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Electrocoagulation in Wastewater Treatment: A Review

Thea Caroline Granhus
Water and Environmental Technology

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Summary

Water is our most valuable resource and essential to humans. Still, a large part of the worlds population does not have access to safe water and sanitation services. More than 80% of the generated wastewater is not treated. At the same time, new contaminants emerge. To cope with these problems, new and improved technology is needed. Electrocoagulation is an emerging treatment method. It is easy to operate, environmentally friendly, and remove a broad spectrum of pollutants from various water types.

This thesis is a review of applications, treatment effects, technological schemes, and design parameters of electrocoagulation. Current literature on electrocoagulation has been carefully evaluated to conclude on its state on these topics.

Electrocoagulation is not a new technology, but is performed limited research on the topic, which is apparent throughout this review. Analysis is mainly performed in short periods and small scale. However, these results are promising. Electrocoagulation has several advantages to other treatment processes, and emerge as a possible alternative. There is evidence of a good treatment efficiency of several pollutants from a broad spectrum of water quality. The treatment process is versatile; it is possible to use at different stages of the treatment process and has several applications. Design of the electrocoagulation unit is proven to affect both treatment efficiency and operation cost.

Sammendrag

Vann er vår mest verdifulle ressurs og essensielt for mennesker. Fortsatt har en stor del av verdens befolkning ikke tilgang til trygt vann og sanitærtjenester. Mer enn 80 % av det genererte avløpsvannet blir i dag ikke behandlet. Samtidig dukker nye forurensninger og miljøforurensninger opp. For å takle disse problemene er det nødvendig med ny og forbedret teknologi. Elektrokoagulering er en lovende metode. Det er lett å drifte, miljøvennlig og fjerner et bredt spekter av miljøgifter fra forskjellige vanntyper.

Denne oppgaven er en gjennomgang av applikasjoner, behandlingseffekter, teknologiske ordninger og designparametere for elektrokoagulering. Nåværende litteratur om elektrokoagulering er nøye evaluert for å konkludere om tilstanden til disse temaene.

Elektrokoagulering er ikke en ny teknologi, men det er gjennomført begrenset forskning på emnet, noe som er tydelig under hele denne gjennomgangen. Analysene hovedsakelig gjennomført i korte perioder og i liten skala. Disse resultatene er imidlertid lovende. Elektrokoagulering har flere fordeler til andre behandlingsprosesser, og fremstår som et mulig alternativ. Det er bevis på god behandlingseffektivitet for flere miljøgifter fra et bredt spekter av vannkvalitet. Behandlingsprosessen er allsidig; det er mulig å bruke på forskjellige stadier av behandlingsprosessen og har flere bruksområder. Det er påvist at design av elektrokoagulasjonsenheten påvirker både behandlingseffektivitet og driftskostnader.

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List of Acronyms

BOD	Biological oxygen demand
CIP	Ciprofloxacin
COD	Chemical oxygen demand
DOC	Dissolved organic carbon
MBR	Membrane bioreactor
MLSS	Mixed liquor suspended solids
S/V	Surface to volume
SiC	Silica carbide
SS	Suspended solids
TOD	Total oxygen demand
TP	Total phosphate
UASB	Upflow anaerobic sludge blanket

1. Introduction and background

In this chapter, motivation, background, and the theoretical foundations essential to this research are presented. This information is to give a short introduction to a better understanding of the research done in this thesis.

1.1 Ensuring access to water and sanitation for all

Water is our most valuable resource and is essential for all aspects of life and sustainable development, with adequate quality and quantity. Most of the earth is covered in water, but only a small amount is fresh water available to humans. Globally, agriculture withdraws 69% of all water used annually, industry 19% and households 12% (United Nations, 2018). Every time humans withdraw water, we risk polluting our freshwater sources. Wastewater from agriculture, industry, and households have to be adequately treated before it is discharged back into freshwater sources. However, today 4,5 billion people do not have safely managed sanitation services and wastewater treatment, as well more than 80% of the worlds generated wastewater is released without any treatment(United Nations, 2018). Releasing untreated wastewater degrades the water quality and our ecosystems, which leads to further reduction of freshwater availability globally. Each day, over 1,000 children die from diarrheal diseases which can be prevented with better water and sanitation services (United Nations, 2018). One of the UNs 17 sustainable development goals for 2030 is to ensure access to water and sanitation for all.

Lack of wastewater treatment is mainly a problem in developing countries, but also developed countries experience a degradation of freshwater quality. Degradation of water quality happens due to inadequate treatment of wastewater, the rise of emerging pollutants and intrusive species, and unmonitored overflow (United Nations, 2018). Inadequate treatment is often a result of economical challenges with building and operating wastewater plants and technology which are not able to remove enough of and the required pollutants. To cope with these challenges, a more integrated approach to allocating and managing water resources is necessary. These measures include protecting ecosystems as societies and economies are dependent on them. An essential part of this is to improve wastewater treatment and secure safe reuse of water. To achieve these goals, new and improved technology needs to be implemented globally, to increase treatment e□ciency and to make treatment methods more available (United Nations, 2018). One possible technology for the future is electrocoagulation. Electrocoagulation shows promising results on treatment e□ciency on several important pollutants, while it also has benefits regarding, for example, operation costs and that it is easy to operate (Mollah et al., 2001). These characteristics make it a promising technology for the future. Therefore this thesis is a review of the e□ects of electrocoagulation in wastewater treatment.

1.2 Challenges in wastewater treatment

The sustainable development goals report by United Nations (2018) regarding goal 6, clean water and sanitation, states that freshwater quality is at risk globally. The report proclaims how water quality should be monitored globally to secure adequate quality for drinking water, irrigating, and preservation of ecosystems and biodiversity. The report presents parameters that should be monitored in water sources, these are shown in table 1.1.

Table 1.1: Water quality parameters

Parameter	River	Lake	Groundwater
Dissolved-oxygen	x	x	
Electrical conductivity	x	x	x
Nitrogen	x	x	
Nitrate			x
Phosphorus	x	x	
pH	x	x	x

Source: United Nations (2018)

To maintain an adequate water quality level in freshwater sources, water has to be treated accordingly, depending on the recipient, and further pollution has to be avoided.

In Norway, water quality and treatment requirements are decided by law and controlled by the county. The requirements are set not to degrade water quality in the recipient. Treatment plants which discharge water into lakes or rivers from more or equal to 2000 pe or discharge water into the sea from more or equal to 10 000 pe have to follow these requirements. Generally, these laws require wastewater treatment plants to remove 90% phosphorous, 75% chemical oxygen demand, 70% biological oxygen demand, and 50% suspended solids (Forurensningsforskriften, 2004). All new and upgraded wastewater plants are required to have at least secondary treatment. Local governments have the authority to set higher discharge requirements than the general ones. Wastewater treatment plants also have to test for parameters as heavy metals, hydrocarbons, phthalate, and oil residue.

1.2.1 Contaminants of emerging concern

(Kreuzinger et al., 2019) defined emerging contaminants as *"Contaminants of emerging concern are substances/compounds that at present are not commonly monitored, but when present are suspected to have adverse ecological and human health effects"*. These substances/compounds come from everyday products used in modern society. The compounds include sanitary, household, and personal care products, as well as antibiotics and pharmaceutical-active compounds (Gavrilescu and Asachi, 2014). Contaminates are transferred to aquatic environments through wastewater and urban run-off. Effluent from wastewater treatment plants from several continents show high levels of these emerging pollutants (Pal et al., 2010). Modern wastewater treatment plants are designed to remove degradable carbon and nutrients, not large concentrations of emerging organic components. As a result, these contaminants are released into receiving water bodies (Kreuzinger et al., 2019).

The presence of pharmaceuticals in aquatic environments is a concern as it can lead to antimicrobial resistance, which is a significant concern to human health. Therefore, European Comission (2019) issued a report containing a strategic approach to limiting pharmaceuticals in the environment. The report states that discharge from treatment plants is one of the most significant contributing factors to the release of pharmaceuticals in the environment.

1.2.2 Aluminum residue

A frequently used coagulant for water and wastewater treatment is aluminum. As a consequence, there is aluminum residue present in drinking water, wastewater effluent, and sludge, after coagulation (Driscoll and Letterman, 1988). WHO (1998) advise to reduce aluminum residue from water and wastewater treatment as much as possible considering the viable health concerns, linking it to Alzheimer if consumed by humans. Regulations set by the European Union has a limit of 0.2 mg/L of aluminum in water (Pedersen, 2018), while WHO (1998) recommends values below 0.1 mg/L for drinking water.

Sludge contains large amounts of phosphorus, which is vital in plant growth. Therefore they use sludge as a source of phosphorus in agriculture. Aluminum residue in sludge makes the phosphorus less available to plants (Clarkson, 1967). These problems are linked to the use of aluminum as a coagulant in wastewater treatment (Odegaard, 2012). Measures like optimal pH at the point of treatment, reduced excessive dosages of aluminum, and sufficient mixing of coagulant is recommended to reduce aluminum residue (WHO, 1998).

1.3 Coagulation in wastewater treatment

The treatment process in a wastewater treatment plant is dependent on the water quality of the influent and treatment requirements of the effluent. From this information, a combination of processes can be determined to meet the requirements.

Wastewater treatment plants usually use a combination of physical, chemical, and biological processes to meet treatment requirements. The different processes can be divided into three categories, primary, secondary and tertiary treatment depending on efficiency. Primary treatment removes larger, suspended particles with physical processes and is commonly used as a pre-treatment. In some areas, this is used as the only treatment, for example, in rural areas in western Norway. Secondary treatment removes nutrients and remaining suspended solids, most commonly using chemical or biological treatment, or a combination of the two. The last step is the tertiary treatment. This step involves removing remaining inorganic compounds and bacteria, viruses, and parasites (Odegaard, 2012).

1.3.1 Coagulation

Coagulation is a well known and used method for wastewater treatment to remove suspended particles and colloids. Traditionally coagulates are divided into two different groups, aluminum-based or iron-based. The coagulant is mixed into the water, which leads to the forming of larger particles, flocs. Floc formation occurs due to the destabilization of colloids reducing the repulsive forces by collision with counterions. Counterions derives from the metal coagulants, which hydrolyze when they are added to water, forming metal hydrolysis species. The type of hydrolysis species which are formed, depends on factors like pH and concentration of anions in the water(Vepsäläinen and Sillanpää, 2012). Larger particles are then easier to remove through processes as settling, flotation, or filtration.

There are four main coagulation mechanisms in aqueous solutions (Vepsäläinen and Sillanpää, 2012).

- Compression of electrical double layer
- Adsorption destabilisation
- Inter-particle bridging

- Precipitation and enmeshment mechanism

1.3.2 Electrocoagulation

Electrocoagulation is an alternative method to conventional coagulation, using electricity instead of chemical reagents to remove suspended particles and colloids. It eliminates the need to add massive amounts of expensive chemicals, but also reduces storage needs at the treatment plant as well as transportation costs (Dohare and Sisodia, 2014). The technology has been around since the late 1800s. The first unit was established for the treatment of wastewater in London in 1889 (Vepsäläinen and Sillanpää, 2012).

Advantages and disadvantages

The technology is renowned for its ability to treat industrial wastewater by being versatile and environmentally friendly (Dohare and Sisodia, 2014). It offers a coagulation method that eliminates the need for transportation and storage of chemicals, thus making electrocoagulation an emerging option for decentralized water treatment (Holt et al., 2005). Electrocoagulation uses a smaller amount of metal for the same removal efficiency, doses ranging below 120 mg/L (Kuzhel and Nehrii, 2019) while conventional coagulants are ranging between 150 mg/L to 600 mg/L (Guida et al., 2007).

Electrocoagulation is easy to operate and requires a minimal amount of maintenance as the operational set up is simple, with no moving parts (Mollah et al., 2001). This does reduce operational and maintenance costs. The technology can handle a broad variety of water quality and pollutants, and still provide high removal efficiency. It can remove multiple contaminates in one operation, and also work as a disinfectant. However, the sacrificial electrodes have to be changed regularly, since it is dissolved into the wastewater during the treatment process (Sillanpää and Shestakova, 2017). There is also a chance of forming an impermeable oxide film on the cathode, leading to loss of efficiency. Both of these aspects may increase operation cost. In areas where electricity is not abundant and reliable, electrocoagulation can be an expensive and unreliable treatment method.

As Mollah et al. (2001) states in their review, by eliminating the addition of chemicals, electrocoagulation produces a smaller amount of sludge than by conventional coagulation. The sludge also tends to be easier to settle and de-water. In addition, there will be a reduced possibility of secondary pollution and a need for neutralization of excess coagulants. These properties make it easier to reuse the generated sludge from the treatment process.

Removal efficiency and specific pollutants removed by electrocoagulation is well researched on a laboratory scale. Design, mechanisms, combination with other processes and full-scale operation have received less attention. This gives an uncertainty to the long term and full-scale operation of electrocoagulation, and may be one of the main reasons the technology is not more widespread used (Moussa et al., 2017).

Theory of electrocoagulation

Electrocoagulation is based on the generation of coagulant in situ by using a sacrificial anode. An appropriate anode material, usually aluminum or iron, is electrolytically oxidized adding charged ions to the wastewater leading to particle suspension and breaking of emulsions. Charged ions will react with opposite charged ionic species suspended in the water. The method can remove metals, colloidal solids and particles and soluble inorganic pollutants from different aqueous media. These mechanisms are similar to conventional coagulation, but require a smaller amount of coagulant to achieve the same removal efficiency (Mollah et al., 2001).

Metal is dissolved at the anode as following for respectably iron and aluminum.

$$Fe(s) \rightarrow Fe^{n+}(aq) + ne^{-} \tag{1.1}$$

$$Al(s) \rightarrow Al^{3+}(aq) + 3e^{-} \tag{1.2}$$

Iron can dissolve into two different forms, divalent *Fe(II)* or trivalent *Fe(III)*. Following *Fe(II)* can oxidize to *Fe(III)* if the reduction potential and pH is in a certain range. aluminum can only be dissolved into trivalent *Al(III)*.

The dissolved metal can then react with the water. *Me* represent both aluminum and iron

$$Me^{m}(aq) + nH_2O \leftrightarrow Me(OH)^{m+n} + nH^{+}(aq) \tag{1.3}$$

At the cathode hydrogen-gas is formed through the reduction of water.

$$2H_2O + 2e^{-} \rightarrow H_2(g) + 2OH^{-}(aq) \tag{1.4}$$

The formation of gas at the cathode works as a flotation method, separating the sludge from the aqueous solution. Hydroxide formed in equation 1.4 reduces the pH of the solution.

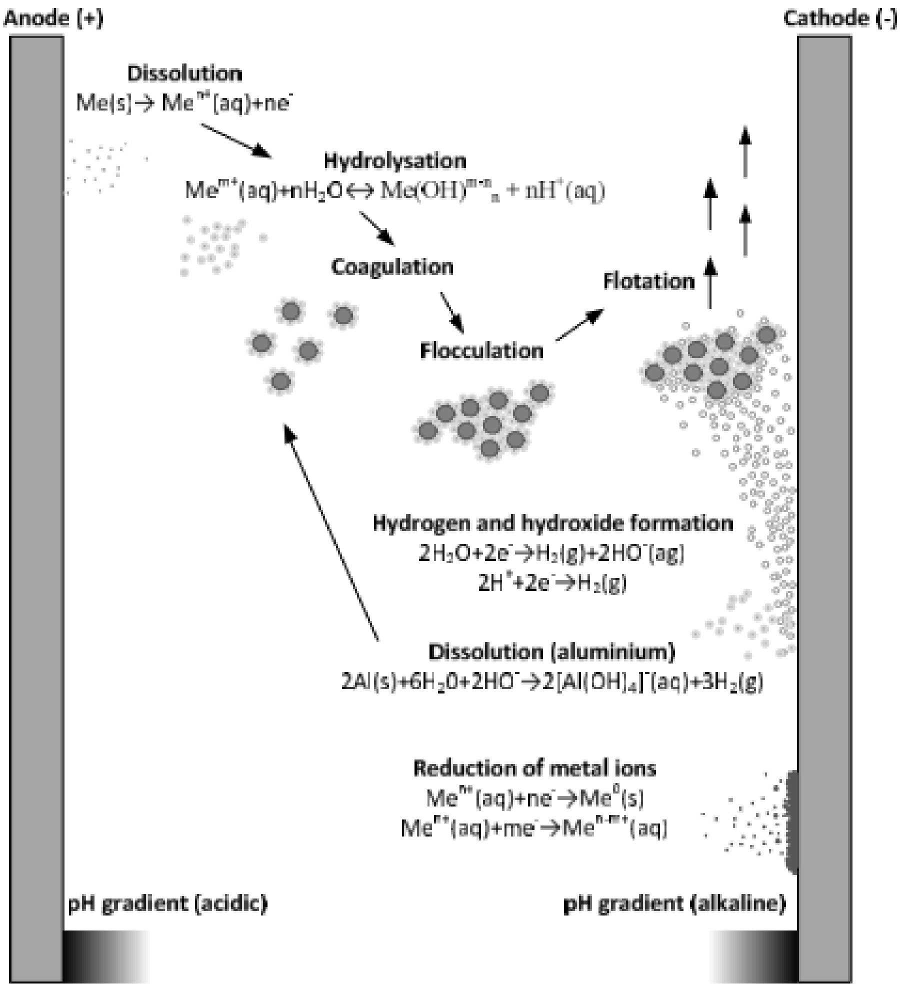

Figure 1.1: Schematic overview of main reactions in electrocoagulation (Vepsäläinen and Sillanpää, 2012)

Estimation of coagulant doses

Estimation of the coagulant doses generated by the electrocoagulation process can be calculated using Faraday's law (Sillanpää and Shestakova, 2017).

The amount of generated coagulant can be calculated with:

$$m = k \cdot i \cdot t \tag{1.5}$$

where

k electrochemical equivalent of coagulant $g/A \cdot s$

i current A

t time s

The electrochemical equivalent of the coagulant, k, can be calculated by:

$$k = \frac{M}{Q \cdot z} \tag{1.6}$$

where

M molar mass of coagulant g/mol

Q Faraday constant $A \cdot s/mol$

z number of electrons involved in the reaction

Estimated coagulant doses are highly theoretical. Picard et al. (2000) studied cathodic dissolution with an aluminum and a stainless steel electrode. Their research suggests that coagulant dissolve from both the cathode and the anode when an aluminum electrode is used. Resulting in a dissolution of over 100% of the theoretical value. Due to the increased dissolution of aluminum, a higher formation of hydrogen was observed. Both dissolution of aluminum and following, hydrogen formation, increased exponentially with higher current intensities. With the iron electrode, dissolution rates followed Faraday's law.

Treatment parameters

Several operational parameters affect the treatment efficiency of electrocoagulation. Vepsäläinen and Sillanpää (2012) present the most important parameters as

- **Electrode material**

  Electrode material is an essential factor as it defines which ions and metal hydroxides are formed at the anode during electrocoagulation. Usually, either iron or aluminum is used, in some cases inert material as cathodes. Therefore different electrode materials will have distinct applications and treatment efficiency at various aqueous media.

- **pH**

  Electrocoagulation can work at various pH as active coagulation species can be formed at alkaline, neutral and acidic conditions. However, treatment efficiency does depend on pH as it affects the formation of hydroxide species, the conductivity of the solution, electrode dissolution, and the ζ-potential of colloidal particles in aqueous solutions. Optimal operation pH depends on electrode material and target pollutants. pH of the solution will also increase during electrocoagulation, the amount largely depending on treatment time.

- **Current density**

Current density is defined as

$$J = \frac{I}{A} \tag{1.7}$$

where

J Current density A/m^2

I Current A

A Cross sectional area m^2

Current density has a direct correlation to the amount of electrochemical reactions that are taking place at the electrode surface, impacting the amount of coagulant dissolved from the anode.

- Treatment time

  Treatment time has an impact on the amount of coagulant, which dissolves into the aqueous solution. Therefore, treatment time does have a significant influence on treatment e□ciency.

- Temperature

  Higher temperatures contribute to less forming of sediments on the electrode surface and do facilitate the dissolution of passivization films. Temperature will a□ect the dissolution rate of electrodes, depending on the material.

Other then these five, several other parameters will have an impact on treatment e□ciency, electrode potential, concentration of pollutants, and concentration of anions.

1.4 Related work - Overview of existing reviews

There is written numerous reviews on electrocoagulation. An overview of current reviews can be found in table 1.2. A majority of these reviews do not cover a specific topic in electrocoagulation, but rather give a general overview of the technology. Nine of the reviews are focused on the removal of one or several specific pollutants, while the rest focus on mechanics, optimization of the process or reactor design.

This review includes design parameters, such as electrode distance, electrode configuration and unit design. These are not broadly review yet. It is also seen in a perspective to Norwegian laws and regulations.

Table 1.2: Overview of existing reviews on electrocoagulation

Author	Published	Name	Scope
An et al. (2017)	Science of the Total Environment	Emerging usage of electrocoagulation technology for oil removal from wastewater: A review	Oil removal
Bharath et al. (2018)	International Journal of ChemTech Research	A Review of Electrocoagulation Process for Wastewater Treatment	General
Butler et al. (2011)	International Journal of ChemTech Research	Electrocoagulation in Wastewater Treatment	General
Chellam and Sari (2016)	Water	Aluminum electrocoagulation as pretreatment during microfiltration of surface water containing NOM: A review of fouling, NOM, DBP, and virus control	Fouling, NOM, DBP, virus
Chen (2004)	Separation and Purification Technology	Electrochemical technologies in wastewater treatment	General
Chen et al. (2020)	Chemosphere	Quantitative contribution study and comparison between electrocoagulation, anode-electrocoagulation and chemical coagulation using polymer-flooding sewage	Polymer flooding sewage
Emamjomeh and Sivakumar (2009)	Journal of Environmental Management	Review of pollutants removed by electrocoagulation and electrocoagulation/flotation processes	Pollutants removed
Feng et al. (2016)	Environmental Science: Water Research and Technology	Electrochemical technologies for wastewater treatment and resource reclamation	Pollutant removal, resource reclamation
Garcia-Segura et al. (2017)	Journal of Electroanalytical Chemistry	Electrocoagulation and advanced electrocoagulation processes: A general review about the fundamentals, emerging applications and its association with other technologies	General
Ghernaout et al. (2019)	Journal of Environmental Science and Allied Research	Electrocoagulation Process: A Mechanistic Review at the Dawn of its Modeling	Mechanisms
Ghernaout (2019)	Journal of Environmental Science and Allied Research	Virus Removal by Electrocoagulation and Electrooxidation: New Findings and Future Trends	Virus
Hakizimana et al. (2017)	Desalination	Electrocoagulation process in water treatment: A review of electrocoagulation modeling approaches	Modeling approaches
Holt et al. (2005)	Chemosphere	The future for electrocoagulation as a localised water treatment technology	Reactor design/operation
Kabdaşlı et al. (2012)	Environmental Technology Reviews	Electrocoagulation applications for industrial wastewaters: a critical review	Industrial wastewater application
Kobya et al. (2020)	Environmental Technology and Innovation	A review on decontamination of arsenic-contained water by electrocoagulation: Reactor configurations and operating cost along with removal mechanisms	Arsenic removal
Mollah et al. (2001)	Journal of Hazardous Materials	Electrocoagulation (EC)- Science and applications	General
Mollah et al. (2004)	Journal of Hazardous Materials	Fundamentals, present and future perspectives of electrocoagulation	Optimisation
Moussa et al. (2017)	Journal of Environmental Management	A comprehensive review of electrocoagulation for water treatment: Potentials and challenges	General
Sadik (2019)	Advances in Chemical Engineering and Science	A Review of Promising Electrocoagulation Technology for the Treatment of Wastewater	General
Sahu et al. (2014)	Environmental Science and Pollution Research	Treatment of wastewater by electrocoagulation: A review	General
Song et al. (2017)	Chemical Engineering Journal	Electrocoagulation treatment of arsenic in wastewaters: A comprehensive review	Arsenic removal

1.5 Scope and research questions

The initial plan for this thesis was to conduct laboratory studies of electrocoagulation and electrocoagulation as a part of a pilot plant. Because of the outbreak of SARS-Cov-2 in Norway, all universities were closed from March 12th. Thus, it was not possible to finish the planed laboratory experiments. The scope of this thesis was therefore changed to a literature review in the end of March.

Appendix A of this thesis consists of laboratory trials conducted before March 12th, and is a part of the initial scope of this thesis.

The purpose of this thesis is to review the effects of interventions in electrocoagulation. To do so, three relevant research question has been developed.

- How can the applications of electrocoagulation be classified by wastewater source/type, industry, technological scheme etc.?
- What are the wastewater treatment effects of electrocoagulation in different applications?
- What are the existing designs of electrocoagulation units and which electrochemical mechanisms are involved in their functioning?

2. Methods

In this chapter, methods used to conduct the literature review are presented. The literature review has been structured according to the Cochrane Handbook for Systematic Reviews of interventions by Higgis et al. (2019) and the PRISMA Statement by Moher et al. (2009).

2.1 Eligibility criteria

To write a literature review many references have to be evaluated. References have to be on a certain level to ensure a proficient quality. Therefor eligibility criteria for the references are set to evaluate all references before they are included in the review.

The eligibility criteria evaluated are

- Relevance to topic and match to research questions
- Originality
- Year published
- Impact factor of journal published in

The information flow through the different phases of the review has been organized with the PRISMA flow diagram by Moher et al. (2009).

2.2 Information sources

For the reference search electronic databases had been the main source of literature, this includes Research Gate, Science Direct, Mendeley, and Google Scholar. A literature search was performed at the start of the review, for two weeks, in March and April 2020.

After the two initial weeks of literature search, additional references were found in the reference list of already included literature.

2.3 Search strategy

As there still has been done relatively limited research on electrocoagulation there is also a limited amount of literature. Therefore only "electrocoagulation" was used as a search word on all databases. No other limits were applied during the initial literature search.

On some topics, the initial literature search did not provide enough resources. In these cases, a more specific literature search was conducted. It was performed with the same method as described earlier, but a topic-specific search word was added after electrocoagulation. For example "electrocoagulation", "textile wastewater".

2.4 Study records

Below the exact method used to find, screen and evaluate resources is described, using already presented information in chapter 2.1, 2.2 and 2.3.

2.4.1 Data management

To manage records and data Mendeley is primarily used. Mendeley provides reference management, which is easy to use for searching specific topics and making notes. For some references there is done a data collection in spreadsheet form, using excel. This is mainly used for overview purposes and comparing information between different literature.

2.4.2 Selection process

The first selection process, screening, was done when adding references from information sources to Mendeley. As described above a broad search was performed at several information sources. Initially all literature containing "electrocoagulation" in the title or abstract were added to the Mendeley library. This initial search resulted in a library of more than 100 references.

References have then been evaluated with regards to the evaluation criteria in section 2.1 on-the-go, as it have been relevant to include them in the literature review. The criteria are designed to filter out irrelevant and outdated information, as well as research that is not performed at an adequate level. Older literature is included when there is still consensus about the information presented in more recent research. The focus has mainly been on different types of wastewater treatment, but literature on surface water, groundwater, and drinking water has been included when the research has been found especially relevant to the topic.

For literature which pass the evaluation criteria, there is performed a brief literature search through their reference list to expand the resource library.

2.4.3 Study selection

This section describes screened and included literature by numbers, and main reasons for exclusion.

Overview of reviews

For table 1.2, 26 reviews were screened. Two reviews were excluded due to lack of relevance on the topic. The 24 remaining reviews were assessed for eligibility. Of the 24, 3 references were excluded for various reasons. Resulting in including 21 references in this part.

Review

For the literature review 191 articles were screened and 79 unique articles included into the review. Main reason for excluding articles were lack of originality, to similar information and results compared to already included articles, and outdated information.

3. Results

This chapter is a review of the effects of interventions in electrocoagulation. The goal of this review is to answer the research questions presented in section 1.5.

- How can the applications of electrocoagulation be classified by wastewater source/type, industry, technological scheme etc.?
- What are the wastewater treatment effects of electrocoagulation in different applications?
- What are the existing designs of electrocoagulation units and which electrochemical mechanisms are involved in their functioning?

Following is a broad spectrum of current literature on elctrocoagulation presented. It is divided into four separate parts; applications, technological schemes, treatment effects, and design and operation.

3.1 Applications of electrocoagulation

Electrocoagulation can be used to treat a wide range of surface water, groundwater, and wastewater. The technology is gaining popularity but is still not commonly used. This section presents applications of electrocoagulation in wastewater treatment. Below, the five most mentioned wastewater types, in context with electrocoagulation, is included.

3.1.1 Domestic wastewater

Domestic wastewater is used water from office buildings, households, industry, and businesses; in many cases, this also includes urban runoff in areas with combined sewer (Read, 1997).

Ensano et al. (2019) researched removal of conventional parameters as chemical oxygen demand (COD), dissolved organic carbon (DOC), phosphates, nitrates and organic matter from domestic wastewater as well as pharmaceutical residue. They concluded that electrocoagulation is an efficient treatment method for domestic wastewater to remove both conventional parameters and bio pollutants. The removal efficiency increased with current density and treatment time.

Devlin et al. (2019) compared treatment efficiency of electrocoagulation with aluminum, iron, and magnesium electrodes to conventional coagulation with metal salts. They found that for iron and aluminum removal efficiency was as good as conventional coagulation with equal metal doses. Magnesium electrodes were less efficient, removing more than seven times fewer orthophosphates than iron and aluminum at optimum conditions.

Tian et al. (2017) and Tian et al. (2018) investigated removal of phosphates from domestic wastewater. In both studies 100% removal were obtained with two different electrocoagulation reactors.

3.1.2 Wastewater from the oil industry and mining

Electrocoagulation has been especially popular in the oil, gas, and mining industry to treat produced water for reuse (Capocelli, 2015). According to Allision and Mandler (2018), 2% of the annual water use in the US is in the mining industry, including gas and oil extraction. In the oil industry, they use water to lubricate and cool the drill and remove debris like mud and rock. The produced water from these industries contains oil residue, salts, chemicals from fracking or drilling, heavy metals, and natural contaminates from rocks (Capocelli, 2015). Electrocoagulation destabilizes oil emulsion by charge neutralization, leading to bonding of particles creating bigger flocs that are easier to separate (An et al., 2017), making it possible to remove up to 99% of the oil emulsions (Bian et al., 2019).

Electrocoagulation is also known to be efficient in removing heavy metals (Mouedhen et al., 2008) and salts (Bian et al., 2019). Chen et al. (2020) reported a treatment efficiency of 80 % removal of oil emulsion by treating polymer-flooding sewage from tertiary oil recovery technology with electrocoagulation. El-Naas et al. (2009) researched removal of sulfates and COD from petroleum refinery wastewater, achieving removal of 93% and 63%, respectively. Aluminum electrodes achieved higher removal efficiency than iron or steel electrodes. Jing et al. (2020) experienced high removal efficiency when treating mineral processing wastewater by electrocoagulation with iron electrodes. They obtained 82.8 % removal of COD with optimum conditions.

3.1.3 Textile industry wastewater

Wastewater from the textile industry contains high concentrations of dyes, suspended solids, organic matter, and chemicals (Sadik, 2019). Biological treatment and conventional coagulation have shown poor results in treating the textile wastewater containing high color and COD levels, as well as high pH (Thakur and Chauhan, 2018).

Singh et al. (2019) investigated the removal of COD and color from simulated textile water containing malachite green dye. They used a dual-stage electrocoagulation process with aluminum electrodes. The process, followed by a settler obtained 92.6 % removal of COD and 93.11 % removal of color with a current density of 165 A/m^2. Khorram and Fallah (2018) researched the treatment of textile wastewater using aluminum electrodes. At optimum conditions, they achieved a 97% removal of color and 40% removal of COD. The removal e□ciency of COD is low because of high levels of dissolved additives such as sodium oxalate, polyvinyl alcohol, and acetic acid in the wastewater.

Aygun et al. (2019) researched the treatment of reactive dyebath wastewater with aluminum and iron electrodes. Removing 92.0 % color and 80.9 % COD for iron electrodes, and 85.8 % for color and 76.9% for aluminum at optimum conditions. Energy consumption for iron electrodes were slightly less than for aluminum, 1.56 € $/m^3$ and 1.84 € $/m^3$, respectively. Due to the high initial pH of textile wastewater, HCl was used to reduce the pH before treatment. The operational cost is as following; 2% energy consumption, 28% electrode, and 70 % chemicals. Energy consumption is insignificant compered to total treatment costs.

Merzouk et al. (2010) applied a alectrocoagulation-electroflotation process. Aluminum electrodes were able to remove 79.7 % COD, 85.5 % suspended solids, 76.2 % turbidity and >93% color with optimum conditions. Afanga et al. (2020) found similar results when comparing several di□erent electrochemical combinations. Electrocoagulation-electroflotation combination had the highest removal e□ciency, with notable better results than electrocoagulation alone.

3.1.4 Food industry wastewater

Water is an important part of producing food in terms of irrigation, as a part of the raw materials or directly used as an ingredient (Kirby et al., 2003). Sardari et al. (2018) research the application of electrocoagulation in treating poultry processing wastewater. Wastewater from poultry production usually contains higher levels of fats, oil and grease, COD, biological oxygen demand (BOD) and suspended solids than regular domestic wastewater. Using aluminum electrodes, they were able to achieve up to 94% removal of fat, oil and grease, 87% of BOD, 59% of COD, and 84% of total suspended solids. Bayar et al. (2011) obtained a removal e□ciency of 85 % COD and 98 % turbidity at optimum conditions using aluminum electrodes. Kobya et al. (2006) compared aluminum and iron electrodes. Aluminum electrodes obtained the highest removal e□ciency for COD with 93 %, while iron electrodes obtained the best removal e□ciency for oil and grease with 98%.

Asselin et al. (2008) investigated the removal of organic compounds from slaughterhouse wastewater. Steel electrode obtained the highest removal efficiency. With optimum conditions they were able to remove 86±1% BOD, 99±1% oil and grease, 82±2% COD, 89 ±4% suspended solids and 90 ±4% turbidity.

The treatment of wastewater from dairy production was investigated by Tchamango et al. (2010). Electrocoagulation with following filtration performed as good as conventional coagulation, removing 61 % COD, 89 % phosphorus, 81 % nitrogen and 100 % turbidity. However, effluent treated with electrocoagulation had lower conductivity and a neutral pH in contrast to conventional coagulation. Lactose removal was insignificant, which may be the reason for the relatively low removal of COD. Bassala et al. (2017) found similar results removing 80 % COD, 98 % phosphorus and 100 % turbidity from dairy wastewater. Akansha et al. (2020) obtained 86.4 % removal of COD using a combination of iron and aluminum electrodes.

3.1.5 Pulp and paper mill wastewater

Pulp and paper mill processing requires water in several stages of the process. The effluent from this process contains substances as wood debris and soluble wood material, as well as chemicals and bleaching agents (Pokhrel and Viraraghavan, 2004). Pandey and Thakur (2020) investigated the treatment effect and sludge quality of recycled fiber paper mill wastewater treated by electrocoagulation. They obtained a removal efficiency of 95 % COD and 67 % color. The generated sludge showed good compaction characteristics for utilization and easy disposal. Wagle et al. (2020) researched the removal of tannin/lignin, organic compounds from plants and trees, and color. Using a sacrificial iron electrode and low current densities, they achieved >70% removal of tannin/lignin and >90% removal of color. Electrocoagulation is an energy and cost-effective option for Kraft paper mill wastewater. Sridhar et al. (2011) concluded that electrocoagulation is suitable to treat bleaching plant effluent for reuse. They obtained 94%, 90% and 87% removal efficiency for color, COD and BOD, respectively.

Camcioglu et al. (2017) compared the treatment effect of aluminum and iron electrodes for COD, color, turbidity, and suspended solids removal and the energy consumption. They found that aluminum electrodes were more efficient than iron, and had a lower energy consumption. Barhoumi et al. (2019) investigated the removal of Humic acid from synthetic paper mill wastewater. Optimum operating conditions gave 93% removal efficiency of Humic acid. Following, they performed experiments with electrocoagulation combined with adsorption by active carbon. Achieving the same removal efficiency, they could reduce treatment time from 10 min to 2 min, and the energy consumption was significantly lower. Thus, emerging as a promising technology for the treatment of pulp and paper mill wastewater at a reduced cost. Jaafarzadeh et al. (2016) combined electrocoagulation and UV to remove organic pollutants. Electrocoagulation alone was able to remove 61% COD, but did not affect biodegradability (BOD/COD ratio). In combination with UV, there was a significant increase in biodegradability.

3.1.6 Summary

Electrocoagulation has proven to be an effective treatment method for several different wastewater types. There is consensus that electrocoagulation is an efficient technology for treating wastewater from the oil industry and mining. The technology has been applied in full scale and operated over more extended periods successfully.

It is a promising technology for treating domestic, food industry, textile and, pulp and paper mill wastewater. There is evidence that electrocoagulation can provide high treatment efficiencies for these wastewater types. However, there is not presented much literature on full-scale and long term operation.

3.2 Technological schemes with electrocoagulation

A treatment process is applied in combination with other processes or alone. A combination of processes can be determined by the influent quality and the effluent regulations. This is described further in section 1.3. This section highlights research on how electrocoagulation is applied as different parts of the treatment process.

3.2.1 Pre-treatment

Pre-treatment is the first step in wastewater treatment. The goal of this step is to remove anything that might interfere with subsequent treatment. This is important when using for example membranes. Membranes are prone to scaling and fouling, which leads to decrease in permeate flux. Therefore Millar et al. (2014) evaluated if pre-treatment by electrocoagulation could reduce these problems when treating coal seam water with reverse osmosis. Coal seam water contains high levels of bicarbonate, chloride, sodium, and fluoride (Rebello et al., 2017). Electrocoagulation were able to remove components responsible for scaling, 100% calcium, 87.9% magnesium, 99.3% strontium, 100% barium and 98.3% silicates. Fluoride and boron removal was not as efficient, removing 44% and 13.3%, respectively. Poor removal of these was likely due to high pH. da Silva et al. (2015) investigated electrocoagulation as pre-treatment for reverse osmosis as well. They applied the technology to oily wastewater. Electrocoagulation with aluminum electrodes provided high removal efficiencies, and did reduce fouling. The permeate flux was reduced less than 10% over the experiment. Thus, working well as pre-treatment for

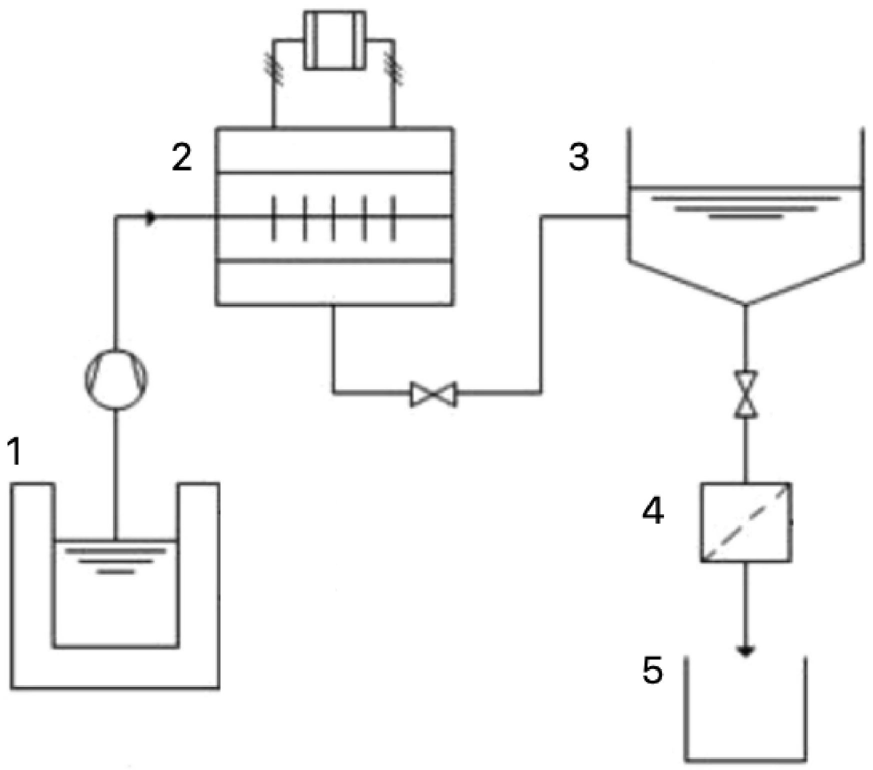

Figure 3.1: Simplified flowsheet presenting the process used by da Silva et al. (2015). 1. Wastewater tank, 2. Electrocoagulation, 3. Sedimentation tank, 4. Reverse osmosis, 5. Sludge tank

oily wastewater treated by reverse osmosis. Sefatjoo et al. (2020) found similar results. Hybrid electrocoagulation-filtration as pre-treatment improved reverse osmosis recovery with 25% compared to only filtration as pre-treatment.

Isik et al. (2020) investigated electrocoagulation and electroflotation as pre-treatment for pistachio processing wastewater treated with fungal. Aluminum, iron, and stainless steel electrodes were used for electrocoagulation. Stainless steel provided the highest reduction in COD, while aluminum electrodes gave the best removal of phenol. However, compared electroflotation was a better option as pre-treatment. Electrocoagulation inhibited fungal growth with solubilized Cr and Ni ions, reducing the total treatment effect.

Bagga et al. (2008) researched if electrocoagulation with iron electrodes could work as pre-treatment for microfiltration of surface water. They concluded that iron electrocoagulation is not applicable as pre-treatment before microfiltration. The process generates more soluble Fe^{2+} than insoluble Fe^{3+}, resulting in only small improvements on fouling characteristics compared to untreated water.

3.2.2 Main treatment process

There is a limited amount of literature that actively focus on electrocoagulation as the main treatment process, even though researchers may have this as their intention. Research on this is not included if their purpose is not clearly stated.

In the oil industry and mining, electrocoagulation has been used as the main treatment process. Electrocoagulation have proved high removal efficiencies in removing oil emulsions and salts (Bian et al., 2019), COD (Jing et al., 2020), sulfates (El-Naas et al., 2009), and heavy metals (Mouedhen et al., 2008). The effluent achieves sufficient quality for reuse in the industry.

Decentralized water treatment plays a vital role in many rural areas. Electrocoagulation is a promising technology for this since it is easy to operate, also remotely, and does not need delivery or storage of chemicals (Holt et al., 2005). St-Onge et al. (2020) investigated if electrocoagulation can be used as primary treatment of surface water in Africa, powered by solar panels for areas without a regular power supply. They were able to remove 92,1 % of turbidity from domestic wastewater, collected after secondary treatment, using solar panels as power supply, without batteries or charge controllers. Treatment efficiency directly correlates with solar radiation intensity. The study suggests that it is feasible to use solar electrocoagulation for decentralized water treatment. Holt et al. (2005) came to the same conclusion after batch experiments and modeling electrocoagulation behavior.

3.2.3 Post-treatment

Post-treatment is the last treatment step before discharge. Zodi et al. (2011) investigated electrocoagulation as post-treatment following a grid for solids removal, a primary settling treatment, and secondary biological treatment by activated sludge. They aim to remove non-biodegradable organic pollutants and arsenic from paper mill wastewater. Experiments show a reduction in COD and DOC up to 68% and 46%, respectively. There was a decrease in the aromaticity of organic pollution and lignin related pollution, as well as a fair decrease in arsenic. Overall, aluminum electrodes obtained the best removal efficiency, while sludge characteristics were better with iron electrodes.

Nguyen et al. (2014) used electrocoagulation as post-treatment for domestic wastewater from a hybrid biological reactor. Using only the hybrid biological reactor, the mean value for phosphorus was 2 mg/L, which were over the guidelines of 0.2 mg/L. By applying electrocoagulation as a post-treatment step, phosphorus residue in the effluent decreased to a mean value of 0.11 mg/L. Total removal efficiency for phosphorus was 97.23–100% over the 171 operation days with natural fluctuations in the influent concentration.

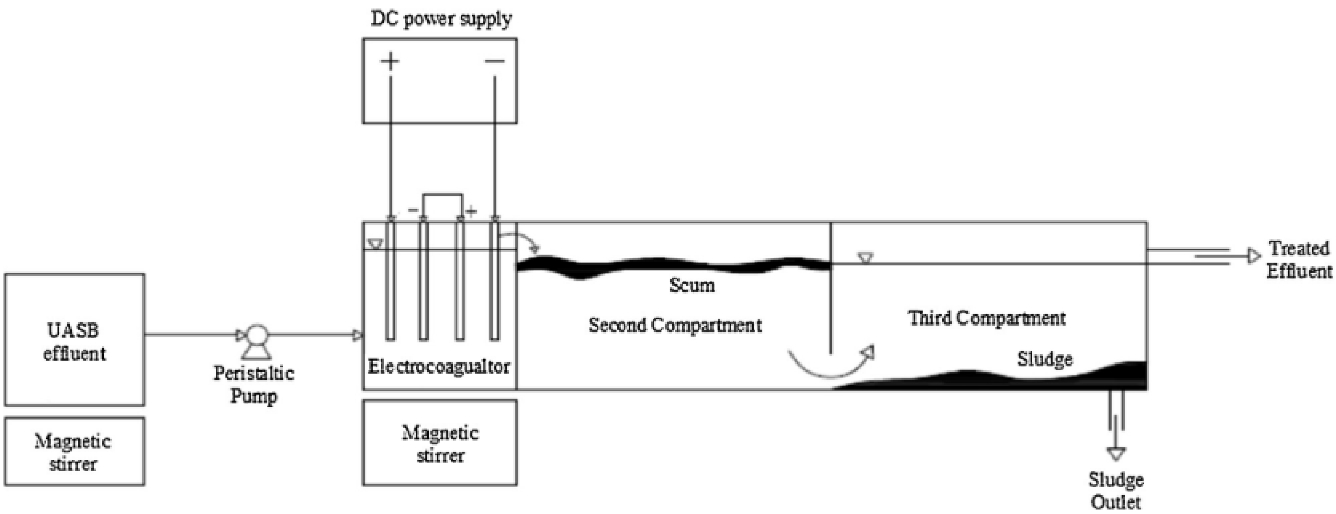

Figure 3.2: Flowsheet of the treatment process applied by Makwana and Ahammed (2016)

Makwana and Ahammed (2016) researched the potential of using electrocoagulation as post-treatment for domestic wastewater, after an up-flow anaerobic sludge blanket reactor(UASB). They operated electrocoagulation as continuous flow with aluminum electrodes. At optimum conditions, the e□uent does comply with Indian regulations for COD, BOD, phosphates, pH, and suspended solids levels in domestic wastewater e□uent. They also observed a >99.8 % reduction of total and fecal coliforms.

Tsioptsias et al. (2015) successfully applied electrocoagulation as post-treatment for biologically treated molasses wastewater. Molasses wastewater is e□uent from the food industry, as distillery or sugar and yeast production. It is usually dark in color, and contains large amounts of substances of intractable nature. From experiments, they obtained 97% color removal and up to 60% COD and nitrogen removal. There is a linear correlation between COD and color removal. Electrodes made of iron and iron filings were compared and found to be as e□ective. Iron filings electrodes is a low-cost alternative to iron electrodes.

3.2.4 Summary

Electrocoaguation with aluminum electrodes is found to reduce fouling and scaling on membranes. It is therefore a good alternative as pre-treatment before membranes and filters. It can be used as the main treatment process for some applications, depending on e□uent requirements. There is clear evidence that it works well on in oil industry and mining. It also shows promising results in surface water. Electrocoagulation as main treatment do have limited literature available. As post-treatment is suitable to use after biological treatment. Electrocoagulation do have a high removal e□ciency, even when initial pollutant concentration is low.

3.3 Treatment effects of electrocoagulation

One of the most critical factors in the development of a new water treatment technology is the treatment efficiency of different pollutants. Emamjomeh and Sivakumar (2009) concludes that electrocoagulation can remove a variety of unwanted dissolved particles and suspended matter from aqueous solutions. A summary of the removal efficiency found in the literature introduced in this section is presented in table 3.1.

Table 3.1: Overview of the treatment effects of electrocoagulation

Pollutant	Highest removal efficiency	Water type	Author
Broad spectrum antibiotics (CIP)	100%	Hospital	Ahmadzadeh et al. (2017)
	100%	Hospital	Yoosefian et al. (2017)
Vetrinary antibiotics	3.1 - 100 %	Domestic	Baran et al. (2018)
Theraputic drugs	50%	Domestic	Ensano et al. (2019)
COD	51%	Artificial	Nguyen (2020)
	60.5%	Landfill leachate	Mallesh (2019)
	62.64%	Oleochemical	Azli and Azoddein (2020)
	82.8%	Mineral processing	Jing et al. (2020)
	91%	Oily	Safari et al. (2016)
	92%	Industial	Yavuz and Ögütveren (2018)
	94.3%	Metalworking	Kobya et al. (2019)
Phosphates	98%	Domestic	Tian et al. (2017)
	98%	Domestic	Tian et al. (2018)
	99%	Domestic	Omwene and Kobya (2018)
	99%	Domestic, surface	Franco et al. (2017)
	100%	River	Attour et al. (2014)
Suspended solids	66.12%	Oleochemical	Azli and Azoddein (2020)
	98%	Textile	Naje et al. (2016)
	98%	Textile	Afanga et al. (2020)
Nitrogen	21%	Domestic	Devlin et al. (2019)
	60%	Molasses	Tsioptsias et al. (2015)
	81%	Dairy	Tchamango et al. (2010)
Turbidity	96%	Textile	Naje et al. (2016)
	98%	Domestic	Bracher et al. (2020)
	99.52%	Surface	Afiatun et al. (2019)
Color	93.11%	Textile	Singh et al. (2019)
	92%	Reactive dybath	Aygun et al. (2019)
	99%	Printing ink	Papadopoulos et al. (2019)
Virus and bacteria	99.997%	Artificial	Zhu et al. (2005)

3.3.1 Contaminants of emerging concern

Ahmadzadeh et al. (2017) investigated the removal of Ciprofloxacin (CIP) from hospital wastewater using electrocoagulation with aluminum electrodes. CIP is a broad-spectrum antibiotic. Their experimental research obtained a removal efficiency of 88.57 % with an initial concentration of 32.5 mg/L. For their analyses using real wastewater with an initial concentration of $154 \pm 6 \mu g/L$, there was zero CIP left after treatment.

Yoosefian et al. (2017) found similar results removing CIP using iron electrodes. With optimal conditions and an initial concentration of 60 mg/L, they achieved 100 % removal. Experiments with real hospital wastewater also had removal of 100 % with an initial concentration of $381 \pm 4 \mu g/L$.

Baran et al. (2018) investigated the removal of efficiency of ampicillin, doxycycline, sulfathiazole, and tylosin from wastewater. These are antibiotics used in veterinary medicine. They used a steel electrode, wastewater, and distilled water with added Na_2SO_4 for control purposes. Results show that ampicillin, sulfathiazole and tylosin have a low removal efficiency of $3,6 \pm 3,2$%, $3,3 \pm 0,4$% and $3,1 \pm 0,3$%, respectively. Doxycycline, on the other hand, is removed 100 %.

The removal of therapeutic drugs with electrocoagulation was researched by Ensano et al. (2019). They investigated the removal of diclofenac, carbamazepine, and amoxicillin from domestic wastewater. They chose these drugs as they regularly are found in wastewater. For all drugs, they achieved removal efficiency between 40-50 % using a continuous power supply. They performed the same experiment using an intermittent power supply with 5 min on and 20 min off intervals. Removal efficiency declined with approximately 15 %, while energy consumption declined 96% using the same operational parameters.

3.3.2 COD

Removal of COD is an integral part of wastewater treatment. Yavuz and Ögütveren (2018) were able to remove 92% of the initial COD using iron electrodes. Jing et al. (2020) found similar results using iron electrodes on mineral processing wastewater. Under optimal conditions, they obtained 82.8 % removal.

Nguyen (2020) performed experiments with artificial wastewater and aluminum electrodes. In optimized conditions, they were able to remove 51% of the COD from the solution. Azli and Azoddein (2020) achieved a removal rate of 62.64% treating oleochemical wastewater. Safari et al. (2016) also used aluminum electrodes when they investigated the removal efficiency from oily wastewater, achieving removal rates of $91 \pm 0,2$% for COD. Both Nguyen (2020) and Safari et al. (2016) found that removal efficiency was highly dependent on the initial pH of the aqueous solution. Mallesh (2019) investigated the treatment of landfill leachate with electrocoagulation with aluminum electrodes. Optimum conditions gave a removal rate of 60.5 %.

Kobya et al. (2019) performed experiments with both iron and aluminum electrodes. Experiments were performed on metalworking wastewater with a continuous electrocoagulation reactor. For aluminum electrodes optimum of EC time are 50 min, flow rate of 0.010 L/min, and current density 90 A/m^2 achieved 92.6% removal efficiency. While for iron electrodes, optimum conditions were 40 min treatment time, flow rate of 0.05 L/min, and current density 90 A/m^2. This configuration removed 94.3 % COD. These results favored iron electrodes in terms of treatment efficiency and operating cost.

3.3.3 Phosphates

Electrocoagulation has shown to be efficient in the removal of phosphates from aqueous media (Emamjomeh and Sivakumar, 2009). Franco et al. (2017) showed that electrocoagulation with aluminum electrodes could remove up to 99% of phosphates in surface water and wastewater with a treatment time below 60 minutes. These results were achieved with pH ranging from 5 to 8.8. The removal efficiency was mainly dependent on the power applied and the conductivity of the solution. Attour et al. (2014) used aluminum electrodes in a batch reactor to remove phosphorus from river water. With optimal operating conditions, phosphorus was removed.

Omwene and Kobya (2018) studied phosphorus removal through electrocoagulation in a batch reactor with both aluminum and iron electrodes. Their research was focused on comparing the two materials for phosphorus removal. With both electrode materials, they investigated when the concentration of phosphates went below 0.01 mg/L. From their experiments, they found that aluminum electrodes provided a higher removal efficiency in a shorter period than iron electrodes. The metal-to-phosphorus ratio was lower for aluminum than iron.

Tian et al. (2017) investigated the removal of phosphates by electrocoagulation with polarity change using an inert graphite electrode and aluminum sacrificial anode. They found that with polarity change of 5 seconds, 90±1% were removed after 10 minutes. For 10 seconds, the results were 95±2%. More extended polarity change periods did not increase the treatment efficiency further. Control experiments with a reactor without inert electrode were performed and showed removal of 67±2% after 5 minutes and 87±1% after 10 minutes. After 30 minutes, the removal was 98±1%, proving that longer treatment time increases removal efficiency, when not using an inert electrode. Tian et al. (2018) performed similar experiments using an iron anode, and inert titanium electrode with a migration electric-field assisted electrocoagulation system. With iron anode, they achieved a removal of 98% with polarity change of 10 seconds and <6 minutes treatment time. Without conventional electrocoagulation, 15 minutes of treatment was required to obtain the same removal efficiency. With the same system, they also tested an aluminum anode. For the same phosphorus removal, 7 minute treatment time was necessary. For conventional electrocoagulation, 20 minutes of treatment time was needed to achieve 98% removal.

3.3.4 Suspended solids

Naje et al. (2016) researched the removal of suspended solids from textile wastewater using a rotating anode electrocoagulation unit. With only 10 min reaction time, they achieved 98% removal. Afanga et al. (2020) achieved 98% removal of suspended solids from textile wastewater in a combination electrocoagulation-electroflotation process. Azli and Azoddein (2020) found a significantly lower treatment efficiency using aluminum electrodes at optimum conditions treating oleochemical wastewater. Highest achieved treatment efficiency was 66.12%.

3.3.5 Nitrogen

Devlin et al. (2019) compared iron, aluminum, and magnesium electrodes for treating domestic wastewater. They achieved the highest removal of nitrogen using magnesium electrodes. Magnesium reduced nitrogen content with 21% at optimum conditions. Iron and aluminum achieved removal efficiencies around 10%. Tsioptsias et al. (2015) treated molasses wastewater with high nitrogen content using iron and copper electrodes. This method was able to remove between 50-60% of the total nitrogen. Tchamango et al. (2010) were able to remove up to 81% of the nitrogen from dairy effluent by an aluminum anode.

3.3.6 Turbidity

Afiatun et al. (2019) investigated the reduction of turbidity from surface water. By using aluminum electrodes, their highest achieved removal efficiency was 99.52 %. Bracher et al. (2020) used aluminum electrodes to treat domestic wastewater for reuse. Optimum conditions resulted in 98% removal. Turbidity in the effluent was measured to be below 6 NTU, which complies with Brazilian standards and guidelines for reuse. Naje et al. (2016) were able to remove up to 96 % turbidity from textile wastewater with a rotating aluminum anode.

3.3.7 Color

Removal of color were researched by Papadopoulos et al. (2019). They treated printing ink wastewater with aluminum and iron electrodes. Aluminum electrodes obtained the highest removal efficiency of 99 %, while iron electrodes were the most cost-efficient alternative. Aygun et al. (2019) achieved similar results, removing 92% color with aluminum electrodes compared to 85.8% using iron electrodes. Singh et al. (2019) obtained 93.11% removal of color from textile wastewater using a dual-stage electrocoagulation with aluminum electrodes.

3.3.8 Viruses and bacteria

Removal of viruses and bacteria is especially necessary when treating drinking water and water for reuse. Zhu et al. (2005) studied the removal of MS2 bacteriophage using electrocoagulation with iron electrodes as pretreatment, followed by microfiltration. While the use of microfiltration alone gave less than a 0,5-log removal (32%), They achieved more than 4-log removal (99.997%) with electrocoagulation as pretreatment. Electrocoagulation as pretreatment outperformed conventional coagulation on all dosages and pH levels used in this research.

Electrocoagulation reduces Cyanobacteria levels in water. The combination of coagulation, flotation by gas bubbles, and disinfection by electrochemical species, e□ciently reduce the amount present concluded Monasterio et al. (2014). A combination of the highest current density and lowest flow rate applied in these experiments gave the highest removal e□ciency.

3.3.9 Summary

Electrocoagulation is a suitable method to remove a broad spectrum of pollutants present in di□erent wastewater types. There is evidence that the technology does achieve high removal e□ciency for suspended solids, phosphates, color, and turbidity. The removal e□ciency of COD with electrocoagulation is highly dependent on initial pH, and therefore provides inconsistent removal rate. Electrocoagulation is e□cient in removing some contaminants of emerging concern, but can not be viewed as an adequate treatment method for all contaminants. There is not enough evidence to conclude on the treatment e□ect on viruses, bacteria and nitrogen.

3.4 Designs and operation principles of electrocoagulation treatment units

Today there is not one dominant design of electrocoagulation units. Numerous di□erent designs are used for di□erent applications on laboratory, pilot and industrial scale. There is is a lack of research done on systematic reactor design as well as comparative design studies (Holt et al., 2005). Therefore this chapter will focus on *what are the existing designs of electrocoagulation units and which electrochemical mechanisms are involved in their functioning?*

Electrocoagulation is dependent on several operating parameters as pH, conductivity, current density, treatment time, and temperature. These parameters are further explained in section 1.3.2. This part will highlight important parameters in the design of

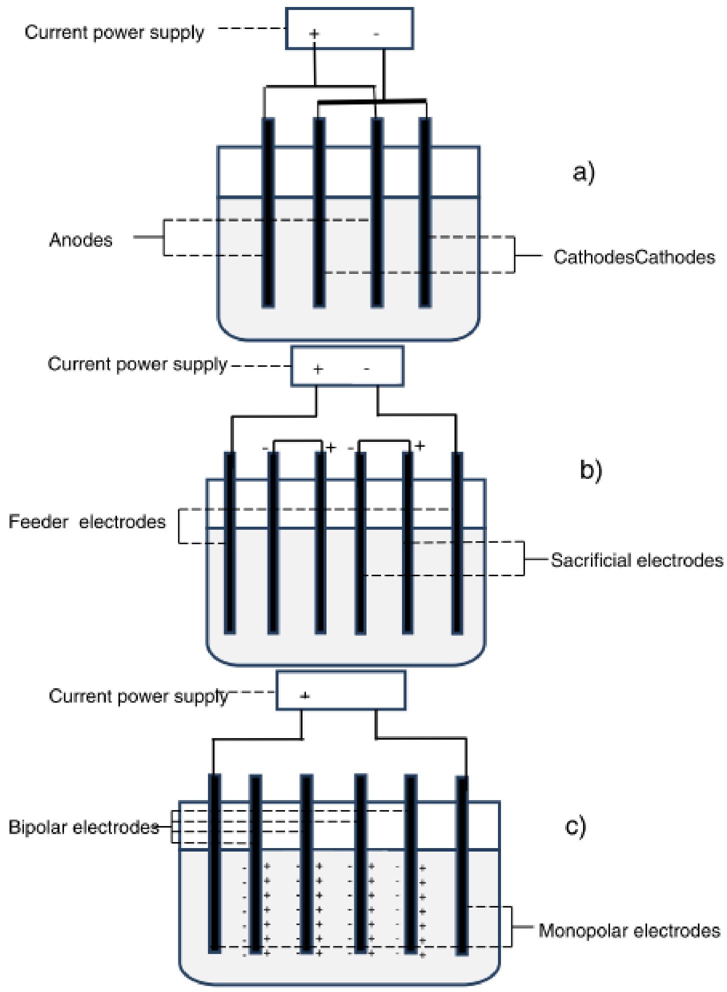

Figure 3.3: a) Monopolar electrodes in parallel b) Monopolar electrodes in series c) Bipolar electrodes in series Source: Hakizimana et al. (2017)

the unit, rather than operating parameters.

3.4.1 Electrode design

Electrode geometry and arrangement plays an essential role in the treatment e□ciency of an electrocoagulation unit. Hakizimana et al. (2017) presents three main electrode arrangements, presented in figure 3.3. The first arrangement is monopolar electrodes in parallel connection. This arrangement consists of anodes and cathodes placed alternatively. They all have the same anodic/cathodic potential, which leads to each pair creating an electrolytic cell with the same voltage. The second arrangement is monopolar electrodes in series. Sacrificial electrodes are placed in pairs which are internally connected. They are not connected to the two outer electrodes. This arrangement gives an equal current through all electrodes. The last arrangement is bipolar electrodes in series. Two outer electrodes are connected to the power supply, while the other electrodes are bipolar and can work as both cathodes and anodes.

Khaled et al. (2019) compared the performance of monopolar electrodes in parallel and bipolar electrodes in series. Their research found that electrode configuration did not affect aluminum electrode consumption when only electrode configuration was changed. However, bipolar electrodes had a higher removal efficiency, but also a higher energy consumption than the monopolar electrodes in parallel. This is a result of more anodic oxidation with bipolar configuration. The increased energy consumption is due to the long distance between electrodes connected to the power supply, increasing the IR-drop. IR-drop is further explained in section 3.4.2. Bipolar configuration also increased the pH and temperature of the effluent more than monopolar configuration. Cruz et al. (2019) found similar results regarding treatment efficiency when comparing monopolar electrodes in parallel and bipolar electrodes in series.

As well as electrode arrangement, electrode geometry also plays a role in treatment efficiency. Cruz et al. (2019) compared electrodes formed as plates and rods, as well as bipolar and monopolar electrode arrangements. They found that plates performed better than rods with both monopolar and bipolar configuration. Aljaberi (2019) also research different electrode geometries. He found a correlation between electrode geometry and energy consumption, thus impacting the operation cost of the electrocoagulation unit.

3.4.2 Electrode distance

Distance between the electrodes influences the IR-drop, thus affecting the magnitude of current and following the energy consumption (Sillanpää and Shestakova, 2017). IR-drop is the drop in potential energy due to the resistance of the aqueous solution. According to Ohms law, the ohmic potential is

$$\Delta E_{ohm} = -IR \tag{3.1}$$

where

- I Current
- R Ohmic resistance

The ohmic resistance is defined as

$$R = \frac{l}{\rho A} \tag{3.2}$$

where

- l Interelctrode distance

ρ Solution conductance

A Working electrode area

Attour et al. (2014) reported that a reduction of the interelectrode distance shows an increase in the treatment efficiency of phosphates. Shorter distances give a reduced ohmic resistance and a smaller IR-drop. Similar results were found by Huda et al. (2017) and Merzouk et al. (2010) for reducing color and turbidity, respectively. Khaled et al. (2019) found that reducing the electrode distance from 2 cm to 0.5 cm increased the removal efficiency of cadmium by almost 10%, as well as reducing treatment cost and energy consumption. Martínez-Villafañe et al. (2009) experimented with three different interelectrode distances for removal of arsenic from underground water. Reducing the interelectrode distance increased the turbulence between electrodes. Turbulence influence the reaction rate of the electrocoagulation process. An increase in the distance gave slower movement, reducing the reaction rate and increasing treatment time. However, if the gap between electrodes is too narrow, fluid and solids transfer is obstructed, and the removal rate reduced (Sridhar et al., 2011).

3.4.3 Surface to volume ratio

Surface to volume (S/V) ratio is the relationship between the electrode area and the volume of treated liquid. Studies have shown that when the S/V ratio increase, the optimal current density decrease, which does affect treatment cost(Mameri et al., 1998). A larger S/V ratio provides a higher treatment efficiency at a lower treatment cost due to better dissolution of electrodes, according to Khaled et al. (2019). An increase promote electrical transport, ensure higher dissolution of aluminum and increase the resistance of the electrochemical cell. Martínez-Villafañe et al. (2009) found that the consumption of electrodes increases significantly when the S/V ratio decrease. This design parameter is considered one of the most important in regards to scale up (Hansen et al., 2007).

3.4.4 Reactor design

The design of the reactor itself regarding geometry influences important operation parameters as bubble path and size, flow regime, floc formation, and flotation/settling characteristics (Hansen et al., 2007). Today there is no standard design of electrocoagulation units, but there are several typical designs. Open rectangular units are common, as well as cylindrical (Sillanpää and Shestakova, 2017).

Holt et al. (2005) describes how reactor design practice can be divided into three core design decisions, batch or continuous operation, the role played by electrolytically generated bubbles, and the means of separating the aggregated pollutant. Batch operation is operated with a fixed treatment volume per treatment cycle. With batch operation conditions, change over treatment time as an additional coagulant is added. Continuous operation units have a continuous flow of wastewater through the unit. Operating at close to steady-state conditions. Separation can be done by flotation, settling or filtration. The amount of electrolytically generated bubbles is influenced by current density. High current density increase gas formation, working as flotation. Low current density reduces gas formation, and in consequence, favors settling.

Hansen et al. (2007) tested three different electrocoagulation units to compare their performance in removing arsenic from wastewater. The units were a modified flow sedimentation basin, turbulent flow reactor, and batch airlift reactor. The modified flow sedimentation basin used steel electrodes, while the two other used iron electrodes. All units had different volumes, electrode configuration, S/V ratio, and flow. The modified flow and the airlift reactor obtained similar results at a current density of 1.2 A/dm^2, removing over 98% arsenic. The turbulent flow reactor obtained a lower removal efficiency than the two others. Iron to arsenic ratio was lower, due to higher levels of arsenic in the inflow. Thus, reducing removal efficiency. However, all three units showed efficient removal despite their different configuration. They concluded that the most important parameters were S/V ratio and to avoid anode passivization. Anode passivization can be reduced by applying current reversal or salt addition.

3.4.5 Summary

There is conses that electrode distance, electrode configuration and surface to volume ratio is important parameters for removal efficiency and operation cost of electrocoagulation units. It is likely that also the geometry of electrodes do have an effect on these aspects. There is not one design that researchers can prove to be more cost efficient or provide higher treatment effects than others. There is however, an agreement that unit geometry and design impact several aspects of wastewater treatment with electrocoagulation.

4. Discussion

This chapter discusses the results found in chapter 3 with regards to theory and the research questions. Electrocoagulation has been applied to numerous wastewater types, with significant differences in composition. There have been most research on and full-scale use for the treatment of industrial wastewater.

Domestic wastewater does typically contain high levels of phosphorus, which is one of the pollutants electrocoagulation shows the highest and most stable removal efficiencies. Electrocoagulation also provides good removal of color, suspended solids, and turbidity. Removal efficiencies are ranging above 80% for most configurations. For the treatment of domestic wastewater, also removal of COD, nitrogen, virus, and bacteria is an important parameter. Removal of COD by electrocoagulation is highly dependent on water composition and operational parameters. This fact can be observed in table 3.1, as the removal efficiency is in a wide range from 40% by Ensano et al. (2019) to 94,3% by Kobya et al. (2019). These fluctuations can be observed even within the treatment of similar wastewater types. Several researchers concluded that COD removal is dependent on the initial pH of the wastewater. There is a limited amount of research on the removal of nitrogen, virus, and bacteria by electrocoagulation. Current research shows promising results on the removal of viruses and bacteria and the use of electrocoagulation as a disinfectant mechanism. Current research on nitrogen removal show inconsistent results, where for some wastewater types performance is good (80%)(Tchamango et al., 2010), and for others poor (10%)(Devlin et al., 2019). To conclude on the treatment efficiency of nitrogen, virus, and bacteria by electrocoagulation, more research has to be performed. Because of these characteristics and current treatment requirements, domestic wastewater treatment with electrocoagulation has to be in combination with other treatment processes. Combinations have so far received limited research. However, recent studies have found electrocoagulation to be efficient as post-treatment after biological treatment. Electrocoagulation does provide high removal efficiency, even when initial concentration is low, and handles fluctuations in influent concentration well. In these characteristics, electrocoagulation performs better than conventional coagulation. Biological treatment has high removal efficiency for nitrogen and COD. Thus, biological treatment combined with electrocoagulation emerges as a good combination, which should receive more research.

Domestic wastewater can contain high levels of emerging contaminants. Electrocoagulation shows promising results in removing emerging contaminants as Ciprofloxacin and Doxycycline. For these two antibiotics 100 % removal efficiency were obtained with several different operation configurations (Ahmadzadeh et al., 2017; Yoosefian et al., 2017; Baran et al., 2018). While for other emerging contaminants, there is a small to no reduction in concentration after treatment. Mechanisms that remove contaminants of emerging concern should receive more attention to quantify which contaminants can be removed by electrocoagulation.

Electrocoagulation has been successfully applied in full scale for the oil industry and mining to treat produced water for reuse. In this application, electrocoagulation has been used as a primary treatment. Electrocoagulation efficiently separates emulsified oil, petroleum hydrocarbons, suspended solids, heavy metals from effluents. It is providing high removal rates on-site. Reverse-osmosis is also a suitable treatment method for oily wastewater but has challenges with fouling and scaling, reducing the permeate flux. Electrocoagulation with aluminum electrodes as pre-treatment has proven to reduce fouling and scaling, increasing the life span and reducing maintenance costs of membranes. Electrocoagulation has proven to be more effective than conventional coagulation to reduce fouling and scaling.

Textile industry wastewater has obtained low removal efficiency with conventional treatment methods. Electrocoagulation is a promising technology for this application, proving high removal efficiencies in small scale experiments. Therefore, electrocoagulation is an emerging application for this type of wastewater. A combination of electrocoagulation and electroflotation seems to provide higher removal efficiencies than electrocoagulation alone.

Food industry and pulp and paper mill wastewater both contain large amounts of organic components and suspended solids, which electrocoagulation does remove efficiently. Wastewater from the food industry also contains high levels of oil and grease, which electrocoagulation is proven to remove successfully. Laboratory scaled experiments have proven effective for these to wastewater types.

While operational parameters have been thoroughly researched and tested, the design of electrocoagulation units has not. Generally, there is a lack of research on design, and existing research is hard to compare. Bipolar electrodes performers better in terms of removal efficiency, while monopolar electrodes are more cost-efficient than bipolar. Better removal efficiency is caused by higher anodic oxidation with bipolar configuration.

Monopolar configuration, on the other hand, uses less electric power to obtain the same removal efficiency as a bipolar configuration. Reduced need for power happens due to a smaller IR-drop. As seen in 3.3, the distance between electrodes directly connected to the power supply, is smaller in a) than c). Shortening the interelectrode distance reduces the drop in potential energy due to resistance in the aqueous media. A longer distance requires a higher current density to obtain the same removal efficiency as the reaction rate and turbulence decrease. Thus, they also need a higher voltage, which increases treatment costs.

Surface to volume ratio is concluded by several researchers to be the most crucial parameters when scaling up electrocoagulation units (Holt et al., 2005; Hansen et al., 2007). The parameter is important because of its effect on treatment cost. A small S/V ratio requires a higher current density to dissolve the same amount of metals as a larger S/V ratio. A large S/V ratio also promotes electrical transportation and reduce the consumption of electrodes as dissolution improves. With the high consumption of electrodes, they have to be changed more frequently - increasing operation costs.

The design of reactors has not been adequately researched to conclude on a optimal design. There is, on the other hand, some critical parameters. These parameters are batch or continuous operation, and the role played by generated gas, and how pollutants are separated from the water. The role played by the generated gas bubbles, and separation method is closely connected. Hydrogen gas formation is dependent on current density, increasing the current density also increase gas formation, and opposite. There is a possibility to utilize the gas formation to ensure flotation or settling in the electrocoagulation unit, reducing the need for a secondary separation process.

5. Conclusions

Ensuring access to water and sanitation for all is one of the UNs goals of sustainable development towards 2030. Today, the lack of safe access contributes to thousands of deaths every day and increase the pollution of the environment. In this thesis, applications, treatment effects, technological schemes, and design parameters of electrocoagulation has been evaluated according to current literature.

Electrocoagulation has been applied in the treatment of domestic, oil industry and mining, textile, food production, and pulp and paper mill wastewater. It has advanced the most in oil and mining because of its high removal efficiency of relevant pollutants, easy operation, and is a low-cost alternative. For the oil industry and mining, electrocoagulation alone provides sufficient treatment for reuse. This industry is also the only one where electrocoagulation has been applied in full-scale and operated for a more extended period. In the treatment of domestic, textile, food production, and pulp and paper mill wastewater, research is performed on a laboratory scale and in a shorter time. There is, however, good agreement that electrocoagulation could be useful in treating these wastewater types. This is especially the high removal of phosphorus from domestic wastewater and good removal efficiencies for textile wastewater, which conventional methods can not provide.

There is high evidence that electrocoagulation can efficiently be applied as pre-treatment before membranes and post-treatment after biological. So far, there is limited research on electrocoagulation as the primary treatment process. Most promising is electrocoagulation before membranes. These properties has in several studies, proved to reduce scaling and fouling on membranes. Improving the treatment efficiency of membranes and reducing maintenance and operating costs. As post-treatment, there is high evidence that electrocoagulation still provides high removal efficiencies, when the initial concentration of pollutants is low. It also handles fluctuations in pollutant concentration well.

Several pollutants can achieve high removal efficiencies with electrocoagulation. There is evidence that phosphates can be removed up to 90-100%, color 85-99%, turbidity 96-

99%, suspended solids 66-98%, and COD 51-94%. Results for contaminants of emerging concern show that some can be removed (ciprofloxacin, doxycycline), while other drugs obtain little to no removal with electrocoagulation. There is little evidence on nitrogen removal efficiencies as there are few studies, and those which exist have different results. There is evidence that electrocoagulation can work as a disinfection of viruses and bacteria. However, this area needs more research.

Critical design parameters in electrocoagulation is electrode distance, electrode configuration, and surface to volume ratio. There is evidence that these parameters are crucial to the removal efficiency and operational costs of electrocoagulation. There is not enough evidence to conclude on a favorable design of units.

5.1 Future work

In the future, the industry should focus on larger-scale and more extended period studies, to test the good results found in laboratory-scale experiments. This research is especially important for the textile industry, where current treatment methods are not a good option.

Future research should be focused on design and utilizing bubble formation. These two are essential to be able to make electrocoagulation more cost-efficient and commercialized.

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Appendix A. Laboratory trials

The first part of this thesis consists of laboratory trials performed on a pilot plant and raw water. The amount of the experiments was drastically reduced from what was planned due to the situation regarding SARS-CoV-2. Therefor only experiments on activated sludge test of raw water were completed.

A.1 Background

A.1.1 Biological

Treatment of wastewater by biological processes rely on bacteria and microorganisms to consume organic substances. The goal is to make the treatment tank work as a system that decomposes organic substances into sludge and clump together into bigger flocs, which can Quickly be removed (Odegaard, 2012).

Biological treatment can be used both anaerobic and aerobic, or as a combination of the two to obtain a su□cient removal of the target pollutants (Odegaard, 2012).

Activated sludge

Activated sludge consists of suspended biological flocs in an aerated tank(Odegaard, 2012). Aerobic microorganisms digest organic matter and form flocs in the process. The liquid left after the flocs are removed is relatively free from suspended particles and organic matter.

An activates sludge system usually consists of a tank containing mixed liquor, which is aerated with oxygen or air. Following the aeration tank is a mechanism to separate the sludge from the liquid - usually a settling tank.

A.1.2 Membrane filtration

Membrane filtration is a physical treatment process separating particular substances from water. The process does not alter the substances thermally, biologically or chemically. Pore size determine the purification efficiency of the membrane ranging from 0.1 μm in microfiltration to 0.001 μm in reverse osmosis (Pinnekamp and Friedrich, 2003). Because of the small pores, transport through the membrane is pressure-driven.

Membranes has a compact design, requiring a smaller area than processes with similar treatment effect (Pinnekamp and Friedrich, 2003). Generally, membranes have a high treatment efficiency with complete retention of particles.

Ceramic membranes

In wastewater treatment, ceramic membranes are widely used. The inorganic material works with a broad spectrum of pH and temperature due to its high resilience to heat and chemicals (Liu et al., 2019). Therefore, making it possible to clean the membranes with strong chemicals, to remove fouling. The membrane will not encounter irreversible structural changes that may affect the filtration capacity(Liu et al., 2019). These characteristics give ceramic membranes a longer life span than other inorganic membranes (Pinnekamp and Friedrich, 2003).

Membrane fouling

One of the main challenges using membranes in wastewater treatment is membrane fouling. Domestic wastewater contains both organic and inorganic substances that are restrained by the membrane. With operating time this will develop a covering layer. The covering layer will increase the filtration resistance, which reduces the permeate flow through the membrane. Increased resistance is a result of adsorption, pore blockage, and covering layer formation (Pinnekamp and Friedrich, 2003).

Membrane fouling can be divided into three categories determined by the cause of the formation of the covering layer. These three groups are biological fouling, colloidal fouling and scaling (Pinnekamp and Friedrich, 2003). Biological fouling is the growth of microorganisms at the membrane, causing a biofilm layer to decrease the membrane flow (Chang et al., 2019). The accumulation of colloids at the membrane surface, colloidal fouling, lead to the forming of a film at the membrane surface, reducing the filtration capacity (Pinnekamp and Friedrich, 2003). The last category is scaling, which is a result of inorganic precipitation forming a coating on the membrane (Pinnekamp and Friedrich, 2003).

A.1.3 Membrane bioreactor

Membrane bioreactors (MBR) is a combination of biological treatment and membrane filters. The process uses a membrane as separation of sludge in an activated sludge bioreactor. Membranes separate all of the suspended solids from the aqueous solution by using membranes with pore openings, usually around 0.04 μm (Odegaard, 2012). There are two main types of MBRs, with submerged membranes or with a separate membrane downstream from the bioreactor.

MBRs provide high treatment efficiency, with good effluent quality. This characteristic makes it highly relevant in areas that practice reuse of wastewater (Odegaard, 2012).

A.2 Materials

In this section, materials used for laboratory trials are described, the pilot plant and raw water collection.

A.2.1 Pilot plant

For these experiments, a pilot plant located at The Norwegian University of Life Sciences was used. The plant receives domestic wastewater from parts of the student accommodation. It consists of three processes, biological, coagulation, and membrane filtration.

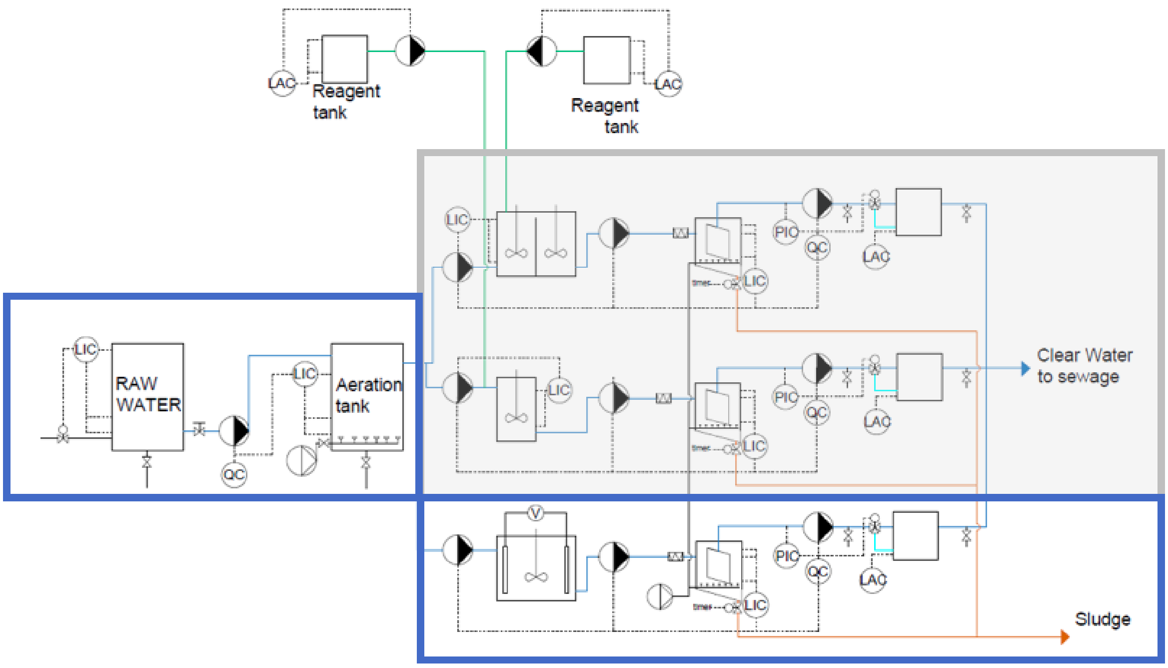

Figure A.1: Pilot plant

Raw water

Raw water is collected from the sewer connected to parts of the student accommodation at the university. To fill the raw water tank, the sewer is partially blocked to raise the water level in the utility hole. A sieve is placed in the sewer to filter out larger particles. Then raw water is pumped to a tank.

Table A.1: Raw water quality

Parameter	TSS (mg/L)	COD unfiltered (mg/L)	COD filtered (mg/L)	TP(mg/L)	Turbidity (NTU)
Amount	510	150.2	891	11.3	260

Biological

The first step in the pilot plant is an activated sludge bioreactor. It consists of an aerated tank with suspended biological flocs. The process is aerobic; therefore, it removes only particulate nitrogen.

The biological unit has a volume of 195 L and an operation volume of about 150 L.

Coagulation

After biological treatment, the flow is divided into three flows, enabling to test three di erent methods of coagulation simultaneously. In this particular project, flow number three, shown in figure A.1, is used for electrocoagulation.

The electrocoagulation unit is constructed as a cylinder consisting of three aluminum tubes, descending in size, placed inside of each other. With this construction tubes are able to work as both cathodes and anodes. Tube one and three are connected to a power supply, which leads to a flow of current from tube one and three to tube two, as shown in figure A.2. The power supply provides a direct current. It is connected to a computer, which controls current, period change of the cathode/anode, and energy consumption.

First and third tube has a wall thickness of 3 mm, and a diameter of 60 mm and 16 mm accordingly. The second tube has a wall thickness of 3 mm and a diameter of 40 mm. Wall thickness of tube two is larger than the others because the current is going from one and three to tube number two; this means that the walls of tube number two are working layers (Kuzhel and Nehrii, 2019).

The unit is connected to a small pump, with a separate power supply, controlling the wastewater flow through the unit.

Membrane filtration

The last step of the pilot plant is membrane filtration, in this plant, a flat sheet silica carbide (SiC) microfiltration membrane is used. The membrane has a pore size of 0,1 μm.

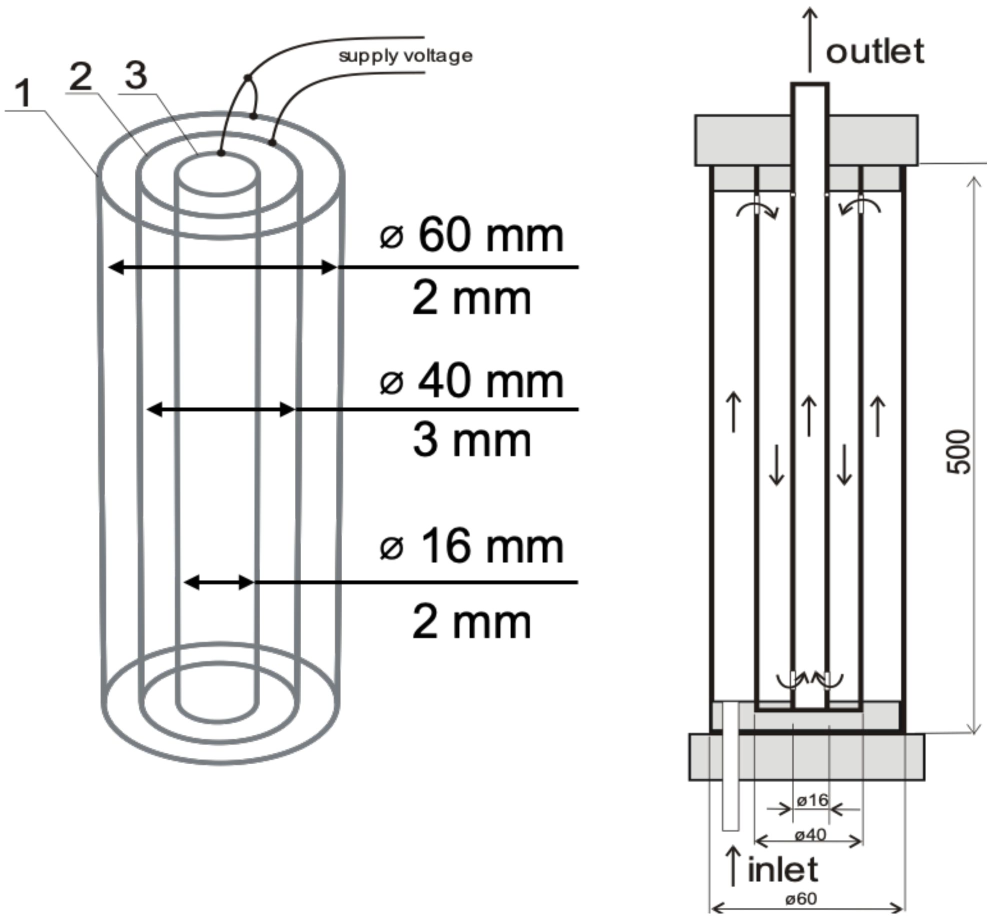

Figure A.2: Electrocoagulation unit

A.3 Methods

In this chapter, measurement methods are explained, and the structure of the experiments presented. The chapter explains how this research is conducted and which methods are used. Chosen methods will affect the results of the research.

A.3.1 Parameters

In order to evaluate the performance of the treatment processes, several parameters of the wastewater is measured at different stages of the treatment process. The difference between influent and effluent gives the treatment efficiency.

Total oxygen demand

Total oxygen demand (TOD) is measured using LAR QuickCODLab. The instrument is calibrated using C8H5KO4 (Potassium hydrogen phthalate). The method is based on thermal oxidation of the sample at a temperature of 1200 Celsius.

Chemical oxygen demand

Chemical oxygen demand (COD) is measured using Hach Lange COD kits in the corresponding range. 2 ml is added to the cuvette, mixed and digested at 148 degrees Celsius for 2 hours. Results are read after at least 30 minutes of cool-down with a spectrophotometer.

pH

pH affects the efficiency and performance of the treatment steps. The treatment steps can also change the pH throughout the process. Therefore this parameter has to be monitored. pH is measured with WTW ProfLine pH meter 3110, which is calibrated with buffer solution every day before use.

Turbidity

Measurements of turbidity is an indirect method to estimate the amount of suspended particles in the solution. Turbidity is measured by sending light through the sample and register the amount that is reflected. Usually, turbidity is measured in NTU.

For measurements of turbidity, Hach 2100N Turbidity Meter is used, with corresponding cuvettes. The cuvettes are kept in deionized water.

Total phosphate

Total phosphate is measured using Systea EasyChem analyser.

TP samples are mixed with potassium peroxide sulfate and concentrated sulphuric acid. Samples are then digested at 120 degrees Celsius for 30 min in an autoclave before they are analyzed.

Suspended solids

Suspended solids (SS) determines the amount of suspended particles in the water. SS is measured by filtering a sample, then drying the filter. The filter is weighed before and after to determine the amount of solids retained by the filter.

A.4 Activated sludge test

The biological process at the pilot plant was first started in December 2019 with sludge from Søndre Follo Renseanlegg. After running the biological plant for six weeks, there was still insufficient removal of COD, below 80%, with a three-day refilling cycle. In order to determine why the treatment was not sufficient several options were investigated.

Chemical oxygen demand to mixed liquor suspended solids ratio

By visual inspection, there was an inadequate amount of sludge in the biological tank, which would make the COD to mixed liquor suspended solids (MLSS) ratio too low, following reducing the removal of COD. MLSS in a biological tank should be around 2-4 g/L (van Loosdrecht et al., 2016).

Sludge quality

In order to obtain removal of COD with biological treatment, microbes in the sludge has to be alive and fed regularly. To start the biological plant, sludge was collected from a nearby treatment plant, which uses biological treatment, December 23rd. Due to the pilot plant tank being damaged, the sludge was filled in a provisional tank and mixed with raw water until January 4th. This inconvenience could lead to reduced sludge quality by microbes dying and resulting in a reduced treatment efficiency.

Raw water quality

van Loosdrecht et al. (2016) states that for microbial growth in biological treatment, an adequate amount of macro- and micronutrients are required for cell synthesis. This includes macronutrients as nitrogen and phosphorus and micronutrients such as potassium, sodium, calcium, and iron, among others. These nutrients are supplied through addition to raw water. Biodegradable organics supplied through raw water are converted into new biomass through anabolism and oxidized to generate energy for biomass growth.

To refill the biological plant, raw water collected in a 1000 L tank was used. The water was collected 5-6 weeks prior to the activated sludge experiments. Quality and the density of nutrients and biodegradable organics of the raw water was a concern as it contained a large amount of greywater. Therefore, less black water, and subsequently less nutrients and biodegradable organics. An insufficient amount would have an impact on treatment efficiency.

A.4.1 Methods

An activated sludge test was performed in order to investigate if there was sufficient COD removal in the biological process. MLSS was measured in accordance with the method described in chapter A.3.1. Experiments were conducted in accordance with “Experimental Methods in Wastewater Treatment” by van Loosdrecht et al. (2016) chapter 2.5.

Sludge was collected from the biological tank and placed in a reactor tank used for the experiments. It was placed with continuous stirring and DO levels above 2 mg/L. After stabilization of pH and DO wastewater substrate were added. Samples were collected every 5 min for the first 30 minutes. After 30 minutes, samples were collected every 10 min. Then after 60 minutes, samples were collected every 15 minutes. Total sampling time was 4 hours. COD tests were conducted immediately after sampling; the rest of the sample was placed in the fridge at a temperature around 4 degrees Celsius until TOD measurements were conducted. pH was measured and adjusted continuously during the experiments to keep a stable value.

Activated sludge tests were performed with raw water and synthetic raw water. This was done in order to eliminate the raw water quality as the reason for inadequate treatment.

The synthetic wastewater was prepared as following per liter.

- 107 mg NH4Cl
- 40 mg Na2HPO4
- 90 mg MgSO4·7H2O
- 14 mg CaCl2·2H2O
- 36 mg KCl
- 1 mg yeast extract
- 0.3 mL of a trace element solution (that includes per liter 10 g EDTA, 1.5 g FeCl3·6H2O, 0.15 gH3BO3, 0.03 g CuSO4·5H2O, 0.12 g MnCl2·4H2O, 0.06 g Na2MoO4·2H2O, 0.12 g ZnSO4·7H2O, 0.18 g KI and 0.15 g CoCl·6H2O)

A.4.2 Results

Activated sludge tests were conducted in three different ways in order to be able to conclude if there was sufficient activity in the sludge to remove the desired amount of COD.

Mixed liquor suspended solids

Mixed liquor suspended solids (MLSS) is the amount of suspended solids in the bioreactor when activated sludge and raw water is mixed. In the biological tank, MLSS was measured to be 1.16 g/L.

Test 1 - real wastewater

The first experiment was conducted using 1.4 liters of raw wastewater and 1.4 liters of activated sludge. Samples were not filtered. After 4 hours, there is no decrease in TOD.

The small increase in TOD seen in figure A.3 most likely occur due to measurement uncertainty.

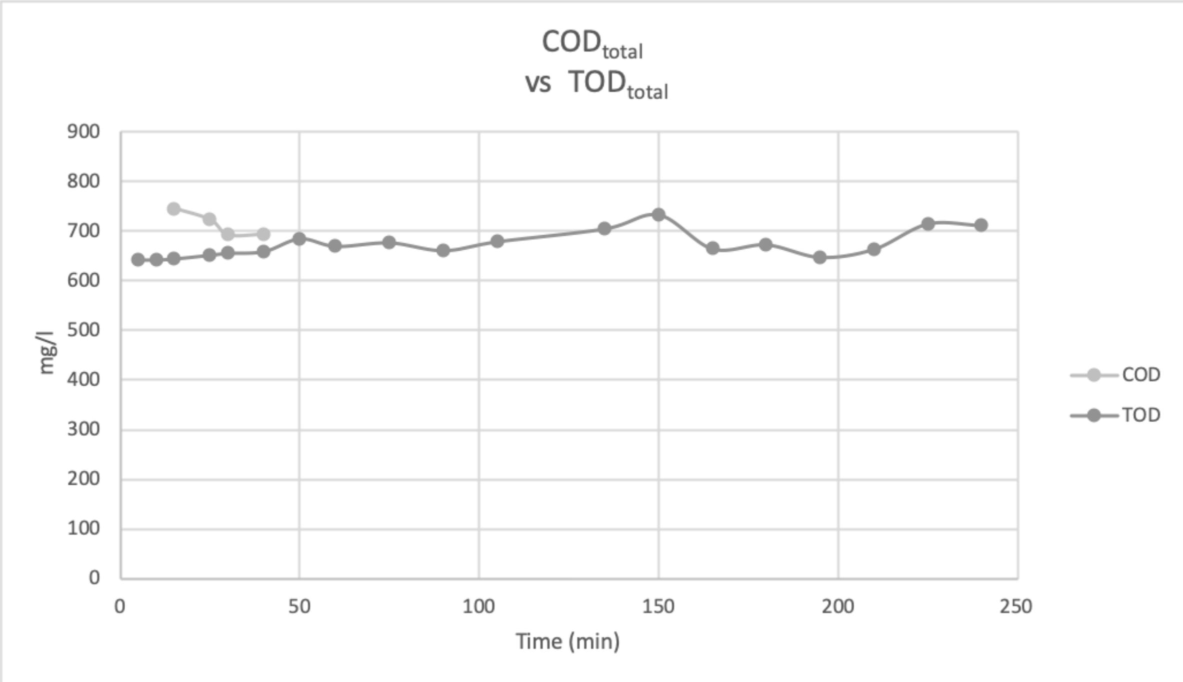

Figure A.3: COD and TOD measurements from the first test

Test 2 - real wastewater

The second experiment was also conducted using 1.4 liters of raw wastewater and 1.4 liters of activated sludge. In this experiment, all samples were filtered immediately after sampling. There is only a small decrease of COD after 1 hour, from 52.9 mg/l to 49 mg/l (7.4%). Removal of COD after 50 minutes was 4.7 %.

Test 3 - synthetic wastewater

For the third experiment, 1.5 liters of activated sludge and 1.5 liters of synthetic wastewater was used. The initial COD concentration of the synthetic wastewater was estimated to be around 400 mg COD/L according to the recipe used for the synthetic wastewater from chapter A.4.

Samples were filtered before measuring COD. After 50 min the removal was 8 mg COD/L (3.4 %) after 4 hours removal was 45 mg COD/L (19%).

A.4.3 Discussion

Measured MLSS in the biological process is 1.16 g/L, way below the recommended amount of 2-4 g/L. Consequently, there is less biomass per g COD added through the raw water, which would lead to reduced treatment e□ciency.

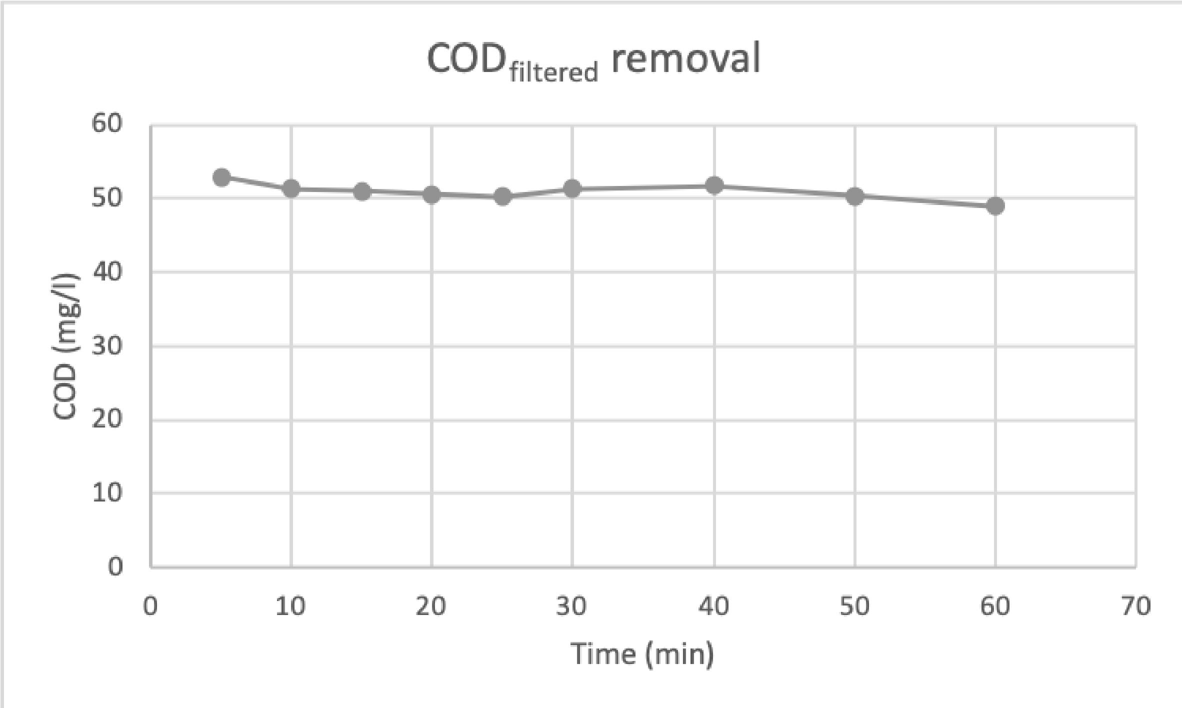

Figure A.4: COD removal in test two

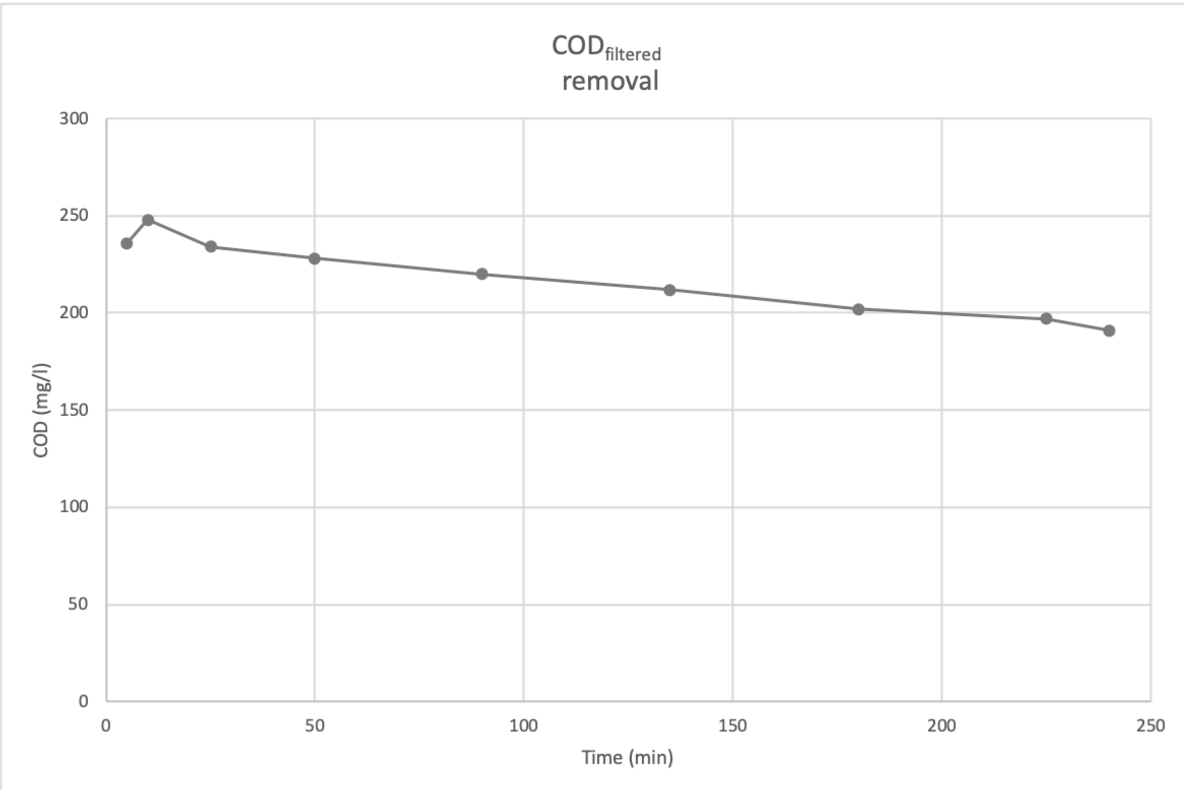

Figure A.5: COD removal with synthetic wastewater

The results show a small to no decrease in COD and TOD levels for the three experiments over the 4 hour experiment time. These numbers are less than expected for the activated sludge experiment, indicating that the process is not operating as it should.

In experiments 2 and 3, COD removal is at 4.7% and 3.4% after 50 minutes. There is an insignificant difference in the removal of COD in the experiment using raw water versus synthetic wastewater. This fact excludes the raw water quality as a non-working parameter.

A.4.4 Conclusion

There is not enough removal of COD in the biological plant due to insufficient sludge quality and a too low biomass to COD ratio. In order to obtain a higher removal of COD, it is recommended to restart the plant with new sludge collected from an operating biological plant.

Norges miljø- og biovitenskapelige universitet
Noregs miljø- og biovitskapelege universitet
Norwegian University of Life Sciences

Postboks 5003
NO-1432 Ås
Norway