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Electrocoagulation for drinking water applications

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Electrocoagulation for drinking water applications

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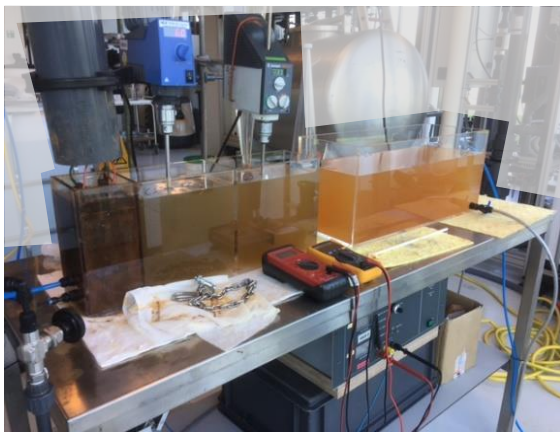
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Managementsamenvatting

Elektrocoagulatie goed alternatief voor coagulatie met FeCl₃

Auteur(s) Julian Muñoz Sierra, Wolter Siegers, Ron Jong

Op dit moment is coagulatie met ijzerchloride de meest gebruikte coagulatietechniek in Nederland. Elektrocoagulatie zou een interessant alternatief kunnen zijn, omdat er geen chemicaliën nodig zijn en de economische aspecten naar verwachting beter zijn. Bij elektrocoagulatie (EC) worden opofferingselektroden van ijzer opgelost in water door het leveren van een stroom, en het opgeloste ijzer werkt dan als coagulant. In een kleine pilotopstelling van 20 l/h zijn oppervlaktewater (PWN), terugspoelwater (Vitens) en nitraathoudend grondwater (WML) getest met elektrocoagulatie en vergeleken met de praktische resultaten van de huidige FeCl₃-dosering. Troebelheid, fosfaat en gesuspendeerde vaste stoffen werden goed verwijderd uit oppervlaktewater en spoelwater dat TOC bevatte. Uit het grondwater werd bijna geen nitraat en sulfaat verwijderd. De investeringskosten voor elektrocoagulatie zijn hoger dan voor het doseren van ijzerchloride, maar de totale OPEX voor EC zou veel lager zijn, bij het energieproces van voor 2022. Het pilotonderzoek en de studie toonden aan dat elektrocoagulatie een goed alternatief is voor de huidige dosering van ijzerchloride. De efficiëntie van het proces kan verder worden verhoogd door de bezinking van de gevormde vlokken door modellering te verbeteren. Er moet meer inzicht komen in de structuur van de investeringskosten van EC. Bij een grotere installatie wordt nu aangenomen dat er meerdere kleine EC-units parallel aan elkaar worden geplaatst.



De elektrocoagulatie pilot in de KWR testfaciliteiten. Op de tafel van links naar rechts: het elektrodencompartiment, de flocculatietanks en de bezinktank.

Belang: alternatieve coagulatiemethode biedt mogelijk voordelen, zoals geen chemische dosering

Op dit moment is coagulatie met ijzerchloride de meest gebruikte coagulatietechniek in Nederland. Elektrocoagulatie zou een interessant alternatief kunnen zijn, omdat er geen chemicaliën nodig zijn en de economische aspecten naar verwachting beter zijn. Bij elektrocoagulatie (EC) worden opofferings-elektroden van ijzer opgelost in water door het leveren van een stroom. Het opgeloste ijzer werkt

dan als coagulant. De belangrijkste redenen om elektrocoagulatie te overwegen in plaats van conventionele coagulatie zijn (i) geen chemische dosering, (ii) er ontstaan beter bezinkbare vlokken, (iii) lagere operationele kosten, (iv) minder impact op het milieu en (v) geen pH-daling en chloortoename in het behandelde water.

Aanpak: literatuurstudie, pilotonderzoek en desktopstudie

Na literatuuronderzoek en een desktopstudie is een pilot uitgevoerd. In een kleine pilotopstelling van 20 l/u zijn oppervlaktewater (PWN), terugspoelwater (Vitens) en nitraathoudend grondwater (WML) getest met elektrocoagulatie en vergeleken met praktijkresultaten van de huidige FeCl_3 -dosering.

Resultaten: elektrocoagulatie geeft veelbelovende resultaten met oppervlaktewater en spoelwater

Elektrocoagulatie geeft veelbelovende resultaten bij de behandeling van oppervlaktewater en spoelwater. Nitraat en sulfaat worden nauwelijks gereduceerd. De investeringskosten zijn hoger, maar de operationele kosten van EC zijn aanzienlijk lager wanneer we uitgaan van de oude energieprijzen. Troebelheid, fosfaat en zwevende deeltjes werden goed verwijderd uit oppervlaktewater en spoelwater met EC (> 91%), TOC werd voor ongeveer 39% verwijderd. Uit het grondwater werd bijna geen nitraat en sulfaat verwijderd. Voor Vitens zijn de investeringskosten 6% hoger voor elektrocoagulatie. Voor PWN was het verschil in investeringskosten groter vanwege het ontbreken van een schaalfactor in de berekening van de EC-kosten. Voor oppervlaktewater van PWN zouden de totale OPEX voor de EC 1785 k€/jaar bedragen, vergeleken met ongeveer 2885 k€/jaar voor conventionele coagulatie, waarbij het grootste aandeel bestaat uit kosten voor chemicaliënverbruik. Voor Vitens zijn de

operationele kosten van EC bij 26 m³/h 27,8 k€/jaar (53%) lager dan de dosering van ijzerchloride bij de behandeling van spoelwater.

Toepassing: elektrocoagulatie is een goed alternatief, efficiëntie nog verhogen

Elektrocoagulatie kan op de gebieden waterkwaliteit, kosten en hanteerbaarheid, eenvoud en veiligheid concurreren met de huidige coagulatietechniek, dosering van ijzerchloride. Het pilotonderzoek en de studie hebben aangetoond dat elektrocoagulatie een goed alternatief is gebruik van ijzerchloride. De efficiëntie van het proces kan verder worden verhoogd door de bezinking van de gevormde vlokken te verbeteren. Modelleren kan hierbij helpen, waarvoor in de literatuur aanknopingspunten zijn gevonden. Samen met een leverancier van EC-installaties moet meer inzicht worden verkregen in de opbouw van de investeringskosten van EC. Bij een grotere installatie wordt nu uitgegaan van meerdere kleine EC units (50 m³/h) parallel aan elkaar worden geplaatst. Dit leidt tot (veel) te hoge kosten voor grote installaties, zoals bij PWN in deze studie (8000 m³/h).

Rapport

Dit onderzoek is beschreven in het rapport *Elektrocoagulatie voor drinkwatertoepassingen* (BTO 2023.057).

Summary

Electrocoagulation (EC) is a process in which iron sacrificial electrodes are dissolved in water by delivering a current, and the dissolved iron then acts as a coagulant. It has been seen as a promising alternative to conventional chemical coagulation in drinking water treatment. The major reasons to consider electrocoagulation over conventional coagulation are (i) no chemical dosing, (ii) the creation of more settleable flocs, (iii) cheaper operational costs, (iv) a smaller environmental effect, and (v) no pH fall and chloride increase in the treated water. Although electrocoagulation has been successfully applied to wastewater, it is still unknown whether it can be applied to drinking water (in the Netherlands) with specific targets such as turbidity, TOC, sulphate, phosphate, and nitrate removal. Therefore, a literature review of the state of the art for drinking water was carried out.

In this laboratory study, the potential of electrocoagulation was investigated for treating three different types of water: surface (basin) water from PWN, backwash water from rapid sand filters of Vitens, and groundwater from WML. The objectives of the research were to assess the effectiveness of removing turbidity, phosphate, TOC, nitrate, sulphate, and suspended solids, at two different electrode gap distances, and to quantify sludge production and costs associated with the process.

The results showed that electrocoagulation was capable of controlled dissolved iron dosing for all three types of water, regardless of the iron dosage or electrode gap distance (0.5 and 1 cm). However, for the groundwater, which targeted nitrate and sulphate removal, the removal efficiencies were not substantial. For surface and backwash waters, the removal efficiencies for turbidity, phosphate, and suspended solids were approximately 94%, 91%, and 96%, respectively, while TOC removal was only about 39%. Furthermore, in contrast to previous literature findings, the setup configuration in this study did not produce heavy, easily and quickly sedimentable flocs.

The investment and operational costs for EC and conventional ferric chloride dosing were determined. The flocculation and flocs removal unit (lamella settling) accounts for the largest share of investment costs in both cases. The investment cost difference for Vitens is 4 k€ higher for electrocoagulation when considering about 65 k€ for the EC unit. For PWN, a larger difference in investment cost (1043 K€) was calculated for electrocoagulation. The operational costs of the EC are 27.8 k€/year (53%) lower than ferric chloride dosage in the case of treating 26 m³/h backwash water from Vitens. For surface water of PWN, the operational costs are 1911 k€/year higher for electrocoagulation due to energy consumption cost of the EC being about 3765 k€/year at an actual cost of 0.35 €/kWh. If this cost goes back in the future to values of about two years ago of 0.07 €/kWh, then the total OPEX for the EC would be 1785 k€/year compared to about 2885 K€/year of conventional coagulation in which the main contribution is due to chemicals consumption costs. At an energy price of 0.17€/kWh, both EC and conventional coagulation operational costs would be similar.

Based on the experimental results, it was found that flocculation and settling could be further enhanced after electrocoagulation (EC), leading to improved removal efficiencies for suspended solids, turbidity, phosphate, and NOM. Modeling techniques can be employed to identify the process's key parameters, which can serve as the foundation for a theory-based design of the installation. Furthermore, sharing knowledge between technologists, engineers from project partners, and technology suppliers can facilitate a more accurate estimation of the investment costs associated with a drinking water EC installation. Energy efficiency (KWh/kg Fe), the use of larger modules/electrodes for EC at high capacity (as in the case of PWN), and the feasibility of the technology given prevailing energy prices are critical factors that need to be carefully considered.

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1 Introduction

1.1 Project Description

Electrocoagulation (EC) is a water treatment process mainly used to treat highly polluted water streams. In this project, after a brief literature study, the efficiency is studied of EC for the removal of suspended matter, natural organic material, iron and nitrate from respectively IJsselmeer water, backwash water from rapid sand filters and groundwater. A desk study on the application of EC for drinking water production based on the experimental results and water types is also carried out, comparing it with current practice and indicating the potential or drawbacks.

1.2 Importance and purpose

Electrocoagulation (EC) is a technique for applying direct current to sacrificial electrodes that are submerged in an aqueous solution. EC is a straightforward and efficient technique to remove the flocculating agent produced by electro-oxidation of a sacrificial anode and generally made of iron or aluminum. Electrocoagulation is currently mainly used in wastewater treatment, for the removal of for example, dyes, dissolved toxic organic compounds and cyanides (see Figure 1-1, Larue et al 2003). Heavy metals and phosphate can also be easily removed with this technique. Excipients such as polyelectrolyte or other flocculation aids (e.g. FeCl_3) are usually not needed.

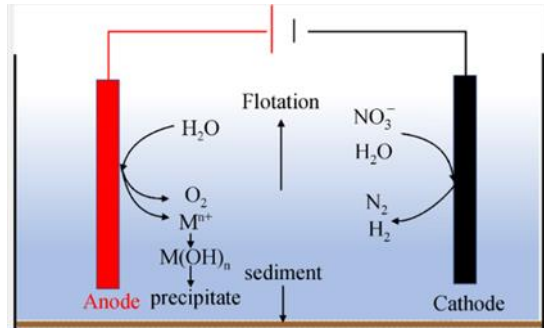


Figure 1-1 Principle of electrocoagulation

The metal of the anode (usually $\text{Fe}(0)$) dissolved by forming ions, which act as coagulant. Electrolysis at the cathode creates hydrogen gas and nitrogen gas is created by reducing nitrate present in the water. Oxidation/reduction of dyes and OMVs present in the water can also take place on the electrode surface. The resulting flocs can be relatively small, part of which can be separated by means of sedimentation or flotation. Further separation from the aqueous phase can be done by post-treatment with filtration, for example sand filtration or membrane filtration.

The conventional coagulation process by dosing iron chloride (FeCl_3), used by PWN to treat IJsselmeer water, consumes a large amount of flocculant annually and also caustic soda to neutralize the pH. The chloride content also increases due to the dosed ferric chloride. With EC, iron is dosed with slow-dissolving iron electrodes and the pH does not need to be neutralized, which will reduce the cost of chemicals and transportation movement. In addition, the chloride content in the drinking water will not increase.

In the case of groundwater, the emphasis is on improving the sedimentation of suspended matter in complex rinsing water of rapid sand filters, to which currently ferric chloride is dosed for improving iron and/or manganese removal in rapid sand filters, which are already equipped with a flocculant dosage. EC is able to dose iron to water without affecting the pH and will therefore have a different effect on flocculation than dosing iron chloride.

Another property of EC that has been described in the literature is the ability to convert nitrate into the harmless nitrogen gas. WML is curious whether EC can be a solution to reduce the nitrate content in drinking water.

In addition to the aforementioned benefits, EC may be able to break down some of the organic micropollutants present in the water.

1.3 Aim

Electrocoagulation involves the formation of precipitating Fe-hydroxide salts in situ by dissolving sacrificial Fe (0) anodes. The Fe (0) oxidizes to dissolved Fe (II), which further oxidizes to Fe(III) in the presence of oxygen, resulting in the precipitation of Fe flocs in water.

The primary objective of this project is to gain insight into the removal yields of suspended solids, natural organic matter, iron, and nitrate from IJsselmeer water, rinsing water, and groundwater using electrocoagulation. By determining the necessary design and settings of the process, overall cost estimates can be made and compared with the current process design.

This research report aims to investigate the applicability of electrocoagulation for treating water from surface water basins, backwash water from rapid sand filtration, and anaerobic groundwater from water utilities such as PWN, Vitens, and WML. During the electrocoagulation process, the most relevant water quality and operational parameters were measured using fast analysis kits and verified with certified laboratory methods, as necessary. The results of the laboratory research will determine whether the electrocoagulation process can achieve high removal efficiencies, providing a basis for possible follow-up pilot installation research.

1.4 Output and application

By measuring the relevant water quality parameters after applying EC to different water types on a laboratory scale, the potential of electrocoagulation for drinking water production can be tested. Analysis of water quality parameter data in combination with optimization of the operational conditions of the EC installation, provide insight into the usability and practical usability as a technology for drinking water production from surface and groundwater. In addition, an exploratory cost estimate is made based on the optimized conditions for electrocoagulation, which is compared with the conventional or current costs. This information contributes to an understanding of the potential and scalability of electrocoagulation in practice, resulting in a valuable input for the project partners, which can be used in decision-making about the water purification process and for initiating further research with this technique.

Based on the experience gained during this research with the operational management of the electrocoagulation technique, recommendations will be made for further optimization and / or the necessary follow-up research. In addition, the potential of electrocoagulation for application to three drinking water sources used / researched in the Netherlands (groundwater, surface water, backwash water) to remove their corresponding targets has been described.

2 Electrocoagulation in drinking water production

This chapter provides insight into the most relevant literature on the main topics that the water utilities Vitens, WML and PWN are interested on, mainly, the application of iron electrocoagulation (EC) for the drinking water production (e.g., in-situ generation of Fe as flocculant) as an alternative to conventional coagulation-flocculation (e.g., dosage of FeCl_3).

2.1 Fundamentals of electrocoagulation

2.1.1 Electrocoagulation principle and process mechanisms

Electrocoagulation (EC) is an electrochemical technique used for treating water and wastewater containing various contaminants (Holt et al. 2005, Mollah et al. 2004, Moussa et al. 2017). In this method an electric current is passed through metal electrodes (iron, aluminum, etc.,) resulting in generation of coagulant species due to electrolytic oxidation of sacrificial anode. It is considered as a one low-cost and chemical-free method of Fe(0) electrolysis, involving the in-situ generation of Fe(III) precipitates (Roy et al. 2021). Depending on solution pH, the metal ions produced from the anode through electrochemical dissolution undergo hydrolysis. This hydrolysis results in formation of hydroxide precipitates (having capability to remove pollutants by adsorption/settling) and other ions metal species that acts as coagulants. Also, the reactions happening at cathode allows pollutant removal by means of deposition on cathode electrode or through flotation due to evolution of H_2 gas at the cathode.

EC process comprises of in-situ production of coagulating ions and consists of the following three successive stages: (i) coagulant formation through electrolytic oxidation of the sacrificial electrode (anode), (ii) destabilization of contaminants, suspensions of particulates, and breaking of emulsions and (iii) flocs formation. The hydroxides, oxyhydroxides and solid oxides produced by EC have active surfaces that allow the pollutants to get adsorbed (Mollah et al. 2004). The EC process uses electrodes like iron, aluminum, zinc etc., for the in-situ generation of metallic coagulants. The commonly used electrodes for EC are made of aluminum (Al) or iron (Fe) materials owing to their easy availability at low price and also the precipitates formed of these metals are non-toxic in nature. Generally, direct current (DC) is used for release of metal ions, hence the anode and cathode are made of same materials (Hakizimana et al. 2017, Mollah et al. 2004).

The EC process mechanism is represented in Figure 2-1.

When a current is applied from an external power source, at anode oxidation reaction takes place where as at cathode reduction reaction takes place. When the electrolytic oxidation of the anode occurs, the metallic anode dissociates into di or tri valent metallic ions and releases equivalent number of electrons (Eq. 4). These metallic migrate to an oppositely charged ions and aggregation occurs due to charge neutralization. The anode also leads to the water reaction/oxidation releasing hydrogen ion and oxygen to the aqueous system (Eq.5). Also, if the water solution contains chloride ions, oxidation of chloride ions to chlorine gas can also take place at anode under high anode potential (Eq. 6). At the cathode, reduction of water takes place. The water molecules dissociates into H^+ ions and OH^- ions by water molecules electron interaction (Eq. 7) (Nidheesh and Singh 2017).

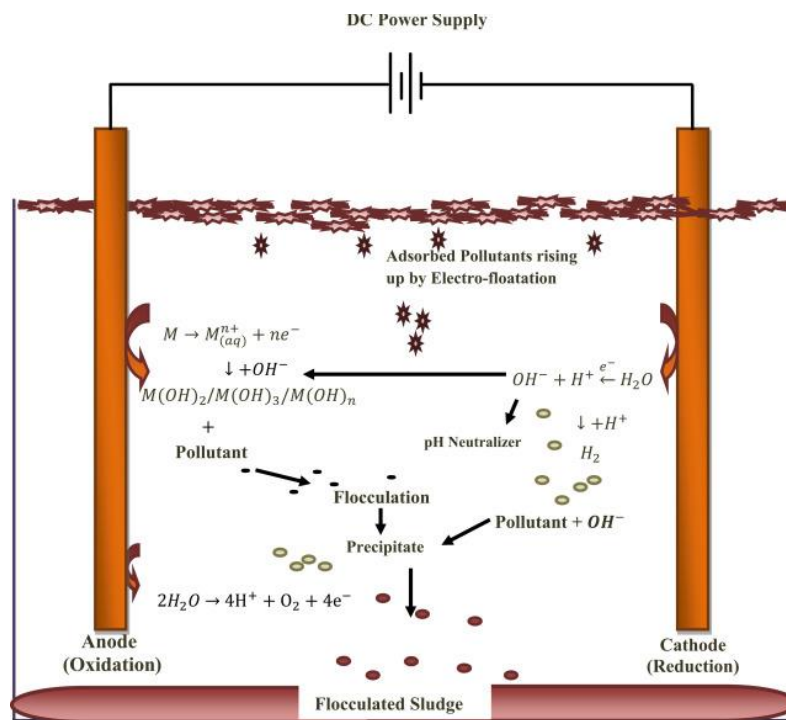


Figure 2-1 Pollutant removal mechanism by EC process (Nidheesh and Singh 2017).

At anode:

- (1) $M (s) \rightarrow M^{n+} (aq) + ne^{-}$
- (2) $2H_2O \rightarrow 4H^{+} (aq) + O_2 (g) + 4e^{-}$ ($E^0 = 1.23 \text{ V}$)
- (3) $2Cl^{-} \rightarrow Cl_2 + 2e^{-}$ ($E^0 = 1.36 \text{ V}$)

At cathode:

- (4) $nH_2O + ne^{-} \rightarrow (n/2) H_2 (g) + nOH^{-} (aq)$

The metallic ions formed in the anode combine with the hydroxide generated at the cathode to form metallic hydroxides, which are very good adsorbents for pollutants. The generation of monomeric or polymeric and soluble or insoluble (amorphous) metallic hydroxides depends on the pH of the aqueous solution (Anantha Singh and Ramesh 2013). The formed amorphous $M(OH)_n$ metallic hydroxides have large surface areas with very high adsorption capacity, form bonds with pollutants, and form flocs (Hakizimana et al. 2017, Kobya et al. 2006, Nidheesh and Singh 2017). The metallic flocs enlarge the size from micro flocs to macro flocs, attaining the self-settling size. The hydrogen gas generated in the cathode and oxygen generated in the anode play a vital role in the removal of macro flocs which never settle by gravitation. The gas generated takes up the micro flocs to the top of the aqueous solution, called electro-floatation.

In an EC process, when a current is applied to the electrodes (e.g., Fe or Al) the following two reactions take place:

- From the anode, Fe or Al ions get released into the solution that hydrolyzes to form polymeric iron or aluminum hydroxide. These polymeric hydroxides act as very good coagulating agents. The metal ions produced at the anode coagulate with the negative particles produced near the cathode. Electrophoretic motion carries the negative particles towards the anode. The contaminants present in a water source are then treated in the EC reactor by chemical reactions and precipitation or by physical and chemical attachment to colloidal materials being generated by the electrode erosion. The coagulants containing the

contaminants can then be removed either by sedimentation and filtration or by electroflotation. Hence, in this process no chemical coagulants are added externally as coagulating species are generated in-situ.

- Electrolysis of water also takes place simultaneously in the EC reactor generating H₂ gas at the cathode that helps in attracting and floating the flocculated particles through natural buoyancy (Deniel et al. 2008, Mollah et al. 2004).

Other than the above two processes, the following physiochemical phenomena can also take place in the EC reactor:

- Impurity reduction at the cathode.
- Colloidal particles can get discharged and coagulate.
- Ion migration in the solution due to electrophoretic motion.
- H₂ bubbles produced at the electrodes can cause electroflotation of the coagulated particles.
- Metal ions reduction at the cathode.

When a constant DC current is applied to the electrode or electrode assembly in an EC reactor, the amount of metal ions released from the electrodes into the solution depends on the current (I) passed through electrolytic solution or the total charge loading (q) (total charge passed through solution by the current), and can be measured using Faraday's law as shown in equation 5. The charge loading (q) can be measured based on the electrode area (A_e), active solution volume (V), current density (J) and electrolysis time (t) as shown in Eq. 6. The charge dosage rate (dq/dt) is proportional to the rate of metal dissolution into a unit volume of solution during electrolysis and can be depicted as in Eq. 7 (Amrose et al. 2013, Kobya et al. 2011)

$$(5) \quad W = qM / nF = ItM / nFV = (IM / nFQ)$$

$$(6) \quad q = JtA_e / V = I t / V$$

$$(7) \quad dq / dt = I / V = JA / V$$

where,

W = amount of dissolved electrode material (mg/L); I = current (mA); t = electrolysis time (min); M = molecular weight of metal; F = Faraday's constant (96,485 C/mol); n = number of transferred electrons; V = solution volume (L), q = Total charge loading (C/L), J = Current Density (mA/cm²), A = electrode area (cm²), A_e = Active electrode area (cm²), Q = flow rate (L/s) in a continuous flow reactor.

2.1.2 Iron electrocoagulation

When Fe is used as electrode, Fe²⁺ ions get released into the solution due to oxidation, which then gets oxidized to insoluble Fe³⁺ ions by the dissolved oxygen present in the solution. The Fe³⁺ ions formed by electrolytic oxidation then combine with OH⁻ ions to form monomeric species such as Fe(OH)₃ and polymeric hydroxy complexes such as Fe(H₂O)₅(OH)²⁺, Fe(H₂O)₆³⁺, Fe₂(H₂O)₈(OH)₂⁴⁺, Fe(H₂O)₄(OH)₂⁺ and Fe₂(H₂O)₆(OH)₄⁴⁺ depending on pH (Mollah et al. 2004).

As presented in Eq.8, following equation 5, the Fe concentration (mg/L) can be calculated by Faraday's law of electrolysis:

$$(8) [Fe] = \frac{I * t * M}{n * F * V} * FE = \frac{I * M}{n * F * Q} * FE$$

With $n = 2$ for oxidation to Fe(II), $M = 55845$ mg/mole for Fe, I is the constant current applied for a time t (s) to a volume V (L) in a batch reactor or at a flow rate Q (L/s) in a continuous flow reactor. FE is the Faradaic efficiency (0 – 1) to correct for deviations from ideal Fe oxidation.

The comparison of experimental and theoretical iron concentration can be referred to as Faradic efficiency, which is defined as the ratio between the experimental concentration and theoretical calculation as shown in Eq. 9 (Khaled et al. 2019).

$$(9) \text{ Faradic efficiency (FE)} = C_{\text{experimental}} / C_{\text{theoretical}} \quad (\text{FE} = \text{Faradaic efficiency } (0 - 1))$$

where, $C_{\text{experimental}}$ = Dissolved iron concentration in the solution measured experimentally (mg/L)

$C_{\text{theoretical}}$ = Iron concentration calculated using Faraday's law (mg/L)

A Faradic efficiency greater than 1 could have resulted due to chemical dissolution of the electrodes (Khaled et al. 2019). Also, corrosion and oxidation of the electrode surface under the influence of chloride ions. The Fe(III) precipitates generated by EC can be removed by rapid sand filtration or gravitational settling.

2.2 Operational factors affecting EC

2.2.1 pH

The initial pH can influence the EC process. It mainly depends on the type of contaminants, anode material, and reaction mechanisms. Several studies showed that pH has a substantial impact on the EC process and treatment effectiveness. The type of EC-causing species and the solubility all depend on pH (Mouedhen et al. 2008).

2.2.2 Electrolysis time

The duration of EC treatment directly affects the EC process, i.e., the generation of coagulants. Increased electrolysis time leads to the increase of the generation of complex ions, and hydrogen bubbles (through electrodisolution of the anode, and reduction in the cathode). However, the removal efficiency only increases with time until an optimal value. The removal efficiency becomes constant outside the optimum time limit due to excess coagulant (Al-Qodah and Al-Shannag 2017). Although a slight increment in the duration could increase the removal efficiency in light of the price and process suitability, the optimum duration for the EC is usually considered 20–30 min, especially for iron EC (Lakshmanan et al. 2009).

2.2.3 Current Density (CD)

CD controls the rate at which coagulant is produced by EC, the evolution of gas, the formation of bubbles, the size of the floc, and the EC operating costs resulting in faster pollutant removal (Amrose et al. 2014). Increased current causes faster anode disintegration, results in increased precipitation, and more bubble production, which accounts for more flocculation and coagulation, resulting in a higher removal (Essadki et al. 2008). The number of flocs produced is a function of the amount of treatment time and the CD used. High CD increases the quantity and rate of floc generation that collects suspended solids and increases the amount of sludge generated. In addition, excess

metal hydroxide flocs are hard to float, resulting in operational shortfalls (Koby et al. 2016). In addition, using high CD, much of the energy is lost in the water electrolysis reaction and heating due to the joule effect. Therefore, it is crucial to operating the device at an optimum CD, which depends on the water matrix (Alam et al. 2021).

2.2.4 Mode of Electricity Application

The constant supply of electricity ensures the continuous supply of metal species, which are responsible for the charge neutralization and eventual precipitation of suspended pollutants and simultaneous adsorption of dissolved organic matter and other chemical species. However, intermittent electricity supply can reduce the consumption of energy and electrode material, since the metal dissolution stops between the gaps. As a consequence, intermittent electrical supply limits anode passivation, which is the phenomenon that occurs when the surface of an anode becomes chemically inactive or coated with a layer of oxide that prevents further oxidation or electrochemical reactions from occurring (Alam et al. 2021).

2.2.5 Electrode

The electrocoagulation process revolves around the vital role of electrodes. Consequently, parameters and configurations, including electrode materials, spacing/gap, shape, and arrangement might impact the performance, which are discussed below.

Material

Several materials such as Al, Fe, Zn, Ag, Na, Mg, Si, Ca, Sr, and Cd can be used as electrodes in EC (Sahu et al. 2014). In addition, using graphite and lead oxide as electrode materials for EC is also reported. These materials are different in terms of their chemical and physical properties. Al and Fe electrodes are the most frequently used materials for the electrode in electrocoagulation applications because they are inexpensive, non-toxic, readily available, and proven reliable to produce Fe^{+3} and Al^{+3} coagulants.

Electrode Spacing or gap distance

Electrode spacing is directly linked to the functioning of the EC, as it can alter electrostatic attraction, the in-between residence time, turbulence, mass transfer and finally, pollutant removal efficiency. For instance, the electrostatic force between metal hydroxide flocs would be very high at low spacing, resulting in small compact flocs; therefore, degradation of flocs will occur due to intensive collision (Daneshvar et al. 2004). On the other hand, at higher electrode spacing, the time for agglomeration of produced metal hydroxide is enough with fewer flocs degradation (larger, more loose flocs), and hence, the removal efficiency increased. However, if the spacing increased beyond the optimum range, the removal efficiency decreased due to the decreased floc formation and increased potential drop (Nasrullah et al. 2012). However, the impact on the electrode gap distance will depend on the water matrix as well.

Electrode Arrangement

Normally series/parallel monopolar or bipolar systems of electrodes are used for EC at industrial scale. In a complex EC system, three different electrode arrangements are reported (Mollah et al. 2004)

1. Monopolar parallel electrodes (MP-P): In the MP-P setup, both oppositely charged electrodes are joined to one another and farther to the outer circuit. As the currents divide in this arrangement, the potential difference becomes lower (Moussa et al. 2017) (Figure 2-2a).
2. Monopolar-series electrodes (MP-S): In the MP-S setup, each internal pair of sacrificial electrodes are joined to one another making an equal amount of current supply in each. However, the voltage is additive here (Figure 2-2 b).
3. Bipolar series electrodes (BP-S): In the BP-S arrangement, two of the outer monopolar electrodes are joined to the external circuit, and internal bipolar sacrificial electrodes are without a connection. Here, on the positive side, oxidation takes place, and the cathodic reaction takes place on the negative side (Demirci et al. 2015)(Figure 2-2 c).

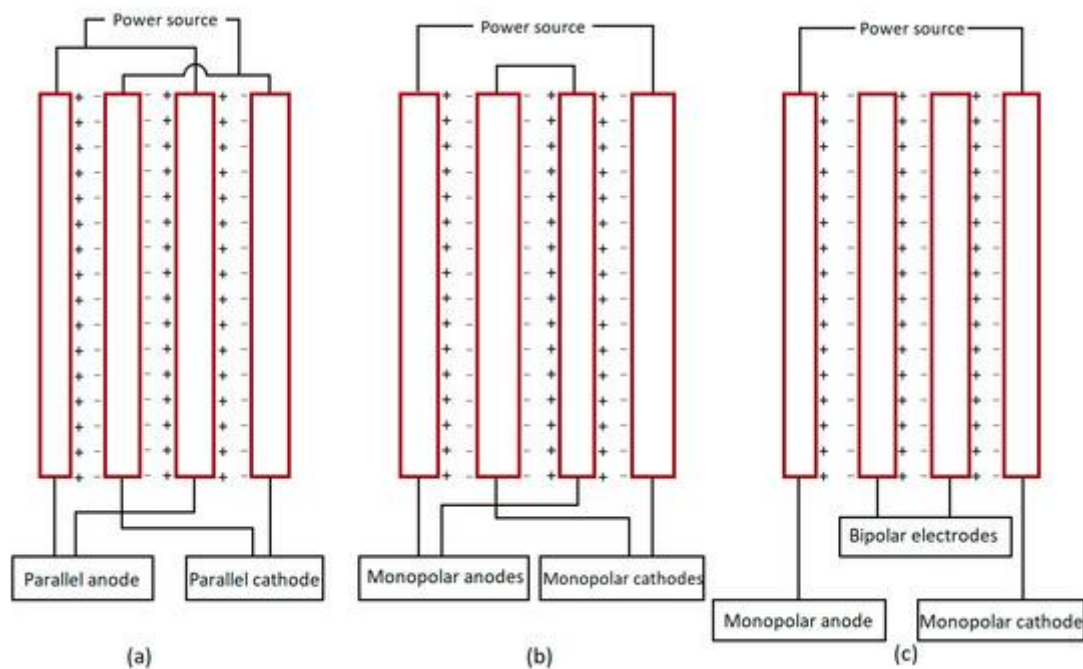


Figure 2-2 Different electrodes arrangements in an electrochemical cell: (a) MP-P; (b) MP-S; (c) BP-S.

Electrode Shape

Electrodes have been used in several shapes such as rectangular, circular, cylindrical, punched hole type in which rectangular shape is the most common because they can be arranged in parallel plates. Rectangular electrodes are also relatively easy to manufacture, clean, and maintain compared to other electrode shapes such as cylindrical or spherical. Their flat shape allows for easy access and cleaning of the electrode surface, which can reduce the likelihood of fouling or scaling, and their simple design makes them cost-effective and easy to replace if necessary. Generally, they are placed vertically. However, in some studies, horizontal placement of electrodes is also reported, which enhanced the mixing efficiency (Tchamango and Darchen 2018).

2.2.6 Mode of Power Supply

Supplies in the form of DC power are generally used for electro-coagulation cells. Several studies have been carried out using DC sources except for a very few studies where the AC source had been implemented (Al-Qodah and Al-Shannag 2017). However, the use of DC means that the anode is oxidized, and a layer of oxide is developed on the cathode called as cathode passivation. This passive layer results in the decrease in the current flow between both electrodes, and the efficiency of EC decreases. Passivity results in overpotential, which leads to a large amount of energy utilization. Several study results with supply in the form of AC were promising. (Mollah et al. 2004) noticed that AC supply ensures an adequate life of electrodes by cyclic energy that hinders DC's conventional consumption. The AC and DC supply reaction on cadmium removal from water through electrical coagulation has been examined by (Vasudevan and Lakshmi 2012). The results have shown that less energy and greater removal efficiency were achieved using an AC power supply.

2.3 Electrocoagulation in drinking water production

EC has been shown to effectively remove a number of common drinking water contaminants such as arsenic (Roy et al. 2021), fluoride (Mameri et al. 2001, Zhu et al. 2007), clay (Holt et al. 2002), cobalt (Mansour et al. 2012) and virus (Zhu et al. 2005). In this section, insight from previous research about removal efficiency is presented:

2.3.1 Iron electrocoagulation

McBeath et al. (2020) evaluated recently the efficiency of electrocoagulation at a pilot-scale as an alternative drinking water treatment technology to conventional coagulation. Using iron anodes (A1008 cold-rolled steel iron), the influence of different parameters such as dissolved Fe (concentration), current density and inter-electrode gap on the reduction of natural organic matter (NOM) was studied using lake raw water. The four dissolved iron concentrations investigated were 27.8, 38.2, 51.1 and 60.8 mg/L, achieved by applying constant current at 16.0, 22.0, 29.4 and 35.0 A, respectively. Two inter-electrode gaps were investigated (1 and 2 mm). When operating in 2 and 4-cell configurations, the electro-active reactor volumes were 93.3 and 217.7 cm³ for 1 mm inter-electrode gap and 186.6 and 435.4 cm³ for 2 mm inter-electrode gap, respectively. At a 10 L/min flow rate in 2 and 4-cell configuration, the respective reactor residence times in the electro-active volumes were 0.56 and 1.31 s for the 1 mm inter-electrode gap and 1.12 and 2.61 s for the 2 mm inter-electrode. Cathode electrodes consisted of cubic crystal stainless steel alloy. Both electrodes were 16-gauge thickness (1.519 mm) and had a single side active surface area of 310.8 cm². A flow rate of 10 L/min was fixed.

The results showed a clear effect of Fe dosage on the removal of DOC and UV254. DOC decreased by 29.2±5.6%, 35.1±4.2%, 37.1±3.4% and 37.2±4.2%, while UV254 was decreased by 38.7±3.0%, 48.8±2.5%, 52.5±1.2% and 54.7±0.9% for Fe dosage of 27.8, 38.2, 51.1 and 60.8 mg/L, respectively, for all current densities and inter-electrode gaps tested. A greater reduction in UV254 was yielded compared to dissolved organic carbon, indicating better removal of larger molecular weight fractions of NOM. As Fe dosages increased from 27.8 to 60.8 mg/L, specific ultraviolet absorbance decreased from 1.92±0.14 to 1.60±0.10 L/m•mg respectively, from an initial raw water value of 2.21 L/m•mg.

No significant differences were observed for the effect of current density and inter-electrode gap in NOM removal, however Fe dosage was the primary variable dictating the process' effectiveness. Although large variations in energy requirements were yielded, depending on the operating potential (dictated by the current and resistance of the electrochemical cell), no large variations were observed for the corresponding NOM removal (DOC and UV254 reduction differences were observed to be negligible). In general, conditions that yielded the highest removal of NOM, a 1 mm gap and 4-cell configuration, had energy requirements between 0.480 and 0.602 kWh/m³ of water treated. These results indicate that similar NOM removal performances can be achieved at both energy intensive and energy non-intensive conditions.

Lower energy requirements (0.11 kWh/m³) for an iron EC process for NOM removal has been reported at a small-scale batch but no fair comparison to full scale system can be made (Alimohammadi et al. 2017). Based on (McBeath et al. 2020) results, a set of preferred operating parameters could therefore minimize the energy requirements without compromising the NOM removal efficiency of the process. Also to minimize energy, charge dosage and charge dosage rate are selected as the main process-control parameters for the iron dosage, directly linked to the electrolysis time and current intensity, respectively (Amrose et al. 2013, Ghernaout et al. 2019, Van Genuchten and Peña 2017). Charge dosage is defined as the total electric charge per unit volume applied to a given water sample, while charge dosage rate is defined as the speed of application of electric charge (Charge Dosage per unit time). In this way charge dosage [C/L] represents the total dose of current, while charge dosage rate [C/L/min] represents the current dosing speed.

2.3.2 NOM removal

The presence of high concentrations of natural organic matter (NOM) and total organic carbon (TOC) in e.g., groundwater can impact the speciation of aqueous Fe(II) by complexation (Sundman 2014). The type and concentration of NOM also influence the effectiveness of the treatment. Successful removal of NOM using EC has also been demonstrated at lab scale (Dubrawski et al. 2013, Dubrawski and Mohseni 2013b, Jiang et al. 2002). Also, (McBeath et al. 2020) showed that iron EC is effective at removing organic matter.

There are several factors that can influence the selectivity of an EC process to predominantly produce one species in greater quantities, including anode potential, pH, temperature, dissolved oxygen, $[\text{Fe(II)}]:[\text{Fe(III)}]$ and the concentration and ratios of ions comprising the water matrix. It has also been shown that different species can have varying affinities to function as effective coagulants for the removal of organic matter (Dubrawski and Mohseni 2013a).

Dubrawski and Mohseni (2013b) examined three EC design parameters for NOM removal in a bench-scale batch reactor: current density (i) ($2.43\text{--}26.8\text{ mA cm}^{-2}$), charge loading rate (CLR) ($100\text{--}1000\text{ C L}^{-1}\text{ min}^{-1}$), and flocculation methodology ("fast" and "slow"). The results showed increased NOM removal at lower current density due to a higher current efficiency (see Eq. 9), and increased NOM removal at lower CLR, due to less pollutant-coagulant contact time and greater pH increase. Changes in the current density were obtained by changing the electrode surface area, while maintaining the current constant. At $i = 26.8\text{ mA cm}^{-2}$, DOC and UV-abs-254 reductions were 67% and 66%, respectively, while i in the range of $2.43\text{--}13.4\text{ mA cm}^{-2}$ had average removals of 73% and 88% for DOC and UV-abs-254, respectively. An optimum operating current density of 10 mA/cm^2 was suggested, while i of 6.7 mA cm^{-2} and less show diminishing NOM removal for greater electrode areas. Additionally, the greatest NOM reductions were found to occur at lower charge loading rates (Dubrawski and Mohseni 2013b). A CLR of $1000\text{ C L}^{-1}\text{ min}^{-1}$ led to DOC and UV-abs-254 reductions of 61.8% and 40.1%, respectively, while reductions were an average of 67.3% and 85.1%, respectively, for CLR values from 200 to $500\text{ C L}^{-1}\text{ min}^{-1}$. It must be pointed out these findings are specific to the water matrix used, the type of NOM and its concentration.

Research involving the effect of three NOM sources (Suwannee River (USA) DOC = 13.79 mg/L , Nordic Reservoir (Norway) DOC = 9.03 mg/L , and a natural source Lost Lagoon (Canada), DOC = 13.31 mg/L) was carried out by Dubrawski et al. (2013). EC was found to predominantly remove NOM with higher concentrations of large hydrophobic and high molecular weight fractions. Moreover, relative reduction in DOC with two different initial NOM concentrations (13.79 and 21.59 mg/L) was not significantly different, performing equally for either treatment at the same EC operating conditions.

Other relevant research investigated the iron hydr(oxide) speciation phenomena during an EC process applied to natural waters, where species were identified *in-situ* using Raman spectrophotometric analysis. Three predominating species were identified to be green rust (GR) ($\text{FeIIIxFeIIy(OH)}_{3x+2y-z}(\text{A}^-)_z$; $\text{A}^- = \text{Cl}^-; 1/2\text{SO}_2^{-4}$ $\text{FeIIIxFeIIy(OH)}_{3x+2y-z}(\text{A}^-)_z$; $\text{A}^- = \text{Cl}^-; 1/2\text{SO}_4^{-2}$), lepidocrocite ($\gamma\text{-FeOOH}$) and magnetite (Fe_3O_4). GR speciation occurred at low current density and intermediate charge loading rate conditions, while magnetite was found to predominate during processes at high current density and charge loading rate (CLR). All other conditions yielded the formation of lepidocrocite. Further analysis compared the varying effectiveness of the three species functionality as a coagulant chemical. It was suggested that GR was the preferred iron species due to its ability to most effectively reduce NOM concentrations (Dubrawski and Mohseni 2013a).

Aluminum EC research has been focused on the effect of current density and metal loading (dose of coagulant) on the removal of aquatic humus, and its comparative performance to conventional alum coagulation (Vik et al. 1984). Researchers found no significant difference in removal performance between EC and chemical coagulation, however higher residual aluminum concentrations after the EC process were present, which was attributed to the high pH of the water. Other researchers have found that aluminum EC outperformed aluminum sulphate coagulant; achieving 20% greater reductions in dissolved organic carbon (DOC) while using synthetic water samples. When natural surface waters were tested, similar contaminant removal was achieved with both EC and chemical coagulation (Jiang et al. 2002).

NOM of different origins (commercial, terrestrial and natural water) were treated by electrocoagulation (EC) process using aluminum, iron and hybrid electrodes (Al+Fe) (Ulu et al. 2015). The lowest effluent concentration of DOC was obtained as 5.05 mg L^{-1} with hybrid electrode for natural NOM source at its original pH 7.3. In addition, among the metal types, the best UV-abs-254 removal efficiency was obtained as 92.4% with 0.0312 cm^{-1} by hybrid

electrode. The color removal efficiency of water occurred successfully by Al and hybrid electrodes. Aquatic NOM source was the most resistant to EC treatment with DOC reduction of 71.1%, 59.8%, and 68.6% for Al, Fe and hybrid electrodes, respectively. SUVA value was reduced to below 2 for three NOM sources studied. In another similar study, Ulu et al. (2014) found that the highest treatment efficiency for humic acids achieved 87% ($\text{DOC}_{\text{treated}}$ 2.1 mg/L) at an initial pH of 4 with iron electrodes.

Although EC has been previously researched for drinking water applications, much of the research has been bench-scale, batch (Ben-Sasson et al. 2013, Mahvi et al. 2011, Vepsäläinen et al. 2009) operations using synthetic water with humic acid and/or reverse-osmosis isolated NOM. A very limited number of studies incorporating continuous flow reactors exist and although successful, these remain at small bench-scale operations with flow rates below a practical capacity for piloting (McBeath et al. 2020).

2.3.3 Nitrate removal

EC has been applied widely to remove nitrate from different types of waters including drinking water (Hashim et al. 2017, Kumar and Goel 2010, Lacasa et al. 2011b). The pH of the solution often increases in EC unlike conventional coagulation (CC) where the pH decreases. Therefore, there is no need for post-neutralization in most cases with EC. The mechanism for EC is simple and no moving parts are required. However, the electrodes need to be replaced from time to time and electricity consumption most likely is greater than in CC in this case.

Hashim et al. (2017) results obtained indicated that the EC reactor reduced the nitrate concentration from 100 to 15 mg/L after 55 min of electrolysis at initial pH of 7, aluminum electrodes, a gap between electrodes of 5 mm, CD of 2 mA/cm², a flow rate of 250 ml/min, and at operating cost of 0.455 US \$/m³.

Lacasa et al. (2011b) showed that nitrate adsorption onto growing metal hydroxide precipitates appears to be the primary mechanism behind the observed nitrate removal efficiency. The same amount of nitrate removal was obtained using both iron and aluminum electrodes. Kumar and Goel (2010) achieved a removal efficiency of 84% for nitrate at 25V using tap water or distilled water after 3 hours of electrolysis, with an initial nitrate concentration of 300 mg/L. When applied potential was 10 V, 50.3% nitrate removal was achieved in 6 h. To avoid reduction in electrocoagulation efficiency, they instructed to regularly change the polarity of electrodes.

Hossini and Rezaee (2014) investigated the removal of nitrate from an aqueous solution by the process of electrocoagulation, using aluminum/graphite as the anode/cathode electrodes. The results showed that by applying electric current of 0.14 A for 120 minutes, the nitrate content would reduce down to 97%. More nitrate reduction is obtained with higher conductivities (e.g NaCl).

Apshankar and Goel (2020) determined operating conditions for maximizing nitrate removal using electrocoagulation, settling and filtration with four different types of waters: double distilled (DD) water, ground water (GW), tap water, and untreated water from the local water works plant. The best nitrate removal efficiencies were obtained at high initial concentrations of nitrate, high EC and settling times and high pH. The best removal efficiency of 38.2% was observed with double distilled (DD) water after EC and settling, since there were no competing ions to interfere with the removal of nitrate from the raw water. EC and settling time were 4 h each. Initial pH was adjusted to ≈ 10 . The voltage applied was low (10 V), with an energy consumption of 8.3 Wh/L. The average conductivity was around 1000 $\mu\text{S}/\text{cm}$ because of the high initial nitrate concentration (104 mg/L).

Karamati Niaragh et al. (2017) investigated the effect of the main EC process and water quality parameters on the performance of a continuous electro-coagulation (EC) process for nitrate removal efficiency and its operating costs. Aluminum plate electrodes, with a total effective area of 364 cm², were connected in monopolar parallel mode. The optimum conditions of initial nitrate concentration, inlet flow rate, current density and initial pH were found to be 100 mg/L, 50 mL/min, 80 A/m² and 8, respectively. As a result, the observed nitrate removal efficiency under these conditions was 61.7%. The operating costs were evaluated as 1.28 US\$/g NO₃-removed.

Ano et al. (2020) showed that high current intensity, high electrolysis time, low initial pH as well as the use of aluminum electrodes, are favorable to high nitrate removal. The optimal conditions obtained are the following: intensity 1.80 A, time 33.0 min and pH 8.73. Under these conditions, 73.8 % of nitrate was removed against 12.02 mg/L of ammonium and 1.8 g of metallic residues generated for an operating cost of 1.3 \$US/m³.

Moradi and Ashrafizadeh (2020) evaluated the removal of nitrate from tapwater by electrocoagulation-flotation (ECF) process. An ECF reactor with a working volume of 8 L was used. The effect of different arrangements of anode/cathode electrodes (Al/Fe, Fe/Al, and SS/Fe) were assessed together with the initial pH of the aqueous solution, process duration (ET), distance of electrodes, electric potential, and the initial concentration of nitrate. The results showed that the percent removal of nitrate increases with the initial concentration of nitrate, ET, the initial pH of the solution, and voltage. In addition, based on the results obtained, ET and ED had the greatest effects on the nitrate removal by the electrocoagulation flotation process. As such, to achieve the maximum removal of nitrate, decreasing the distance of electrodes can be more economic than increasing other variables such as voltage and ET. Finally, the results of this research showed that at optimum conditions (initial concentration of nitrate = 120 mg/L; solution pH = 8; voltage = 17 V; ED = 3 cm; ET = 240 minutes), the removal of nitrate by the Al/Fe electrodes arrangement, 93% removal was achieved.

EC has been used to remove nitrate from different types of waters, including drinking water. Unlike conventional coagulation, the pH of the solution often increases in EC, meaning that post-neutralization is not needed. The electrodes, however, need replacement, and electricity consumption is typically greater than in conventional coagulation. Studies have shown that nitrate removal efficiency increases with high initial concentrations of nitrate, high settling times and high pH, and that higher conductivities yield greater nitrate reduction.

2.3.4 Phosphate removal

Hashim et al. (2019) investigated phosphate removal from river water by using aluminum EC. The results showed that 99% of phosphate was removed within 60 min of electrolysis at an initial pH of 6, inter-electrode distance of 0.5 cm, current density of 6 mA/cm², initial concentration of phosphate of 100 mg/L, and minimum operating cost of 0.503 US \$/m³. The electrochemical cell produced enough H₂ gas to generate 4.34 kWh/m³ of power.

Lacasa et al. (2011a) showed that pH increases with current density, indicating adsorption onto metal hydroxide. In the case of aluminum electrodes, the coexistence of both direct precipitation and adsorption onto metal is observed, whereas in the case of iron electrodes, the adsorption mechanism is less effective. Due to the higher solubility of the iron phosphate, iron hydroxide was the main product of coagulation when iron electrodes were used. The optimum conditions to remove phosphates by electrocoagulation are at the lower current densities (<3.0 mA cm⁻²) for iron and aluminum electrodes because it is favored the direct metal phosphate precipitation.

Bektaş et al. (2004) under low phosphate concentrations (10–50 mg l⁻¹) and EC duration equals to 5 min, the removal degree is 81–90%. The increase of the EC duration gives better results of the removal parameter. Thus, the 10 min EC process duration demonstrates the 70 and 94% phosphate removal for 200 and 100 mg l⁻¹ solutions, respectively.

Attour et al. (2016) indicated that the kinetics of the phosphate removal was found to be accelerated by continuous adjustment of the pH. The total removal of phosphate was obtained after 140 minutes without pH adjustment and after only 40 minutes with adjustment of pH every 10 min. About 50% was reached after 20 min of electrolysis with an initial phosphate concentration of 100 mg/L and a current of 10 mA/cm². The choice of the aluminum electrode connection modes was found to strongly affect the efficiency of the treatment, the Faradic yield and the consumption of energy. The mono-polar in parallels connection mode was found to be the most economic whereas bi-polar in parallels was found to favor a higher kinetic rate of treatment.

Franco et al. (2017) examined the effects of treatment parameters (initial pH, initial conductivity, power input, and initial P concentration) on the ability of the EC process to remove P in solutions with initial P concentrations less than 2 mg/L. The total removal of phosphate was obtained after 140 minutes without pH adjustment and after only 40 minutes with adjustment of pH every 10 min. Higher conductivity was demonstrated to have a higher P removal efficiency. 81% removal of P was achieved after 10 min of reaction time.

Vasudevan et al. (2008) used EC for the removal of phosphate from drinking water using mild steel as the anode and stainless steel as the cathode. The results showed that the maximum removal efficiency of 98% was achieved at a current density of $0.50 \text{ mA}\cdot\text{cm}^{-2}$ at a pH of 6.5.

In summary, various studies have shown that EC is an effective method for removing phosphate from water. The optimal conditions for phosphate removal are generally lower current densities and longer durations of EC. The type of electrode used, as well as the connection modes, can affect the efficiency of the treatment. Additionally, adjusting the pH during the EC process can accelerate the removal kinetics.

2.3.5 Removal of bacteria/viruses

Previous studies have demonstrated the effect of Fe-EC in the removal of a wide range of microorganisms from bacteria to viruses in different water matrices, mainly for drinking water applications (Delaire 2016, Ghernaout et al. 2019, Heffron et al. 2019a, Heffron et al. 2019b, Ryan et al. 2021).

Bacteria removal by Fe-EC was investigated by (Delaire 2016). The results show that removal is primarily due to bacteria encapsulation in Fe(III) flocs and removal by gravitational settling, while inactivation by germicidal reactive oxidants remains limited in the presence of HCO_3^- and at $\text{pH} > 7$. Fe(III) precipitates are found to adhere to the surface of bacterial cells, primarily through interactions with bacterial phosphate groups, resulting in bacteria enmeshment in precipitate flocs.

The effect of major groundwater ions is interpreted in light of this mechanism: the presence of Ca and Mg reduce bacterial removal by competing with complexing bacterial phosphate groups; Si and NOM, which do not strongly compete with phosphate groups for sorption to Fe(III) precipitates, do not affect attenuation; by contrast, P decreases attenuation significantly, except in the presence of bivalent cations, which can bridge between P sorbed to precipitates and bacterial phosphate groups. Finally, Fe-EC is shown to be equally effective towards Gram-positive and Gram-negative bacteria, smooth and rough alike, likely due to the universal presence of phosphate moieties on bacterial cell walls. Altogether, results show that Fe-EC can effectively remove all types of bacterial contamination from a range of groundwaters.

Ghernaout et al. (2019) reviewed the mechanisms for disinfection by applying EC. EC alone is effective for bacteria and algae removal to a certain extent (Barışçı and Turkay 2016, Ghernaout et al. 2008). (Tanneru and Chellam 2012) established that for surface water carrying 5 mg-C/L NOM, EC/MF only eliminated 1.5 log of MS2 viruses at 13 mg Fe/L, whereas 6.5 log elimination was evaluated from synthetic water at an identical Fe injection. The existence of 5 mg-C/L of Suwannee River-humic acid reduced virus elimination by roughly 4-log at pH 6.4 and approximately 2-log at pH 7.5 by EC/MF compared to FeSO_4 coagulation/MF (Chellam and Sari 2016). Therefore, it was suggested that NOM exacerbated virus elimination through complexing anode dissolved Fe(II), hindering its oxidation to insoluble Fe(III). The amount and constitution of NOM seem to have made crucial contributions to influencing microorganism elimination/deactivation in surface and groundwaters.

Al EC can perform better than Fe in eliminating viruses from NOM-containing surface waters because of effective $\text{Al}(\text{OH})_3(\text{s})$ precipitation and enmeshment while Fe did not significantly precipitate following electrolysis upon these situations (Tanneru et al. 2013). Aluminum electrodes can produce more reactive species, such as hydroxyl radicals, the aluminum hydroxide flocs have a higher positive charge than iron hydroxide flocs, which can improve the

coagulation of negatively charged viruses. However, prolonged flocculation/contact times would be required to attain important deactivation through Fe/Al EC of surface water (Chellam and Sari 2016).

Boudjema et al. (2014) studied the likely treatment of Oued El Harrach river water by Al EC. A 99% reduction in fecal coliforms and *E. coli* was obtained and a near-total inactivation of fungi was reached.

On the other hand, (Llanos et al. 2017) compared the efficiency of EC in two steps of the urban water cycle: drinking water production and wastewater regeneration. They found that Al EC is an effective method to eliminate turbidity and TOC from surface water and to deplete turbidity and *E. coli* from urban treated wastewater.

Fe EC has been shown to effectively remove a wide range of microorganisms from bacteria to viruses in different water matrices. The removal of bacteria by Fe-EC is primarily due to bacteria encapsulation in Fe(III) flocs and removal by gravitational settling. Al EC can perform better than Fe in eliminating viruses from NOM-containing surface waters due to effective aluminum hydroxides precipitation and enmeshment.

2.4 Cost implications in drinking water production

The appeal of EC as a potential water treatment technology is due to the elimination of the chemical supply chain required for conventional chemical coagulation, it's easier operation and maintenance, as well as reported low operating costs (Gu et al. 2009). Most of the EC research has been focused around two anode metal types: aluminum (Al) and iron (Fe). Although both metal types have been shown to be effective for the removal of a number of contaminants, some research has suggested that Fe provides the lowest residual metal concentration post-EC and filtration (Dubrawski et al. 2013).

Apart from the costs of electricity, the expenses of chemicals and sacrificial electrodes are the main components used in calculating the cost of an electrochemical device. Thus, energy usage, complete dissolution of the electrode, any cost add up of chemical substances (adjustment of the conductivity or altering solution pH), and cost of disposal and transportation of sludge are all part of the EC process costs. However, the use of a renewable energy source and process optimization could reduce the overall cost.

Bicudo et al. (2021) estimated cost based on a simplified operational cost estimate approach for each individual Fe-EC experiments performed. This study considered the consumed electric power and metallic iron as input parameters. As for 2019 average energy cost in The Netherlands for a medium size consumer = 0.0679€/kWh (Eurostat, 2019), and the steel S235 cost = 0.21€/kg (MEPS International Ltd, 2019). For a given Fe-EC experiment, in which U is the applied voltage (v), I is the current intensity (mA), t is the treatment time (h) and V is the volume of the cell (m^3), then the consumed power can be estimated as:

$$P[\text{kW}\cdot\text{hm}^3]=U\cdot I\cdot t/V$$

Then, the operational cost for each particular experiment is determined by the amount of consumed power and the amount of dosed iron (described in SI-Faraday's equation), multiplied by their respective unit costs:

$$\text{Cost } (\text{€}/\text{m}^3) = P [\text{kW}\cdot\text{hm}^3] \times 0.0679 \text{ €}/\text{kWh} + m_{\text{Fe}} (\text{kg}) \times 0.21 \text{ €}/\text{kg}$$

$$C_{\text{energy}} = Uq$$

$$C_{\text{energy}} = Uit/V$$

where C_{energy} = Consumption of electricity per m^3 of water treated (Wh/m^3); U = Potential of the cell/EC reactor (V);
 q = Charge dosage (C/L), i = current applied (mA), t = electrolysis time (sec)

Karamati Niaragh et al. (2017) made an economic evaluation in which electrical energy consumption, electrode material and chemical costs were calculated as the major operating costs using Equation ():

$$\text{OPEX} = aC_{\text{energy}} + bC_{\text{electrodes}} + cC_{\text{chemicals}}$$

where C_{energy} (kWh/ g target-removed), $C_{\text{electrodes}}$ (kg M/ g target-removed) and $C_{\text{chemicals}}$ (g chemicals/ g target-removed) are quantities consumed in the experiments on target (e.g., NO_3 , PO_4) removal. The a , b and c are the coefficients of the market, as follows:

- Coefficient a : industrial electrical energy price [$\text{US}\$/\text{kWh}^{-1}$].
- Coefficient b : wholesale electrode material price [$\text{US}\$/\text{kg}$]
- Coefficient c : industrial major chemicals added [$\text{US}\$/\text{kg}$ chemical added]

To obtain the mass depletion of the electrodes, the amount of the dissolved electrode theoretically relies on current intensity and reaction time, which is calculated according to Faraday's law.

EC is a potential water treatment technology that eliminates the need for conventional chemical coagulation, making it easier to operate and maintain with most likely lower operating costs.

2.5 Electrocoagulation sludge valorisation

Rajaniemi et al. (2021) reviewed the potential applications from EC sludge, mainly from wastewater. As an adsorbent, EC sludge has shown suitability for the removal of some impurities. EC sludge displays some characteristics similar to those of metallic oxides/hydroxides, which are well-known adsorbent materials.

Iron oxides are among the most commonly used pigments. Sludge produced from EC treatment with Fe electrodes contains iron oxides and thus may be used as a pigment. Meanwhile, Al is an important component of concrete materials. Sludge produced from EC treatment with Al electrodes contains a high Al content and therefore offers a potential application in construction materials. However, few studies have investigated the use of EC sludge in construction materials. These studies have found that EC sludge could constitute the mass of building blocks to a certain extent. The use of EC sludge in many construction areas remains unexplored, such as in ready-mix concrete, environmental concrete products (e.g., draining channel, curbstone, and parking element), plastering, and possibly in a new mineral wool product for insulation.

3 Materials and Methods

This chapter described the materials and methods used for both the experimental work with EC and three different water types and the desk study for cost calculations and scalability of the system.

3.1 Iron Electrocoagulation

EC experiments were carried out with a laboratory-scale electrocoagulation setup (design by QStone Capital, made by KWR's workshop). Figure 3-1 shows a drawing and photo of the EC laboratory set-up (drawing made by QStone Capital, the Perspex pilot unit produced by Industrial Plastics Europe in Nieuwegein, eventually compiled by KWR). The laboratory setup was previously used in a TKI study in which different types of water were tested with EC (Siegers and Jong 2021). The experiments were carried out at KWR's test hall. For each experiment, approximately 500 L of water was taken from drinking water locations of PWN (Andijk), Vitens (Sint Jans klooster) and WML (De Beitel), respectively, and stored at room temperature (18°C) in a stainless steel tank that was connected to the laboratory setup.

The EC laboratory setup is originally made of three compartments: the first compartment contains the electrodes and air intake (turbulence for mixing when using heavily polluted water), the second compartment was used for flocculation and the third compartment for the settling of the flocs. A 4th compartment has been added for the present research to improve the sedimentation process. In addition, agitators were added to compartments 2 and 3 to promote flocculation, the stirring speeds were set at 60 and 30 rpm respectively. The volume of the compartments (up to the outlet) are 2.08, 4.35, 4.55 and 20 L respectively. The flow rate through the pilot was approximately 20 ± 2 L/h where the contact time in these compartments was 6.2; 13.1; 13,7 and 20 min (± 10 %), correspondingly, and a total of about 25 hours per experiment.

For the pumping of the water from the stainless steel tank, a magnetic impeller pump type Iwaki MD-30AZ was used, which was set to the desired flow rate by a frequency converter (manufactured KWR) and a needle valve.

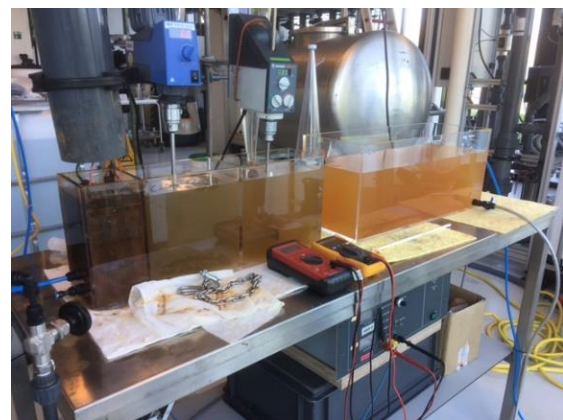
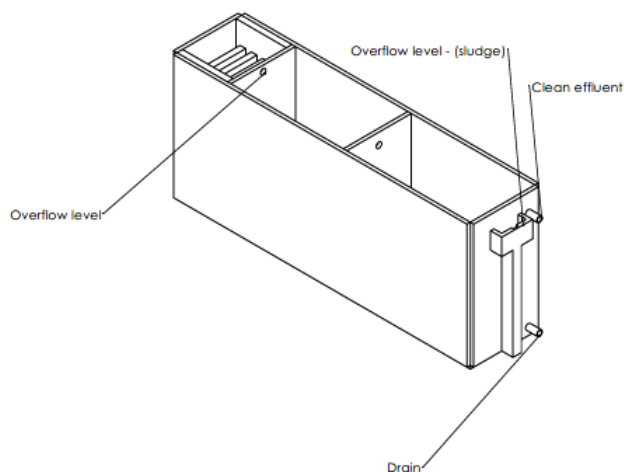


Figure 3-1 Drawing of the original pilot and photo of the EC laboratory setup.

For the electrodes, for both the anode and the cathode, low-quality unalloyed construction steel was used (10 mm thick steel 37, purchased from Van Hengstum Metaal in Soest, costs € 2.88 per electrode). The composition of the steel was Fe maximum 0.17% C, 1.4% Mn, 0.045% P, 0.045% S and 0.009% N (see Appendix III). The electrodes were connected to a power source (Alpha+elektronika, type AL854) with which a certain voltage (V) and current (I) could be transferred over the cathode and anode electrode when they were submerged in the water in compartment 1.

Dosage of iron is a function of current (A) and voltage (V), and the voltage is applied between the electrodes using a monopolar configuration. The iron dosed by dissolving electrodes is proportional to the total charge passed between electrodes. In the electrode compartment (see Figure 3-2) 2 distances are applied: 5 and 10 mm. Both the distance, the applied potential and the type of water determine the amount of iron that will dissolve.

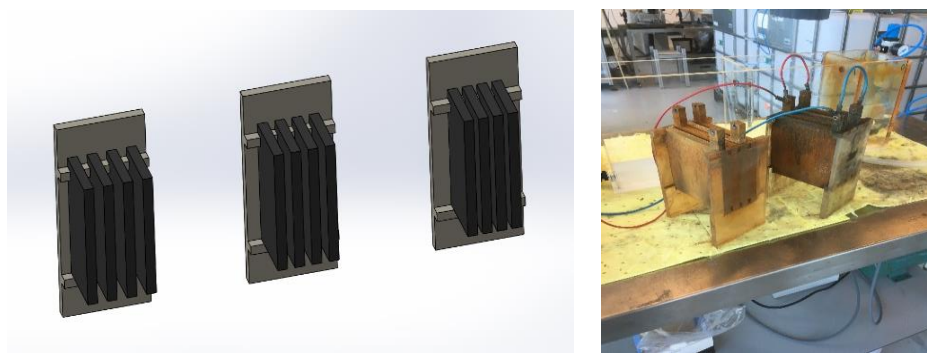


Figure 3-2 Drawing and photographing of the (used) electrodes with different distance/gap between the electrodes (0.5 and 1 cm)

The oxidation from Fe(II) to Fe(III) takes place via the oxygen in the water, theoretically this conversion of oxygen can be calculated and depicted in Table 4, Appendix I. Based on this calculation, 0.72 mg of oxygen is consumed per mg Fe(II) for complete oxidation to Fe(III).

3.2 Electrochemical conversion nitrate with graphite electrodes

Because during the experiments with WML water during the removal of nitrate, an undesirable amount of iron also entered the water, an additional experiment was carried out. Using graphite electrodes, an attempt was made to electrochemically convert the nitrate to gaseous nitrogen, assuming that no materials from the electrodes would dissolve. To this end, graphite electrodes (bulk density > 1.8 g/cm³, 10 by 10 cm, 4 pieces, Fruugo.nl) were purchased and placed in a 3 L beaker with 1 cm electrode distance (see Figure 3-3). Direct current has been applied to the electrodes with crocodile clamps. At 3 different applied voltage, the course of the nitrate concentration in the water has been followed in time.

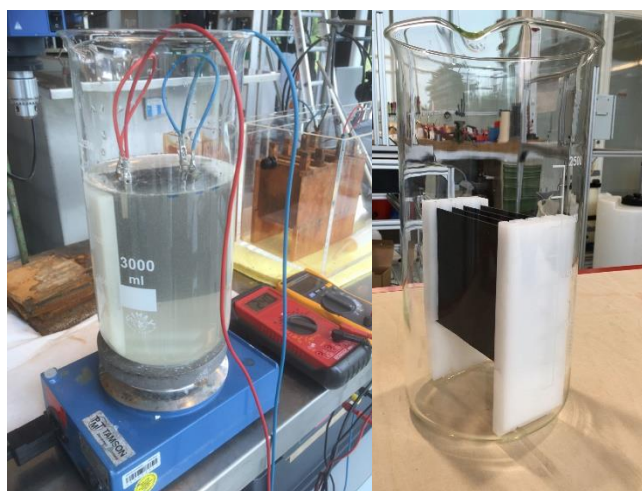


Figure 3-3 Photos of the graphite electrodes placed in a 3 L beaker during and for an experiment with WML water

3.3 Data of the water types examined

The water types studied are (1) surface basin water (PWN) for research into the removal of phosphate and suspended particles, (2) backwash water from a rapid filter (Vitens) to investigate whether the current iron chemical dosage could be replaced by electrocoagulation and (3) anaerobic groundwater (WML), in particular for research into nitrate removal without the addition of chemicals.

3.3.1 PWN surface water

PWN makes drinking water in Andijk from water from the IJsselmeer. From the IJsselmeer, water is led into reservoirs, after which it is pumped to the treatment after a certain residence time (on average about 21 days). During the stay in the reservoir, all kinds of chemical and microbiological processes take place that lead to both quality improvement (in particular sedimentation of solid material) and to quality deterioration (growth and death of algae and bacteria). The water contains (organically bound) phosphate and suspended solids which are removed after the inlet during the water treatment by a conventional coagulation – sedimentation process. The water used for the experiments was sampled at the inlet to the treatment plant.

3.3.2 Vitens sand Filtration backwash water

At Sint Jansklooster, groundwater is used to make drinking water. After aeration, the water is filtered with rapid sand filtration. When backwashing these sand filters, water with a high iron content is released. This backwash water required a 45 mg Fe/l as ferric chloride dosed for rapid settling of the iron and allows reuse part of the backwashing water. There is a lamella settler used to drain the sediment. The water used for the experiments was sampled from a flushing pipe.

3.3.3 WML anaerobic groundwater

WML uses groundwater for the production of drinking water at OPB De Beitel. The raw water of the Roodborn extraction field was sampled at well PP118 (near Eys). The aerated and anaerobic water from this well contains about 44 mg/l NO_3^- and no iron. The high nitrate content is undesirable for drinking water and difficult to remove. In addition to nitrate, the water also contains sulphate and can be called relatively hard.

3.4 Analysis

The following analytical methods have been used for the determination of key parameters of the raw and treated water after EC followed by sedimentation time. The methods are used depending on the type of water tested. The methods were applied directly in the pilot installation and by the KWR laboratory.

- Fe was measured with Hannah test kit¹, type HI93721-01 reagent and HI83399 spectrophotometer, verified on a number of samples with an ICP-MS method (KWR);
- Nitrate, ortho- and total PO₄ were analyzed with Hach Lange test kit (type LCK 340 and LCK 349 respectively);
- pH and electrical conductivity (EGV) were measured with Radiometer and Hach Lange sensors;
- Suspended solids were determined via the method KWR LAM-046;
- Turbidity was measured with Hach Lange type 2100Qis
- Sedimentation of flocs were measured using an Imhoff settling cone (see Figure 3-4) for a period of 72 hours.



Figure 3-4 Photo of an Imhoff flask, empty and filled within the experiments.

3.5 Cost and scalability calculations

With an Excel calculation sheet from QStone Capital, data required for the evaluation have been calculated. Figure 3-5 shows an example of a printout of QStone Capital's calculation sheet.

Input from the experiments are: the current [A], conductivity [mS/cm], flow rate of the pilot [l/h] and measured iron dose [mg/l Fe]. Input for practical applications are: flow rate [m³/h], pH [-], TSS [mg/l], temperature [°C], electrode gap distance [m], dry matter content [%] and iron cost [€/kg]. Key technology parameters such as the coagulant dose [kg/h], the current [A], the voltage [V], the energy consumption [kWh/m³] and the sludge production [kg/m³] are calculated.

¹ Note: the Hach-Lange test kit LCK 320 that was initially used with the PWN water turned out not to be suitable/reliable for the measurement of Fe in water

EC Coagulator Calculator			V7_20201127		
PROJECT: VITENS			2-mrt-23		
<i>Basic Data Wastewater</i>					
Company information			Lab Results (After Optimisation)		
Avg flowrate Wastewater	26 m ³ /hr	Type of test	Pilot Scale		
Avg Conductivity	0,55 mS/cm	Metal	Fe		
Avg pH	7,3	Current	0,72 A		
TSS	540 mg.l	AVG Conductivity	0,528 mS		
Temperature	12 °C	Flowrate Pilot Optimisation	0,02 m ³ /hr		
Production Schedule	7 Days/Week	Coagulant Dosage test scale	43 mg/l		
Operational Days	365 Days/Year				
Summary					
Test Results		EC Reactor Overview		Sludge Production	
Coagulant Dosage test scale	43 mg/l			Total Fe2+ Production	0,65 kg/hr
Coagulant Production Lab / Pilot Scale	0,043 kg/m ³			Total Ironsludge (+ Hydroxide)	1,21 kg/hr
Coagulant Production Full Scale	1,12 kg/hr			SS retention	0,54 kg/m ³
Total Current	936 A	m between plates	0,005 m	Dry Matter %	50%
Total Voltage (for reaction)	3,23 V			Total Sludge Production	1,17 kg/m ³
Energy per m ³ waste	0,12 kwh/m ³			Total Dry Matter Production	0,59 kg/m ³
				Water from sludge	15,25 l/hr
				Waste to Oxidant ratio	0,4
				% Oxidant in sludge	52% Fe
<i>Constants and Factors</i>					
Chemical / Physical Constants			Electrochemical Constants		
Molecular Weight Iron	55,85 g/mol	Mol Electron Release during Fe EC	3 e ⁻		
Molecular Weight Aluminum	26,98 g/mol	Mol Electron Release during Al EC	3 e ⁻		
Density Iron	7860 kg/m ³	Faraday Constant	96485 C/mol		
Density Aluminum	2702 kg/m ³				
Iron costs	€ 0,55 €/kg				
U Fe	1,41 V				
U AL	1,67 V				

Figure 3-5 Example of the QStone Capital calculation sheet (this excel is not shared, calculations are based on QStone Capital data)

The investment costs of the electrocoagulation installation have been estimated extrapolating data used by QStone Capital with an accuracy of ± 30%. Investment costs do not include control cabinet, cabling and mounting in the basin. The substantiation of EC's operating costs is based on information using QStone Capital calculation sheet and calculations from KWR. Based on this information and previous data/EC experience, KWR has calculated the energy costs [€/m³ and €/year], the iron costs [€/m³ and €/year], the energy efficiency [kWh/kg Fe], etc. Furthermore, for the current practice, the investment and operating costs of conventional coagulation-flocculation with FeCl₃ dosage followed by lamella sedimentation were determined with the Cost Standard drinking water tool of Royal HaskoningDHV (Bakker, 2015).

4 Results and Discussion

This chapter presents the main results obtained from the experimental work and discusses their implications in terms of cost calculations for implementation and achieved removal efficiencies of the different targeted compounds/parameters per water type (see Table 1).

Table 1 Characteristics of the 3 water types

Water type	Fe [mg/L]	PO4 [mg/L]	TOC [mg/L]	Conductivity [uS/cm]	pH	Turbidity [FNU]	N-NO3 [mg/l]
PWN: Surface water	0.007	0.285	4,9	550	8.03	5.7	N.D
Vitens : backwash water from rapid sand filters	173	N.D	N.D	536	7.58	1000	N.D
WML: groundwater	0.0	N.D	N.D	611	8.0	0.09	48

4.1 PWN surface water

4.1.1 Release of iron, decrease in oxygen and formation of iron sludge

Figure 4-1 shows the iron concentration after EC at different varying applied current at 0.5 and 1.0 cm electrode gap distance. The Fe dissolution appears to be linear with the applied current with a coefficient of determination $R^2 > 0.98$ independently of the electrode gap distance, and the dissolution at a distance of 1 cm seems to be higher when the applied current is higher.

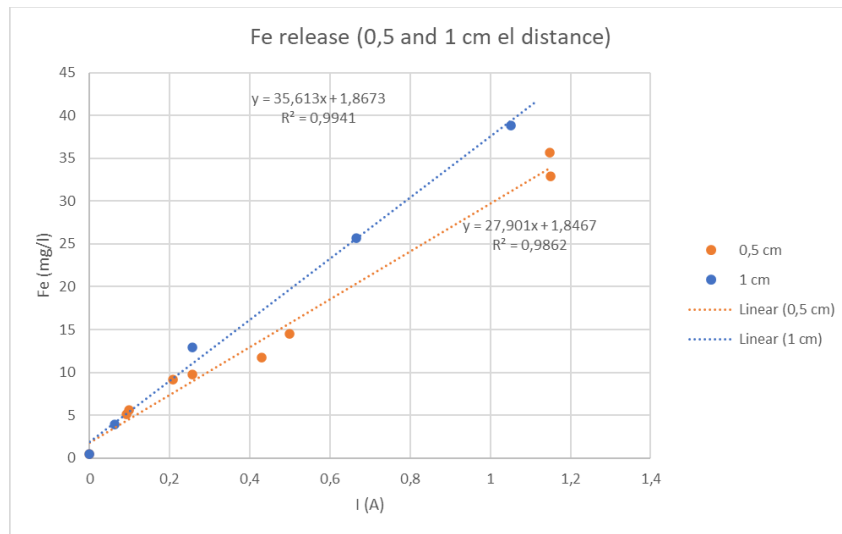


Figure 4-1 Release of iron at different applied current over the iron electrodes with different electrode gap distances.

In Figure 4-2 the oxygen concentration measured in compartment 2 is shown. As expected, the oxygen decreased due to Fe(II) undergoes oxidation by releasing an electron to form Fe(III).

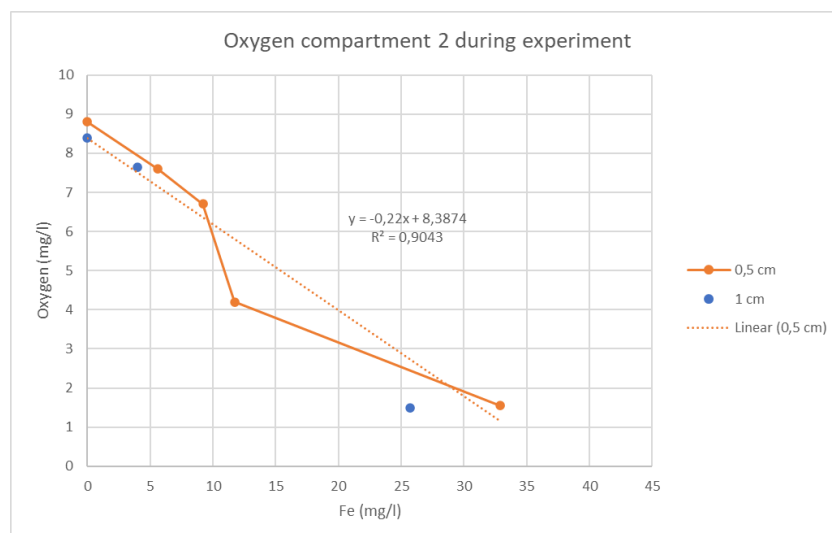


Figure 4-2 Oxygen concentration in compartment 2 measured during the experiments.

By comparing this decrease in oxygen with the maximum theoretical decrease (1 mg/l Fe : 0.72 mg/l O₂), it appears that in the second compartment not all the Fe(II) has yet been converted to Fe(III) since the measured decreased ratio was approximately 1 mg/l Fe : 0.23 mg/l O₂. This decrease in oxygen can be an undesirable side effect of using EC method if a certain concentration of oxygen in the water is desired, which can be controlled by aerating in another stage without affecting the floc formation and sedimentation.

Figure 4-3 shows the amount of iron sludge in ml/l resulting from 72 h in a Imhoff sedimentation cone. Approximately 0.76 ml/l of sludge is produced per 1 mg/l Fe dosed by EC. Under full-scale operational conditions, it is expected to be about 0.10 kg/m³ of sludge being produced. The results from the electrode distance gap of 1 cm where consistent and behaved linearly as the Fe concentration dose increased. At a dose of 20 mg Fe/l, about 12 ml/l of sludge settled down.



Figure 4-3 Amount of iron sludge measured after 72 h Imhoff sedimentation cone.

4.1.2 Turbidity, phosphate and TOC removal

Figure 4-4 to Figure 4-6 shows the turbidity, total phosphate and TOC decreased in water for each of the tests performed, measured after 72 h of settling.

The turbidity was removed by 94% at the maximum iron doses applied for both 1 and 0.5 electron gap distances, 39 mg Fe/l and 33 mg Fe/l correspondingly. In the case of 1 cm electron gap distance, a dose of 26 mg Fe/l decreased the turbidity by 87%.

In the case of total phosphate, already at low applied dosages of 4 mg Fe/l and 5.6 mg Fe/l, the phosphate removal was above 91%, reaching the limit of detection of the analysis (0.025 mg/l), and therefore no differences were determined between the electrode gap distances on this matter.

TOC decreased was more prominent with the electrode gap distance of 0.5 cm. A TOC removal of 39% was achieved after applying 32 mg Fe/l compared to a similar removal but with a higher dose of 39 mg Fe/l at 1 cm electrode gap distance.

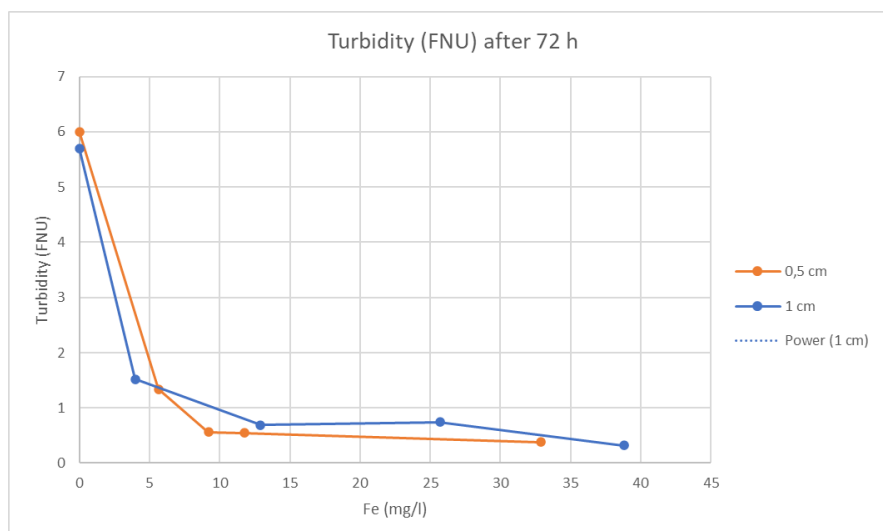


Figure 4-4 Turbidity at different iron concentrations (dose) after 72 h of settling.

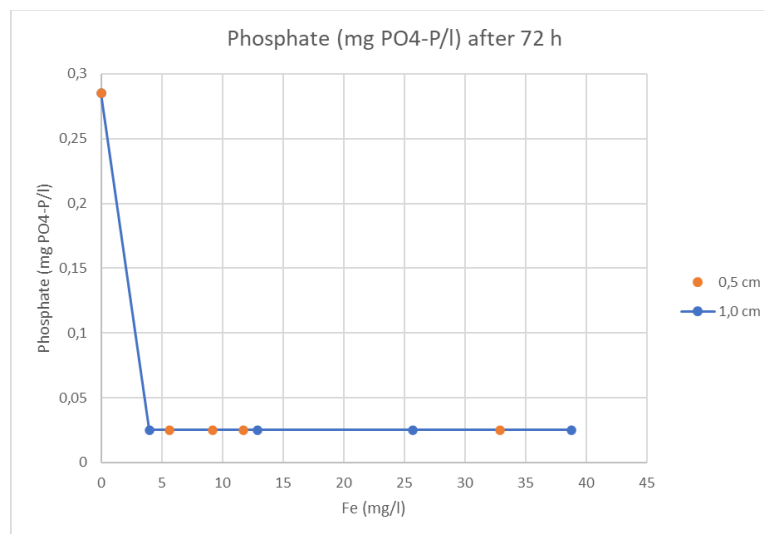


Figure 4-5 Total phosphate at different iron concentrations (dose) after 72 h of sedimentation

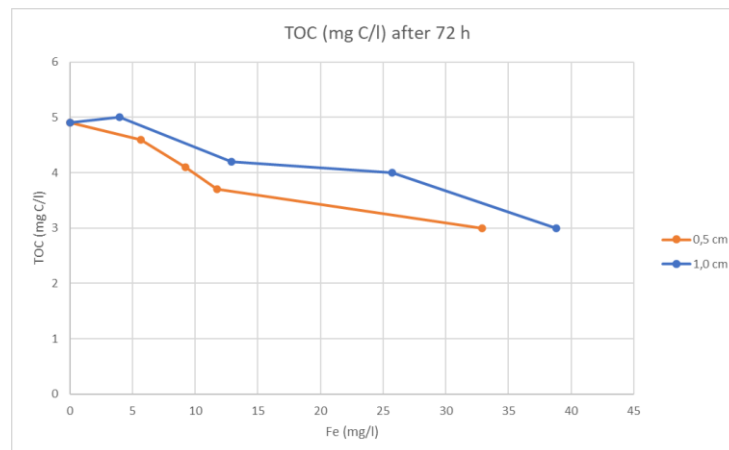


Figure 4-6 TOC concentration at different iron concentrations (dose) after 72 h sedimentation

From the result after settling of the flocs, it was found that there was also a comparable reduction in TOC, turbidity and total phosphate by applying different iron dosages at both electrode gap distances. However, the dosing of iron (as coagulant or via electrocoagulation), if well-sedimentable (heavier) flocs are formed, will ensure a faster and higher decrease in these parameters. It should be noted that the lab set-up used does not achieve optimal flocculation, and this could be better investigated with a pilot on location. It was observed that generally better flocculation occurred at higher iron doses.

Figure 4-7 shows an example of flocculation (small flocs formation) in compartments 2 and 3 during the experiments.

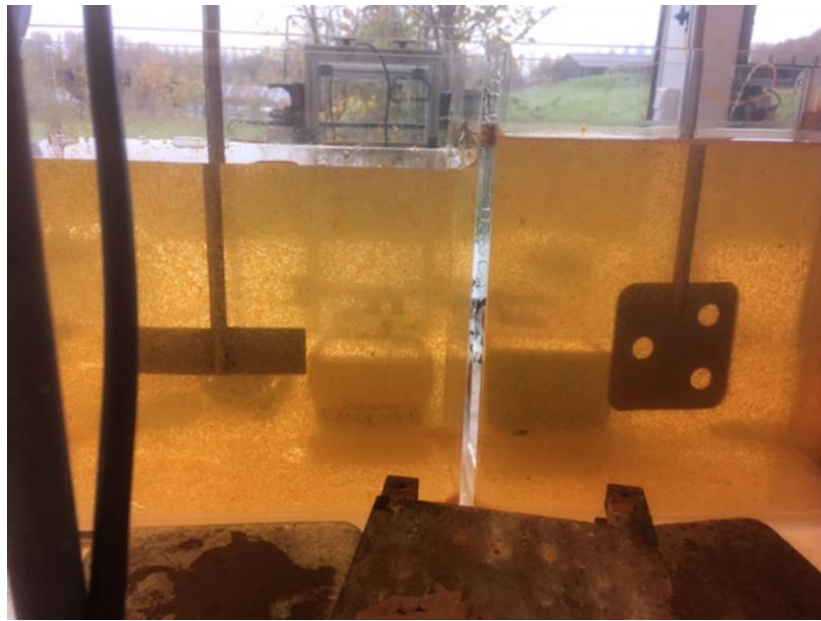


Figure 4-7 Flocculation in compartments 2 and 3 (right to left) during the experiment with approximately 12 mg Fe/l

4.2 Vitens backwash water from rapid sand filters

4.2.1 Release of iron, decrease in oxygen and formation of sludge amounts

For the Vitens case, backwash water from rapid sand filters was used to perform EC experiments. From water analysis it was determined that the backwash water already contained a relatively large amount of iron, approximately 170 mg Fe/l. This can also be observed in Figure 4-8 and Figure 4-9, in which the water appearance and iron concentrations are denoted.

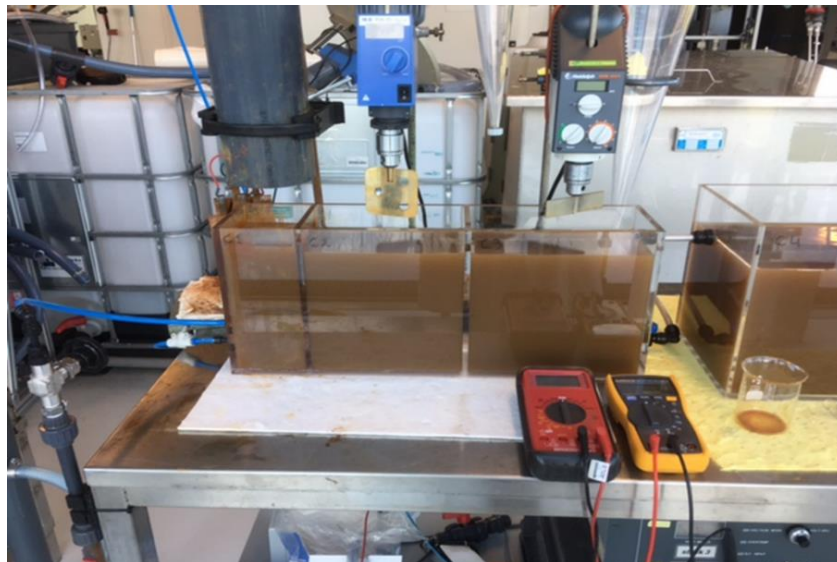


Figure 4-8 Backwash water from rapid sand filter in the EC setup during an experiment.

Figure 4-9 up to Figure 4-11 present the iron release, the oxygen concentration and the amount of iron sludge formed, respectively.

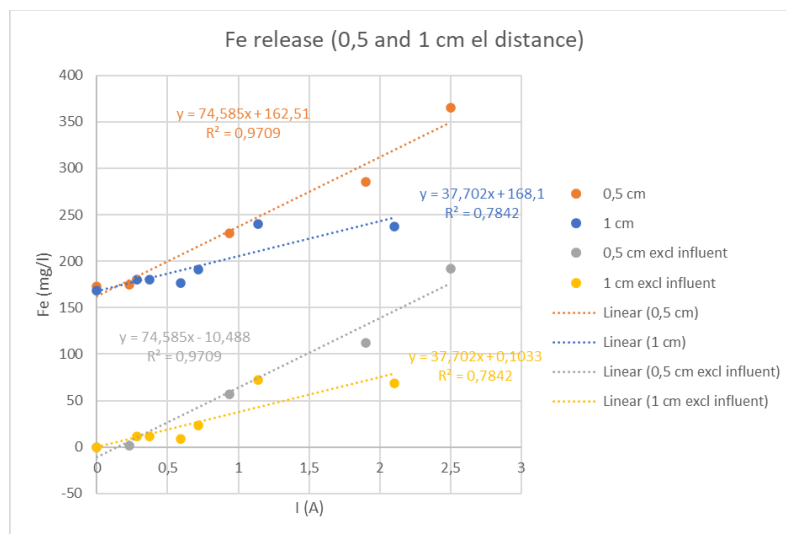


Figure 4-9 Release of iron at the applied current over the iron electrodes with different electrode gap distances.

Contrary to what it was observed with the surface water from PWN in the previous section, with the backwash water, the electrode gap distance of 0.5 cm showed to be more effective to release iron at a lower applied current and therefore consuming less energy (see Figure 4-9), also fitting a linear behavior. This might be explained by the difference in water quality, specially on the iron concentration in the water.

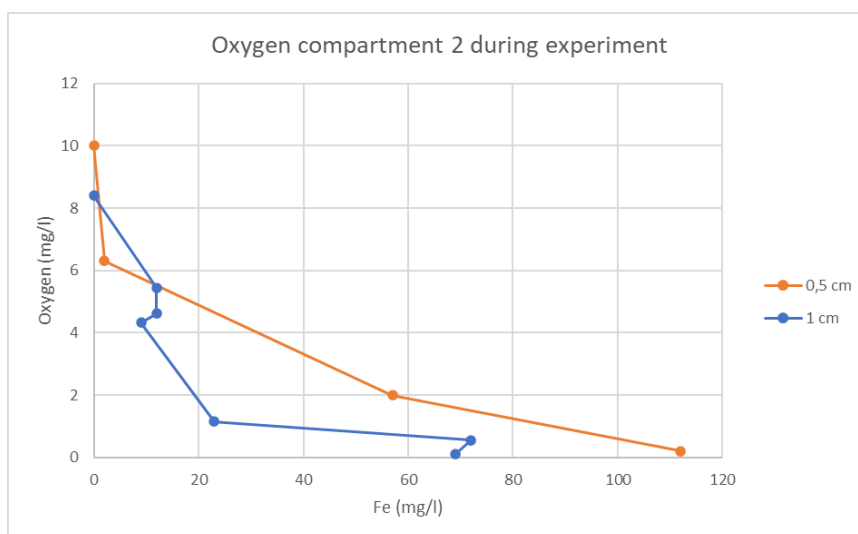


Figure 4-10 Oxygen concentration in compartment 2 during the experiments

The oxygen concentration decreased from 8.4 mg/L and 10 mg/L to 0.1 mg/L and 0.2 mg/L, respectively showed the consumption of oxygen for the oxidation of iron. Only at very high iron dosage the oxygen gets limited in the water.

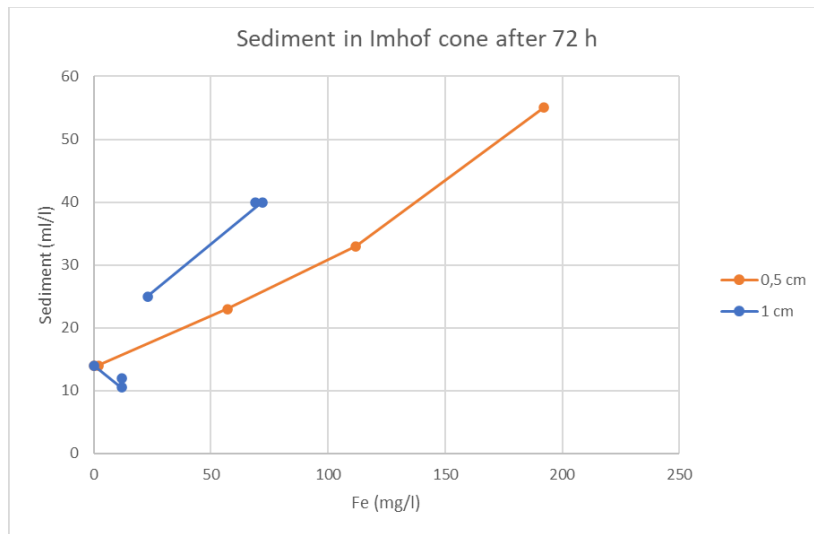


Figure 4-11 Amount of iron sludge measured after 72 h imhoff sedimentation cone.

Figure 4-11 shows the amount of iron sludge in ml/l resulting from 72 h in a imhoff sedimentation cone. Approximately 0.21 ml/l of sludge is produced per 1 mg/l Fe dosed by EC. Under full-scale operational conditions and a dose of 43 mg/L, it is expected to be about 1.17 kg/m³ of sludge being produced. The results from the electrode distance gap of 0.5 cm where consistent and behaved linearly as the Fe concentration dose increased. At a dose of 57 mg Fe/l , about 23 ml/l of sludge settled down.

4.2.2 Turbidity and suspended solids removal

Figure 4-12 and Figure 4-13 show, respectively, the turbidity and suspended solids in the treated water by EC after 3 h of settling. The samples were taken in compartment 4.

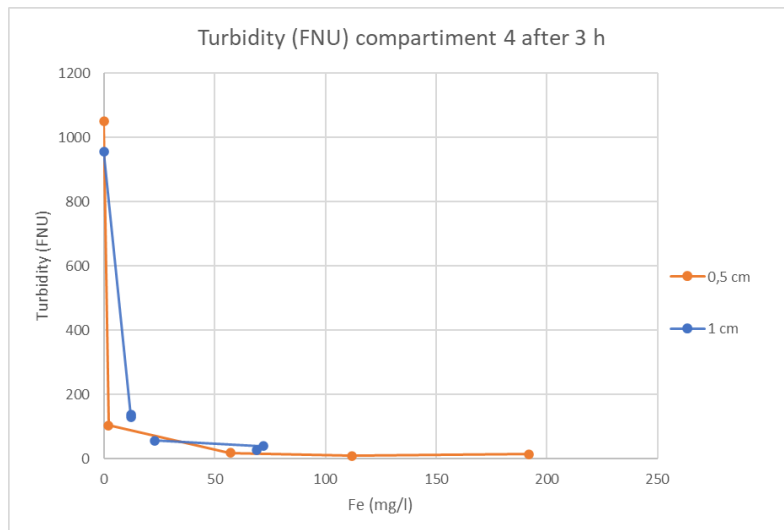


Figure 4-12 Turbidity at different iron concentrations (dose) after 3 h of settling.

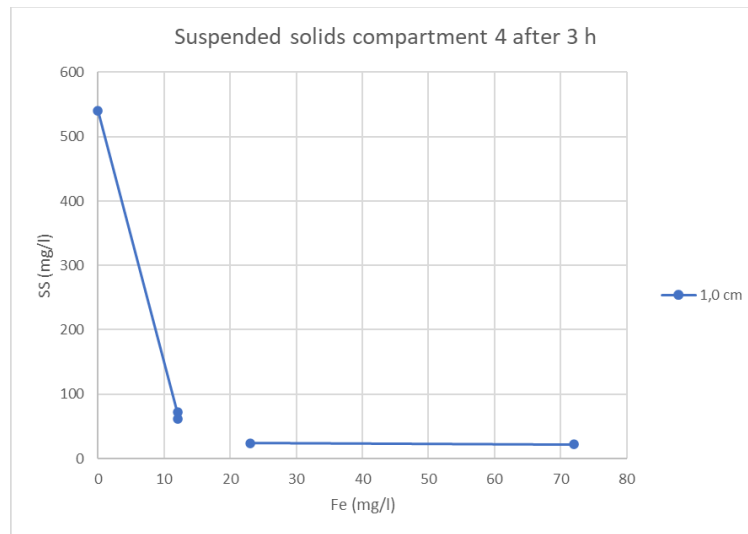


Figure 4-13 Suspended solids after 72 h sedimentation, measured only at 1 cm electrode gap distance.

The turbidity removal efficiency varied from 94% to 98% for both electrode gap distances, confirming that EC is effective and might be even further optimized.

The removal of suspended solids was determined for the tests with 1 cm of electrode gap distance, which was about 96% after 3 hours of settling for iron doses higher than 23 mg Fe/l. However, when the dose was increased to 72 mg Fe/l, the suspended solids removal was not increased.

Flocculation was not properly investigated with the setup used and with the tested water types. The iron concentration affects the formation of the flocs. At relatively high concentrations, significantly more settleable flocs were formed, but even at this dosage relatively many small non-settled flocs remained in the water. In addition, the pH also plays a role in the formation of the flocs, but the pH of the water meets the desired pH for the process, which is approximately 7.6 before and 8.4 after coagulation. The advantage claimed in the literature (see Table 5, Appendix II) that better settleable flocs are formed with electrocoagulation than with chemical coagulation has not been established with these experiments.

4.3 WML groundwater

4.3.1 Release of iron, decrease in oxygen and sludge amount

In Figure 4-14 the applied current against the dissolution of iron in groundwater at different electrode gap distances is shown. The linear regression indicates that the electrode gap distance requires slightly less current to produce the same amount of iron dosage from the EC system.

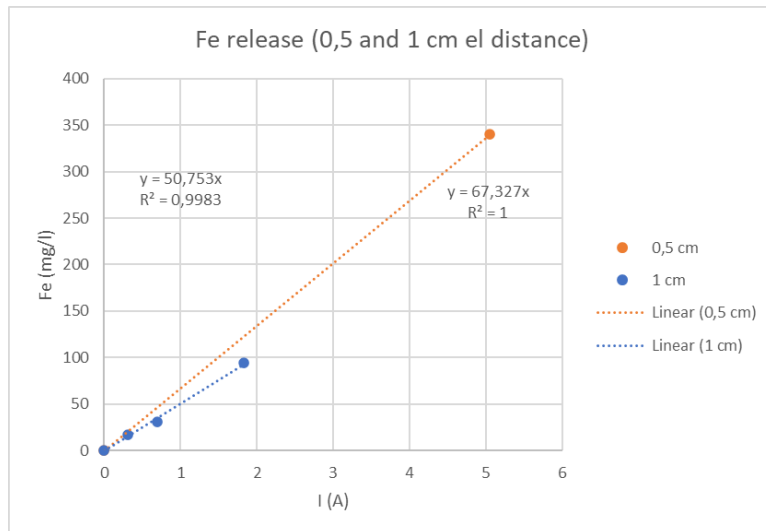


Figure 4-14 Release of iron at different applied current over the iron electrodes with different electrode gap distances.

Moreover, the oxygen concentration in the compartment 2 indicated a consumption of oxygen from 2.4 mg/l to 0.12 mg/l with a dose of 94 mg Fe/l (Figure 4-15).

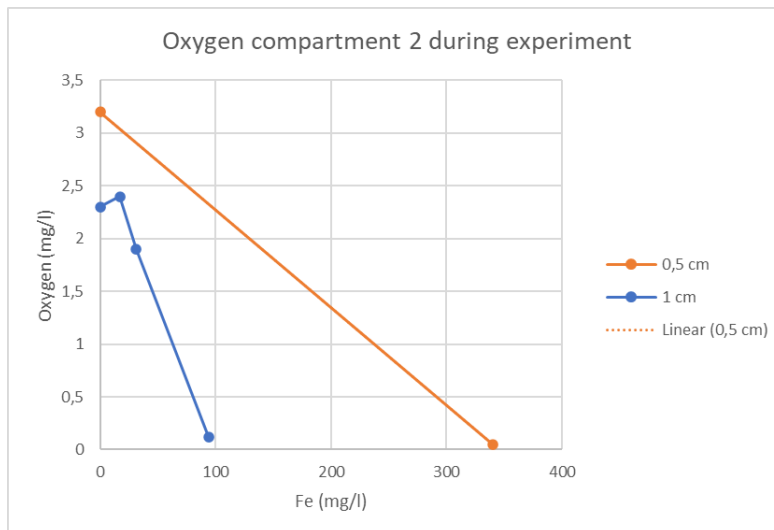


Figure 4-15 Oxygen concentration in compartment 2 during the experiments



Figure 4-16 Amount of iron sludge measured after 72 h Imhoff sedimentation cone.

Figure 4-16 shows the amount of iron sludge in ml/l resulting from 72 h in a Imhoff sedimentation cone. Approximately 0.17 ml/l of sludge is produced per 1 mg/l Fe dosed by EC. The results from the electrode distance gap of 0.5 cm where consistent and behaved linearly as the Fe concentration dose increased. At a dose of 94 mg Fe/l , about 15 ml/l of sludge settled down at an electrode gap distance of 1 cm, while 60 ml/l of sludge were produced at a 340 mg Fe/l dose at 0.5 cm of electrode gap distance.

4.3.2 Nitrate and sulphate removal

The target for WML groundwater is to remove nitrate and sulphate. Figure 4-17 and Figure 4-18 show, respectively, the concentrations of nitrate and sulphate after 24 h of settling after the EC treatment.

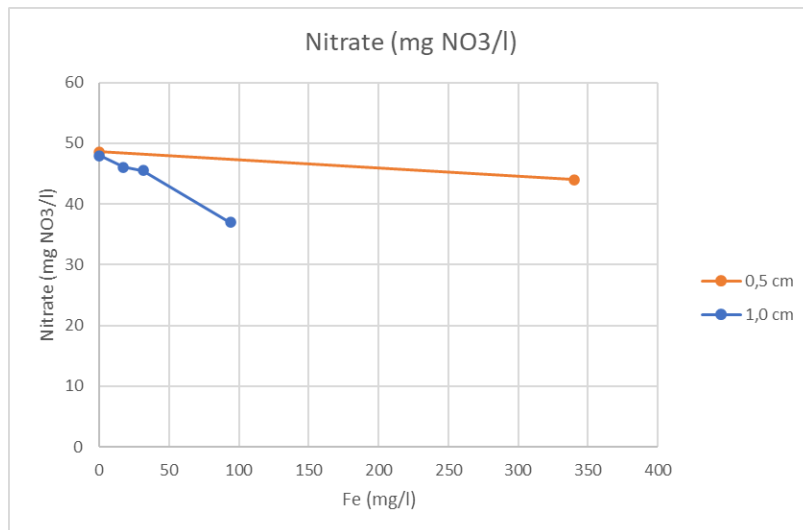


Figure 4-17 Nitrate concentration after EC treatment and 24 h sedimentation at two electrode gap distance.

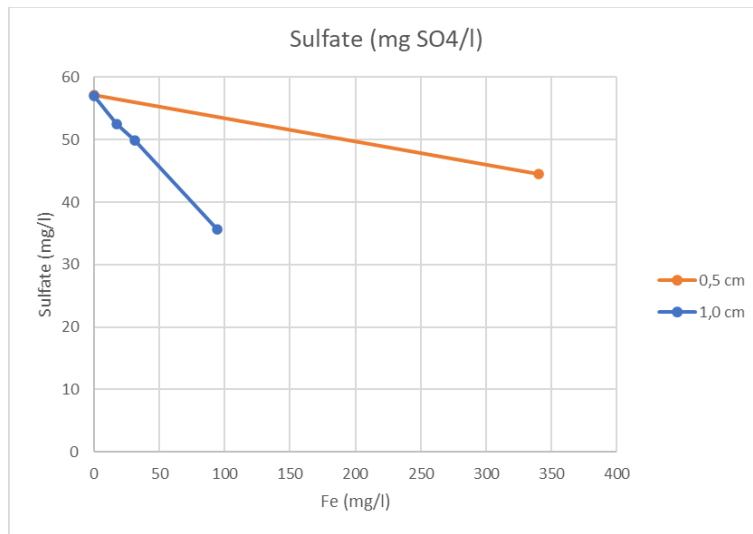


Figure 4-18 Sulphate concentration after EC treatment and 24 h sedimentation at two electrode gap distance.

Nitrate removal efficiencies of only 23% and 9% were achieved when applying high doses of iron as 94 mg Fe/l and 340 mg Fe/l at 1 cm and 0.5 cm of electrode gap distance, respectively. Similarly, sulphate removal efficiencies achieved were only of 38% and 22%. Clearly, the electrode gap distance of 1 cm favored both the nitrate and sulphate concentration decrease compared to 0.5 cm electrode gap distance.

Since the removal of nitrate and sulphate with iron electrocoagulation was not satisfactory, in agreement with the project partners, it was decided to try another method. Based on the literature review, aluminum electrocoagulation could improve the nitrate removal significantly, however, the amount of aluminum sludge created will hampered the quality of the treated water for further purification. Therefore, another method was chosen to be tried out. Specifically, electrolysis using graphite electrodes was proposed as a possible option for proof of concept tests.

Figure 4-19 shows decrease in nitrate concentration during the electrolysis time at three different voltages applied, using the graphite electrodes. It appeared quite rapidly that nitrate removal was only possible to a limited extent (up to approximately 4%) with these electrodes, which was rather disappointing. In addition, the electrodes were found to release material in each experiment (yellow color, see Figure 4-20), possibly hindering the conversion of nitrate. However, this was not quantified but the material settled down after some time. Therefore, further investigation was stopped.

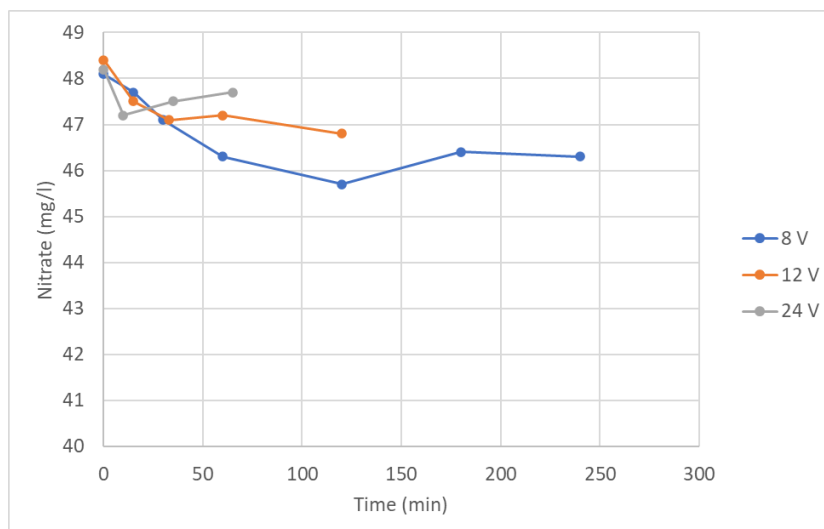


Figure 4-19 Reduction of nitrate by applying the electrochemical conversion with graphite electrodes

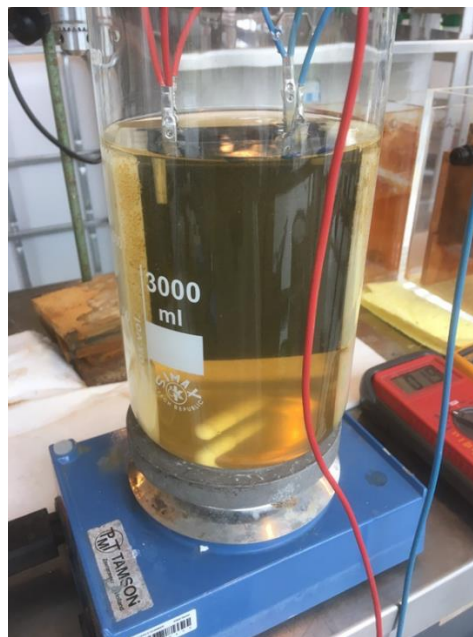


Figure 4-20 Yellow color of the water after carrying out the electrochemical experiment

4.4 Cost estimation and scalability

This section provides an overview of a calculated cost comparison between iron electrocoagulation and conventional coagulation/sedimentation dosing ferric chloride, by translating the experimental results into an operational scale of the system.

Iron chloride dosing is currently the preferred technique used in water treatment to add an iron flocculant to water. The investment and operating costs of ferric chloride have been determined with the Cost Standard drinking water of RHDHV (Bakker, 2015). For small scale systems, extrapolation has been used. The investment costs of the electrocoagulation installation have been estimated extrapolating data used by QStone Capital (see section 3.5).

For these calculations, we have considered two practical scenarios in which significant removal efficiency of targeted parameters was achieved (1) sand rapid filter backwash water treatment at Vitens, with a capacity of 26 m³/h and (2) treating surface water from the IJsselmeer at PWN (Andijk) with a capacity of 8.000 m³/h.

For both cases, the results of the electrocoagulation experiments were taken from the linearization of the tests results (current vs Fe dosage) taking the same actual Fe dosage, in which 43 mg/l Fe dissolved in the water was used for Vitens and 20 mg/l Fe dissolved in the case of PWN.

It is assumed that flocculation takes place with a residence time of 20 minutes, followed by a lamella settling with a design surface load 0.9 m³/h / m². These are common practice values in the Dutch water sector.

The electrodes for electrocoagulation cost about 0,55 €/kg Fe, while 40% iron chloride (FeCl₃) costs were assumed to be 2,40 €/kg Fe for Vitens and 1,60 €/kg Fe (based on actual costs). The energy costs used were 0,350 €/kWh for PWN and 0,247 €/kWh for Vitens (current energy costs, but they are exceptional high given the energy crisis in

2022). Normal price of energy before was about 0,07 €/kWh for water companies in the Netherlands). The summary of these input values are presented in Table 2.

The storage capacity of 40% ferric chloride is 12 days (single-walled), with a strategic reserve of 15%, 100% reserve rack pumps and 1 emergency/eye wash is provided. Transport costs of ferric chloride and caustic soda are not included in the calculations.

Table 2 Input data provided by Vitens and PWN for comparison with EC.

	Vitens	PWN
Flowrate [m ³ /h]	26	8000
Fe dose[mg/L]	43	20
Cost (FeCl ₃ 40%) [Euro/kg]	0,33	0,22
Other Chemicals consumption:		
PE (Praestol 2515, Wispro)	Average 5 mg/L	0,15 mg/L (w), 0,05 mg/L (s)
Cost (euro/kg)	7,56	5,9*
Energy cost [euro/kWh]	0,247	0,35

*Assumed value by RHDHV tool.

provides an overview of the determined costs. They are classified as follows:

- Calculation QStone Capital, based on the EC spreadsheet in which the results of experiments are filled in;
- Calculation KWR, calculated with the results from the spreadsheet of QStone Capital and experiments;
- Investment costs RHDHV, floc formation, lamella settling and chemicals (ferric chloride and polyelectrolytes) storage and dosing;
- Operating costs RHDHV, floc formation, lamella settling and chemicals (ferric chloride and polyelectrolytes) storage and dosing;
- Investment costs of the electrocoagulation installation for both Vitens and PWN are extrapolated from the cost curves determined with previous data shared from by QStone Capital for other Dutch drinking water companies.

As it can be seen from the pilot research values, a higher current of 0.72 A but a lower voltage of 3.23 might be used for the case of Vitens, compared with a current of 0.61 A and a voltage of 5.05 V of PWN. The difference lay on the electrode gap distance used, the iron dose and the water characteristics from the rapid sand filter and the surface water. Correspondingly, at PWN the energy needed for EC is higher, of about 0.15 kWh/m³ compared to 0.12 kWh/m³ with backwash water from rapid sand filter (see Table 3). However, due to the high suspended solids and the high dose of iron, the sludge production is significantly higher with 1.17 kg/m³ compared to 0.1 kg/m³ with surface water at PWN. Therefore, the cost of energy per m³ of treated water by EC is about 0.053 euro/m³ for surface water and 0.029 euro/m³ for backwash water from rapid sand filtration. The latter is also due to the energy per kg of iron calculated based on the experimental results for PWN is high of about 7.7 kWh/kg Fe, while for Vitens is only 2.7 kWh/kg Fe. Here, there might be room for optimization in the case of surface water.

Table 3 also indicates that all the costs of the Vitens case, due to the lower capacity, are significantly lower overall than those of PWN. If we now look at both scenarios separately, the largest items of the investment costs for a flocculation with EC and one with iron chloride appear to be almost the same in both cases. This is mainly because the flocculation and flocs removal unit, which accounts for the largest share of investment costs, is equal. Therefore, the difference in the case of Vitens is 4 k€ when considering about 65 k€ for the EC unit compared with 61 k€ for the dosing units (no data for the polyelectrolyte PE dosage). In the case of PWN, the difference is significant when comparing 1300 k€ of the EC to 257 k€ for the dosing units, since the amount of EC modules and invertors needed for 8000 m³/h is high (64 modules).

At Vitens, the EC unit consists of 1 modules of 150 plates of 15 k€ (excl. plates), 3 invertors of 15 k€ and a blower of 2.5 k€. c. For PWN, the possibility of having bigger modules might decrease the investments cost, but no cost data for these bigger modules is available. At PWN these are 64 modules of 150 plates of 15 k€ each (excl. plates), 16 invertors of 20 k€ and a blower of 20 k€. The plate spacing based on the experiment results was 0.5 cm for Vitens and 1 cm for PWN and the stand time of the iron plates is 24 months. In the investment costs of EC, the control box, cabling and assembly of the electrodes in the basin are not included.

The operating costs of the EC process are 27.8 k€/year (53%) lower than ferric chloride dosage for the case of Vitens with 26 m³/h. For PWN the situation is the opposite, since in this case of 8000 m³/h the difference is 1911 k€/year of operational cost less for the conventional coagulation-flocculation. The reason is that the energy consumption cost of the EC will be about 3765 k€/year at an actual cost of 0.35 €/kWh. If this cost goes back in the future to values of about two years ago of 0,07 €/kWh, then the total OPEX for the EC would be 1785 k€/year compared to about 2885 k€/year of conventional coagulation in which the main contribution is due to chemicals consumption costs. Clearly, the energy costs will determine the feasibility of the EC regarding OPEX. At an energy price of 0.17€/kWh, both EC and conventional coagulation costs have similar operational costs.

The operational costs do not include the costs of manpower for business operations and maintenance. The electrodes of the EC are estimated to be replaced every 2 years, but it is unknown how much time the change will take. This time effort should be compared to processing a supply of iron chloride once every 12 days.

It must be pointed out that by dosing ferric chloride, the chloride concentration of the water increases by 81.9 (Vitens) and 38.2 mg/l (PWN) respectively.

Table 3 Overview of costs of iron electrocoagulation and ferric chloride dosage

	Unit	Vitens		PWN	
Design flow rate	m ³ /h	26		8.000	
Design iron dose	mg/l Fe	43		20	
	mg/l FeCl ₃	124,9		58,1	
Water quality					
Conductivity	mS/cm	0,528		0,550	
pH		7,30		8,03	
Temperature	°C	12		15	
TSS	mg/l	540		10	
		EC	FeCl ₃	EC	FeCl ₃
Pilot research					
Flow rate	l/h	20		20	
Current	A	0,72		0,61	
Voltage	V	3,23		5,05	

Electrode gap distance	cm	0,5		1	
Iron dose	mg/l Fe	43		20	
Calculation QStone Capital					
Energy	kWh/m ³	0,12		0,15	
Iron hydroxide formation	kg/m ³	0,54		0,01	
Sludge production (50% dry matter)	kg/m ³	1,17		0,1	
Calculation KWR					
Cost of energy	€/m ³	0,029		0,053	
Cost iron electrodes	€/m ³	0,024		0,011	
Energy efficiency**	kWh/kg Fe	2,70		7,7	
Increase in water chloride content	mg/l Cl	0	81,9	0	38,2
Investment costs RHDHV					
Flocculation	K€	122*	122*	14.795	14.795
Lamella settling	K€	496*	496*	23.387	23.387
EC Unit	K€	65 ³		1.300 ³	
Dosing unit FeCl ₃	K€		28*		97
Dosing unit NaOH (pH correction)	K€		33*		100
Dosing PE	K€				60
<i>Subtotal</i>	K€	683	679	39.482	38.439
Operating costs RHDHV					
FeCl ₃	K€/year		23,5		2233
NaOH	K€/year		N.D		97
PE	K€/year		9,1		201
Electrodes	K€/year	5,4		769	
Energy flocculation/lamella	K€/year	13,2	13,2	263	263
Energy dosing	K€/year		7,1		92
Energy – EC	K€/year	6,5		<u>3765</u>	
<i>Subtotal</i>	K€/year	25,1	52,9	4796	2885

* Costs determined by power/polynomial function from data, based on flow. RHDHV tool is not yet available for a range of 26 m³/h flow rate.

** Energy efficiency was calculated taking into account the energy needed for EC (KWh/m³) and the dosage of Fe (kg Fe/m³).

When calculating the investment costs, it was assumed that the flocculation time in electrocoagulation and dosing of ferric chloride is the same. The same applies to the load on the lamella sedimentation. For optimization of sedimentation the experiments are suggested to be carried out in a demo scale and continuous period of evaluation, so that the questions regarding flocculation effectiveness can be solved as well as refining the cost estimations.

5 Conclusions and recommendations

The following conclusions can be made out of this study:

- Electrocoagulation is technically feasible for the controlled dosing of iron to water of three different types and characteristics: surface water, groundwater and backwash water. The dosage of iron differs per water type and electrode gap distance (0.5 and 1 cm); and different applied currents are needed for achieving the same iron concentrations.
- For PWN surface water:
 - The maximum turbidity removal efficiency achieved was 94% for both 1 and 0.5 electrode gap distances. TOC removal of 39% was achieved after applying 32 mg Fe/l and 39 mg Fe/l at 0.5 and 1 cm electrode gap distances. In the case of total phosphate, already at low applied dosages of 4 mg Fe/l and 5.6 mg Fe/l, the phosphate removal was above 91%.
 - Approximately 0.76 ml/l of sludge was produced per 1 mg/l Fe dosed by EC. Under full-scale operational conditions, this is expected to be about 0.10 kg/m³ of sludge being produced.
 - The cost of EC energy per m³ of water treated was estimated to be at 0.15 kWh/m³ at a dosage of 20 mg Fe/l, with an applied current of 0.61 A, 5.05 V at an electrode gap distance of 1 cm. The cost of electrodes were 0.011 €/m³ at an energy efficiency of 7.7 kWh/kg Fe, which is rather high. Further optimization to reduce this energy efficiency might be necessary.
 - Electrocoagulation investment costs for treating 8000 m³/h are about 1043 K€ higher compared to ferric chloride dosage, mainly coming from the EC composed by 64 modules of 150 plates each, 16 invertors and blower.
 - The operational costs are 1911 k€/year higher for EC compared to ferric chloride dosage. Energy consumption cost of the EC will be about 3765 k€/year at energy cost of 0.35 €/kWh. If this energy cost is reduced to normal prices of 0,07 €/kWh of drinking water companies in the Netherlands, then the total OPEX for the EC would be 1785 k€/year compared to about 2885 K€/year of conventional coagulation. EC operational costs are lower when energy prices in the range of 0.07 to 0.17 €/kWh are taken into account.
 - During the experiments no compact, heavy and easily settleable flocs were formed. Therefore, flocculation needs to be investigated further because of the limitations of the laboratory setup.
- For the backwash water of rapid sand filtration of Vitens:
 - The turbidity removal efficiencies obtained varied between 94% and 98% for both electrode gap distances.
 - The removal of suspended solids was determined for the tests with 1 cm of electrode gap distance, which was about 96% after 3 hours of settling for iron doses higher than 23 mg Fe/l.
 - Approximately 0.21 ml/l of sludge was produced per 1 mg/l Fe dosed by EC. Under full-scale operational conditions and a dose of 43 mg/L, the expected sludge being produced is about 1.17 kg/m³.

- The cost of EC energy per m³ of water treated was estimated to be at 0.12 kWh/m³ at a dosage of 43 mg Fe/l, with an applied current of 0.72 A, 3.23 V at an electrode gap distance of 0.5 cm. The cost of electrodes were 0.024 €/m³ at an energy efficiency of 2.7 kWh/kg Fe.
- The investment cost for a EC unit composed by 1 modules of 150 plates, 3 invertors and a blower was determined about 65 k€ , which resulted to be 4 k€ higher compared to conventional coagulation.
- The operational costs of the EC were 27.8 k€/year, which is 53% lower than conventional coagulation treating 26 m³/h backwash water from rapid sand filters in Vitens.
- This water type is the most suitable for EC of the three analyzed, also taking into account the overview of Pros and Cons of EC (see Appendix II, Table 5). It remains unclear whether flocs formed by EC are larger and more stable compared to conventional coagulation/flocculation with doing FeCl₃.
- For the groundwater water of WML:
 - Nitrate removal efficiency of maximum 23% was achieved when applying high doses of iron as 94 mg Fe/l and 340 mg Fe/l at 1 cm and 0.5 cm of electrode gap distance, respectively. Similarly, sulphate removal efficiencies achieved were only of 38% and 22%. Clearly, the electrode gap distance of 1 cm favored both the nitrate and sulphate concentration decrease compared to 0.5 cm electrode gap distance.
 - Since unsatisfactory results were obtained with EC, electrolysis with graphite electrodes was chosen for a proof of concept test. However, the removal of nitrate was disappointing reaching only about 4% removal after an electrolysis time of about 2 h. Further research with focus on electrocoagulation removal of nitrate needs to be carried out, since in literature there are some reported promising results.

The following recommendations were formulated:

- Since the results of the experiments showed that flocculation and settling can be further optimized, and therefore the removal efficiencies for suspended solids and turbidity are expected to increase. Modeling would be an option to help on determining key parameters for the EC process couple to flocculation/sedimentation, forming the basis for a theory-based installation design.
- Additionally, knowledge exchange between technologists and engineers from the project partners and EC technology suppliers can provide a more accurate estimate of the investment costs of a drinking water EC installation.
- Points of attention include the energy efficiency (kWh/kg Fe) which was higher than 7 in the case of surface water (PWN), and larger modules/electrodes for EC at this large capacity, in which the feasibility of the technology is relying on energy prices between 0,07 – 0.17 €/kWh.
- On-site pilot continuous experiments with longer duration are expected to provide better insights into the application of EC in the drinking water sector, and are recommended for further investigation specially with the backwash water from rapid sand filters of Vitens, in which EC seems more directly suitable to be applied.

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I Annex: Theoretical calculation of the oxidation of Fe(II)

Table 4 Theoretical calculation of the oxidation of Fe(II)

Theoretical decrease in oxygen due to complete conversion of Fe(II) to Fe(III)			
[Fe]	55,85	g/mol	
[O ₂]	32	g/mol	
$\text{Fe} + \text{O}_2 + 2 \text{H}_2\text{O} \rightarrow \text{Fe(OH)}_2$		Dissolving electrode	
$4 \text{Fe(OH)}_2 + \text{O}_2 + 2 \text{H}_2\text{O} \rightarrow 4 \text{Fe(OH)}_3$		Conversion to Fe(III)	
1 mol Fe	2 mol O ₂	Consume 1,25 mol O ₂ for 1 mol Fe	
1	8,4	mg/l	Present dissolved oxygen in water
0,017905	0,2625	mmol/l	in mM
0,017905	0,02238138	Conversion ratio (1:1.25)	
1	0,72	Decrease in mg/l oxygen per mg/l iron	
10	7,2	Maximum oxygen decrease (mg/l) when dissolving 10 mg/l iron	


II Annex: Pros and Cons of Electrocoagulation

Table 5 Pros and Cons from EC (adapted from (Moussa et al. 2017))

Pros	Cons
Since no chemicals are added, there is no chance of secondary pollution due to high concentration of chemicals as in CC/CF.	Regular replacement of sacrificial anode used in EC is necessary since the anode dissolves into the solution
Gas bubbles produced from EC facilitates can remove pollutants by floating them, so they could be easily collected by skimmers.	Cathode passivation can occur which decreases the efficiency of the EC process
EC is easily operated due to the simplicity of its equipment hence, complete automation of the process is possible	In some areas where electricity is not abundant, the operating cost of EC can be expensive
Wastewater treated by EC gives clear, colorless and odorless water	Electricity consumption most likely is greater than in CC.
Flocs formed by EC are much larger than CC/CF and more stable, hence they are easily separated during filtration	Less suitable for water with conductivities lower than 250 uS/cm, total suspended solids less than 20 mg/L of turbidities less than 25 NTU.
EC produces much less sludge volume than CC/CF and the sludge formed is more stable and non-toxic	
Small colloidal particles are removed by EC since the applied electric current makes collision faster and facilitates coagulation	
No need for post-neutralization in most cases with EC due to pH of the solution often increases .	
The mechanism for EC is simple and no moving parts are required.	
Elimination of the chemical supply chain and storage required for conventional chemical coagulation.	

III Annex: Composition of electrodes

Source: <https://www.hetraco.com/images/Materialen/Werkstof/1.0037.pdf>

		 Hetraco B.V.									
Werkstofnummer: 1.0037											
DIN	St 37-2	ASTM									
AISI		UNS									
Handelsnaam	Fe360	SAE									
Alloy		BS	S 235JR								
EN	S235JR	AFNOR	E24-2								
Opmerking:											
Chemische Samenstelling:											
Element	%C	%Si	%Mn	%P	%S	%Cr	%Mo	%Ni	%Nb	%N	%Cu
min	0		0	0	0					0	
max.	0,17		1,40	0,045	0,045					0,009	
Element	%W	%V	%Al	%B							
min											
max.											
Beschrijving:											
Ongelegeerd constructiestaal											