



Effect of low frequency ultrasonic irradiation on the sonoelectro-Fenton degradation of cationic red X-GRL

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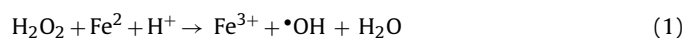
ABSTRACT

This work evaluates the effect of low frequency ultrasonic irradiation on the sonoelectro-Fenton (SEF) oxidation process in an acid aqueous medium. Ultrasonic irradiation significantly increases the H₂O₂ production rate and reduces the time needed to reach the maximum H₂O₂ concentration. In addition, ultrasonic irradiation has a considerable effect on the degradation of the cationic red X-GRL in the SEF process. A pseudo-first-order model was used to simulate the experimental results, and this revealed that the decolorization rate increased with the ultrasonic power in the SEF process. Furthermore, both TOC removal efficiency and mineralization current efficiency were greatly promoted in the SEF process compared with the electro-Fenton (EF) process. These results prove that the SEF process is a promising technology in terms of colored wastewater treatment.

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1. Introduction

The textile industry currently consumes large amounts of water and chemicals in its finishing and dyeing processes, and a large quantity of dye wastewater is being generated. This presents a serious environmental problem due to its stability when exposed to sunlight, oxidizing agents and microorganisms [1]. Therefore, there is an urgent need to develop effective methods for the treatment of this wastewater. Advanced oxidation processes (AOPs) have been proven to be efficient methods for the degradation of dyes because of the oxidizing hydroxyl radicals ($\cdot\text{OH}$) produced [2–5]. Among these AOPs, Fenton oxidation is particularly attractive because of its simplicity without the requirement of special equipment and its high efficiency in removing dye [6]. The principal reaction of the Fenton oxidation process yields hydroxyl radicals through hydrogen peroxide (H₂O₂) catalyzed by ferrous ion (Fe²⁺) in acidic solution (reaction (1)). Subsequently, the hydroxyl radical non-selectively oxidizes most organic, as well as some inorganic compounds.



Compared with traditional Fenton oxidation, electro-Fenton (EF) oxidation has attracted growing concern [7,8]. The H₂O₂ can be generated by the reduction of dissolved oxygen (reaction (2)), and Fe²⁺ can be generated by the addition of outside processes or

by the electroreduction of ferric salts in situ (reaction (3)). Hence, the H₂O₂ can be coupled with Fe²⁺ to continuously produce the Fenton's reagent for either the degradation or synthesis of organic compounds [9].



Recently, several other advanced oxidation processes have been introduced to the EF process in an effort to create more efficient systems [10–12]. Ultrasonic (US) irradiation has been reported to be an advanced oxidation process, which leads to the rapid degradation of chemical contaminants in water [13,14]. The main effects of the ultrasonic irradiation arise from acoustic cavitation, namely the formation, growth, and implosive collapse of bubbles in a liquid, which produces unusual chemical and physical environments [15]. Hence, a hybrid technique called the sonoelectro-Fenton (SEF) process, which combines sonochemistry and the EF process, has been proposed to enhance the removal of refractory organic compounds from water, and this is receiving continuously increased interest. Abdelsalam and Birkin [16] proposed a possible combination of sonoelectrochemistry and Fenton's reagent, but their method did not take advantage of the key step in the EF process. Oturan et al. [12] reported that, for the first time, the sonoelectrochemically assisted Fenton's reaction with the continuous in situ electrogeneration of the Fenton's reagent for the removal of organic pollutants in an aqueous medium. However, their work did not provide a detailed description of the improvement yielded by the ultrasonic irradiation.

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Based on the above discussion, the aim of this paper is to investigate the effects of low frequency ultrasonic irradiation (20 kHz) on the sonoelectro-Fenton degradation of cationic red X-GRL. The electrogeneration of H_2O_2 and the comparative degradation of the azo dye in the SEF were studied by comparative experiments with the EF process. The activated carbon fiber (ACF) was used as a cathode due to its characteristics of adsorption, conductivity and catalysis [17]. The azo dye cationic red X-GRL was chosen because of its wide usage in the textile, plastic and varnish industries, and its lack of biodegradability by conventional biological processes [18].

2. Materials and methods

2.1. Materials and chemicals

The ACF was obtained from the Anshan Senxin Activated Carbon Fiber plant (Liaoning, China), and the physical properties of the BET surface area ($1532.9 \text{ m}^2/\text{g}$), pore volume ($0.774 \text{ cm}^3/\text{g}$), average pore size (20.198 \AA) and thickness (0.6 mm) were provided by the manufacturer. A titanium mesh coated with metal oxides (RuO_2) was used as the anode.

Ultrasonic apparatus was employed in the former investigations and definite parameters were described in detail [19–21]. Briefly, it worked at an operating frequency of 20 kHz and supplied adjustable ultrasonic power of 80, 120 and 160 W measured by a calorimetric method [22].

The azo dye cationic red X-GRL used in the present work was purified from industrial cationic red X-GRL by extraction with methanol at 50°C . Its structural formula is depicted in Fig. 1.

Sodium sulfate anhydrous (Na_2SO_4 , 0.05 M) and heptahydrated ferrous sulfate ($\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$, 5 mM) were selected as the supporting electrolyte and catalyst, respectively. Sulfuric acid (H_2SO_4 , 1 M) and sodium hydroxide (NaOH , 1 M) were used to adjust the pH of the solution to optimum pH 3 according to our previous studies. The chemicals mentioned above were of analytical grade, and all solutions were prepared with ultra-pure water at room temperature.

2.2. Sonoelectro-Fenton system

The experiments were conducted in an open, undivided glass vessel, and the size of the reactor vessel was $130 \text{ mm} \times 80 \text{ mm} \times 170 \text{ mm}$. The total liquid volume in the reactor was 1 L, which was jacketed cooler to thermostat at $27 \pm 1^\circ\text{C}$. The dimension of the ACF, which was used as the cathode, was $100 \text{ mm} \times 90 \text{ mm}$. The same geometrical working area of the RuO_2/Ti mesh was used as the anode. The two electrodes were installed in parallel, and the distance between them was 50 mm. The horn of the ultrasonic apparatus was dipped into the solution between the cathode and the anode, from the top of the vessel at a depth of 20 mm. Because of the high price of pure oxygen, and to increase the competitiveness of the SEF process, we used air which was fed to the cathode at a flow rate of $450 \text{ cm}^3/\text{min}$. The current and amount of charge passed through the solution were measured and displayed continuously throughout the electrolysis using a DC

power supply (LWDQGS, PS-1505D). The electrolysis was carried out under galvanostatic conditions.

Experiments with ultrasonic irradiation alone were performed with the same setup, but without the application of current to the electrodes.

2.3. Analytical methods

The pH value of the solution was measured with a PHS-3C digital pH-meter. The samples were withdrawn from the vessel at regular time intervals and were filtrated in qualitative paper before being analyzed. Each experiment was performed in triplicate, and average values and standard deviations were obtained.

The ACF can present an adsorption effect because of the high porosity of the ACF surfaces. To eliminate the adsorption effects of the dye on the ACF cathode, the ACF cathode was immersed in a dye solution of a certain concentration (37.5 or 150 mg/L) for 24 h before it was used in the EF and SEF processes.

The hydrogen peroxide concentration was determined spectrophotometrically using a T6 UV/vis spectrophotometer (PGeneral, China) by the iodide method (detection limit of $\approx 10^{-6} \text{ M}$) [23]. The cationic red X-GRL concentration was determined spectrophotometrically at $\lambda_{\text{max}} = 530 \text{ nm}$, and the decolorization efficiency was evaluated using the formula $[1 - C/C_0] \times 100\%$, where C_0 and C were the initial concentration before the reaction and the concentration during the reaction, respectively.

The mineralization of the initial and electrolyzed cationic red X-GRL samples was monitored for the abatement of the total organic carbon (TOC), which was determined with a Shimadzu TOC-V_{CPH/CPN} total organic carbon analyzer. From this data, the mineralization current efficiency (MCE) for the treated solutions at a given time was calculated by using the following equation:

$$\text{MCE} = \frac{\Delta(\text{TOC})_{\text{exp}}}{\Delta(\text{TOC})_{\text{theor}}} \times 100 \quad (4)$$

where $\Delta(\text{TOC})_{\text{exp}}$ is the experimental TOC removal, and $\Delta(\text{TOC})_{\text{theor}}$ is the theoretical TOC abatement, assuming that the applied charge (=current \times time) is only consumed in the mineralization reaction.

3. Results and discussion

3.1. Hydrogen peroxide production with different current densities in the absence ultrasonic irradiation

The formation of hydrogen peroxide by reducing oxygen at the cathode is crucial to the EF process. The ACF is an effective cathode material, which generates more H_2O_2 than other cathodes [7]. Hence, an investigation of H_2O_2 production in our system was carried out in the absence of Fe^{2+} . A series of experiments was performed to investigate the effect of the applied current on the generation of hydrogen peroxide by using different current densities.

As can be seen from Fig. 2, during the initial 120 min of electrolysis, the H_2O_2 concentration presented a gradual rise of all of the current densities, and after approximately 120 min, it reached an almost constant concentration. This can be ascribed to the decomposition of hydrogen peroxide (reactions (5)–(7)) [7,24,25], even though no Fe^{2+} was added to the system. However, the H_2O_2 concentration did not increase with the applied current densities. The maximum H_2O_2 concentration was obtained at applied current density of 8.89 mA/cm^2 , but it decreased when the applied current density increased to 13.33 mA/cm^2 . For the larger current density (13.33 mA/cm^2), a higher voltage must be supplied to the system, which quickens the H_2O_2 decomposition either at the anode (reactions (6) and (7)) or in the medium directly (reaction (5)) [7]. Moreover, competitive electrode reactions, such as the discharge

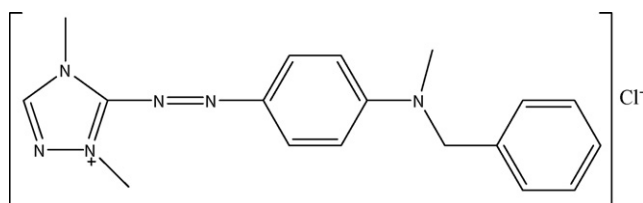


Fig. 1. Structural formula of cationic red X-GRL.

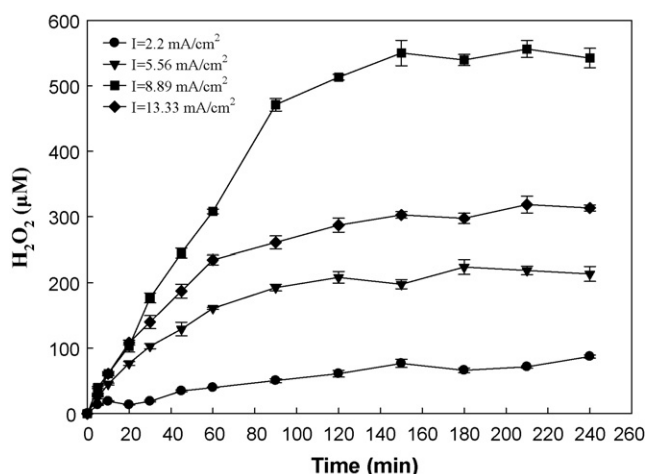


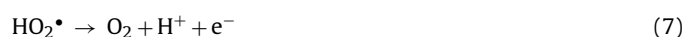
Fig. 2. H_2O_2 concentration with electrolysis time at different current densities using ACF cathode. Operating conditions: pH 3.0; Na_2SO_4 0.05 M; temperature 27 °C.

of O_2 (reaction (8)) and H_2 (reaction (9)) also inhibit the reactions for H_2O_2 generation [26]. The related reactions are as follows:

In the medium:



At the anode:



At the cathode:



3.2. Effect of ultrasonic irradiation on the generation of hydrogen peroxide

It should be noted that aqueous phase sonolysis is likely to result in the formation of hydrogen peroxide, which may be formed by the recombination of hydroxyl radicals (reactions (11) and (12)) [27–29]. Moreover, if the solution is saturated with air or oxygen, peroxy ($\cdot\text{O}_2\text{H}$) and more hydroxyl radicals are formed in the bubble, and the recombination of the former at the interface and/or in the solution bulk results in the formation of additional hydrogen peroxide (reactions (13)–(16)) [27,29]. Hence, hydrogen peroxide generated by ultrasonic irradiation alone was determined with the power of 160 W, and these results are shown in Fig. 3. As can be seen, the H_2O_2 concentration produced by ultrasonic irradiation alone appears to increase linearly with the irradiation time ($R^2 > 0.94$, irradiation time 180 min) before eventually reaching a plateau of 55.57 μM at reaction time 180 min. These results are in agreement with those reported in previous studies [30,31]. In addition, Fig. 3 also shows the H_2O_2 concentration produced at the ACF cathode coupled with the ultrasonic irradiation at 80, 120, and 160 W. The ultrasonic irradiation increased the hydrogen peroxide production rate [32] and reduced the time required to reach the maximum H_2O_2 concentration from 120 to 60 min. The accumulated H_2O_2 increased slightly from 549 μM to 581, 581, and 623 μM when the electroreduction process was coupled with the ultrasonic irradiation at 80, 120, and 160 W, respectively. At the higher output power, more H_2O_2 was accumulated. These outcomes are in agreement with a previous study by González-García [33].

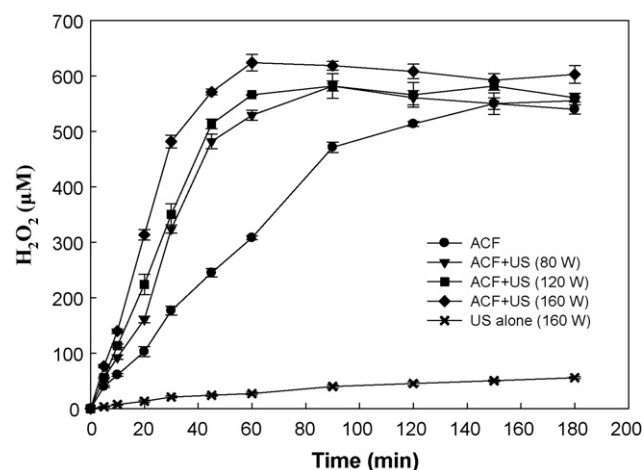
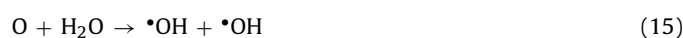


Fig. 3. Effect of ultrasonic irradiation on the H_2O_2 concentration with electrolysis time. Operating conditions: ultrasonic frequency 20 kHz; current density 8.89 mA/cm^2 ; pH 3.0; Na_2SO_4 0.05 M; temperature 27 °C.



According to these results, the ultrasonic irradiation had a positive effect on the production of hydrogen peroxide. The H_2O_2 concentration produced in electroreduction process coupled with the ultrasonic irradiation is evidently higher than their sum at the beginning of the 60 min. One possible explanation for this could be the agitation via cavitation, which is considerably effective in enhancing the mass transport of dissolved oxygen to the electrode and H_2O_2 from the cathode surface to the bulk solution [12]. Hence, the use of ultrasonic irradiation can shorten the time to the maximum value of the H_2O_2 concentration compared with the electroreduction process in the absence of ultrasonic irradiation. However, it can hardly increase the maximum value of the H_2O_2 concentration in the system, because the side reactions of H_2O_2 decomposition (reactions (6) and (7)) also accelerate simultaneously.

3.3. Effect of ultrasonic irradiation on the decolorization efficiency

Hydroxyl radicals may be generated during ultrasonic irradiation by the reaction (10). Thus the ultrasonic degradation of cationic red X-GRL was experimented with a power of 160 W. The results showed that the decolorization efficiency of cationic red X-GRL was only 2% after 180 min. The color removal barely increased with the degradation time. Therefore, dye effluent cannot be degraded by low frequency ultrasonic irradiation (20 kHz) alone under the experimental conditions. The decolorization of cationic red X-GRL in the EF and SEF processes with different ultrasonic power is shown in Fig. 4. The operating conditions used here were the optimum combination of the system parameters, which were previously determined for the electroreduction of hydrogen peroxide. Within 60 min of electrolysis, the decolorization efficiency of the cationic red X-GRL attained 74.91% in the EF process and 83.39%, 85.40% and 86.39% in the SEF process when the ultrasonic power was 80, 120 and 160 W, respectively. This suggests that ultrasonic

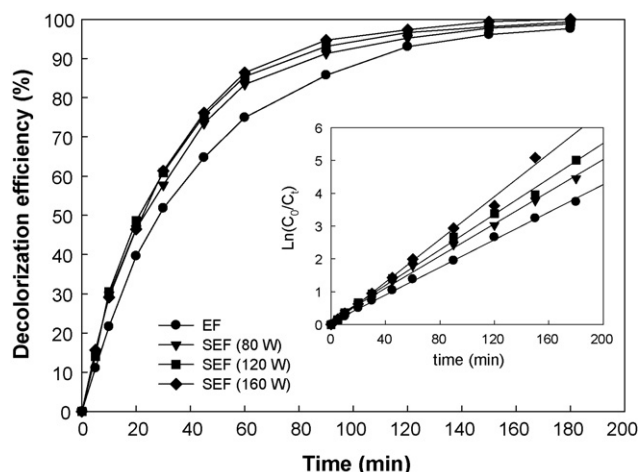


Fig. 4. The decolorization efficiency of cationic red X-GRL with electrolysis time in the EF and SEF processes. Operating conditions: ultrasonic frequency 20 kHz; dye concentration 37.5 mg/L; Fe^{2+} 5 mM; current density 8.89 mA/cm²; pH 3.0; Na_2SO_4 0.05 M; temperature 27 °C.

irradiation has a considerable effect on the color removal of cationic red X-GRL.

In order to quantitatively describe the effect of ultrasonic irradiation on the reaction kinetics of our experiments, we applied a pseudo-first-order model to characterize the degradation of the cationic red X-GRL in the EF and SEF processes. The apparent rate constant (k_{app} , min⁻¹) and regression coefficient (R^2) were calculated. For example, the apparent rate constant was 0.021 min⁻¹ ($R^2 = 0.9982$) in the EF process, whereas it was 0.0245 min⁻¹ ($R^2 = 0.996$), 0.027 ($R^2 = 0.9956$) and 0.0326 min⁻¹ ($R^2 = 0.995$) in the SEF process with the ultrasonic power of 80, 120 and 160 W, respectively. It is evident that the apparent rate constant rapidly increased when the ultrasonic irradiation was introduced into the EF system. The apparent rate constant was higher at a higher ultrasonic power, which suggests that the reaction rate of the SEF process could be accelerated with an increase in ultrasonic power. This trend did not quite agree with the results reported by Oturan et al. [12]. In their trials, when the power density was larger than 240 W/L, the worse results were achieved. In our case, the ultrasonic power density was 80, 120 and 160 W/L respectively, all of which were less than 240 W/L. Therefore, the degassing effect did not appear. On the other hand, a reactor with a different configuration and geometry may cause a discrepancy [34].

The effect of ultrasonic irradiation on dye degradation from aqueous solutions can be explained physically. The ACF cathode adsorbed several layers of compounds, such as the dye molecule, ferrous iron and ferric complexes, on the surface during a long period of time in the experiments. Thus, the electroreduction from the dissolved O_2 to H_2O_2 and the ferric reduction reaction were prevented. However, the ultrasonic irradiation can activate the cathode by providing cavitation at and near the electrode surface [35]. The continuous cleaning and activation is especially important for the ACF cathode to provide a good electrocatalytic activity. Therefore, this could mainly explain the better performance of the ACF cathode in the SEF process. However, the enhancement of dye degradation was not evident with the increase of ultrasonic power, and this may mainly be explained by the small increment of hydroxyl radicals. Firstly, the low frequency ultrasound was used in our study, which can hardly generate large amounts of hydroxyl radicals. Even when increasing the ultrasonic power, the additional amount of hydroxyl radicals was very small. Secondly, the H_2O_2 concentration produced in the electroreduction process coupled with the ultrasonic irradiation increased

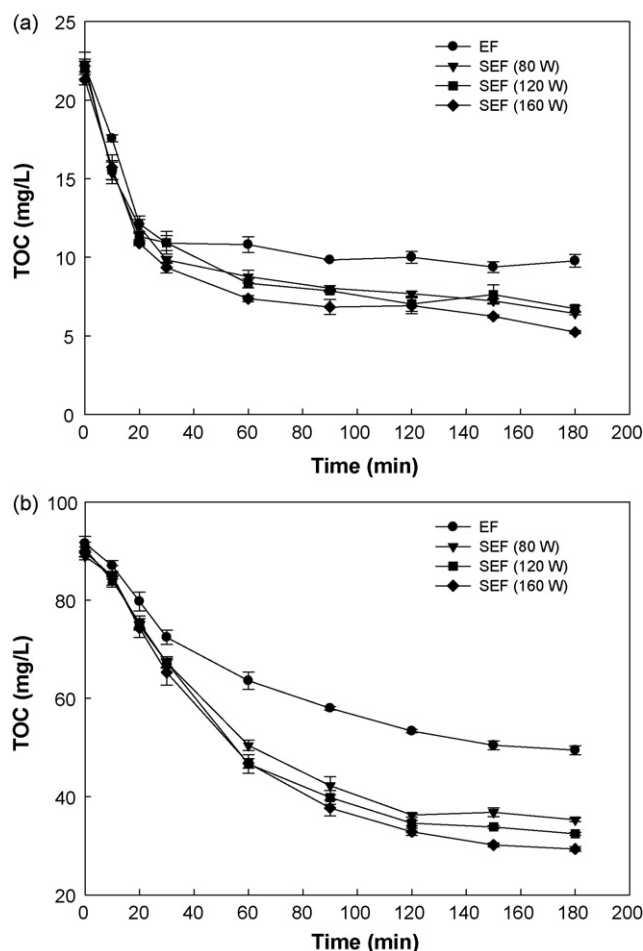


Fig. 5. The reduction of the TOC with electrolysis time in the EF and SEF processes. Operating conditions: dye concentration (a) 37.5 mg/L and (b) 150 mg/L; ultrasonic frequency 20 kHz; Fe^{2+} 5 mM; current density 8.89 mA/cm²; pH 3.0; Na_2SO_4 0.05 M; temperature 27 °C.

slightly with the ultrasonic power. In other words, even when increasing the ultrasonic power, there was no significant increase in the amount of the hydroxyl radicals yielded by the hydrogen peroxide.

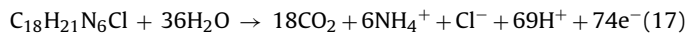
3.4. Effect of ultrasonic irradiation on the mineralization efficiency

In order to clarify the description of the effect of ultrasonic irradiation on the mineralization of cationic red X-GRL, a series of experiments was carried out at different initial dye concentrations under EF and SEF conditions. Fig. 5 shows the comparative TOC decay in the treatment with two different initial dye concentrations (37.5 and 150 mg/L). In the EF process, during the 180 min reaction time, the percentage of TOC decay was 56% and 45.99% when the initial dye concentration was 37.5 and 150 mg/L, respectively. Meanwhile in the SEF process with an ultrasonic power of 80 W, the percentage of TOC decay was 67.58% and 60.96% for the initial dye concentrations of 37.5 and 150 mg/L, respectively. This indicated that better TOC removal could be achieved in the SEF process for both of the concentrations of dye solutions. Furthermore, the mineralization extent of the cationic red X-GRL in the SEF process was significantly enhanced with the increased ultrasonic power. TOC removal efficiency of 76.52% and 68.45% could be achieved for the two concentrations after the same reaction time when a higher ultrasonic power (160 W) was applied.

Overall, the TOC removal efficiency in the SEF process was higher than in the EF process, which indicated that the SEF process performed better for mineralization. Nevertheless, in both processes, the TOC removal rate decreased dramatically after 120 min. This may be attributed to the formation of degradation products. Zhou and He [36] investigated the degradation of cationic red X-GRL by electrochemical oxidation, and degradation intermediates were detected. The main degradation intermediates identified were *N*-phenylmethylene-benzenamine, benzaldehyde and *N*-methyl-aniline. These products were further degraded to light molecule intermediates and organic acids. During the experiments, the pH value of the solution decreased to a final value of 2.3–2.5 with the reaction. It indicates the formation of organic acids. The same intermediate products were reported in other AOPs [18,37]. Hence, in the EF process, the breakage of the azo group was relatively easy, while further degradation intermediates to organic acids was rather difficult. Similarly, the same degradation intermediates could be deduced in SEF process, because ultrasound irradiation had no impact on the reaction pathway [38]. Therefore, it is not difficult to understand that the mineralization rate of the cationic red X-GRL becomes slower even with the application of ultrasonic irradiation.

3.5. Effect of ultrasonic irradiation on the mineralization current efficiency

To evaluate the various current efficiencies resulting from the ultrasonic irradiation in the SEF process, the MCE was applied. The mineralization reaction of the cationic red X-GRL can be written as follows:



The effect of the ultrasonic irradiation on the MCE for the EF and SEF processes with various ultrasonic powers was calculated from Eq. (4), assuming that the overall mineralization of the cationic red X-GRL was accomplished via the reaction. The MCEs of the two different initial dye concentrations given above were compared and are presented in Fig. 6.

As illustrated in Fig. 6b, at the higher concentration (150 mg/L), there is a rapid increase of the MCE in all trials before 60 min of reaction time. This suggests a rapid destruction of products which were more easily oxidizable than the initial compound [39]. The MCE was significantly enhanced by the ultrasonic irradiation from 43.92% in the EF process to 56.20% in the SEF process with an ultrasonic power of 160 W. In addition, the MCE gradually increased with the ultrasonic power to reach maximum values of 49.57%, 53.01%, and 56.20% for 80, 120, and 160 W, respectively. Unlike the tendency illustrated in Fig. 6b, the MCE value in Fig. 6a underwent a dramatic drop immediately at the beginning of the SEF process. As mentioned above, the progressive drops in the MCE at the long electrolysis times in the SEF process could be the consequence of the more difficult destruction of stable intermediate products.

4. Conclusions

Comparative experiments were performed to demonstrate the effect of ultrasonic irradiation applied in the EF process. The positive effect of the ultrasonic irradiation on the electrogeneration of H_2O_2 was evidenced by the increasing hydrogen peroxide production rate and the reduction of the time to a maximum value for the H_2O_2 concentration. These effects may be the result of the enhanced mass transport. In addition, the effect of the ultrasonic irradiation on the decolorization and mineralization of the cationic red X-GRL in the sonoelectro-Fenton process was also investigated. Compared with the electro-Fenton process, a higher decolorization efficiency and TOC removal efficiency for the dye solution can be achieved with ultrasonic irradiation, and these increase with ultrasonic power. This is due to the continuous cleaning and activation on the activated carbon fiber cathode. However, the formation of stable intermediate products could decrease the mineralization efficiency and current efficiency. Overall, it can be concluded that low frequency ultrasonic irradiation has a positive effect on the degradation of the dye effluent when combined with the electro-Fenton process.

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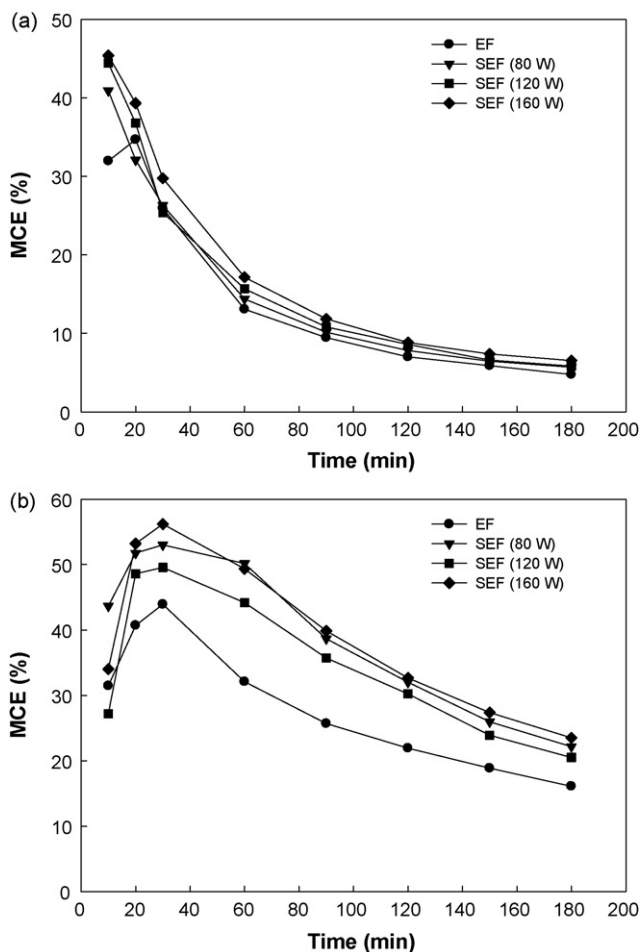


Fig. 6. Variation of MCE calculated from Eq. (4) with electrolysis time. Operating conditions: dye concentration (a) 37.5 mg/L and (b) 150 mg/L; ultrasonic frequency 20 kHz; Fe^{2+} 5 mM; current density 8.89 mA/cm²; pH 3.0; Na_2SO_4 0.05 M; temperature 27 °C.

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