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ORIGINAL ARTICLE

Improvement of Methylene Blue removal by electrocoagulation/banana peel adsorption coupling in a batch system



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KEYWORDS

Electrocoagulation; Banana peel; Adsorption; Methylene Blue; Kinetics; Energy consumption **Abstract** The present work studies the improvement of Methylene Blue (MB) from aqueous solutions by an electrocoagulation (EC)/banana peel (BP) adsorption coupling process. The kinetics of this coupling process was studied using different amounts of adsorbent material in order to identify the most appropriate dosage, for enhancing wastewater treatment. The influence of current density on the removal efficiency and energy consumption of the EC/BP coupling process was also determined.

The coupling between electrocoagulation and BP showed that the addition of an appropriate BP dosage resulted in the enhancement of the removal rate of dyes especially at lower current densities, and in a considerable reduction in the contact time compared to the conventional simple EC process and simple adsorption process, having achieved efficiency removal of approximately 99%.

Based on the high removal efficiency, short contact time and low energy consumption, the EC/BP coupling method could be recommended instead of the conventional simple EC.

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1. Introduction

The disposal of textile wastewater is currently a major problem from a global viewpoint [1]. It has gained increased attention because the extensive release of synthetic dyes has caused considerable environmental pollution [2]. Importantly, the

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E-mail address: helarc2008@gmail.com (H.P. de Carvalho). Peer review under responsibility of Faculty of Engineering, Alexandria University. accumulation of dyes in water can seriously damage food chains of human and animals [3,4]. Gupta Suhas [5] pinpointed the need of some systematized studies on separation/degradation processes of dyes, and the consideration of wastewater treatment at the source underlines the importance of developing technologies that are simple, reliable, adaptable and relatively cheap. In this way, an increasing interest has been shown in combining process such as electrocoagulation, electro-oxidation, adsorption, ozonation [6] and reverse osmosis [7].

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The electrocoagulation process provides a simple, reliable, and cost-effective method for the treatment of wastewater without the need for additional chemicals, and thus, without the production of secondary pollution [8–11]. It also reduces the amount of sludge which needs to be disposed [12]. The separation by electrocoagulation of pollutants such as hydrocarbons [13,14], dyes [15–18], heavy metal ions [19,20], leachate [21], and various other ions such as boron [22] and fluorine [23] has been reported.

EC utilizes a direct current source between metal electrodes partially submerged in electrolyte. The electric current causes the dissolution of the metal plates into the wastewater [24]. The most widely electrode materials are aluminum and iron, because they are cheap and very effective [25]. In the case of electrocoagulation with Al, the anodic reaction leads to soluble Al^{3+} :

$$Al_{(s)} \to Al^{3+} + 3e^{-} \tag{1}$$

and the cathodic reaction produces hydroxide ion and H₂ gas:

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

Some researchers consider that reaction shown in Eq. (3) occurs to form amorphous $Al(OH)_3$ (gel) [26,27]. The flocs formed of the amorphous $Al(OH)_3$ have large surface area, which is beneficial for rapid adsorption of soluble organic compounds and trapping of colloidal particles [24].

$$Al^{3+} + 3OH^{-} \rightarrow Al(OH)_{3(gel)}$$
(3)

Other researchers suggest that the reaction shown in Eq. (4) takes place at the metal-solution interface before $Al(OH)_{3(gel)}$ can be formed in solution. The soluble $Al(OH)_4^-$ species is a precursor for Al_{13} polymer formation, the most effective specie of polyaluminum coagulant [28–30]. The Al_{13} polymer removes soluble organic compounds by charge neutralization and sweep flocculation [31]. Decolorization efficiency mainly depends on the total dissolved aluminum (Al_T) and the ratio of Al_{13} polymer to Al_T [24]. In fact, dye removal from wastewaters is a combined effect of the actions of $Al(OH)_{3(gel)}$ and Al_{13} polymer [26,32].

$$\mathrm{Al}^{3+} + 4\mathrm{OH}^{-} \to \mathrm{Al}(\mathrm{OH})_{4}^{-} \tag{4}$$

On the other hand, Avsar et al. stated that the main disadvantage of conventional EC consists in the formation of an impermeable oxide film on the cathode [33], resulting in higher energy consumptions and lower efficiencies [33–37]. One of the ways to enhance conventional EC systems aside from the changing polarity of electrodes [38] has been suggested by Narayanan and Ganesan, who reported the use of granular active carbon (GAC), which might be a more efficient and faster separation technique compared to conventional EC [39]. Similar studies have also been reported by Secula et al. [40,38]. On the other hand, coupling the electrocoagulation technique with BP adsorption was not reported before, and it deserves attention because BP is an effective and low-cost coagulant and therefore ideal for application in developing countries.

The main aim of the present work was to investigate the efficiency of coupling the electrocoagulation technique with BP adsorption for removing Methylene Blue from aqueous solution. Cell voltage and solution pH were monitored during the electrocoagulation process. The unit energy demand

(UED), the unit electrode material demand (UEMD) and the charge loading (Q_e) were also determined and discussed. A pseudo-first order kinetic model was tested on both simple EC and EC/BP coupling processes to determine the values of rate constant and half time under various experimental conditions. Based on the rate constant values and energy consumption (E_{con}), optimization of the coupling process by adding different amounts of BP was evaluated. The influence of current density on the enhancement of removal efficiency and E_{con} of the EC/BP coupling process was established.

2. Experimental

2.1. Materials

Bananas were purchased at a local market (Changchun, China) and were manually peeled immediately. The banana peels were washed with distilled water to remove the surface adhered particles and water-soluble materials. Then it was sliced, spread on trays and over dried at 60 °C for 48 h. The dried slices were ground and sieved to obtain a particle size range of 60 mesh and stored in plastic bottle for further use. No other chemical or physical treatments were used prior to experiments.

Methylene Blue (MB, C.I. No. 52015, $C_{16}H_{18}CIN_3S \cdot 3H_2O$, MW = 373.90 gmol⁻¹), was purchased from Shanghai Jinsui Bio-Technology Co., Ltd. (Shanghai, China) (Fig. 1).

Solutions of MB (volume 1 L) were prepared before each experimental run by dissolving certain amounts of dye in ultrapurified water.

To adjust the solution conductivity, a weight of 1 g of NaCl (Beijing Shiji, Beijing Chemical works, China) was dissolved in synthetic dye solutions (volume 1 L).

2.2. Electrocoagulation experiments

The electrocoagulation reactor consisted in a parallel-plate electrocoagulation cell provided with two facing electrodes and with six perforated tubes attached to its bottom to maintain a uniform gas flow and stirring into the cell. The experimental setup used in this study is shown in Fig. 2. The effective area of the each electrode plate was 97 cm². The anode and cathode were positioned vertically and fixed at a distance of 1 cm from each other. The electrodes were aluminum and stainless steel (SS-304) plate connected to a digital DC power supply (WYJ, 0–30 V; 0–5 A; DC Regulated Power Supply Double Way Output, TESTMART, Shanghai, China). The electrodes were polished with fine-grained emery paper, washed with 1 N H₂SO₄ and then with distilled water before each run. All the runs were performed at room temperature 20 ± 1 °C.



Figure 1 Chemical structure of Methylene Blue.



Figure 2 EC experimental setup: 1 -anode (aluminum); 2 -cathode (stainless steel); 3 -reactor; 4 -aquarium air pumps with two outlets; 5 -perforated tubes; 6 - DC power supply.

The experiments were carried out in batch mode. In each run (time interval of 1 h), 1000 cm^3 of 50 mg/L dye solution was placed in the electrochemical cell and three aquarium air pump with two outlets (SOBO SB-648, max output: $2 \times 4 \text{ L/min}$, Zhejiang, China) was used to supply needed air to the bottom of the reactor through the perforated tubes. The current density was set to a desired value and then coagulation was started. The pH of the solution was measured by a PHS-3E pH meter (Shanghai INESA and Scientific Instrument Co., Ltd, China).

During experiments samples were taken at different time intervals (5; 10; 15; 20; 25; 30; 40; 50; 60 min). Approximately 5 mL of sample was taken, allowed to settle, filtrated by means of Whatman 0.45 μ m filters, and then analyzed. For all the experiments, a total of 60 cm³ samples were taken to assess the treatment progress.

The concentration of MB was determined at 665 nm using a SDPTOP UV2600PC UV/VIS Spectrophotometer (Shanghai Sunny Hengping Scientific Instrument Co., Ltd, China).

The color removal efficiency (Y(%)) was calculated from

$$Y = (C_o - C_t) / C_o * 100$$
(5)

where C_o is the concentration of dye before electrocoagulation (mg/L), and C_t is the concentration of dye after t min of electrocoagulation (mg/L).

The concentration of dye was determined using the initial calibration curve which was recorded after spectrophotometric measurement of the solution absorbance for dye standard concentration at the specific wavelength corresponding to the maximum absorption of dye.

The FTIR spectrum of the dried BP was obtained on an FT-IR Shimadzu 8400 s spectrophotometer. Scanning electron microscopy with energy dispersive X-ray analysis (SEM–EDX) of the adsorbent before and after adsorption was examined by a Shimadzu XD-3 Scanning Electron Microscope equipped with EDX analyzers.

2.3. Unit energy and electrode material consumptions, charge loading and energy consumptions

The most important costs of electrocoagulation technology are related to the consumption of electrical energy and electrode material [38].

When EC tests are conducted in galvanostatic regime, i.e., the current intensity is maintained constant, the cell voltage varies. Therefore, the energy consumption related to the amount of removed dye, unit energy demand (UED, kW h/kg) [41,42], can be determined by means of the following relationship:

$$\text{UED} = \frac{I \cdot \int_0^t U \cdot dt}{1000 \cdot V \cdot C_o \cdot \frac{Y_t}{100}} \tag{6}$$

where U – cell voltage, (V); I – current intensity, (A); t – time, (h); V – volume of treated solution, (m³); Y_t – color removal efficiency at time t, (%).

The generation of coagulant during EC process leads to the consumption of electrode material that can be estimated based on Faraday's law [43] as follows:

$$\text{UEMD} = \frac{I \cdot t \cdot A}{n \cdot F \cdot V \cdot C_o \cdot \frac{Y_t}{100}} \tag{7}$$

where UEMD – unit electrode material demand, (kg/kg); t - time, (s); n - number of electrons involved in oxidation/reduction reaction; <math>F - Faraday's constant, (C/mol); A - atomic mass of electrode material, (g/mol).

The charge loading (Q_e) and energy consumption (E_{con}) per gram of dye molecule were calculated using Eqs. (8) and (9) [44].

$$Q_e = It_{\rm EC}/C_o \tag{8}$$

$$E_{\rm con} = U I t_{\rm EC} / C_o \tag{9}$$

where Q_e – charge loading per gram of dye (A h g⁻¹), $E_{\rm con}$ – specific electrical energy consumption per gram of dye (W h g⁻¹), C_o – initial dye concentration (g), I – current (A), U – cell voltage (V), and $t_{\rm EC}$ – electrolysis time (h).

2.4. Determination of rate constant

In the processes investigated, conventional electrocoagulation and coupling, the rate of MB removal is proportional to the concentration of dye, the amount of aluminum hydroxides formed and BP added [45] as follows:

$$-dC/dt = kCC_{\rm ads} \tag{10}$$

where *C* is the concentration of dye (kg/m³), and C_{ads} refers to the concentration of adsorbents present in the system (kg/m³), aluminum hydroxides in case of conventional EC, and aluminum hydroxides and BP in the case of the coupling process. Since the generation of aluminum hydroxide can be assumed constant for a given current density, Eq. (10) can be reduced to pseudo first order kinetics:

$$-dC/dt = k_1 C \tag{11}$$

The integration of Eq. (11) results in

$$\ln(C_o/C) = k_1 \cdot t \tag{12}$$

The rate constant k_1 can be estimated from the plot $\ln(C_o/C)$ versus electrocoagulation time.

3. Results and discussion

The effect of BP material on the removal of MB by electrocoagulation was evaluated. MB can be correctly adsorbed onto the considered BP. The EC technique also proved to be very effective and relatively cheap compared to the simple adsorption process by BP. By coupling BP adsorption with the EC technique it was expected that the overall rate constant of dye removal would be enhanced.

3.1. FT-IR spectroscopy, scanning electron microscopy and energy dispersive X-ray analysis of banana peel

The FT-IR spectra of BP were taken before and after the adsorption of MB to ascertain the possible involvement of the functional groups on the surface of BP in the adsorption of MB (Fig. 3). Fig. 3a shows a number of peaks, indicating the complex nature of the adsorbent. Bands appearing at 3313.4, 2920.3, 2850.6, 1734, 1613.6, 1317.4, 1035.2 and 884.6 cm⁻¹ in Fig. 3a were assigned to OH stretching, C–H stretching of alkane, C–H and C=O stretching of carboxylic acid or ester, COO⁻ anion stretching, OH bending, C–O stretching of ester or ether and N–H deformation of amines respectively [46]. Out of these, carboxylic acid and hydroxyl groups played a major role in the removal of MB. On the other

hand, Fig. 3b reveals that some new bands appeared, some remained and some were shifted after MB sorption. The bands at 3500, 3313.5, 2920.3 and 1613.6 cm⁻¹ were shifted to 3846.3, 3336, 2906.3, 1622.9 cm⁻¹ after MB adsorption. This is an indication that OH, C–H, C=C and C=O, carbonyl group could be involved in the adsorption of MB onto BP.

The SEM image of BP before adsorption (Fig. 4a) shows that the peel particles pores are highly heterogeneous. However, after adsorption, a significant change in the structure of the peel is observed (Fig. 4b). The peels appear to have a rough surface with crater-like pores because they are partially covered by MB molecules.

In order to know the composition of BP, elemental analysis was done with the use of EDX analysis. The EDX analysis of unadsorbed BP, shown in Fig. 4c, showed the presence of various elements along with a high amount of potassium. The difference in peak intensities of each element before and after adsorption (Fig. 4d) as well as the presence of additional



Figure 3 FTIR characterization of BP before (a) and after (b) MB adsorption.



Figure 4 Scanning electron microscopic (SEM) and energy dispersive X-ray (EDX) analysis of (a), (c) unadsorbed and (b), (d) MB adsorbed banana peel.



Figure 5 Abatement rate of Methylene Blue at different values of current density (a) without BP and (b) with 5 g of BP; $[MB]_0 = 50 \text{ mg/}$ L, [NaCl] = 1 g/L.

peaks after adsorption (such as N, Na, and Al), demonstrates the change of the characteristics of the BP, and confirms the adsorption phenomenon.

3.2. Influence of current density

Current density not only influences coagulant dosage but also bubble formation rate, their size and the flocs growth [47]. In order to establish a favorable value of current density for the EC/BP coupling process, four values of current density, 2.5, 5, 7.5 and 10 mA/cm², were chosen to be applied during simple EC and EC/BP processes to evaluate the effect on decolorization over a wide range of current density parameters. In Fig. 5, the abatement rate produced by the two processes investigated is compared.

Compared to the simple EC technique, the addition of a quantity of 5 g/L of BP in the electrocoagulation cell resulted in faster decolorization. The highest enhancement of



Figure 6 Evolution of removal efficiency in the absence and at different doses of BP. $[MB]_0 = 50 \text{ mg/L}$, [NaCl] = 1 g/L, $CD = 2.5 \text{ mA/cm}^2$.

decolorization by adding BP was obtained at low current density values, although even at high current density values the separation process was significantly improved. The rapid removal of MB can be attributed to the fact that the system suffered both reactions simultaneously, electrocoagulation and also adsorption process. In all the experimental investigations presented below, a current density of 2.5 mA/cm² was therefore applied in order to demonstrate the effects of the BP dose and dye initial concentration on the removal efficiency and operations related to the consumption of electric energy.

3.3. Influence of BP dosage

Fig. 6 compares the evolution of the removal efficiency of dyes for simple EC and for EC/BP coupling at different BP dosages demonstrating the influence of the amount of BP on the removal efficiency.



Figure 7 Pseudo-first kinetics of EC/BP coupling process at several doses of BP. $[MB]_0 = 50 \text{ mg/L}$, [NaCl] = 1 g/L, $CD = 2.5 \text{ mA/cm}^2$.

 Table 1
 Kinetic parameters of simple electrocoagulation and electrocoagulation/BP coupling for several amounts of BP.

BP concentration (g/L)	Pseudo-first order model			
	$k_{0-10} \ (\min^{-1})$	$t_{1/2}$ (min)	$k_{10-60} \ (\min^{-1})$	$t_{1/2}$ (min)
0.0	0.02181	31.781164	0.00661	104.863416
0.2	0.03248	21.340738	0.0181	38.295424
0.4	0.12687	5.463444	0.00536	129.318504
0.8	0.09256	7.488626	0.00945	73.348908
2.0	0.16841	4.115831	0.01967	35.238799
3.0	0.2159	3.210501	0.01383	50.119102
5.0	0.33553	2.065828	0.00379	182.888438
6.0	0.24742	2.801500	0.0101	68.628434
7.0	0.2667	2.598977	0.00763	90.844978



Figure 8 Activity–pH diagram for Al(III) species in equilibrium with Al(OH)₃ (amorphous).

It can be seen that adding low amounts of BP, e.g. 0.2 g/L, does not increase significantly the removal efficiency compared with no addition of BP. The addition of a quantity of BP above 2 g/L or more has a strong influence on dye removal from the very beginning of the electrocoagulation/BP process. As the BP dosage is increased, maintaining the other parameters constant, the removal efficiency of MB becomes significantly higher and the contact time for obtaining 97% removal efficiency decreases until 5 min. Although higher dosages of BP above 5 g/L lead to a low level of the remaining MB, we should take into account that an excessive amount of coagulant may provoke not a very effective procedure, as the BP will remain without coagulant, dissolved in water [48].

Fig. 7 shows the results of the kinetics investigations of electrocoagulation/BP coupling at different doses of BP. It shows that, in the first 10 min of coupling process, the MB depletion is a pseudo-first-order reaction and also shows that a pseudo-first order reaction might describe the behavior for the system over the 10–60 min period. However, in this case the values of rate constant (k) were lower compared with the first 10 min of the electrocoagulation (Table 1). This was probably due to the fact that during oxidation there is a formation of intermediates that are more difficult to remove [49]. A similar behavior was observed by other researchers [50,51].

3.4. Variation of solution pH during EC/BP coupling

The role of pH in EC process is very important [52]. pH affects $Al(OH)_3$ stability in the solution [52]. Fig. 8 demonstrates different forms of $Al(OH)_3$ relative to the pH and concentration of Al^{3+} ions in the media [53]. In the high and low pH, $Al(OH)_3$ is in its charged form and is soluble in water, hence, cannot be used for EC. But in neutral pH, $Al(OH)_3$ is stable and insoluble in the water and available for pollutant adsorption from water [52].

Some studies reported in the literature claim that controlling pH in iron-based electrocoagulation processes leads to higher efficiencies [42]. However, the mechanism of electrocoagulation depends mainly on the nature of the pollutant [54,55]. Recently, Secula et al. [56] found that the addition of acid solution to control the pH value of the treated solution in fact limits the development of the flocs generated.

Fig. 9 shows the influence of the amount of the BP on the solution pH during the EC/BP coupling process. As expected, the increase in BP dose leads to a low final pH of the treated effluents, making it possible for the EC/BP coupling process



Figure 9 Variation of pH value during EC/BP coupling process, at several doses of BP ranging between (a) 0 and 2 g/L, (b) 3 and 7 g/L. $[MB]_0 = 50 \text{ mg/L}, [NaCl] = 1 \text{ g/L}, CD = 2.5 \text{ mA/cm}^2.$

to be conducted at a more favorable pH value (neutral pH), consequently, creating stability of $Al(OH)_3$ in the water for pollutant adsorption.

3.5. Influence of the dye initial concentration

This parameter was investigated in the range of 50–300 mg/L. Fig. 10 shows that the increase of the dye initial concentration causes a reduction in color removal conversion. With increasing the dye initial concentration while amounts of the coagulant are constant, the adsorption capacity of flocs becomes insufficient and a reduction in color removal conversion will occur [52].

3.6. Unit energy demand, electrode material consumptions, charge loading and energy consumptions

3.6.1. Unit energy demand

Fig. 11 presents the influence of different amounts of BP on the evolution of UED determined according to Eq. (2).

It can be seen that adding amounts of BP results in an increase of UED values with contact time, except for quantity of BP between 5 and 7 g/L which had significant fluctuations in the values of UED, and consequently had a negative influence on dye removal once the removal efficiency decreased. It is more notorious especially at durations above 20 min of EC/BP coupling.

3.6.2. Electrode material consumptions

Fig. 12 shows the influence of different amounts of BP on the evolution of UEMD determined according to Eq. (3).

It can be seen that the longer contact time of the system applied, the weight of the electrode consumed in the simple EC and EC/BP coupling process has been increased. It also notorious that amount in excess of BP (above 5 g/L) leads to significant decreases in the electrode consumption and consequently the removal efficiency of MB dye also decreases.



Figure 10 Influence of initial concentration of the dyes on the color removal conversion. $[NaCl] = 1 \text{ g/L}, \text{ CD} = 2.5 \text{ mA/cm}^2.$



Figure 11 Variation of UED as a function of BP dose at several values of contact time.



Figure 12 Evolution of unit electrode material unit (UEMD) during EC/BP coupling as a function of BP dose at several values of contact time.

3.6.3. Charge loading

Fig. 13 shows the influence of different amounts of BP on the evolution of Q_e determined according to Eq. (4).

As expected, an increase of contact time leads to significant increase in the Q_e value. It can be seen that when the amount of BP in solution increases, the Q_e value increases. Also, adding amounts of BP above 5 g/L led to large fluctuations in the values of Q_e , affecting negatively the efficiency of the EC/BP coupling process.

3.6.4. Energy consumptions

The variation of electrical energy consumption (determined according to Eq. (5)) with the contact time for simple EC and for EC/BP coupling at different BP dosages is presented in Fig. 14. It is shown that the electrical energy consumption



Figure 13 Evolution of charge loading (Q_e) during EC/BP coupling as a function of BP dose at several values of contact time.



Figure 14 Variation of energy consumption in the absence and at different doses of BP. $[MB]_0 = 50 \text{ mg/L}$, [NaCl] = 1 g/L, $CD = 2.5 \text{ mA/cm}^2$.

ranging from 0 to 1.65 W h g⁻¹ increased proportionally with contact time (0–60 min) for both electrocoagulation processes (simple EC and EC/BP coupling). In first 5 min, the energy consumption for the EC/BP coupling process with the optimal dosage did not differ with the process of simple EC which is roughly 0.07 W h per gram of dye, having increased in relation to simple EC for contact time above 10 min.

4. Conclusions

In this work we discussed the effects of dye removal by EC/BP coupling on decolorization and energy consumptions in comparison with simple EC. It was found that the removal of MB by electrocoagulation using aluminum (anode) and stainless steel (cathode) and adsorption using BP are feasible processes.

The addition of BP as adsorbent resulted in a rapid increase in the removal efficiency of MB especially at lower current densities and operating times compared with the conventional electrocoagulation process. The success of this coupling process is linked to the specific chemical and textural properties of the material and the best conditions found were used for the electrocoagulation/banana peel coupling.

The removal rate and energy consumption were found to increase when the current density was increased from 2.5 to 10 mA/cm^2 .

Increasing the dosage of BP from 0.2 to 7 g/L improved the kinetics of the coupling process rapidly (5 min was sufficient to remove almost all color $\approx 97\%$). However, in terms of energy consumption and removal efficiency a dosage of 5 g/L is optimal, and further increases resulting in insignificant increase and reductions in energy consumption, affecting negatively the efficiency of the EC/BP coupling process.

It was shown that the EC/BP coupling technique for MB removal is considerably cheap, and is suitable for developed and developing countries.

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