



Comparison of electrocoagulation using iron and aluminium electrodes with chemical coagulation for the removal of a highly soluble acid dye

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ABSTRACT

The decolorization of a synthetic textile wastewater was investigated using electrocoagulation (EC) in a batch electrochemical cell. Orange II, a typical mono-azo acid dye, was used. Aluminum and iron electrodes were compared. The respective effects of operational parameters (initial dye concentration, current density, electrolysis time) and wastewater properties (initial pH and conductivity) were analyzed by comparing the performance of color removal, energy input, electrode mass consumption and the amount of sludge produced. Operating costs were deduced. A comparison with chemical coagulation (CC) using the same amount of metal cations as in EC was also carried out. Experimental results showed that EC maximized decolorization (up to 98%) in comparison to CC (limited to 53%). For EC, iron electrodes exhibited the highest decolorization yield and minimized simultaneously energy requirements, the amount of floc and operation costs in comparison to aluminium. As voltage vs. current curves did not differ significantly between Al and Fe, higher current was required with Al electrodes to achieve a similar decolorization yield. This was partly explained by differences in the decolorization mechanism, as Al electrodes seemed to promote the electroreduction of the azo bond of Orange II, contrary to Fe electrodes.

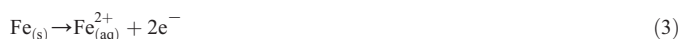
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1. Introduction

Electrocoagulation (EC) is a versatile method for water and wastewater treatment that relies upon the electrochemical dissolution of sacrificial metal electrodes (usually iron or aluminium) into soluble or insoluble species that enhance the coagulation, the adsorption or the precipitation of soluble or colloidal pollutants [1]. As the coagulant is formed progressively in situ when the anode corrodes due to a fixed current or cell potential, EC differs strongly from conventional chemical coagulation (CC) in which the coagulant is added at once. The electrolytic reactions during EC with Al electrodes are metal dissolution (anode) and water reduction (cathode):



For iron electrodes, Eq. (2) prevails at the cathode and it is usually admitted that anodic $\text{Fe}_{(s)}$ oxidation is first limited to Fe(II) cations:



Then, Fe(III) cations are assumed to be obtained from the oxidation of Fe(II) on the electrode, or in the bulk with dissolved oxygen or pollutants [2,3], although alternative mechanisms were proposed [4]. Other possible anodic reactions include water oxidation,



and, in the presence of chloride anions, their oxidation into $\text{Cl}_{2(g)}$ in acidic medium or ClO^{-} anions in alkaline water [5]. All these reactions are pH-dependent. As the electrogenerated cations can react with hydroxide anions resulting from hydrogen evolution at the cathode, Al(III) cations can gradually form monomeric soluble hydroxides or oxyhydroxides that polymerize into insoluble $\text{Al}(\text{OH})_{3(s)}$ when pH is between 6 and 8 [3,6]. The same stands for iron electrodes, except that the insoluble hydroxide species $\text{Fe}(\text{OH})_{3(s)}$ is stable between pH 5 and pH 10 [3,4].

Electrocoagulation has been successfully applied for the treatment of various agricultural [7], industrial [8,9] and urban wastewaters [10], but also for water potabilization [5,11]. EC is, indeed, able to remove a wide variety of pollutants, such as heavy metals [3,12], various anions

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[5,13,14], but also suspended oils droplets and particles [15,16], and organic compounds including dyes [2,17,18]. EC presents many advantages over alternative techniques, such as high efficiency at low capital and operation costs, simplicity and compactness of the equipment required and easiness of process control that results in robustness. In addition, EC eliminates or reduces the need for additional chemicals, and has been reported to decrease drastically sludge production in comparison to CC, with sludge that settles or even flotates easily due to H₂ microbubbles generated at the cathode [17]. Due to the low power requirements of EC, the use electricity from renewable energy sources has also been suggested [19].

EC presents, however, some drawbacks. First, high water conductivity is required to reduce power requirements, which is usually not achieved for water potabilization. Key problems are also the lack of general design of batch or continuous electrocoagulation reactors [20] and the complexity of the mechanisms involved in pollution abatement during EC. For example, cations released at the anode can act through charge neutralization or complexation with anionic species, double layer compression with colloidal particles, but also co-precipitation with Al(OH)_{3(s)} or Fe(OH)_{3(s)}, or pollutant entrapment in these compounds during their precipitation, as well as direct adsorption on the surface of the precipitates. In addition, direct electrode attachment of pollutants was reported [21], but also direct and indirect electrochemical reactions, and oxydoreduction in the bulk with metal cations [3]. As a result, the efficiency of EC strongly depends on alkalinity and co-existing species, particularly anions [22].

For the last decade, the textile industry has had to face up to the social and environmental challenges resulting from the aesthetic impact (strong color, suspended particles...) and the toxicity of wastewater textile effluents. Many dyes are, indeed, toxic to human health and the environment [17]. Roughly, it is estimated that up to 15% of the dyes are released in wastewater due to their limited fixation efficiency [23]. Even though the biological treatments are the cheapest and the simplest, they cannot be applied directly to most textile wastewaters because many dyes are also toxic to the microorganisms involved in these processes. At the moment, alternative physical/chemical methods are used, among which chemical coagulation [24,25] is the most common. However, they present all severe limitations. For instance, CC requires the addition of chemicals that decrease pH and, therefore, may impair the efficiency of pollution abatement [24]. CC also produces large volumes of sludge, requires high treatment times and exhibits a limited effectiveness in the presence of soluble dyes [24,25]. Adsorption [26] and advanced oxidation processes AOP (ozonation, photocatalysis, Fenton, electro-oxidation... [27–29]) constitute another alternative. However, AOP are often expensive and lead sometimes to byproducts that can be more toxic than primary pollutants, although chemical oxygen demand (COD) is reduced. Similarly, adsorption effectiveness is limited and can be improved only using a hybrid CC/adsorption process [26]. EC appears, therefore, as a balanced compromise that is able to remove simultaneously insoluble (dispersive) and soluble dyes, but also COD, turbidity, and suspended solids [17,18].

About 50% of all commercial dyes belong to the azo-dye class [30]. Orange II dye is a mono-azo acid dye, highly water-soluble (116 g/L), widely used in the textile industry in the form of a sodium salt. It is, therefore, able to attach strongly through hydrogen bonding, ionic bonding and van der Waals forces, to cationic groups in natural and synthetic fibers. As a result, Orange II has often been selected as a representative example of the mono-azo acid dye class in studies dedicated to textile wastewater treatment involving EC [2,31,32]. In these works, the influence of wastewater properties (initial pH, conductivity, initial dye concentration) and operation parameters of EC (current density or cell voltage, operation time) is usually investigated. However, a comparison between iron or aluminium electrodes is only available in [31,32] in the potentiostatic mode; similarly, comparisons between CC and EC on Orange II dye are limited. A careful analysis of the literature also shows that energy requirements

and cost analysis of EC operation are hardly reported, even for other dyes, and that electrode mass loss and sludge production are taken into account only in a few papers [33,34].

Consequently, the objective of this study is to investigate the performance of EC for decolorization of a synthetic textile wastewater containing Orange II and to propose a rational and systematic comparison between iron and aluminium electrodes as a function of the above-mentioned wastewater properties and operation parameters. This comparison discusses not only color abatement, but also energy requirements, electrode mass consumption, sludge production and cost analysis. A comparison with conventional chemical coagulation using various coagulants is also provided.

2. Materials and methods

The synthetic textile wastewater was prepared using Casablanca tap water, a moderately alkaline water (120 mg/L CaCO₃), the detailed properties of which are reported in [35]. Orange II, also denoted Acid Orange 7, was supplied by *Sigma-Aldrich Co.* (USA) in the form of sodium salt of the sulfonic acid (Fig. 1); it was added in tap water without further treatment to achieve initial dye concentrations [AO7]₀ between 20 and 200 mg/L. Sodium chloride NaCl (*Fluka AG*, Switzerland) was added to adjust the initial conductivity (κ) of the synthetic wastewater. A minute addition of 0.1 N H₂SO₄ or NaOH solutions (*Fluka AG*, Switzerland) was used to adjust the initial pH, which was varied between 3 and 11. Conductivity was measured using a CDM210 conductimeter equipped with a CDC866T electrode (*Radiometer Analytical*, France), while pH was obtained using a pHM220 pH-meter (*Radiometer Analytical*, France). The Orange II dye content was measured using UV-vis spectrophotometry (*J.P. Selecta*, Spain) at 480 nm. A calibration curve based on absorbance measurements was set up and was linear in the range 0–50 mg/L. For chemical coagulation, aluminium sulphate Al₂(SO₄)₃·18H₂O and ferric chloride FeCl₃·6H₂O (*Sigma-Aldrich Co.*, USA) were used for comparison with EC using Al and Fe electrodes, respectively.

A cylindrical electrocoagulation cell of 250 mL internal volume (V) was equipped with two parallel plates of rectangular shape (4.7 cm × 4.7 cm × 0.1 cm; electrode gap $e = 1.5$ cm), used as anode and cathode, respectively. Either iron or aluminium electrodes could be used in order to compare the influence of electrode metal. Experiments were carried out at room temperature (20 ± 1 °C). Before each run, both electrodes were cleaned, immersed in 0.01 M HCl for 2 min and, amply rinsed with water, air-dried and weighted before use. During EC, a DC power supply (*BK-Precision*, USA) was used in the intensiostat mode; cell voltage (U) was measured. Current (I) was varied in order to achieve current density values (j) between 7.5 and 65 mA/cm². Magnetic stirring at 300 rpm maintained moderate mixing during EC near the electrodes, while preventing the destruction of the flocs during their formation. Electrolysis time (t) was varied between 2 and 10 min. Then, flotation and settling

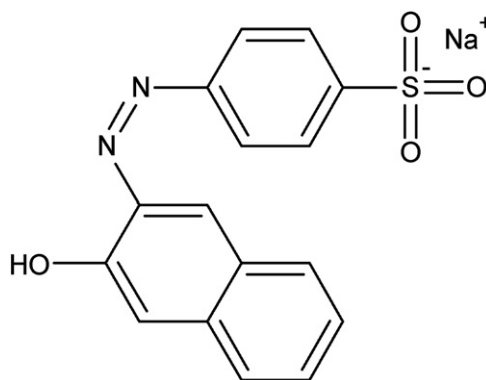


Fig. 1. Molecular structure of Orange II sodium salt (HOC₁₀H₆N = NC₆H₄SO₃Na).

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