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Electrochemical degradation of methylene blue using electrodes of stainless steel net coated with single-walled carbon nanotubes

Yao Xing Liu[a,b,*], Zai Yi Liao[a], Xiao Yun Wu[a], Chun Ju Zhao[a], Yu Xin Lei[a], Dao Bin Ji[a]

[a] *Department of Environment Engineering, College of Hydraulic and Environmental Engineering, China Three Gorges University, Hubei Province, Yichang 443002, China, Tel./Fax: +86 0717 6392318; email: yxliu19830218@ctgu.edu.cn*
[b] *Hubei Provincial Collaborative Innovation Center for Water Security, Wuhan, 430070, China*

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ABSTRACT

In the present study, an electrochemical technique was used for degradation of methylene blue (MB) from aqueous solutions on a laboratory scale using a homemade plexiglass cell. Electrodes comprising a stainless steel net coated with single-walled carbon nanotubes (SWCNTs@SSN) were used as both the anode and the cathode. The effects of various parameters, including the supporting electrolyte type, solution pH, electrochemical time, applied voltage, electrolyte concentration, and initial MB concentration, on the MB decolorization efficiencies were investigated. The percentage of chemical oxygen demand (COD) removal was also investigated. The supporting electrolyte type, applied voltage, and electrolyte concentration have obvious effects on the degradation of MB, and the solution pH and initial MB concentration have minor effects on the degradation. As the initial MB concentration was varied from 100 to 400 mg L^{-1}, the percentages of COD removal of all the investigated solutions were higher than 86.0%, and the general current efficiency (GCE) increased from 1.79 to 6.38%. The electrical energy consumption (E_c) increased with increasing applied voltage. The results of the study prove that the method is a potential technique for the treatment of azo dye wastewater.

Keywords: Electrochemical; Methylene blue; Electrode; Stainless steel net; Single-walled carbon nanotubes (SWCNTs); Dyestuff wastewater

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1. Introduction

Dye wastewater is one source of environmental pollution. The discharge of this wastewater can cause esthetic problems, and it also damages the quality of the receiving body of water. In addition, since some dyes are considered carcinogens or mutagens [1], the dye wastewater must be treated before it is discharged.

Many methods have been used to treat dye wastewater, including biological treatment [2], adsorption [3,4], coagulation [5], advanced oxidation process [6–8], chemical oxidation [9,10], etc. However, conventional biological treatments are usually ineffective for dye degradation because of the presence of aromatic rings, and high biochemical stability. The method of adsorption requires regular regeneration of adsorbents, and chemical oxidation and advanced oxidation processes have high costs and present some operational problems.

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*Corresponding author.

Hence, it is necessary to find a new method that is not only highly efficient but also cost-efficient.

In recent years, electrochemical oxidation has attracted more attention owing to its wide application, simple equipment, easy operation, high efficiency, and environmental compatibility [11,12]. It is a relatively new trend in dye wastewater treatment and various electrode materials have been tested, such as boron-doped diamond (BDD) [13–15], PbO_2 [16,17], SnO_2 [16,18,19], Pt [20], and mixed metal oxide (RuO_2) [21]. However, these electrodes are only used on a laboratory scale for research on the degradation mechanism. In addition, SnO_2 and PbO_2 have the common drawbacks of short service life and the release of toxic ions because they can react with hydroxyl radicals ($HO^{\bullet}$) generated when a voltage is applied. So far, there have not been reports of studies on field-scale use of these electrodes. Because these electrodes are either expensive or their production processes are complicated, they are not suitable for use on the field level.

Azo dyes are the largest group of dyes, and they are widely used in the textile industry [22]. In our previous work [23,24], a stainless steel net coated with single-walled carbon nanotubes (SWCNTs@SSN) was used to remove hexavalent chromium and lead ions, and it showed excellent chemical stability and conductivity. In this present work, SWCNTs@SSN was used as both anode and cathode, and methylene blue (MB) was chosen as the representative dye to investigate the feasibility of using SWCNTs@SSN as electrodes to degrade azo dye from an aqueous solution. The key parameters of the degradation process, such as the electrolyte type, initial solution pH, electrolyte concentration, applied voltage, and initial MB concentration, were investigated.

2. Materials and methods

2.1. Materials and reagents

Analytical grade Na_2SO_4, NaCl, H_2SO_4, NaOH, and methylene blue ($C_{16}H_{18}ClN_3S\cdot 3H_2O$) were used (Sinopharm Chemical Reagent Co., Shanghai, China). Ultrapure water (18.2 MΩ cm) was obtained from a water purification system (Unique-R30, Research Scientific Instruments Co., China). SWCNTs were purchased from the Institute of Solid State Physics of the Chinese Academy of Sciences (Hefei, China). The SSN (AISI304), which had 40 meshes and a wire diameter of 0.2 mm, was obtained commercially. A stock MB solution (2000 mg L^{-1}) was prepared by dissolving MB in ultrapure water. Working MB-containing solutions of desired concentrations were obtained by diluting the stock solution with ultrapure water.

2.2. The electrolysis cell and preparation of the SWCNTs@SSN electrodes

The experiments were carried out on a laboratory scale in a homemade plexiglass cell. The dimensions of the electrolysis cell were 9.0 × 6.0 × 5.0 cm. An illustration of the experimental setup is shown in Fig. 1. The applied voltage was supplied by an electrochemical analytical instrument (CHI630B, Shanghai Chenhua Instrument Co., China). A saturated calomel electrode (SCE) was selected as the reference electrode, and two SWCNTs@SSN electrodes were used as the cathode and the anode. A magnetic stirrer (JBZ-14, Shanghai Kang-Yi Instruments Co., China) with a rotational speed of 300 rpm was used to maintain the electrolytic homogeneity of the solutions.

The preparation of SWCNTs@SSN is described in our previous work [23]. To reiterate, the SSN was cut into 7.5 × 5.0 cm pieces, and the pieces were rinsed with a sulfuric acid solution (1:10, v/v) and ultrapure water to remove the dirt on the surface. The pieces were then dried at 40℃ to obtain clean SSN. One gram of the SWCNTs, which had a pore size of 1–2 nm, length of 5–30 μm, and specific surface area of more than 380 $m^2 g^{-1}$, was refluxed in a mixed solution of nitric and sulfuric acids (1:3, v/v) at 130℃ for 45 min. The pH of the mixture was then changed to neutral by washing with ultrapure water. The mixture was centrifuged and the SWCNTs were dried in a constant-temperature drying oven (DHG-9146A, Shanghai Jing-hong Instrument Co., China). Next, the treated SWCNTs were ultrasonically dispersed in 200 mL of *N*,*N*-dimethylformamide, resulting in a suspended solution of the SWCNTs. Two clean pieces of the SSN were immersed in parallel into this

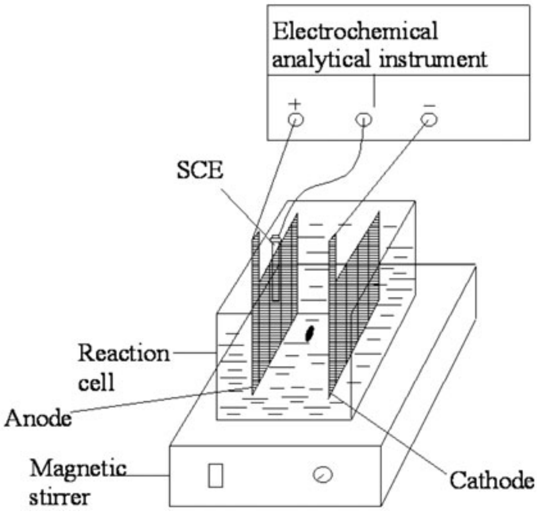

Fig. 1. Illustration of the experimental set up.

suspension. A direct current (dc) voltage of 40 V was applied between the two pieces of SSN. A deposit of SWCNTs appeared on the SSN used as the anode. The SSN anode was then removed from the suspension and dried at 120℃ to evaporate the solvent. Thus, a SWCNTs@SSN electrode was obtained.

2.3. Electrochemical treatment

The pH of the MB solution, determined with a pH meter (UB-7, Denver Instrument, USA), was adjusted using diluted NaOH and H_2SO_4 solutions. The effects of the experimental parameters, including the supporting electrolyte type, solution pH, electrochemical time, applied voltage, electrolyte concentration, and initial MB concentration on the decolorization efficiency, were studied. The volume of the solution used was 240 mL, the distance between the anode and cathode was 3 cm, and the MB concentration was 300 $\mathrm{mg\,L^{-1}}$. Samples of the MB solution were collected at designated intervals for analysis of the MB concentration and chemical oxygen demand (COD) removal.

2.4. Analytical methods

A UV–vis spectrophotometer (TU1810, Beijing Puxi, China) was used to monitor the MB concentration at 662 nm. The COD measurements via the dichromate titration technique ($\mathrm{COD_{cr}}$) were obtained using the standard method [25]. The decolorization efficiency (η) was calculated by the change in absorbance using Eq. (1):

$$\eta = \frac{A_0 - A_t}{A_0} \times 100\% \tag{1}$$

where A_0 and A_t are the absorbance value of MB initially and at a given time t, respectively. The specific energy consumption (E_c, in $\mathrm{kWh\,m^{-3}}$) was calculated using Eq. (2):

$$E_c = \frac{UI_t}{3600\,V} \times 10^{-3} \tag{2}$$

where U is the average cell voltage (V), I is the current (A), t is the treatment time (s), and V is the volume of the treated wastewater ($\mathrm{m^3}$). The general current efficiency (GCE) for the anodic oxidation of MB was calculated from the values of COD using Eq. (3) [21,26]:

$$\mathrm{GCE}\,(\%) = \frac{(\mathrm{COD_0} - \mathrm{COD_t})}{8It} FV \times 100\% \tag{3}$$

where COD_0 and COD_t are the COD initially and at time t (in g $\mathrm{O_2\,L^{-1}}$), respectively, I is the current (A), F is the Faraday constant (96,487 $\mathrm{C\,mol^{-1}}$), V is the volume of the electrolyte (L), and 8 is the oxygen equivalent mass (g $\mathrm{eq^{-1}}$).

3. Results and discussion

3.1. Effect of experimental parameters

3.1.1. Effect of electrode assembly used

The comparison of SSN and SWCNTs@SSN on the degradation MB was carried out under the experimental conditions, solution pH 7.0, applied voltage 2.0 V, current density 3.3 $\mathrm{mA\,cm^{-2}}$, NaCl concentration 1.0 $\mathrm{g\,L^{-1}}$, and MB concentration 100 $\mathrm{mg\,L^{-1}}$. After 60 min treatment, the MB decolorization efficiencies were 93.3 and 95.7% with the SSN and SWCNTs@SSN as both cathode and anode, respectively. The MB decolorization efficiencies had no obvious difference between them. However, when the SSN and SWCNTs@SSN were used more than three times, obvious oxidation phenomenon was found on the SSN surface, and no obvious change was found on SWCNTs@SSN. The results indicated that the SWCNTs@SSN was a stable electrode compared with the SSN.

3.1.2. Effect of different supporting electrolytes

In order to investigate the effect of different supporting electrolytes on MB degradation, NaCl and Na_2SO_4 were added to the MB solution, separately, as the supporting electrolyte. The results are shown in Fig. 2. Higher MB decolorization efficiency and shorter electrolysis time were obtained when NaCl was used as the electrolyte, possibly because the active chloride could be generated via direct oxidation of the chloride ion and water at the electrode surface, as follows:

$$\text{Cathode reaction: } 2H_2O \rightarrow 2OH^{\bullet} + 2H^+ + 2e^- \tag{4}$$

$$\text{Anode reaction: } 2Cl^- \rightarrow Cl_2 + 2e^- \tag{5}$$

$$\begin{aligned}\text{Bulk solution reaction: } Cl_2 + 2H_2O &\rightarrow HOCL \\ &\rightarrow H^+ + OCl^-\end{aligned} \tag{6}$$

$$\text{Bulk solution reaction: } HOCL \rightarrow H^+ + OCl^- \tag{7}$$

Thus, the presence of NaCl in the solution would lead to the production of additional oxidants such as Cl_2,

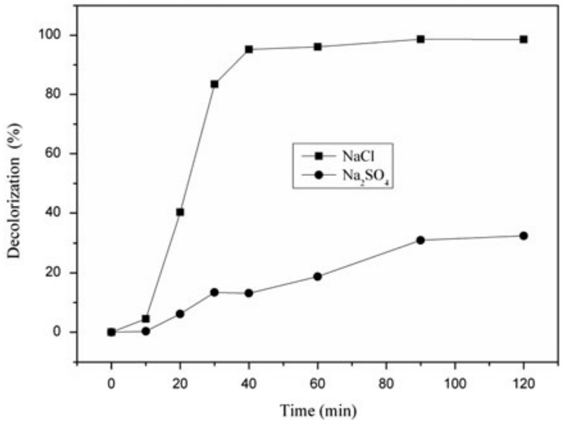

Fig. 2. Effect of different supporting electrolytes on the decolorization of MB. Experimental conditions: solution pH 7.0; applied voltage 2.0 V; current density 3.3 mA cm^{-2}; NaCl concentration 2.0 g L^{-1}; and Na$_2$SO$_4$ concentration 2.0 g L^{-1}.

HOCl, and OCl$^-$. These active chlorides favor oxidation cleavage of the azo bond, which is the most active site in the dye molecule [27].

The decolorization of MB in the presence of Na$_2$SO$_4$ is attributed to the generation of peroxodisulfates (reaction in Eq. (8)) that can oxidize organic dyes:

$$2SO_4^{2-} \rightarrow S_2O_8^{2-} + 2e^- \tag{8}$$

These peroxodisulfates are powerful oxidants and they can act as a mediator for the oxidation of organic compounds. The oxidation rate of organic compounds is significantly affected by temperature, and the oxidation rate increases with the increase in temperature [28]. In this work, since the experiment was carried out at ambient temperature, the decolorization efficiency of MB was low.

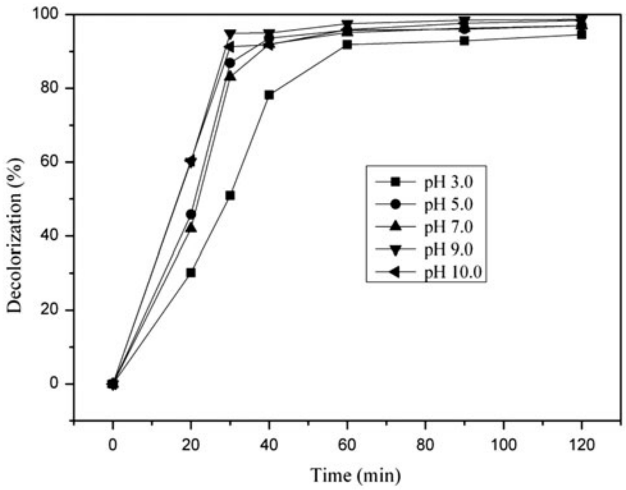

Fig. 3. Effect of solution pH and electrolysis time on the decolorization of MB. Experimental conditions: applied voltage 2.0 V; current density 3.3 mA cm^{-2}; NaCl concentration 1.0 g L^{-1}; and initial MB concentration 300 mg L^{-1}.

3.1.3. *Effect of solution pH and electrolysis time*

Solution pH is an important factor for dye wastewater treatment. There have been many reports on the effect of solution pH in anodic oxidation, but the results are diverse and even contradictory owing to the different organic structures and electrode materials used [26,29–31]. In our study, the effect of solution pH on the decolorization of MB was carried out using NaCl with a concentration of 1.0 g L^{-1} as the supporting electrolyte, an applied voltage of 2.0 V, a current density of 3.3 mA cm^{-2}, and a solution pH that varied from 3.0 to 10.0. The results in Fig. 3 indicate that the decolorization rate of MB increased with increasing solution pH. For example, the decolorization of MB increased from 50.9 to 91.3% when the solution pH was increased from 3.0 to 10.0 after 30 min of electrolysis. However, all MB solutions at various solution pH levels studied had higher decolorization efficiency after 60 min of electrolysis. The pH of the MB solution prepared by dissolving MB in ultrapure water was about 7.0, which was a suitable value for MB decolorization according to the results of Fig. 3. Thus, the prepared MB solution was used directly with no further adjustments in the following study.

The effect of electrolysis time on the decolorization efficiency of MB was analyzed over a period of 0–120 min, with the same operating conditions for the analysis of the effect of solution pH. We observed in Fig. 3 that MB decolorization showed a sharp increase during the first 30 min at the investigated pH levels, and most decolorization of MB reached equilibrium in less than 60 min. Thus, an electrolysis time of 60 min was adopted in the following study.

3.1.4. *Effect of applied voltage*

Fig. 4 shows the progress in MB decolorization with time at five different applied voltages of 1.0, 1.5, 2.0, 3.0, and 4.0 V, with an SCE as the reference electrode and corresponding current densities of 1.27, 2.75, 3.30, 3.32, and 3.35 mA cm^{-2}. The MB decolorization efficiency increased from 7.3 to 93.8% as the applied voltage was increased from 1.0 to 4.0 V after 30 min of electrolysis treatment. As expected, the

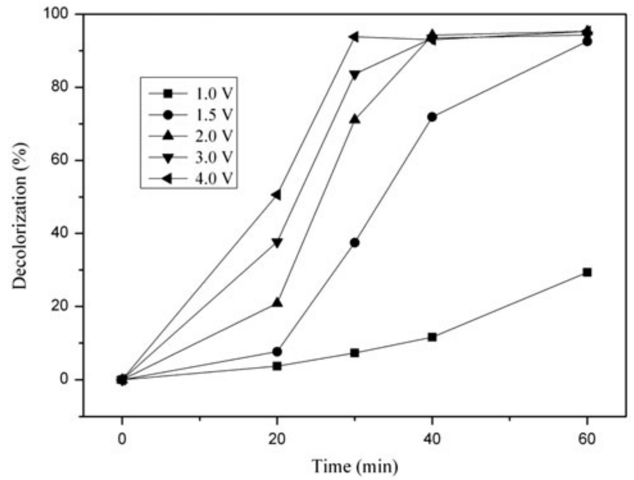

Fig. 4. Effect of applied voltage on the decolorization of MB. Experimental conditions: solution pH 7.0; NaCl concentration 1.0 g L^{-1}; electrolysis time 60 min; and initial MB concentration 300 mg L^{-1}.

larger the applied cell voltage, the higher the MB decolorization efficiency. Moreover, almost all the decolorization efficiencies were higher than 92.6% after 60 min at all applied voltages except 1.0 V. Taking both decolorization efficiency and energy consumption into account, we determined that 2.0 V was the optimized applied voltage.

3.1.5. Effect of supporting electrolyte concentration

The effect of supporting electrolyte concentration on the decolorization of MB was investigated for various MB concentrations between 0.5 and 5.0 g L^{-1}. The results (Fig. 5) showed that the efficiency of MB decolorization increased from 37.3 to 95.2% when the concentration of NaCl was increased from 0.5 to 2.0 g L^{-1} after 40 min of electrolysis. However, there was no additional effect of NaCl when its concentration was increased beyond 2.0 g L^{-1}. In other words, the optimal supporting concentration was 2.0 g L^{-1}.

3.1.6. The effect of the initial MB concentration

In view of the fact that the wastewater discharged from different industries has different concentrations of MB, it is very important, from a practical point of view, to investigate the effect of the initial concentration on the degradation efficiency of MB during the electrochemical process. Fig. 6 shows that effect as the MB concentration was varied from 100 to 400 mg L^{-1}. High MB degradation rates were obtained for all four MB concentrations studied. The initial MB concentration had no obvious effect on MB degradation. For example, when the initial MB concentrations were 100 and 400 mg L^{-1}, the surplus MB concentrations were 6.0 and 15.9 mg L^{-1}, respectively, after 60 min of electrochemical treatment; the corresponding decolorization efficiencies were 93.9 and 96.0%, respectively.

Fig. 7 shows the trend of COD concentration and removal efficiency during the electrolytic process under various experimental conditions. The results show that the COD removal efficiency decreased slightly with increasing initial MB concentration. For example, a COD removal efficiency of 86.7% was obtained when the MB concentration was 100 mg L^{-1}, and a COD removal efficiency of 84.1% was obtained when the MB concentration was 400 mg L^{-1} after 60 min of electrolysis. Although the COD degradation

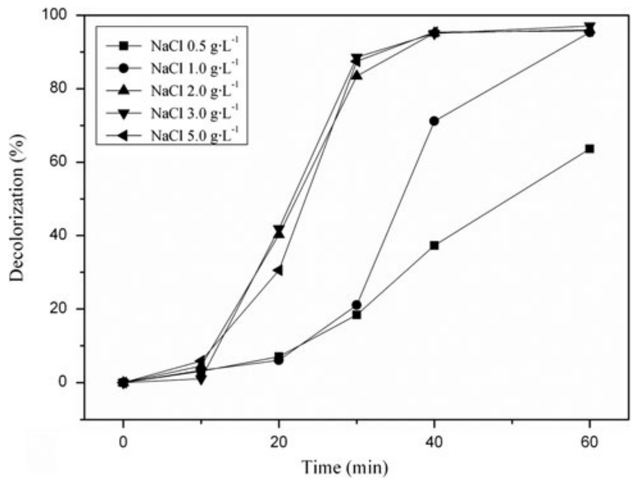

Fig. 5. Effect of supporting electrolyte concentration on the decolorization of MB. Experimental conditions: applied voltage 2.0 V; solution pH 7.0; electrolysis time 60 min; and initial MB concentration 300 mg L^{-1}.

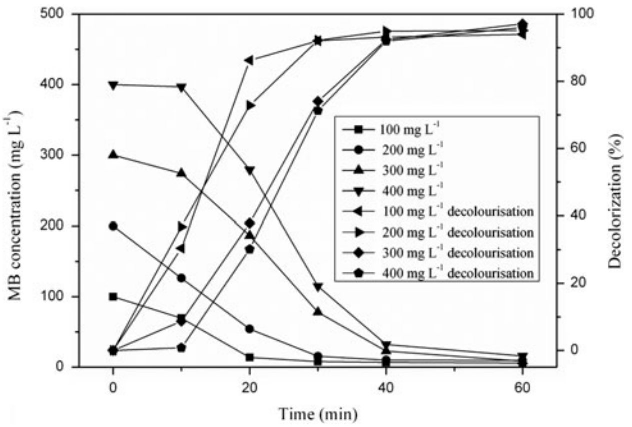

Fig. 6. Effect of initial MB concentration on the decolorization of MB. Experimental conditions: solution pH 7.0; applied voltage 2.0 V; current density 3.3 mA cm^{-2}; NaCl concentration 1.0 g L^{-1}; and electrolysis time 60 min.

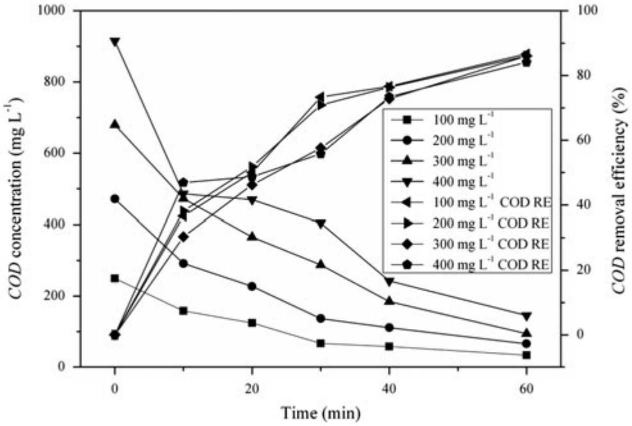

Fig. 7. COD removal in the MB solution. Experimental conditions: solution pH 7.0; applied voltage 2.0 V; current density 3.3 mA cm^{-2}; NaCl concentration 2.0 g L^{-1}; and electrolysis time 60 min.

efficiency decreased slightly for a higher initial MB concentration the absolute COD removal amount was increased, which resulted in the enhancement of GCE at the higher initial concentration. For example, when the initial MB concentration was increased from 100 to 400 mg L^{-1}, the value of GCE increased from 1.79 to 6.38, indicating that a higher initial dye concentration led to higher mineralization efficiency. A possible explanation could be that a higher MB concentration in the bulk solution resulted in not only the higher reaction rate of indirect oxidation, but it also avoided problems of mass transport for direct anodic oxidation on the SWCNTs@SSN surface [26].

From the above, higher MB decolorization was obtained using the SWCNTs@SSN as electrode, and no blue color increased phenomenon was found in treated MB solution after 2 d, the similar results were also obtained by Panizza et al. [21] and Ghauch et al. [6]. Panizza et al. compared the full UV spectra of water solution of MB from beginning to the end of electrochemical treatment; some colorless intermediates were found during the process of electrochemical treatment MB, and the intermediates can disappeared after about 15 min of the electrolysis. According to the study of Panizza et al. and Ghauch et al., the process of MB degradation was that the MB was oxidized firstly to colorless intermediates and then to carbon dioxide.

3.2. *Electrical energy consumption*

The major operating cost of wastewater treatment is associated with electrical energy consumption during the electrochemical degradation process. The value of E_c was calculated according to Eq. (2) at various applied voltages. The other operating conditions were: an MB concentration of 300 mg L^{-1}, supporting electrolyte NaCl concentration of 1 g L^{-1}, and applied voltage of 2.0 V. The results shown in Fig. 8 indicate that when all the studied MB solutions reached 95% of the decolorization, the value of E_c increased with increasing applied voltage. For example, the minimum E_c was 0.34 kWh m^{-3} at an applied voltage of 1.0 V, but it increased to 1.88 kWh m^{-3} at an applied voltage of 4.0 V because more side reactions, such as hydrogen and oxygen evolution, competed with pollutant mineralization. This scenario has already been suggested by many authors.

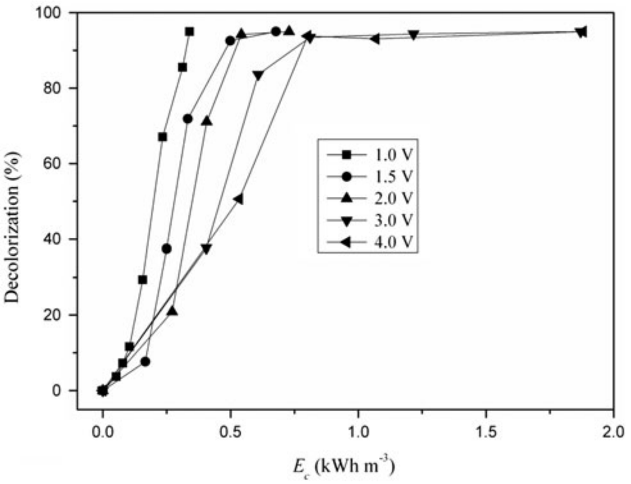

Fig. 8. Evolution of the electrical energy consumption against the decolorization efficiency during the electrochemical process at different applied voltages. Experimental conditions: solution pH 7.0; NaCl concentration 1.0 g L^{-1}; electrolysis time 60 min; and MB concentration 300 mg L^{-1}.

4. Conclusions

In the present work, the electrochemical degradation of MB was carried out using SWCNTs@SSN as the anode and cathode. The optimal conditions were 2.0 V for the applied voltage, 2.0 g L^{-1} for the electrolyte's NaCl concentration, and 60 min for the electrolysis time. Under these experimental conditions, the decolorization of MB increased with increasing applied voltage and NaCl concentration, the solution pH had a slight influence on the decolorization of MB, while the initial MB concentration had no obvious effect on the decolorization and COD removal. The electrical energy consumption increased with increasing applied voltage. These results suggest that the proposed electrochemical oxidation process is a promising approach for the degradation of MB.

Acknowledgments

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References

[1] Q. Zhuo, H. Ma, B. Wang, F. Fan, Degradation of methylene blue: Optimization of operating condition through a statistical technique and environmental estimate of the treated wastewater, J. Hazard. Mater. 153 (2008) 44–51.

[2] E. Dalentoft, P. Thulin, The use of aerobic selectors in activated sludge systems for treatment of wastewater from the pulp and paper industry, Water Sci. Technol. 35 (1997) 181–188.

[3] M. Matheswaran, T. Karunanithi, Adsorption of Chrysoidine R by using fly ash in batch process, J. Hazard. Mater. 145 (2007) 154–161.

[4] S. Preethi, A. Sivasamy, S. Sivanesan, V. Ramamurthi, G. Swaminathan, Removal of safranin basic dye from aqueous solutions by adsorption onto corncob activated carbon, Ind. Eng. Chem. Res. 45 (2006) 7627–7632.

[5] R.J. Stephenson, S.J.B. Duff, Coagulation and precipitation of a mechanical pulping effluent—I. Removal of carbon, colour and turbidity, Water Res. 30 (1996) 781–792.

[6] A. Ghauch, A.M. Tuqan, N. Kibbi, S. Geryes, Methylene blue discoloration by heated persulfate in aqueous solution, Chem. Eng. J. 213 (2012) 259–271.

[7] U. Bali, E. Catalkaya, F. Sengul, Photodegradation of Reactive Black 5, Direct Red 28 and Direct Yellow 12 using UV, UV/HO and UV/HO/Fe: A comparative study, J. Hazard. Mater. 114 (2004) 159–166.

[8] A. Aleboyeh, Y. Moussa, H. Aleboyeh, Kinetics of oxidative decolourisation of Acid Orange 7 in water by ultraviolet radiation in the presence of hydrogen peroxide, Sep. Purif. Technol. 43 (2005) 143–148.

[9] M. Muthukumar, D. Sargunamani, N. Selvakumar, J. Venkata Rao, Optimisation of ozone treatment for colour and COD removal of acid dye effluent using central composite design experiment, Dyes Pigm. 63 (2004) 127–134.

[10] K. Dutta, S. Mukhopadhyay, S. Bhattacharjee, B. Chaudhuri, Chemical oxidation of methylene blue using a Fenton-like reaction, J. Hazard. Mater. 84 (2001) 57–71.

[11] C.A. Martínez-Huitle, E. Brillas, Decontamination of wastewaters containing synthetic organic dyes by electrochemical methods: a general review, Appl. Catal. B-Environ. 87 (2009) 105–145.

[12] M. Zhou, Q. Dai, L. Lei, Long life modified lead dioxide anode for organic wastewater treatment: Electrochemical characteristics and degradation mechanism, Environ. Sci. Technol. 39 (2005) 363–370.

[13] M. Wu, G. Zhao, M. Li, L. Liu, D. Li, Applicability of boron-doped diamond electrode to the degradation of chloride-mediated and chloride-free wastewaters, J. Hazard. Mater. 163 (2009) 26–31.

[14] B. Balci, N. Oturan, R. Cherrier, M.A. Oturan, Degradation of atrazine in aqueous medium by electrocatalytically generated hydroxyl radicals. A kinetic and mechanistic study, Water Res. 43 (2009) 1924–1934.

[15] P. Cañizares, A. Gadri, J. Lobato, B. Nasr, R. Paz, M.A. Rodrigo, C. Saez, Electrochemical oxidation of azoic dyes with conductive-diamond anodes, Ind. Eng. Chem. Res. 45 (2006) 3468–3473.

[16] A.M. Polcaro, S. Palmas, F. Renoldi, M. Mascia, On the performance of Ti/SnO_2 and Ti/PbO_2 anodesin electrochemical degradation of 2-chlorophenolfor wastewater treatment, J. Appl. Electrochem. 29 (1999) 147–151.

[17] N.B. Tahar, A. Savall, Electrochemical degradation of phenol in aqueous solution on bismuth doped lead dioxide: a comparison of the activities of various electrode formulations, J. Appl. Electrochem. 29 (1999) 277–283.

[18] C. Comninellis, C. Pulgarin, Electrochemical oxidation of phenol for wastewater treatment using SnO_2 anodes, J. Appl. Electrochem. 23 (1993) 108–112.

[19] S. Stucki, R. Kötz, B. Carcer, W. Suter, Electrochemical waste water treatment using high overvoltage anodes Part II: Anode performance and applications, J. Appl. Electrochem. 21 (1991) 99–104.

[20] M. Hamza, R. Abdelhedi, E. Brillas, I. Sirés, Comparative electrochemical degradation of the triphenylmethane dye Methyl Violet with boron-doped diamond and Pt anodes, J. Electroanal. Chem. 627 (2009) 41–50.

[21] M. Panizza, A. Barbucci, R. Ricotti, G. Cerisola, Electrochemical degradation of methylene blue, Sep. Purif. Technol. 54 (2007) 382–387.

[22] C. Máximo, M.T.P. Amorim, M. Costa-Ferreira, Biotransformation of industrial reactive azo dyes by Geotrichum sp. CCMI, Enzyme Microb. Tech. 32 (2003) (1019) 145–151.

[23] Y. Liu, D. Yuan, J. Yan, Q. Li, T. Ouyang, Electrochemical removal of chromium from aqueous solutions using electrodes of stainless steel nets coated with single wall carbon nanotubes, J. Hazard. Mater. 186 (2011) 473–480.

[24] Y. Liu, J. Yan, D. Yuan, Q. Li, X. Wu, The study of lead removal from aqueous solution using an electrochemical method with a stainless steel net electrode coated with single wall carbon nanotubes, Chem. Eng. J. 218 (2013) 81–88.

[25] S.E.P.A. China, Monitoring and Analysis Methods of Water and Wastewater, 4th ed. China Environmental Science Press, Beijing, 2002.

[26] M. Zhou, H. Särkkä, M. Sillanpää, A comparative experimental study on methyl orange degradation by electrochemical oxidation on BDD and MMO electrodes, Sep. Purif. Technol. 78 (2011) 290–297.

[27] S. Song, J. Fan, Z. He, L. Zhan, Z. Liu, J. Chen, X. Xu, Electrochemical degradation of azo dye C.I. Reactive Red 195 by anodic oxidation on Ti/SnO_2–Sb/PbO_2 electrodes, Electrochim. Acta 55 (2010) 3606–3613.

[28] D.A. House, Kinetics and mechanism of oxidations by peroxydisulfate, Chemical Rev. 62 (1962) 185–203.

[29] X. Chen, G. Chen, Anodic oxidation of Orange II on Ti/BDD electrode: Variable effects, Sep. Purif. Technol. 48 (2006) 45–49.

[30] G. Lissens, J. Pieters, M. Verhaege, L. Pinoy, W. Verstraete, Electrochemical degradation of surfactants by intermediates of water discharge at carbon-based electrodes, Electrochim. Acta 48 (2003) 1655–1663.

[31] Á. Anglada, A.M. Urtiaga, I. Ortiz, Laboratory and pilot plant scale study on the electrochemical oxidation of landfill leachate, J. Hazard. Mater. 181 (2010) 729–735.