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ELECTROCOAGULATION IN WASTEWATER TREATMENT

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ABSTRACT

Within the past decades,water scarcity is one of the greatest challenges faced by humans, sothe treatment and reuse of wastewater generated daily from household, sanitary and industrial activities iswidely practiced and encouraged.Electrocoagulation has emerged as an effectivetechnology for the treatment ofdifferent categories of wastewater. The electrode materials should non-toxic to human health and environment and these usually involve aluminium or iron, but new materials are also used.The efficiency of electrocoagulation depends on current density, gap between anodes and cathodes, electrodes arrangement, composition and shape, initial pollutant concentration, composition and pH of the solution, electrolysis time.This paper aims to reviewthe mechanisms involved in wastewater electrocoagulationand the results of electrocoagulation applied in the treatment of wastewater from different sources.

INTRODUCTION

Fast urbanization and industrialization are generating large amounts of wastewater, which in turn reduces the availability of fresh water. Thedisposal of wastewater to the receiving environment without an appropriate treatmentis threatening the water qualityand human health worldwide. Wastewater treatment plants receive complex mixtures of organic, inorganic pollutants,heavymetals, colorants and nutrients from municipal, sanitary, agro-zootechnical andindustrial sources.

The organic pollutants found inwastewaters are pharmaceuticals, proteins, lignin, insecticides, fungicides, herbicides, phenols, biphenyls, polycyclic aromatichydrocarbons, halogenated aromatic hydrocarbons, formaldehyde, detergents,greases, oils, normal hydrocarbons, alcohols, aldehydes, ketones. Common inorganic water pollutants are heavy metals, nitrates, sulfates, phosphates, fluorides,and chlorides.

Wastewater can be treated physically, chemically andbiologically, or with a combination of those methods. Physical treatment achievesthe separation of pollutantsfrom wastewater without causing a significant change in thechemical or biological characteristics of the treated water.Chemical treatment(adsorption, ion exchange, coagulation, and coagulation-flocculation) is efficient but it requires large amounts of chemical reagents and theyproduce sludge. Biologicaltreatment(activated sludge, algae, anaerobic-aerobic) uses microorganisms for the biodegradation of pollutants inwastewater, aiming to reduce theorganic content and nutrients, but the efficiencyissometimes limited due to the sensitivity ofmicroorganisms to some complex chemical species, longtreatment time, largesurface treatment area and

non-capability of removing some toxicelements.

The electrochemical technologies rely on physico-chemical processes and have received great consideration for their capability in treating different types of wastewater with different electrode material and configuration [28]. The employment of electrochemical technologiescan be carried out easily, with complete automation, without direct addition of chemical agents[2], and without secondary pollutiondue to high concentration of chemicals.

Electrochemical treatmentsincluding electrochemical oxidation, electrocoagulation, peroxi-coagulation, anodic oxidation, electro-Fenton, are very effective in treatingmunicipal, livestock and industrial wastewater [27, 44].

Electrocoagulation(EC) was first developed andpatented in 1906 by A. E. Dietrich for the treatment of bilge water from ships.In 1984, in the US, a large scale drinking water treatment by EC was implementedfor the first time[6].ECis an environmentallyfriendly technology that combines the benefits of coagulation,flotation and electrochemistry.EC has been applied for the treatment of domestic, agro-industrial and industrialwastewaters.EC has been successfully employedin removing suspended solids, colloidal material and metals, as well asother dissolved solids [3].EC treatment gives clear, colorless and odorless water. Even the smallest colloidal particles in wastewater can be removed by EC, because the appliedelectric current makes collision faster and facilitates coagulation.

Compared to other electrochemical methods and chemicalcoagulation/flocculation, EC has lower costs, is simpler to operate, it generatesless sludge [17], it doesn't require additional chemicals to enhance the process [14], it has amenability to automation,ease of control and environmental compatibility [45]. There are also disadvantages of EC, such as the consumptionof the electrode with time and its corrosion, so regular electrode replacement must be made for better performance [9].EC is an accelerated corrosion process.After a certain time, an impermeable oxide layer is formed on thecathode, preventing effective transmission of current within thesystem, leading to higher power costs and lower efficiency. Thisproblem is enhanced when using Al electrodes [20].To preventthe passivation of the electrodes and the formation of sediment on theirsurface, electrodes polarity should be changed at regular intervals of time [14].

MATERIAL AND METHOD

The EC electrodes are mostly made of non-hazardous, widely available and cost effective materials such as Al and Fe [45], mild steel and stainless steel [23] which istraditional anodically soluble metals. Efficiency of EC using Fe electrodes is limited in real wastewater, due to the absence of sufficient dissolved oxygen required for converting electrolytically generated ferrous ion into ferric ion [32]. Fe (II) is a weak coagulant compared to Fe (III) due to its lower positive charge [28].In addition to Al and Fe, dimensionally stable anodes, such as PbO_2, SnO_2, graphite, boron-doped diamond electrodes, the most common insoluble anodes, have higher chemical resistance and efficiency in wastewater treatment [40]. New electrode materials such as magnesium may contribute to nutrient recovery, as magnesium-phosphate complexin addition to enhanced operations like stabilization [26].

Typically, anyEC equipment is composed of DC-powered cathode and anode electrodes, which are partially submerged into a wastewater tank (Fig.

1). Reactor design consists of two or more conducting electrodes that are immersed in a tank filled with an electricity-conducting liquid called the electrolyte [1].The EC system works in batch or continuous mode, by treating a fixed volume of waste effluent per process cycle or by treatment of a continuous flow of the waste stream.Some EC systems are fitted with fast mechanical stirrers (Fig. 2) to help the coagulant dispersion [39].

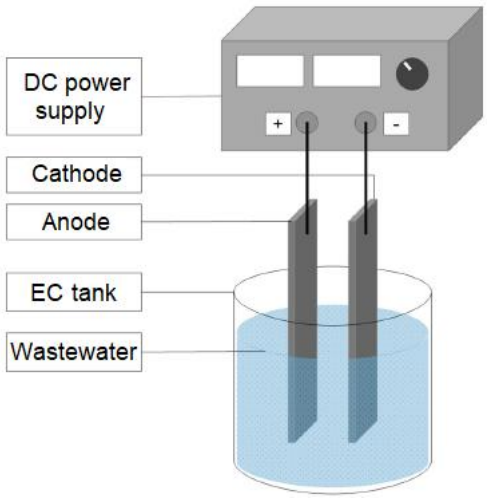

Fig. 1. Simple EC system [39]

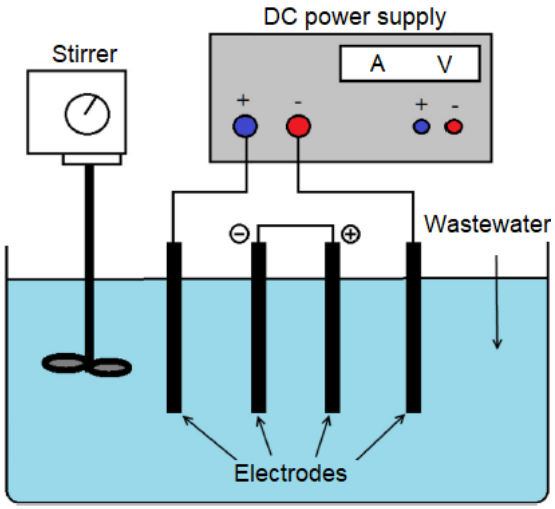

Fig. 2.EC system with stirrer[30]

In EC systems, coagulants are generated in situ through electrolytic oxidation. Electrolysis is based on the dissolution of (sacrificial) anode when an external current, so the dissolution is the result of the application of potential difference across the electrodes [9]. During electrolysis in aqueous solutions at pH values close to neutral, the organic compound anions can be oxidized directly on the anode [33].

Ions of Al^{3+} and Fe^{2+} flowing from the sacrificial anode generate the coagulants: $Al(OH)_3$, respectively$Fe(OH)_3$ which bind the pollutants. Once in solution, the Al^{3+} and Fe^{2+} ions react with OH^- groups and hydroxides are formed, which will further entrap the pollutants by electrostatic attraction or by complexation (Fig. 3).

With Al electrodes, reactions that take place in the EC systemare [31]:

Anode:

$$Al_{(s)} \rightarrow Al^{3+} + 3e^- \quad (1)$$

Cathode:

$$3H_2O + 3e^- \rightarrow 1.5H_{2(g)} + 3OH^- \quad (2)$$

Once Al has solubilized, longer-chain Al hydroxidescan develop depending on contact time and pH [13]. $Al^{3+}$and OH^-ions react to form various monomeric species which will transform into amorphous form $Al(OH)_3(s)$.

Hydroxide formation:

$$Al^{3+} + 3H_2O \rightarrow Al(OH)_3^+ + 3H^+ \quad (3)$$

The$Al(OH)_3(s)$ flocs have large surface areas which adsorb the soluble organic compounds and trap the colloidal particles. As observed in Figure 4, the removalof flocsin the EC cell takes place by sedimentation (the heavier flocs settle at reactor bottom) and electroflotationdueto H_2 small bubbles generated from the cathode [21].

Similarly, when using Fe at the anode in basic wastewater, Fe^{2+} is generated by the anodic oxidation ofiron while OH^- is produced by the cathodic reduction of H_2O at the cathode, as seen in reactions 4 and 5 [50]:

Anode:

$$Fe_{(s)} \rightarrow Fe^{2+}_{(aq)} + 2e^- \quad (4)$$

Cathode:

$$2H_2O_{(l)} + 2e^- \rightarrow H_{2(g)} \uparrow + 2OH^-_{(aq)} \quad (5)$$

Fe^{2+} is an active coagulant precursor which forms insoluble iron hydroxide

$Fe(OH)_{2(s)}$ and $Fe(OH)_{3(s)}$ (reactions 6 and 7) which act as coagulant/flocculent for the suspended solids and lead to high density flocs which will settle afterward.

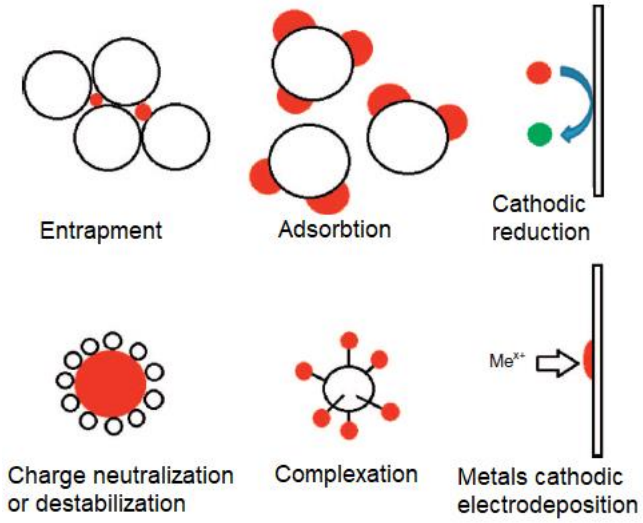

Fig. 3.Pollutantsremoval by EC [18]

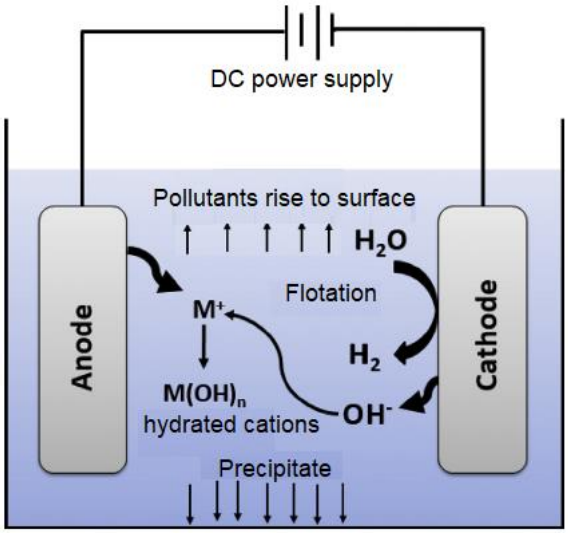

Fig. 4. Basic processes in EC cells [28]

Hydroxide formation:

$$Fe^{2+}_{(aq)} + 2OH^{-}_{(aq)} \rightarrow Fe(OH)_{2(s)} \quad (6)$$

$$Fe_{(s)} + 2H_2O_{(l)} \rightarrow Fe(OH)_{2(s)} + H_{2(g)} \uparrow \quad (7)$$

The production of H_2 at the cathode helps the suspended particles to float on the surface (electro-flotation) and thenon-soluble particles that settle according to their size and density can be removed by filtration [50].

When using Mg at the anode [13]:

$$Mg_{(s)} \rightarrow Mg^{2+} + 2e^{-} \quad (8)$$

At the cathode, regardless the electrode material, the reaction is[13]:

$$2H^{+} + 2e^{-} \rightarrow H_{2(g)} \uparrow \quad (9)$$

There are many available shapes, sizes and numbers of electrodes, but in practice the most used ones are rectangular-shaped plates [19]. Plate electrodes are a block of plates grouped perpendicularlyand separated by washers so that a gap existsbetween adjacent plates where the solution flows perpendicularly to the direction the electric current flow [14].

The need forwide electrode surface area to overcome themetal dissociation rate is solved by using monopolar-parallel (MP-P), monopolar-series (MP-S) or bipolar-series (BP-S)electrodes connections (Fig. 5). The choice of the appropriate electrode connection is determinedby the removal efficiency and treatment cost [28].

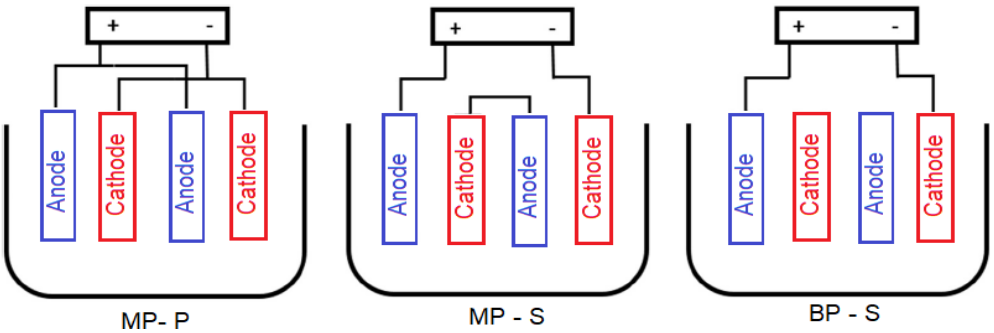

Fig. 5. Possible configurations of electrode connections[28]

EC is influenced by thedensityof applied current, reactiontime, nature of anode/cathode, wastewater pH and the distance between the electrodes [36]. Current density is the most important parameter for controllingthe reaction rate

within the electrochemical reactor. Current density determines the coagulant dosage at the anode and the formation of H_2gas at the cathode [25].An important factor affecting EC performance, particularly the coagulation mechanism,is the pH which governs the hydrolyzed metal species formed in electrolyte media [34]. Inter-electrode distance is an important variable with regards to EC's operational costs. At high values of effluent conductivity, greater inter-electrode distanceis recommended [11]. Consumption of energy and electrode during the EC treatment are important and affect the economic feasibilityof the process [45]. Less energy is consumed with decreasing the gap between electrodes. As thedistance between electrodes decreases, more gas bubbles are generated,leading to higher mass transfer and higher reaction rate between coagulants and pollutants [19].

EC in combination with alternative treatment methods is a safe and effective way to remove pollutants andhas been applied for the treatment of:polluted drinking waterlike the removal of fluoride from drinkingwater, textile industry wastewater (EC-nanofiltration and EC-advanced oxidation process), reuse of textile wastewater (EC-ozone, Fig. 6), treatment and reuse of laundry wastewater (EC-electroflotation, Fig. 7), brackish water (EC-reverse osmosis), landfill leachate (EC-biofiltration), etc.

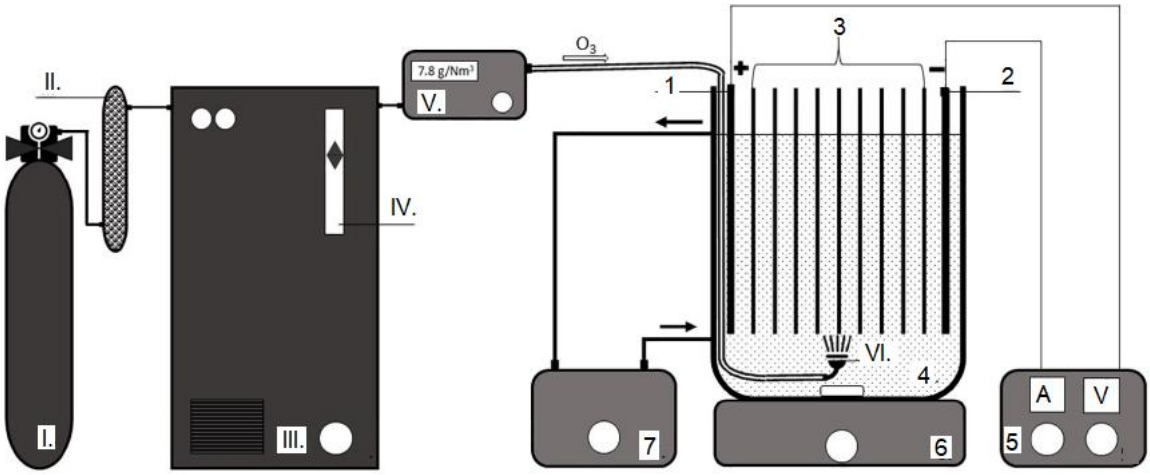

Fig. 6. Combined EC - O_3 process [7]

O_3 supply system: I. gas (O_2) cylinder; II. gas dryer; III. O_3 generator; IV. rotameter; V. O_3-meter;VI. diffuser; EC system: 1. mono-polar anode; 2. mono-polar cathode; 3. bi-polar electrodes; 4. electrochemical cell; 5. power supply; 6. magnetic stirrer;7. circulation pump

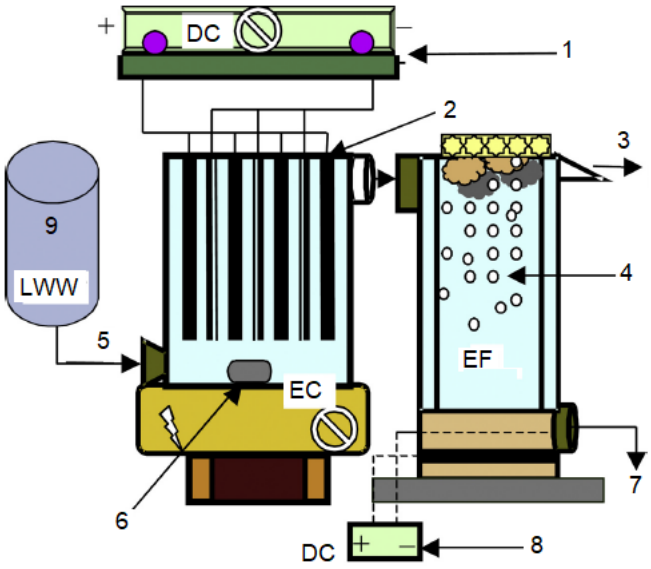

Fig. 7. Combined EC - EF system [14]

1. DC power apply for EC cell; 2. EC cell; 3. floated sladge; 4. EF cell; 5. inlet LWW; 6. magnetic bar-stirrer; 7. purified LWW; 8. DC power apply for EF cell; 9. feed sludge tank

RESULTSAND DISCUSSIONS

So far, electrocoagulationwas successfully employed in treating municipal wastewater, dairy wastewater,slaughterhouse wastewater, restaurant wastewater, canola-oil refinery effluent, palm oil mill effluent, distillery wastewater, carwash wastewater, tannery wastewater, industrial estate wastewater, metal plating/ electroplating wastewater, textile wastewater, printing ink wastewater, pulp and paper mill wastewater, leachate wastewater, heavy oil / petroleum refinery wastewater, de-fluoridation of groundwater, removal of polyethylenemicrobeads (microplastic contaminants) from wastewater, recovery of microalgae and plant extracts.

EC is effective in removing organic matter, turbidity, color, phenol, phosphate, heavy metals, oils and greases [34], refractorypharmaceutical compoundsfrom synthetic wastewater [16] and veterinary antibiotics [4] known as emerging contaminants (they are biologically active, can persist and bioaccumulatein the environment).

Solar powered EC system was applied successfully forthe municipal wastewater and the removal efficiencies for current density of 48 A/m^{2}and hydraulic detentiontime of 16 minwere 90% for COD,94.56% for turbidity and 49.78% for TDS [31].

EC was found to be effective for the removal of polyethylene microbeads(microplastics) from wastewater streams. Removal efficiencies over 90% were found at pH values ranging from 3 to 10. The optimum removal efficiency of 99.24% was foundat a pH of 7.5 [37].

Baran et. al. (2018) have used EC with low-carbon steel electrodes and Al anode, and obtained a decrease in concentration of ampicillin, doxycycline, sulfathiazole and tylosin in vererinarywastewater decreased 3.6 ± 3.2%, ~100%, 3.3 ± 0.4% respectively 3.1 ± 0.3%. Doxycycline was the only antibiotic effectively removed from wastewater during electrocoagulation [4].

Different electrode configurations were reported in the treatment of dairy wastewater: iron parallel plate electrode [41], aluminium two parallel platesand platinized titaniumelectrodes [5], rectangular iron cathode compartment and parallel plate anodes [43].

Boudjema et al. (2016) have found that EC is particularly effective in the removal of natural organic matter [8].Cheballah et al. (2015) have reported 95.95% effectiveness of CODremoval from industrial wastewater using EC [10]. ECwasalso tested as a pretreatment for olive mill wastewater, in which most phenolic compounds were polymerized.Biodegradability was measured by the BOD_5/COD ratio, whose value must be less or equal to 0.5. After EC, the solubleCOD decreased to 33.6% of the initial value (initial COD = 36,900 mg/L)and the BOD_5/COD ratio increased to 0.58 (initialBOD_5/COD = 0.33). This proves the ability ofEC using iron electrodes to eliminatethe soluble compounds in olive mill wastewaters [24].

EC was applied in a comparative study to treat canola oil refinery wastewaters. Total chemical oxygen demand (TCOD), soluble chemical oxygen demand(sCOD), total organic carbon (TOC), dissolved organic carbon (DOC) and total suspended solids (TSS) weremeasured. EC was significantly successful in removing suspended andcolloidal pollutants and could remove > 90% TCOD and 80% of TOC at current densities between0.91-13.66 mA/cm^2. The maximum removal of dissolved organic pollutants was relativelylow: 75% for sCOD and 74% for DOC [42].

When treating textile wastewatercontaining a variety oftextile dyes and complex chemicalsby EC combined with alternative treatments, a retention rate > 68% was observed for

BOD, COD, turbidity, color,suspended solids, Cl^-, NO_3, SO_4, NH_3[46].In an EC process using Fe-Al as composite electrode atits optimized operating conditions, virtuallycomplete color and 90% COD removal efficiency was achieved [45].EC using Fe and Al electrodes was applied for the treatment of printing ink wastewater with high COD values (10,000 mg/L). 75% COD removal, respectively ~99% color removal rates were obtained [35].

Dura and Breslin (2019)have conducted an EC test for simultaneous removal of phosphates, Orange II and Zn ions from a synthetic wastewater. Two stainless steel anodes, AISI 420 and AISI 310, and pure Fe were compared at a current density of 11.7 mA/cm^2.88% and 99%removal efficiencieswere observedwith AISI 420 and pure Fe, and significantly lower values(30%) were obtained with AISI 310 who performed well in Zn^{2+}removal due to itsremoval as $Zn(OH)_2$[15].

Deghles and Kurt (2016) have used combined EC/electrodialysis process to treat tannery effluent. The EC unit had five pairs of electrodes (Fe or Al) inmonopolar parallel mode. A bipolar membrane electrodialysis with platinized titanium electrode as anode and cathode in a pilot scale was used to treat EC effluent. Fe electrodes achieved a removal efficiency of 87% COD, 100% NH_3-N, 100% Cr and 100% color. Al electrodes achieved a removal efficiency of 92% COD, 100% NH_3-N, 100% Cr and 100% color [12].

Kabdasli et al. (2010) investigated the effectof H_2O_2 addition to the EC (combined EC / Fenton) process in order to enhance the organic matter removal efficiency for a metal plating wastewater. The highest COD and TOC removal efficiencies were obtained for the combined EC / Fenton process in the presence of 20 mM H_2O_2, when the organic matter mineralization level increased from 50% to 70%, with complete removal of heavy metals [23].The combination of EC and ozonation, both asone-step ($EC+O_3$) and two-step ($EC \rightarrow O_3$) treatments, gave very good results in color removal and moderate results in the mineralization of RB5 aqueous solution and wastewater containing this dye [7].

Nanseu-Njiki et al. (2009) have applied EC to treat synthetic solutions containing Hg(II) and anodic redissolution in the differential pulse mode as coupled electroanalysis after the EC treatment, which allowed to optimize the EC parameters to obtain Hg removal of99.95% [29].

Xu et al. (2009) have found that by using multi-staged EC, 99% removal from boron concentration was completed after the fifth stage (from 500 mg/L to less than 0.5 mg/L) at current density of 62.1 A/m^2 [47].

Zaroual et al. (2009) have obtained 91% removal efficiency for treating Cr(III) with Al anodes by EC [48].

CONCLUSIONS

Electrocoagulation isan efficient wastewater treatment that implies reactions of oxidation and reduction, in which the suspended, emulsified, or dissolved pollutants are destabilizeddue to the application of electric current to theelectrolytic solution.

The pairs of electrodes are usually made of Al or Fe, but advancements in research have allowed new materials to be tested and applied with good removal rates of pollutants. The coagulating ions are generated in situ by the dissolution of the sacrificial anode, which resultsin much less sludge generation compared to other treatment methods. Different electrode materials give different effectiveness ofwastewater treatment by electrocoagulation.

Electrocoagulation combines the benefits of coagulation,flotation and electrochemistry.This emerging technology has been widely applied to treat different wastewater, andis capable of high removal efficiencies of color, turbidity, COD and BOD.To increase the removal of pollutants, electrocoagulation can be combined with other treatments.

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