



## Purification treatment of dyes wastewater with a novel micro-electrolysis reactor



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### ABSTRACT

A novel micro-electrolysis reactor bringing iron-carbon filler to cycle flow was developed in this study, called as an internal circulation micro-electrolysis (ICE) reactor, which can overcome the disadvantage of conventional fixed bed (CFB) reactor. In order to investigate the performance of the ICE reactor, a practical dyes wastewater was selected as target wastewater. The removal efficiency of chemical oxygen demand (COD) and chroma of the CFB reactor with commercial hardening resistance iron-carbon filler (CRIF) are only 23% and 40%, respectively. However, for the ICE reactor with a common iron-carbon mixture (CIM), the removal efficiency of COD and chroma are 73% and 98.5%, respectively. The improvement efficiency of biodegradability index of dyes wastewater for the ICE reactor with the CIM is 23 times, which is about fourfold as much as that of the CFB reactor with CRIF. The ICE reactor with CIM can stably keep a high treatment performance for 60 days continuous operation. However, the performance of CFB reactor with CIM decreases sharply. The results indicate that the ICE reactor has an excellent performance for the pretreatment of dyes wastewater, which should be due to the elimination of hardening, blocking and inactivation occurring in the CFB reactor with CIM by the circulation action.

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### 1. Introduction

Internal micro-electrolysis (IE), which can effectively improve the biodegradability index (BI, the ratio of five-day biological oxygen demand (BOD<sub>5</sub>) to chemical oxygen demand (COD)) of wastewater, is an excellent pretreatment technology for high-concentration non-readily degradable organic industrial wastewater, such as dyes wastewater, petrochemical wastewater, pharmaceutical wastewater, landfill leachate and coking wastewater [1–4]. The materials of IE consisting of iron chip and granular activated carbon (GAC) can form microscopic galvanic cells in an electrolyte solution [5]. Some products (such as active hydrogen, hydrogen peroxide, hydroxyl radical and ferric flocculants) generating at the electrode have high activity, and can bring about redox reaction and electrocoagulation with many pollutants, which can induce the decoloration and degradation of pollutants [6–9]. Therefore, the IE process has been extensively used in many kinds of wastewater because of no consumption of electric power resources, low cost, convenient operation and maintenance [10,11]. However, some disadvantages have been found during the practical application of traditional IE process [7]. The

iron-carbon mixture (ICM) is easy to harden, block and passivate after operating for a period of time [12], which brings about channeling and drifting flow phenomenon and then reduce the processing efficiency [13,14]. Moreover, the mixture needs to be unloaded and reloaded because of the consumption of iron chips, which not only leads to the immense waste of labor and financial resource but also interrupt the continuous operation. In order to solve the above defects, an internal circulation micro-electrolysis (ICE) reactor has been developed.

Due to dyes are considered as stable, stubborn, colorant, and potentially carcinogenic and toxic pollutants, their discharge will bring about serious environmental, aesthetical and health problems [15]. Therefore, the decontamination of dyes effluents from the textile and dyes industries should be effectively treated before they are discharged into the environment. Biological technology known as the cheapest ways is strikingly ineffective for the dyes wastewater due to their low biodegradability [16–18]. Therefore, it is necessary to develop effective pretreatment technologies to remove/reduce the toxic substances before biological treatment. Some technologies have been developed for the potential application in the pretreatment of poor biodegradable and even toxic wastewater [17,19,20]. Electrochemistry has been used to pretreat the wastewater containing methylene blue with its BI improved by 16 times [21]. Anaerobic processes have been also applied to enhance the BI of textile wastewater [22]. In addition, advanced

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oxidation technologies have been also employed to improve dyes wastewater treatment [23–25]. However, either removal efficiency and/or treatment cost have restricted the wide application of these pretreatment methods. Therefore, the dye was selected as a target pollutant in this study due to its biodegradable characteristics.

The objective of this study is to investigate the performance of the ICE reactor for the pretreatment of dyes wastewater and supply a theoretical basis for this technical potential of ICE for the treatment of toxic industrial wastewater. Therefore, the influencing factors and the treating efficiency of practical dyes wastewater with ICE process were investigated.

## 2. Materials and methods

### 2.1. Materials

Iron chips obtained from our school metalworking practice factory were passed through a sieve with 20 meshes. The sifted iron chips were first soaked in 10% NaOH solution for 2 h to remove the surface grease, then dipped in 5% hydrochloric acid solution for 30 min to remove the surface oxidation, and finally rinsed by tap water until pH was close to neutral. GACs (1–2 mm) were purchased from the Sinopharm Chemical Reagent Beijing Co., Ltd. Before use, the GACs were cleaned by tap water, and then soaked in the prepared dyes wastewater for 72 h to eliminate the adsorption influence. The dyes wastewater, of which the main organic component was azodyes stuff, was taken from Zhejiang Longsheng Group Co.Ltd. The characteristics of the raw dyes wastewater were described in Table 1. Other chemical reagents were of analytical grade except for special mentioning in the paper.

### 2.2. ICE reactor

The ICE reactor was made of plexiglass, shown in Fig. 1. The feed was introduced into the lower part of the iron-carbon bed through a casing tube, and then discharged through an inlet distributor. Microelectrolytical degradation reaction took place upwards through the iron-carbon bed. The iron-carbon bed was kept in a downward motion by an air-lift pump taking out the passivated and fully reaction iron-carbon mixture from the bottom. The iron-carbon mixture was subjected to a thorough mechanical agitation by the action of air bubbles in internal recycle tube, which made the iron chips to be activated. With the action of air-lift pump, the iron-carbon mixture was automatically and continuously taken out of the iron-carbon bed, washed and returned to the upper part of the iron-carbon bed. The treated wastewater was discharged from the iron-carbon reactor through overflow weir.

### 2.3. Experimental procedure

A series of experiments of iron-carbon microelectrolysis were carried out to investigate the influence of main operating parameters on COD removal and decolorization efficiency of dyes wastewater. The iron-carbon ratio (Fe/C), pH, reaction time and aeration amount were selected as the main operating parameters in the

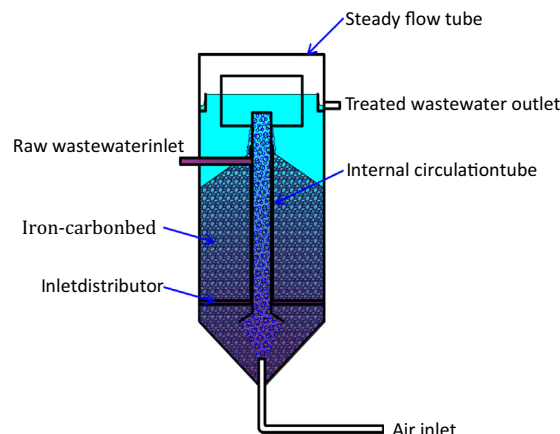


Fig. 1. Schematic of the ICE reactor.

paper. The practical dyes wastewater with adjusted pH value was fed into the ICE reactor filled with CIM. For identifying the superiority of ICE reactor, the treatment efficiency of dyes wastewater were investigated for both a conventional reactor with commercial hardening resistance iron-carbon filler (CRIF) and a conventional fixed bed (CFB) reactor with CIM. The samples obtained from the effluents of ICE reactor were first adjusted pH value by adding NaOH to about 9, and then filtered. COD, BOD<sub>5</sub> and chroma of the leach solution were determined according to the national standard methods of China [26]. The surface morphologies of Fe before and after 60 days continuous operation in ICE reactor and CFB reactor were characterized by scanning electron microscopy (SEM, JSM-5600LV, Oxford, UK) with an accelerating voltage of 20 kV.

## 3. Results and discussion

### 3.1. Influence of operating conditions

The wastewater quality parameters of the industrial effluent usually fluctuate, such as pH [27]. A large of research show that reaction time, Fe/C and aeration amount have important influence on COD removal and decolorization efficiency of IE reaction [28,29]. Therefore, the effect of reaction time, pH, Fe/C and aeration amount on COD removal and decolorization efficiency of dyes wastewater in the IE reactor were investigated in this work.

#### 3.1.1. Effect of reaction time

Fig. 2 shows the influence of reaction time on the removal efficiency of COD and chroma of dyes wastewater in the ICE reactor. The results showed that with the reaction time increasing, the removal efficiency of COD and chroma increases. The decoloration efficiency rises rapidly and reaches at 98% with 90 min. However, the removal efficiency of COD increases slowly and is 70% with 90 min. This indicates that the extension of reaction time can effectively improve the degradation efficiency of dyes wastewater in ICE reactor. However, the longer reaction time induces the more iron to dissolve and the larger reactor volume, adding the cost of wastewater treatment [30]. Therefore, in the following experiments, the 90 min reaction time was selected.

#### 3.1.2. Effect of pH

Fig. 3 shows the effect of pH on the removal efficiency of COD and chroma of dyes wastewater treated by ICE reactor. The removal efficiency of COD and chroma increase slightly when pH rises from 2 to 3.5, and then decrease with pH over 3.5. It is well known that the acidic condition is conducive to the microscopic

Table 1  
The characteristics of the raw dyes wastewater.

	Maximum	Minimum	Mean
COD (mg/L)	2000	1000	1300
BI	0.015	0.000	0.01
Chroma (multiple)	16,384	1024	4096
pH	3	1	2

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