



Electrochemical decolorization of textile dyes and removal of metal ions from textile dye and metal ion binary mixtures

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ABSTRACT

The electrochemical textile dye decolorization and removal of metal ions from [textile dye|metal ion] binary mixtures of [AB29|Cu(II)], [RR2|Cu(II)], [AR97|Ni(II)], and [RB4|Ni(II)] were investigated using iron anodes and Na₂SO₄ electrolyte in a batch electrochemical reactor. The independent parameters of current density, electrolyte concentration, initial solute concentrations, and reaction temperature were operated in the range of 0.8–4.0 mA/cm², 0–2 g/L, 0–400 mg/L, and 25–45 °C, respectively. The experimental data were optimized using response surface methodology (RSM), the approximation functions were obtained and the optimum operating conditions for the electrochemical decolorization and metal ions removal from [textile dye|metal ion] binary mixtures were determined. The quadratic models for textile dye decolorization and metal ion removal responses were well satisfied the assumptions of the analysis of variance (ANOVA). At optimized reaction conditions, the operating regions were determined for the efficiencies over 95% textile dye decolorization along with metal ion removal in [AB29|Cu(II)], [RR2|Cu(II)], [AR97|Ni(II)], and [RB4|Ni(II)] binary mixtures. The mean energy consumption values were evaluated between 29.2–38.1 kWh/kg dye decolorization and 14.2–22.5 kWh/kg metal ion removal at response surface optimized operating conditions.

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1. Introduction

Industrialization facilities have unfortunately resulted in increased contamination of the environment by textile dyes and heavy metal ions which can be often found together. Industrial electroplating, acid mine wastewaters, metal finishing, and electronic industries contain various kinds of toxic substances and heavy metals [1,2]. Moreover, waste printed circuit boards (PCBs) contain plenty of toxic materials including heavy metals, hence heavy metal contamination becomes a serious problem due to dramatic growth in electronic industry [3,4]. Heavy metal ions reveal hazardous effects when discharged to the environment and they are not biodegradable unlike organic pollutants [5]. Most of the uncontrolled release of metal ions such as arsenic, cadmium, chromium, copper, mercury, nickel, lead, and zinc are dangerous to human health and the environment as well [6]. Textile dyes are organic chemical substances that are used in coloring of textile materials by dyeing and printing of fibers, yarns and woven/knit apparel. The relatively low fixation efficiency of the dyes results in effluent color, which is not easily removed in treatment systems. Although many methods of color removal exist, none of them works in every case [7]. The wastewater containing toxic com-

ponents such as heavy metal ions and textile dyes from various industries should be treated with effective and efficient methods before being discharged receiving environment of seas, lakes and rivers.

The presence of textile dyes and heavy metal ions in liquid effluents are posing unacceptable influence on both human health and the environment [6]. Aksu et al. [8] indicated the resistance of many synthetic dyestuffs to biological degradation due to their complex aromatic molecular structures whereas heavy metal ions cannot be biodegraded. The authors reported that textile dye color and heavy metal removal by conventional biological processes are difficult and incomplete [8]. In the literature, several chemical, physicochemical and biological techniques including chemical precipitation, adsorption, biosorption, microfiltration, reverse osmosis, coagulation, ion-exchange, electrochemical methods, membrane separation, etc. have been utilized for the removal of textile dyes and heavy metal ions from industrial wastewaters.

Generally, the conventional methods utilized for heavy metal separation are, NaOH or Na₂CO₃ precipitation followed with further filtration, and FeSO₄ or Al₂(SO₄)₃ coagulation followed with further sedimentation [1,2,6,9,10]. However, they result in low effectiveness and high process cost for low level heavy metal containing wastewaters [6], and chemical coagulation can cause secondary pollution by additional chemical substances as well [1,10]. Moreover, chemical precipitation can become ineffective in the presence of strong complexing agents, and high buffer capacity provided

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Nomenclature

COD	chemical oxygen demand concentration (mg/L)
$d\%$	textile dye decolorization (%)
df	degrees of freedom
E	mean energy consumption (kWh/kg dye decolorization, kWh/kg metal ion removal, kWh/kg COD removal)
i	current (ampere)
J	current density (mA/cm ²)
$m\%$	metal ion removal (%)
$[\text{metal ion}]_0$	initial metal ion concentration (mg/L)
n	number of experiments
p	number of model parameters
R^2	coefficient of determination
R^2_{adj}	adjusted coefficient of determination
V_m	mean cell voltage (volt)
V_R	electrolyte volume (L)
SS	sum of squares
t	reaction time (min)
T	reaction temperature (°C)
$[\text{textile dye}]_0$	initial textile dye concentration (mg/L)
x_i, x_j	independent variables
V	variance
\hat{y}	predicted value from the model (response)
<i>Greek letters</i>	
η	response
$\beta_0, \beta_j, \beta_{ij}, \beta_{ij}$	constant, linear, quadratic, and interaction coefficients in the approximating model
σ^2	residual mean square from ANOVA table
Δt	reaction time (h)

by complexing agents requires excessive amounts of chemicals to neutralize alkalinity [11].

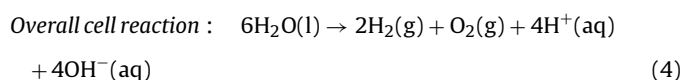
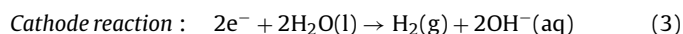
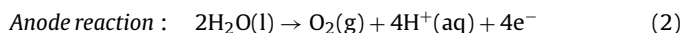
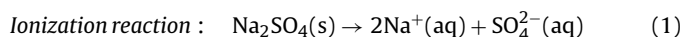
In recent, electrochemical techniques are promising alternative wastewater treatment methods and replacing the conventional processes. The limitations in conventional processes forced several studies on the use of electrocoagulation for the treatment of industrial effluents which is found particularly useful in wastewater treatment [1,12]. The electrocoagulation process involves the generation of coagulants in situ by dissolving electrically using sacrificial anodes of aluminum or iron [12]. Simultaneously, the metal ions generation takes place at the anode with the evolution of hydrogen gas from the cathode [12]. In the electrocoagulation treatment, some of the organic molecules and suspended solids removed by metallic hydroxide sedimentation or H₂ flotation.

The objective of this study was in order to investigate the electrochemical textile dye decolorization and metal ion removal from the binary mixtures of [Acid Blue 29|Cu(II)], [Reactive Red 2|Cu(II)], [Acid Red 97|Ni(II)], and [Reactive Blue 4|Ni(II)] using iron anodes and Na₂SO₄ electrolyte in a batch electrochemical reactor. The influence of operating parameters such as initial dye concentration, initial metal ion concentration, electrolyte concentration, current density and reaction temperature on the removal efficiency of textile dye color and metal ions were analyzed. In the study, pH adjustment, coagulant addition and any other physicochemical processes were not employed in order to enhance the electrochemical treatment performance. This binary mixture study contributes valuable information on the interactions influencing the textile dye decolorization and metal ions removal from binary mixtures, as well as introducing possible underlying mechanisms of the electrochemical process.

2. Electrochemical treatment phenomena

The electrochemical reactions that take place during the electrochemical treatment are rather complicated and electrochemical removal mechanisms of contaminants could not be clearly expressed in many cases. In general, the electrochemical reactions occur simultaneously with competitive and/or cooperative reaction mechanisms, and the pollutants are thought to be removed from wastewater by direct and indirect electrochemical mechanisms [13–15]. In direct electrochemical treatment, molecules were electrooxidized at the anode and electroreduced at the cathode forming smaller intermediate by-products. Although direct anodic oxidation is capable of removing pollutants from wastewater, the direct oxidation reactions of molecules on inert anodes are very slow due to reaction kinetics and limiting reactions [13,14]. The direct oxidation rate of organic pollutants is dependent on the electrocatalytic activity of the anode, on the diffusion rate of organic compounds to the active sites of the anode, and the applied current density [16].

In this study, the electrochemical treatment of binary mixtures were investigated in the presence of Na₂SO₄ supporting electrolyte using iron anodes, and possible electrochemical oxidation and reduction reactions are outlined in Eqs. (1)–(19). Once, Na₂SO₄ dissolves in the aqueous medium it ionizes into Na⁺ and SO₄²⁻. However, water has a greater tendency to be oxidized than SO₄²⁻, and it has a greater tendency to be reduced than Na⁺ electrochemically [17]. During the electrolysis, H⁺ ions are produced at the anode and OH⁻ ions are produced at the cathode. Sodium sulfate does not participate in this electrolysis that it is not consumed at the electrodes. The function of Na⁺ and SO₄²⁻ ions is to sustain the electrical conductance in the reaction medium. Therefore, the net electrolysis reaction is the decomposition of water with O₂ evolution at the anode and H₂ evolution at the cathode [17].



In the electrochemical treatment of contaminants using iron (Fe) anodes, Fe²⁺ ions are added into the solution with sufficient applied voltage by the anodic dissolution reaction [15], and some of the organic molecules and suspended solids captured by iron hydroxides [18–21]. This process is known as electrocoagulation (EC). Coagulation is the phenomenon of agglomeration in colloidal suspension in which charged particles are neutralized by collision with counter ions that is followed by sedimentation [22]. In the electrocoagulation (EC) process, the coagulant is generated in situ by electrolytic oxidation of a sacrificial anode material. In EC process, charged pollutants are removed from wastewater by either reaction with opposite charged ions or by flocculation with generated metallic hydroxides within the reaction effluent [22]. In electrocoagulation, metal ions, colloidal solids and particles, and soluble inorganic pollutants can be removed from aqueous reaction medium by highly charged polymeric metal hydroxide species [22]. The metal hydroxide species neutralize the electrostatic charges on suspended solids and oil emulsions in order to facilitate the coagulation and/or flocculation which results in a separation process [22].

It is known that electrode material greatly influences the effectiveness of the electrocoagulation process [23]. In the literature, iron has been reported to be the most effective electrode material in situ electrogeneration of Fe(II) hydroxides [23]. There are

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