

The decolorization and mineralization of orange II by microwave-assisted ball milling

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ABSTRACT

This study proposed an integrated technique of reduction coupled with an oxidation process in order to acquire simultaneously both decolorization and mineralization of orange II under the condition of microwave-assisted milling. Experimental variables of initial dye concentration, iron dosage, microwave power, solution pH and initial H₂O₂ concentration were systematically studied. Under the optimal operational parameters (100 mg/L aqueous solution of pH 3 containing 400 mg/L H₂O₂ while controlling microwave power at 400 W), the results showed that the decolorization efficiency is up to 91% after reaction for 2 min and the total organic carbon removal efficiencies were 72.7% and 80.5% at a reaction time of 10 min and 60 min, respectively. It indicated that the decolorization and mineralization of orange II were largely enhanced by the reduction of zero-valent iron in the ball milling process and the oxidation of hydroxyl radicals generated by hydrogen peroxide. It suggested that microwave-assisted ball milling technology has potential application for degradation of azo dye in wastewater.

Key words | ball milling, decolorization, microwave, mineralization, orange II

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INTRODUCTION

The wastewaters of textile, printing and dyeing, and leather processing industries generally contain various dyes, of which the largest amount is azo dye. This dye's main feature is a nitrogen–nitrogen double bond (–N=N–); it has a high degree of color and is non-degradable, particularly toxic and potentially carcinogenic (Pinheiro *et al.* 2004; Ghoneim *et al.* 2011; Cai *et al.* 2015). A variety of traditional methods such as biodegradation (Kudlich *et al.* 1996), photo-catalytic degradation (Kertész *et al.* 2014), chemical oxidation (Moon *et al.* 2011) and adsorption (Mittal *et al.* 2005) has been used to treat wastewater. However, there are shortcomings in these techniques; for example, the activated carbon method results in transferring the dyes to another place; photo-catalytic degradation treatment is energy consuming and limited in treating amount; and the conditions of the biological process in microorganism treatment are difficult to control to reach a satisfactory level (Cao *et al.* 1999).

As we all know, the Fenton method is an effective method of azo dye wastewater treatment (Kušić *et al.* 2007). Disadvantages of the Fenton process include production of large amounts of sludge and formation of a high concentration of anions in the wastewater (Moon

et al. 2011). Recently, considerable attention has been paid to zero-valent iron, because it has potential for application in groundwater treatment and site remediation (Cao *et al.* 1999; Roy *et al.* 2003; Mielczarski *et al.* 2005; Lin *et al.* 2014). Some researchers have treated dye wastewater with zero-valent iron/H₂O₂; zero-valent iron (ZVI) is used as a solid form of iron instead of iron salts to eliminate the adverse effect of Fenton method. Oh *et al.* reported that the reduction products of TNT (2,4,6-trinitrotoluene) and RDX (hexahydro-1,3,5-trinitro-1,3,5-triazine) are more rapidly and completely mineralized by a sequential iron treatment and Fenton oxidation process (Oh *et al.* 2003). Chang *et al.* reported that ZVI and UV/H₂O₂ process could make acid black 24 wastewater completely decolorized and mineralized (Chang *et al.* 2006). However, the cost of nanoscale ZVI/H₂O₂ is high and the removal efficiency of conventional ZVI/H₂O₂ is not high because ZVI is easily passivated and agglomerated. The application of microwaves in wastewater treatment has been increasing in the recent years (Zhang *et al.* 2007a, 2007b; Mao *et al.* 2015; Wang & Wang 2016). Wang *et al.* reported that a microwave synergistic electro-Fenton oxidation process can

rapidly reduce orange II dye (Wang *et al.* 2012). Yang *et al.* reported that a microwave-enhanced Fenton-like process is a promising treatment technology for pharmaceutical wastewater (Yang *et al.* 2009). Therefore, on the one hand, the formation of a new iron ball reaction layer and ZVI particles during the ball milling process makes the reaction proceed. On the other hand, compared with common heating mode, microwave heating can decrease activation energy, reduce reaction time and improve the rate of reaction, and so on (Remya & Lin 2011; Peng & Hwang 2014; Chen *et al.* 2016).

Microwave-assisted ball milling was based on a solid-liquid ball milling technique in which a ball milling machine is placed in a microwave oven (Chen *et al.* 2015). The coupling effect of mechanical milling and microwaves enables the production of certain nanocrystal materials at room temperature, such as magnetic ferrite (Chen *et al.* 2013a, 2013b).

The main objective of this study was to develop an integrated advanced reductive/oxidative process for the reduction of orange II. The factors affecting the decolorization of orange II were investigated. In addition, we discuss the degradation mechanism of orange II, the total carbon content (TOC) removal rate and the ultraviolet-visible spectrum.

EXPERIMENT

Materials

Experimental materials were orange II, iron ball ($\Phi = 1\text{--}1.5$ mm), iron powder, 30 wt% H_2O_2 , NaOH, H_2SO_4 (AR), deionized water and other reagents.

Methods

Microwave-assisted ball milling reacted in a special tetrafluoroethylene mill tank (diameter 200 mm, height 90 mm, total volume about 1,300 mL) (Chen *et al.* 2015). For the process of degradation of orange II by microwave-assisted ball milling, a certain amount of iron balls ($\Phi = 1\text{--}1.5$ mm) and orange II solution were placed into a special tetrafluoroethylene milling pot; the liquid height was recorded and the milling pot was placed into a special microwave oven. The stirring rod was adjusted to a height of 4 mm from the bottom of the pot. The microwave oven was opened and then the milling process was started. The stirring speed was 200 r/min constantly. The power of the microwave oven was 400 W. Samples were extracted at special times, then centrifuged and filtered. Before sampling, the liquid

height was kept the same as the original height. The concentration of the test solution was measured using a Shimadzu UV-2550PC UV spectrophotometer from the absorbance at $\lambda_{\text{max}} = 484$ nm, and the TOC of orange II was measured by a Shimadzu TOC-VCSN analyzer. The reaction apparatus is shown in Figure 1.

Color and TOC removal ratios (%) were calculated as follows:

$$\text{Decolorization efficiency} = \left(\frac{1 - C_t}{C_0} \right) \times 100\%$$

where C_0 and C_t are the concentration of orange II dye before and after reaction, respectively,

$$\text{TOC removal efficiency} = \left(\frac{1 - \text{TOC}_t}{\text{TOC}_0} \right) \times 100\%$$

where TOC_0 and TOC_t are the TOC values before and after reaction, respectively.

RESULTS AND DISCUSSION

The effect of orange II concentration

Figure 2 indicates that the decolorization efficiencies decrease with increasing orange II concentration. After

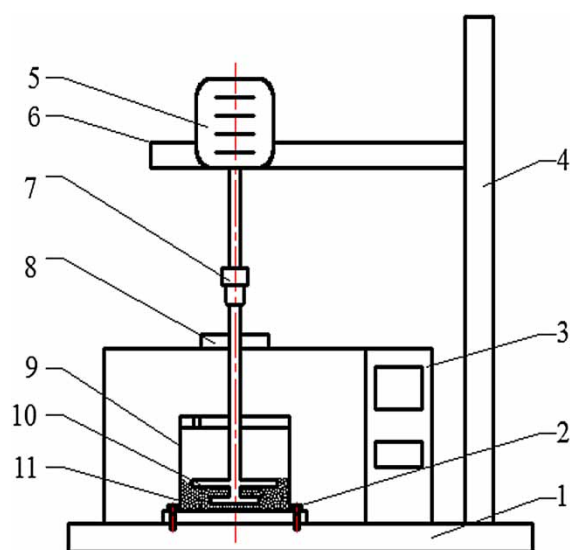


Figure 1 | Schematic diagram of microwave-assisted ball milling device. 1: substructure; 2: screw; 3: microwave oven; 4: support; 5: electric motor; 6: beam; 7: universal joint; 8: microwave oven lid; 9: milling tank; 10: stirring rod; 11: iron ball.

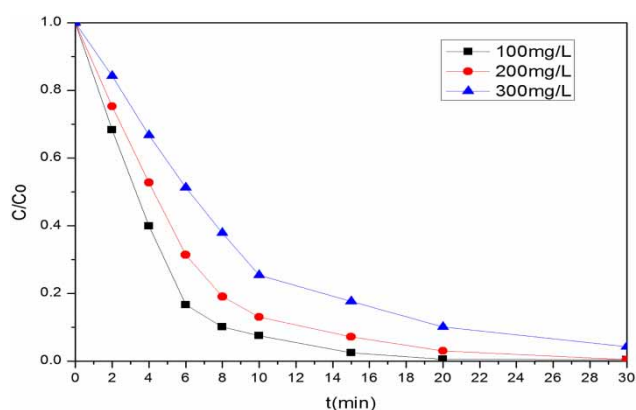
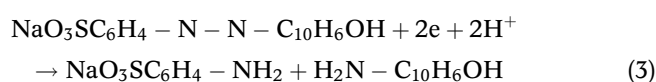
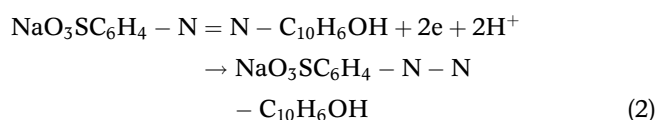


Figure 2 | Effect of the initial orange II concentrations on decolorization efficiency of orange II (pH = 7, microwave power = 400 W).

10 min, the color removal percentage was about 92.56, 87.03 and 74.71% at initial dye concentrations of 100, 200 and 300 mg/L, respectively. Because the reaction involves mainly the adsorption of dye onto the iron surface and the subsequent surface reaction, the adsorption capacity of iron is limited when the iron ball is fixed, and the size of the iron surface area is the key to decolorization: the higher the solubility of the solution, the smaller the relative reaction area. Also, the dye molecules on the iron surface area would hinder other dye molecules in the bulk solution from being adsorbed and reduced on the iron surface (Zhang *et al.* 2005; Fan *et al.* 2009). There is still a high degradation rate for a high initial concentration, because microwave-assisted ball milling produced a new layer of ZVI by the collision with the iron ball surface, so that the reaction proceeds rapidly. For the decolorization mechanism of orange II, some article pointed out (Feng *et al.* 2000; Roy *et al.* 2003; Zhang *et al.* 2005):



When the dye molecules are adsorbed on the surface of the iron ball, elemental iron as a donor of electrons combined with H^+ in the solution makes the $(-\text{N}=\text{N}-)$

double bonds break, after which they are transformed into sulfanilic acid and 1-amono-2-naphthol (Moon *et al.* 2011).

Effect of iron powder dosage

As can be seen from Figure 3, the decolorization efficiency increases with the iron powder dosage increasing. At 4 min, the color removal efficiencies were 60.01, 71.7, 86.7, and 90.7% by iron powder dose of 0, 10, 20 and 40 g/L, respectively. Figure 4 shows that TOC removal rate was 18.09% for the orange II solution at 30 min. However, TOC removal efficiency did not significantly increase until the end of the experiment (30 min) after the addition of iron powder. It is because more ZVI provides substantially more surface active sites to accelerate the initial reaction, resulting in more iron ions colliding with azo dye molecules to remove color. The decolorization was limited by the iron

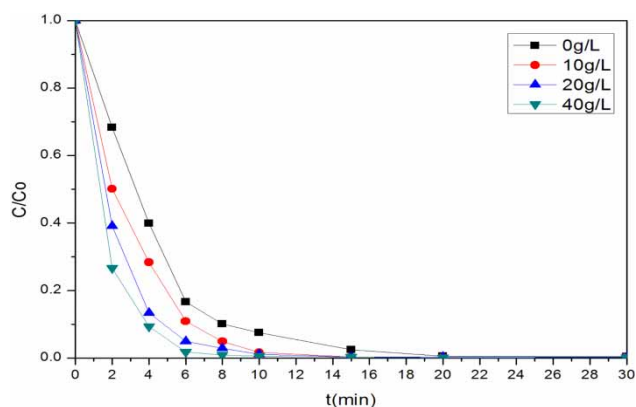


Figure 3 | Effect of ZVI dosage on the decolorization efficiency of orange II (pH = 7, microwave power = 400 W, $C_0 = 100$ mg/L).

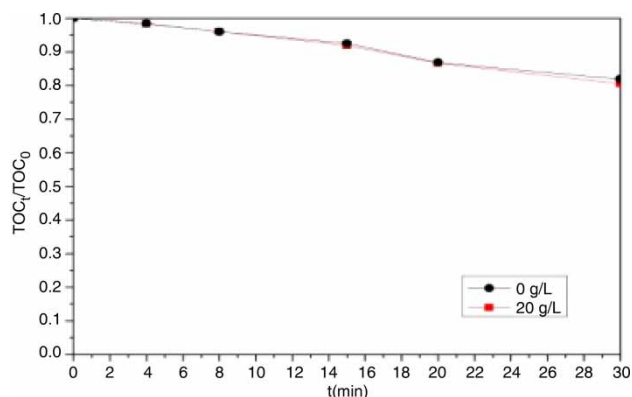


Figure 4 | Effect of ZVI dosage on the TOC removal rate of orange II (pH = 7, microwave power = 400 W, $C_0 = 100$ mg/L).

surface area and diffusion of dye molecule to the iron surface. The increased total surface area and availability of more ZVI sites are the reason for the rise in decolorization efficiency with increasing ZVI dosage (Chang *et al.* 2006; He *et al.* 2012). Moreover, due to the constant stirring and peeling actions in ball milling, iron powder was not easily agglomerated and passivated, which makes the reaction proceed rapidly. However, too much iron powder will cause waste and make the solid-liquid separation operation cumbersome.

Effect of microwave power

As shown in Figure 5, the decolorization efficiency improved with the increase of the microwave power. Within 6 min, the decolorization efficiency was found to be 53.1, 60, 83.5 and 89.8%, respectively, for the microwave power of 0, 200, 400 and 600 W. Compared with common heating mode, microwave heating can decrease activation energy, reduce reaction time and improve the rate of reaction. On the one hand, iron ball surfaces absorb microwave energies, and a lot of 'hot spots' form simultaneously; the temperature of these 'hot spots' can ordinarily reach 1,200 °C and above. On the other hand, H₂O₂ is a strong oxidant with the capability of absorbing microwaves, and can generate some active free radicals. Due to both thermal and non-thermal effects, not only the orange II can be destroyed but also the benzene rings and naphthalene rings of it can be destroyed quickly (Wang & Wang 2016; Zhang *et al.* 2007a, 2007b). It is noteworthy that the greater the power is, the higher the internal temperature of the reaction system will get, which will affect the life of the reactor.

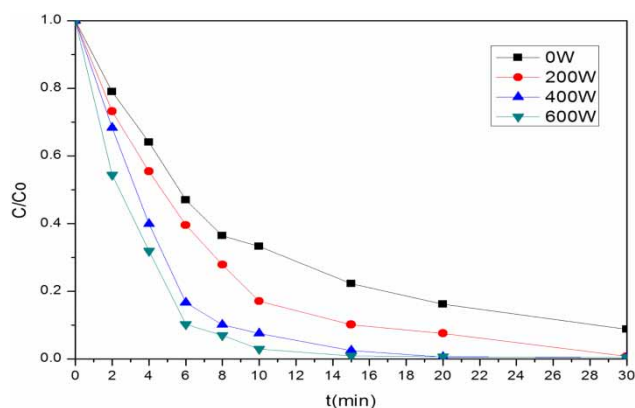
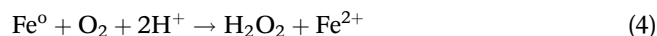


Figure 5 | Effect of microwave power on the decolorization efficiency of orange II (pH = 7, C₀ = 100 mg/L).

Effect of pH

The pH value of the solution is the most important factor that affects the decolorization of azo dye in the ZVI system. As shown in Figure 6, the decolorization efficiency of orange II decreases with increasing pH value. The decolorization process finished at 6, 6, 10 and 15 min for pH value of 2, 3, 5 and 7 with decolorization efficiency of 98.8, 98, 91.4 and 97.6%, respectively. The main reason is that the indirect oxidation of dye by strong oxidants generated by iron (Equations (4), (5) and (8)) was responsible for dye decolorization rather than the direct reductive decolorization on iron surface. The hydrogen peroxide could react with the Fe²⁺ (Equation (5), Fenton reaction) and produce strong oxidants such as hydroxyl radical. Azo chromophore can be decolorized by •OH radical attack. Besides, as the reaction proceeds, pH value becomes larger. Most iron oxides accumulate on the iron surfaces and inhibit the dye decolorization rates (Joo *et al.* 2005; Noradoun & Cheng 2005; Chang *et al.* 2009).



Effect of H₂O₂ concentration

Figure 7 shows that the decolorization efficiency gradually increases with the increase of the initial concentration of H₂O₂. At 2 min, the decolorization percentage was 71.4, 76.99, 87.2 and 87.7% for initial dye concentrations of 0, 100, 200 and 400 mg/L, respectively. When hydrogen

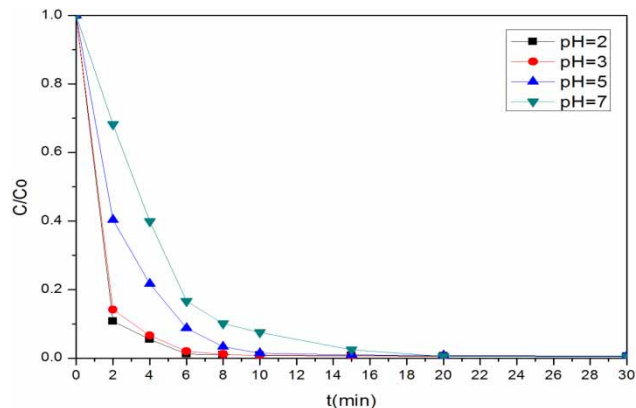


Figure 6 | Effect of solution pH on the decolorization efficiency of orange II (microwave power = 400 W, C₀ = 100 mg/L, pH = 3).

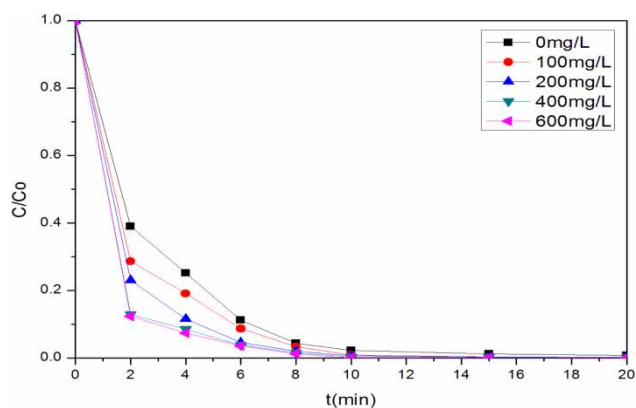
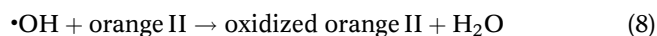
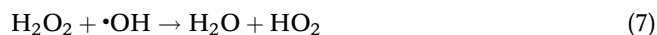
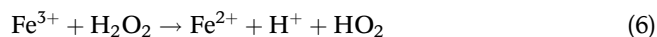


Figure 7 | Effect of the concentration of H_2O_2 on the decolorization efficiency of orange II (microwave power = 400 W, $C_0 = 300 \text{ mg/L}$, $\text{pH} = 3$).

peroxide concentration increased from 0 to 400 mg/L, orange II concentration decreased rapidly. However, when hydrogen peroxide loads exceed 400 mg/L, decolorization efficiency would slow down and level off. According to Equation (5) the ferrous ions would react with hydrogen peroxide to generate hydroxyl radicals. The amount of generated hydroxyl radicals depends on hydrogen peroxide concentration. However, the fact is that in some conditions very high H_2O_2 concentration values lead to a decrease in the final discoloration, and rate of mineralization is possibly due to the competition between these species for hydroxyl radicals (Equations (6)–(8)). Indeed, $\bullet\text{OH}$ radicals are quite non-selective, reacting with the organic matter present but also with other species (Tang & Chen 1996; Ramirez *et al.* 2009; Fu *et al.* 2010).



Ultraviolet-visible spectrum analysis

It is observed from Figure 8 that the strong absorbance peak at $\lambda_{\text{max}} = 484 \text{ nm}$ in the visible region originates from a conjugated structure formed by the azo bonds. The other two bands at 230 and 310 nm in the ultraviolet region are ascribed to the benzene rings and naphthalene rings of the dye, respectively. As the reaction time is increased from 0 to 12 min, the bands at λ_{max} decrease quickly, which

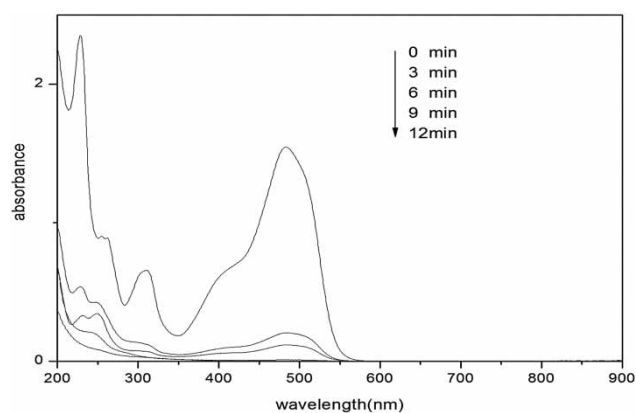


Figure 8 | UV-vis spectra of orange II during the decolorization process ($\text{pH} = 3$, $\text{H}_2\text{O}_2 = 400 \text{ mg/L}$, $C_0 = 100 \text{ mg/L}$ (diluted five times), microwave power = 400 W).

shows the cleavage of the azo bonds and the decomposition of orange II. The intensities of the bands at 230 and 310 nm decrease gradually, which reveals benzene rings and naphthalene rings were destroyed under the influence of hydroxyl radicals (Stylidi *et al.* 2003; Moon *et al.* 2011; Zhang *et al.* 2012).

The performance of decolorization and mineralization in microwave-assisted ball milling

Figure 9 shows decolorization efficiencies were 91.38% at 2 min, and the decolorization and TOC removal rates of orange II solution were approximately 100% and 72.5% in reaction time of 10 min under the optimal operational parameters, while TOC removal rate reached 80.5% at 60 min. It suggested decolorization efficiencies are faster than the TOC removal rate at 10 min. This is because $\bullet\text{OH}$ radicals attack first the $-\text{N}=\text{N}-$ double bonds,

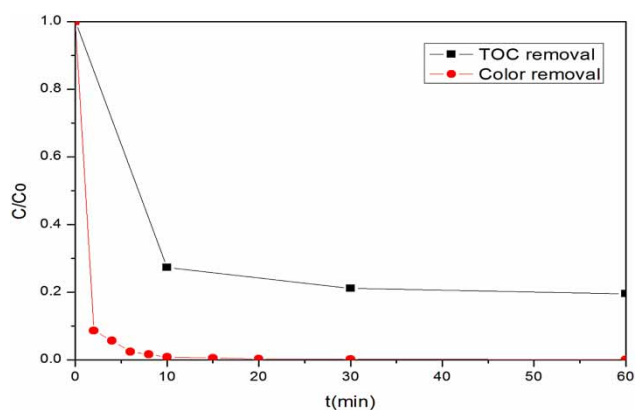


Figure 9 | The decolorization and TOC removal efficiency of orange II under microwave-assisted ball milling ($\text{pH} = 3$, $\text{H}_2\text{O}_2 = 400 \text{ mg/L}$, $C_0 = 100 \text{ mg/L}$, microwave power = 400 W).

Table 1 | Comparison of different methods

Methods	Concentration (mg/L)	Decoloration efficiency (%) (2 min)	Removal efficiency of TOC (%) (60 min)	Reference
Microwave + ball mill + H ₂ O ₂	100	91.3	80.5	This study
Microwave + polyaniline	100	45.4	65	Riaz <i>et al.</i> (2014)
ZVI + H ₂ O ₂	105	/	53	Moon <i>et al.</i> (2011)
Microwave electrodeless lamp +TiO ₂ + H ₂ O ₂	100	5	20	Zhang <i>et al.</i> (2008)

which are of the lowest energy, then destroy the intermediate product (benzene ring and naphthalene ring), consequently causing decolorization/degradation of the orange II solution (Ghoneim *et al.* 2011).

Comparison of different methods

As can be seen from Table 1, compared with other experimental methods, microwave-assisted ball milling is a more efficient method for the decolorization and mineralization of orange II. The results demonstrated the microwave-assisted ball milling method is environmentally friendly since it does not involve the use of harmful chemical reagents. In addition, it is easy to handle and the reactor used is simple.

CONCLUSIONS

In this work, microwave-assisted ball milling was applied for the removal of orange II in wastewater. Parameters affecting the removal of orange II were investigated. Results showed that the microwave-assisted ball milling could remove orange II effectively. The results obtained under the optimal operational parameters (100 mg/L aqueous solution of pH 3 containing 400 mg/L H₂O₂ while controlling microwave power at 400 W) showed that the decolorization rate was 91.3% after 2 min and the TOC removal efficiency was 72.7 and 80.5% with reaction time of 10 and 60 min, respectively. The results indicated that microwave-assisted ball milling can significantly reduce color and TOC in dye wastewater.

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