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Removal of Rhodamine B from aqueous solution using graphite–graphite electro-Fenton system

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ABSTRACT

Removal of Rhodamine B (RhB) using graphite–graphite electro-Fenton (EF) system is investigated in the present study. The effect of various operational parameters on RhB removal was studied. Experimental results showed that RhB was removed completely by the reaction with $^{\cdot}$OH generated from graphite cathode. The optimum applied voltage was determined as 8 V. Higher RhB removal efficiency values were obtained with rising electrode area and depth of electrode immersion. But, RhB removal was decreased with increase in RhB concentration. The optimal electrode spacing was obtained as 4 and 5 cm. At optimal conditions, 99.2% of RhB removal was obtained after 180 min of electrolysis. The present study shows that graphite–graphite EF system was an efficient tool for removing dyes from aqueous solutions.

Keywords: Dye removal; Electro-Fenton; Graphite; Rhodamine B; Advanced oxidation process

1. Introduction

Hazardous substances removal from wastewater is a major problem in the world. In recent years, advanced oxidation processes (AOPs) were used to remove such pollutants from wastewater. AOPs are defined as the oxidation processes in which the hydroxyl radicals ($^{\cdot}$OH) are derived in sufficient quantity to effect wastewater treatment [1,2]. AOPs are recommended when wastewater components have a high chemical stability and/or low biodegradability [3]. The efficacy of AOP process depends strongly on the rate of generation of the free radicals along with the extent of contact of the generated radicals and/or chemical oxidants with the contaminant molecules and the efficient design should aim at maximizing both of these quantities [2]. A chemical wastewater treatment using AOPs can produce a complete mineralization of pollutants to CO_2, water, and inorganic compounds, or at least their transformation into more innocuous products [3].

Among the AOPs, Fenton process has received a great attention in recent years. Fenton process generates $^{\cdot}$OH by the reaction between ferrous ion and hydrogen peroxide in an acidic medium (Eq. (1)). The advantages of the Fenton process are a low capital cost, easy operation, and nontoxic by-products [1]. However, the Fenton process produces a huge amount of ferric hydroxide sludge, which requires additional separation and disposal [1]. In order to rectify this defect, electro-Fenton (EF) [1,4–6], photo Fenton [7], sono Fenton [8], hydrodynamic cavitation combined

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Fenton [9], photo EF [10], sono photo Fenton [11], and sono EF [12] processes have been developed. EF process consists of adding Fe^{2+} with the simultaneous production of H_2O_2 upon the reduction of O_2 on the electrodes [4]. H_2O_2 is electrogenerated in acidic solutions by two-electron reduction of oxygen on the cathode surface according to Eq. (2) [5]. This electrogeneration of H_2O_2 depends more on cathode material. Several cathodes such as stainless steel [1,6], activated carbon fiber [13], boron doped diamond [14], commercial graphite felt [15], carbon felt [16], graphite [17], reticulated vitreous carbon [18,19], and carbon sponge [20] are some of the recently used working electrodes in wastewater treatment. Due to low cost, graphite has traditionally used for the electrochemical production of hydrogen peroxide [13].

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + OH^- + HO^{\cdot} \tag{1}$$

$$O_2 + 2H^+ + 2e^- \rightarrow H_2O_2 \tag{2}$$

Generally, materials like Pt [6,13], boron doped diamond [20], Ti–IrO_2/RuO_2 [21], etc. were used as anode in most of the recent studies. Because, from water oxidation, the hydroxyl radicals will produce at the surface of a high oxygen overvoltage anode (Eq. (3)) [22]. But the cost of these materials is very high. Therefore, an alternative low cost EF system is necessary for the treatment of wastewater.

$$H_2O \rightarrow HO^{\cdot} + H^+ + e^- \tag{3}$$

Few studies have been reported that graphite can be used as both electrodes in EF process [23]. This system has many advantages over other EF systems. If two different materials are used as electrodes, the anode particle will form a layer on cathode. This may affect the efficiency of the EF process. But in graphite–graphite EF system, the new graphite layer on cathode will enhance the efficiency of the system. Also, it will not affect the homogeneity of the system. Therefore, the aim of this present study is to test the efficiency of the graphite–graphite EF system for the removal of Rhodamine B (RhB) from aqueous solution. RhB is a basic dye which is widely used as a colorant in textiles and food materials and as biological stains in biochemical laboratories at high concentrations [24]. It is estimated that 10–15% of this dye is lost in the effluent during dyeing process [25]. RhB is dangerous if swallowed by human beings and animals, causes irritation to the skin, eyes, and respiratory tract [26]. The carcinogenicity, reproductive and developmental toxicity, neurotoxicity, and chronic toxicity towards humans and animals have been experimentally proven [24,27]. Accordingly, effluent of industries containing this nonbiodegradable toxic dye must be treated before discharging into water bodies [28]. RhB was removed effectively from the aqueous solution by various AOPs such as photo catalysis [29], hydrodynamic cavitation [30], sono photocatalysis [31], Fenton [32], etc. To our best knowledge, the RhB removal performance of graphite–graphite EF system was not tested upto yet. The main purpose of the present study is to analyze the RhB removal behavior at various operational parameters in graphite–graphite EF system.

2. Materials and methods

2.1. Materials

Synthetic solution was prepared by dissolving analytical grade RhB dye stuff (Loba Chemie, CI 45,170) used without further purification, in distilled water. Other chemicals of AR grade ($FeSO_4{\cdot}7H_2O$, $NaHCO_3$, and H_2SO_4) were obtained from Merck. All the preparations and experiments were conducted at room temperature. Graphite plate was supplied by Anabond Sainergy Fuel Cell India Private Limited, Chennai, Tamilnadu, India. A UV/Vis spectrophotometer (Lambda 25, PerkinElmer, USA) using silica cell of path length 1 cm was used for the determination of RhB concentration. The pH of the solution was monitored using multiparameter PCSTestr 35 (Eutech, Singapore). The current input was supplied by a DC power supply (Make: Beetech, 0–5 A and 0–30 V).

2.2. Electrolytic system

All electrolyses were conducted in an open, undivided, and cylindrical cell of 1,000 ml capacity and diameter of 10.4 cm. The working volume of 10 mg/l RhB solution was 750 ml. Graphite plates of effective area 25 cm^2 (5×5 cm) were used as both cathode and anode. The distance between electrodes was kept 4 cm. Prior to the electrolysis, air was bubbled through the aqueous solution using commercially available "fish aerator". This was continued till the end of the experiments. This continuous aeration saturates the acidic RhB solution with oxygen and maintains this saturated level till the end of electrolysis. This dissolved oxygen is responsible for the in situ production of H_2O_2 at cathode as in Eq. (2). Then the pH of the solution was adjusted to 3 using 0.5 N H_2SO_4·2.5 mg/l of $NaHCO_3$ was chosen as supporting electrolyte in all the experiments. Series of experiments were conducted to investigate the effects of applied voltage, current, initial dye concentration, area of electrode, depth of immersion of electrode, and distance between electrodes, etc. The RhB samples were taken at regular intervals and the concentration of RhB was measured using UV–Visible spectrometer.

3. Results and discussions

3.1. Effect of applied voltage and current

The influence of applied current and voltage on RhB removal was performed by electrolyzing 10 mg/l of RhB solution of pH 3 containing 10 mg/l Fe^{2+}, 2.5 mg/l $NaHCO_3$ at voltage ranging from 2 to 20 V (Current was increased from 0 to 0.1 A according to increase in applied voltage). The effect of voltage and current on RhB removal is shown in Fig. 1. As seen in Fig. 1, the removal of RhB increases by increasing the amount of applied voltage and current upto 8 V and 0.03 A. For example, the RhB removal increases from 88.5 to 98.8% after 180 min of electrolysis for the increase of voltage from 2 to 8 V. Because, higher applied current increases the quantum of hydrogen peroxide produced, thus increasing the number of hydroxyl radicals in the electrolyte medium, which are highly reactive and responsible for the removal [6]. This is also due to the higher electro-regeneration of ferrous ion from ferric ion with increasing current, which increased the efficacy of Fenton chain reactions [33]. Since RhB is a cationic dye, increase in applied voltage attracts more dye molecules towards cathode, which is the working electrode of EF process. Therefore, the collision frequencies between the dye molecules and electro-generated $^{\cdot}OH$ increases. This enhances the efficiency of EF process. However, the removal of RhB decreases after 8 V and 0.03 A. This is due to the competitive electrode reactions in the electrolytic cell. At higher current, the competitive electrode reactions such as the discharge of oxygen at the anode (Eq. (4)) and the evolution of hydrogen at the cathode (Eq. (5)) would become pronounced [33]. These reactions inhibit main Fenton reactions. Therefore, 8 V and 0.03 A is considered as the optimal conditions for voltage and current for further studies.

$$2H_2O_2 \rightarrow 4H^+ + O_2 + 4e^- \tag{4}$$

$$2H^+ + 2e^- \rightarrow H_2 \tag{5}$$

3.2. Effect of initial dye concentration

The effect of RhB concentration on its removal via EF process at voltage = 8 V, current = 0.03 A, $[Fe^{2+}]$ = 10 mg/l, and $[NaHCO_3]$ = 2.5 mg/l is shown in Fig. 2. Fig. 2 shows that the rate of RhB removal decreases with increase in initial dye concentration. With increase in initial RhB concentration from 10 to 40 mg/l, the removal efficiency of graphite–graphite system after 180 min of electrolysis decreased from 98.8 to 84.1%. This fact might be due to the competitive consumption of hydroxyl radical by the generated intermediates at high initial dye concentration [34]. Also, at higher dye concentration, the generation of $OH^{\cdot}$ radicals on the surface catalyst is reduced since the active sites may be occupied by dye ions and because the number of dye molecules is increased but not the $OH^{\cdot}$ radical

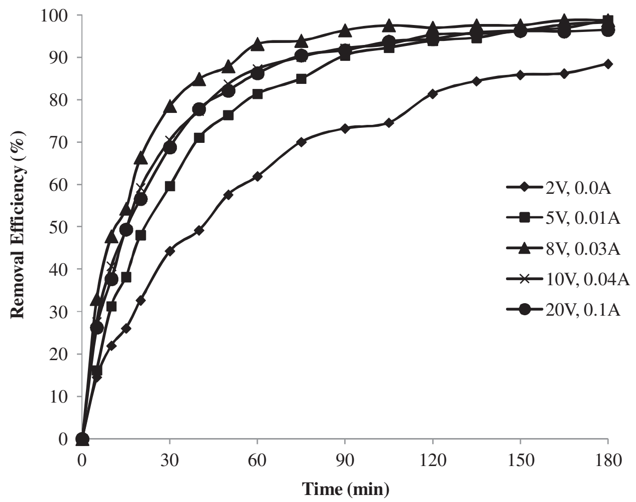

Fig. 1. Influence of applied voltage and current on RhB removal kinetics.

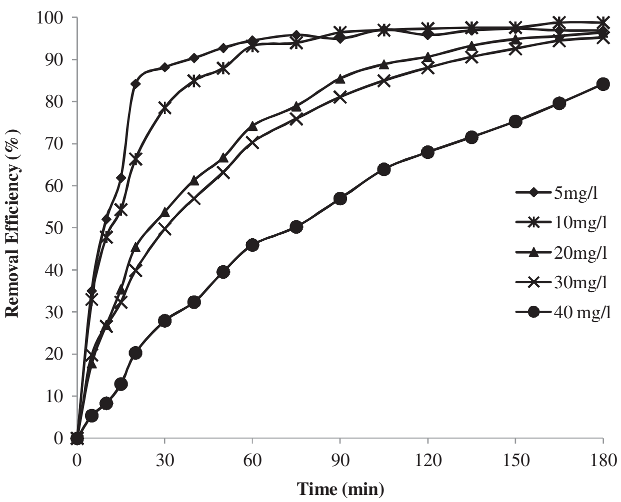

Fig. 2. Influence of initial RhB concentration on removal.

molecules number [35]. Although the decolorization efficiency of EF system decreased with initial RhB concentration, the absolute dye removal has increased. For example, when the RhB concentration was increased from 10 mg/l to 40 mg/l, the removal of RhB concentration after 60 min of electrolysis has been increased from 7.8 to 16.1 mg/l, respectively. The quantity foam formed during electrolysis also increased with initial dye concentration. The foam was observed upto 60 min for 10 mg/l, while in the case of 40 mg/l RhB solution, the foam was retained even after 180 min of electrolysis.

3.3. Effect of electrode area

To clarify the effect of electrode area on dye removal, 10 mg/l of RhB solutions was electrolyzed at different graphite area ranging from 10 cm^2 and 25 cm^2. The depth of immersion electrode was kept constant as 5 cm in all the experiments. The applied voltage, [Fe^{2+}], [$NaHCO_3$], and pH for the removal experiments was 8 V, 10 mg/l, 2.5 mg/l, and 3, respectively. The results obtained are shown in Fig. 3. It can be seen from Fig. 3 that RhB removal rate decreases with decreasing the area of electrode. For example, the RhB removal was obtained as 98.8 and 93.9% for the area of 25 and 10 cm^2, respectively. The H_2O_2 generation in carbonaceous electrodes is mainly depends on pore volume. With the increase in graphite area, the volume of pores also increases. Consequently, the rate of H_2O_2 generation increases with increase in electrode area. This leads to the greater removal of RhB in higher electrode area. Similar result was reported by Daneshvar et al. [36] for the removal of Orange II.

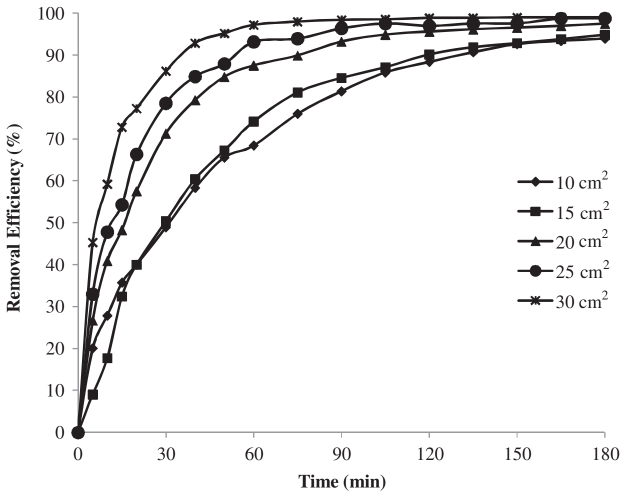

Fig. 3. Effect of graphite area on the removal kinetics of RhB.

3.4. Effect of depth of electrode immersion

Depth of electrode immersion on RhB removal was tested by changing the dimensions of electrodes with constant effective area of 25 cm^2. The following sizes such as, 3.57 × 7, 5 × 5, and 6.8 × 3.7 cm were selected as the effective dimensions of graphite electrodes. During the electrolysis the other operating parameters were kept constant throughout the experiments. It was found that the RhB removal rate increased with increase in depth of immersion. For example, the RhB removals were 70.64, 78.5, and 81.99% for the depths of 3.57, 5, and 6.8 cm, respectively after 30 min of electrolysis. Generally, the size of air bubble increases as it comes out from the bottom. Surface area of the air bubble is also an important parameter for the mass transfer of oxygen from the liquid phase to electrode surface. The mass transfer of oxygen increases with increase in surface area. The surface area of small bubbles is higher than that of coarse bubbles. These bubbles can be trapped effectively inside the active pores of graphite plate. It is well known that the large gas bubbles rise in a plug-flow manner and small gas bubbles are entrained within liquid recirculation and back mixing [37]. Also, increase in depth of immersion of the electrode results in increasing the contact time between air bubble and graphite surface. Therefore, for higher depth of immersion, electrodes have sufficient time to adsorb the oxygen than in lesser depth of immersion. Another important parameter affecting the mass transfer of oxygen is the bubble rising velocity. Due to higher surface area, the friction between the finer bubbles and the liquid is very high and results in very much slower rising velocity than coarse bubbles. Thus, with the increased friction with the small bubble, much more oxygen will get trapped into the graphite surface in comparison to the same volume of air with coarse bubbles. Matheswaran and Moon [38] compared the mass transfer rate of bubble column and bubble diffuser for the ozonation of phenol and reported that higher mass transfer rate was achieved by means of reduced bubble size in the bubble column reactor. Since, these air bubbles are the main sources of production of H_2O_2, the H_2O_2 production also increases with increase in depth of immersion. Hence, the RhB removal rate also increases with increase in depth of immersion of electrodes. In the case of low electrode immersion depth, dissolved oxygen present in the solution is the main source for the production of H_2O_2. With the production of H_2O_2, a concentration gradient has been developed between the solution and near cathode surface. Therefore, the oxygen present in the solution moves towards the cathode. This enhances the H_2O_2 production.

Bubble reactor is a better option for scaling-up of EF process. Rosales et al. [39] carried out decolorization of Lissamine Green B and Azure B under EF process using bubble reactor and reported higher removal efficiency at residence time of 30 min. The efficiency of RhB removal by graphite–graphite EF system increased with the increase in depth of immersion. This will help in scaling-up of this process using bubble reactor.

3.5. Effect of distance between electrodes

The effect of electrode distance on RhB removal is shown in Fig. 4. The experiments were conducted using 6.8 × 3.7 cm electrodes at solution pH of 3 and voltage of 8 V. It can be observed from Fig. 4 that RhB removal rate increased with decreasing the gap between electrodes from 6 to 4 cm. This decrease of the distance between the electrodes leads to a decrease of the ohmic drop through the electrolyte and then an equivalent decrease of the cell voltage and energy consumption [40]. The longer distance between the electrodes also inhibits the mass transfer of Fe^{3+} to cathode surface, which results in the reduction in electro regeneration of Fe^{2+} [41]. Therefore, the production of hydroxyl radical reduces at higher electrode gaps and results in the reduction in RhB removal rate. The RhB removal was nearly same when the distance between electrodes is between 4 and 5 cm. For example, the RhB removal after 180 min of electrolysis was 99.2% for 4 cm distance and it was 99.5% for 5 cm distance. But, further decrease in distance between electrodes less than 4 cm leads to decrease in dye removal. RhB removal was reduced to 96.5% after 180 min of electrolysis for the distance of 3 cm. This is due to the oxidation of electro-regenerated Fe^{2+} to Fe^{3+} at anode as in Eq. (6) [33]. This oxidation inhibits the Fenton chain reactions and results in less dye removal efficiency. It was also observed that the current was increased with decreasing distance between electrodes. For example, the currents for 5 and 3 cm gap were observed as 0.02 and 0.09 A, respectively. The optimum current for the present study was obtained as 0.03 A. For currents higher than 0.03 A, two major scavenging reactions as given in Eqs. (4) and (5) will occur in the electrolytic reactor. This also decreases the dye removal efficiency of the EF system at shorter inner electrode gap.

$$Fe^{2+} \rightarrow Fe^{3+} + e^- \qquad (6)$$

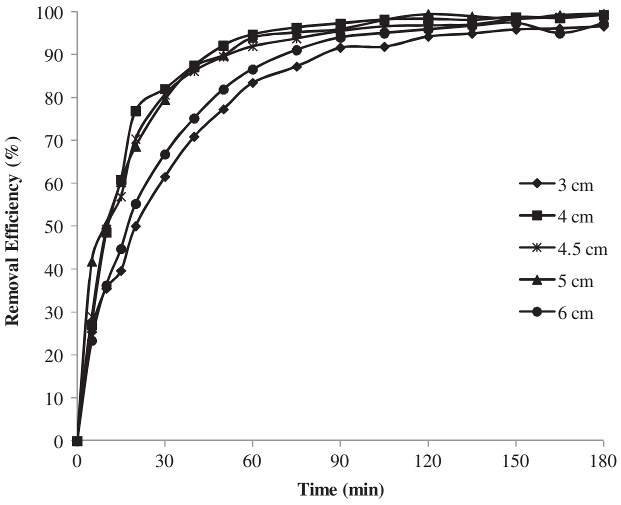

Fig. 4. Effect of distance between graphite electrodes on RhB removal.

4. Conclusions

A synthetic solution containing RhB has been effectively removed by graphite–graphite EF system in acidic medium. Efficiency of this system was increased by increasing the electrode area and depth of immersion. RhB removal was inversely proportional to initial dye concentration and was optimal at electrode spacing in between 4 and 5 cm. At the operational conditions such as applied voltage of 8 V, solution pH of 3, catalyst concentration of 10 mg/l, electrode area of 25 cm^2, and inner electrode spacing of 4 cm, 99.2% of RhB removal was obtained after 180 min of electrolysis. From this study, graphite–graphite EF system seems to be an environmentally friendly process to remove dyes from aqueous solution.

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