Electrocoagulation process for removing dyes and chemical oxygen demand from wastewater: operational conditions and economic assessment – a review

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ABSTRACT

Electrocoagulation (EC) technique gained a significant attention owing to its efficiency in removing colors and contaminations in industrial effluents discharged from numerous industries as textiles, pulp and paper industry, landfill leachate, food processing industry and tannery industry, etc. These effluents are characterized by a wide range of chemical oxygen demand (COD), heavy metals, suspended solids, turbidity and color levels. Most published works focused on the optimization of the operational parameters of EC in order to enhance the simultaneous abatement of soluble and colloidal pollutants. Indeed, EC is influenced by several operation parameters such as pH, applied current density, electrodes material and configuration, conductivity of the solution, electrode gap, electrolysis time and mixing rate. The present work handles the implied mechanisms in removing such contaminants from wastewater together with discussing the major operating parameters influencing the EC performance. Special emphasis is accorded to reducing COD and colors from wastewater. Comparing economically of EC with other alternatives is also conducted aiming at providing a reference for process selection. Economic assessment gives the opportunity for further optimizing the appropriate technology of wastewater treatment through the different strategies of cost optimization.

Keywords: Electrocoagulation (EC); Chemical oxygen demand (COD); Color; Operational parameters; Economic assessment; Operating cost (OC)

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1. Introduction

Due to the rapid increases in the world population and the new patterns of water consumption, the available water quantities are becoming a crucial problem. Many industrial processes are piloted using a huge amount of water for various purposes (e.g., as a scrubbing, a coolant, a raw material, etc.), which is discharged as wastewater. Discharging indiscriminately these effluents might affect negatively canals and rivers that are being constantly polluted [1,2]. Water pollution is consequently one of the most difficult problems of the twenty-first century. Thus, wastewater needs to be treated properly to decrease or eradicate pollutants until the water can be recycled in the industrial processes to promote sustainability.

Textiles and printing industries are considered the major water pollution sources, due to huge water demands and production of effluents containing significant concentrations of surfactants, organic matters, suspended solids, and dyes [3]. Included in such dyes, azo dyes are known for their acute color, pH ranged between 6–10 [4], and high levels of chemical oxygen demand (COD), which is the indirect measure of the amount of oxygen consumed in the chemical oxidation of the biodegradable organic compounds, non-biodegradable compounds, and inorganic oxidizable compounds present in water. Treating these colored wastewaters poses a main trouble for the industry besides their devastatrice effects on the environment due to their high toxicity and long degradation time. Purifying these effluents is therefore vital ecologically and economically.

For removing color and COD from wastewater, several treatment technologies have been developed during the last decades founded either on physical, chemical, or biological phenomena. Biological treatment was efficiently employed in removing COD [5]. Nevertheless, it become nowadays insufficient mainly in reducing color. This is due to the fact that most of that dyes are resistant to biodegradation because of their design requirements to withstand both oxidizing and reducing conditions. Further, most commercial dyes are usually too complicated in their chemical structures and non-biodegradable because of their chemical nature and molecular size [6,7]. In addition, the dyes toxicity has an inhibiting effect on the bacterial development.

Many researchers have demonstrated the possibility to obtain an acceptable removal efficiency of these pollutants by operating physico-chemicals methods like adsorption, precipitation, chemical degradation, ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO) [8–10]. However, such techniques may simply transfer the pollutants to another phase rather than destroy them, therefore it imposes the problem of discharging a high concentrate effluent. In addition to that, these technologies are very time-consuming [11].

Advanced oxidation processes (AOPs) (e.g., Fenton and photo-Fenton oxidation, ozone-based oxidation, photocatalytic and ultraviolet (UV)-based oxidation treatments, etc.) have received great attention for removing organic pollutants and dyes [12,13]. Despite of that, all of these techniques present complex chemistry mechanisms that need to be carefully manipulated. They require also a restrictive conditions to occur. However, their most difficult drawback is likely about what is called "by-products" formation (such as organochlorine compounds, chlorate, and perchlorate with UV/chlorine treatment, and bromate formation during ozone oxidation [14,15].

Recently, there is an increasing demand for more eco-friendly approaches to deal with wastewater treatment in relation with energy utilization and efficient remediation as well. Much attention has been paid to technologies for treating pollutants from effluents especially those by electrochemical methods owing to their advantages such as high efficiency [16,17], operating at ambient temperature without the need of temperature control [18] and environmental compatibility. As an electrochemical technique, electrocoagulation (EC) process was developed as an attractive option for dealing with organic and inorganic, soluble and insoluble pollutants from wastewater effluents including heavy metal ions, total suspended solids (TSS), COD, biochemical oxygen demand (BOD), and dyes [19,20]. EC technique is characterized by simple equipment and a direct current source applied between metal electrodes immersed in polluted water [21]. The electrical current induces the dissolution of metal plates (usually in iron(Fe) or aluminum(Al)) into wastewater. The metal ions, at an appropriate pH, can form flocs (metallic hydroxides, $Fe(OH)_{3(s)}$ or $Al(OH)_{3(s)}$) that destabilize and aggregate the suspended particles or precipitate and adsorb dissolved contaminants [22]. EC process provides efficient color and COD removal from wastewater without excessive chemical additions and subsequent secondary pollution and decreased amount of precipitate or sludge that requires to be removed [23,24] (Table 1). Despite all these advantages, using electrical energy, as the heart of the electrochemical technology, seems to be a limit for a large-scale EC application. Based on aforementioned, adopting the suitability of EC technique for wastewater treatment becomes even more difficult before designing and implementing the treatment plant itself. The selection criteria should be based on technical, environmental and economical scales. However, since the majority of COD and dye treatment technologies likely meet the discharge standards to the receiving environment, the economic factor is predominant in decision making. In this context, comparing the operating costs (OCs) of different technologies is required to determine the most appropriate process should be applied.

The present work is designated to review the EC treatment of COD and dyes together with highlights the mechanisms of their removal. The effects of various operating parameters such as initial pH, current density (CD), electrolysis time (*t*), electrodes materials, conductivity (supporting electrolyte), electrodes arrangement and stirring velocity (SV) on the removal efficiency of pollutants from wastewater are discussed. Despite to the high amount of research works dedicated to the treatment of various effluent wastewaters by EC, very limited investigations have been considered cost effectiveness. Thus, an economic evaluation has been performed in order to compare the EC process to other technologies.

2. Principles of electrocoagulation

The EC process has successfully been used for treating water and wastewater [44,21]. Meanwhile, EC process

Table 1
EC process used for eliminating color and COD

References	Type of wastewater	Conditions	COD removal (%)	Color removal (%)
[25]	Surface wastewater	CD = 25 A/m^2; t = 50 min; pH = 7; Fe–Fe	93	–
[26]	Olive mill	CD = 10 mA/cm^2; t = 60 min; pH = 4.8; Fe–Fe	80	–
[27]	Reactive Red 120	CD = 75 A/m^2; t = 15 min; pH = 7; Fe–Ti	–	96
[23]	CI Reactive Blue 25	CD = 2.5 mA/cm^2; t = 90 min; Al–Al	–	97
[28]	Dairy industry	CD = 61.6 A/m^2; t = 21 min; pH = 5; 8 Al plates	57	
[18]	Textile industry	pH = 9; t = 120 min; CD = 10.4 mA/cm^2; 4 Mp*–Al	69. 64	72.8
[24]	Acid Black 194 (AB194)	CD = 100 A/m^2; t = 60 min; pH = 4; Al–Al		100
[29]	Unsanitary landfill leachate (LL)	CD = 194.2 A/m^2; t = 67.64 min; pH = 7.23; Fe–Al	43	
[30]	Poultry slaughterhouse	CD = 30 A/m^2; t = 40 min; pH = 4; Al–Al	86	
[31]	LL wastewater	pH = 7.83; CD = 525 A/m^2; d = 1 cm	51.75	
[32]	Methyl orange (MO)	CD = 64 A/m^2; pH = 7.25; NaCl = 1.6 g/L; d = 2 cm; Fe–Fe	–	83
[33]	Reactive Black dye	pH = 11; t = 30 min; Al–Al	–	98.23
[34]	Yellow 145 dye	pH = 3; NaCl = 1.6 g/L; t = 10 min; Al–Al		98.24
[35]	Textile wastewater	CD = 80 A/m^2; pH = 7.1; t = 10 min; Fe–Al	59	86
[36]	Reactive Red 195 dye	CD = 400 A/m^2; Natural pH; t = 10 min; Fe–Fe	74	98
[37]	Swine slaughterhouse wastewater	CD = 25 mA/cm^2; t = 100 min; pH = 2; Al–Fe	92	
		Fe–Fe	74	
		Al–Al	97	
[38]	Tannery wastewater	CD = 200 A/m^2; t = 20 min; Fe–Fe; pH = 7; Al–Al; pH = 6	81 75	98 98
[39]	Licorice processing wastewater	CD = 350 A/m^2; t = 81.8 min; NaCl = 300 mg/L; SV** = 45 rpm; Fe–Fe	89.4	90.1
[40]	Reactive Red 120 dye	CD = 134.5 A/m^2; pH = 6.8; t = 69.62 min; d = 1.77 cm; Fe–Fe (two pairs)	93.47	81.31
[41]	Table olive debittering	CD = 211 A/m^2; pH = 4.4; t = 58.9 min; Al–Al	75.3	–
[42]	Indigo dye	U = 47 V; pH = 7.5; 2 L/min; 30 Fe parallel electrodes	–	94.083
[43]	Mineral processing wastewater	CD = 192.3 A/m^2; pH = 7.1; t = 70 min; Fe–SS***	77.62	

*Mp: Monopolar configuration;
**SV: Speed velocity;
***SS: Stainless steel.

has been widely used to decolorize various types of dye containing solutions such as disperse, reactive and acidic dyes [34,45]. EC is a separation technique in which both physical and chemical mechanisms for pollutants removal are involved [46,47]. The basic principle of EC comes from "electrolysis", which means to break substances apart using electricity. Therefore, EC consists of electrodes that are arranged in pairs of two, with one serving as an anode and the other as a cathode. When the electrodes are connected to an external power source, the anode material is electrochemically and rapidly corroded due to its oxidation. These conductive metal plates are commonly known as 'sacrificial electrodes'. The electrochemical dissolution of sacrificial metal electrodes, usually Fe or Al, occurs in an electrolyte (water or salt-melting solution) [42]. The transfer of ions between two electrodes generates a coagulant *in situ* by the dissolution of metal from the anode with simultaneous formation of hydroxyl ions (OH$^-$) and hydrogen gas (H$_{2(g)}$) at the cathode. When this happens, the particulates are neutralized by formation of hydroxide complexes, which then form the basis of agglomerates. A stirrer is set to keep the liquid and slurries uniform in the reactor (Fig. 1).

Although EC seems similar to chemical coagulation, an EC process has many differences as it involves many chemical and physical phenomena that use consumable electrodes to supply coagulating ions *in situ* the wastewater. The EC process generally consists of the following mechanisms that occur serially [48,49]:
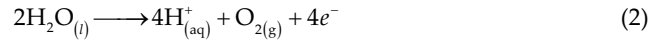
- Oxidation of the anode and reduction of the cathode resulting in metal ions (coagulating agents) associated with O$_2$ microbubbles generation (at the anode) and H$_2$ with OH$^-$ ions;
- Formation of coagulants: The metallic cations interact with OH$^-$ ions and form metal hydroxides. The floc formed through the coagulation has a good adsorption ability to bind the pollutants;
- Adsorption of pollutants on coagulants: neutralized matter is aggregated and adsorbed on metal hydroxides to form larger flocs;
- Removal of contaminants: Some of the neutralized matter flocculated is removed by sedimentation or through electroflotation (EF) by lifting them to the surface with the H$_2$ gas generated in the system.

The complexity and number of possible interactions of an EC process are highlighted in Fig. 2, which shows the main mechanisms of pollutants removal.
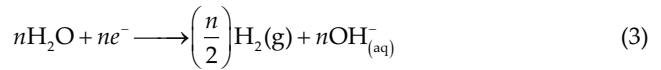
2.1. Anode and cathode reactions

During the EC technique, when an electric potential is applied from an external power source, the anode metal is dissociated to di- or tri- metallic ions and discharges proportional number of electrons (n). Water reacts with the anode and releases hydrogen ions (H^+) and oxygen gas ($O_{2(g)}$) in the
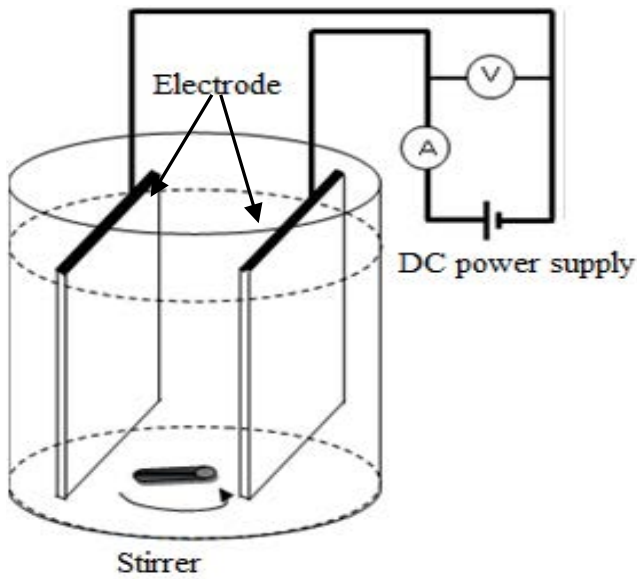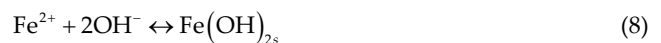

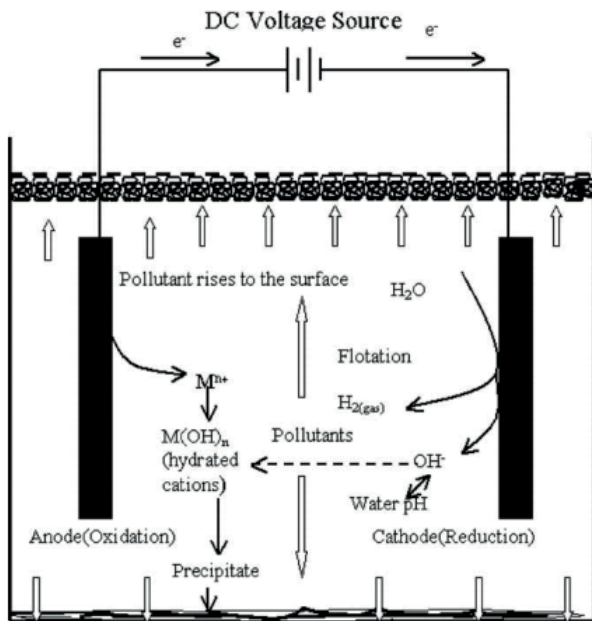Fig. 1. Schematic presentation of the EC batch reactor.

Fig. 2. Schematic diagram of a bench-scale two-electrode electrocoagulation (EC) cell [48].

system. The reactions occurring at the anode and cathode are given below as Eqs. (1)–(3):

$$M_{(s)} \longrightarrow M^{n+}_{(aq)} + ne^- \tag{1}$$

$$2H_2O_{(l)} \longrightarrow 4H^+_{(aq)} + O_{2(g)} + 4e^- \tag{2}$$

The cathode will be subjected to reduction of water, which generates $H_{2(g)}$ and OH^- ions:

$$nH_2O + ne^- \longrightarrow \left(\frac{n}{2}\right)H_2(g) + nOH^-_{(aq)} \tag{3}$$

Metal ions formed at the anode react with hydroxyl ions liberated from the cathode to produce various monomeric and polymeric species, which (at an appropriate pH) can form wide range of coagulated species and metal hydroxides $M(OH)_n$:

$$M^{n+}_{(aq)} + nOH^-_{(aq)} \to M(OH)_n \tag{4}$$

2.2. Electrodes used in EC process and their governing reactions

Iron (Fe) and aluminum (Al) are the mostly used anode materials in EC technique. Fe (or Al) is dissolved from the anode generating corresponding metal ions, which are almost immediately hydrolyzed to polymeric iron or iron hydroxide.

2.2.1. Iron(Fe) electrode

Both the common two oxidation states of iron species might be dissolved from the anode and exist in aqueous environment, namely ferrous (Fe^{2+}) and ferric (Fe^{3+}) ions [Eqs. (5) and (6)] [50]:

$$Fe \to Fe^{2+} + 2e^- \tag{5}$$

$$Fe \to Fe^{3+} + 3e^- \tag{6}$$

Significant OH^- production from the cathode causes an increase in pH during electrolysis leading to the formation of monomeric and polymeric species in solution [Eqs. (7)–(9)]. Fig. 3 presents the predominance zone diagram of Fe^{2+} and Fe^{3+}, respectively, and their hydrocomplexes depending on pH.

$Fe(OH)_2$ precipitates at pH > 5.5 and remains in equilibrium with monomeric species $Fe(OH)^+$ from pH 9.5 up to 11.4 and with $Fe(OH)_3^-$ in pH comprised between 11.8 and 14:

$$Fe^{2+} + OH^- \leftrightarrow Fe(OH)^+ \tag{7}$$

$$Fe^{2+} + 2OH^- \leftrightarrow Fe(OH)_{2s} \tag{8}$$

$$Fe^{2+} + 3OH^- \leftrightarrow Fe(OH)_3^- \tag{9}$$

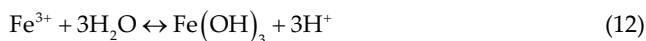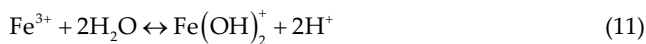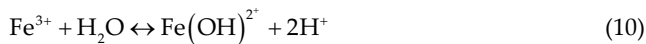Fig. 3. Predominance zone diagram for Fe^{2+} (a) and Fe^{3+} (b) chemical species in aqueous solution [51].

As depicted in Fig. 3, the monomeric species $Fe(OH)_3$ coagulates at pH > 1.0 and remains the unique species present in the solution in the range of pH between 6.2 and 9.6. $Fe(OH)_3$ is in equilibrium with Fe^{3+} up to pH 2.0, $Fe(OH)_2^+$ from 2.0 up to 3.8, $Fe(OH)_2^+$ from 3.8 up to 6.2 and $Fe(OH)_4^-$ from pH equal to 9.6. [Eqs. (10)–(13)]:

$$Fe^{3+} + H_2O \leftrightarrow Fe(OH)^{2+} + 2H^+ \tag{10}$$

$$Fe^{3+} + 2H_2O \leftrightarrow Fe(OH)_2^+ + 2H^+ \tag{11}$$

$$Fe^{3+} + 3H_2O \leftrightarrow Fe(OH)_3 + 3H^+ \tag{12}$$

$$Fe^{3+} + 4H_2O \leftrightarrow Fe(OH)_4^- + 4H^+ \tag{13}$$

Fe^{2+} can be oxidized to Fe^{3+} depending on dissolved oxygen concentration and pH value of solution. In acidic media, Fe^{2+} oxidizes slowly in contact with dissolved oxygen [Eq. (14)]. In neutral or alkaline media, Fe^{2+} is transformed into ferrous hydroxide that is oxidized by oxygen to form ferric hydroxide [Eq. (15)] [52].

$$Fe^{2+} + O_2 + 2H_2O \rightarrow Fe^{3+} + 4OH^- \tag{14}$$

$$4Fe(OH)_2 + O_2 + 2H_2O \rightarrow 4Fe(OH)_3 \tag{15}$$
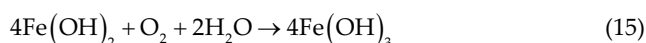
However, there are some ambiguities on the literature about the mechanism of iron dissolution from anode to produce Fe^{2+} or Fe^{3+} and their hydrolysis products.

Lakshmanan et al. [53] affirmed that, regardless of initial pH and current, 80%–95% of iron was present as Fe^{2+} confirming that this ion was the primary species formed during electrolysis. Tchamango et al. [54] suggested that Fe^{2+} is not totally transformed into Fe^{3+} since $Fe(OH)_2$ co-exists with $Fe(OH)_3$. Based on the current intensity as a function of applied voltage variation and the pertinent literature, Ghernaout et al. [46] suggested three mechanisms for acid, neutral and alkaline pH. For pH 2, Mechanism 1 explains $Fe(OH)_{2(s)}$ formation; for pH 7, Mechanism 2 concerns both the varieties $Fe(OH)_{2(s)}$ and $Fe(OH)_{3(s)}$ production; and for pH 12, Mechanism 3 is characterized by $Fe(OH)_{3(s)}$ apparition.

2.2.2. Aluminum (Al) electrode

During EC using Al electrodes, several reactions take place at the surface of Al anode. Dissolution of Al occurs in the anode and reduction of water happens to form $H_{2(g)}$ at the cathode [55].

$$Al \rightarrow Al^{3+} + 3e^- \tag{16}$$

The hydrolyzed aluminum ions can form monomeric species such as $Al(OH)_2^+$, $Al(OH)_2$, $Al(OH)_3$ that are more dominant species depending on the pH conditions [56].

At acidic condition [Eq. (17)]:

$$Al^{3+} + 3H_2O \rightarrow Al(OH)_3 + 3H^+ \tag{17}$$
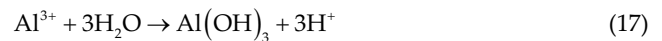
At alkaline condition [Eq. (18)]:

$$Al^{3+} + 3OH^- \rightarrow Al(OH)_3 \tag{18}$$

In neutral pH, $Al(OH)_{3(s)}$ is stable and insoluble in the water. It is the major compound responsible for a rapid adsorption of soluble organic compounds and trapping of colloids [57,58]. The significant increase of the local pH in the vicinity of the cathode due to the formation of hydroxyl ions or the consumption of hydronium ions/protons induces the corrosion of aluminum by water [59] [Eq. (19)]:

$$2Al + 6H_2O + 2OH^- \rightarrow 2Al(OH)_4^- + 3H_2 \tag{19}$$
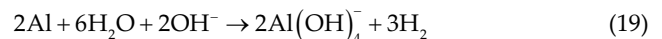
$Al(OH)_4^-$ can participate to remove some pollutants from wastewater by reacting with cations, thus neutralizing their charge and reducing their solubility, or it can be transformed into $Al(OH)_3$ [56]:

$$Al(OH)_4^- \rightarrow Al(OH)_3 + 3OH^- \tag{20}$$
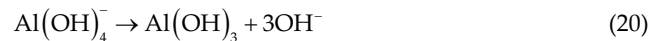
The species availability of Al^{3+} at various pH is shown in Fig. 4.

3. Mechanisms and kinetics of dye and COD removal

The sacrificial metal anodes are used to continuously produce polymeric hydroxides in the solution. Several interaction mechanisms are possible between dyes or contaminant molecules present in wastewater stream and hydrolysis products being generated by the electrode erosion. Two major interaction mechanisms have been considered: precipitation and adsorption for dye or pollutant (L) [61,47].
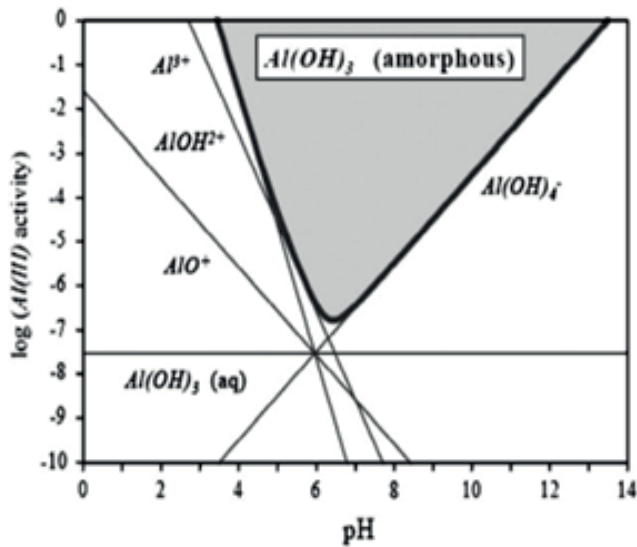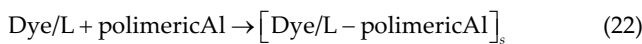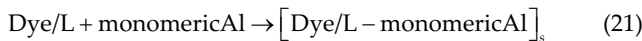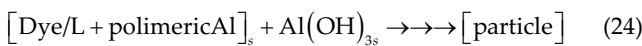
Fig. 4. Diagram of solubility of Al^{3+} species as a function of pH [60].

Fig. 5. Mechanisms of pollutant removal [51].

Precipitation:

$$Dye/L + monomericAl \rightarrow \left[Dye/L - monomericAl\right]_s \quad (21)$$

$$Dye/L + polimericAl \rightarrow \left[Dye/L - polimericAl\right]_s \quad (22)$$

Adsorption:

$$Dye/L + Al(OH)_{3s} \rightarrow \left[particle\right] \quad (23)$$

$$\left[Dye/L + polimericAl\right]_s + Al(OH)_{3s} \rightarrow \rightarrow \rightarrow \left[particle\right] \quad (24)$$
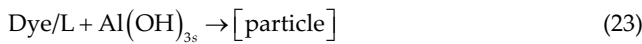
The same mechanism is also valid for iron.

Fig. 5 gives the most important mechanisms of pollutant removal by EC.

Many researchers have studied the kinetic of pollutant adsorption during EC process. Various adsorption isotherms models were used in the literature such as Langmuir [62], Freundlich [63], Dubinin and Radushkevich [64], Redlich and Peterson [65] and Sips [66].

Langmuir isotherm:

$$Q_{eq} = \frac{Q_0 b C_e}{1 + b C_e} \quad (25)$$

where: Q_{eq} is the amount of the dye adsorbed at the equilibrium time, Q_0 is the maximum amount of the dye molecule per unit weight of the coagulant, C_e (mg/g) is the concentration of the dye or pollutant remaining in solution at equilibrium, and b is an equilibrium constant (dm³/mg).

Freundlich isotherm:

$$Q_{eq} = K_F C_e^{1/n} \quad (26)$$

where: K_F and n are the Freundlich constants related to the adsorption capacity and intensity of the sorbent, respectively.

Redlich–Peterson isotherm:

$$Q_{eq} = \frac{Q_0 C_e}{1 + K_R C_e^{\beta}} \quad (27)$$

where: K_R and β are the Redlich–Peterson parameters.

Dubinin–Radushkevich isotherm:

$$Q_{eq} = Q_m e^{-\beta \varepsilon^2} \quad (28)$$

where: Q_m is the theoretical saturation capacity (mol/g), β is a constant related to the mean free energy of adsorption per mole of the adsorbate (mol²/J²), and ε is the Polanyi potential given by Eq. (29):

$$\varepsilon = RT \ln\left(\frac{P_s}{P}\right) \quad (29)$$

where: P_s (atm) and P (atm) are the saturation vapor pressure and the equilibrium pressure of adsorbate molecules at the temperature T, respectively. Also, R (8.314 J/mol K) is the universal gas constant and T (K) is the absolute temperature.

Sips isotherm:

$$Q_{eq} = \frac{Q_0 \left(k_s C_e\right)^{ms}}{1 + \left(k_s C_e\right)^{ms}} \quad (30)$$

where: k_s is the Sips isotherm model constant and m_s is the Sips isotherm model exponent.

Table 2 presents results obtained on kinetic adsorption modeling of dye and COD in literature using EC.

Moreover, some researchers used the zeta potential as a tool to more understand the mechanism of pollutant removal. In fact, Zaroual et al. [71] measured the zeta potential of a basic textile effluent treated by electrocoagulation with two iron electrodes. The value of zeta potential was about –30 mV of the formed flocs and remains stable at

600 mV and pH = 10.6. The obtained value was compared with the zeta potential of Fe(II) and Fe(III) hydroxide resulting from iron ions synthetic solutions. The results showed that the zeta potential of $Fe(OH)_2$ is −37 mV which was comparable with the one of electrocoagulation tests. Then, the formed flocs are Fe(II) hydroxide. The stability of zeta potential exhibits that there isn't any chemical interaction between the pollutants and iron hydroxide. The removal could be explain by (i) the pollutants are possibly enmeshed in the main pores of iron hydroxide, and or (ii) hydrogen bonding and the van der Waals interactions between flocs and pollutants. The last mechanism was also confirmed by Golder et al. [72]. These authors treated two sulfonated azo dye Trypan blue (TB) and Orange G (OG) by electrocoagulation using Al and Fe electrodes. The results showed that the removal efficiency in case of OG is lower compared with that in TB for both Fe and Al electrodes since TB dye contains more sulfonate groups compared with OG which facilitate TB removal by electrostatic attraction. The authors suggested the following equations of dye removal:

$$M(OH)_n \rightarrow M(OH)_{n-1}^+ (aq) + OH_{(aq)}^- \tag{31}$$

$$Dye - SO_3Na \rightarrow Dye - SO_3^- (aq) + Na_{(aq)}^+ \tag{32}$$

$$M(OH)_{n-1}^+ (aq) + Dye - SO_3^- (aq) \rightarrow M - (OH)_{n-1} O_3 S - Dye(s) \tag{33}$$
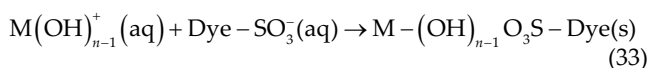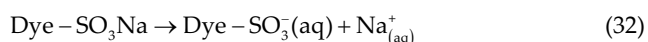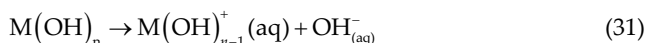
Besides, Casillas et al. [73] studied the mechanism of COD removal by EC using Fe electrode plates of some selected organic compounds. They were observed that: (i) the final COD can be increased, especially with acid compounds that react with Fe ion to form soluble products and remain in the solution; (ii) some compounds (like glucose, lactose, isopropyl alcohol, phenol, and sucrose) were not removed and remain in the solution; (iii) the COD was partially removed in the case of organic salts and sodium oxalate. They suggested that Fe^{2+} and Fe^{3+} are more acidic than Na^+, hence OH^- stay with $Fe(OH)_2$ and/ or $Fe(OH)_3$ to form insoluble iron hydroxides. Some other compounds form soluble and/or insoluble compounds with Fe ion that reduce the removal efficiency of COD. For the fourth case where the COD was highly removed, mainly the portion of COD include into suspended solids, fecal coliforms, turbidity, fats oil and grease, etc.

4. Advantages and drawbacks of EC

4.1. Advantages

- No additional chemical required, and so minimum chance for secondary pollution caused by chemical substances.
- EC needs simple equipment, easily operable and easy controllable since the EC reactor is controlled electrically.
- EC process destabilizes and removes even the smallest colloidal particles, and gives clear, colorless and odorless treated water.
- Sludge formed by EC is minimal compared to capital cost (CC) and tends to be readily settable.

- Gas bubbles produced during electrolysis can enhance the pollutant removal by floating them on the top of the solution.

4.2. Drawbacks of EC

- The sacrificial anode needs a regular replacement, since it dissolves into the solution.
- Required, in some cases, a salt addition (as a supporting electrolyte) to enhance the solution conductivity.
- The use of electric energy can be impediment for process scale up especially in some areas where electricity is not abundant.
- Cathode passivation that occurs by the formation of an impermeable oxide film on the surface, may lead to the loss of EC performance.

Nevertheless, related to electric energy consumption, some alternatives may be conveniently used in areas where electricity is not available, like the solar energy that is free to use and has abundant in availability [74,75].

Concerning the cathode passivation, many researchers studied the possibility to prevent the formation of oxide film on cathode electrode surface. Mao et al. [76] investigated the effect of the use of alternating pulse current (APC) rather than DC as a power type supply for EC cell on electrode passivation. The morphology of the electrode surface after APC and DC was characterized by scanning electron microscopy (SEM). The results show that electrode demonstrates a regular microstructure and less disordered pores when the APC was used. However, when the DC was used, many aggregates of microcrystals show that aluminium oxide may be formed. Eyvaz et al. [77] used the same technique to prevent the electrode passivation during the treatment of winery wastewater. They observed that by using the DC system that the COD, turbidity and color removal efficiencies increase until a certain CD and then decrease due to the cathode passivation. However, removal efficiencies increase in APC system after an optimum operation time that belongs to DC system, and APC provide 40% more COD removal than DC. Pi et al. [78] demonstrated the effectiveness of electrolysis with periodic reversal (PREC) of the electrodes to inhibit electrode passivation. Indeed, the results show that the PREC has a smaller rate of resistance increase (2.48×10^{-4} cm^{-2}/min) compared with conventional EC without PREC (7.72×10^{-4} cm^{-2}/min).

5. Reactor design

The reactor design is considered as the most important feature of EC process, since it affects the hydrodynamic of reactor, including fluid flow regime, mixing, and flotation or settling effectiveness. Thus reactor design has a great impact on the efficiency and removal rates. In addition, the design phase should consider the operating mode of EC reactor, as batch or continuous mode. Batch EC reactor is characterized by dynamic behavior where the pollutant and coagulant concentration, and pH level vary over time. Whereas, in continuous mode, the EC reactor is stable in performance. The conventional EC reactor is consisted on

Table 2
Adsorption isotherm of pollutants and dye removal in electrocoagulation (EC) unit

References	Wastewater (COD/dye)	Isotherm of adsorption			
[67]	Pharmaceutical wastewater; COD/turbidity removal; Al–Al; pH: 4–10; CD = 20–80 mA/cm^2; t = 10–30 min	Freundlich K_F = 0.0059; n = 0.577; R^2 = 0.972 Dubinin–Radushkevich Q_m = 306.12 mg/L; β = 0.013 kJ/mol; E = 0.0062 kJ/mol; R^2 = 0.901			
[68]	4 Fe electrodes; CD = 35.7 A/m^2, pH: 6.5	Langmuir		Freundlich	
	Malachite green	Q_{max} = 39.76 mg/g; b = 0.086 L/mg; R^2 = 0.97		K_F = 6.16; $1/n$ = 0.45; R^2 = 0.97	
	Dye Remazol yellow	Q_{max} = 11.69 mg/g; b = 0.015 L/mg; R^2 = 0.9926		K_F = 0.55; $1/n$ = 0.45; R^2 = 0.983	
[69]	14 parallel Al and Fe electrodes; pH: 7.7; 8 A	Langmuir		Freundlich	
		Al	Fe	Al	Fe
	Setazol Black TNN	Q_{max} = 555.5 mg/g; b = 0.146 L/mg; R^2 = 0.983	Q_{max} = 303.30 mg/g; b = 0.429 L/mg; R^2 = 0.942	K_F = 126.47; $1/n$ = 0.35; R^2 = 0.901	K_F = 80; $1/n$ = 0.532; R^2 = 0.844
[22]	Al–Al electrodes; pH: 7; t = 10 min; 1 A	Langmuir		Freundlich	
	Non-anionic dye	Q_{max} = 5.66 mg/g; b = 23.79 × 10^{-3} L/mg; R^2 = 0.989		K_F = 57.18 × 10^{-2}; $1/n$ = 0.405; R^2 = 0.986	
	Cationic dye	Q_{max} = 4.68 mg/g; b = 6.29 × 10^{-3} L/mg; R^2 = 0.990		K_F = 13.44 × 10^{-2}; $1/n$ = 0.599; R^2 = 0.978	
	Anionic dye	Q_{max} = 2.9 × 10^{-3} mg/g; b = 3.7 L/mg; R^2 = 0.980		K_F = 5.35 × 10^2; $1/n$ = 0.6871; R^2 = 0.981	
[70]	Basic dye rhodamine B; 2 pairs of steel electrodes; CD = 71.5 A/m^2; pH: 7.0	Freundlich K_F = 42.47; $1/n$ = 0.35; R^2 = 0.961 Langmuir Q_{max} = 136.36 mg/g; b = 0.2 L/mg; R^2 = 0.974 Redlich–Peterson Q_0 = 143.51 L/mg; K_R = 0.96; β = 0.243; R^2 = 0.973 Dubinin–Radushkevich Q_m = 306.12 mg/L; β = 0.013 kJ/mol E = 0.0062 kJ/mol; R^2 = 0.978 Sips Q_{max} = 139.95 mg/g; K_R = 0.266 L/mg; m_s = 2.07; R^2 = 0.982			

rectangular shape with a submerged rectangular electrode. However, other cell patterns have been used for EC reactor.

Generally, the batch process was used for the operating parameters optimization, which serves as guidelines to operate the continuous mode. Ardhan et al. [79] optimized the removal efficiency of Reactive blue 21 from wastewater by EC reactor operating in batch mode. The optimized parameters obtained were used to operate a cylindrical tube EC reactor under continuous mode. The electrodes were assembled in a concentric configuration. A hollow iron tube with a diameter of 3.2 cm was used as the cathode, and a hollow iron tube with a diameter of 5.1 cm was used as the anode. The results showed that the color and COD removals from both reactors were found to be within 10%.

In the aim to improve fluid intermixing, some investigators have studied the effectiveness of cylindrical type. El-Ashtoukhy et al. [80] studied the treatment of paper mill effluents by EC cell using a cylindrical agitated vessel equipped with four rectangular baffles, fixed to the container. A cylindrical lead sheet used as anode, while a concentric cylindrical stainless steel sheet screen as a cathode. The COD removal efficiency was about 97% and the percentage of decolorization and the energy consumption were ranged between 53% to 100% and 4 to 29 kWh/m^3, respectively. Ortega et al. [81] studied the removal of Indigo dye from aqueous media by EC reactor on batch and flow mode with different cell geometry. The batch mode was performed in a glass reactor with carbon steel rectangular electrode. However, the flow reactor consisted of a tubular reactor with a volume of 2.450 L. A helical electrode with cross section was used as anode with a central rod used as cathode measuring 1.07 m. Both anode and cathode are made of carbon steel. They found that the removal of Indigo was about 93% by using a continuous tubular reactor against 89% on conventional EC cell. López et al. [82] investigated the use of a new cartridge type EC reactor with a three dimensional anode electrode made of steel wool with a cylindrical form. The reactor has a volume 650 mL,

outer diameter of 2.5 and 25 cm of height. The cathode is an aluminum rod placed at the axis of the cartridge. The new configuration ensures a high removal efficiency of a textile dye by 99% associated with an energy consumption of 0.68 kW/h m³ in continuous mode of operation (flow rate: 13.3 L/h), which is very similar to those obtained in conventional batch reactor with iron electrode.

In order to improve solid/liquid separation and to reduce energy consumption by mechanical mixing, some researchers studied the combined of EC reactor with air diffuser. Indeed, Ammar et al. [83], investigated the use of a novel split-plate airlift/EC batch reactor for the treatment of petroleum refinery wastewater. The reactor has a cylindrical geometry and composed by two rectangular split-plates with two aluminum electrodes inserted between the two split-plates of airlift. The experimental results showed that the COD percentage removal could achieve up to 93.1% and the energy consumption was 1 kWh/g COD removed at a current density of 11.3 mA/cm² within 30 min of treatment. Elhafdi et al. [84] treated a mixture of real textile dye by operating a pilot external loop airlift EC reactor with maximum capacity of 150 L. A combination of aluminum and iron electrodes are placed in the riser section of air lift/EC reactor. The electrodes have a rectangular shape with surface area of 1,050 cm². The results showed a good removal of color and COD reached 80% and a lower specific energy consumption (50 kWh/kg dye) at electrolysis time of 20 min and pH: 6.9.

Moreover, in order to increase the surface specific electrode area, the design of fixed bed EC reactor has been used. Rodrigues et al. [85] investigated the use of a vertical fixed bed EC rector to remove color. The reactor was composed by a homogenization region, composed of polyethylene particles and fixed bad region filled with spherical metal particles (aluminum or iron) with a diameter of 2.4 mm. The reactor operated on batch and continuous mode. The batch mode revealed that the best color removal efficiency by 95% was achieved with Fe anode particulate under a pH 3 and current of 6 A associated with specific energy consumption about 33.7 Wh/g. Therefore, these conditions were used for the continuous process (24.3 L/h). The removal efficiency was 98% of color with specific energy consumption about 36.5 Wh/g. The authors concluded that the fixed bed EC reactor has achieved approximately the same removal efficiency compared to other studies, which they used a flat anode, but in a shorter treatment time, only 0.2 min.

6. Parameters influencing the EC process

The effectiveness of the EC process depends on many operational parameters such as conductivity of water/wastewater and pH of the solution, electrolysis time (t), current density (CD). In addition, other characteristics might be influencing the efficiency of EC such as type of metal electrodes, arrangement of electrodes, and distance between the electrodes (d). The effect of these parameters is reported during the purification of different industrial effluents, particularly in wastewater containing high amount of COD such as textiles, olive mill wastewater, petroleum refinery effluent, and dairy wastewaters, etc.

6.1. Effect of initial pH

The initial pH is one of the most important parameters that control the EC process performance [86,87]. The pH has a significant influence on the Al and Fe species distribution and therefore determines the type of hydroxide of metal cations in the EC system and also determines the interaction between the formed coagulants and the molecules of dye and pollutant in the solution (Table 3). Consequently, it influences the mechanism of the removal of contaminants from the aqueous system.

Using Fe electrode, the formation of iron hydroxyl complexes taking place after oxidation at the anode was strongly dependent on pH. From Table 3, the higher value of removal efficiency was observed specially at neutral value of pH [43,89]. The formation of monomeric species of $Fe(OH)_3$ at this pH was the responsible for the removal of the major part of the impurities in wastewater. For highly alkaline solutions, especially for pH more than 11, the quantity of soluble hydroxides (such as $Fe(OH)_4^-$) increases. $Fe(OH)_4^-$ ions are not able to destabilize the pollutant species, and thus their presence directly affects the removal of COD. Nunez et al. [35] reported that the COD removal efficiency is close to zero and 20% of dye removal at initial pH about 12. It was observed also that at acidic pH, the removal efficiency of dye and COD also decreased since OH^- ions generated at the cathode were neutralized by H^+ ions, limiting the formation of iron hydroxide [91]. Irki et al. [32] reported that the effectiveness of the treatment and the decolorization rates are decreased below 72% when the initial pH range from 9 to 12.

Also, from Table 3, when the sacrificial electrode was Al, the good removal of COD and dye was observed generally in weakly acidic medium (pH: 4–6) and weakly alkaline medium (pH = 6–8). Tak et al. [86] studied the effect of initial pH on the removal of COD from livestock wastewater using four Al electrodes. They observed that the best COD removal can be obtained at the range of 4–8 of the initial pH. Kobya et al. [90] reported that the highest removal efficiencies have been obtained with Al in acidic medium with pH < 6, while Fe was more efficient in neutral and alkaline medium especially between 6 < pH < 9 [90]. Ozyonar et al. [96] treated a domestic wastewater by EC. The results showed that the maximum removal of COD was found to be 72% by Al electrode at initial pH about 7.8.

Hernández et al. [94] analyzed the Al species associated with the aqueous pH (Fig. 6). At a pH below 3.5, the Al ion is the predominant specie. However, at a pH 4–9.5, the predominant Al chemical species is $Al(OH)_{3(s)}$, which has a large surface area as coagulant for a rapid adsorption of soluble organic compounds and dye. At pH greater than 10, $Al(OH)_4^-$ fraction increases and it becomes the dominant species. Such anion is unable to form flocs, and hence, there is no reduction in COD and color.

It should be noted that the pH value, where the removal efficiency was on maximum, does not always correspond to a good choice especially when the economic reason was considered [35]. Color removal seemed not very influenced by pH variation as COD removal. Shah et al. [34] treated a Yellow 145 dye by EC process using Al and Fe as sacrificial electrodes [34]. They showed that the optimal initial pH

Table 3
Influence of pH value on color and COD removal (SS: Stainless steel)

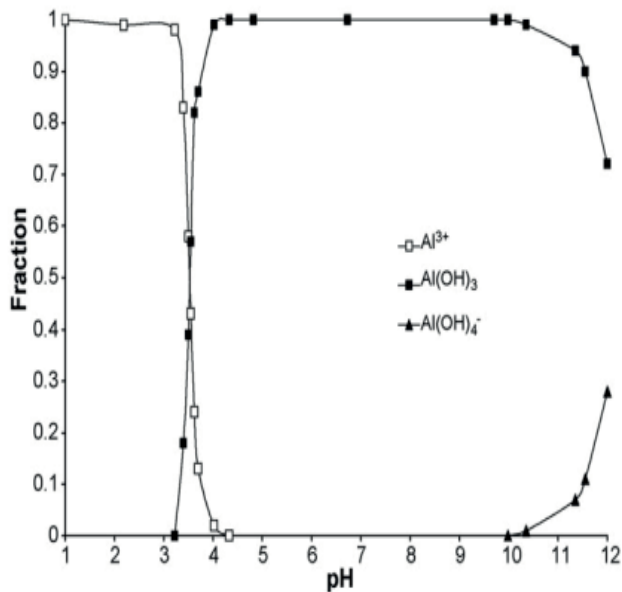| References	Wastewater specification and electrode used	Optimum initial pH	COD removal (%)	Color removal (%)
[88]	Basic dye solutions Anode–cathode: Fe-Steel	Between 5.5 and 8.5	–	–
	CD = 60 A/m²; t = 5 min C.I. Basic Red 46 (BR46)		99	98
	CD = 80 A/m²; t = 5 min C.I. Basic Blue 3 (BB3)		75	85
[89]	10 Fe electrodes Textile wastewater CD = 85–95 A/m²; t = 30 min	Neutral pH	52–70	85–90
[78]	Pair Al electrodes MO synthetic waste CD = 1,850 A/m²; t = 14 min	Optimum pH range of 6–8. Removal decreases at initial pH extremes of 3 and 10	–	82
[86]	Four Al electrodes Livestock wastewater CD = 300 A/m²; t = 30 min	pH 4 for the maximum removals. pH = 8 for economic reason	93	95.2
[32]	Pair of Fe electrodes MO synthetic wastewater CD = 64 A/m²; t = 12 min	Initial pH between 5 and 8, optimum value 7.25	–	83
[35]	Textile industry Fe–Al; CD = 80 A/m²; t = 10 min	pH = 7.1	59	86
[90]	Textile wastewater Four Fe electrodes CD = 150 A/m², t = 10 min	Acidic medium with: pH < 6 highest removal efficiencies obtained with Al	65	–
	Four Al electrodes CD = 80–100 A/m²; t = 10 min	Neutral and alkaline medium, especially between 6 < pH < 9 for Fe	77	–
[91]	C.I. Disperse Yellow 3 CD = 83 A/m²; t = 45 min		–	98.96
	Pair Al electrodes	pH: 3–10		
	Pair Fe electrodes	pH: 6.5	–	96.28
[92]	Egg processing effluent Pair Al electrodes CD = 20 mA/cm²; t = 30 min	pH = 6	89	–
[93]	Direct Black 22 Acid Red 97 Pair Al electrodes CD = 50 A/m²; t = 5 min	pH = 5–8	68.33 (Al)	~90
	Pair Fe electrodes CD = 50 A/m²; t = 5 min	pH = 8	58.33 (Fe)	–
[34]	Yellow 145 dye		–	98.24
	Pair Al electrodes 15 V; t = 10 min	pH = 3	–	–
	Pair Fe electrodes 15 V; t = 30 min	pH = 5	–	99.21
[43]	Mineral processing wastewater Fe–SS electrodes; t = 70 min; CD = 19.23 mA/cm²	pH = 7.1	82.8	–
[94]	Industrial wastewater 3 A; t = 60 min		46–50	–
	Al anode	Maximum removal observed at pH 6–8	–	–
	Fe anode	Maximum removal observed at pH = 8	53	–
[87]	Petroleum refinery wastewater Al–SS; CD = 12 mA/cm²; t = 60 min	pH = 7	96.8	–
[95]	Distillery spent wash Pair of Fe electrodes 3 A; t = 5 h	pH = 7	66.7	–

Fig. 6. Aluminium species distribution as a function of pH [94].

was found to be 5 and 3 for Fe and Al electrodes, respectively. Some investigators claim that the Fe electrode as a sacrificial anode generates a yellow color and increases turbidity of water.

Nevertheless, aluminium is generally more toxic than iron to human body and environment [97]. Therefore, the choice of adequate electrode for treatment was related to many factors, mainly those linked to the environmental requirements and economic consideration.

Moreover, several studies deals with pH buffering effect during EC, meaning that at an initial pH < 7, the pH will rise; however, at pH > 8.5, the pH will decrease [90,98]. For instance, Chen et al. [98] noted a large increase in final pH when the initial pH ranged between 3 and 7, but insignificant increase when the initial pH was greater than 8. The pH evolution during the EC process can be attributed to the balance between the production of hydroxyl ions in the cathode and its consumption by the ultimate transformation of soluble aluminium compounds into aluminium hydroxides. Two other mechanisms have been reported in the literature [99]: (i) when iron electrode was used, it may have an ion exchange between hydroxide ions in iron hydroxide and chloride or sulfate ions present in solution, (ii) stripping of carbonic acid as CO_2 by hydrogen bubbles. Nevertheless, the pH change during application can be a problematic especially when the pH value moved from the optimum.

6.2. Current density

EC process is linked to the electric current intensity and applied potential. It can be driven under the galvanostatic or potentiostatic mode. For the galvanostatic mode, the current flow is controlled; while for the potentiostatic mode, the potential is accurately controlled. Current density (CD) is defined as current applied per unit surface area of the electrode. In EC process, the reaction rate may be controlled by

the applied CD. Thus, it determines the production rate of the coagulant from anode and the hydrogen bubbles generation from cathode and adjusts the rate and size of the bubble production, and hence affects the growth of flocs [100,101]. In general, removal efficiency increases by more CD applied, a fact that has been observed by many authors [38,39]. The amount of pollutant removed is influenced by the quantity of metal cations released by the anode and therefore of $M(OH)_{3(s)}$ adsorbent generated, which is related to the time and CD. The amount of anode material that goes into the solution is given by Faraday's law [Eq. (34)] [102]:

$$m_{\text{theo}} = \frac{I \cdot t \cdot M}{Z \cdot F} \tag{34}$$

where m is the theoretical amount of metal produced (g), I is the current flow (A), t is the contact time (s), M the molecular weight of anode (g/mol), Z the number of electrons transferred in the reaction at the electrode, and F the Faraday's constant (96,500 C/mol electrons).

In addition, the current efficiency (CE) may be calculated according to Eq. (35) [103]:

$$CE = \frac{m_{\text{exp}}}{m_{\text{theo}}} \times 100 \tag{35}$$

where m_{exp} is the experimental value of electrode mass released into solution. It may be obtained by the electrode mass difference before and after EC treatment.

From the literature, a wide range of CDs are applied in EC treatment, varying between 4 and 180 mA/cm² [87,104,105] depending on the amount of pollutants and dye to be removed from wastewater. As it is mentioned above, increasing CD can contribute positively to the removal efficiency. Increasing coagulant and bubbles generation rate leads to the decrease of bubble size, resulting in a rapid removal of COD and color [89,106]. Nevertheless, further increase in the CD causes an excessive amount of oxygen evolution that inhibits the mass transfer to the electrode surface and cause a reduction it the removal efficiency of OM [107]. Alam et al. [108] have stated that the flux of $O_{2(g)}$ and $H_{2(g)}$ on the electrodes' surface increases and their mean bubble size decreases with the increase in the CD up to 100 A/m². A further increase in CD above this value causes an increase in the size of free moving bubbles. Thus, further increase in CD above optimal condition did not lead to an increase in COD and dye removal efficiencies. Jing et al. [43] showed that the COD removal rate first increases from 49.41% to 77.46% as the CD rises to 24.73 mA/cm² and then declines to 60.42% with the increase of CD. Ozyonar et al. [109] investigated the effect of CD on Disperse Blue 60 removal by EC treatment. They indicated that the highest dye removal (99%) was achieved at the CD of 80 mA/cm², and then, a decrease in color removal efficiency was observed with raising the CD to 100 mA/cm². Ogedey and Tanyol [29] treated an unsanitary landfill leachate (LL) by EC using Fe et Al as anode and cathode, respectively. They noted an increase of COD removal with an increase in CD up to 25 mA/cm². Further increase in CD above this value will reduce the efficiency of COD removal.

In addition, when CD continuous to increase, the temperature of treated wastewater increases significantly, hence

causing the instability of coagulation phenomenon that can leads to a low EC efficiency. Giwa et al. [110] pointed out that when the CD increases from 988 to 3,951 A/m^2, the temperature of sample increased up to 46°C caused by the electrical energy wasting in heating the water. In addition, increasing CD can reverse the charge of the colloids thereby conducting to a decrease in the removal efficiency and reduces the lifespan of the electrodes. Consequently, an optimum CD should be determined considering other operating parameters.

Moreover, as the CD is linked to the time of treatment through the Faraday's law [Eq. (34)], the best removal was generally depicted at a higher CD value and a shorter operation time or at a low value of CD and a high operation time. Secula et al. [111] treated a sample of indigo carmine dye with iron electrode, the removal was about 96% after 3 h of electrolysis time at 10.91 A/m^2, and it was about 99.9% after 60 min at 54.57 A/m^2. Nguyen et al. [112] reported that the removal of COD, from an artificial wastewater under 40 min of electrolysis time and 15 V, was 35%. Whereas, it did not exceed 25% at 60 min of electrolysis time and 5 V.

It is noteworthy to mention that the energy consumption increases with increasing CD. Therefore, the COD and/or color removal and the energy consumption should be considered simultaneously. Electric energy consumption as a function of operation time t may be determined using Eq. (36):

$$\text{Energy consumption } \left(\text{kWh/m}^3\right) = \frac{U \times I \times t}{V} \tag{36}$$

where I is the electrical current (A), V is the sample volume (m^3), U is the electrical potential (V), and t is the EC time (s).

6.3. Electrolysis time (t) and electrode material

Electrolysis time (t) also influences the treatment efficiency of the electrochemical process. The color and the pollutant removal efficiency depends directly on the concentration of metal ions produced. EC involves two stages namely: destabilization and aggregation [89]. The first stage is usually short; whereas, the second one is relatively long. For a fixed CD, the concentration of metal ions produced by the electrodes for removing contaminant is directly dependent on the EC time. If the duration of electrolysis increases, the concentration of ions and their hydroxide flocs will increase resulting in an increases in the pollutant removal efficiency [33,106,113]. The formation of coagulant in an appropriate and sufficient quantity depends not only on the operation time but also on the anode material [94]. Obviously, selecting electrode material is one of EC operating parameters that not only have impacts on the performance of the process, but it is also affected the cost. Typically, Al and Fe electrodes are preferred for their easily availability, their low cost, and their high electrode dissolution rates. Nevertheless, other types of electrode martial were reported in the literature (e.g., Ag, As, Ba, Ca, Cd, Cr, Cs, Fe, Mg, Na, Si, Sr, Zn, SS, MS, etc.) [21,114].

Tables 4 and 5 show results obtained by researchers in dealing with COD and color from different types of wastewater regarding the electrolysis time (t) and electrode material.

Considering Tables 4 and 5, we can conclude the following important remarks:

- The color and COD removal efficiency depend directly on the concentration of ions produced by the electrodes which depends deeply on the electrolysis time (t). Thus, when the electrolysis period increases, an increase occurs in the concentration of ions and their hydroxide flocs. Cruz et al. [37] monitored the electrode consumption as a function of time of Al and Fe electrodes. The electrode consumption increases from 1 to 9 kg/m^3 and from 0.5 to 9 kg/m^3 when time increase from 20 to 180 min for Fe and Al, respectively.

Cestarolli et al. [118] found that the removal of Eriochrome black dye increases from 0% to 95% when t reaches 60 min. Hossain et al. [89] reported that increasing operating time from 10 to 50 min will increase the removal of COD from 23.97% to 79.86%. The same result was obtained by other researchers [33,120].

- Some investigators found that the dye and COD removal efficiencies reach a maximum value faster by using Fe electrode than Al. In fact, Ainchu et al. [119] reported that approximately 97.17% of vat dye solution removal efficiency was obtained by Al electrode at 30 min against 20 min using Fe electrode, under the same operating parameters. Gündüz and Atabey [36] reported that a 100% of colour removal was obtained at 5 min by using Fe electrodes for Reactive Red 195 decolorizing, against 80% at 10 min by using Al. Jing et al. [43] treated a mineral processing wastewater by EC. Two types of electrodes were tested (Al–SS and Fe–SS). The results showed that a maximum removal of COD was found to be 62.7% by Fe–SS. Golder et al. [72] treated the Trypan blue (TB) dye by EC with Al and Fe electrodes. During the first 30 min of electrolysis, about 67% of TB was removed with Fe electrodes. While to achieve the same removal efficiency, Al electrodes take about 50 min. Presumably, these finding can be explained by the difference of removal mechanism between Fe and Al. In fact, the removal of dye and COD from solutions using Al anode electrode is mainly by EC, while it is by the combination of EC and electro-oxidation (EO) when using Fe electrode [116].
- The opposite trend was obtained by some researchers. Actually, Ghernaout et al. [91] found a 98.96% of C.I. Disperse Yellow 3 decolorizing by using Al electrode against 96.28% for Fe electrode. Cruz et al. [37] studied the removal of COD by different combination of electrodes: Al–Al, Fe–Fe and Al–Fe. The results showed that the amount and rate of COD removal was Al > Al–Fe > Fe, despite the fact that Fe has the highest electrode consumption. They explained that results in terms of higher reactivity and large negative potential of Al than the Fe. Ozyonar and Aksoy [122] investigated the effect of electrode type on COD removal efficiency of EC process, Al and Fe electrodes were experimented in eight different combinations (Fe–Al–Fe–Al, Fe–Al–Al–Fe, Al–Fe–Fe–Al, Al–Fe–Al–Fe, Fe–Al–Al–Al, Al–Fe–Fe–Fe and Al–Al–Al–Al or Fe–Fe–Fe–Fe). They attained a high COD removal efficiency of 80.79% with Al–Al–Al–Al combination

Table 4
Effect of electrolysis time (*t*) and electrode material on color removal

Reference	Conditions	Color removal (%)	Observations
[33]	Pair Al electrodes pH: 7; 1.5 A	98.23	Colour removal increases from 40% to 85% by increasing *t* from 5 to 10 min. Maximum removal at 25 min and remain constant
[106]	Methylene blue (MB) pH: 5.5; CD = 50 A/m^2 Four Al electrodes Four Fe electrodes	At *t* = 24 min; 98 (Al); 100 (Fe)	When *t* increases from 4 to 12 min; color removal efficiency increased: – From 24% to 70% for Al – From 28% to 80% for Fe
[91]	Organic dye C.I. Disperse Yellow 3 (DY); pH: 6.5; CD = 83 A/m^2 Pair Al electrodes Pair Fe electrodes	 96.28 98.96	When *t* increases from 15 to 45, color removal efficiency increases: – From 97.95% to 98.96% for Fe – From 9.49% to 96.28% for Al
[115]	Methylene blue (MB) Pair Fe electrodes 8 mA/cm^2; pH: 12 Pair Fe electrodes Pair Al electrodes	92 97.33 97.17	An increase in *t* from 10 to 60 min yields an increase in the removal efficiency from 68% to 92%. After 20 min, only 10% further removal could be achieved
[116]	Tartrazine dye CD = 120 A/m^2; pH: 5.78 Fe–Fe Al–Al Al–Fe Fe–Al	 99.21 62 50 99	 An increase in *t* from 2 to 5 min: efficiency of color removal increases from 49.53% to 99.21% For other combination *t* was fixed at 6 min
[117]	Paper wastewaters CD = 10 mA/cm^2; pH: 7 Al–Al Al–Fe Fe–Al Fe–Fe	 85 80 60 45	Colour removal increases by increasing *t* from 10 to 45 min. Above this time, removal efficiency remains constant: – From 9% to 85% – From 9% to 80% – From 8% to 60% From 4% to 45%
[36]	Reactive Red 195 CD = 20 mA/cm^2; pH: 8 Fe–Fe Al–Al Zn–Zn	At *t* = 45 min 100 99 97	For an increasing of *t* from 0 to 5 min, the decolorization efficiency was reached 100 75 25
[118]	Eriochrome black Four Al electrodes 7 V, pH: 7	97	By increasing *t* from 0 to 60 min, the removal increases from 0 to 97%. Above 60 min, the removal efficiency remains constant
[119]	Vat dye solution pH: ND; CD: ND Al–Al–Al–Al; pH: 5; Voltage (V); 30 Fe–Fe–Fe–Fe; pH: 5; Voltage (V); 30	 At *t* = 30 min; 98.23% At *t* = 20 min; 97.33%	The removal efficiency from 5 to 15 min was 0% for Fe and increase from 50% to 97% for Al

after 10 min of treatment and at initial pH equal to 6. These results were in harmony with those obtained by [122–124].

Zongo et al. [125] reported that using Fe electrodes generates a very fine brown particles that are less prone to settling than the gel floc formed with Al, on treating of COD from textile wastewater. The same observation has been reported by Nasrullah et al. [105]. Therefore, in some cases a downstream treatment should be added to the EC system using Fe electrode.

- It should be noticed also that the dye and COD removal increases with time until reaching a maximum. Above this value, the efficiency remains constant or decreases despite the time increasing [33,118]. This can be

Table 5
Effect of electrolysis time (*t*) and electrode material on COD removal

Reference	Conditions	% COD removal	Observations
[91]	Textile wastewater Ten Fe electrodes CD = 100 A/m²; pH: 10.4	79.82	From 10 to 30 min, the removal of COD increases from 23.97% to 79.86% Beyond 30 min, removal remains constant
[120]	Oily wastewater 3 Fe (anode) – 3 Al (cathode) pH: 7; 4.5 V	85.7	Removal increases from 42.4% to 83.3% with *t* increasing from 10 to 40 min. Beyond 40 min, the COD removal remains approximately constant
	pH: 7; 10.5 V; *t* = 40 min Different combination: Fe–Fe	95.6	
	Al–Fe	96.9	
	Fe–Al	98	
	Al–Al	99.1	
[37]	Swine slaughterhouse wastewater CD = 25 mA/cm²; pH: 2		Removal increases with *t* for all electrode materials. For Al and Al–Fe, the removal remains constant above 100 min, equal to 90% and 80%, respectively. For Fe electrode, the removal was constant above 160 min with 80% efficiency
	Al–Al	90	
	Fe–Fe	80	
	Al–Fe	80	
[121]	Olive processing wastewater pH: 4.4; CD = 7.5 mA/cm²		
	Al–Al (120 min)	48	For Al–Al, under pH: 4, CD = 15 mA/cm², high removal efficiency of COD during the first 60 min. Above this time, the removal efficiency remained almost constant
	Fe–Fe (120 min)	35	
	SS–SS (120 min)	42	
	Cu–Cu (120 min)	32	
[113]	Carwash wastewater Four Al electrodes	88	From 5 to 50 min of *t*, COD removal percentage increases: – From 81% to 88% for Al electrode – From 82% to 89% for Fe electrode
	pH: 6; CD = 1 mA/cm² Four Fe electrodes pH: 8; CD = 3 mA/cm²	89	
[43]	Mineral processing wastewater pH: 7.1; *t* = 50 min; CD = 13.74 mA/cm²		
	Al–SS	37.7	
	Fe–SS	62.7	Under 19.23 mA/cm²; pH: 7.1; When *t* increases from 15 to 70 min; COD removal increases from 23% to 77.62%. Beyond 70 min, %COD remains constant (Fe–SS)
[105]	Sewage wastewater 1816 A/m²; pH: 7; *t* = 10 min		Under 1,816 A/m²; pH 7; the increase of *t* induces an increase in COD removal until 20 min. After this time, COD removal remained constant
	SS–SS	98.07	
	Al–Al	97.64	1,816 A/m²; time 0.5 h; pH 7; inter-electrode distance 10 mm.
	Fe–Fe	96.14	Green and brown color appeared with Fe electrode

explained by the fact that (i) the maximum adsorption capacity of coagulant was reached or (ii) after a certain period of time, metal ions and their hydroxide flocs cover the electrode surface. This can cause electrode passivation and consequently the concentration of metal ions and their hydroxide flocs became constant. Moreover, the faster removal of hydroxide ions by the gas bubbles generated in the cell could also reduce the removal efficiency.

Nevertheless, the COD and dye removal efficiencies and their variability depend not only on the electrode material and time, but also on other parameters linked to the reactivity and solubility of formed compounds and the final pH [73].

6.4. Conductivity

In general, as a SE, a salt is added to obtain the desired conductivity of the water or wastewater in EC process.

Solution conductivity affects both the CE and electrical energy consumption in electrolytic cells. Therefore, it is necessary to investigate the effect of wastewater conductivity on EC processes for treating COD and dye. The SE consists of anions (chloride, sulfate, nitrate, etc.) and cations (sodium, potassium, ammonium, etc.) that are contained in the waste or added for sufficient conductivity of the medium [126]. Izquierdo et al. [127] reported that Al electrodes can be dissolved in all the SEs, whereas Fe dissolution is little with nitrate salts and potassium sulfate. These results were in agreement with those obtained by literature [32], the best removal efficiency was obtained for $CaCl_2$, KCl, NaCl whereas $NaNO_3$ appears to be an inhibitor for color removal when Fe anode electrode was used. Huang et al. [128] reported that increasing the NaCl concentration from 1 to 2.5 g/L, under 60 A/m² of CD, leads to an increase of the Al dissolution from 28 to 33 mg that corresponds to 118% of faradic efficiencies. Jing et al. [43] observed that the addition of Na_2SO_4 has no effect on the COD removal efficiency. Thus, the most electrolyte used to increase conductivity in EC process is NaCl for its well-known advantages such as:

- Its low cost and availability;
- The presence of chloride ions can destroy the metal oxide passivation layer formed on the electrode surface by the presence of other anions (e.g., HCO_3^- and SO_4^{2-}) that causing lower efficiencies and higher energy [129,130].
- If the chloride anions are present, they may be oxidized into Cl_2 [130]. The last-mentioned can conduct to generating HOCl that can take part in oxidizing the soluble OM [131].

$$2Cl^- \rightarrow Cl_2 + 2e^- \tag{37}$$

$$Cl_2 + H_2O \rightarrow HOCl + Cl^- + H^+ \tag{38}$$

$$HOCl \rightarrow OCl^- + H^+ \tag{39}$$

Sridhar et al. [92] reported that the presence of NaCl has a considerable effect on the percentage of COD and BOD_5 removal up to 1.5 g/L; beyond that, there is no significant reduction in COD and BOD_5. The same result was obtained by Özyonar et al. [132], as the COD and color abatement increases from 71% and 98% to 80% and 99% (pH = 6 and 75 A/m²) with increasing conductivity for 1 to 1 mS/cm during the first 4 min. Above this time, the removal remained constant. Parallely, the authors stated that the energy costs decrease with increasing conductivity. This result was in good harmony with those reported by literature [133]. Hendaoui et al. [42] reported a slight increases in decolorization with low conductivity using NaCl as SE; however, this increase becomes insignificant at a conductivity over than 2,000 μS/cm, then a significant decreased obtained when the conductivity value exceeded 15,000 μS/cm. In fact, a high electrical conductivity was reported having an adverse effect on the color removal. Nunez et al. [35] found a small color removal when treating water with a strong conductivity (51,200 μS/cm), similar result was reported in literature [134]. It has been shown that a high conductivity influences the kinetics and the equilibrium between charged particles during the reactions, which are attributed to the alterations in the ionic strength [135].

However, some researchers founded that introducing SE has neglect or no effect on COD removal efficiency. Eryuruk et al. [136] noted that there is no any positive effect of adding SE on COD removal efficiency. Tak et al. [86] observed that increasing the NaCl concentration from 0.5 to 1.0 g/L slightly increased color and COD removal, and a further increase beyond 1 g/L resulted in a retardation of treatment efficiencies. According to Naje et al. [137], by the increase of NaCl concentration from 0.02 to 0.1 kg/m³, the COD and color removal efficiencies were approximately unchanged. Can et al. [135] reported that by increasing conductivity, the decolorization efficiency decreases as the conductivity increases from 200 to 4,000 μS/cm. The effect of conductivity was related also to the electrode material. In fact, Kobya et al. [90] found that the COD removal efficiency is slightly reduced with the Al electrode whereas slightly enhanced for the Fe electrode when the conductivity increases from 1,000 to 4,000 μS/cm. The same observation was reported by literature [138]. Solution conductivity affects the CE and consumption of electrical energy in electrolytic cells. It was reported that increasing solution conductivity resulted in the reduction of cell voltage under constant CD that caused a decrease in electrical energy consumption [122]. On the contrary, Safari et al. [120] reported that by increasing the SE concentration from 0.25 to 1.25 g/L, the energy consumption increased from 2.63 to 13.24 kWh/m³.

Bejjany et al. [139] have established a correlation between the energy consumed per unit volume and the initial conductivity:

$$W_c \left(Wh/m^3 \right) = 8.036 \sigma_0 \tau_e \tag{40}$$

where (W_c) is the energy consumption and (σ_0) is the initial conductivity of the solution.

6.4.1. Inter-electrode distance (d)

The impact of inter-electrode distance (d) on EC process performance is well studied. The increase in the distance between the electrodes connected to DC current supply created an ohmic drop that results from the ohmic resistance of the electrolyte R, which can be expressed as follows:

$$R_{ohm} = \frac{d}{S.k} \tag{41}$$

where d is the inter-electrode distance, k is the water conductivity and, S is the active anode surface (m²).

From Eq. (41), it is seen that the ohmic resistance increased by increasing the distance between the electrodes at constant anodic surface area and conductivity of solution. At a constant voltage, when the ohmic resistance between electrodes increases, the current passed through electrodes decreases, thereby the rate of anodic oxidation decreased. Therefore, the removal efficiency of COD and color will decrease. Ghosh et al. [140] reported that the increase of d will increase the ohmic resistance and decrease the percentage of dye removal. Dalvand et al. [141]

investigated the effect of *d* on color removal. They showed that when *d* increases from 1 to 3 cm, color removal efficiency decreased from 98.59% to 90.43%. Naje et al. [137] have studied the effect of three inter-electrode distances on color, COD and TS removal. They noticed that the efficiency decreases with increasing the inter-electrode gap. Similar results were also observed by researchers [142] during the treatment of LL. A decrease in the treatment efficiency was observed when the distance between the electrodes was increased from 1 to 4 cm. The better performance was obtained with *d* = 1 cm (60.5% and 47.5% for COD and color, respectively). Niazmand et al. [121] reported that an increase in *d* had an undesirable effect on removal efficiency, especially for COD. Other studies have also reported that the decreasing in the electrode distances are more favorable for the removal of color and COD [116,143].

On the contrary, it has been observed that with the increases in *d*, the percentage removal of color and COD increased. Shankar et al. [144] concluded that the removals of COD, TOC, and color increase from 56% to 65%, 59% to 65%, and 88% to 94%, respectively, when the electrode gap increased from 1 to 3 cm. The optimum distance between electrodes was found to be 1.5 cm. These results are in good harmony with those obtained by [92]. They found that when the electrode distance increased from 1 to 3 cm, the removal of turbidity, COD and BOD_5 increased by about 13%, 12% and 12%, respectively; further increase beyond 3 cm in *d* will decrease the removal efficiency. In fact, when *d* is small, the solid and fluid transfer will be hindered, which may significantly affects the settling and flotation thereby can leads to decrease the removal efficiency.

As it is aforementioned, *d* affects the ohmic resistance, and hence the energy consumption. Nandi and Patel [145] studied the effect of electrode gap on the energy consumption. They noted that the energy consumption increased from 3 to 9 kWh/kg Fe when *d* increases from 1 to 3 cm, under constant CD of 13.9 A/m². Maghanga et al. [146] reported that the energy consumption decreases from 1.2 to 1.15 Wh as *d* increase from 2 to 5 mm, and then increase beyond 5 mm. Hence, the electrode gap is an important parameter that should be optimized to enhance the performance of EC process.

6.4.2. Stirring velocity

The stirring velocity (SV) is a crucial operating factor that should be studied since it affects the performance of EC process. The main function of mixing is to disperse the coagulant and homogenize the EC reactor, which may enhance the mass transfer kinetics and the removal efficiency. Furthermore, the increase in SV rates can contribute to reduce the diffusion layer thickness formed at the electrode surface and increase the collision between metallic ions and hydroxides. Nevertheless, high speed rates, even though it lead to a homogeneous distribution in the EC cell, it may lead to flocs fragmentation in the reactor that are becoming difficult to settling and removing. Sridhar et al. [92] studied the effect of SV on the treatment of egg processing effluent. They reported that the removal of COD increased from 70% to 85% when the SV increased from 50 to 150 rpm, respectively. Beyond 150 rpm, no significant

increase in COD nor BOD_5 removal was observed. The energy consumption decreased from 23 to 15 kWh/m³ as the SV increased. Similar results are obtained by Abbasi et al. [39], who observed that the increasing in the mixer intensity, from 30 to 60 rpm, positively affects the COD and color removal. Naje et al. [137] found that a high removal efficiency of COD (90%), TSS (92%) and color (94%) was obtained when the reactor was operated at 500 rpm, and further increase in SV (750 rpm) leads to a decrease in the reactor performance (COD, 86%, TSS, 88% and color, 92%). Bayar et al. [147] investigated the effect of varying the SV on the performance of COD removal. They reported that the varying SV below or beyond 200 rpm leads to a decrease the pollutants removal and consequently, an increase in the energy consumption will be obtained at both relatively.

6.4.3. Electrode configuration

In EC device, the electrodes may be configured as monopolar (Mp) or bipolar (Bp) in parallel connection or serial connection. In the Mp parallel system, all anodes are connected with other anodes in the cell; and similarly all cathode s are also connected to each other. In the Mp serial configuration, each pair anode–cathode is internally connected but without connection with the outer electrodes.

In the case of Bp systems, only the outer electrodes are connected to the power source with no interconnections between the other electrodes. Each one of the electrodes, excepting the external ones, present different polarity (Fig. 7).

In the EC cell, the electrode arrangement affects not only the removal efficiency but also the energy consumption and the cost. The COD and color removal efficiencies and OCs of Mp and Bp configurations are discussed in several studies. Demirci et al. [18] investigated the influence of electrode arrangement on the removal of color from textile wastewater using four Al electrodes for 60 min reaction time under the same CD 10.4 mA/cm². For Bp connection, the color removal efficiency attained 73.6% with OC of 7.541 €/m³. However, for the Mp in serial connection, the color removal was about 73.1% and the OC was around 5.896 €/m³; in Mp parallel connection, the color removal efficiency reached 72.8% with total OC 2.985 €/m³. Thus, the choice of Mp parallel configuration exhibited high removal efficiency that is negligibly lower than that of Bp and Mp in serial connection but an OC significantly lower than other mode of arrangement. Kobya et al. [149] reported that the Mp parallel mode is the most cost-effective for Al and Fe electrodes for the treatment of COD from textile wastewater compared to Bp and Mp serial mode. However, Fe was the preferred electrode material in EC with Mp parallel system under 30 A/m² of CD and 15 min of *t*, the treatment cost was 0.245 against 0.4 $/m³ using Al electrode. Wang et al. [150] found that the Al electrode in Mp connection was more efficient in COD removal by 62% with an energy consumption of 3.82×10^{-4} kWh compared to Fe electrode by 53% with energy consumption of 3.53×10^{-4} kWh. It was observed also that Fe arranged as Bp configuration gives a very low COD removal (7%). Sharma et al. [101] treated a domestic wastewater using EC cell with Mp parallel connection mode (Fe–Al), which resulted
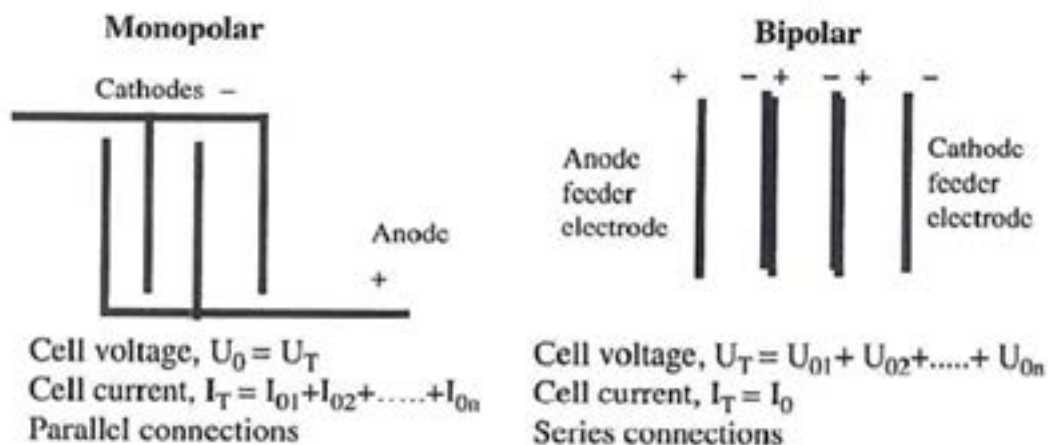
Fig. 7. Monopolar (Mp) and bipolar (Bp) electrode connections [148].

into a removal efficiency of 91.8% of COD at 30 min under 1.25 mA/cm^2 of CD, associated with a very low energy consumption of 0.017 kWh/m^3.

However, Alimohammadi et al. [151] investigated the effect of electrode arrangement on the removal of OM. They found that the Bp electrode connection leads to a percentage of removal of natural OM about 100% compared to Mp electrode arrangement. Moreover, Nasrullah et al. [152] studied the effect of electrode orientation and arrangement on COD removal efficiency from palm oil mill effluent. Three types of electrodes orientation were investigated including vertical orientations, horizontal with anode on the top, and horizontal with anode at the bottom. The best COD removal efficiency was obtained with vertical orientation by 57%. The highest treatment efficiency was obtained by using Mp serial mode arrangement that is able to remove 65% of COD compared to 61% for Mp parallel and 56% for bipolar configuration. They linked the removal efficiency to the amount of anode dissolution. In fact, the anode dissolution were found to be 5.0872, 3.2996 and 3.0176 g for Mp serial, Mp parallel and bipolar configuration, respectively.

Nevertheless, the obtained results aforementioned depend deeply on the (i) electrode material and their number used as anode, cathode and extra plates between anode–cathode on EC cell and (ii) matrix of wastewater. In fact, Ozyonar et al. [153] investigated the influence of four types of combination of Mp parallel arrangement (Al–Al–Al–Al; Al–Fe–Al–Fe; Fe–Fe–Fe–Fe; Fe–Al–Fe–Al). The best removal efficiency of RO 16 dye was 99.87% using the Fe–Al–Fe–Al configuration with CD of 100 mA/cm^2 and 15 min of t. The energy consumption was about 0.772 kWh/m^3. Bellebia et al. [154] reported that using four Al electrodes connected in Mp parallel mode leads to 98.8% on removal of marine blue eurionyle MR under 5 min of t and 1.41 mA/cm^2. The energy consumption was equal to 1.4 kWh/kg dye.

## 7. Sludge and treated water characterization

The sludge characterization produced during EC process is important for two main reasons. The first one is to get a better understanding of by-products for disposal or significant potential reuse. The second reason is the fact that sludge treatment and disposal are among the major contributors to the OC. The EC sludge produced after treatment can be characterized by Fourier-transform infrared spectroscopy (FTIR), SEM with energy-dispersive X-ray spectroscopy (SEM/EDX) and X-ray diffraction (XRD) analysis.

Ozyonar and Korkmaz [155] characterized the sludge produced after EC process with iron electrodes for domestic wastewater treatment. The SEM/XRD analysis reveals the amorphous structure of the sludge and the presence of Fe_3O_4 (magnetite), FeO (vuestite) and FeO(OH) (goethite) in the dry sludge obtained after EC treatment. The chemical composition was determined using EDX, it was observed that it is composed essentially by Fe (57%), C (8.53%), O (31.09), and P (3.19%). The amorphous nature of sludge was also observed by Sahu [156] during the treatment of sugar industry wastewater by EC with iron electrode. The XRD analysis shows also the existence of magnetite (Fe_3O_4) 52%, goethite FeO(OH) 24% and other compounds. In addition, Un and Aytac [157] investigated the sludge produced after EC process with iron electrodes for textile wastewater by XRD. According to the results, the only by-product of EC is maghemite (Fe_2O_3) that can be produced from ferric hydroxide ($Fe(OH)_3$) according to Eq. (42) [73]:

$$2Fe(OH)_3 \rightarrow Fe_2O_3 + 3H_2O \tag{42}$$

The magnetite and goethite are also derived from $Fe(OH)_3$ [Eqs. (43) and (44)] [73]:

$$2Fe(OH)_3 + Fe(OH)_2 \rightarrow Fe_3O_4 + 4H_2O \tag{43}$$

$$Fe(OH)_3 \rightarrow FeO(OH) + H_2O \tag{44}$$
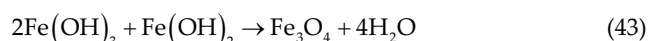
Elhafdi et al. [84] analyzed the EC flocs formed after the treatment of a real textile effluent. The X-ray fluorescence (XRF) analysis showed that solid metal formed by the anode dissolution presents an important part of total mass of

sludge. In fact, the percentage of Fe_2O_3 was 44.6% in the case of Fe anode and 49.6% of Al_2O_3 in the case of Al cathode.

Afanga et al. [158] analyzed the sludge produced after textile wastewater treatment by EC process with iron electrodes for dye and COD removal. The SEM micrograph reveals that the sludge presents an aggregate with non-uniform distribution of particle sizes varying from 4 to 120 μm. The EDX analysis shows that the sludge contains various metallic elements such as, chromium, sodium, nickel and manganese, which required a subsequent treatment.

Mohammed et al. [159] investigated the effect of initial pH on obtained sludge after the treatment of textile wastewater. The infrared spectrum shows that the peaks of functional groups of treated textile wastewater at acidic pH of 4.13 are more clear and sharp than that at alkaline pH of 10.2. These results can be attributed to the difference on mechanism removal, which could be precipitation in acidic pH and via adsorption and precipitation at alkaline pH. Aoudj et al. [160] studied the sludge produced during EC with Al electrode in the absence and in the presence of Direct red 81 ($C_{29}H_{19}N_5O_8S_2Na_2$) using FTIR analysis. The results show a significant spectroscopic changes with apparition of new bands in presence of dye. An intense band at 3,452 cm^{-1} is attributed to stretching vibrations of OH groups. Rodrigues et al. [85] analyzed the sludge formed after the treatment of royal blue color by a fixed bed EC reactor using spherical aluminum and iron particles. The SEM images showed that the flocs formed when using iron have a smoother surface than those obtained by using aluminum. The XRD patterns revealed the presence of boehmite AlO(OH) in the sludge formed when using aluminum anode. In the case of the sludge generated by using iron particles, the XRD revealed the presence of the goethite

In addition to the sludge characterization, a few researchers focused on the analysis of treated water in terms of residual concentrations of anode materials. Indeed, the metallic ions released from anode dissolution should be controlled in order to ensure that EC meets the discharge standards and complies with environmental regulations. Usuga et al. [161] studied the evolution of iron and aluminum ions in the treated solution with time and pH during the treatment of crystal violet dye. When iron electrode was used, the concentration of iron ion decreases from 3.3 to 1 mg/L by increasing the electrolysis time from 3 min to 5 min and the pH of the solution increased to around 7.4. However, by using aluminum electrode, the aluminum ion increases from 17 to 58 mg/L when the electrolysis time increases from 5 min to 60 min, the pH increased from 5.9 to 8 in 5 min. The authors attributed the low iron concentration in solution to the poor solubility of iron species in the considered range of the pH. While, the ion aluminum solubility is minimum when pH is approximately equal to 6.3, and as the pH increases, the soluble $Al(OH)_4^-$ species will be formed. Nunez et al. [35] studied the removal of pollutants from textile industrial wastewater and examined the possibility of reusing the wastewater treated by EC in the process of wool dyeing. For that aim, the level of iron in the treated water was analyzed, it was found lower than 0.2 mg/L. The treated water was then used in dyeing process. The authors concluded that there are no differences in the color of the fabrics dyed with treated and standard

water. Elhafdi et al. [84] have analyzed the content in aluminum and iron of treated water generated from the treatment of textile effluent using EC reactor by aluminum and/or iron electrodes. Under a pH of 6.9 and CD of 31.4 mA/cm^2, the concentration of metallic ions were 1.82 and 1.985 mg/L for aluminum and iron, respectively.

8. Economic assessment

Nowadays, the need of efficient technologies in terms of cost and proper sanitation are among the reasons that will drive the choice for wastewater treatment plant, especially when industrial implementation is concerned. Increasing number of technologies and hybrid processes for water and wastewater treatment pose an enormous challenge for a critical economic assessment of these processes concerning their operational costs (OCs) such as energy used for pumping, stirring and aeration, chemical utilization, sludge disposal, etc., and investment in building materials.

Many technologies have been used for the treatment of dye and COD containing wastewater such as biological, physical, chemical and electrochemical techniques. Considering the environmental limitations of each of the above treatment methods, different combinations and hybrid approaches for dye and COD removal were investigated (i.e., ozone (O_3)/UV, O_3/H_2O_2, EC-electro-oxidation (EO), etc.). Most studies focused essentially on the removal efficiency and the influence of operating conditions on the conducting of the process. There are a few studies discussing the economic side of operation. Such economic analyses can be beneficial to evaluate the profitability of implementing an adequate technology to treat the wastewater and to reach the desired efficiency, also to decide about the feasibility of the treatment at an industrial scale.

Most industries of wastewater treatment are seeking to reduce production costs wherever possible. Since the water process world's electricity consumption accounts for the 2%–3% [162], the issue of energy management as well as removal efficiencies have been increasingly alluring attention. Energy consumption is required at every stage of treatment for the good functioning of the treatment plant. The energy key performance indicators (KPIs) were defined, referred to volume of treated wastewater (m^3), kg of COD removed or dye removed [41,163]:

$$KPI_1 = \frac{U \cdot I \cdot t}{60 \cdot V} \left(kWh/m^3 \right) \tag{45}$$

$$KPI_2 = \frac{KPI_1}{C_0 - C_t} \left(kWh/kgCOD_{removed} \text{ or } kWh/kgDye_{removed} \right) \tag{46}$$

where: C_0 and C_t are the initial and final concentrations of the COD or dye at the end of reaction (kg/m^3).

The total OCs have different forms depending on the technologies used during the treatment. Fig. 8 gives a summarize of main technologies used for decolorization and COD removal and different elements in cost accounting.

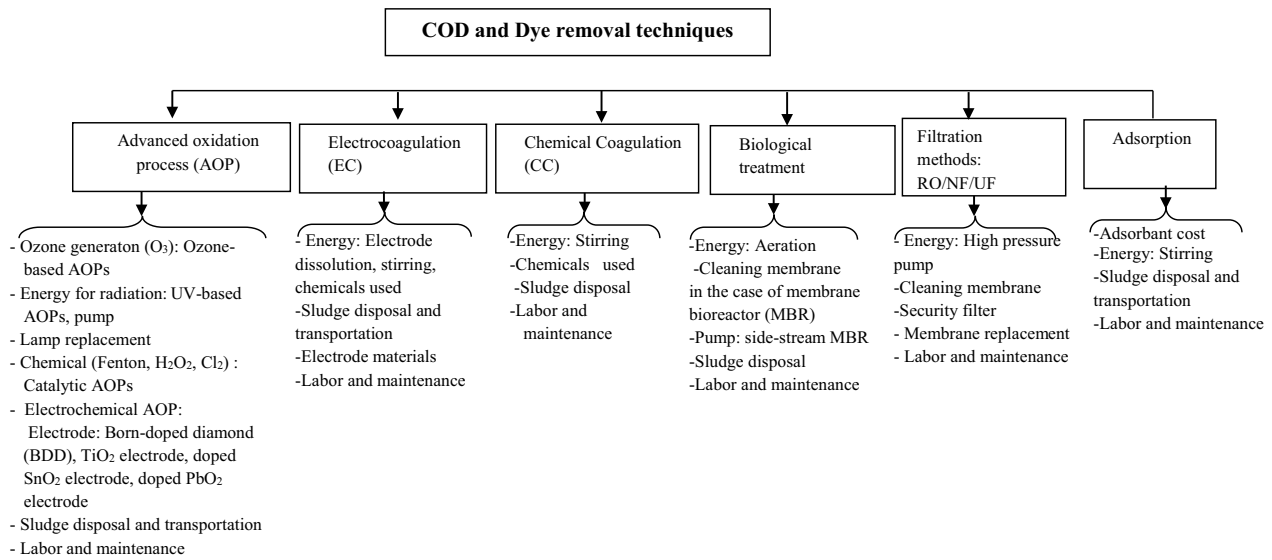An example of calculation of OC for EC cell is given by Eq. (47):

Fig. 8. Operating cost (OC) accounting in different technologies.

$$OC(\text{unit price}/m^3) = a.C_{energy} + b.C_{electrode} + c.C_{chemical} \\ + d.C_{sludge} + MCD \tag{47}$$

where: C_{energy} is the amount of energy consumed for electrochemical cell and other electrical accessories (e.g., sludge pumping) (kWh/m³); $C_{electrode}$ is the electrode consumption (kg$_{electrode}$/m³); $C_{chemicals}$ is the chemical consumption for pH adjustment (kg/m³); C_{sludge} (kg/m³) is the amount of produced sludge (kg$_{sludge}$/m³); MCD is the maintenance cost and depreciation (unit price/m³); a (unit price/kWh), b (unit price/kg$_{electrode}$); c (unit price/kg), and d (unit price/kg$_{sludge}$) indicate the unit price of electrical energy, consumed electrode, chemicals intake, and sludge disposal, respectively. To the OC, a constant capital cost (CC) can be added [164], which covered the cost of equipment (electrolytic cell, mixing apparatus, etc.). Therefore, the total cost is the sum of OC and CC. Table 6 gives the energy consumption and OC of EC and other technologies for different types of wastewater treatment.

8.1. Textile wastewater

The various processes involved in textile manufactories generate huge amount of liquid and solid wastes. The amount and composition of these wastewaters vary following many different factors such as the dyeing process and chemicals used. There are many techniques reported in the literature for textile wastewater treatment.

Najé et al. [137] treated a real textile wastewater by EC using Al electrodes. They indicated a good efficiency removal of COD (92.6%) and color (96.5%). The total OC of the EC process was approximately 1.76 US$/m³ (considering: electrical energy cost, electrode consumption cost, sludge disposition cost, and chemicals cost). The energy consumption was about 8.49 kWh/m³. The energy consumption was little increased when operated EC-EO

process accompanied by a small increase in efficiency (93.5% COD and 97.5% color). Deghles [38] compared the efficiencies and OC using Al and Fe for the treatment of tannery wastewater by EC. Under 20 min of *t* and a CD of 20 mA/cm², EC provided around 75% and 98% of COD and color removal efficiency, respectively, for Al electrode and 81% and 98% of COD and color removal efficiency, respectively, for Fe electrode. He obtained an OC about 0.88 and 0.90 $/m³ for Al and Fe, respectively, associated with the same energy consumption equal to 6.6 kWh/m³. The OC was calculated considering only electrode and energy consumption. Kobya et al. [165] treated a textile dye house wastewater by continuous EC, which provided a decolourization of around 85% and 77% for Fe and Al, respectively. The OC includes all expenses associated with energy, electrode material and chemical costs. It was found equal to 1.562 $/m³ for Fe electrode and 1.851 $/m³ for Al electrode which approximately the same energy consumption. Bayramoglu et al. [166] conducted a comparative study between EC and CC. They showed that the best removal efficiency of COD was obtained by EC process (approximately 63%) and an OC 3.2 cheaper than CC.

AOPs are widely used for the treatment of pollutants and decolorization of wastewater. AOPs are based on the *in situ* generation of oxidizing radical species such as hydroxyl radicals (·OH) and characterized by a high reactivity toward a wide range of pollutants. Khorram and Fallah [167] investigated the energy consumption related to EC and photocatalytic process for the COD and dye removal from industrial dyeing wastewater. Under the optimum conditions, decolorization and COD removal for EC and photocatalytic process were 85.57% and 34.48%, and 81.39% and 67%, respectively. The energy consumption was very high in the case of photocatalytic process of 178.61 kWh/m³ compared to the EC of 0.0032 kWh/m³.

Hybrid systems were also reported in the literature for the treatment of Dye and COD. Bilińska et al. [168] studied the combination of EC and O_3 that is considered as a strong

Table 6
Energy consumption and operating cost (OC) of EC and other technologies for different types of wastewater

References	Wastewater	Technology	Operating conditions details	COD removal (%)	Color removal (%)	Energy consumption (kWh/m³)	OC
	Textile wastewater						
[166]		EC (batch)	Al; Fe; CD = 30 A/m²; t = 15 min	65 (Fe) 63 (Al)		– For Fe: 0.7 – For Al: 0.9	0.25 $/m³ 0.4 $/m³
[18]		EC (batch)	Al; Fe; t = 120 min; CD = 104 A/m²	69.3 (Al) 64.1 (Fe)	90.9 (Al) 80.0 (Fe)		6.439 €/m³ (Fe) 4.739 €/m³ (Al)
[137]		EC (batch)	Six Mp Al electrodes Five inner electrodes (Bp–Al) pH = 6; I = 0.6 A; t = 90 min	92.3	96	8.49	1.76 US$/m³
		Electrocoagulation-Electro-oxidation (EC-EO)	Mp Ti/Bp Al pH = 6; I = 0.6 A; t = 90 min	93.5	97.5	9	1.69 US$/m³
[165]		EC (continuous)	Al–Al–Al; Fe–Fe–Fe 0.01 L/min; CD = 65 A/m²; t = 80 min	85 (Fe) 77 (Al)		11.2 (Fe) 11.17 (Al)	1.562 $/m³ (Fe) 1.851 $/m³ (Al)
[167]		EC (batch)	Fe 8 electrodes CD = 50 A/m²; t = 40 min	34.8	85.57	0.0032	
		AOP: photocatalytic process (batch)	pH = 5.8; Aeration: 2 L/min; t = 40 min	67	81.39	178.61	
[38]		EC (batch)	pH = 6 and 7; t = 20 min; 4 Mp-Al CD = 20 mA/cm² 4 Mp-Fe	75 81	98 98	6.6 6.6	0.88 $/m³ 0.90 $/m³
[166]		CC (batch)	1,500 mg/L FeCl₃·6H₂O; pH = 7; t = 25 min	71			0.67 $/m³ (FeCl₃·6H₂O) 0.75 $/m³ (Al₂(SO₄)₃·18H₂O)
			1,500 mg/L Al₂(SO₄)₃·18H₂O; pH = 6; t = 25 min	59			
[187]		Coagulation with FeSO₄ (batch)	450 mg FeSO₄/L; pH = 12	62	99		0.2 $/m³
		Coagulation with FeCl₃ (batch)	500 mg FeCl₃/L; pH = 12	64	61		0.19 $/m³
		Ozonation (O₃) (batch)	15 min ozonation; pH = 11; 20 mg O₃/min	43	97		4.94 $/m³
		Fenton (Fe/H₂O₂) (batch)	450 mg FeSO₄/L; 200 mg/L H₂O₂; pH = 3	78	95		0.59 $/m³
		Fenton-like (batch)	200 mg FeCl₃/L; 200 mg/L H₂O₂; pH = 3	64	71		0.57 $/m³

Ref	Scale	Process	Conditions	Removal	Removal	Value	Cost
[188]		UV/H$_2$O$_2$	960 mg/g of H$_2$O$_2$; 0.5 g/L RB5 Lamp UV 150 W; pH = 7; t = 270 min		100	177	
		UV/H$_2$O$_2$-biological treatment	pH: 7; 543 mg/H$_2$O$_2$ g; t = 420 min		100	~123.9	
[10]		Ozone-UF-RO (primary stage) (continuous)	O$_3$ concentration: 80–100 g/m^3; Oxidation time: 3.20–3.29 h; Pressure UF: 0.15 MPa; Pressure RO: 1.0–1.2 MPa	99.17 ± 0.88	99.11 ± 1.6	0.28	0.293 USD/m^3
[168]		EC (batch)	10 A; t = 8 min		85	60.98	
		O$_3$ (batch)	t = 50 min		90	28.29	
		EC + O$_3$ (batch)	0.057; t = 18 min		90	115.3	11.86 USD/m^3
		EC O$_3$ (batch)	10 and 0.164 A; t = 18 min; 0.28 mg/L		90	56.29	5.80 USD/m^3
[9]	Lab-scale membrane	UF/tight NF2	26 L/m^2 h; 3 L/min	78.3	DFZ436:20 DFZ525:51.6 DFZ620:47.6		0.16 $/m^3
		Loose NF1/NF2	27 L/m^2 h; 3 L/min	47.3	DFZ436:100 DFZ525:100 DFZ620:100		0.17 $/m^3
		NF1/RO	65 L/m^2 h; 3 L/min	97.9	DFZ436:100 DFZ525:100 DFZ620:100		0.31 $/m^3
		UF/NF2/RO	72 L/m^2 h; 3 L/min	98	DFZ436:100 DFZ525:100 DFZ620:100		0.21 $/m^3
[10]		Ozonation (batch)	t = 20 min; 384 mg/L	33	94		0.37 €/m^3
		Coagulation using polymer coagulants (PP) (batch)	PP-concentration: 190 mg/L	18	91.4		0.5 €/m^3
		Coagulation using polymer coagulants (CP) (batch)	CP-concentration: 180 mg/L	40	92		0.45 €/m^3
		CP with pre-ozonation (batch)	t = 5 min of pre-ozonation CP-concentration: 165 mg/L	30	94		0.35 €/m^3
		PP with pre-ozonation (batch)	t = 5 min of pre-ozonation PP-concentration: 165 mg/L	20	95.7		0.36 €/m^3
[42]		EC (continuous)	pH = 7.5; U t = 47 V; 2 L/min 30 Fe parallel electrodes		94.08		0.0927 USD/m^3

(continued)

Table 6 Continued

References	Wastewater	Technology	Operating conditions details	COD removal (%)	Color removal (%)	Energy consumption (kWh/m³)	OC
	Dairy wastewater						
[173]		MBR-NF (lab scale pilot)	Hollow fiber module; 0.70 L/h Sludge retention time (SRT): 60 d; 1 Nm³ air/h Permeability NF: 23 L/h m² bar	100	99	17	9.08 (lifespan membrane 1 y) 5.91 (lifespan membrane 7 y)
[171]		EC (continuous)	Al–Al; CD = 150 A/m²; 20 mL/min; pH = 7.03	92		2–3.5	
[172]		Sequencing batch reactors (SBR) (industrial plant)	CD = 519 m³/d	42.7		2.05–3.3	
[189]		EC (batch)	Two Fe electrodes pH = 4.5; t = 60 min; Electric current = 1.5 A	96.36	94.9		1.04 US$/m³
[17]		EC (batch)	Al–Al–Al–Al; CD = 50.5 A/m²; t = 5.06 min; pH: 5	98.9	–	–	0.42 €/m³
			Fe–Fe–Fe–Fe; CD = 65 A/m²; t = 5.21 min; pH: 5	98.84			0.54 €/m³
[190]	High organic wastewater	EC (batch)	Fe–Al; pH: 7.7; t = 60 min; Electric current = 0.5 A	55			0.08–0.24 $/m³
[174]	LL	EC (batch)	Al–Al	51 (pH: 5; 40 mA/cm²; t = 20 min)	88 (pH: 11; CD = 10 mA/cm²; t = 25 min)		5.01–6.24 $/m³
			Fe–Fe	39 (pH = 3; 100 A/m²; t = 5 min)	81 (pH: 11; CD = 10 mA/cm²; t = 25 min)		0.28–5.58 $/m³
[175]	LL	NF (bench-scale filtration module)	Q = 120 L/h; Pressure = 8 bar	91	49		3.35 $/m³
[191]	Waste metal cutting	EC (batch)	CD = 60 A/m²; t = 25 min Al; pH: 5 Fe; pH: 7	93 92			– Fe: 0.497 $/m³ – Al: 0.768 $/m³

Ref.	Wastewater	Process	Operating conditions				Cost
[147]	Pistachio industry wastewaters	EC (batch)	5 Al–5 Al; pH: 6; 60 A/m²; $t = 180$ min	60.1		39.6	
[192]	Egg processing industry wastewater (COD)	EC (batch)	Fe–Fe or Al–Al, $t = 10$ min for Al; $t = 15$ min for Fe; CD = 66 A/m² for Al–Al; CD = 91 A/m² for Fe–Fe	99 (Al) 96 (Fe)		14.2 (Al) 15.1 (Fe)	
[178]	Beer industry	EC (batch)	Al–Al CD = 75 A/m²; $t = 60$ min	46.5	96.7	6.75	0.994 $/m³
		Sono-electrocoagulation (SEC) (batch)	Al–Al CD = 75 A/m², $t = 60$ min; Ultrasound frequency: 37 kHz	60.5	99.2	7.020	1.032 $/m³
		SEC-cross-flow membrane (CFM) (NF) (continuous)	Al–Al; CD = 100 A/m²; $t = 60$ min; Ultrasound frequency: 37 kHz; 800 mL/h	62.6	99.7	8.120	1.830 $/m³
[20]	Metal working wastewater	EC (continuous)	Fe–Fe–Fe–Fe; $t = 40$ min; CD = 90 A/m²	94.3			3.09 $/m³
[164]	Moquette industry wastewater	EC (continuous)	Al–Al; CD = 210 A/m²; $t = 25$ min Fe–Fe; CD = 210 A/m²; 25 min	86 90.4		6.1 2.4	
[179]	High concentration wastewater from an oil and gas field	Combined sequencing batch reactor (SBR) and a ceramic membrane bioreactor (CMBR) (continuous)	VSBR: 360 m³; VCMBR: 17 m³; HRTSBR: 24 h; HRTCMBR: 1 h	99			1.04 USD/m³
[193]	High-strength organic wastewater	EC (continuous)	Fe–Fe; 2 A; $t = 10$ min; 3 m³/d	91		13.33	3 $/m³
[41]	Table olive debittering wastewater (COD)	EC (batch)	Al–Al; CD = 199 A/cm²; $t = 58.9$ min	75.3		14.92	5.88 USD/m³

Table 6 Continued

References	Wastewater	Technology	Operating conditions details	COD removal (%)	Color removal (%)	Energy consumption (kWh/m³)	OC
[194]	Slaughterhouse wastewater	EC (batch)	Al–Al–Al–Al; CD = 100 A/m²; pH 4; t = 20 min	78.3			2.757 $/m³
			Fe–Fe–Fe–Fe; CD = 100 A/m²; pH 6; t = 20 min	76.7			0.872 $/m³
		Chemical coagulation (batch)	$Al_2(SO_4)_3 \cdot 18H_2O$; pH=7; 200 mg Al^{3+}/L	36.4			
			$FeSO_4 \cdot 7H_2O$, pH = 7; 200 mg Fe^{3+}/L	27.6			
	Other industrial influent		$FeCl_3 \cdot 6H_2O$; pH = 7; 100 mg Fe^{3+}/L	37.4			
[195]	Municipal wastewater	Membrane bioreactor (MBR) (pilot plant)	Two reactors: 4.4 m³ HRT = 16 h; SRT = 100 d	94		6.06 (LMH = 19) 4.88 (LMH = 25)	0.49 (LMH = 19) 0.39 (LMH = 25)
[196]	Industrial waste management pond	EC (batch)	Fe/Fe: 3 anodes and 3 cathodes; CD = 110 A/m²; t = 30 min	45.14		3.30	0.29 $/m³
[181]	Peat wastewater	EC (continuous)	3 Al; CD = N/A; t = 120 min		88.43	2.64	23.21 RP/L ≈ 1.6 $/m³
[182]	Urban wastewater	Batch EC-flotation (ECF)	Al; CD = 200 A/m²; t = 6 min; pH: 5	82		5.781	0.82 $/kg COD
[184]	Municipal wastewater	Adsorption (batch)	Adsorbent: nano zero valent Fe (GT-nZVI); pH: 8; GT-nZVI dosage 3.2 g/L; t = 60 min	87.9			0.440 $/m³
[180]	Simulated oil- and gas-produced water	EC (batch)	4MP-Fe electrodes; CD = 100 A/m²	95.5 (0 miles)		4	0.2 $/m³
		Coagulation-flocculation (bench-scale jar testing)	200 mg/L $FeCl_3$ Rapid mixing (200 rpm; 2 min) Slow mixing (25 rpm; 30 min)	95.5 (0 miles)		0.9	0.4 $/m³
[197]	Treated combined industrial effluent	O_3 (batch)	100 mg O_3/L; pH: 8; t = 10 min	96	89	8–12.5	
		Fe/H_2O_2 (batch)	t = 60 min; 20 mg/L $FeSO_4 \cdot 7H_2O$; 179 mg/L of H_2O_2	69	60	2.6–3.4	
		UV (batch)	5 mW/cm²; t = 30 min	79	57	160–295	
		UV/H_2O_2 (batch)	170 mg/L H_2O_2; t = 25 min	91	82	86–120	
		$UV/Fe/H_2O_2$ (batch)	170 mg/L H_2O_2; 20 mg/L $FeSO_4 \cdot 7H_2O$; t = 3 min	100	97	6–11.8	

Ref.	Type	Process	Conditions			Cost
[198]	Urban wastewater	Four membrane bioreactors (MBRs) industrial plant	Aeration: 0.3–0.6 Nm³/m²h Flux: 100, 150, 620, 755 m³/h		92–96	0.8–1.1
[186]	Domestic wastewater	2 MBRs	SRT = 15 d; HRT = 0.84 d	60–83 80–95		MBR1 ≈ 5.4 MBR2 ≈ 3.8 3
		Activated sludge process (AS)s				
[172]	Meat processing wastewater	Two-stage AS system	2,321 m³/d	54.6		2.72–3.23
[183]	Real urban wastewater Synthetic dye wastewater	Al scrap	pH: 7.85; 1.5 A; t = 5.84 min	81		3.55
[140]	Crystal violet solution	EC (batch)	Al–Al; CD = 1,112.5 A/m²; t = 60 min		99.75	10.0125 0.2141 $/m³
[199]	Blue 140 and Direct Red 23	EC (continuous)	25 pairs of Fe electrodes CD = 30 A/m²; 5 min (Direct Red 23) CD = 40 A/m²; 5 min (Blue 140)	93	99	1.42 (Blue 140) 0.69 (Direct Red 23)
[200]	RO 84 dye	EC (batch)	SS electrode, CD = 110 A/m² Fe electrode, CD = 130 A/m²		99.8 89	2.02 2.8 0.58 $/m³ 0.77 $/m³
[153]	Reactive orange 16 (RO 16)	EC (batch)	Fe–Al–Fe–Al, CD = 1,000 A/m²; pH: 6; t = 15 min		99.87	0.772 0.816 €/m³
	Reactive red 239 (RR 239)		Fe–Al–Fe–Al; pH: 6; t = 20 min; CD = 1,000 A/cm²		100	0.955 0.997 €/m³
[79]	Reactive blue 21	EC (continuous)	Five pairs of Fe electrodes CD = 30 A/m², t = 2–15 min Q = 0.75–0.1 L/min	90	99	0.495
[170]	Remazol black B dye	EC (batch)	pH: 3; CD = 100 A/m² Two SS electrodes	19.8	91	4.274 $/m³/kg COD
		EF (batch)	pH: 3; CD = 10 mA/cm² Two SS electrodes; 1 mM Fe²⁺	26	93	3.360 $/m³/kg COD
		EC + H_2O_2 (batch)	pH: 3; CD = 10 mA/cm² Two SS electrodes 750 ppm H_2O_2	63	98.8	1.785 $/m³/kg COD
		EF + H_2O_2 (batch)	1 mM Fe²⁺;750 ppm H_2O_2	53	91	1.998 $/m³/kg COD

oxidant and widely used due to its high reactivity with dyes and good removal efficiencies. Four configurations were adopted for the treatment of industrial dyeing effluent highly polluted by residual dye: O_3 (single treatment), EC (single treatment), EC combined with O_3 carried out simultaneously (EC + O_3), and EC combined with O_3 separately (EC → O_3). The results showed that all configurations were high energy consumers, their energy consummation was equal to 5.08, 6.58, 10.13 and 3.77 kWh/m³ for EC (alone), O_3 (alone), (EC + O_3) and (EC → O_3), respectively. The highest color removal was obtained with (EC → O_3), approximately 98% (for a time more than 20 min) and a moderate COD removal. For the treatment by EC under 8 min, the decolorization was about 90%. The costs for EC + O_3 and EC → O_3 were 11.86 and 5.80 USD/m³, respectively. These results pointed out the advantage of the combined system (EC → O_3) to reduce energy consumption and OC.

Aydin el al. [10] treated a highly colored synthetic-cotton textile wastewater by CC and compared between metal coagulant (ferric chloride and aluminum sulfate), polymer coagulant (polyethylene polyamine (PP) and cyanoguanidine polymer (CP)), and O_3. Even with high dosages, the metal coagulant has no effect on color removal. The degradation of color needs a high dose with O_3 over 15 min, but COD efficiency did not meet the standard discharge (around 25%–35%). The combined application of polymer coagulation with pre-ozonation leads to achieve a high removal of color about 94% and 95% for CP with pre-ozonation and PP with pre-ozonation, respectively. OCs of ozonation, PP with pre-ozonation, and CP with pre-ozonation were found to be 0.37, 0.50, and 0.34 €/m³, respectively (chemical consumption and ozone production costs were taken into account for cost analysis).

Membrane processes are often used for water and wastewater treatment including UF, NF and RO. The last has traditionally been used for the desalination of brackish and seawater. Several studies proved that good removal efficiencies can be achieved by using membrane filtration [8,9] in dealing with dye and COD. Nevertheless, data related to membrane processes costs remain deficient. There are several challenges with implementing membrane technology for treatment of dye and COD mainly, the high cost of chemical cleaning or membrane replacement resulting of severe membrane fouling, higher energy requirement and secondary waste generation. Hence, different pretreatment techniques are always necessary to increase the lifespan of membrane; this may increase the cost of wastewater treatment and limit the widespread of these technologies. Combining ozone and membrane filtration techniques was reported in the literature. The application of pre-ozonation in the wastewater treatment may increase the performance of both UF and RO membranes [169]. Yin et al. [10] operated a continuous hybrid system composed by ozone-UF-RO for the treatment of textile wastewater. They achieved a high pollutant removal efficiencies, approximately 100% for COD and color. The OC includes all expenses associated with energy consumption (O_3, oxygen pumps), machine maintenance, replacement of UF and RO membranes, and labour. The OC evaluated is equal to 0.24 USD/m³ (for the first stage of water reuse system), which is a low cost considered those aforementioned.

Synthetic dye wastewater has been widely used to simulate the industrial textile wastewater. EC was used as a technique of treatment. Suhan et al. [170] studied the removal of COD and color from synthetic colored containing Remazol black B (RBB) dye by applying the EC, electro-Fenton (E-F), EC with adding H_2O_2 (EC + H_2O_2), and (E-F + H_2O_2). The color and COD efficiencies were obtained about 19.8%, 26%, 63%, 53% and 91%, 93%, 98.8%, 98.8% for EC, E-F, EC + H_2O_2, and EF + H_2O_2, respectively. The estimation of OC was performed considering energy consumption, electrode material, chemical consumption, and other costs such as labor and maintenance. The combination of processes leads to a reduction in the OC. Indeed, EC + H_2O_2 and E-F + H_2O_2 have 1.785 and 1.998 $/m³/kg COD, respectively; whereas it was equal to 4.274 and 3.360 $/m³/kg COD for EC and E-F, respectively.

8.2. Dairy wastewater

Dairy wastewater is generated from the transformation of raw milk into other products for human consumption. Dairy effluents are characterized by high organic content, TSS, phosphorus and nitrogen. Purifying these effluents is required in order to reduce environmental damage. EC is widely used as a process for the treatment of dairy wastewater, even though the industrial application still very limited.

Varank and Sabuncu [17] studied the treatment of a real wastewater from milk-processing factory in the EC batch process, using Al and Fe electrodes. They achieved a COD removal efficiency of 98.84% and 98.9% for Fe and Al, respectively. The OC was calculated considering the electrode consumption, electric energy and chemicals consumptions. It was found to be equal to 0.42 and 0.54 €/m³ for Al and Fe, respectively. Aitbara et al. [171] operated a continuous pilot scale of EC with two parallel plate electrodes of Al for the treatment of industrial dairy effluent. They attained a high pollutant removal efficiency of COD (92%). The specific energy consumption, between 2 to 3.5 kWh/m³, can be achieved under specific condition. The value of energy consumption was near to those obtained by literature [172]. In fact, the authors evaluated the efficiency of industrial sequencing batch reactor (SBR) technology for the treatment of the mixed dairy effluents. The system was able to remove around 67% of COD and 99% of BOD_5. The authors calculated the energy consumption considering these items: mechanical treatment, biological treatment, aeration and sludge processing. The energy consumption was evaluated as 2.05 to 3.3 kWh/m³ wherein the biological treatment with aeration and sludge processing present 40% and 30% of total energy consumption.

In the aim of reutilized the treated water, Andrade et al. [173] evaluated the technical and economic feasibility of hybrid process composed by membrane bioreactor (MBR) followed by NF for dairy wastewater treatment. The removal efficiencies were 100% and 99% for COD and dye, respectively. The total cost was very influenced by the lifespan of membrane. Therefore, for the same initial investment, it was shown that the OC decreases from 9.08 R$/m³ of treated water to 5.91 R$/m³ when the lifespan of membrane increases from 1 to 7 y. The cost of electricity is corresponding to approximately 38% of the OC in the case of annual

replacement of membranes and 59% in the case of lifespan membrane of 7 y. The energy consumption was approximately close to 17 kWh/m³ (for a daily average cost per kWh equal to 0.2050 R$/kWh) that is considered a high energy consumption compared to that in EC technology.

8.3. High organic wastewater

There are many technologies that generate a highly organic wastewater (e.g., leachate generated at landfill sites, wastewater from an oil and gas industry, food industry, etc.). Such strongly polluted wastewater is composed of a mixture of high organic and inorganic contaminants. It is always necessary to treat these effluents to prevent environmental damage. EC method was reported in the literature for dealing with LL wastewater. Apaydin and Özkan [174] operated a batch EC unit for the treatment of LL effluent with height amount of COD (7,560–8,893 mg COD L⁻¹) and color (1,800–4,000 Pt-Co). They tested two types of electrodes (Al and Fe). Their findings depicted a color removal efficiency around 88% and 81% for Al and Fe, respectively, after 25 min of t and CD equal to 10 mA/cm². The COD removal reached 51% for Al electrode (pH = 5, CD = 40 mA/cm², t = 20 min) and 39% for Fe electrode (pH = 3, CD = 10 mA/cm², t = 5 min). They evaluated the OC considering the electrode and energy cost, and found around 5.01–6.24 USD$/m³ in the case of Al electrode and 0.28–5.58 USD$/m³ for Fe electrode. It should be noted that the OC is influenced by the LL age [31,124]. LL wastewater was also treated by membrane technology. Almeida et al. [175] investigated the application of bench-scale NF to treat LL wastewater with COD of 2,258 ± 230 mg/L. They showed a 91% of COD removal and between 45% and 49% of color removal. The cost estimation was performed based on the results obtained in bench-scale filtration tests and extrapolated to a full-scale NF plant with feed flow of 1,000 m³/d. The OC estimation was about 3.35 US$/m³ wherein the cost of membrane (exchange and cleaning) represents 22%.

Kobya et al. [20] operated a continuous EC process for the treatment of metalworking wastewater containing high COD of 17,312 mg/L. Removal efficiency was determined to be 94.3% under 40 min of t, 70 min of hydraulic retention time and 90 A/m² of CD. The OC was found to be 3.09 US$/m³ encompassing the energy consumption, electrodes, chemicals and sludge disposal. Their findings are comparable to those obtained by Niazmand et al. [41] who operated with batch EC an olive debittering wastewater characterized by a COD around 20,000 mg/L. The OC and energy consumption were around 5.88 USD/m³ and 14.92 kWh/m³, respectively [41]. Ehsani et al. [164] treated also a high organic load moquette industry wastewater (COD equal to 23,000 mg/L) by continuous EC. The removal efficacy of COD was obtained around 86% and 94.1% for Al and Fe electrode, respectively. The energy consumption was approximately close to 2.4 and 6.1 kWh/m³ for Al and Fe, respectively. However, it should be noted that the treatment of such types of wastewater by electrochemical oxidation process leads to excessive consumption of electrical energy 100 kWh/m³ [176,177].

Combining treatment systems were also used for dealing with such types of wastewater. Dizge et al. [178] evaluated the usage of EC, combination of sono-electrocoagulation (SEC) and a cross-flow membrane system (NF or RO membrane) integrated to SEC (SEC-CFM) for the treatment of brewery wastewater highly polluted. The results showed a slightly improvement of COD removal by the hybrid system SEC-CFM. Indeed, the color and COD efficiencies were obtained about 96.7%, 99.2%, 99.7% and 46.5%, 60.5%, 62.6% for EC, SEC, and SEC-CFM, respectively. The energy consumption was evaluated to be 6.750, 7.020 and 8.120 kWh/m³ for EC, SEC and SEC-CFM, respectively. The increase in energy consumption for the SEC-CFM was due to the high-pressure pump for producing trans-membrane pressure. The OC encompassing the energy consumption, material cost (electrode and membrane). The OC was around 0.994, 1.032 and 1.830 $/m³ for EC, SEC, and SEC-CFM, respectively.

Biological treatment technology was also operated by researchers to deal with high organic load of wastewater. Wei et al. [179] investigated the use of biological treatment by SBR and a ceramic membrane bioreactor (CMBR) in conjunction to treat high concentration oil and gas wastewater (COD ranged between 20,000–76,000 mg/L). The COD removal reached 99% with an OC of 1.04 USD/m³ (the OC derived only from the energy consumption and labor).

8.4. Other industrial wastewaters

Khor et al. [180] compared the effectiveness of EC and CC in the treatment of synthetic oily wastewater. The energy consumption for CC was evaluated taken into account the energy needed for mixing, the energy demand for $FeCl_3$ production and transportation. In the case of EC, the energy consumption encompassing the electric energy for electrode dissolution, the energy demand for iron sheet production and transportation. They showed that the energy demand of 95% COD removal for EC was around 4.3 times that of CC when no transportation was required. Even though the energy demand of EC is much higher than that required for CC, OCs are higher for CC than EC. Indeed, the OC was about 0.2 and 0.4 $/m³ for EC and CC, respectively. This was attributed to the unit price of chemical coagulant that was higher than iron metal. Sari et al. [181] operated a continuous EC unit for the treatment of peat wastewater with a color concentration of 698 Pt-Co. The OC and energy consumption to achieve 88.43% of color removal were 1.6 USD$/m³ and 2.64 kWh/m³, respectively. The OC includes the energy consumption cost, the cost of Al plates, and chemicals. Elazzouzi et al. [182] used a batch EC-flotation cell to treat a real urban wastewater characterized by a COD of 1,000 mg/L. Under optimum conditions (Al electrode, 6 min of t and 20 mA/cm² of CD), the COD efficacy removal was 82%. The electrical energy consumption was about 5.781 kWh/m³. Treating the same real urban wastewater, Elazzouzi et al. [183] investigated the using of scrap Al as the anode. Under optimum conditions of current 1.5 A, t = 5.84 min, the energy consumption needed to achieve 82% of COD was 3.55 kWh/m³. Mahmoud et al. [184] investigated the adsorption process for the removal of COD from municipal wastewater. They used a green synthesis nanoparticles extracted from soft black tea. Under optimum conditions (pH 8, dosage

3.2 g/L, contact time 60 min), the removal efficiency was 87.9%. The OC was calculated as the total sum of chemicals, adsorbent material, and energy consumption, and it was found to be 0.440 \$USD/m³. The biological process includes essentially activated sludge and MBR, which are commonly used in urban wastewater treatment to remove pollutants such as COD, nitrogen, phosphorus [185]. The highest energy consumption is attributed to the aeration system necessary for the secondary treatment and to manage membrane fouling in addition to the pumping system. Skouteris et al. [186] compared the energy consumption by two MBRs with conventional activated sludge for the treatment of municipal wastewater. The energy consumed was about 5.4 and 3.8 kWh/m³ for MBR1 and MBR2, respectively, and the air blowers are the almost-energy consuming, representing 88% of the total energy consumed by MBR1, and 60% of the total energy consumed by MBR2.

From the foregoing discussion we can conclude that:

- The OC is strongly linked to the polluted water matrix; indeed, an operation could be economically reliable for one type but not for the other ones.
- For EC process, the electrical energy consumption and OC depend on the type of electrode material under the same operating conditions.
- The high energy consumption does not mean a high OC.
- Even though the data are not directly comparable due to incomplete data sets (e.g., sludge disposal cost, labor and maintenance are not always included), the EC process was economically reliable compared to other processes and have a best chance for industrial applications.
- Some industrial processes are not applicable alone such as membrane technologies (UF, NF and RO), which need additional steps: up-stream pretreatment to prevent membrane fouling and down-stream for the treatment of brine generated after filtration that leads, in some cases, to an increases of OC.
- Optimized EC-combined system leads to a decreases in energy consumption and OC.
- Despite the good removal efficiency obtained by using AOPs, such techniques are considered highly expensive and involving complex technology.
- Extensive further work, especially in continuous mode, is needed to well conduct the economic study and compare each of these technologies.

9. Conclusion

In the present work, a start-of-the-art of EC process for the treatment of dye and COD focusing on economic assessment is performed. EC is a promising technology for removing contaminants from wastewater. The main advantage of EC lies mainly in its technical simplicity and versatility; and can remove many types of pollutants simultaneously.

The performance of EC process is related to many operating parameters like pH, electrolyte concentration, CD, SV, *d*, and electrodes configuration. For instance, pH is deeply dependent on the electrode material and wastewater characteristics.

The mechanisms of dye and COD removal were evaluated. However, further studies need to be carried out

to more understand the interaction between different phenomena taking place in the electrolytic reactor. Certainly, the development of global model describing complex reactions and interactions between all phenomena happening in the EC process is definitely necessary with a view to facilitate the design, the prediction of pollution removal, and the global optimization tasks.

Applying EC technology for wastewater treatment at industrial scale still very restraint. According to the comparative economic study conducted in this work, the EC process is a highly efficient technology considering the technical and economic performances, compared to others alternatives processes. The energy consumption during the EC process was less than for the AOPs and even compared to the traditional methods (biological for COD removal).

Even if there are some investigations on the cost effectiveness of EC process, they are not enough and further research is required on the actual full-scale OCs based on the guidelines from a lab-scale experiments.

One of the challenges of EC system is using of electric energy, which can be lowered through different management strategies like the integration of renewable energy (solar modules or wind modules). Furthermore, more efforts should be devoted to develop a novel hybrid system of EC-coupled process, in order to better exploit the advantage of different systems of dye and COD treatment. Such hybrid systems can meet the environmental and economic requirements (improving energy recovery) as well as the possibility of water reuse. Thus, it is necessary to develop a model to describe the kinetic of treatment of such hybrid systems.

Conflicts of interest

The authors declare that they have no conflicts of interest.

Acknowledgements

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