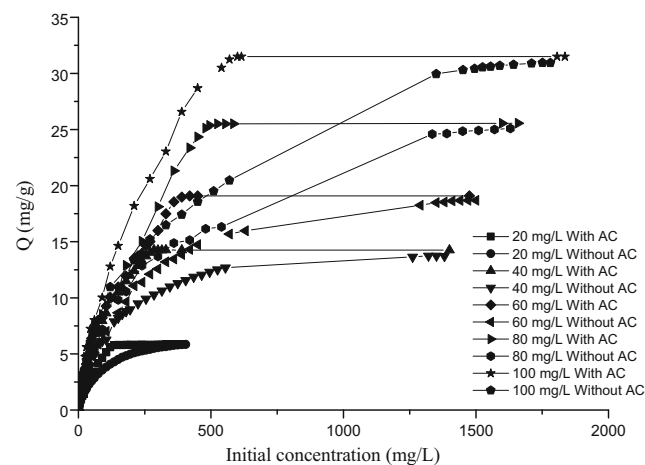


Fig. 3 Effect of initial concentration on the adsorption capacity. Temperature: 25 °C, current density: 0.017 A/cm², pH: 2, contact time: 2 h, mass of activated carbon: 0.5 g, volume: 150 mL

increased the speed of movement of the molecules of the indigo carmine.

Influence of initial pH

The initial solution pH constitutes a significant parameter in any adsorption study. The influence of pH on indigo carmine adsorption by activated carbon was investigated by tests performed on solutions of different pH (from 2 to 10) as shown in Fig. 4. The dye adsorption in the presence and absence of AC raised as the pH of the solution reduced over the pH range 2–10. The results show that the equilibrium adsorption capacity is maximum at pH 2 for both methods (5.60 mg/g with AC and 4.09 mg/g without AC).

Acidity is a very important parameter in the adsorption reaction. The pH of solution will control the amplitude of the electrostatic charges conferred by the ionized dye molecules (Ould Brahim et al. 2014; Rajurkar and Desa 2015). The indigo carmine and the activated carbon can have functional groups could be protonated or deprotonated to form various surface charges at different pH, which leads to electrostatic attraction or repulsion between the charged adsorption and adsorbents (Sivaramakrishna et al. 2014; Ben Douissa et al. 2014). In case of this study, the attraction of the positively charged surface of the AC at acidic pH on the negative sites of this anionic dye (Ben Hamissa et al. 2013) is independent of alternating current.

Influence of salt addition

The study of salt addition is intended, on the one hand, to simulate the industrial dye baths, on the other hand, to investigate the effect of ionic strength on adsorption phenomena. In fact, textile industry often added salts to improve the fixation

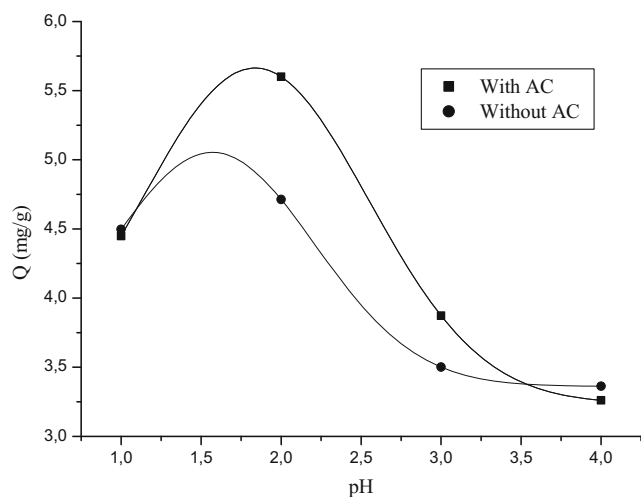


Fig. 4 Effect of pH on the adsorption capacity. Temperature: 25 °C, current density: 0.017 A/cm², initial concentration: 20 mg/L, contact time: 2 h, mass of activated carbon: 0.5 g, volume: 150 mL

of the dye on the tissue. As shown in Fig. 5, the adsorbed amount of the indigo carmine decreased when adding 0.5 g NaCl for adsorption in the presence and absence of AC.

In theory, when the electrostatic forces between the surface of the adsorbent and the adsorbate ions are favorable, a raise in ionic strength decreases the adsorption capacity. Inversely, when the electrostatic forces are repulsive, a raise in the ionic strength increases the adsorption capacity (Newcombe and Drikas 1997; Alberghina et al. 2000). The similar results were obtained with our both processes. The addition of 0.5 g NaCl at pH 2 allowed a significant decrease in the amount adsorbed of 28.5 and 11 %, respectively, in the presence and absence of AC. Sodium ions Na⁺ highly mobile appear discharging anionic dyes and decrease their attachment to the positive surface sites of activated carbon (Krika et al. 2012).

Influence of temperature

The indigo carmine elimination capacity by the activated carbon was also investigated at various temperatures (298, 313, and 353 K). The study of the effect of temperature on the adsorption of the indigo carmine by activated carbon for initial concentration of 20 mg/L and a pH 2 is shown in Fig. 6. A raise of temperature is followed by a raise of adsorption. Therefore, the viscosity of the solution decreased and the mobility of the adsorbate increased.

A small raise of adsorption capacity of the indigo carmine by activated carbon was observed with increasing temperature. This result indicates that the adsorption was endothermic (Ncibi et al. 2008b; Bilgin Simsek et al. 2015). This behavior may be due to the increase of adsorbate ions mobility with temperature which lightly improves the elimination of indigo carmine from the solution by activated carbon (Al-Khatib et al. 2012). Similar performances were got in other studies

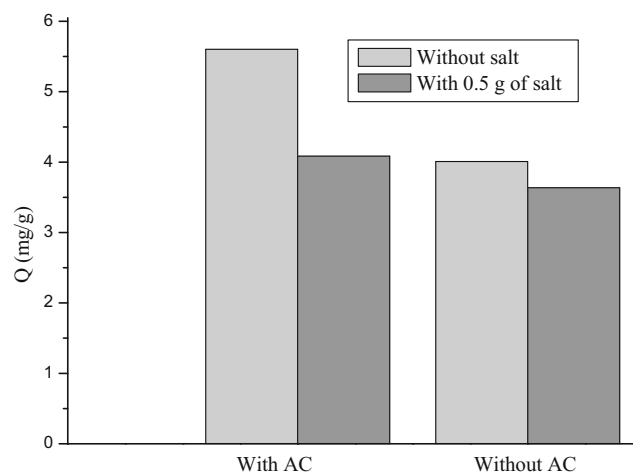


Fig. 5 Effect of salt addition on the adsorption capacity. pH: 2, temperature: 25 °C, current density: 0.017 A/cm², initial concentration: 20 mg/L, contact time: 2 h, mass of activated carbon: 0.5 g, volume: 150 mL

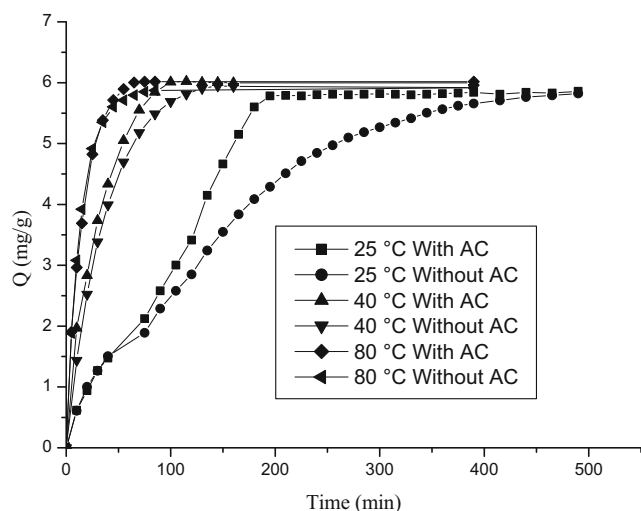


Fig. 6 Effect of temperature on the adsorption capacity. pH: 2, current density: 0.017 A/cm², initial concentration: 20 mg/L, contact time: 2 h, mass of activated carbon: 0.5 g, volume: 150 mL

for the adsorption of Congo red dye onto date palm leaf base (Ghadah 2014) and adsorption of remazol brilliant blue by seed activated carbon (Ahmad et al. 2015).

Thermodynamic analysis

Thermodynamic parameters such as free energy change (ΔG°), enthalpy change (ΔH°), and entropy change (ΔS°) for the adsorption of indigo carmine by activated carbon were calculated according to the Eqs. 3 and 4 (Ucun et al. 2008):

$$\Delta G^\circ = -RT \ln K_C \tag{3}$$

$$\Delta G^\circ = \Delta H^\circ - T \Delta S^\circ \tag{4}$$

where

- R is the gas constant (8.314 J mol⁻¹ K⁻¹)
- T is the absolute temperature (K)
- K_C is the apparent equilibrium constant defined according to the Eq. 5 (Yurtsever and Sengil 2009):

$$K_C = \frac{C_{ad,eq}}{C_{r,eq}} \tag{5}$$

where

- $C_{ad,eq}$ is the concentration of adsorbed dye at equilibrium (mg/L)
- $C_{r,eq}$ is the remaining concentration dye at equilibrium (mg/L).

The thermodynamic was studied at 298, 313, and 353 K. The thermodynamic parameters like Gibbs free energy ΔG° (kJ mol⁻¹), enthalpy ΔH° (kJ mol⁻¹), and entropy ΔS° (J mol⁻¹ K⁻¹) were shown in Table 2.

The negative value of ΔG° shows the spontaneous nature of adsorption of indigo carmine onto activated carbon for all investigated temperatures (Acosta et al. 2016; Aljeboree et al. 2014; Bouhamed et al. 2012). ΔG° is varying from -8.63 to -11.21 kJ mol⁻¹, and from -7.85 to -11.43 kJ mol⁻¹ in the presence and absence of AC, respectively. Furthermore, these values being within the range -20 to 0 kJ mol⁻¹ (Ben Hamissa et al. 2010), this process can be considered as physisorption. ΔH° has a positive value which confirms the endothermic character of the adsorption process (Ben Hamissa et al. 2013; Bouhamed et al. 2012). The positive value of ΔS° suggests the increasing randomness at solid/liquid interface in the course of the adsorption process taking place in the internal structure of indigo carmine removal onto activated carbon (Bouhamed et al. 2015; Ben Hamissa et al. 2013; Bouhamed et al. 2012). Similar findings have been found by other investigators for the adsorption of dyes on various adsorbents (Travlou et al. 2013; Sun et al. 2013; Salleh et al. 2011; Matheswaran and Karunanithi 2007). These results indicate also that the alternating current has no significant influence on the thermodynamic of the adsorption, the values of the free energy are maintained essentially the same.

Kinetic data analysis

To forecast the mechanism of the indigo carmine adsorption, two kinetic models were analyzed: the pseudo-first-order and the pseudo-second-order models.

The model of pseudo-first-order and the pseudo-second-order equation (Saleh 2015; Ho 2004; Lagergren 1898; Ho and McKay 1998) were employed to fit the experimental data of the colorant adsorption

Table 2 Thermodynamic parameters for the adsorption of indigo carmine onto activated carbon at different temperatures

C ₀ (mg/L)	ΔH° (kJ mol ⁻¹)	ΔS° (J mol ⁻¹ K ⁻¹)	Temperature (°K)		
			298	313	353
			ΔG° (kJ mol ⁻¹)		
With AC	17.475	81.630	-6.195	-8.449	-11.218
Without AC	17.551	82.040	-6.560	-8.022	-11.448

before to attain equilibrium. The linearized form of this model is determined by Eq. 6:

$$\log(Q_e - Q) = \log(Q_e) - \frac{k_1 \cdot t}{2.303} \tag{6}$$

where

- t is the time (min)
- k_1 is the equilibrium rate constant of pseudo-first-order sorption (min^{-1}).

This equation can be also written according to the Eq. 7:

$$Q = Q_e(1 - \text{Exp}(-k_1 \cdot t)) \tag{7}$$

and define $h_1 = k_1 Q_e$ as the initial adsorption rate expressed in ($\text{mg g}^{-1} \text{min}^{-1}$) is the initial adsorption rate (Lagergren 1898).

Contrary to the pseudo-first-order model, the pseudo-second-order model is used to a larger interval of time (Ncibi et al. 2006; Ho and McKay 1999). The pseudo-

second-order is founded on the adsorption capacity of the solid phase is described by the Eq. 8:

$$\frac{t}{Q} = \frac{1}{k_2 \cdot Q_e^2} + \frac{t}{Q_e} \tag{8}$$

where

- k_2 is the equilibrium rate constant of pseudo-second-order adsorption (g/mg min)
- $h_2 = k_2 \cdot Q_e^2$ as the initial adsorption rate expressed in ($\text{mg g}^{-1} \text{min}^{-1}$).

The kinetics of indigo carmine adsorption was studied before reaching equilibrium by both models previously described. The modeling results expressed by the rate constants, the calculated equilibrium adsorption capacities, and the linear regression coefficients obtained at all concentrations were presented in Table 3.

The correlation coefficients for the first-order kinetic model obtained at all studied initial concentration and without AC were low. While the second-order model gives satisfactory fit with the experimental data related to the adsorption of indigo carmine onto activated carbon with high R^2 (0.992). These

Table 3 Constants of adsorption kinetics of indigo carmine into activated carbon in the absence and presence of AC

Kinetic models	Initial indigo carmine concentration (mg/L)				
	20	40	60	80	100
Without AC					
Pseudo-first-order					
Calculated Q_{e1} (mg g^{-1})	5.633	12.930	18.147	24.992	31.150
k_1 (min^{-1})	0.011	0.006	0.004	0.003	0.002
R^2	0.990	0.975	0.974	0.988	0.984
h_1	0.0635	0.083	0.077	0.076	0.077
Pseudo-second-order					
Calculated Q_{e2} (mg g^{-1})	7.089	14.968	20.935	29.905	37.866
k_2 ($\text{mg g}^{-1} \text{min}^{-1}$)	0.002	0.0006	0.0002	0.0001	0.00007
R^2	0.998	0.993	0.992	0.995	0.991
h_2 ($\text{mg g}^{-1} \text{min}^{-1}$)	0.084	0.125	0.109	0.098	0.101
With AC					
Pseudo-first-order					
Calculated Q_{e1} (mg g^{-1})	5.898	13.678	17.971	25.310	31.976
k_1 (min^{-1})	0.015	0.012	0.007	0.004	0.004
R^2	0.998	0.970	0.982	0.981	0.990
h_1	0.091	0.144	0.125	0.115	0.138
Pseudo-second-order					
Calculated Q_{e2} (mg g^{-1})	7.038	15.665	21.243	28.424	36.89
k_2 ($\text{mg g}^{-1} \text{min}^{-1}$)	0.002	0.0008	0.0004	0.0002	0.0001
R^2	0.992	0.977	0.988	0.988	0.977
h_2 ($\text{mg g}^{-1} \text{min}^{-1}$)	0.113	0.207	0.171	0.161	0.1905

Table 4 Kinetic parameters obtained by fitting the experimental data with BSf(n,α) equation

Kinetic models	Initial indigo carmine concentration (mg/L)				
	20	40	60	80	100
Without AC					
BSf: <i>R</i> ²					
<i>n</i> = 1	<i>0.99967</i>	<i>0.99822</i>	<i>0.99622</i>	<i>0.99495</i>	<i>0.9876</i>
<i>n</i> = 1.5	0.99925	0.99706	0.98982	0.98554	0.97
<i>n</i> = 2	0.99894	0.99602	0.98114	0.97503	0.95938
With AC					
BSf: <i>R</i> ²					
<i>n</i> = 1	<i>0.99797</i>	<i>0.98728</i>	<i>0.99612</i>	<i>0.99157</i>	<i>0.99092</i>
<i>n</i> = 1.5	0.99634	0.98121	0.99429	0.98983	0.98558
<i>n</i> = 2	0.99501	0.97426	0.99272	0.98870	0.98163

The highest *R*² values are in italics

results demonstrate that the pseudo-second-order model is able to describe satisfactorily the kinetic behavior of indigo carmine adsorption by activated carbon and without AC. Similar phenomenon processes have been found in the adsorption of direct dyes on activated carbon prepared from date pits-A (Mahmoudi et al. 2015), sawdust (Malik 2004), and adsorption of Congo red dye on activated carbon from coir pith (Namasivayam and Kavitha 2002).

As regards the indigo carmine adsorption onto activated carbon in the presence of AC, the order of kinetic model is poorly defined. In fact, the first-order kinetic model was obtained at 20 and 100 mg/L, while at 40, 60, and 80 mg/L, the second-order model was better. In order to improve the kinetic study and determine accurately the order of kinetic adsorption of indigo carmine onto activated carbon, the Brouers–Sotolongo fractal (BSf) kinetics modeling has been used.

On the other hand, the initial adsorption rate (*h*₁ and *h*₂) and the rate constant (*k*₁ and *k*₂) were found to increase using AC. This result indicates that the rates of indigo carmine

adsorption onto activated carbon are much faster when using AC.

BSf(n,α) kinetics modeling

Brouers–Sotolongo model generalized fractional kinetic equation was designed to obtain a universal function for the kinetics of complex systems characterized by stretched exponential and/or power law behaviors. The Brouers–Sotolongo model unifies and generalizes previous theoretical attempts to describe the “fractal kinetic.” Many details on the mathematical development of this model and its application are presented in Brouers and Sotolongo (Brouers 2014a, b; Brouers and Al-Musawi 2015). The pseudo BSf (n,α) sorption kinetics equation is given by Eq. 9:

$$Q_t = Q_e \left[1 - \left(1 + (n-1) \left(\frac{t}{\tau_c} \right)^\alpha \right)^{\frac{-1}{n-1}} \right] \tag{9}$$

where

- α as the fractal time exponent
- n* is an effective non-integer reaction order
- τ_c a characteristic time
- Q*_e is the sorbed quantity at saturation
- Q*_t is the sorbed quantity at any time

Equation 9 is a solution of a fractal differential equation (Brouers and Al-Musawi 2015; Brouers and Sotolongo-Costa 2006) and the half sorption time (the time at which half of the sorbed material has been adsorbed is a function of the three quantities *a*, *n*, and τ_c (Eq. 10):

$$\tau_{50\%} = \tau_c \left(\frac{2^{(n-1)} - (n-1)}{n-1} \right)^{1/a} \tag{10}$$

Kinetic parameters were calculated from fits of BSf(n, α) model and they are showed in Tables 4 and 5. The best-fit

Table 5 Kinetic parameters obtained by fitting the experimental data with BSf(1,α) equation

Kinetic models	Initial indigo carmine concentration (mg/L)				
	20	40	60	80	100
Without AC					
BSf					
τ _c	114.995	186.275	267.309	347.973	389.440
α	0.7582	0.70241	0.7329	0.8324	0.8588
<i>R</i> ²	0.9997	0.9982	0.9962	0.9949	0.9876
With AC					
BSf					
τ _c	64.29842	114.382	171.401	243.779	235.430
α	1.02798	0.76678	0.80459	0.79946	0.95681
<i>R</i> ²	0.99797	0.98728	0.99612	0.99157	0.99092

Table 6 Mathematical equations of the used isotherm models

Isotherm model	Equation
Brouers-Sotolongo	$Q_e = Q_{max} (1 - \exp(-k_w * Ce^\alpha))$ (Brouers et al. 2014; Brouers et al. 2005)
Langmuir	$Q_e = Q_{max} * k_L * \frac{Ce}{1 + k_L * Ce}$ (Langmuir 1916)
Freundlich	$Q_e = k_F * Ce^{1/n}$ (Freundlich 1906; Yang et al. 2016)
Temkin	$Q_e = \frac{RT}{bt} \ln(kt * Ce)$ (Temkin 1941)

model was selected according to the nonlinear regression correlation coefficient (R^2) and listed in Table 5.

Based on the R^2 correlation coefficients provided by pseudo-first-order, pseudo-second-order (Table 4), and BSf model (Table 5), it is clear that the BSf model is the best-fitting kinetic model for all studied initial indigo carmine concentrations and for both processes.

We have verified that for this type of well-behaved experimental data the other regression parameters (covariance, adjusted Rsquare ...) of the nonlinear fitting program of “mathematica 10” software follow the same order.

Moreover, it appears that with the data reported in Table 4, the largest values of the recursion parameter R^2 are obtained with the BSf(1, α) function for both processes. A similar phenomenon has been found by other researchers (Ben Hamissa et al. 2013; Ncibi et al. 2008a).

However, it would be easy to compare τ_c values calculated from BSf(1, α) in the presence and absence of AC. As a matter of fact, for 20 mg/L in the presence of AC, the τ_c (64.30) is

Table 7 Isotherm parameters of Brouers-Sotolongo, Langmuir, Freundlich, and Temkin models referred to the adsorption of indigo carmine in absence of AC and in presence of AC

Isotherm model	Parameters			
With AC				
Brouers-Sotolongo	Q_{max} (mg/g)	k_w (L/mg)	α	R^2
	76.39	0.24	1.03	<i>0.994</i>
Langmuir	Q_{max} (mg/g)	k_L (L/mg)		R^2
	87.82	0.28		0.971
Freundlich	k_F (mg/g)	n		R^2
	24.46	3.05		0.795
Temkin	b_t (KJ/mol)	k_t (L/mg)		R^2
	0.140	2.96		0.930
Without AC				
Brouers-Sotolongo	Q_{max} (mg/g)	k_w (L/mg)	α	R^2
	75.29	0.23	0.98	<i>0.980</i>
Langmuir	Q_{max} (mg/g)	k_L (L/mg)		R^2
	85.57	0.26		0.961
Freundlich	k_F (mg/g)	n		R^2
	23.41	3.15		0.781
Temkin	b_t (KJ/mol)	k_t (L/mg)		R^2
	0.145	2.70		0.916

The highest R^2 values are in italics

clearly shorter than the τ_c in absence of AC (114.99). A similar result has been found for all concentration. Hence, the results from BSf(n, α) showed clearly that the AC allows an increase of the speed of the adsorption reaction.

Adsorption isotherm modeling

In order to optimize dyes adsorption process, it is useful to find the best correlation for the equilibrium curves.

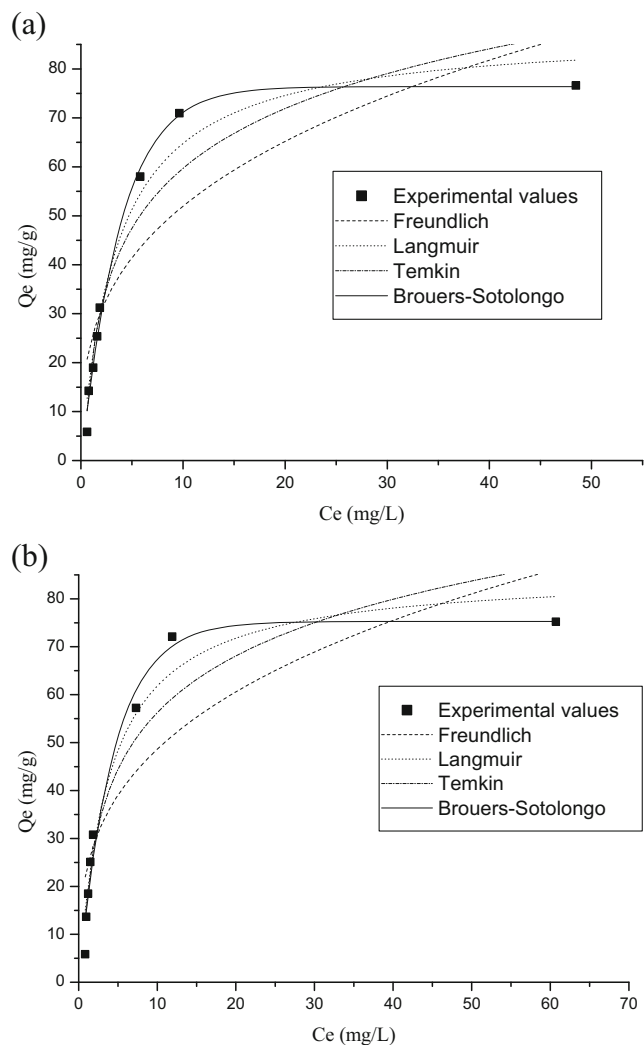


Fig. 7 Isotherm modeling of indigo carmine into activated carbon. **a** In the absence of AC. **b** In the presence of AC

Table 8 Comparison of adsorption capacities between coupling of electrochemical process with adsorption and other adsorption study using activated carbon for IC

Sorbents	Precursor	Q_m (mg/g)	References
Activated carbon	–	76	This study
	–	57.32	Secula et al. 2011
	KOH	13.45	Odogu et al. 2016
	<i>Citrus reticulata</i> peels	71.07	Rehman et al. 2014
	Bottom ash	78.81	Mittal et al. 2006
	De-oiled soya	174.88	Mittal et al. 2006
	Rice husk ash	29.28	Lakshmi et al. 2009
	Date stones	298.10	Ben Amor et al. 2015

The isotherms modeling were investigated using four equilibrium models: Langmuir, Freundlich, Temkin, and Brouers-Sotolongo. The mathematical expressions are listed in Tables 6 and 7. The graphic correlation between the experimental data and the theoretical models for the adsorption process is shown in Fig. 7a and b for indigo carmine in the presence and absence of AC, respectively. The isotherms fit and their parameters were, respectively, followed and calculated using nonlinear regressions analysis.

First, Table 7 shows that the experimental data were better described by the Brouers–Sotolongo for both processes. In fact, the Brouers–Sotolongo isotherm model presents the best fit by offering the highest nonlinear R^2 (0.988 without AC and 0.994 with AC). In addition, results show clearly that Freundlich model is not appropriate to fit the isotherm curves for adsorption of indigo carmine onto activated carbon. The Freundlich results show that the utilization of this model gives the lowest R^2 (0.7814 without AC and 0.7949 with AC). Similar results were obtained in other studies (Brouers and Al-Musawi 2015).

Comparison between coupling of electrochemical process with adsorption and other adsorption process using activated carbon for IC.

To determine the efficiency of our process (coupling of electrochemical process with adsorption process) compared to others adsorption process to eliminate IC from aqueous solutions, a comparison was realized according to the adsorption capacity (Q_m) (Table 8).

Table 8 shows that the coupling of electrochemical process with adsorption process could be an effective method to eliminate IC when compared to the other study using activated carbon to remove IC.

Several studies have found the values of Q_m for the adsorption of IC lower than the coupling of electrochemical process with adsorption process such as Secula et al. (2011); Odogu et al. (2016); Rehman et al. (2014), and Lakshmi et al. 2009.

Conclusion

The absorption tests of the indigo carmine on the activated carbon (FILTRASORB 200) were carried out in the absence and in the presence of AC in different conditions of AC voltage, pH, temperature, and concentration. The effectiveness of the AC in the increased speed of adsorption was demonstrated. The best dye adsorption was found at pH 2; it is 99 % for the highest initial concentration 100 mg/L and at a voltage of 15 V. The addition of NaCl decreased greatly the amount adsorbed because of the reduction of the electrostatic forces between the adsorbate and the adsorbent. The thermodynamic study showed that the adsorption of indigo carmine is a spontaneous and endothermic process and while the kinetic study showed that the process follows a kinetic of BSf(1, α) model. The adsorption isotherm studies showed that the Brouers–Sotolongo model presents a good fit to the experimental data. AC has a little effect on the parameters of the adsorption equilibrium; however, it has a great effect on the speed of adsorption that it accelerates. Therefore, we succeed to improve the performance of FILTRASORB 200 by increasing the speed of the adsorption process and reduce the processing time.

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