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ORIGINAL RESEARCH PAPER

A comparative study for the electrochemical regeneration of adsorbents loaded with methylene blue

Ines Bouaziz[1,2], Morched Hamza[2], Ridha Abdelhedi[2], André Savall[1], Karine Groenen Serrano[1,]*

[1] *Laboratory of Chemical Engineering, University of Toulouse, CNRS, INP, 118 route de Narbonne 31062 Toulouse cedex, France*

[2] *Laboratory of Electrochemistry and Environment, National Engineering School, Sfax University, BP 1173, 3038 Sfax, Tunisia*

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ABSTRACT

The electrochemical regeneration of methylene blue-saturated activated carbon, Nyex®1000 and sawdust has been studied and the performances in terms of capacity of adsorbent regeneration have been compared in this work. The adsorption isotherms were investigated. The results showed that the adsorption of methylene blue onto the investigated adsorbents obeyed Langmuir's model. The electrochemical oxidation of methylene blue beforehand adsorbed was studied using a boron doped diamond anode. The electrochemical regeneration efficiencies, under the same experimental conditions, of the activated carbon and Nyex®1000 were significantly less than 100% which were much lower to that of sawdust. Indeed the electrolysis tends to activate the sawdust because all the regeneration efficiencies obtained, whatever the applied current intensity, are higher than 100 %. Increasing treatment time would also result in a better regeneration of sawdust. This study confirmed that the coupling adsorption onto sawdust and electrochemical degradation is a potential technique for the efficient elimination of low concentration organic dyes from wastewater.

Keywords: Electrochemical regeneration, adsorption, methylene blue, Sawdust, Nyex®1000, Activated carbon, wastewater treatment.

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INTRODUCTION

Pollution caused by industrial wastewaters has become a common problem for many countries. The search for alternative wastewater treatment technologies has been intensified due to a growing public concern about health and related environmental issues of trace levels of pollutants [1-2]. Dyes are one of the common organic pollutants in wastewaters. Over 7×10^5 tones and approximately 10,000 different types of dyes and pigments are produced world wide annually [3]. Most of these compounds are visually detected even in low concentrations, carcinogenic, mutagenic, and can cause a severe health hazard to human beings.

Due to the low biodegradability of colored compounds, a conventional biological treatment process is not very effective. Dyes laden wastewaters are usually treated by physical or chemical processes such as adsorption [4], membrane filtration [5], ion exchange [6], ozonation [6], Fenton oxidation [6] and so on. However, these methods have different color removal capabilities, produce secondary waste and involve capital costs [6].

In recent years, the electrochemical method for wastewater treatment has attracted a great

* Corresponding Author Email: serrano@chimie.ups-tlse.fr

deal of attention, especially regarding the electrochemical oxidation [7-11]. This process was applied successfully to the total destruction of different persistent organic pollutants and in particular of dyes. It has been determined that the nature of the anode material is the main factor that affects the process [7]. In fact, with the use of nanostructured boron doped diamond (BDD) anode, many refractory organic compounds can be completely mineralized with high efficiency due to electrogenerated hydroxyl radicals from water discharge without addition of any reagent [9,11]. Although electrochemical oxidation is an attractive means for wastewater treatment, the low current efficiency is still a critical problem for the treatment of dilute solutions because of mass-transfer limitations. Therefore, organic pollutants in industrial wastewater should be concentrated to obtain a large quantity of organic pollutants before carrying out electrochemical degradation. Consequently, a single electrochemical process alone may not be adequate for the treatment of organic compounds. Hence, the researchers are attempting a combination of two or more treatment methods to overcome these limitations.

In this study, the adsorption has been chosen as preconcentration step and the electrochemical process allows the desorption and destruction of the adsorbed organic matter, restoring the adsorptive capacity of adsorbent.

Electrochemical regeneration of activated carbon (AC), the most popular adsorbent, has been widely investigated [12-16]. The first report on the electrochemical regeneration of activated carbon was by Owen and Barry [12] who achieved regeneration efficiencies of up to 61%. Whilst other researchers have demonstrated that efficiencies of electrochemical regeneration of activated carbon were often significantly less than 100% [13-20]. Moreover, the rate of adsorption and desorption of organic compounds from loaded adsorbent is often governed by intra-particle diffusion which requires long adsorption and regeneration periods [13]. Research in the development of alternative adsorbents has been performed. Over the last few years, Brown et al. [17-19] have worked on an alternative approach to adsorption and electrochemical regeneration based on a novel, non-porous, highly-conducting carbon-based adsorbent material (called Nyex®). It was shown that this adsorbent can be rapidly and fully electrochemically regenerated with low energy consumption.

In most cases, the regeneration of adsorbent by electrochemistry was studied in the presence of sodium chloride. However, using NaCl the main drawback is the formation of hazardous organochloride by-products during the electrolysis. That is why sodium sulfate has been chosen in this study.

We have recently studied the coupling of adsorption onto sawdust, chosen as a by-product of furniture industry without commercial value, and electrochemical oxidation in the presence of Na_2SO_4 [20]. The electrochemical oxidation of methylene blue adsorbed onto sawdust led simultaneously to its degradation and sawdust regeneration for the next adsorption. It was also observed that multiple adsorption and electrochemical regeneration cycles led to an enhancement of adsorption capacity of the sawdust.

The efficiency and performance of the different electrochemical regeneration methods cannot be directly compared to each other if the lay out of the electrochemical reactor, the adsorbate, and/or electrodes are different. To the best of our knowledge, no comparison including the electrochemical regeneration of AC, Nyex® and sawdust has been reported to date. This work is intended to fill this gap and make a comparative study between electrochemical regeneration of the three organic saturated adsorbents under the same experimental conditions. For this purpose, the treatment of aqueous solution of methylene blue (MB) by coupling adsorption onto commercial activated carbon, Nyex®1000 and sawdust, and electrochemical oxidation on BDD anode has been studied.

MATERIALS AND METHODS

Materials

Chemicals

MB, a cationic dye with a chemical formula of $C_{16}H_{18}N_3$ and molecular weight of 319.85 g mol^{-1}, was chosen as a model organic contaminant. MB was purchased from Merck, and it was used as received without further purification. MB solutions were prepared with ultra pure water.

All other chemicals used were of analytical grade (Acros Organics). Na_2SO_4 (0.1M) was used as the supporting electrolyte.

Adsorbents

Three adsorbents were used in this study: sawdust, commercial A.C. and Nyex®1000.

The red wood sawdust, a low cost material with BET surface equals to 0.4m^2/g was obtained from a furniture factory in Sfax, Tunisia.

The commercial A.C. was supplied by Merck (reference 2514). The specific surface area of this adsorbent is 980m^2/g [21].

Both adsorbents were washed with distilled water several times to remove the suspended particles, then dried in a hot air oven at 373 K. The dried adsorbents were sieved and the final sizes of the particles retained were in the range of 0.5 to 1.12 mm and 0.4 mm for sawdust and A.C., respectively.

The third adsorbent used in this work was a graphite intercalation compound, Nyex®1000 supplied by Arvia Technology Ltd. The specification for Nyex®1000 provided by Arvia Technology Ltd and mentioned by Mohamed et al. [19] indicates a carbon content of 95 wt%, with particle diameters in the range of 100 to 700 µm. The BET surface area of Nyex®1000 was found to be 1m^2/g, which is not far to that of sawdust and very small compared to the commercial activated carbon. Its conductivity is about 0.16 $\Omega^{-1}cm^{-1}$.

Experimental methods

Adsorption experiments

The adsorption isotherms studies were performed by batch adsorption technique.

In the case of Nyex®, various known quantities of adsorbent were added to 100 mL of 100 mg/L of MB solution in a 500 mL flask at 303 K.

The adsorption isotherms of sawdust were investigated, at three different temperatures 303, 313 and 323 K, by adding 0.5 g of sawdust into 50 ml of different initial concentrations of MB. For all experiments, the agitation time was one hour to reach equilibrium before analysis.

The adsorptive capacity, q, defined as the mass of adsorbate per gram of adsorbent (mg/g), can be calculated from the initial (C_0) and final (C_f) concentrations of MB according to Eq.1:

$$q = \frac{(C_0 - C_f)V}{m} \quad (1)$$

Where V is the volume of solution used and m is the weight of adsorbent.

To reduce the number of adsorption steps and make sure that the adsorbent has reached saturation before electroxidation experiments, the saturation of adsorbent has been performed in a (30 cm x 1.5 cm) glass column, packed with a known mass of adsorbent sandwiched between two layers of glass wool was used. MB solution with the initial concentration of 300 mg/l (in the case of sawdust and activated carbon) and 100 mg/l (in the case of Nyex) was pumped into the column at a constant flow rate of 0.3 mL/min.

Desorption experiments

Desorption experiments were conducted to quantify the long-term non-electrochemical desorption of MB. For simple desorption studies, the loaded adsorbent after drying at room temperature was contacted with 200 mL of 0.1mol/L Na_2SO_4 solution. The solution was stirred at a temperature of 303 K until the release process reached equilibrium.

Electrolysis

Batch reactor coupling with adsorption

The loaded adsorbent was removed from the column of adsorption and stirred with Na_2SO_4 solution. Immediately, a constant current was applied in the same reactor at 303 K using a Meteix d.c. power supply. The mixture was stirred constantly during the electrochemical degradation of adsorbed and released MB. A nanostructured BDD electrode with a geometric area of 7 cm^2 was used as anode. The counter electrode was a cylindrical mesh of platinum (67.5 cm^2).

The regeneration efficiency of the adsorbent, R_e, by electrolysis on BDD anode was defined as the ratio between the capacity of adsorption of the sawdust after the electro-regeneration, q_r and the initial capacity of adsorption q_i:

$$R_e = \frac{q_r}{q_i} \times 100 \quad (2)$$

Where q_r is the capacity of adsorption of the regenerated adsorbent (mg of MB/g of adsorbent) and q_i is the initial capacity of adsorption of fresh adsorbent under the same adsorption conditions.

It should be noted that for the batch desorption and electrochemical experiments, 1g of loaded adsorbent was contacted with 200 ml of Na_2SO_4 solution in the case of sawdust and activated carbon. Whereas due to the low adsorption capacity of Nyex® and in order to obtain the same range of desorbed MB concentration, 16 g of MB saturated Nyex® were used in 200 ml of Na_2SO_4. Moreover, a high concentration of MB solution was used in this study to reach the saturation of adsorbent in short time. All the experiments have been performed at natural pH.

Analytical techniques

The concentration of MB in the solution was determined using a Hewlett-Packard UV-visible spectrophotometer at a wavelength of 660 nm. A previously established linear Beer–Lambert relationship was used to determine the concentration. In a range of 0-8 mg/L of MB, the molar extinction coefficient obtained was: ε= $6.3x10^4$ L mol^{-1} cm^{-1} at 660 nm at 298 K. The relative standard deviation has been determined as 2%.

RESULTS AND DISCUSSION

Adsorption

Batch adsorption studies at 303 K have been performed previously to investigate the kinetic and equilibrium isotherm of MB adsorption onto sawdust [14]. Experimental data obtained at 303 K were compared with models of Langmuir (Fig.1) and Freundlich (Supplementary materials). These models suggest interactions between the adsorbed molecules, assuming a heterogeneous surface with a non-uniform distribution of heat of adsorption on the surface in the case of Freundlich model. The Langmuir model supposes a monolayer with a homogeneous distribution of sorption sites and sorption energies without interactions between the adsorbed molecules. In order to optimize the design of a sorption system to remove MB from wastewater, it is important to establish the most appropriate correlation from the equilibrium curve. Both models are commonly used for investigation of the sorption of a variety of dyes on sawdust and activated carbon.

The linear form of Langmuir equation is given by Eq. 3:

$$\frac{C_e}{q_e} = \frac{C_e}{q_m} + \frac{1}{K_L q_m} \quad (3)$$

C_e is the concentration of the adsorbate in the solution at equilibrium (mg/L), q_e and q_m are the capacity of adsorption (mg g^{-1}) at equilibrium and maximum, respectively and K_L is the Langmuir isotherm constant (L mg^{-1}). The parameters K_L and q_m can be obtained by plotting C_e/q_e versus C_e.

As shown in Fig. 1, the adsorption isotherms of MB onto Nyex® and sawdust fit very well with the theoretical Langmuir isotherms (R^2>0.996 for sawdust and Nyex) whereas low correlation coefficients (R^2<0.949) were obtained using the model of Freundlich. This result is similar to that found by Mohammed et al. [22] for the adsorption of Acid Violet onto Nyex®1000.

As the adsorption isotherms of MB onto activated carbon were extensively studied in the literature [23], the maximum adsorption capacity of MB onto activated carbon was determined, in this study, with the column adsorption only. Consequently, the highest value of the quantity of MB adsorbed was obtained for the commercial activated carbon (285 mg/g) followed by the sawdust (23 mg/g) and then by Nyex® (1.4 mg/g) which is close from that observed for the sorption of Acid violet onto the same adsorbent [23]. The high adsorption capacity of the activated carbon is due to its very high surface area (980m^2/g). However

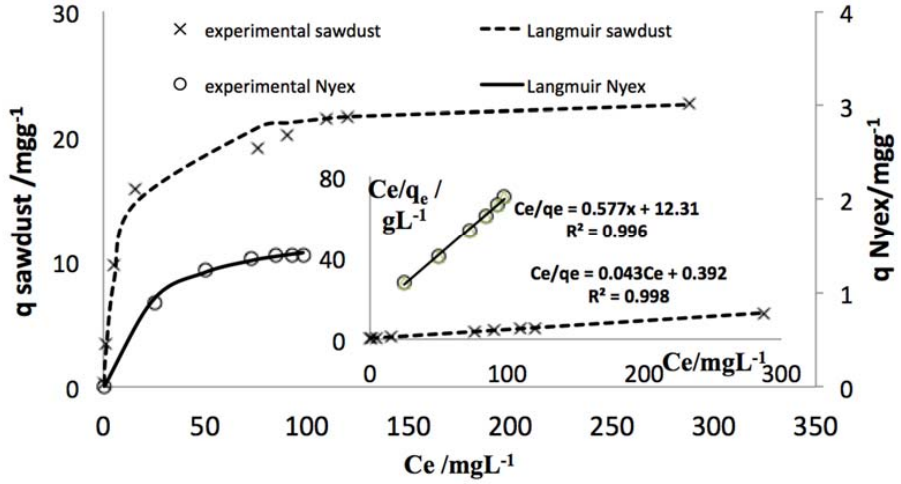

Fig. 1: Adsorption isotherms of MB onto sawdust and Nyex® at 303 K. Comparison of experimental data (symbols) with Langmuir model (lines). Inset panel: Linear form of Langmuir isotherms. Operating conditions: T=303 K. For Nyex®: C°=100 mg/L; V=100 mL; m_{nyex}=0.5-8 g. For sawdust:$m_{sawdust}$ = 0.5g, V=50mL, C°=35-512 mg/L

the sorption capacities of sawdust and Nyex® are inversely proportional to their specific surface area (0.4 and 1m²/g, respectively). The comparison of Langmuir isotherm constants for adsorption of MB calculated in this work with those determined by Hameed et al. [23] for activated carbon are shown in Table 1.

Table 1 shows that the adsorption capacity (q_m) of the different adsorbents for MB is comparable to the maximum adsorption obtained from adsorption isotherms (Fig. 1) and/or column adsorption. However, if the adsorptive capacity (q_m) is normalized with the specific area, it is found that the sawdust is able to adsorb the highest mass of dye per surface unit (q_m = 58 mg/m²), against Nyex (q_m = 1.7 mg/m²) and activated carbon (q_m = 0.3 mg/m²).

These results again show that sawdust, having the lowest surface area, can be an attractive option for the removal of dyes from dilute industrial effluents. It indicates that the adsorption capacities depend not only on the specific surface area or porosity but also on the surface chemistry of the adsorbents. To evaluate the influence of the temperature of the adsorption capacity of the sawdust, adsorption isotherms were performed at 313 and 323 K and shown in Fig. 2. It can be observed that adsorption decreases with an increasing temperature. Same results were obtained by many authors for the adsorption of dyes on various adsorbents [24-25]. This can be explained by the exothermicity and spontaneity of the adsorption process and by the weakening of bonds between dye molecules and active sites of adsorbents at high temperatures.

All adsorption isotherms are fitted well with the Langmuir model. The Langmuir constants and correlation coefficients calculated from the linear form of Langmuir equation are given in Table 2.

Table 1: Comparison of Langmuir constants and desorption parameters for adsorption of methylene blue onto sawdust, Nyex® and activated carbon at 303 K.

Adsorbent	Langmuir constants			Desorption parameters	
	q_m (mg/g)	K_L (L/mg)	R^2	Desorbed amount of MB (%)	[MB] desorbed (mgL^{-1})
Sawdust	23.2	0.11	0.998	33	38
Nyex®1000	1.7	0.05	0.996	10	10.5
Activated carbon	294.1[23]	0.13[23]	0.999 [23]	0.17	2.3

Table 2: Langmuir parameters for the adsorption of methylene blue onto sawdust at 303, 313 and 323 K. Operating conditions: see Fig. 2

Temperature	q_m (mg/g)	K_L (L/mg)	r^2
303 K	23.2	0.11	0.998
313 K	16.1	0.10	0.997
323 K	13.1	0.09	0.999

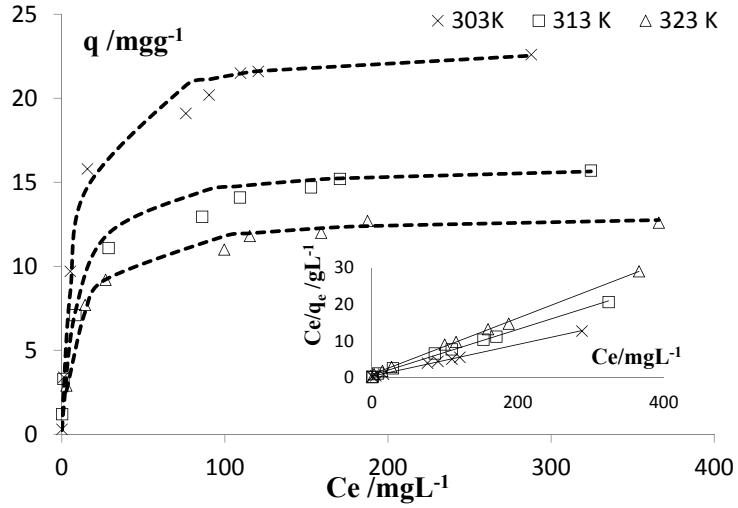

Fig. 2: Adsorption isotherms of MB onto sawdust at three different temperatures 303, 313 and 323 K. Comparison of experimental data (symbols) with Langmuir models (lines). Inset panel: Linear form of Langmuir isotherms. Operating conditions: msawdust=0.5g, V=50 mL, C°= 12-600 mg/L.

Thermodynamic parameters of MB adsorption onto the sawdust samples, including free energy change (ΔG°), enthalpy change (ΔH°) and entropy change (ΔS°), were calculated using the following equations:

$$\Delta G°=-RT\ \ln K \quad (4)$$

$$\Delta G° = \Delta H° - T\Delta S° \quad (5)$$

Where T is the temperature (K), R is universal gas constant (8.314 J mol^{-1} K^{-1}), and K (l g^{-1}) is an equilibrium constant defined as the product of the Langmuir constants q_m and K_L.

The values of ΔH° and ΔS° were calculated from the intercept and slope of ΔG° versus T according to Eq. (5) by linear regression analysis. The results are listed in Table 3.

Due to the values of the free energy obtained in Table 3, one can deduce that physisorption of MB takes place on sawdust. Indeed, it's known that for physisorption, the change in free energy for adsorption of MB onto sawdust ranges between -0.4 kJ $mol^{-1}$and -2.36 kJ mol^{-1}. It is known that, the absolute magnitude of the change in free energy for physisorption is between -20 kJ $mol^{-1}$and 0 kJ mol^{-1} and for chemisorption the corresponding range is -80 kJ mol^{-1}-400 kJ mol^{-1} [26]. The negative value of ΔH° and ΔS° confirm that the adsorption process is exothermic and renders the molecule distribution more orderly in the adsorbent than in the solution.

Desorption

Batch desorption experiments of MB from loaded sawdust in Na_2SO_4 aqueous solution have been previously performed [20]. Similar experiments were carried out with the commercial A.C. and Nyex® in order to compare the results and quantify the long term desorption of MB from these three adsorbents in the Na_2SO_4 solution without electrolysis. The comparison is shown in Fig. 3 at 303 K.

The calculated desorption rates are shown in Table 1. In the case of activated carbon, the desorption equilibrium was reached in 20 min and the desorbent amount of MB was 0.44 mg/g corresponding to 0.17% of the adsorbed dye. The low desorption capacity of activated carbon suggests that chimiosorption is involved. The desorbed amount of MB from the Nyex® at equilibrium, after 10 min, was relatively higher, 0.13 mg/g corresponding to 10% of the MB adsorbed. The highest value of the desorbed amounts of the dye, at the equilibrium in 15 min, was obtained from the sawdust and was close to 7.6 mg/g, accounting for about 33% of the adsorbed amount of MB.

Table 3: Thermodynamics parameters for the adsorption of methylene blue onto sawdust at different temperatures. The negative values of ΔG° indicate the spontaneous adsorption of MB onto sawdust samples.

Temperature	ΔG° (kJ mol^{-1})	ΔH° (kJ mol^{-1})	ΔS° (kJ mol^{-1} K^{-1})
303 K	-2.36	-32	-0.1
313 K	-1.23		
323 K	- 0.4		

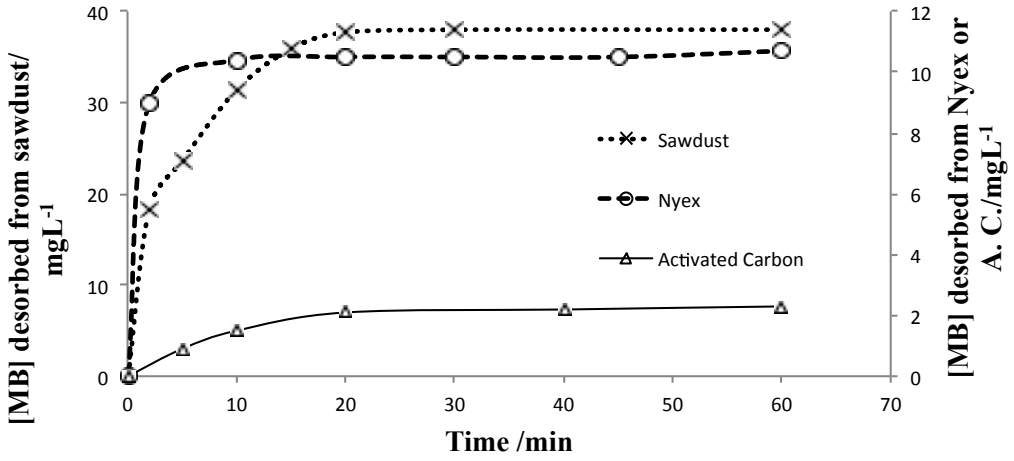

Fig. 3: Desorption kinetics of MB from sawdust, Nyex® and Activated Carbon at 303 K. Operating conditions : 1 g of saturated sawdust or A.C. and 16 g of Nyex®, V=200 mL, [Na_2SO_4]= 0.1 mol/L, T=303 K.

Electrochemical regeneration

The adsorbent after saturation with MB was contacted with Na_2SO_4 solution. Immediately, a constant current was applied in the same reactor at 303 K and the temporal variation of the concentration of MB in solution has been measured. Fig. 4 shows the comparison of the concentration profile for a single desorption (full symbols) and for the electrolysis (empty symbols). It appears that the speed of appearance of MB and the MB concentration in the solution are less important with electrolysis. This difference may be explained by the fact that a part of desorbed MB is oxidized on BDD surface. Consequently the instantaneous variation of MB concentration during the electrolysis results on the speed difference of MB desorption from the sawdust and oxidation of MB desorbed.

The efficiencies of the electrochemical regeneration on the three adsorbents have been compared in Fig. 5 at different applied current intensities and different regeneration time. One can observe that for a regeneration time of 600 min for an applied current density of 0.215 A/cm^2, the regeneration efficiencies of activated carbon and Nyex® were low, only 35 and 44%, respectively, in comparison with the one of sawdust (240%).

The limitation in the electrochemical performance of activated carbon can be attributed to the nature of the activated carbon and, more specifically, by its characteristics of micropores structures. Indeed, the narrow microporosity that favors MB uptake may also hinder subsequent MB desorption (as demonstrated in paragraphs 3-2), and consequently the regeneration efficiency achieved by electrochemistry. These trends have been observed by other researchers. For example Narbaitz et al. [27] have found that the electrochemical regeneration reached only 8-15% in the case of adsorbed natural organic material (NOM) on granular activated carbon. The authors have been attributed the low efficiency to the high irreversibility of NOM adsorption onto A.C.

Fig. 5 shows also that the regeneration efficiency of Nyex cannot exceed 50% whatever the treatment time in the range of 120 to 616 min and the applied current densities used. The low electrochemical regeneration efficiencies were surprising in comparison with the ones obtained by Brown and co-workers [17-19,22]: they have demonstrated that the Nyex®, which considered as the first adsorbent specifically designed for electrochemical regeneration, can be rapidly and fully regenerated by electrochemical oxidation. For example, they observed a regeneration efficiency of 100% in 10 min by passing a charge of 25 C g^{-1} and suggested that the organic compounds were oxidized during regeneration without being released. However, it should be noted that it is difficult to compare with our results because the design of the electrochemical cell is different (they used a bed of loaded Nyex) whereas in the present work, the Nyex particles were in suspension in the electrochemical batch cell. Moreover amount of sodium chloride were added

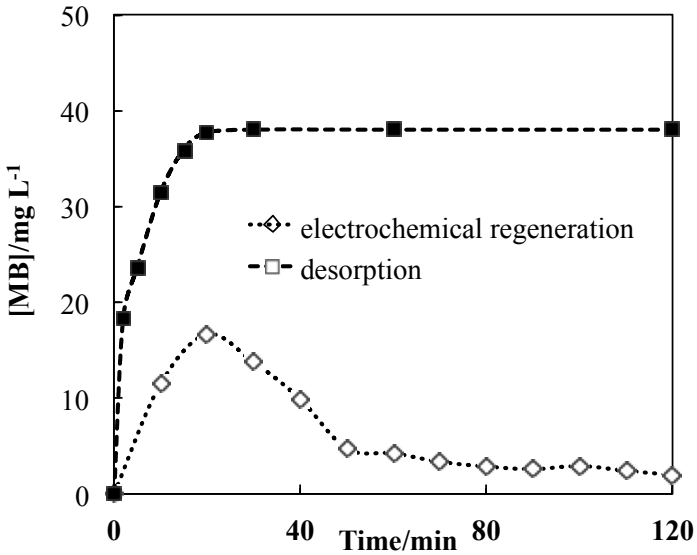

Fig. 4: Comparison of variation of MB concentration during desorption (full symbol) and electrochemical regeneration (empty symbol). [Na_2SO_4] =0.1 M, $m_{sawdust.}$ = 1 g, q° = 21 mg/g. anode: BDD, i = 0.215 A/cm^2, V = 100 mL, T = 303 K, anode DDB , cathode Platinum mesh.

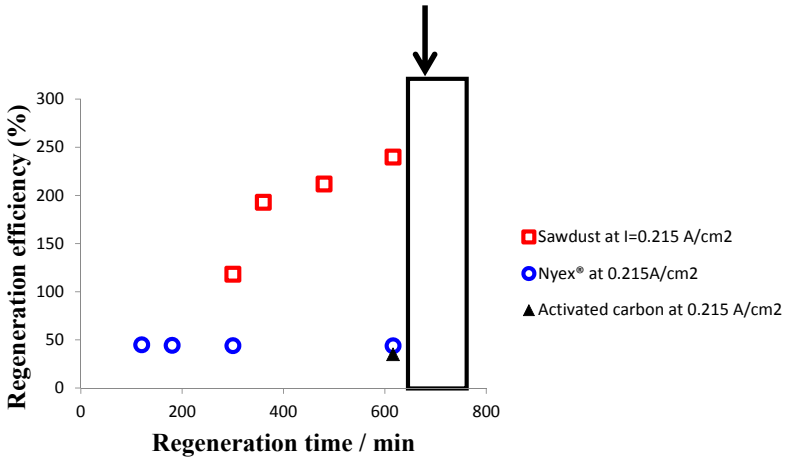

Fig. 5: Comparison of regeneration efficiency of different adsorbent with electrolysis time and applied current intensity. Operating conditions: m_{ads} = 1g for AC and sawdust, 16 g for Nyex®, V= 200 mL, [Na_2SO_4] =0.1 M, T=303 K.

in their cell and electrogenerated hypochlorite ions can react chemically with MB.

The electrochemical treatment of saturated sawdust shows efficiencies higher than 100% with an increase of the regeneration efficiency with regeneration time. This result is similar to that found in our previous work for the lowest current density (0.05 A/cm^2) [20]. Thus, this present work confirms our earlier studies showing that the modification of adsorption of sawdust depends on the electrical charge passed. However in this study, it was observed that the regeneration efficiency increases slightly for regeneration time more than 480 min. It seems that it tends to reach a maximum value. These data also confirm the hypothesis suggested in the previous work that the modification of the surface chemistry of sawdust causes the increase in adsorptive capacity after regeneration [20]. Moreno-Castilla has evidenced that the adsorption capacity depends on the surface chemistry of the adsorbent [27]. Hence, the characterization of surface functional groups of sawdust before and after electrochemical treatment is the subject of on-going research.

As final remark, it appears clearly that, under our experimental conditions, the electrochemical regeneration is largely attributed to the desorption of the organic substances from the adsorbents, the desorption is enhanced under polarization.

CONCLUSION

In order to treat dilute wastewater, the coupling of adsorption and electrochemical oxidation can be considered as a promising approach. It is the first time that a study is devoted on the suitability of the electrochemical regeneration of three adsorbents: Activated carbon, Nyex® and sawdust.

The thermodynamic study shows that the isotherms of the three adsorbents follow the Langmuir model. The maximum adsorption capacity obtained for MB onto the virgin sawdust reached 23 mg/g which is 16.4 times higher than the one obtained onto Nyex® and 12.4 times lower than the one obtained onto AC. The desorption experiments confirm that on A.C. the low desorption occurs due to an intra-particles diffusion controlled process. However on sawdust, 33 % of the MB adsorbed amount was desorbed after 15 minutes of contact with the electrolyte. The desorbed amount from Nyex® corresponds to 10 %. The study of the electrochemical regeneration of adsorbents has evidenced that only sawdust can be completely regenerated and even more: the capacity of adsorption is enhanced after electrolysis. By contrast, whatever the electrical charge used, it appears that electrolysis cannot prevent the deterioration of the adsorption performances of Nyex and A.C.

This present work has confirmed that the coupling adsorption onto sawdust and electrochemical treatment is a potential technique for an efficient elimination of low concentration organic dyes. However, the development of an effective electrochemical reactor with a very small volume is needed to improve the performance of electrochemical oxidation (to increase the average current efficiency). Further work is also needed to characterize surface chemistry of the sawdust after

regeneration, optimize the various parameters and reduce the regeneration charge passed per gram of sawdust.

CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this manuscript.

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