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Review

Efficacy of Electrocoagulation Treatment for the Abatement of Heavy Metals: An Overview of Critical Processing Factors, Kinetic Models and Cost Analysis

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Abstract: The electrocoagulation (EC) process introduces coagulants by electrochemical means, and is widely adopted for removing heavy metals, besides other contaminants, such as organic pollutants, suspended and dissolved solids, colloidal materials, etc. However, its capability can vary significantly, depending on the operating conditions. Although most of the investigations so far are limited at the laboratory level with artificially prepared solutions or industrial effluent lacking full- and field-scale studies, the success of the process depends a lot on optimizing the process variable. It has been found that the current density (typically 1–20 mA/cm²), type of electrode (generally aluminum or iron) and minimum electrolysis time are the key process parameters that influence performance. Furthermore, key mechanisms involved in the EC process, including charge neutralization, reduction-oxidation and precipitation/co-precipitation, are crucial for pollutant abatement. This review presents a detailed study undertaking all significant parameters that play a crucial role in the EC process, its mechanism, and improving the efficiency of this process by optimization of these parameters, along with suitable kinetic models.

Keywords: toxic metals; electrochemical coagulation; electrolytic conductivity; optimization; water energy nexus; energy consumption



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1. Introduction

A clean and reasonable water supply to meet the needs of the growing global population has become a great challenge of this century [1,2]. Surface and groundwater, which are a major source of drinkable water supplies, besides desalinated water in water-scarce areas, are commonly contaminated by various pollutants, primarily heavy metals [3,4]. Increasing urbanization and expansion of industries, such as metallurgical industries, electroplating units, mining operations, electronic manufacturing units, fertilizer industries and leather industries, have resulted in discharging a considerable amount of wastewater containing high concentrations of trace metals, besides other contaminants, into the environment [5,6]. Several reports of heavy metal contamination of drinkable water supplies worldwide are available in the literature, of which a few are tabulated below in Table 1. There are

numerous reports on health effects, due to the presence of these trace metals in water supplies [7,8]. Intake of chromium, lead, copper, nickel and cobalt may cause leukaemia, anaemia, hypertension, heart problems and cancer [9,10].

Table 1. Worldwide reports of some important trace metals, along with their source of contamination.

S. No	Heavy Metals	Country/Region	Source Type	Contamination Source/Reason	Reference
1.	Chromium, lead, copper, manganese and iron	North Greece	Surface water used for drinking	Effluents discharged from industries and leaching with soil	[11]
2.	Arsenic, iron, lead, copper and zinc	Amiata and Siena, Italy	Drinking water supplies	Leaching with water-distributing pipes	[12]
3.	Chromium, lead, copper, cadmium and zinc	Dipsiz stream, Turkey	Surface water	Coal-fired power plant	[13]
4.	Arsenic and manganese	South Vietnam and Cambodia	Groundwater	Geogenic	[14]
5.	Cadmium, chromium, lead, copper and nickel	Greece	Drinking water supplies	Geogenic and anthropogenic activities	[15]
6.	Lead, arsenic, cadmium and zinc	Dakahliya Governorate, Egypt	Tap water	Corrosion in plumbing fixtures	[16]
7.	Arsenic	Coastal areas and 64 districts, Bangladesh	Tube wells of shallow depth	Subsurface contamination	[17]
8.	Chromium, lead, copper, cadmium and nickel	Bangladesh	Buriganga river	Effluents from tanneries, other industrial outlets and pesticides	[18]
9.	Arsenic, chromium, and selenium	Makkah, Kingdom Saudi Arabia	Groundwater	Geological activity	[19]
10.	Cadmium and lead	Egypt	Water distribution system	Battery, steel and plastic making industries	[20]
11.	Lead	Southwest Bangladesh	Rainwater harvesting structures	Dissolutions from atmosphere	[21]
12.	Cadmium, Lead	Lagos State, Nigeria	surface water	scrap yard waste	[22]
13.	Cadmium, lead and iron	Kumasi district, Ghana	Groundwater from boreholes	Industrial waste from paint industries, municipal sewage and anthropogenic waste	[23]
14.	Chromium, zinc, copper, manganese, nickel, lead and iron	Goiânia, Brazil	Leachate	Sanitary landfill	[24]
15.	Chromium, zinc, copper, manganese, nickel and iron	North Mathiatis mine Nicosia, Cyprus	Open cast mine	Mine drainage	[25]
16.	Nickel and iron	Qazvin, Iran	Thermal power plant	Boiler, air preheater and washing	[26]
17.	Copper	Eco-city, Bohai bay	Sewage samples	Wastewater treatment plant	[27]

These toxic heavy metals cannot be easily removed from the wastewater without the use of advanced treatment technologies [28]. The practiced methods for removing heavy metals are adsorption, chemical precipitation and aquatic plants [29]. However, most of

these have serious drawbacks, such as low efficiency, long treatment time or high operating cost. For example, advancements in the nano-technological field for water purification and treatment have also engrossed attention [30], although there are a few limitations, including high cost. Membrane filtration techniques have been widely used, but these also have disadvantages, such as membrane fouling and high cost. Therefore, effective and low-cost techniques need to be employed before discharging industrial effluents. Among conventional methods adopted, electrochemical-based approaches are currently used worldwide to remove metal ions because of their unique merits, such as the low consumption of chemicals, cost-effectiveness and simplicity. Electrocoagulation (EC), which is one of the electrochemical methods, has been successfully adopted for the remediation of wastewater from a wide range of pollutants, such as heavy metals [26], organic matter [31], textile dyes [32] and pharmaceutical wastewaters [33]. Preferring the EC method over conventional methods needs proper evaluation and in-depth analysis of various indices like a comparative assessment of the process based on operation compatibility, energy requirement, operational cost, sludge handling, etc. [34]. Therefore, there is a need for a feasible and cost-effective technology for the target removal of trace metals [35].

The EC method has certain advantages over other conventional methods. For example, although the chemical coagulation and EC methods have the same removal mechanism [35], the EC method is better than the chemical coagulation in terms of chemicals consumption, dissipation of coagulants [36], separation of flocs and sludge and sludge quality [37,38].

A few studies on heavy metal removal by conventional treatment methods on the comparative ground are shown in Table 2.

Table 2. Comparative assessment of heavy metal removal by various conventional methods adopted.

S. No	Treatment Technique	Target Metal Ion	Efficiency %	Advantages	Disadvantages	References
1.	Adsorption	Cd ²⁺ & Cr ⁶⁺	55–60	Low cost, simple technique	Moderately efficient, Regenerative power declines substantially	[39]
2.	Nano-adsorption	Pb ²⁺ , Cr ⁶⁺	90–99%	Selective removal, Highly efficient	Costly, bulk production is not possible	[40]
3.	Coagulation and chemical precipitation	Cu ²⁺ , Cr ³⁺ , Pb ²⁺ & Zn ²⁺	99.3–99.6	Easily operational, Ease of sludge settling & dewatering	Costly, high consumption of chemicals, huge sludge production	[41]
4.	Nano filtration	Cu ²⁺	96	Highly efficient, reliable	Low anti-compacting ability	[42]
5.	Ion exchange	Cu ²⁺ & Ni ²⁺	99.14–99.33	Selective removal, high regenerability	Costly disposal problems with regenerative fluids	[43]
6.	Electrochemical	Ni ²⁺ , Cr ⁶⁺	98–100	Highly efficient, no chemical requirement	High operational cost	[44]

Electrochemical method-based water treatment was first noticed and reported long ago in 1889 in England. However, the first application of electrolysis was patented in 1904. Since then, treatment using EC has grabbed attention worldwide. On a larger scale, EC was first used in the USA, way back in 1946, for treating contaminants in drinking water. However, in the later half of the twentieth century, extensive work was undertaken to investigate the process mechanism involved. In the last two decades, the EC process has become quite popular, and it has been used in treating several types of wastewater and water, such as the removal of arsenic-containing smelting wastewater [45], mining and metal-processed wastewater [46], distillery-processed wastewater [47], nitrate-containing

wastewater [48], paper and pulp industry wastewater [48], textile mill wastewater [49], tannery wastewater [50] and laundry wastewater [51]. Accordingly, numerous publications have stated the wide applicability of EC in removing heavy metal ions from water and wastewater [52]. These heavy metals include chromium, nickel, arsenic, cadmium, mercury, lead, copper, zinc, iron, etc. A sample of studies on optimizing and removing these heavy metal ions using the EC method has been tabulated in Table 3 below.

Table 3. A sample of the studies that optimized operating conditions for heavy metal removal using the EC method.

S. No	Metal Ion	Initial Conc. (ppm)	pH	Electrode Material	Current Density (A/m ²) or Current (A)	%Removal Efficiency	References
1.	Ni & Fe	-	8.1	Fe-Fe	1.5 A	99	[26]
2.	Cr, Ni and Cu	-	6	Al-Al	12	84.55, 89.65 and 95.16	[27]
3.	Cr and Cu	Cu ₀ = 20	4 & 6	Al-Al	11.57	93 and 99.4	[53]
4.	Fe, Zn, Mn, Cu, Ni, Cd and Cr	-	2.6	Al-Al	20 mA/cm ²	100	[25]
5.	SO ₄ ²⁻	-	2.4	Fe-Fe	200	10	[54]
6.	Cd	-	7	Fe/Cu/Zn	25 mA/cm ²	99.73, 99.9, 82	[55]
7.	Cr	887	6	Fe-Fe	-	100	[56]
8.	Ni	300	4–8	Al-Al	400	100	[57]
9.	As(III)	50	7.5	Al-Al	60	92.2	[58]
		50	4	Fe-Fe	5.4	98.42	[59]
10.	Hg	41	3–7	Al-Fe	40	99.95	[60]
		20	3–7	Al-SS	30	99	[61]
11.	Pb	41	6	Al-Fe	23	98.2	[62]
		2	7	Fe-Fe	80	99.3	[63]
12.	Zn	75	4–8	Al-Al	400	100	[64]
		20.4	9.5	Fe-Fe	40	97	[52]

Although the EC method is a self-sufficient process in treating heavy metals with only a few exceptions, it is used as a hybrid process, along with some other treatment methods in a continuous phase [52]. However, several studies have used the EC method either as a pre-treatment or a polishing step. For instance, color and chemical oxygen demand (COD) removal from UASB (up-flow anaerobic sludge blanket) pre-treated poultry manure wastewater was carried out using EC as a polishing step. Under the highest substrate loading with HRT (Hydraulic Retention Time) of 8 days, the maximum COD removal attained was 81%, suggesting inadequate treatment by UASB alone as per environmental regulations. Similarly, a recent study conducted by Stylianou, et al. [25] showed significant removal of heavy metals. The results showed a 99.9% removal of Fe, Zn, Mn, and Cu, and 98%, 96%, and 88% for Ni, Cd and Cr, respectively. In another study conducted by Xu, et al. [27], keeping the initial pH of 6, they found a removal efficiency of 95.16% of copper. Thus, EC may be used as a polishing step for various types of wastewater, indicating its suitability to be applied as a preceding or polishing step.

The present work aims to review and analyze past research on heavy metal removal using the EC method. Additionally, it aims to define the role of different key parameters in EC treatment.

2. EC Theory and Mechanism

Overall, the EC method has a quite complex mechanism that includes various simultaneous physio-chemical reactions to generate the coagulants, as shown in Figure 1. The EC process involves in situ production of coagulating ions, with three consecutive phases: (i) production of coagulant by electrolytic oxidation of dissolving electrode (generally Al or Fe), (ii) destabilization of target pollutant, particulate suspension and (iii) aggregation of destabilized particles together to form flocs. The general mechanism undergoing inside an EC cell with dissipative electrodes of metal M can be described in the form of chemical reactions as follows:

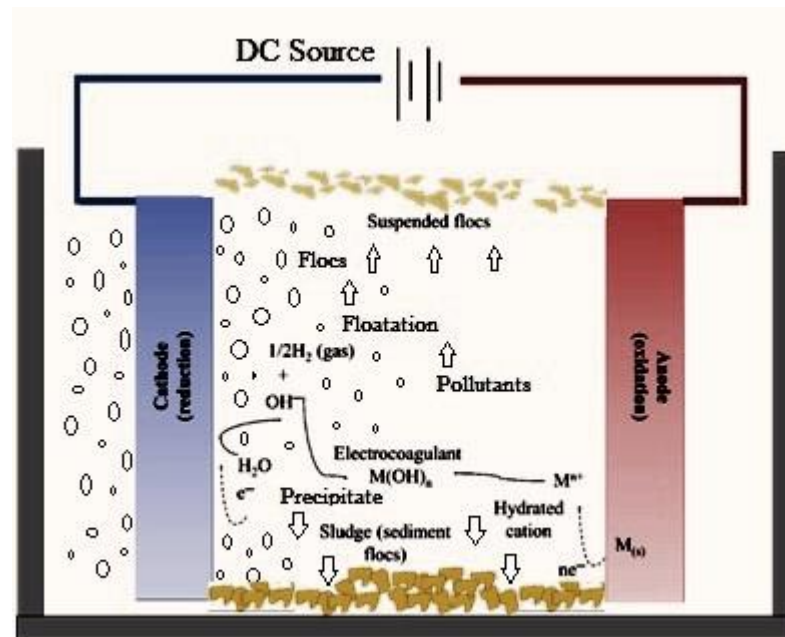
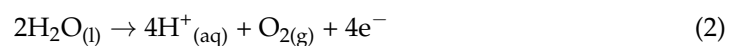
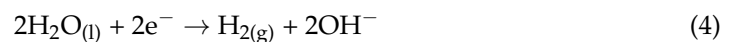


Figure 1. A schematic illustration of a conventional EC reactor.

At the anode:



At the cathode:

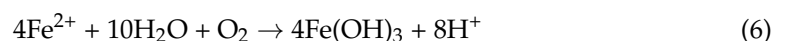


For iron electrodes, $Fe_{(aq)}^{3+}$ ions are formed by electrochemical oxidation, which further reacts to form resultant hydroxide [$Fe(OH)_3$] and polyhydroxide species, namely: $Fe(H_2O)_6^{3+}$, $Fe(H_2O)_5(OH)^{2+}$, $Fe(H_2O)_4(OH)_2^{+}$, $Fe_2(H_2O)_8(OH)_2^{4+}$ and $Fe_2(H_2O)_6(OH)_4^{2+}$, based on the pH conditions. These hydroxides/polyhydroxy metallic complexes show strong empathy towards distributed ions and particles, so they start coagulation. The gases evolved, resulting in the up-flowing of the coagulated particles. The reactions may be shown as follows:

At anode:



Precipitation:



At cathode:



Overall reaction:



In addition to Fe^{3+} production, several reactions occur simultaneously in an EC system. These include:

- formation of H_2 gas at the cathode (Equation (4))
- rise of pH as a result of hydroxyl ion formation (Equation (4))
- reduction of metal ions on the cathodes.

Many researchers have proved the production of O_2 at the anode according to Equation (2) above, though, this does not occur in usual conditions (electrochemical potential) in EC as anodic dissolution obeys Faraday's rule [65]. However, in alkaline conditions, the dissolution of the iron anode is lesser than intended, as per Faraday's law, which specifies that several other electrochemical reactions occur in these environments.

3. Factors Affecting EC and Their Significance

Most of the studies undertaken in the past have undertaken only a few parameters to optimize the EC process. However, to attain maximum removal efficiency with minimum energy consumption, various factors that influence the process, such as the initial pH, applied current, operational time, initial metal ion concentration, electrode material, electrode size, spacing and arrangement, the conductivity of the solution, agitation rate and nature of power supply (DC or AC). All these parameters have their particular role to play and need to be adjusted accordingly for the target removal of contaminants. An insight into these important factors and their role in EC is discussed in detail below.

3.1. Initial pH

pH plays a deterministic role in electrochemical coagulation. It is observed that pH does not remain still during the treatment, but mostly increases as reaction time proceeds [66]. The effect of pH depends on the type of pollutant, the material of anode used, and the reaction mechanism involved. Therefore, understanding the role of the pH in the reaction mechanism is important to achieve the best performance of the EC units [52]. Several studies have explored the role of pH in the performance of EC units. For instance, the effect of pH (from 2 to 10) on the removal of mercury, nickel and lead from solutions was investigated by [67]. The latter used a solution with an initial concentration of 10 mg/L of the mentioned metals, a current density of 0.15 mA/cm², a treatment time of 30min, a temperature of 305 K and an anode and cathode made of magnesium and galvanized iron, respectively. Increasing the pH beyond the optimum value (7) negatively influenced the removal of Hg, Ni and Pb. A similar trend was noticed in the study of Kim, et al. [68], who used two different electrodes (aluminum and iron) to remove Cu, Ni, Zn and Cr from wastewater. The results showed that both electrodes sowed the best removal of the mentioned heavy metals at a neutral or slightly alkaline pH value. However, Xie, et al. [69] reported that the removal of silver using a graphite anode and aluminum cathode is not much influenced by the pH value, where it was noticed that the best removal of the silver was achieved after 20 min, regardless of the pH value. The outcomes of these studies confirmed the fact that the effect of pH on the performance of the EC units depends on several factors, including the type of electrodes and pollutants. Generally, the effects of the pH on the performance of the EC method are attributed to its impact on the species of the coagulants. The literature indicated that changes in the pH value led to the generation of different species of coagulants. For example, in iron electrodes, Fe^{2+} is the dominant specie when the $\text{pH} < 8$, and then $\text{Fe}(\text{OH})_2$ is dominant. The same behavior is noticed with aluminum electrodes. Therefore, the removal efficiency will be changed according to the type of the dominant specie of coagulants [70]. In conclusion, the pH of the solutions being treated by the EC units plays a significant role in the deterioration of the species and charge of the fresh coagulants, which in turn significantly determines the removal efficiency.

3.2. Role of Electrode Material

Selecting and employing feasible electrode material is essential to maximize the efficiency of the EC process. Besides conventional electrode materials, including Fe, Al, Cu, Mg, Sr and Zn, a few other materials, such as graphite and lead oxides, may seldom be used in the EC process as insoluble anodes. Despite the efficiency, easy availability and cost-effectiveness of conventional electrode materials, they have high oxygen evolution potential characteristics. Therefore, many materials were utilized in the EC method to remove pollutants. For example, Safwat [71] investigated the efficiency of zinc (Zn) and titanium (Ti) electrodes for the removal of oils, total dissolved solids (TDS) and COD from solutions at various experimental conditions (current density, spaces between electrodes and time). The results showed that the best oil removal efficiency was 14% by the Zn electrode and 19% by the Ti electrode, and the removal of COD was 50% and 46% by Zn and Ti electrodes, respectively. TDS removal efficiencies were 9% and 19% for Ti and Zn electrodes, respectively. These results showed that Zn and Ti electrodes could remove pollutants when used in the EC process. Hafez, et al. [72] used three different materials, Al, Fe and Zn, as electrodes to remove hardness and dissolved silica from wastewater under different experimental conditions. The obtained results showed the Al electrodes were better than the Fe and Zn electrodes, because Al removed 55.36% and 99.54% of hardness and dissolved silica. However, the low cost, better dissolution and availability of Fe and Al electrodes made them favorable in wastewater treatment practices. Reátegui-Romero, et al. [73] used iron electrodes to remove several metals (Fe, Mn, Cu, Zn and Cd) at pH between 6.6 to 8, a current density of 22.35 A/m², and an application time of 45 min. The results showed the removal efficiency of Fe and Mn at 99.17% and 99.97%, respectively, whereas the removal efficiency of Cu, Zn and Cd was about 99.99%. In another study, Khosa et al., 2013 [74] used aluminum and iron electrodes to remove Ni, Pb and Cd. The results showed the Fe electrodes achieved a better removal; the Al electrodes removed 92.3%, 91.5% and 89.4% for Ni, Pb and Cd, respectively, while the Fe electrodes removed 99.0%, 98.3% and 96.8% of Ni, Pb and Cd, respectively.

The effects of the electrode materials on the performance of the EC method are related to many factors. For example, the chemical and physical properties of electrode materials are different in terms of ions' size, the ions' migration speed in solution, oxidation potential, ion-OH bond polarity and structure of their hydroxide compounds. Therefore, the adsorption capacity of the coagulants is different from one material to another [71,72,75]. The main conclusion of this short literature review is that the material of electrodes determines the chemical composition and adsorption capacity of the coagulants and the size of flocs, which determines the removal of the EC method.

3.3. Role of Current Density

The influence of the applied current density in the EC process is quite noteworthy, as it controls both bubble generation rate and coagulant dosage. The latter two parameters are very important because, firstly, the bubble generation rate determines the removal paths. Increasing the applied current increases the number of the generated bubbles, which results in the predomination of the floatation path in the separation of pollutants, while the low current densities lead to a low production of bubbles that makes the sedimentation path the predominant separation path. Additionally, the high current density increases the anodic dissolution that increases the formation of hydroxo-cationic complexes, thereby improving the removal efficiency (Liu et al., 2010). A considerable number of studies investigate the effects of the current density on the removal of pollutants by the EC units. For example, Manilal et al. [76] applied the EC method to remove oil and chemical oxygen demand (COD) from water, and the experiments were carried out using mild steel electrodes under different conditions, including the current density (between 0.25 to 0.75 A/dm²). The results showed the current density significantly affects the removal of the pollutant; it was noticed the removal of oil and COD increased from about 55% to about 90% when the current density increased from 0.25 to 0.75 A/dm². The authors explained this increase

in the removal of oil and COD with the current density by the rise in the production of the coagulants [$\text{Fe}(\text{OH})_3$ ions] that, in turn, produce more precipitates and consequently improve the removal of pollutants. A similar trend was noticed by Mena, et al. [77] during the removal of fluoride from water using aluminum electrodes under different current densities (in the range between 3 and 15 mA/cm^2). The results showed that increasing the current density from 3 and 15 mA/cm^2 considerably improved the removal of the fluoride from about 60% to about 80%, respectively. The authors also explained the increase in the removal of fluoride with the current density by the increase in the production of the aluminum ions, which improves the removal efficiency. The same effect of the applied current density on the removal of heavy metals (arsenic) from the water was noticed by Müller, et al. [78]. The results showed that the residual concentration of arsenic decreased from about 750 to 10 $\mu\text{g}/\text{L}$ when the current density increased from 1.72 to 0.49 mA/cm^2 after 180 min. The given reason for the increment in arsenic removal with the current density by the increase in the production of $\text{Fe}(\text{OH})_3$ ions that improve the removal of arsenic. The conclusion from these studies is that the effect of the current density on the efficiency of the EC method is related to the production rate of the coagulants from the anodes, which speeds up the removal of pollutants.

3.4. Operational Time

Operational time is another key parameter in EC, as the coagulant concentration generated due to electrolysis is time-bound and directly dependent on the electric charge added per unit volume. With longer durations, more hydroxyl radicals are generated, resulting in more availability of metallic polymer species in the form of hydroxides as adsorbents, thus increasing the removal efficiency [79,80]. However, the removal efficiency of pollutants increases with electrolysis time until it reaches saturation at an optimum condition [59]. This operation time is well associated with current density and thus needs to be optimized, as the removal efficiency desired depends upon the availability of coagulant floc for the target removal of pollutants. The effect of the operating time on the performance of the EC units was widely investigated in the literature; for example, Balarak, et al. [81] used a bench-scale EC unit to remove amoxicillin from water under different operating parameters and noticed increasing the treatment time significantly improves the removal of amoxicillin. The results showed that increasing the treatment time from 10 to 100 min increased the removal of amoxicillin by about 58%. Similar results were noticed in the removal of other pollutants, such as total organic carbon [82], orange II dye [83], Cr [84] and COD [85]. The given reason for the improvement of pollutant removal with the increase in the treatment time is more coagulants will be produced over time, which improves the removal of the targeted pollutants [81,86].

It can be concluded from the reviewed studies that it is essential to provide an adequate period for the EC treatment to generate coagulants, hence, to complete the coagulation of the pollutants.

3.5. Initial Concentration

The amount of metal ions present in an aqueous solution is another important parameter in the EC method. Several studies have been performed in the past to inspect its influence on the performance of the EC method [87,88]. For example, the effect of initial arsenic concentration on its removal efficiency was investigated by Chen, et al. [89] by preparing solutions with an initial concentration of zinc ranging from 50–2000 mg/L at a pH of 2.9–7.4 and a current density of 8.3 mA/cm^2 . It was observed that higher removal efficiency was achieved at lower initial concentration values, and it was found the residual concentration of zinc in the solution increased from 0 to about 1250 mg/L when the initial concentration of zinc increased from 50 to 2000 mg/L , respectively. Moreover, the removal rate was observed initially to be quite high in the early 10 min of the process; however, the rate declined afterwards. Moreover, it was found that amount of coagulant generated was almost the same having no effect of varying the initial concentration, due to which

the generated coagulant was insufficient for the complete removal of metal ions at higher initial concentrations. Thereby, with a higher initial concentration of the ions present, there is more power consumption to attain the complete removal of target metal ions. These findings agree with another study carried out in treating Cr(VI) with initial concentrations of 329.55 to 1170.45 ppm using an EC unit supplied with iron electrodes [90]. The results showed increasing the initial concentration of Cr(VI) from 329.55 to 1170.45 ppm decreased the removal efficiency from 99.9% to 84.6%, respectively. The reviewed studies indicated that a decrease in the efficiency of the EC units with the increase of the concentration of the pollutant is due to insufficient production of the coagulant to remove the high concentrations, but this problem can be solved by increasing either the treatment time or the applied current density, which allows the anodes to produce more coagulants and thereby complete the removal of the high concentrations of the pollutants. The conclusion here is the decrease in the efficiency of the EC units with the increase in the concentration of the pollutant is not related to the performance of the EC reactor; it is related directly to the production rate of the coagulants. Therefore, this problem can be overcome by applying high current density to accelerate the dissolving of the anodes or by increasing the treatment time.

3.6. Electrode Arrangement

Typical arrangement of electrodes inside an EC reactor may be either bipolar or monopolar, such arrangements have been tried in various studies carried out in the past as shown in Figure 2. Several studies have comparatively reported the removal efficiency and operating cost from the monopolar and bipolar arrangement [91–93]. For instance, a group of researchers (Khaled et al. [94]) employed both monopolar and bipolar configurations using 4 aluminum electrodes for the removal of cadmium, with results showing rapid removal of up to 95% taking just 30 min, in comparison to when only two electrodes were used, where it took 45 min for almost the same performance. The removal efficiency obtained from the bipolar configuration was 92%, whereas 87% was obtained from the monopolar configuration. More often, effective treatment costs are incurred, and a high removal efficiency rate makes bipolar configuration more favorable. In another study, the removal efficiency of 99.84% and 89.58% for Cr (III) from bipolar and monopolar configuration after 20 min of EC at a current density of 25 mA/cm² was reported [95]. In an earlier alternate study, the same removal efficiency from monopolar and bipolar configuration was found but reported bipolar configuration as energy and cost-effective [63].

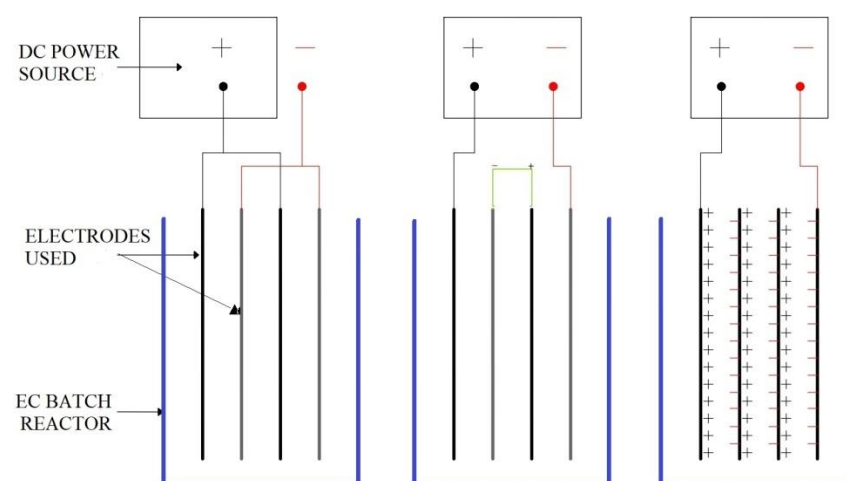


Figure 2. Schematic view of EC system with monopolar electrodes in parallel and monopolar electrodes in series and bipolar electrodes in series connections.

3.7. Electrode Spacing

Spacing between electrodes is considered a vital parameter in the design of the reactor for the efficacy of the EC process, because it determines the energy consumption and reactor

size [96]. Generally, the removal efficiency increases with the increase in electrode spacing until it reaches the optimum distance; then, the travel time of ions between electrodes increases resulting in reduced removal efficiency. Additionally, the literature shows that the potential developed in the EC units due to internal resistance (IR), is directly proportional to electrode distance; as the electrode spacing increases, the IR drop increases [75]. This drop in the IR increases power consumption and decreases the efficiency of the EC units. Therefore, optimizing the distance between electrodes is vital in the EC units [97]. Hence, most literature included the distance between electrodes as a crucial parameter. For example, several experiments were conducted by Mohammadi et al. [98] to measure the effects of different spaces between electrodes (3, 5, 6 and 7 cm) on the removal of nitrogen using an EC unit with iron and aluminum electrodes. It was found that closely spaced electrodes achieved higher removal efficiency; increasing the distances between electrodes from 3 to 7 cm decreased the removal of nitrogen from about 53 % to 20% and increased the power consumption from 4.75 to 19.01 KWh/kg of nitrogen, respectively. The authors explained the decrease in the performance of the EC unit by the fact that short distances decrease the IR drop, which facilitates the travel of ions between electrodes and minimizes power consumption. The relationship between the distance between electrodes and the IR is shown by the following equation [99]:

$$IR = \frac{\text{Cuurent (A)} \times \text{Distnace between electodes (m)}}{\text{Effective anode area (m}^2\text{)} \times \text{Specific conductivity (10}^3\text{ mS/m)}} \quad (9)$$

The same results were noticed by other researchers, such as Naje et al. [100], Maghanga et al. [99], and Liu, et al. [101], and they justified the drop in the removal efficiency with the increase in the distance between electrodes by the reduction in the IR and the long travel time for the ions between the electrodes. The conclusion here is that the distance between electrodes must be optimized not because it affects the removal efficiency but because it also affects the power consumption, which determines the operating cost of the EC units.

3.8. Agitation

To sustain a uniform distribution of coagulants and pollutants inside an EC cell, stirring or agitation at a particular intensity is generally employed [102]. Obviously, by agitating the solution, the velocity attained by generated ions due to anode oxidation increases accordingly, which enhances the contact between the pollutants and the coagulants and thereby improves the removal efficiency. However, increasing the stirring speed more than the optimum value leads to breaking the fresh flocs, negatively affecting the removal efficiency [94,103]. The influence of agitation speed on the EC process has been investigated by numerous researchers in the past. For example, Khaled et al. [94] demonstrated the positive outcome of agitation on cadmium removal from wastewater by EC in which observations were taken at 0, 300, 450, and 600 rpm and found moderate agitation most effective. In this investigation, the maximum attainable removal efficiency was 95% and 99.5%, achieved after 30 min and 60 min, respectively. A slight decrease was noticed in the removal efficiency was observed at higher speeds, 450 rpm and 600 rpm, which could be due to the breaking of flocks and excessive agitation. Similarly, Bao et al. [103] investigated the effect of stirring speed on the removal of three types of perfluoro-alkane-sulfonic acids (PFBS, PFHxS and PFOS) from groundwater using an EC unit having Al and Zn electrodes. The experiments were conducted at stirring speeds of 0 to 1000 rpm, and results showed that a stirring speed of 400 rpm achieved the best removal of PFBS, PFHxS and PFOS (87.4%, 95.6% and 100%, respectively). The decrease in the removal of these pollutants at high stirring speeds was due to the development of a high shear force within the solution that destroys the hydroxide flocs, while the low efficiency at low stirring speeds is attributed to the insufficient contact between coagulants and pollutants (insufficient mixing). The same trend was found by Liu et al. [104] who reported that increasing the stirring speed more than the optimum value (500 rpm) decreases the removal efficiency of perfluoro-octanoic acid from water, and explained this reduction in the removal efficiency by the

prevention of adsorption of the pollutant by the fresh coagulants. It can be concluded here that low or high stirring speeds are not useful for the EC treatment, and these effects can be summarized as follows:

1. High stirring speeds prevent the adsorption of pollutants on the fresh flocs and also break down the aggregates, which decreases the removal efficiency;
2. Low stirring speeds do not create a uniform distribution of pollutants and coagulants in the solution, which also decreases the efficiency of the EC unit.

4. Kinetic Models Applied in EC

Although it has quite a complex mechanism, including various physio-chemical reactions, the adsorption is only one step in the whole electrocoagulation (EC) process. The complexes produced during EC as a result of oxidation in the form of metal hydroxide act as an adsorbent for pollutants in the water and wastewater. These adsorption and complexation steps in the EC process can be modelled using suitable models, such as the first-order, second-order, Dubinin–Radushkevich model, Elovich model, Temkin model, Pseudo-first-order or Pseudo-second-order [52,105]. Each of these models has its peculiarities and significances. For instance, the Pseudo-first-order model describes the adsorbate uptake and is based on solid adsorption capacity, while the Pseudo-second-order model undertakes chemisorption involving the exchange of electrons between metal ions and OH^- . For instance, Yilmaz et al. [106] carried out a statistical analysis, along with an empirical model, to explain the boron removal from wastewater stream employing aluminum electrodes in EC. They reported that the reaction rate followed the pseudo-second-order model quite well.

The pseudo-first-order model is as follows:

$$\frac{dq}{dt} = k_1 (q_e - q) \quad (10)$$

where k_1 is the first-order rate constant, q is the adsorption capacity at any time “ t ” and q_e is the adsorption capacity on attaining equilibrium. Integrating and linearizing this equation, the resulting Equation is as follows:

$$\ln(q_e - q) = \ln(q_e) - k_1 t \quad (11)$$

$\ln(q_e - q)$ is plotted against “ t ” and linear regression is carried out to obtain the best fit. The values of q_e and k_1 were acquired from the slope and intercept of the plots accordingly.

The Pseudo-second-order model is given as follows:

$$\frac{dq}{dt} = k_2 (q_e - q)^2 \quad (12)$$

where k_2 is the second-order rate constant. The following equation can be obtained by integrating and rearranging Equation (12):

$$\frac{t}{q} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \quad (13)$$

The graph of “ $\frac{t}{q}$ ” vs. “ t ” is plotted, and linear regression is carried out to obtain the equation. q_e and k_2 values are depicted from the slope and intercept of the plots accordingly.

Several researchers have applied kinetic modelling in the adsorption of heavy metals in the EC process. Xi, et al. [107] applied both first and second-order models in the removal of cadmium using EC and found better values of correlation coefficients in the case of second-order model plots. Similarly, in another research carried out on the removal of mercury, lead, and nickel using EC, the second-order model was found to be more suitable in explaining the adsorption of all three metal ions over $\text{Mg}(\text{OH})_2$ by employing magnesium

anode [108]. However, in another investigation carried out employing iron electrodes in EC for heavy metal removal, the Pseudo-second-order model was found to fit well [52]. In the same manner, several other researchers have explored the suitability of these models in their studies, a few of which are summarized in Table 4. Thus it may be concluded that various models are used to explain the adsorption step of heavy metal in EC. This may be attributed because the interaction between pollutants and coagulant ions depends on their nature, properties, and operating conditions.

Table 4. Some used kinetic models to explain the adsorption of heavy metal ions in the EC process.

S. No	Target Metal Ions (Pollutants)	Anode Employed	Kinetic Model	Reference
1.	Cu, Zn and Ni	Iron	Pseudo-second-order	[52]
2.	Hg, Ni and Pb	Magnesium	Second-order	[109]
3.	As	Iron and Aluminum	First-order	[110]
4.	Co and Mn	Aluminum	First-order	[111]
5.	Ni and Zn	Stainless steel	Pseudo-first-order	[112]
6.	As	Iron	Pseudo Second order	[113]
7.	Cu, Pb and Ni	Aluminum and Iron	Pseudo-first-order	[114]
8.	Cd	Magnesium	Second-order	[108]
9.	Pb	Magnesium	Second-order	[63]
10.	Fe	Iron	Second-order	[115]

5. Cost Analysis

Cost information and estimation are available in very few researches employing EC. Considerable work at the laboratory and pilot scale had been conducted using EC processes for treating metal-containing wastewater, and it has been speculated that the cost is quite competitive with traditional chemical precipitation and other techniques. The main constraint in the large-scale applicability of the EC process is its operating cost [116]. For wider applicability in treating various industrial effluents, it is necessary to make EC a low-cost technique. Increasing energy prices is an important issue that discourages the applicability of the EC process on a field scale [117]. Moreover, electrodes get sacrificed in dissipating coagulants and thus need to be replaced from time to time, which adds up to the overall operational cost of the process. Therefore, optimization of the EC process is a necessary step in making it cost-effective before any practical use, thereby minimizing energy consumption and attaining higher efficiency. For instance, Khan et al. [28] reduced the overall cost of the EC process to a very low amount in treating per g of Cr(VI) by multivariate optimization of process variables. In another research, response surface methodology (RSM), a mathematical approach, was used to optimize and analyze the effect of the process-dependent variables, affecting the treatment efficiency and thus making the process cost-effective [118]. They attained higher efficiencies at lower operating costs, suggesting the successful applicability of RSM and the need for multivariate optimization as a whole in achieving the objectives. A good illustration of cost variation for the EC process was performed by Shaker et al. [119] on nickel removal at different current densities (5, 10, and 15 mA/cm²). They found that the least EC cost was around 0.7 USD/m³ when the current density was five mA/cm² with an initial pH of 9.2. From an economic aspect, a current density of 5 mA/cm² instead of 10 mA/cm² is more feasible, because the difference in the removal efficiency is insignificant.

Few comparative studies on optimizing the removal of heavy metal using EC have been tabulated, indicating the energy consumption as well as operating cost (see Table 5). Another potential advantage of the EC process is the lesser production of sludge which is generally easier to dewater that might be usefully recovered.

Table 5. A summary of previous EC studies investigated the power consumption and operational cost with optimized conditions.

S. No	Metal	Parameters	Removal (%)	Energy Consumption	Operating Cost		References
					Local Currency	USD	
1.	Copper	Applied current 0.26 A, pH 7, Time 5.4 min, Initial Concentration 27.8 ppm	95%	0.903 W-h/g Cu(II)	0.134 INR/g removal	0.002 US\$/g	[120]
2.	Arsenic	Applied current 0.46 A, pH 7, Initial concentration 10 ppm, Time 2 min	95%	3.1 W-h/g total As	0.0974 INR/g removal	0.0013 US\$/g	[121]
3.	Chromium	Applied Current 1.48 A, pH 3, Time 21.47 min Initial Concentration 49.96 ppm,	100%	12.97 W-h/g Cr (VI)	0.956 INR/g removal	0.013 USD/g	[28]
4.	Copper, Zinc, Nickel and Manganese	Current density 25 mA/cm ² , Time 50 min		49 kWh/m ³	€3.43/m ³	3.77 USD/m ³	[122]
5.	Iron	Initial concentration 20 ppm, Time 20 min, pH 6, Current density 1.5 mA/cm ²	98%	3.2 kWh/m ³		0.22 USD/m ³	[116]
6.	Arsenic and Fluoride	Run time -95 min Flow Rate -0.88 L/h				0.358 USD/m ³	[123]
7.	Copper Chromium Nickel	pH 3.0, Time 20 min, Current 10 mA/cm ²	Cu-100% Cr-100% Ni-100%	10.07 kWh/m ³			[44]
8.	Cadmium	Initial concentration 250 ppm, pH 6, Current density 0.25 mA/cm ²	58%	0.363 kWh/m ³	0.42 €/m ³	0.46 USD/m ³	[124]
9.	Zinc and Copper	Initial Concentration 500 ppm, pH 7, Electric potential 40 V.	Zinc-99.5% Copper-99.7%	Zn- 0.095 kWh/g Cu- 0.29 kWh/g			[125]
10.	Domestic Wastewater	Current density-100 A/m ² , pH 7, Time 10 min	COD- 72% Turbidity-98% Phosphorus- 98%	8.9 kWh/m ³		0.86 USD/m ³	[126]
11.	Fluoride	pH-7, Current density 18.51 A/m ²	85%	3.43 kWh/m ³			[127]
12.	Textile Water	Initial concentration 50 ppm, electrode distance 1 cm, time 30 min	98.59%	1.303 kWh/m		0.256 USD/m ³	[128]

6. Conclusion Remarks and Future Perspectives

The EC method has gained wider acceptance as an alternative to conventional wastewater treatment methods, although its potential needs further exploration. Simple apparatus, expedient operation and no use of chemical substances for settling and floc formation are the main advantages of this process. EC had been successfully applied with a variable degree of treatment achieved for various types of water and wastewater streams. The current density, electrolysis time and electrode type must be carefully considered for each application. It is found that this treatment process has been successfully used to treat different types of heavy metal ions, such as Ni, Cu, Cr, Cd, etc. Optimization of this process not only enhances efficiency, but also makes it possible for its use at a larger scale at an affordable cost. The EC process has proven to be a practical, cost-effective technology for

removing most pollutants. However, in the future, more work needs to be done in process optimization, modelling and scaling up this technique to prove its consistency for efficient and large-scale wastewater treatment. Multivariate optimization techniques, such as RSM, have made the applicability of this technique wider. In addition, sustainable end-use of sludge generated is essential, as it has recently become an attractive research area. Beyond that, we may improve and enhance the efficiency of the EC process by combining it with other conventional techniques or using other methods sequentially as pre-treatment or as a polishing step wherever required.

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