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Electrochemical treatment for simultaneous removal of heavy metals and organics from surface finishing wastewater using sacrificial iron anode

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ABSTRACT

Surface finishing wastewater having low pH (~2) and high conductivity (>11 mS/cm) was treated by electrocoagulation (EC) or electrochemical Fenton (ECF) processes using sacrificial iron anodes. Under the same theoretical Fe dosage, the effects of electrolytic time, initial pH, and current density on the simultaneous removal of organic and heavy metal were investigated. A complete metal removal was achieved even at the low current density of 24.2 mA/cm² and short electrolytic time of 4 min, whereas approximately 40% of chemical oxygen demand was removed. Increasing the electric current density did not improve removal efficiencies, but consumed more electric energy. Low current conditions produced a brown color sludge associated with ferric hydroxide. On the contrary, a greenish color sludge was created at a high current due to the formation of ferrous hydroxide. The formation of ferrous hydroxide impacted the treated water quality. The ECF was employed to overcome the low COD removal by the EC, achieving >67% of COD removal. The costs of ECF processes were slightly greater than that of chemical coagulation, but achieving a lot greater heavy metal removals. ECF process can be a promising method for simultaneous removal of heavy metal and organics from complex industrial wastewater.

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1. Introduction

Ferric salt is one of the most common coagulants used, although ferrous salt is less expensive than ferric chloride [1–3]. Previous studies have explored the effectiveness of Fe(II) and Fe(III) on contaminant removal [1,4–6]. Comparing ferric chloride, ferrous sulfate, and alum for the treatment of landfill leachate, Aziz et al. [4] reported that ferric chloride was the most effective coagulant for the removal of contaminants (e.g., suspended solids, color, and organics). For phosphate removal, the effectiveness of ferric has been shown to be superior to that of ferrous [6]. To the best of our knowledge, however, there have been no report on the comparison between ferric and ferrous ions with regard to contaminant removal in electrocoagulation (EC).

The conversion of Fe(II) to Fe(III) is facilitated under an alkaline pH region in the presence of dissolved oxygen (DO). Hence, increasing the electric current intensity can affect the ferrous-to-ferric conversion rate since the rate of ferrous ion generation increases with the electric current intensity and the oxidation of ferrous to ferric might be limited by the replenishing rate of DO. The majority of previous studies did not pay much attention to the effect of electric current intensity/voltage on the conversion of ferrous to ferric ions [7–9]. Meanwhile, the most of EC studies normally examined the effect of the initial pH, allowing solution pH to increase with time due to the reduction of protons to H₂ on the cathode [10–12]. Although the alkaline pH region might be beneficial for the removal of metals [13] and the precipitation of ferrous hydroxide, it can be detrimental to the organic matter removal. Kobya et al. [14] showed that the chemical oxygen demand of textile wastewater was preferably removed at acidic pH values. Also, Can and Bayramoglu [15] showed that removal of dye and two other organic molecules decreased with increasing pH.

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In addition, previous studies reported contaminant removal by Fenton process with hydrogen peroxide being produced through the cathodic reduction of DO and ferrous ions being added chemically and/or generated electrolytically on a cathode [16–18]. It was denoted as an electro-Fenton (EF) process [19,20]. When hydrogen peroxide is added chemically and ferrous iron is generated electrolytically from iron anodes, it is denoted as an electrochemical Fenton process (ECF). Comparing the EC, EF, and ECF for treating textile wastewater, Ghanbari and Moradi [19] reported that the ECF was the most effective treatment method for the removal of chemical oxygen demand and color removal. Yet, there is still the need to conduct further studies for the simultaneous removal of heavy metals and organics by electrochemical methods.

Therefore, this study focused on the treatment of plating wastewater using the EC and ECF processes in terms of the simultaneous removal of heavy metals and organic matter. The effects of electric current density and initial pH on the removal efficiencies of contaminants were investigated under the same theoretical iron dose. The content of ferrous and/or ferric hydroxides in the sludges produced was investigated while discussing the implication of the iron oxide species on the contaminant removal and the treated water quality was discussed. Finally, the economical cost and treatment efficiency of the electrochemical process for the treatment of surface finishing wastewater was evaluated and compared with chemical coagulation.

2. Materials and methods

2.1. Wastewater samples

Wastewater samples were collected from a local surface finishing plant, which generates three different streams of wastewaters, namely Cr-containing, CN-containing, and heavy metal-containing wastewater. Both Cr- and CN-containing wastewaters are pre-treated with reduction and oxidation processes, respectively, to convert Cr(VI) to Cr(III) and to oxidize cyanide ions before being mixed with heavy metal-containing wastewater in an equalization tank. The mixed wastewater (flow rate approximately 30 m³/h) is then treated with traditional chemical coagulation, sedimentation, and sand filtration before being discharged into a local river stream. The wastewater sample used in this work was collected from the equalization tank. Table 1 summarizes the qualities of the wastewater used. The solution pH was acidic (2.0). Zinc was the main heavy metal present in the wastewater at a concentration of 190 mg/L, whereas the nickel, copper, and chromium ions were also present (<1 mg/L). The conductivity (11.4 mS/cm) of wastewater was high enough to apply the EC treatment. For the experiments to examine the effects of electrolytic time and current, the wastewater sample pH was first adjusted to 7. The neutralization step is the pretreatment step employed in the surface finishing plant, and is adopted in this study before EC. Flocs were created after the pH adjustment due to the precipitation of Cr(OH)₃.

Table 1
Qualities of raw wastewater collected from a surface finishing plant.

Item	Value
pH	2
Conductivity, mS/cm	11.35
COD, mg/L	246.81
TOC, mg/L	91.4
Zinc, mg/L	190.10
Total Cr, mg/L	87.50
Iron, mg/L	24.88
Nickel, mg/L	0.94

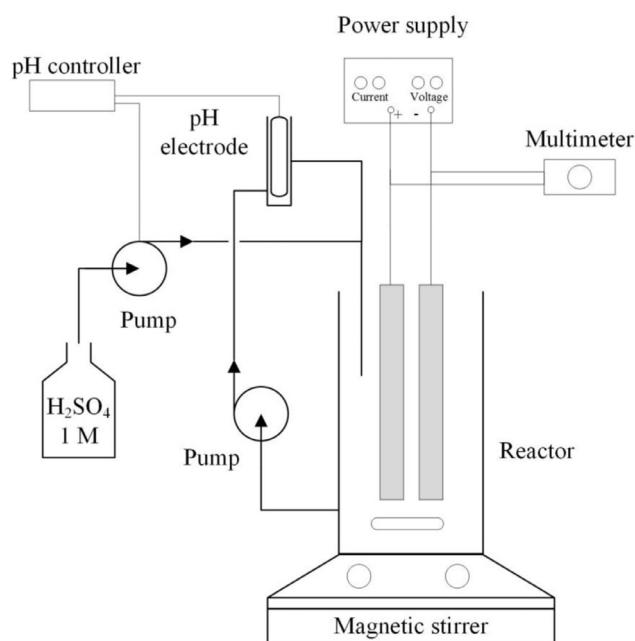


Fig. 1. Schematic diagram of the experimental set-up used for EC and ECF.

2.2. Electrochemical system

A schematic diagram of the experimental setup used in this work is shown in Fig. 1. A 0.25-L Pyrex glass was used as the reactor. Both anode and cathode electrodes were made of iron plates with a dimension of 20 cm x 3 cm x 0.3 cm (effective area of 20.7 cm²), and the spacing between two electrodes was 1 cm. The experiments were conducted at constant current mode using a direct current power supply (GPS-3030DD, Taiwan) with the variation of electric voltage being recorded using a multimeter (CENTER 122, Taiwan). The reactor was mixed with a magnetic stirrer (Cimarec Stirring Hot Plates, MA, USA) and an octagon magnetic stir bar of 51 mm (VWR Spinbar®) at the agitation rate of 100 rpm. The pH value was maintained only for the ECF experiments with a pH controller (PC3200, Suntext Instruments, Taiwan) using 1 M H₂SO₄.

2.3. Electrocoagulation tests

For EC experiments, a 0.2-L wastewater sample was placed in the reactor with the pH being adjusted to predetermined pH values using 5 N NaOH or 1 M H₂SO₄ solution. After the electric current intensity was set to the desired value, the experiment was started by turning on the power supply. At the end of the reaction, the solution was poured into a sedimentation column and set quiescently for 10 min to separate the sludge from the solution. The final pH of the treated water was recorded after the sludge separation. Sampling was done for the determination of the total dissolved iron concentration under completely mixed condition. Samples for heavy metals and COD concentration analyses were collected and filtered through a 0.45 μm membrane filter. Three replicates were performed, and the mean and one standard deviation from the mean were reported.

2.4. Electrochemical Fenton tests

For ECF experiments, the iron dose was determined based on the optimum iron dose obtained from the EC process. The solution pH level (pH 3) was maintained during the reaction using a pH controller. A desired amount of H₂O₂ was added into 0.2 L

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