Removal of fluoride, nitrates and phosphor from drinking water using electrocoagulation: A Review

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Abstract. This paper is intended to provide a review of the published literature on the topic of electrocoagulation technology. Water treatment processes containing inorganic anions (fluoride, nitrates and phosphorous) are discussed, including electrocoagulation technology. Electrocoagulation technology is a simple and environmentally friendly electrochemical technique that produces less sludge compared to other treatments. Coagulant production from dissolution of the sacrificial anode and formation of metal hydroxides. The efficiency of the process depends on the type and arrangement of the electrodes, the distance between the electrodes, the operating time, the pH, and the temperature.

1. Introduction

Although 70% of the land is covered by water, just 0.3% of it is freshwater which is available in rivers and lakes. Whereas 90% of water on earth is saltwater contained in oceans [1]. With the population growth around the world, the need for water has increased. Additionally, within the industrial growth as water is utilised for cooling, heating, and power generation. It is treated as a reactant and solvent and, in getting fuel sources (natural gas and oil). Which led to a rise in the contaminants thrown into rivers [2]. The major contaminants responsible for the occurrence of some environmental changes are inorganic anions (for example, the excess of phosphate concentration results in the growth of algae in the water). Additionally, at rising concentrations, inorganic anions are poisonous and cause a lot of health disorders with varying levels of riskiness, such as cancer and disorders of the nervous and digestive systems. However, these anions are essential to human health at the allowable concentrations. Fluoride conserves teeth from caries and is significant in calcification of tooth enamel [3]. The most necessary indications of water contaminant to wastewater and stiff waste are particles generated from the last step of oxidation of nitrogen components, and from origins of nitrate contamination are the nitrogenous fertilisers utilised in farming for the objectives of soil fecundity [4]. Some industries utilise fluoride and its composites on a large scale, such as the rubber and fertilisers industries, power stations that are working using coal-fired boiler, semiconductor manufacture, mineral treatment, ceramics, and glass industries [5]. Nutrients (phosphates and nitrates) are fundamental factors for the flourishing of microorganisms and plants. However, excessive concentrations of these nutrients lead to the plentiful boom of algae and aquatic flora (eutrophication) which cause risky environmental concerns that harm fish and aquatic organisms, and can be a reason for an imbalance of water equilibrium [6]. Potentially, one of the hazardous troubles caused by immoderate concentrations of nitrate ions in freshwater is the blue child syndrome babies regarded as
methemoglobinemia [7]. The following standards for drinking water have been established on the basis of the unsafe and poisonous nature of fluoride, nitrates and phosphorous as shown in table 1 below.

| Pollutants | Limitations       | References |
|------------|-------------------|------------|
| Fluoride   | 1.5 mg/l          | [8]        |
| Nitrate    | 50 mg/l as (NO\textsuperscript{3-}) | [9]        |
| Phosphorus | 0.01-0.025 mg/l as P\textsuperscript{-} | [10]      |

2. Technology for the removal of inorganic anion contaminants from water

Adsorption, precipitation/coagulation, and membrane techniques, including electrodialysis and reverse osmosis are among the most applied techniques. These techniques have succeeded in removing contaminants from low to high levels. But the expenses and efficiency of these techniques have led to a greater consideration of the options. The technology required definitely depends on the infrastructure (that is, the availability of chemicals, resources and manpower), the characteristics of raw water (that is, the degree and the nature of pollution), the acceptability and manpower of raw water (that is, the degree and nature of the pollution), and affordability [11]. Table 2 shows the advantages and disadvantages of methods for treating drinking water containing inorganic anions.

3. Electrocoagulation

Electrocoagulation is an advanced and economical water treatment technique which has been proven to be an effective technique in depolluting heavy metals, breaking down emulsifiers and removing suspended solids [20]. In 1889, electrocoagulation was applied for the first time in wastewater treatment as a method for generating coagulation on site [21]. Due to the demand for alternative wastewater treatment solutions, a revived interest in electrocoagulation (EC) technology has arisen in recent years [22]. EC may be characterised as the making of coagulants in situ through electrochemical reactions of a “sacrificial” anode that lead to dissociation of the anode in the effluent wastewater. Large-scale coagulated metal hydroxides may be generated through metal ions that destabilise and aggregate particles and condense dissolved contaminants. There are many advantages of EC which include without chemical addition, lower costs of capital, sludge reduction, and low power requirements [23]. However, the EC’s disadvantages are: a necessity to replace the electrodes and high conductivity of wastewater [24].
### Table 2. Methods for treating drinking water containing inorganic anions — advantages and disadvantages.

| Process of treatment | Treatment definition | Advantages | Drawbacks | Reference |
|----------------------|----------------------|------------|-----------|-----------|
| **Biological process** | Treatment based on microbiological degradation (e.g., bacteria) | It prepares energy for itself using dissolved organic matter | It takes a longer time | [12] |
| **Precipitation and Coagulation** | Adding of precipitation and coagulants | Easy to understand and generally utilized technique | Required high chemical dosages and Sludge transfer issue | [13] |
| **Membrane processes** | Microfiltration (MF), Nanofiltration (NF), Ultrafiltration (UF), Reverse osmosis (RO) | Using a semi-permeable membrane, the separation is done | Providing high quality drinking water | Technology with high cost compared to other technologies | [14,15] |
| **Ion-exchange** | The crossing of contaminated water via the resin bed | The ability to reuse the resin in addition to regenerating the resin easily | In the presence of other ions lower removal efficiency of some ions | [16] |
| **Adsorption** | On a solid surface concentration of gaseous or liquid molecules | High removal of pollutant efficiency | Difficulties with regeneration | [17] |
| **Electrodialysis (ED)** | Electric potential-driven membranes | Safety, reliability and non-pollution | Ionic species cannot be removed greatly | [18] |
| **Electrocoagulation (EC)** | On-site generation of coagulants by electrically dissolving metal ions | Produces less sludge and less secondary pollution | Sacrificial electrodes need to be replaced periodically due to oxidation | [19] |
3.1. Principles of electrocoagulation

In the electrocoagulation technique, an electrochemical cell is utilized to treat water. In its simplest form, the electrochemical cell is composed of two electrodes, the cathode, and the anode, immersed in the electrolyte or a conductive solution, and are joined together via a DC or AC power supply that includes a controller and a current source, as seen in the ‘figure 1’ below [25]. By applying the electrical current into electrolytic solution in the electrolysis operation, oxidation and reduction processes occur. The sacrificial anode produces mineral ions that act as coagulation ions that act in situ in aqueous solution. These ions are a result of the dissolution of the anode [26]. The electrodes are generally iron, aluminum, and stainless steel due to they are easily available, inexpensive, and non-poisonous. This leads to its adoption as the main electrode material utilized in the EC method [27]. Arrangement of electrocoagulation systems varies in an EC system, the electrodes can be either one or more anode-cathode couples and their connection is in either a monopole or a bipolar [28]. The anode-generated metallic cations hydrolyze to generate poly hydroxides, hydroxides, and poly hydroxyl mineral compounds together with a powerful converge for anti-ions and dispersed particles, leading to coagulation. In fact, owing to the decrease of the disharmony possibility of the electric twin layer, they will reduce the Surface net charge of suspended colloid particles. Van der Waals force prevails when the particles are near enough to lead to particle agglomerate due to the repulsion of the colloidal particles. In chemical coagulation when mineral salts are utilized in water treatment plants, the two phases of coagulation and flocculation are differentiated on the basis of the time required for each of the processes or physically separated between them. In electrocoagulation, the coagulation and flocculation processes happen simultaneously and the two phases cannot be distinguished [29,30].

The possibility of applying the EC process to remove fluoride, nitrate, and phosphorous from water is shown in the table 3.

Table 3. The possibility of applying the EC process to remove fluoride, nitrate, and phosphorous from water.

| Reference | Contaminant | pH | Type of electrode | Processing time (min) | CD (mA/cm²) | % η |
|-----------|-------------|----|-------------------|-----------------------|-------------|-----|
| [31]      | NO₃        | 7  | Al                | 150                   | 3.2         | 94.41% |
| [32]      | NO₃, PO₄   | 7  | Al                | 90                    | 8           | 100% |
| [33]      | F          | 7.6| Al                | 180                   | 1.5         | 96%  |
| [34]      | NO₃        | 11 | Al, Fe            | 75                    | 10          | 86%  |
| [35]      | F, NO₃     | 7  | Al-Cu             | 90                    | 36          | 87.04,89.70% |
| [36]      | P, N       | 6  | Al, Fe            | 40                    | 30          | 99%, 27% |
| [37]      | P          | 7  | Al                | 120                   | 10          | 97.65% |
| [38]      | PO₄        | 7  | Al, SS            | 90                    | 4.54        | 95%  |
| [39]      | F          | 6  | Al plates         | 25                    | 2           | 98%  |
3.2. Factors influencing the mechanism of electrocoagulation

Many factors are considered to influence the performance of the electrocoagulation and its ability to extract contaminants from waste water and water; this section addresses the most relevant parameters:

3.2.1. Electrode materials. Electrode materials are important factors that have a fundamental role in the EC process. These factors are related to the types of ions released from minerals. Metal ions are utilised as coagulants with a higher charge due to improvements in the coagulation of contaminants. Usually, iron or aluminium electrodes are utilised by reason of the coagulation characteristics of polyvalent ions, low cost, high electro dissolution rates, and ease of availability [40].

3.2.2. Arrangement of electrodes. Electrocoagulation may be influenced by the electrode system that includes the distance between the electrodes and the arrangement of the electrodes. The electrode arrangement consists simply of a cathode and an anode, or it is composed of numerous intricately stable cathodes and anodes in an EC cell. The arrangement of the electrodes may be arranged into bipolar or monopolar electrodes:

- Connection in parallel of monopolar electrodes (MP-P) as seen in ‘figure 2a’. It refers to an electrode configuration consisting of anodes and cathodes alternately located at the similarly cathodic or anodic potential, sequentially. All couple of anode / cathode refers to a tiny electrolytic cell. The reactor involves parallel electrolytic cells that a voltage is equal. Then, the current is additive of all electrolyte cells.

- Connection in series of monopolar electrodes (MP-S) as seen in ‘figure 2b’. All couple of inner electrodes is joined together and has no link to the two external electrodes. This cell configuration leads to the overall voltage being the aggregate of a voltage in all singular cells, while the current flowing in all cells is equal.

- Connection in series of bipolar electrodes (BP-S) as seen in ‘figure 2c’. It contains two monopole external electrodes which are located unconnected dipole inner electrodes. The negative direction is subject to cathodic reactions while the anodic decay occurs on the obverse directions. You can say that the obverse directions of any dipole are counter-charged and every one of its directions works together as a cathode and anode [41]. Generally, monopolar electrodes operate under greater current and lower voltage in contrast to bipolar electrodes operating under a low current and great voltage [42]. Depending on the cost-effectiveness ratio, monopolar electrodes can be considered preferred due to in several states this provides depress energy consumption with greater removal of contaminants [43,44].

When choosing suitable electrodeposition, consideration should be given to the influence of
maintenance cost on the total running cost. The last style, which is simple to use requires less maintenance cost through operation [45].

![Diagram of electrode arrangement](image)

**Figure 2.** Schematic diagram of electrode arrangement: (A) Connection in parallel of monopolar electrodes, (B) Connection in series of monopolar electrodes, (C) Connection in series of bipolar electrodes [46].

3.2.3. **Current density and time of treatment.** The current density is defined as the current for each region of the electrode. The utilised current density is directly proportional to the dissociation of a mineral ion. Besides, the number of metal ions emitted from the electrodes is determined. There is a greater chance of electrical energy decline in water heating when a very high current is utilised, and also a reduction in the current efficiency. The current density additionally locates the size of gas bubbles produced whether from the cathode or anode which in turn, impacts the efficiency of the process. If the treated water quality exceeds the critical value of the current density does not display much improvement. Different factors such as water flow rate, temperature, and pH also affect the choice of an optimal value for current density [47,48]. When knowing current and processing times, the coagulant obtained by electrolysis can be identified. The added electrical charge per volume (coulombs per litre) is directly proportional to the coagulation concentration got by electrolysis. Despite this, the whole quantity of coagulant decayed consists of the decay of aluminium cathodes and the chemical dissolution of the electrodes at low pH. Since the current density impacts the average at which the hydroxyl ion is formed at the cathodes, it impacts the chemical decomposition of aluminium cathodes [48-50]. Using an electrochemical cell consisting of an aluminium cathode and a platinised titanium anode, a study was performed on the dissociation of aluminium from the cathode by utilising a different current density and a steady charge per volume (540 C/L). On the basis of the findings, the amount of aluminium generated rises as the current density decreases. The quantity of aluminium generated above the cathodes is depending more on the time of treatment than on the electrical charge applied per volume. The efficiency of the removal of contaminants rises with a rise in the electrolysis time. The average removal becomes steady and does not rise with rising electrolysis after the optimum electrolysis time. The dissolution of the anode generates mineral hydroxides. There is a rise in the formation of flocks for steady current density for a greater period of electrolysis which leads to a high contaminant removal efficiency as the number of metal hydroxide formation rises with rising electrolysis time [51,52].

3.2.4. **Effect of pH solution.** The pH of the water influences the dissolution of metal hydroxides and the current efficiency of the electrocoagulation process. The release of chlorine may also be impaired if there are chloride ions found. In general, the efficiency of aluminium current is greater than neutral
at either alkaline and acidic situations. A more effective removal of contaminants occurs near a pH 7. However, because of the difference in conductivity, the energy exhaustion is higher at the neutral pH. The pH influence is not important at high conductivity. The pH of the effluent reduces after treatment with electrocoagulation for the alkaline effect but rises for the acidic effect. In the reaction of a cathode, a rise in the pH in the acidic situation leads to the evolution of hydrogen [53,54]. The pH reduces due to the generation of Al(OH)$_3$ at the anode, which leads to the release of H$^+$ ions, in addition to the reaction of oxygen evolution.

3.2.5. Conductivity of solution. The conductivity and ionic strength of water are important factors affecting the efficiency of the current density. There is a reduction in the ohmic resistance of water because of the increase in electrical conductivity, which leads to a rise in the efficiency of the current density. Furthermore, conductivity reduces the time wanted for treatment to reach a specific removal product [55]. Thus, energy exhaustion is decreased. To raise the electrical conductivity, sodium chloride is usually utilised. In order to prevent sedimentation of calcium carbonate which might create an isolate layer on the surface of the electrodes in hard water, chloride anions also help in lowering the harmful impacts of another anions [47]. Slight pollution can result due to the high conductivity of wastewater treatment. There are standards that must be esteemed depending on whether the treated wastewater is devoting either to be flowed out in the environmental system or to be reused. In accordance with the standard criteria for rising conductivity during drinking water treatment by the EC that determine, the maximum concentration of chloride in industrial effluents at 250 mg/L [56].

3.2.6. Effect of temperature. The collected results have shown that with a rise in the temperature from 15 to 45 $^\circ$C, the effectiveness of removing some pollutants, especially phosphorus, rises from 85.16 to 97.65%, and behind this temperature, there is a little impact on the removal efficiency. According to the Stokes-Einstein equation, both the rate of hydrolysis of Al$^3+$ to Al(OH)$_3$ and diffusivity of Al$^3+$ rise with rising temperature, with a subsequent rise in the rate of carrying off the Al$^3+$ mass from the anode surface to the mass of solution. The performance of the electrocoagulation reactor decreases due to the decline in the dissolution of Al$^3+$ ions. And this is explained by the fact that higher temperatures of solution rise the passivation of the cathode and anode through the generation of preventive layers of aluminium oxide, which leads to a decrease in the efficiency of removing some pollutants after 45 $^\circ$C [57,58].

3.2.7. Initial concentration of pollutant. The initial contaminants concentration varies with seasons and locations. It is also a function of the type of contaminants (inorganic or organic, soluble or not, … etc.). At a fixed current density, with increasing initial contaminant concentration, the contaminant removal efficiency generally declines. Because the amount of metal hydroxide mass-generated is not adequate to coagulate the high quantity of contaminant flocs at higher primary contaminant concentrations [59]. At high contaminant initial concentration, the current density does not generally have any significant influence on the removal efficiency, particularly at the first stages of EC [60]. Modirshahla et al. mentioned that the removal efficiency of 4-nitrophenol declined from 99.9 to 88.7% as its initial concentration rises from 15 to 35 mg/L [61], which is the most common direction reported in the literature, regardless of the pollutant considered.

3.2.8. Effect of anions concentration. The nitrate and chloride ions break down the passive layer generated by inhibiting sulfate ions, hence it reduces colloids destabilisation and current efficiency. Deposition of salts on the electrodes when the concentration of salt is high enough. Where the harmful influence of sulfate ions is greatly declined in the presence of chloride ions. The precipitate produces an insulating layer that reduces the current efficiency through rising the potential between the electrodes [45] [62]. With a rise in the conductivity of the solution, the energy consumption of the EC has declined. Because of the rise in the efficiency of the removal of contaminants as the conductivity of the solution is a significant parameter influencing the energy consumption and efficiency of the EC.
the presence of chlorine ions has also been shown to contribute significantly to disinfection of water. Usually, the conductivity of a solution is improved by adding anions in the form of salts such as NaCl [47].

3.2.9. Interelectrode distance. The small distance between electrodes limits or leads to a decrease in the efficiency of pollutant removal because many methods may be influenced (for example flocculation, sedimentation, electroflotation, coagulation, etc.), precipitation and flocs production are also affected by these factors [63]. The high electrostatic influence impedes particle collision and prevents aggregation [61]. The flocs generation also greatly declines through an excessive distance between the electrodes [64]. In the electrocoagulation cell for a complex electrode arrangement, once its size is determined the distance between the electrodes determines the number of electrodes to be placed. Also the distance between the electrodes determines the treatment time for a batch reactor and the residence time between the cathode and anode for a continuous reactor to reach the desired EC efficiency [65].

3.2.10. Power supply type. By electrolytic oxidation of the sacrificial anode in the electrocoagulation process, metal hydroxide ions are generated on-site that acts as a coagulant and these ions remove contaminants from the solution by precipitation. The majority of the research in the literature have utilised direct current (DC) in the EC process. In the electrocoagulation process, when the direct current (DC) is used, it reduces the efficiency of removing contaminants. Because of the formation of an oxidation layer on the cathode, which decreases the current flow between the cathode and the anode, as well as the generation of corrosion on the anode due to oxidation. By using aluminium alloy as cathode electrode and anode has been made studying the effect of direct current (DC) and alternating current (AC) on removing cadmium from water they obtained a removal efficiency of 96.2% and 97.5% with an energy consumption of 1.002 and 0.454 kWh / kL at a pH7 and current density of 0.2 A / dm² using DC and AC, Respectively. The results show that by using alternating current instead of direct current in the electrocoagulation process, the problem of corrosion formation in electrodes is reduced [66].

4. Conclusions
Because of the many pollutants discharged to the river, especially fluoride, nitrates, and phosphorus. These pollutants, if their concentrations exceed the permissible limits, it will lead to many health and environmental problems. It is crucial to choose a treatment appropriate to the type and amount of the pollutant, the cost and efficiency resulting from it. Unlike traditional treatments such as coagulation and sedimentation, electrocoagulation is an alternative treatment process for water and wastewater that is characterised by simplicity in terms of tools and operation, its appropriate cost and efficiency, and the removal of different types of pollutants and does not require the addition of chemicals. It generates coagulants on-site from the dissolution of the anode and the formation of metal hydroxides thus reducing the production of sludge and materials harmful to the environment. Although electrocoagulation has many advantages, but its disadvantages require the periodic replacement of sacrificial anodes.

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