



# Enhanced decolorization of methyl orange in aqueous solution using iron–carbon micro-electrolysis activation of sodium persulfate



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## HIGHLIGHTS

- Decolorization of methyl orange by Fe–C micro-electrolysis with persulfate was tested.
- ICE-activated PS oxidation increased MO removal efficiency and BI of MO wastewater.
- ICE-PS is tolerant to wide range of pH, initial MO concentration, and temperature.
- Ferrous ions (Fe<sup>2+</sup>) were the persulfate activation resource.

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## ABSTRACT

Reactivity of sodium persulfate (PS) in the decolorization of methyl orange (MO) in aqueous solution using an iron–carbon micro-electrolysis (ICE) method was investigated. The effects of sodium persulfate doses, pH, Fe-to-C mass ratios, initial MO concentration as well as the reaction temperature were comprehensively studied in batch experiments. The ICE-PS coupled process was more suitable for wide ranges of pH, initial MO concentration and reaction temperature, accompanied by the reduction of Fe compared ICE. The MO removal efficiency improved substantially by ICE-PS technique, 76.03% for ICE and 91.27% for ICE-PS at experimental conditions of pH 3.0, Fe-to-C mass ratio 3:1, PS addition 10 mM and initial MO concentration 0.61 mM. Furthermore, the biodegradability index (BI) dramatically increased from 0.26 to 0.65. The binary hydroxyl and sulfate radicals that non-selectively degrade MO to the derivatives with small molecules are ascribed to ICE-PS method as detected by the UV–vis spectra. The PS activation resource was Fe<sup>2+</sup> through the hydroxyl radical quenching reaction by the additive tert-butanol (TBA). This study provides an in-depth theoretical understanding of the development and wide commercial application of the ICE technology to refractory industrial dye wastewater treatment.

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## 1. Introduction

Dye effluents containing hazardous materials with high concentrations of organics, recalcitrant macromolecules, and biological toxicity pose serious threats to surface water safety and aquatic ecosystems. Therefore, the elimination of contaminants from dye effluents was intensively investigated in the past few decades (Wu and Wang, 2012; Thai and Ruey, 2013; Matou et al., 2014; Szabolcs et al., 2014; Haque et al., 2015; Aravind et al., 2016). Conventional

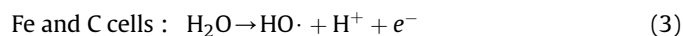
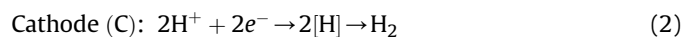
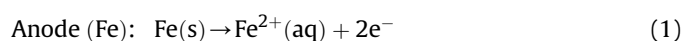
wastewater purification methods such as reverse osmosis (Zhao et al., 2015; Alicia et al., 2016), ultrafiltration (Dasgupta et al., 2016; Lin et al., 2016), adsorption (Regti et al., 2016; Zhu et al., 2017a,b), membrane and magnetic separation (Puspasari and Peinemann, 2016; Song et al., 2016; An et al., 2017), and traditional chemical oxidation (Tichonovas et al., 2013; Wang et al., 2017) can hardly achieve complete detoxification, and have non-flexible applications, high operating costs, limited environmental compatibility. Therefore, advanced oxidation processes (AOPs) have been introduced for the purification of dye effluents (Doumic et al., 2015; Cui et al., 2016; Li et al., 2017).

AOP technologies yield inspiring removal efficiency rates in dye-effluent and other refractory organics treatment (Bilińska et al., 2016; Li et al., 2016a,b; Zhu et al., 2017a,b). The main mechanism

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of the organics decomposition of AOPs is the chemical oxidation by activated radicals, including hydroxyl ( $\text{HO}\cdot$ ), sulfate ( $\text{SO}_4^{\cdot-}$ ), hydroperoxyl ( $\text{HO}_2\cdot$ ), and oxygen ( $\text{O}\cdot$ ) radicals. A wide range of hazardous organic contaminants can be destructed by activated radicals quickly and non-selectively (Harada et al., 2016; Li et al., 2016a,b; Expósito et al., 2017). Among the various activated radicals,  $\text{HO}\cdot$  (standard reduction potential  $E^\circ = 2.80 \text{ V/SHE}$ ), induced by water discharge through retardation of oxygen in the ultrasound, UV irradiation, electric field and Fenton systems is the most common and powerful oxidant extensively applied in organic wastewater treatment (Fontmorin et al., 2016). However, the instability of  $\text{HO}\cdot$  (half-life of  $10^{-9} \text{ s}$ ) restricts the continuous oxidation of organics. Especially, in treating wastewater with low-concentration organics, the mass transfer will inhibit the degradation dramatically.  $\text{SO}_4^{\cdot-}$  radicals have an electron transfer preference that would ensure a prolonged half-life for efficient utilization. Additionally, their remarkable redox potential ( $E^0 = 2.5\text{--}3.1 \text{ V}$ ) guarantees complete oxidation of refractory organics.  $\text{SO}_4^{\cdot-}$  can be generated during the activation of persulfate (PS) (including peroxomonosulfate (PMS) and peroxydisulfate (PDS), with the standard reduction potential of 1.82 V/SHE and 2.01 V/SHE respectively) by heat, ultrasound, ultraviolet irradiation, quinones, transition metals and strong oxidants (Amr et al., 2013; Zhou et al., 2015; Matzek and Carter, 2016; Chen et al., 2017; Ismail et al., 2017; Rayaroth et al., 2017). Recently, binary and complex systems such as electro-ultrasound (Chen and Huang, 2015), ozone-ferrous (Amr et al., 2013; Ge et al., 2016) as well as iron-carbon micro-electrolysis (ICE) processes (Ma et al., 2017) have been reported to have a strong activation effect on PS. Nevertheless, to the best of authors' knowledge, no studies have focused on the identification of PS activation resources (transition metals ( $\text{Fe}^{2+}$ ) or strong oxidants ( $\text{HO}\cdot$ )), in these binary activation systems.

ICE is considered an attractive pretreatment method for high-concentration, low biodegradability index (BI, the ratio of five-day biological oxygen demand ( $\text{BOD}_5$ ) to chemical oxygen demand (COD)) organic industrial wastewater, with the advantages of no electricity consumption, cost savings, and ease of operation and maintenance (Ying et al., 2012; Zhu et al., 2014; Wang et al., 2016; Deng et al., 2017). Organic contaminants would undergo a synergistic decomposition via oxidation reactions with activated radicals and reduction reactions with reducing groups ( $\text{Fe}^{2+}$  and  $[\text{H}]$ ). The ICE mechanism is shown in the following equations:



Coagulation, interception and adsorption reactions can also contribute to organics purification. Unfortunately, ICE has the disadvantages of hardening, blockage and passivity after being operated for a period of time during real-world applications (Han et al., 2016). On the other hand, the BI after ICE was expected to be improved to achieve highly efficient biological processes. Therefore, ICE requires continuous improvement to become a time-efficient, versatile, and adaptive alternative for the pretreatment of organic industrial wastewater.

Methyl orange (MO) is a representative azo dye, which contains an azo group (N, N double bond) attached to substituted benzene rings and has the properties of macromolecule and refractory degradation. It is widely used in paper and textile industries, while its toxic, carcinogenic and mutagenic effects on human beings are still a severe problem. Herein, MO was chosen as the target

pollutant. The objective of this study is to investigate the performance of ICE activation of PS for the pretreatment of MO wastewater and determine the PS activation resources in transition metal ( $\text{Fe}^{2+}$ ) and strong oxidant ( $\text{HO}\cdot$ ) binary activation system. The effects of the critical parameters, including sodium persulfate doses, aqueous solution pH, Fe-to-C mass ratios, initial MO concentrations, and reaction temperature were comprehensively studied in batch experiments. UV absorption spectrum and BI values at different treatment duration were detected. The hydroxyl radical quenching reagent tert-butanol (TBA) was used to identify of the activation resource in the ICE-PS coupled process.

## 2. Experimental

### 2.1. Materials

Iron chips with the particle size of 5–8 mm and coal-based granular activated carbon (GAC) (diameter 2–5 mm) were obtained from Gongyi Ming Jian Chemical Reagent Beijing, China Co., Ltd. Before use, iron chips and GAC were pretreated as follows: First, iron chips were soaked in 10% NaOH solution for 2 h to remove surface grease, then dipped into 5% hydrochloric acid solution for 30 min to remove surface oxidation, and finally rinsed with deionized water until a neutral pH value was reached. GAC was cleaned with deionized water for five times, boiled for three times in 10% KOH solution, and washed with deionized water until pH was close to neutral. Before each experiment, GAC was soaked in the prepared methyl orange wastewater for 72 h to eliminate the effect of adsorption. Methyl orange, tert-butanol (TBA) and sodium persulfate (PS) (analysis pure, A. P.,) were purchased from Changcheng Chemical Reagent Nanchang, China Co., Ltd. The pH of the solution was adjusted using  $\text{H}_2\text{SO}_4$  (98%, Changcheng Chemical reagent Nanchang, China) or NaOH (A.P., Changcheng Chemical reagent Nanchang, China).

### 2.2. Batch experiments

Methyl orange solutions were prepared using deionized water. Two groups experiments with and without PS were carried out with a sequencing batch operating mode in 500 mL beakers. All reactions were initiated with 300 mL methyl orange solutions. The total addition of Fe and GAC is 200 mg. The effect of PS doses was investigated for PS concentration of 0, 4, 6, 8, 10 and 20 mM. The other operating parameters were temperature ( $25^\circ\text{C}$ ), pH (3.0), Fe-to-C mass ratio (3:1), initial MO concentration (0.61 mM). The aqueous solution pH values were designed as 3.0, 5.0, 7.0 and 9.0 with PS doses of 4.0 mM. And the followed reactions were conducted under the optimized pH value. The mass ratio of Fe-to-C ranged from 1:3 to 3:1 based on actual industry experience. Initial methyl orange concentrations of 0.15, 0.31, 0.46, 0.61, 0.92 and 1.22 mM, and the reaction temperatures of 25, 35, 45, 55 and  $65^\circ\text{C}$  were used in the experiments.

The aqueous solutions were shaken by a thermostatic reciprocating shaker at 150 rpm at various reaction temperatures. At different elapsed times, the samples were collected and filtered through 0.22  $\mu\text{m}$  membranes to determine methyl orange concentration,  $\text{COD}_{\text{Cr}}$  and  $\text{BOD}_5$ . The concentration of tert-butanol (TBA) was controlled at 5.0 mM.

### 2.3. Analytical methods

The concentration of methyl orange was determined by UV–Vis spectrophotometer (Shimadzu, Japan) at an absorbance of 462 nm. The MO concentration was analyzed with a pseudo-first-order kinetic model:

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