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Research Article

Decolorization of Methyl Orange (MO) by Electrocoagulation (EC) Using Iron Electrodes Under a Magnetic Field (MF). II. Effect of Connection Mode

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Abstract: This work aims to investigate the electrocoagulation (EC) of methyl orange (MO) using iron electrodes and examine the effect of magnetic field (MF) on EC performance focusing on electrodes connection mode. Experimentally, an electrochemical cell is made in a configuration as simple as possible to impose a MF parallel to the current density and to allow an evaluation of the performance of the EC coupled to the MF. After 12 min of treatment, at pH 7.25, and with a current density of 64 A/m^2, the MO decolorization obtained by EC-MF reaches 95%; this rate is higher than that obtained by the EC alone, which does not exceed 70%. In the MF presence and under optimal conditions, the decolorization increases allowing a gain in energy consumption (36%) from 30 to 19 kWh/kg dye. The decolorization with the bipolar system in series (BP-S) reaches 98% while it reaches 64% and 74% for the mono-polar in series (MP-S) and the mono-polar in parallel (MP-P), respectively. Consequently, the BP-S is more efficient.

Keywords: Electrocoagulation (EC), Methyl Orange (MO), Magnetic Field (MF), Iron, Decolorization, Supporting Electrolyte (SE)

1. Introduction

Many industrial sectors (textiles, plastic industry, paper mills, tanneries, etc.) are heavy users of water and use soluble or pigmented synthetic dyes to color their products; but in the textile industry, the use of synthetic dyes is very important. Among the many families of synthetic dyes, azo dyes are widely used and account for 60 to 70% [1]. Clothing containing these toxic azo dyes will reach consumers, posing a real threat to the environment. On the other hand, the risk of contamination of ecosystems related to the use of this type of dye is more important. Indeed, some of these dyes, which are not removed during effluent treatment, become more toxic when dumped into streams. Regulations on the discharge of wastewater are becoming more stringent and oblige industrialists to treat their effluents. In addition, most synthetic dyes are not very biodegradable and can be a risk factor for health and harm to the environment [2, 3]. It is therefore essential to limit pollution by setting up a suitable treatment system incorporating a decolorization unit. Among all possible remediation methods, the electrocoagulation (EC) process [4] is one of the most promising processes because of its high efficiency and allows the use of less toxic products with environmental compatibility [5-14].

Following the study carried out in Part 1 of this work [1], the EC process seems to be easily ameliorable by coupling EC with magnetic field (MF) [15], which leads us to present a comparative analysis between the results of the EC alone and the EC coupled to the MF. More particularly, we focus on optimizing the operating conditions on the MO decolorization through electrodes connection mode. Liu et al. [16] and Ghernaout et al. [17, 18] showed that the MF improves the EC effectiveness. They indicated that the generated Lorentz force is sufficient to cause sedimentation of colloids with MF strength of 40 mT [16].

Generally, a two-electrode EC cell is not always well appropriate for wastewater treatment, since the dissolution rate of the metal is not suitably exploitable [19-21]. The use of large area electrodes is therefore essential [22]. Improving EC

performance is more than necessary for industrial applications or medium-scale installations [23]. This is usually done using electrochemical cells, connected in series or in parallel, alone or in combination with other types of process (hybrid processes) [24-26].

1.1. Mono-polar Connection in Parallel

Figure 1 (a) shows an EC cell with a pair of anodes placed between two parallel cathodes which are connected to a DC source. The current is shared between all the electrodes as a function of the resistance of the individual cells. This type of process requires a small difference in potential compared to the series connection [25].
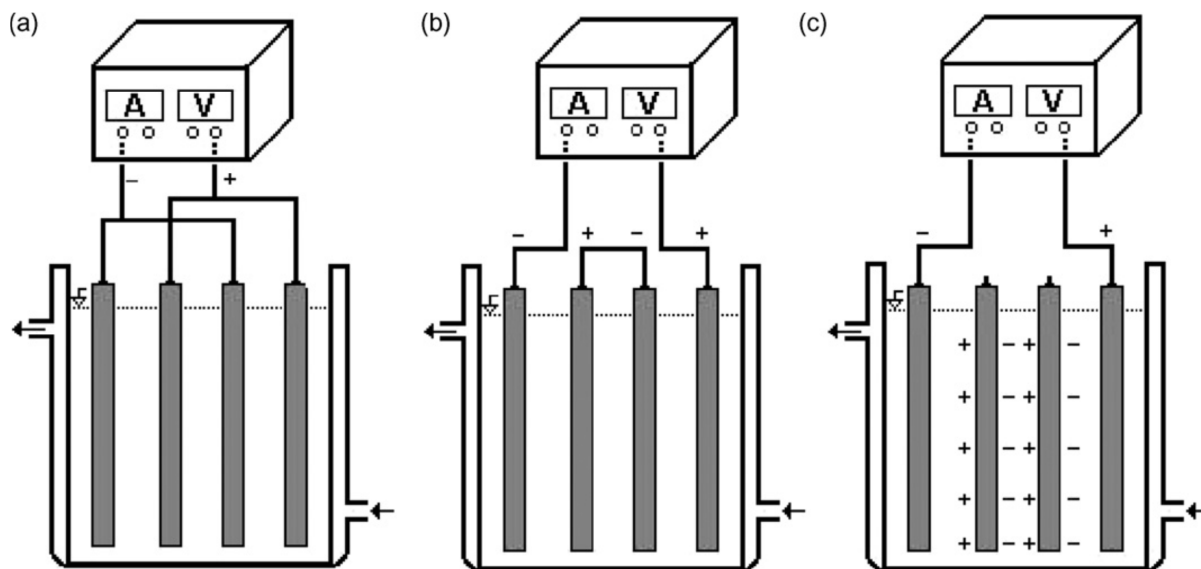

Figure 1. (a) MP-P system, (b) MP-S system, and (c) BP-S system [25].

1.2. Mono-polar Connection in Series

Figure 1(b) shows an EC cell with a pair of anodes interconnected from one to the other and does not interconnect with the outer electrodes. The difference in potential is greater because cell resistance is higher [25].

1.3. Bipolar Connection in Series

As shown in Figure 1(c), there is no electrical connection between the inner electrodes, only the outer electrodes are connected by a power supply. The outer electrodes are single pole and the inner electrodes are bipolar. This connection mode has a simple configuration which facilitates maintenance during operation [25].

Our previous study [1] realized on the optimization of the operating parameters influencing the good functioning of the mono-polar EC, made it possible to determine the optimal operating conditions. The present study focuses on improving the efficiency of the treatment through increasing the active surface by connecting the electrodes in different connection modes: bipolar in series (BP-S), mono-polar in series (MP-S), and mono-polar in parallel (MP-P). This is done in order to

compare the performance of different electrode connection modes in the presence and absence of the MF. The permanent magnets with 0.1 Tesla (T) were placed parallel to the cathode surface and the anode surface, respectively. The electrodes were connected to a direct current (DC) power supply (Elektrolyser, type Elyn1) with an ammeter and voltmeter used to controlling the current and the voltage during the EC process, respectively. The electrode plates were cleaned manually before every each run by abrasion with sand paper and by treatment with 15% HCl acid followed by washing with distilled water.

2. Experimental

2.1. Experimental Device

The EC unit was made of Plexiglas with the dimensions of 60 mm × 80 mm. There are four electrodes used, each one with dimensions of (50 mm × 25 mm × 2 mm) and the distance between them in the EC was 1 cm (Figure 2). The schematic diagram of monopolar and bipolar electrodes in series and monopolar parallel connections is shown in Figure 1.
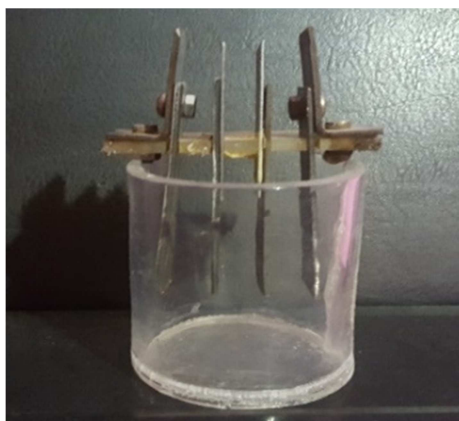
Figure 2. The apparatus of EC.

2.2. Experimental Procedure

Dye Orange III (abbreviated as methyl orange MO) is used for preparing wastewater solution by dissolving it in distilled water. The solution conductivity values were adjusted by adding NaCl as supporting electrolyte (SE) to the 200 mL solution of the synthetic wastewater. The pH of the tested solutions was measured by Hanna pH-meter and adjusted by

adding HCL 0.05 (or 1) N or NaOH 0.05 (or 1) N. At the end of the EC experiments, all samples were filtered through a 0.45 μm pore size syringe filter. The MO concentration (C_{MO}) was measured using a UV/Vis spectrophotometer (SHIMADZU UV-1700 pharma Spec) at a wavelength corresponding to the maximum absorbance of the MO (λ_{max} = 465 nm). The color removal efficiency R (%) was calculated using Eq. (1), where Abs_i and Abs_f are initial and final absorbance, respectively:

$$R(\%) = \left(\frac{Abs_i - Abs_f}{Abs_i}\right) \times 100 \qquad (1)$$

3. Results and Discussion

3.1. Influence of Electrolyte Type and NaCl Concentration

If the electric conductivity of the effluent is low, some SEs are usually added to the solution to ensure sufficient conductivity to conduct the electric current in the medium. The most commonly tested types of SE are: sodium (NaCl) or potassium (KCl) chloride, sodium (Na_2SO_4) sulfate or ($NaNO_3$) nitrate, and calcium chloride ($CaCl_2$). The results obtained are shown in Figure 3.
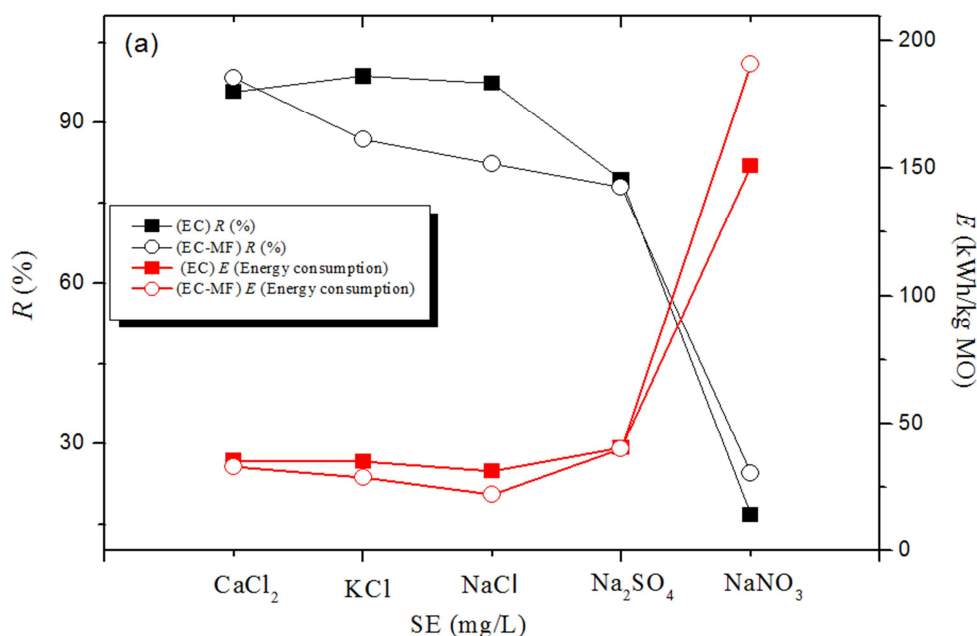

Figure 3. Effect of the SE type on the removal efficiency and consumption of energy (CMO = 15 mg/L, J = 64 A/m2, d = 1 cm, tEC = 15 min). (b) Effect of CNaCl on the removal efficiency and consumption of energy (CMO = 15 mg/L, J = 64 A/m2, d = 1 cm, tEC = 15 min).

The results obtained show that the nature of the SE has a significant influence on the rate of removal of the MO. In the absence of MF, the rates of decolorization reach: 98.47%, 86.88%, 82.34%, 77.95% and 24.44% for $CaCl_2$, KCl, NaCl, Na_2SO_4 and $NaNO_3$, respectively. In the presence of MF, the rates of decolorization reach 95.79%, 98.68%, 97.41%, 79.41% and 16.67%, for $CaCl_2$, KCl, NaCl, Na_2SO_4 and $NaNO_3$, respectively.

The consumption of energy increases considerably with the type of electrolyte used (Figure 3(a)). Indeed, the treatment with EC-MF in the presence of the NaCl as an electrolyte

allows a significant reduction in energy consumption of the order of 21 kWh/kg dye, and the treatment with the EC requires approximately 28.46 kWh/kg dye. The energy consumed during the two treatments of the MO in the presence of $CaCl_2$, KCl and Na_2SO_4 is greater than 35 kWh/kg dye. To avoid this adverse effect, it would be appropriate to use sodium chloride (NaCl) as a SE because the chloride ions can significantly reduce the adverse effects of other anions [27-30]. Actually, the processes of EC and EC-MF become more performant when using NaCl as SE. In order to examine the influence of the conductivity of the solution on the

decolorization rate, the NaCl concentration is varied from 0.3 to 2 g/L (Figure 3(b)). Figure 3(b) shows that the increase in NaCl concentration causes a marked decrease in energy consumed by both EC-MF and EC treatments. The concentration of 1.6 g/L NaCl allows a significant reduction in energy consumption. Indeed, for EC treatment within 15 min, the decolorization rate increases from 74% to 89.87% and the energy consumption decreases from 96 to 20 kWh/kg dye for doses of 0.3 to 1.6 g/L, respectively. A stable decolorization rate is obtained after 15 min of treatment with EC-MF, which is between 96% and 98% for doses between 0.3 and 1.6 g/L. It is found that the increase in NaCl concentration in the presence of MF does not significantly affect the rate of decolorization (Figure 3(b)).

3.2. Influence of Current Density and pH

The most important parameters in the electrochemical processes are the current density and the electrolysis time t_{EC}. The experimental results obtained by the EC treatment show that when the current density decreases from 56 to 32 A/m^2, the decolorization rate observed for 15 min is less than 60% (Figure 4(a)). The results obtained by the EC-MF treatment show that when the current density increases from 56 to 72 A/m^2, the discoloration rate is greater than 80%. When the current density decreases from 48 to 32 A/m^2, the fading rate is less than 68% for a period of 15 min (Figure 4(a)). When the current density is greater than 64 A/m^2, the decolorization rate does not change significantly. The judicious choice of initial conditions (electrolysis time t_{EC} and the current density) will limit the excessive release of hydrogen [31-35]. As the current density increases, the energy consumption decreases (Figure 4(a)). This will increase the temperature by the Joule effect and increase the rate of anodic dissolution [36].
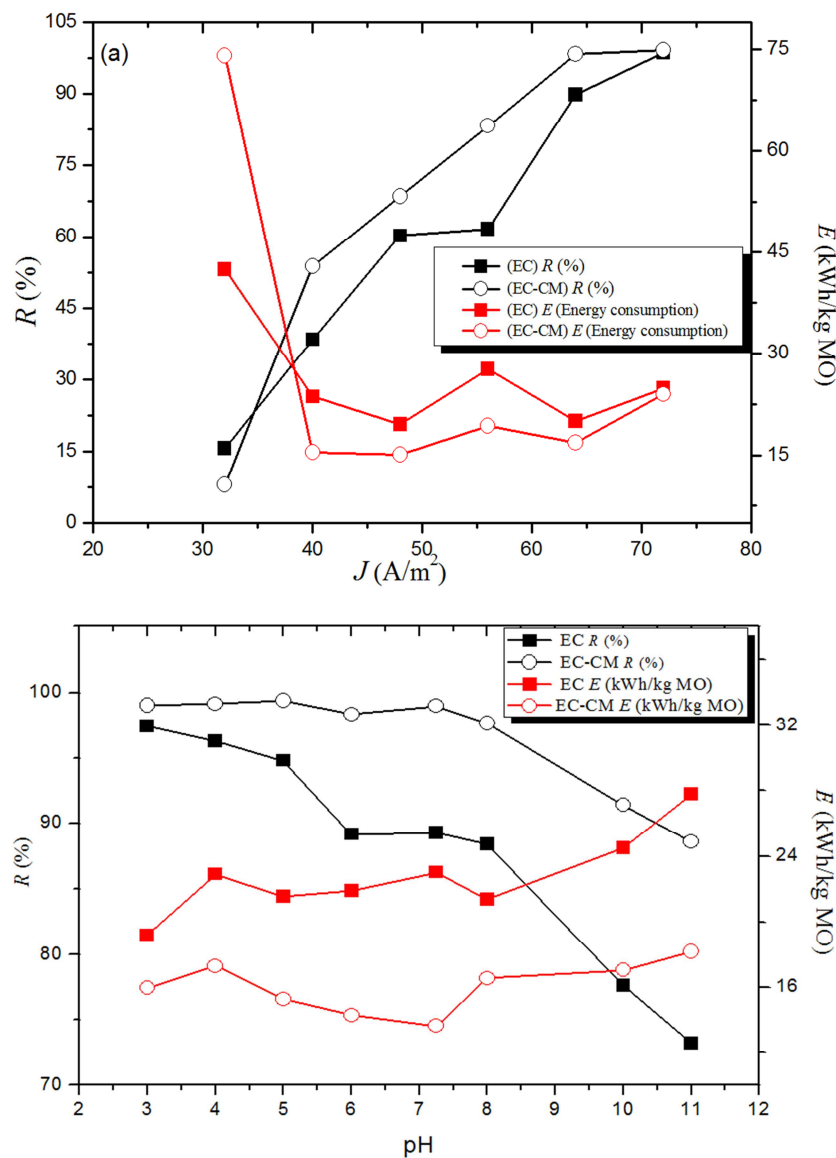
Figure 4. (a) Effect of J on the removal efficiency of MO and consumption energy (C$_{MO}$ = 15 mg/L, C$_{NaCl}$ = 1.6 g/L, d = 1 cm, t$_{EC}$ = 15 min). (b) Effect of pH on the removal efficiency and consumption energy (C$_{MO}$ = 15 mg/L, C$_{NaCl}$ = 1.6 g/L, J = 64 A/m^2, d = 1 cm).

The energy consumed during EC-MF process is less than 24 kWh/kg dye when the current density decreases from 72 to 40 A/m^2. It reaches a maximum value when the density decreases to 32 A/m^2.

To illustrate the influence of the initial pH on the decolorization kinetics by the treatment of EC and EC-MF, a series of experiments on initial pH, varying it between 3 and 12, was carried out (Figure 4 (b)). The decolorization rate is particularly important at pH 7.25, when it is of the order of 92.72% and 70.48% for EC and EC-MF, respectively.

The results show that both treatments contribute to the increase in energy consumption by increasing pH to 11 or decreasing it to 4; this increase is 28 kWh/kg dye and 18 kWh/kg dye for EC and EC-MF, respectively. The energies consumed by EC and EC-MF treatments at pH 7.25 reach 23 kWh/kg dye and 13 kWh/kg dye, respectively. It is also noted that the improvement of the EC by the MF is favored over a wide range of initial pH ranging from 3 to 10 (Figure 4(b)). The EC process is accompanied by an increase in energy consumption at the neutral pH, which favors a better efficiency of the system [37].

3.3. Effect of Inter-electrode Distance and Dye Concentration

Figure 5(a) shows that as the inter-electrode distance increases, energy consumption during EC and EC-CM treatments is increasing. When the inter-electrode distance was maintained constant at 2 cm, the EC and EC-CM treatments recorded for 12 min the highest decolorization levels, with 70% and 95.55%, respectively. When the electrodes of the EC-MF are kept at a distance of 2 cm, the treatment is more efficient with energy consumption around 19 kWh/kg dye. In addition, the efficiency of the treatment of the MO without MF decreases with high energy consumption, which is close to 30 kWh/kg dye. The energy consumption by the EC-MF is less than 19 kWh/kg dye with a distance comprised between 0.8 and 2 cm. When the inter-electrode distance is increased until 3 cm, the decolorization rate decreases to 84%. EC process is more energy consuming at a distance of 2 cm, representing 30 kWh/kg dye; and when the electrodes are kept at a distance between 0.8 and 2 cm, the energy consumption is less than 24 kWh/kg dye (Figure 5(a)).
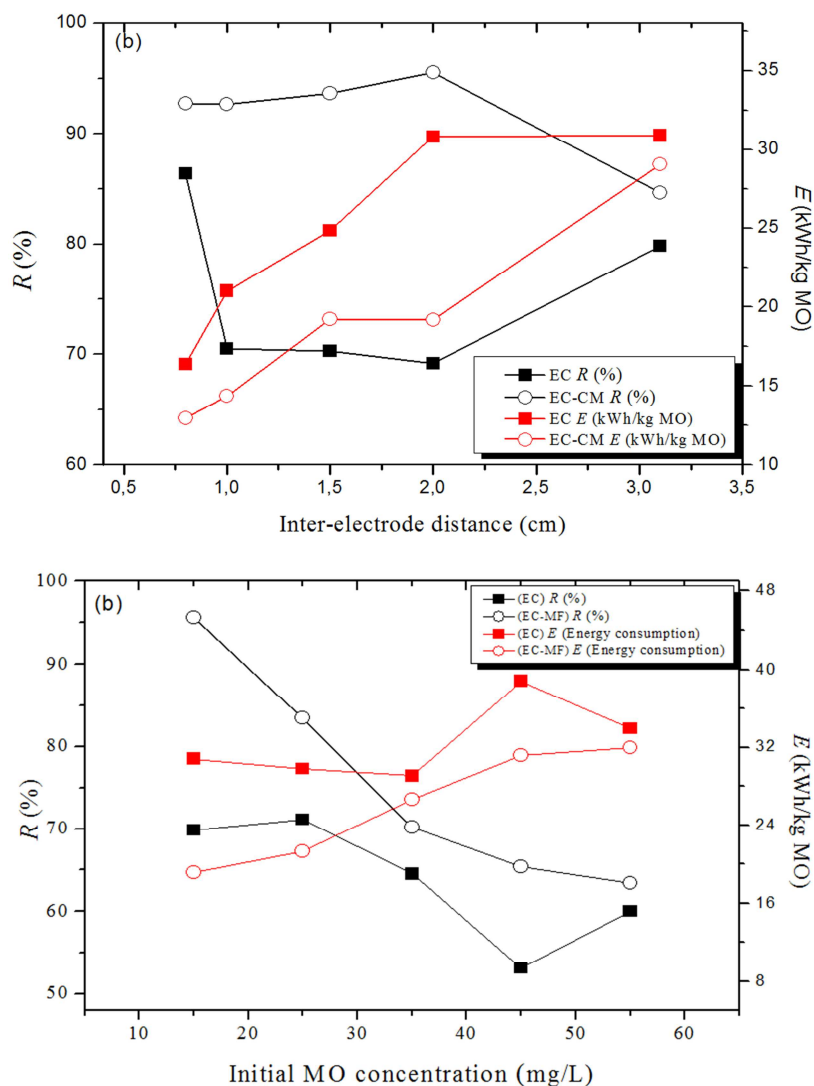
Figure 5. *(a) Effect of the inter-electrode distance on the removal efficiency and consumption of energy (C_{NaCl} = 1.6 g/L, J = 64 A/m^2, t_{EC} = 12 min). (b) Effect of the concentration on the removal efficiency and consumption of energy (C_{NaCl} = 1.6 g/L, J = 64 A/m^2, d = 2 cm, pH = 7.25, t_{EC} = 12 min).*

The energies consumed during MO treatment by EC [38-42] or EC-MF increase up to 31 and 33 kWh/kg, respectively. Then, these energies are stabilized (Figure 5(b)). It is found that the value of the maximum rate of decolorization by the treatment of EC-MF begins to decrease, when the initial concentration increases from 15 to 55 mg/L. The performance of the improvement by the MF decreases significantly to 63% for a concentration of 55 mg/L with the same degree of decolorization as that achieved by the EC (Figure 5(b)).

Table 1 presents a comparison between EC and EC-MF in terms of efficiencies.

Table 1. Comparison between EC and EC-MF in terms of performances.

| Process	C_{NaCl} = 1.6 g/L	J = 64 A/m^2	pH = 7.25	d = 2 cm
EC	82.84%; t_{EC} = 15 min	89.87%; t_{EC} = 15 min	89%; t_{EC} = 15 min	83%; t_{EC} = 15min
EC-MF	97.41%; t_{EC} = 15 min	98.37%; t_{EC} = 15min	92.72%; t_{EC} = 12 min	95.55%; t_{EC} = 12 min

3.4. Influence of the Type of Electrical Connection

Figure 6 shows the effect of the different connection modes on the MO decolorization as a function of time. The BP-S system ensures a removal rate of 98%; while the decolorization reaches 64% and 74% for the mode MP-S and MP-P, respectively. Coupling the different connection modes with the MF shows that the decolorization reaches a maximum of 95% for the BP-S mode; while it arrives at 57% and 69% for MP-S and MP-P, respectively (Figure 7). It is found that the MF disturbs the decolorization of the MO with the connection modes MP-S and MP-P. In the absence of the MF, the BP-S system performs better than the other two systems. The study of the treatment of textile wastewaters using various connection modes shows that the MP-P mode is more appropriate than the other modes, to reduce the chemical oxygen demand value at more than 54% in a neutral medium [24]. Daneshvar et al. [43] showed that azo dye treatment by MP-S mode is more efficient than MP-P mode at 20 A/m^2 density and BP-S elimination efficiency is greater than 90% for density of 90 A/m^2.
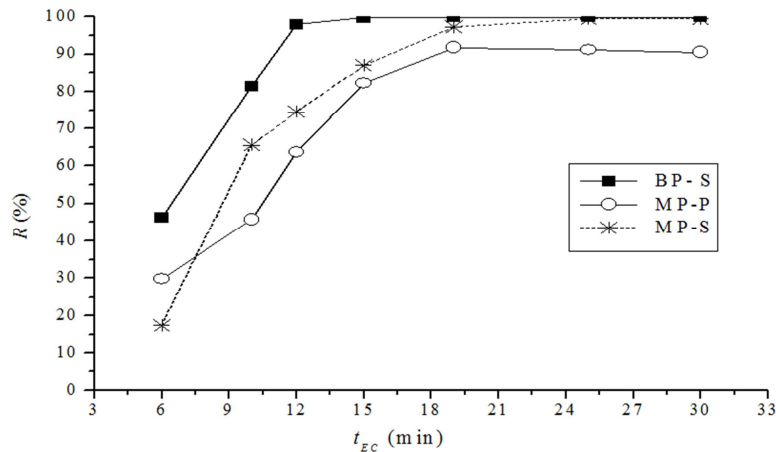
**Figure 6.** *Influence of connection mode on decolorization rate in the absence of MF. C_{NaCl} = 1.6 g/L, d_i = 1 cm, J = 64 A/m^2, pH = 7.25, w = 279 rpm, C_{MO} = 30 mg/L.*
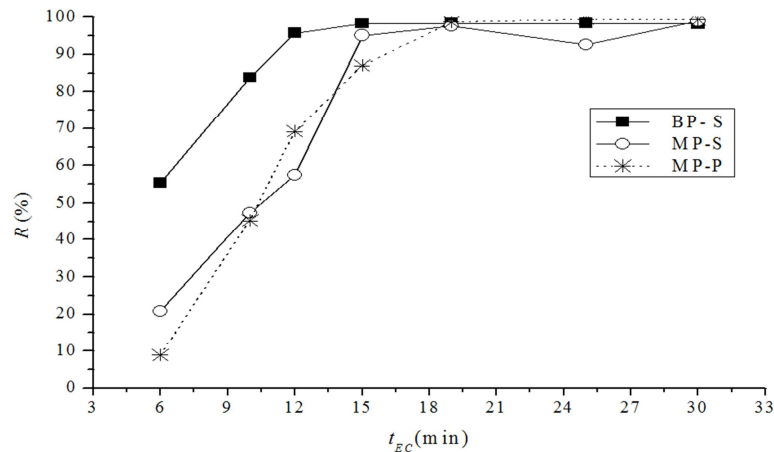
Figure 7. *Influence of connection mode on decolorization rate in the presence of MF. C_{NaCl} = 1.6 g/L, d_i = 1 cm, J = 64 A/m^2, pH = 7.25, w = 279 rpm, C_{MO} 30 mg/L.*

Figure 8 shows that the energy consumption varies with the type of the connection. Indeed, it is found that coupling the BP-S system without and with MF consumes the same energy of a value of 29 kWh/kg dye, while the MP-S and MP-P systems consume the lowest energies, reaching 18 and 14 kWh/kg dye, respectively.
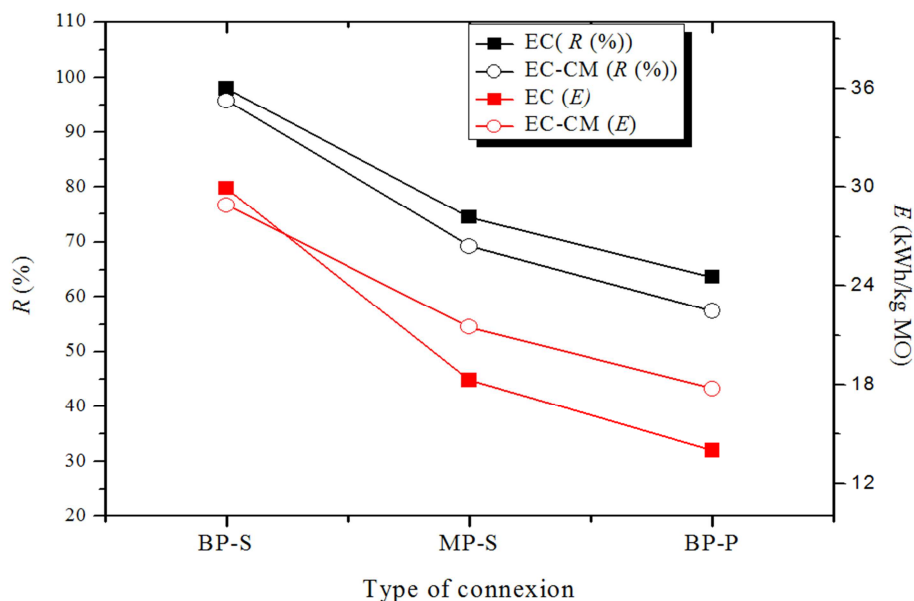
Figure 8. *Evolution of decolorization and energy consumption as a function of connection mode. C_{NaCl} = 1.6 g/L, d_i = 1 cm, J = 64 A/m^2, pH = 7.25, t_{EC} = 12 min.*

4. Conclusion

Coupling EC and MF has made it possible to obtain significant results. Indeed, it is found that during the combined treatment for 12 min, the decolorization rate reaches 91%, while the rate during the treatment with the EC reaches only 70% for a current density 64 A/m^2, a salinity of 1.6 g/L and pH = 7.25. The energy consumption during the EC-MF process is lower compared with the EC alone. The EC-MF process could reduce energy consumption to 36% and therefore the cost of operation. On the other hand, increasing the dye concentration is unfavorable from an energy point of view. Studying the effect of the different modes of connection on the decolorization shows that the maximum of removal rate is reached at 98% with the BP-S system without the MF.

Acknowledgements

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