# Electrocoagulation as an eco-friendly River water treatment method

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## Abstract

Electrocoagulation (EC) is an effective water and wastewater treatment technology; where the coagulants are produced in-situ by electrolytic oxidation of a sacrificial anode. In this technique, pollutant removal is done without adding chemicals; therefore, it remarkably reduces the sludge produced, and consequently reduces the cost of sludge handling. This method has been efficiently used to remove, up to 99%, of a wide range of pollutants such as heavy metals, oil, dyes, and fluoride. Therefore, the EC method could be the cost-effective, safe, and reliable option to face the growing water scarcity. However, like any other treatment method, the EC technology still has some drawbacks that could limit its applications.

This chapter has been therefore devoted to present the principles, history, applications, limitations, advantages and disadvantages of the electrocoagulation technology, the role of key operating parameters on the performance of the EC reactors, and highlight the differences between the traditional coagulation process and EC technology. More importantly, this chapter will highlight the defects of EC technology that need to be enhanced.

Keywords: Electrocoagulation; phosphate and nitrate; heavy metals; advantages and disadvantages.

## **1.** Chapter Introduction

Water is the most essential element to ensure the reliability and sustainability of the ecosystem of our planet. Although 1400 million cubic kilometres of water covers 71% of the earth surface, less than 2.5% of this huge amount is fresh water (Fogden and Wood, 2009). A vast amount of this fresh water is captured in glaciers, snowy mountain ranges, and groundwater, which leaves less than 1% safe for drinking purposes (Fogden and Wood, 2009; Kuokkanen, 2016). Moreover, with the increasing

population and industrial growth that generate billions of litres of polluted wastewater every day, fresh water resources are becoming limited and/or contaminated (Holt *et al.*, 2005; Linares-Hernández *et al.*, 2009). According to the WHO/Unicef (2000), at the turn of the last century, about 1.1 billion people were without access to a safe drinking water source. Furthermore, some forecasting studies confirm that by 2050 the world's population will experience severe drinking water scarcity (Emamjomeh, 2006; Fogden and Wood, 2009). It is believed that by 2050 more than 50% of the world's population will not have an access to safe drinking water sources (Fogden and Wood, 2009).

To reverse this downward trend, a board range of treatment technologies have been developed and applied to treat water and wastewater such as chemical precipitation, biological treatment, electrocoagulation, ion exchange, ultrasound, photo-degradation and membrane filtration (El-Naas *et al.*, 2014; Katal *et al.*, 2014; Swelam *et al.*, 2015; Alattabi *et al.*, 2017b; Alattabi *et al.*, 2017a). Amongst these methods, the electrocoagulation (EC) method has recently gained increasing popularity as a promising alternative to treat polluted water and wastewater because it bears many environmentally and economically attractive merits (Kuokkanen, 2016; Hashim *et al.*, 2017a).

## 2. Definition of EC process

Electrocoagulation is a process of in-situ production of coagulants by applying an electric current through metallic electrodes to remove suspended pollutants in liquors (Heffron, 2015; Hashim *et al.*, 2017c; Shaw *et al.*, 2017). This method consists of three main stages: generation of coagulant agents (destabilising agents), destabilisation of pollutants, and flocs formation (Heffron, 2015). Initially, the destabilising agents (such Al and Fe) are electrochemically generated from the sacrificial electrodes, these agents destabilise pollutants due to providing an opposite electrostatic charge. Once charged, the pollutants start the bridging process forming flocs that can easily be separated from the solution (Emamjomeh, 2006; Heffron, 2015). Therefore, it can be said that the EC method utilises the advantages of three traditional treatment methods: traditional chemical coagulation, flotation, and electrochemistry (Kuokkanen, 2016).

## 3. Principles of the EC process

The principle of the EC process is the application of an electrical current to the solution being treated through sacrificial metallic electrodes to form in-situ coagulating ions (Chaturvedi, 2013; Heffron, 2015; Hashim *et al.*, 2017b). Selection of the material of the metallic electrodes is dependent on several key parameters such as material availability, cost-effectiveness, oxidation potential, toxicity, and the

properties of the targeted pollutant (Chaturvedi and Dave, 2012; Kuokkanen, 2016). The literature shows that several materials, such as stainless steel(Abuzaid *et al.*, 2002), diamond (Cañizares *et al.*, 2008), iron (Malakootian *et al.*, 2010), zinc (Vasudevan *et al.*, 2012), graphite (Raju *et al.*, 2008), and aluminium (Hashim *et al.*, 2015; Vidal *et al.*, 2016; Hashim *et al.*, 2018), were used as electrodes in EC units. However, it is well documented that aluminium is a very effective and efficient, as an electrode material, to remove different pollutants at suitable operating conditions (Emamjomeh, 2006; Essadki *et al.*, 2009; Chaturvedi and Dave, 2012; Heffron, 2015).

As the electrical current starts to flow through the immersed electrodes, the anode starts to dissolve forming coagulating ions, while the cathode generates hydroxide  $(OH^-)$  ions and hydrogen  $(H_2)$  gas (Chen, 2004; Essadki *et al.*, 2009; Mechelhoff *et al.*, 2013; Heffron, 2015). The EC reactions are determined by several operating parameters such as the initial pH, applied current, electrolysing time, solution conductivity, and electrode material (Bard and Faulkner, 2001; Emamjomeh, 2006; Kobya *et al.*, 2014).

In the case of using aluminium (Al) as an electrode material, the anode liberates  $Al_{(aq)}^{3+}$  ions, while the cathode produces H<sub>2</sub> gas (Essadki *et al.*, 2009; Chaturvedi, 2013); these reactions are represented in the following equations (Chen, 2004; Essadki *et al.*, 2009; Heffron, 2015):

#### At the anode:

$$Al_{(s)} \to Al^{3+} + 3e^{-} \tag{1}$$

#### At the cathode:

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

After dissolution, the  $Al_{(aq)}^{3+}$  ions undergo several reactions to yield different monomeric and polymeric substances such as  $Al(OH)^{2+}$ , and  $Al_{13}O_4(OH)^{7+}_{24}$ , which in turn readily coagulate pollutants forming flocs (Adhoum *et al.*, 2004; Essadki *et al.*, 2009; Heffron, 2015). In fact, speciation of aluminium (hydrolysation of dissolved ions) is highly governed by the pH of the solution being treated, where  $Al(OH)^{2+}$  is the prevailing species of aluminium for pH range of 5 to 6,  $Al(OH)^{4-}_{4}$  is the predominant species for pH above 9, while  $Al(OH)_3$  is predominant in neutral or slightly alkaline pH (Vasudevan *et*)

*al.*, 2009; Heffron, 2015). Generally, aluminium speciation is modelled as follows (Holt *et al.*, 2002; Heffron, 2015):

$$Al^{3+} + H_2 0 \rightarrow Al(OH)^{2+} + H^+$$
 (3)

$$Al(OH)^{2+} + H_2O \rightarrow Al(OH)_2^+ + H^+$$
 (4)

$$Al(OH)_{2}^{+} + H_{2}O \rightarrow Al(OH)_{3}^{0} + H^{+}$$
 (5)

$$Al(OH)_{3}^{0} + H_{2}O \to Al(OH)_{4(aq)}^{-}$$
 (6)

# 4. Pollutants adsorption and precipitation in EC method

Since the electrolysing process started, an electrophoretic immigration of negatively charged ions started towards the anode and positive ions move towards the cathode. This electrophoretic movement works to concentrate the negatively charged colloids near to the anode, which in turn greatly increases their collision rate with the coagulant species, and aggregates formation consequently (Den and Huang, 2005; Chaturvedi, 2013; Shim *et al.*, 2014). Colloids in the EC method are aggregated by adsorbing them into the freshly formed flocs, and/or chemical transforming (Zhu *et al.*, 2007; Essadki *et al.*, 2009; van Genuchten *et al.*, 2014). Aluminium and iron hydroxides have a high affinity for dispersed negatively charged colloids, especially metallic ones, that leads them to coagulate forming large aggregates (Gomes *et al.*, 2007; Ferreira *et al.*, 2013; Heffron, 2015). Positively charged metallic ions can bond themselves to  $OH^-$ , such as  $Cu(OH)_2$  and  $Cu(OH)_2$ , leading to coagulation and forming, with aluminium hydroxide, large flocs whose separation is easily obtained (Ferreira *et al.*, 2013). In addition, colloids and hydroxides may also co-precipitate by replacing ions in the floc structure (Essadki *et al.*, 2009; Heffron, 2015).

For instance, fluoride ions readily react with  $Al^{3+}$  ions forming  $AlF_6^{3-}$  (Mameri *et al.*, 1998; Zhu *et al.*, 2007; Essadki *et al.*, 2009) according to the following equation:

$$Al^{3+} + 6F^- \rightarrow AlF_6^{3-} \tag{7}$$

Which could undergo more reactions, in the presence of sodium ions, to form salt as follows:

$$AlF_6^{3-} + 3Na^+ \rightarrow Na_3AlF_6 \tag{8}$$

Chromium removal using iron electrodes is another good example of the high affinity of iron hydroxides metallic ions, where dichromate ions are readily reduced by ferrous ions according to the following equation (Heffron, 2015):

$$Cr_2O_7^{2-} + 14H^+ + 6Fe^{2+} \rightarrow 2Cr^{3+} + 6Fe^{3+} + 7H_2O$$
 (9)

During the final stage of the electrocoagulation process, the formed aggregates will be separated from the liquid being treated by either flotation or sedimentation. Flotation is the separation of particles from an aqueous media by electrolytic gas; while the bubbles are moving upwards, aggregated colloids adhere to these bubbles and float to the surface of the liquid to be skimmed later (Holt et al., 2005; Emamjomeh, 2006; Butler et al., 2011). While sedimentation takes place when the weight of the aggregated colloids become big enough to settle to the bottom of the EC unit due to the gravity, and this will be removed later as sludge (Emamjomeh, 2006; Chaturvedi and Dave, 2012). The dominant one of these two removal paths is determined by the magnitude of the applied current density; sedimentation is the dominant path at low current densities as the number of generated bubbles will not be enough to float the targeted pollutants. While, at high current densities, large amount of the electrolytic bubbles will be generated, which makes flotation is the predominant path (Holt *et al.*, 2005; Chaturvedi and Dave, 2012). The number of bubbles produced in the EC unit explains this shift in removal mechanism, lower current densities produce an insufficient number of bubbles to float aggregates, making sedimentation predominant. As the current density increases, the number of bubbles increases, leading to aggregates floating to the surface, i.e. it gives the flotation path an edge over sedimentation (Holt et al., 2005; Maximova and Dahl, 2006; Chaturvedi and Dave, 2012).

The pollutants removal efficiency (Re%) can be calculated using the following formula (Un *et al.*, 2013):

$$Re\% = \frac{C_0 - C_t}{C_0} \times 100\%$$
(10)

Where  $C_0$  and  $C_t$  are the initial and final concentrations of the studied pollutant.

## 5. Energy consumption of EC units

Energy consumption is one of the most important parameters in water treatment as it determines the operating cost of any method, which in turn determines its applicability (Bayramoglu *et al.*, 2004; Ozyonar and Karagozoglu, 2011). Power consumption for EC units, which highly depends on the gap

between the electrodes and conductivity of liquid being treated, can be calculated using the following Equations (Emamjomeh, 2006; Un *et al.*, 2013):

$$Power(Watts) = V_{cell}. I$$
(11)

Where  $V_{cell}$  and I are the cell potential (volts) and the applied electric current (amperes).

**Power consumption** 
$$(C_{power})(kWh) = \frac{I.V_{cell} \cdot t}{1000}$$
 (12)

Where *t* is the treatment time (hours).

The consumed power per cubic meter of treated solution is:

$$C_{power} (kWh/m^3) = \frac{I.V_{cell} \cdot t}{1000. Vol.}$$
(13)

Where *Vol*. Is volume of treated liquid (m<sup>3</sup>).

#### 6. Dissolving rate of anode

The dissolved mass (dose) of the sacrificial anode is governed, as previously mentioned, by a number of parameters such as the magnitude of the applied current and treatment time. In fact, the applied current and treatment time are the most significant parameters in the anodic dissolving process as they determine the number of liberated ions from the metallic anode (Bayramoglu *et al.*, 2004; Emamjomeh, 2006; Pallier *et al.*, 2011; Vidal *et al.*, 2016). Theoretically, the dissolved amount of the anode can be calculated using Faraday's second Law (Ozyonar and Karagozoglu, 2011; Un *et al.*, 2013; Kobya *et al.*, 2014; Vidal *et al.*, 2016):

$$C_{material} = \frac{I.t.m}{Z.F.Vol.}$$
(14)

Where  $C_{material}$ , *t*, *m*, *Z*, and *F* represent the dissolved mass from the anode (g/L), treatment time (second), molecular weight (g/mol) (26.98 for Al), number of electrons (3 for Al), and Faraday's constant (96,500 C/mol).

Experimentally, the consumed mass of electrodes can be calculated by measuring the difference between the anode before and after each run.

## 7. Operating cost (OC) of EC units

The operating cost of EC process consists of operating and fixed costs. The operating cost could be defined as all expenditures during the operation the treatment process, such as the cost of electricity, consumed chemicals and electrodes material, and employment, while the cost of other parts of the EC plant, costs of equipment and construction, represent the fixed costs (Bayramoglu *et al.*, 2004; Ghosh *et al.*, 2008; Dalvand *et al.*, 2011). However, the operating cost of bench scale EC units (Lab work) includes the costs of energy and materials (Ghosh *et al.*, 2008; Kobya *et al.*, 2010; Ozyonar and Karagozoglu, 2011; Chopra and Sharma, 2012). The following equation was recommended by Kobya *et al.* (2010) to estimate the operating cost of the electrocoagulation process:

$$OC = \gamma_1 C_{power} + \gamma_2 C_{electrodes} + \gamma_3 C_{chemicals}$$
(15)

Where  $C_{power}$  (kWh/m<sup>3</sup>),  $C_{electrodes}$  (kg/m<sup>3</sup>), and  $C_{chemicals}$  (kg/m<sup>3</sup>) are the consumed power, electrode material, and chemicals respectively. While  $\gamma_1, \gamma_2$  and  $\gamma_3$  represent the unit price of power, electrode material, and chemicals respectively.

### 8. Applications of EC method in the treatment of water and wastewater

It is well documented that the EC method has broadly been used in the water and wastewater treatment field to remove different pollutants such as heavy metals, dyes, oil, and bacteria (Genc and Bakirci, 2015; Wan *et al.*, 2011; Ricordel *et al.*, 2014; Vidal *et al.*, 2016). Overall, EC treatment method could remove as much as 95-99% of the targeted pollutants (Gomes *et al.*, 2007; Gao *et al.*, 2010; Butler *et al.*, 2011; Ricordel *et al.*, 2014; Vidal *et al.*, 2016). However, the performance of this technology is influenced by several parameters such as the chemistry of the liquid being treated, chemical properties and concentration of the targeted pollutant, and electrodes (Golder *et al.*, 2007; Dubrawski and Mohseni, 2013; Wan *et al.*, 2011; Kenova *et al.*, 2015).

Therefore, this section is devoted to presenting a quick review of earlier applications of EC technology in water and wastewater treatment. This literature survey provides important information and data on the applications of the EC technology, which can be used as a guide to conduct future experimental works. The applications of EC technology discussed here were divided into the following five groups:

#### I. Removal of dyes

Treatment of water or wastewater containing dyes using EC method has become one of the most attractive methods during the last few decades due to its high efficiency and cost-effectiveness (Kim *et al.*, 2002; Daneshvar *et al.*, 2006; Aoudj *et al.*, 2010; Naje *et al.*, 2016). For instance, Ogutverena *et al.* (1992) used a bipolar packed bed EC unit supplied with soluble electrodes to remediate Acilan Blau dye from wastewater, taken into considerations the influence of several operating parameters such as pH, and current density. 98-100% of dye was removed within 3-5 min and the power consumption was 2.24 kW/m<sup>3</sup>. Jia *et al.* (1999) treated more than 20 types of dyes; such as sulfur brilliant green GB, Vat red F3B, and Vat blue RSN, using electrodes made from active carbon fibre. Overall, 90% of the studied dyes were removed within 60 min of electrolysing at voltage of 25 V and 0.5 g/L of Na<sub>2</sub>SO<sub>4</sub>. Daneshvar *et al.* (2007) investigated the influence of dye concentration, initial pH, current density, and duration of electrocoagulation on the removal of Acid yellow 23 dye. The findings of this study demonstrated that the best removal efficiency (98%) was obtained after 5 min of electrolysing at an initial pH of 6, current density of 11.25 mA/cm<sup>2</sup>, and initial dye concentration of 50 mg/L.

Naje *et al.* (2016) investigated the removal of Imperon violet KB dye from textile wastewater using a rotated bed EC unit (made from aluminium). 98.5% of this dye was removed after 10 min of electrolysing at a current density of 4 mA/cm<sup>2</sup>, initial pH of 4.57, and water temperature of 25 °C.

#### II. Removal of heavy metals

Application of the EC method to treat water and wastewater containing heavy metals has gained substantial interest during the last few years, and removal efficiencies of 95 to 100% have been documented (Öğütveren *et al.*, 1994; Mills, 2000; Yilmaz *et al.*, 2007; Vasudevan *et al.*, 2011; Heffron, 2015).

Öğütveren *et al.* (1994) used a bipolar packed bed EC reactor with steel ring electrodes to treat water containing 100 mg/L chromium, taking into account the influences of some operating parameters such as time of treatment, and the applied potential. The authors found that the chromium was completely removed after 20 min of electrolysing at a current of 160 mA, power consumption of 33.33 kW/m<sup>3</sup>, and 0.25 mole/L of NaCl (as electrolyte). Kumar *et al.* (2004) used three different types of electrodes, namely iron, aluminium, and titanium, to treat water containing 2 mg/L arsenic, III and V. The results of their study showed that the most efficient electrode to remove arsenic was iron followed by aluminium and titanium, where, 99% of arsenic was removed using iron electrodes at a current density

of 1.53 mA/cm<sup>2</sup>, for a pH range 6-8. The simultaneous removal of two heavy metals, zinc and copper, from water using the EC method was investigated by Nouri *et al.* (2010). In this study, a batch flow EC reactor with four aluminium electrodes was applied to treat 5 mg/L of zinc and 5 mg/L of copper at different operating parameters such as the applied potential. The results obtained from this study showed that EC was able to remove 90.2% zinc and 97.7% of copper after 15 min at an initial pH of 7, and cell voltage of 30V. Ouaissa *et al.* (2013) used an EC unit provided with aluminium electrodes to investigate the ability of the EC method to remediate chromium Cr (VI) from synthetic water samples. The authors indicated that 97% of initial Cr (VI) concentration of 100 mg/L was achieved at a current density of 4.03 mA/cm<sup>2</sup>, with an initial pH of 3 to 6.

#### **III. Removal of organic matter**

Since the 1970s, EC technology has been practised in the treatment of organic-rich waters and wastewaters (Deng and Englehardt, 2007), such as the wastewater from coke plants, tanneries, and slaughterhouses, landfill leachate, seawater, and drinking water (Chiang *et al.*, 1995; Tsai *et al.*, 1997; Rao *et al.*, 2006; Labanowski *et al.*, 2010; Hakizimana *et al.*, 2016). For instance, Chiang *et al.* (1995) used EC technology to treat wastewater from a coke plant containing 2143 mg/L of COD (chemical oxygen demand). The obtained results showed that the COD decreased from 2143 to 226 mg/L (89.5% removal efficiency) after 120 min of electrolysing using an EC cell supplied with a lead dioxide coated titanium anode. Remediation of the organic content of tannery wastewater collected from the effluent of a treatment plant for 85 tanneries in India using a batch EC cell. The electrolysing process used three different pairs of electrodes, titanium/platinum(Ti/Pt), titanium/lead dioxide(Ti/PbO<sub>2</sub>), and titanium/manganese dioxideTi/MnO<sub>2</sub>. The obtained results showed that the order of removal efficiency was Ti/Pt > Ti/PbO<sub>2</sub> > Ti/MnO<sub>2</sub>, where the COD concentration decreased from 515 to 189 g/m<sup>3</sup> (which the highest removal efficiency) after 240 min of electrolysing using Ti/Pt electrodes.

Tchamango *et al.* (2010) used a batch EC reactor, provided with aluminium electrodes, to treat synthetic samples of dairy wastewater. The outcomes of this study showed that 61% of the initial COD was removed after 30 min of electrolysing at a current density of 4.3 mA/cm<sup>2</sup>, and initial pH of 6.88 to 7. Aswathy *et al.* (2016) conducted an investigation about the removability of organic matter from synthetic bilge water using the EC method. Synthetic bilge water samples with initial COD of 2.1120 g/L was treated using an EC cell supplied with Al electrodes and magnetic stirrer at different initial pHs (3 to 10), voltages (5 to 10 V), and gaps between electrodes (1 to 5 cm). The obtained results indicated

that the 85% of the COD was removed after 120 min of electrolysing at initial pH of 7, with a mixing rate of 300 rpm, an applied voltage of 10V, and a gap between electrodes of 1 cm.

## IV. Removal of organisms and pathogens

Beside the acknowledged efficiency of the EC technology to coagulate and remove colloids from water and wastewater, it has been found that this technology can eliminate several types of biological pollutants such as bacteria and algae (Chen, 2004; Ricordel et al., 2010; Kuokkanen, 2016). Several trials have been made over the last four or five decades to use the EC method as a disinfection tool for water and wastewater. For instance, Matsunaga et al. (1994) developed a new EC reactor using activated carbon fibre as electrodes to remediate Escherichia coli (E.coli) bacteria from drinking water. Water samples containing 22 cells of *E.coli/L* were treated using this type of electrodes at different voltages and durations, the obtained results showed that the *E.coli* was completely removed from drinking water after 8 hrs of treatment at voltage of 0.8V. The ability of the EC technology to removal algae from water was explored by Alfafara et al. (2002), the researchers used an EC reactor supplied with an aluminium anode and a titanium cathode to treat lake water containing Chorophyll. The experimental work was conducted under different flow patterns (batch and continuous flow), taking into account the effect of several key parameters such as algae loading rate (from 2.4 to 22.9 mg/dm<sup>3</sup>. H), applied power (60 to 155 W/dm<sup>3</sup>). The results showed that, under flow pattern conditions, algae removal efficiency was about 50%. Sarkka et al. (2008) applied the EC technology to de-activate three types of paper mill bacteria (Deinococcus geothermalis, Pseudoxanthomonas taiwanensis and Meiothermus silvanus). After 3 min of electrolysing, using an EC unit with stainless steel and mixed metal oxides electrodes, at current density of 50 mA/cm<sup>2</sup>, these three types of bacteria were efficiently de-activated (> 2 log). Another study about the removal of E.coli from water by the EC method was conducted by Castro-Rios et al. (2014). In this study, a 500 mL batch EC cell having aluminium electrodes was used to treat synthetic water samples containing 10<sup>5</sup> to 10<sup>6</sup> cfu/mL of *E.coli*. The outcomes of this investigation confirmed that a current density of 2.27 mA/cm<sup>2</sup> at initial pH of 4 and 2.5 mg/L of Na<sub>2</sub>SO<sub>4</sub> was enough to reduce the *E.coli* number by 1-log and 1.9-log after 40 min and 90 min respectively.

#### V. Removal of other pollutants

Beside the mentioned applications of EC, the literature survey showed that this technology has been applied to remove a vast number of other pollutants. For instance, Un *et al.* (2013) using a batch EC reactor with aluminium electrodes for defluoridation of 5 mg/L fluoride-containing water. To obtain the

best removal efficiency, the current density was changed between 0.5 to 2 mA/cm<sup>2</sup>, the initial pH range between 4 to 8, and electrolyte concentration between 0.01 to 0.03 moles of  $Na_2SO_2$ . The highest removal efficiency of 97.6% was achieved after 30 min of electrolysing at current density, initial pH, and electrolyte concentration of 2 mA/cm<sup>2</sup>, 4, and 0.01 mole respectively.

Additionally, the EC method was efficiently used to remove fluoride and nitrate from water and wastewater. Hashim *et al.* (2017a) studied fluoride removal from synthetic drinking water samples using EC technology. In this study, fluoride removal was performed using perforated aluminium electrodes taking into accounts the effects of the initial pH (4 to 8), current density(1 to 3 mA/cm<sup>2</sup>), inter-electrode distance (5 to 11 mm), treatment time (0 to 30 min), and the initial concentration of fluoride (10 to 20 mg/L). The results indicated that 98% of fluoride could be removed within 25 min of electrolysing at a current density of 2 mA/cm<sup>2</sup>, and inter-electrode distance of 5 mm.

Hashim *et al.* (2017b) used aluminium-based EC reactor to denitrify synthetic drinking water samples, under batch flow conditions, containing 100 mg/l of nitrate. The denitrification process was optimised for different current densities (1 to  $3 \text{ mA/cm}^2$ ), the inter-electrode distance (from 3 to 10 mm), treatment time (up to 70 min), and nitrate concentration ranging from 0.05 to 0.150 g/L. The authors indicated that the aluminium-based EC reactor removed 85% of nitrate (met the World Health Organization limitations for infants) within 55 min of electrolysing at current density of 2 mA/cm<sup>2</sup>, and inter-electrode distance of 5 mm.

#### 9. Influence of liquid chemistry on the EC method.

In spite of the proven efficiency of the EC technology to remove a vast number of pollutants from aqueous media, its performance is highly influenced, negatively or positively, by the chemistry of the liquid being treated. It has been found that the presence of chloride or magnesium ions in water being treated enhances the performance of the EC units (Huang *et al.*, 2009; Heffron, 2015). Chloride inhibits the formation of the inert metallic film of the anode surface and accelerates anode dissolution rate by pitting corrosion, which enhances both generation of coagulants and current efficiency (Huang *et al.*, 2009). While, the presence of magnesium to a certain level enhances the performance of EC units by making the flocs bigger and denser, which greatly facilitates their precipitation (Heffron, 2015).

Conversely, it has been well documented that the presence of some competitive ions (such as phosphate, silica, or organic matter) and/or the initial pH of liquid being treated can significantly decrease the efficiency of the EC cells (Hu *et al.*, 2003; Heffron, 2015; You and Han, 2016). For instance, Hu *et al.* 

(2003) found, during defluoridation of water samples using aluminium-based EC reactor, that the presence of sulphate ions in water decreases the fluoride removal efficiency. The same negative impact of sulphate on the performance of the EC units was observed by Huang *et al.* (2009), where the latter noticed, during cadmium removal from water, that no reaction took place during 10 min of electrolysing due to the presence of sulphate ions in the water being treated. The authors believed that sulphate ions inhibit the corrosion of Al electrodes, which in turn decreases the removal efficiency. Silicate and phosphate play a very negative role in the electrochemical removal of other pollutants such as arsenic (Roberts *et al.*, 2004; Wan, 2010). Where, during the electrolysing of 0.5 mg/L arsenic containing water reduces arsenic removal by 90%. Moreover, the authors discovered that 3 mg/L of phosphate cause the same influence as 30 mg/L of silicate. Similar findings were obtained by Wan (2010), the latter demonstrated that 0.04 g/L of phosphate, during the electrolysing of 0.1 mg/L of arsenic containing water, could inhibit arsenic removal. The authors explained this decrease in removal efficiency by competition between arsenic ions and phosphate and silicate ions to occupy the active sites on the surface of freshly generated coagulants.

## 10. Environmental advantages of the electrocoagulation method

The electrocoagulation technology could be categorised as an eco-friendly water and wastewater treatment method as it bears several environmentally attractive merits. For example:

1. The EC method does not produce secondary pollutants as it does not required chemical additives, which makes it a green technology (Chaturvedi and Dave, 2012; García-García *et al.*, 2015).

2. In comparison with the traditional coagulation process, the flocs formed by the EC method have very low water content that significantly reduces the volume of the sludge produced (Olmez, 2009; Zodi *et al.*, 2009). It has been found that the EC units produce about 50% less sludge than traditional coagulation processes (Barrera-Diaz *et al.*, 2011). This significant reduction in sludge volume greatly reduces the cost of sludge dewatering and handling, which in turn reduces the operating cost of the EC units (Olmez, 2009).

**3.** Leaching of hazardous pollutants from waste landfills is a serious environmental problem (Abdulredha *et al.*, 2017; Abdulredha *et al.*, 2018). However, the heavy metals leaching from the EC sludge, according to the Toxicity Characteristic Leaching Procedure (TCLP), was within the permissible limits of the Environmental Protection Agency (EPA) (Addy *et al.*, 2011). These facts makes the EC sludge one of the favourable additives for construction materials, for

instance EC sludge was successfully used in the production of concrete (Barrera-Diaz *et al.*, 2011), and cement mortar (Banerjee and Chakraborty, 2005).

**4.** In comparison with traditional chemical and biological treatment methods, the EC units are able to remove very small particles, as the fine charged particles are more easily attracted to the electric field (Mollah *et al.*, 2004).

**5.** Due to the low electricity consumption, the required energy to perform the EC method can be driven from clean-energy resources such as the windmills or an attached solar panel (Chaturvedi and Dave, 2012; Deokate, 2015; Kuokkanen, 2016). Consequently, the EC method could reduce the carbon emission.

6. One of the main by-products of the EC units is hydrogen gas (Nasution *et al.*, 2011; Lakshmi *et al.*, 2013), which is categorised as an eco-friendly fuel (122 kJ/g) (Eker and Kargi, 2010). Therefore, recovery of hydrogen gas is considered to be one of the most important benefits of the EC treatment method, where Phalakornkule *et al.* (2010) reported that 13% of the required power to operate the EC unit can be produced from the generated hydrogen gas.

# 11. Disadvantages of the electrocoagulation method

EC technology, like any other treatment technology, has some drawbacks that could influence its performance. For instance:

**1.** The electrodes should be periodically replaced as they dissolve into the solution due to the oxidation process (Emamjomeh, 2006; Tamne *et al.*, 2015).

2. The formation of an oxide film on the surface of the anode during the electrolysing process reduces the anode dissolution, which in turn reduces the pollutants removal efficiency (Mansour *et al.*, 2013; Hashim *et al.*, 2016). Moreover, this film maximises energy consumption (Heffron, 2015), and limits hydrogen recovery (Yang *et al.*, 2015). However, the negative influence of this film could be reduced by different techniques such as the addition of antipassivation agents (Mechelhoff *et al.*, 2013), aeration or increasing the turbulence (Mohora *et al.*, 2014), and periodically cleaning electrodes (Kuokkanen, 2016).

**3.** The EC technology still has a deficiency in the variety of reactor design (Un *et al.*, 2013); where most of the EC reactors have parallel plate monopolar or bipolar electrode configuration systems.

**4.** In addition, there is a real deficit in the modelling of EC performance, which is very important to design, optimise, and reproduce the performance of the EC units (Kuokkanen, 2016; Hashim, 2017).

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