

Norwegian University  
of Life Sciences

**Master's Thesis 2019 30 ECTS**

Faculty of Environmental Sciences and Natural Resources Management

# **Effects of electrochemical treatment of mixed liquor in submerged ceramic membrane bioreactor**

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Environmental sciences – Sustainable water and sanitation, health and  
development Master's program

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## Abstract

Our most valuable and irreplaceable resource is water. Humanity withdraws 3.928 km<sup>3</sup> of freshwater per year. Approximately 44% is used for agricultural purposes, the other 56% discharged as wastewater (WW). A large amount of this water does not get adequate treatment and even more, have no treatment at all, which cause problems such as eutrophication, water scarcity, waterborne disease etc. Water reuse and circularity is an alternative and successful way to reduce the environmental problems, and scale of water scarcity issues. Membrane bioreactor (MBR) is one of the technologies which allows implementing the water recirculation approach in real life, but the process of membrane fouling reduces the lifespan of modules and make this type of wastewater treatment very costly. To date, there are many ways to control the membrane fouling were checked, but there is no one universal solution. Numerous advantages of electrocoagulation (EC) make this process interesting for investigation in terms of WW treatment and membrane fouling reduction. In this study, the effect of electrochemical treatment of mixed liquor in submerged ceramic membrane bioreactor applied to municipal wastewater have been evaluated. By using the optimal conditions and dosage of 55.96 mg-Al/L it is possible to achieve the high removal efficiency of PO<sub>4</sub><sup>3-</sup>(up to 99%); TSS (up to 98%); Turbidity (up to 98.5%); TOD (up to 70%) and extend the filtration cycle up to 10 times. It was established, that for the optimal dose of Al, the operating cost (energy and electrode costs) for wastewater treatment is 19.6 NOK/m<sup>3</sup>. It can be concluded that combined IFAS/EC/MBR process of wastewater treatment is highly effective, modern, which make this process closer to water reuse target.

Keywords: Coagulation; Electrode; Electrocoagulation; Fouling; Membrane filtration; IFAS; Wastewater

## Abstrakt

Vår mest verdifulle og uerstattelige ressurs er vann. Mennesker forbruker 3.928 km<sup>3</sup> ferskvann per år. Omtrent 44% brukes til landbruksformål, de andre 56% avgis som avløpsvann. En stor del av dette avløpsvannet renses ikke tilstrekkelig før utløp til naturen, og visse deler renses ikke det hele tatt. Mangelen på rensegrad i avløpsvann forårsaker problemer som eutrofiering, vannknapphet, vannbårne sykdommer osv. Vanngjennbruket og en sirkulær tilnærming til vann er alternative og vellykkede måter å redusere miljøproblemer og omfanget av problemer med vannknapphet. Membranbioreaktor (MBR) er en av teknologiene som gjør det mulig å implementere vannresirkulasjonsmetoden i praksis, men prosessen med membranfouling reduserer levetiden til moduler og gjør denne typen avløpsvann svært kostbar. Per dags dato er det mange måter å kontrollere membranfouling, men det finnes ingen universell løsning. Elektrokoagulasjon (EC) kan vise til flere fordeler og er interessant for undersøkelser når det gjelder avløpsvannbehandling og reduksjon av membranfouling. I denne studien har effekten av elektrokjemisk behandling av blandet væske i en nedsenket keramisk membranbioreaktor som brukes for rensing av kommunalt avløpsvann blitt vurdert. Ved å bruke optimale forhold og 55.96 mg-Al / L dosering er det mulig å oppnå høy rensegrad av PO<sub>4</sub><sup>3-</sup> (opptil 99%); TSS (opptil 98%); Turbiditet (opptil 98.5%); TOD (opptil 70%) og utvide filtreringssyklusen opptil 10 ganger. Det ble etablert at med optimal dose Al er driftskostnadene (energi- og elektrodeknaster) for avløpsvann 19.6 NOK/m<sup>3</sup>. Oppgaven konkluderer at en kombinert IFAS/EC/MBR prosess med avløpsvannbehandling er svært effektiv, moderne, noe som gjør denne prosessen nærmere vannåterbruksmål.

Nøkkelord: Koagulasjon; Elektroder; Elektrokoagulation; Fouling; Membranfiltrering; IFAS; Avløpsvann

## Acknowledgments

We would like to express our deep gratitude to our main supervisor, *Professor Harsha Chandima Ratnaweera*, for his advice, instructions, and at the same time criticism and comments that helped us improve the conditions of our research, self-organization and the experiment in general.

Thanks to our co-supervisor Doctor *Zakhar Maletskyi*, who spent a lot of time with us and helped with the structure of the presentation, the experiment and offered his useful ideas about the thesis.

We want to express special thanks to *Professor Slawomir Kalinowski* from University of Warmia and Mazury in Olsztyn (Poland) for the installation designed specifically for our project. Professor Kalinowski gave us a lot of useful advice and put a lot of effort into the possibility of conducting our experiment.

We would like to express special gratitude to the *AECo project*, which represents cooperation and exchange of experience between Norwegian University of Life Sciences (Ås, Norway), Calgary University (Calgary, Canada) and Columbia University (New York, USA). This project provided an opportunity for Roman, to gain great experience, to exchange ideas and knowledge, as well as to get to know Canada and the USA.

We want to say thanks to *Elisabeth Sundheim Hoff*, who helped us very much in matters of logistics, communication with partners/professors outside of Norwegian University of Life Sciences, and other organization moments.

We want to thank *Dinindu Samadhi Ratnaweera* for his help with the translation of our abstract into Norwegian, for his time and effort.

We would like to thank NMBU and its laboratories, especially Fløy 5, for providing the equipment, reagents and access to them. Also, thanks to *Sven Andreas Högfeldt* who always helped and assisted us in the laboratory, as well as *Vladyslav Shostak*, and his father, who developed and assembled the pilot plant which we use.

We would like to express special gratitude to *our beloved families* for their support, faith and patience.

We are grateful to all those who took any part in our project, in supporting us and our research.

Also, we are very happy that we had the opportunity to work together on a common project, this gave us a chance to invest in this master thesis more persistence, ideas and strength.

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## List of Acronyms

BOD – biological oxygen demand  
COD – chemical oxygen demand  
DI water – distilled water  
EC – electrocoagulation  
EPS – extracellular polymeric substances  
IFAS – integrated fixed film activated sludge process  
MBR – membrane bioreactor  
OC – operating costs  
OP – ortho-phosphates  
RO – reverse osmosis  
SMPs – soluble microbial products  
TMP – transmembrane pressure  
TOD – total oxygen demand  
TRT – filtration cycle length  
TSS – total suspended solids  
WHO – World Health Organization  
WW – wastewater  
WWTP – wastewater treatment plant

## Introduction

The Earth population is rapidly growing. At the time of writing this thesis, the number of people was 7.708.828.977 people, and at the time when you are reading, it is most probably more (“World Population Clock - Worldometers,” 2019). The rapid population growth began after the Industrial Revolution in the 18th century in the Western world. Researchers and scientists explain this phenomenon by a decline of mortality, especially childhood and infant mortality, rising living standards, improved nutritional status. In 2050, it is expected to be about 9 billion. Most population follow a logistic model of growth, not an exponential one, and it is expected to stabilize after achieving the carrying capacity. Most likely, this will happen when the resources that humanity uses for its habitual existence will be exhausted to a high extent. Naturally, as the number of people increases, the need for water, energy, electricity, and resources increases as well (Hussain, 2019).

Our most valuable and irreplaceable resource is water. Even though more than 70% of the Earth surface is covered by water, around 97.5 % of it is salty. This water is unsuitable for domestic, agricultural or industrial use, and most animals and humans consume. Of course, it is possible to remove salt by desalination process, but due to the high cost of this treatment, it is usually used in some emergency cases. Of the remaining 2.5-3 %, most of the water is in the form of glaciers or snow, and only 0.01 percent of freshwater is found in lakes, rivers, atmosphere, groundwater, etc. By and large, the amount of fresh renewable water is several times greater than the total requirements for the healthy development and existence of the planet's population. However, only about 31% of this water is directly available for use through seasonal variations and geographic constraints, which make this source even more precious (WWF, n.d.).

In the case where the rate of water withdrawal from the source of supply does not exceed the rate of its stock recovery, we can talk about the rational consumption of resources. However, in terms of sustainable use, we should also take into account water quality in addition to water quantity balance (Rogers, Llamas, & Martinez-Cortina, 2005).

According to the AQUASTAT database (“AQUASTAT database,” 2019), humanity withdraws 3.928 km<sup>3</sup> of freshwater per year. Approximately 44% of this amount is used for agricultural purposes by direct application such as irrigation etc. The other 56% of water discharged as wastewater to the environment. A large amount of this water does not get adequate treatment, and even more, have no treatment at all. Based on the Sustainable development goals (SDG) report in 2015 around 61% of the global population were without managed sanitation services, furthermore 892 million of people still practiced open defecation (“Goal 6: Sustainable Development Knowledge Platform,” 2018). Looking at the overall picture, one can conclude that humanity is far behind the sustainable management of water resources and wastewater treatment (*Water Scarcity and Drought in the European Union*, 2010).

Water pollutants can be divided into two types nonpoint source and point one. Nonpoint pollution is a result of diffusion distribution from different sources, due to this reason, it is complicated to control and regulate this pollution and, in some cases, it is not possible at all. Nonpoint pollution comes from construction sites, field fertilization, farm runoff, and other results of the daily activities of different people. Single source is a result of a determined source of pollution, such as wastewater treatment plant, industrial waste, etc. This type of pollution can be regulated by a human, so the impact should be minimized primarily. Untreated and consequently, unsafe water leads to many human diseases, global problems, and technical issues (Greenfield, 2016). Unclean water could be a cause of human waterborne diseases. WHO reports that more than 1000 children die daily from diarrhea, over 190 million people live with the risk of trachoma

blindness. Water contamination with chemicals such as hydrocarbons, pesticides, persistent organic pollutants can cause cancer, hormonal problems, DNA damage, and so on. Nutrient pollution by nitrogen and phosphates prompted to accelerate the growth of algae, the leading cause of eutrophication. Algae can produce toxins. These toxins are a particular problem for systems used for drinking water because some toxins can cause outbreaks diseases. The bacterial decay of algae consumes oxygen dissolved in water, creating hypoxia, which endangers the damaging effects on fish and water invertebrates. In addition, phosphorus is a nonrenewable resource, stocks of which will be depleted in the next 50 years, which makes this source very important to reuse (Distefano & Kelly, 2017).

Based on the above, one can conclude that it is essential not only to treat wastewater effectively but also use water sources in a sustainable way and circularity approach. Water reuse is an alternative and very successful way to reduce the environmental problems, and scale of water scarcity issues. Water recovery options can significantly affect the agricultural field, reduce the overall level of pollution of the environment as a complete system, reduce greenhouse gas emissions, as well as improve quantity and quality of well-being and health of living organisms (Jost, Dale, & Schwebel, 2019).

In order to reuse wastewater in a wide range of spheres, the quality of water at the outlet should be as high as possible. Membrane bioreactor (MBR) is one of the technologies which allows implementing the water recirculation approach in real life. Especially in the circumstances prevailing in the European Union, where the standards of effluent quality become more strict, and amount of wastewater is continuously increasing in conditions of limited space (“Water Reuse - European Commission,” 2018). With MBR treatment, it is possible to achieve high-quality effluent, reduce the foot-print of water treatment facilities, and save time. But due to the process of membrane fouling which reduces the lifespan of the membrane and increases the capital, operational and maintenance cost in comparison to conventional activated sludge process, the technics which can solve the fouling related problems must be used for further successful implementation (Radjenovic, Petrovic, Majitovic, & Barcelo, 2008).

Sustainable membrane fouling mitigation techniques has been one of the main concerns over the last twenty years. To date, there are many ways to control the membrane fouling were checked, including the following: addition of adsorbents and coagulants, the introduction of aeration through granular materials or air blowers in the MBR tank, quorum quenching. Each of these techniques has its pros and cons, which means that there is no one universal solution for membrane fouling mitigation. For example, the addition of coagulants leads to large flocks formation, which expands the filtration cycle of the membrane and improves the filterability of mixed liquor. However, the coagulant addition can decrease the pH and may affect the bioactivities of mixed liquor. Excess of coagulant can also cause deposition on the surface of the membrane (Zhao et al., 2019).

Electrocoagulation is an alternative method for membrane fouling mitigation. With the advantages, which electrocoagulation can provide to MBR process, such as:

- Small foot-print;
- No needs for chemical delivery and use (small and remote communities);
- Direct coagulant formation during EC process;
- Easy adjusting and operating process (simplification of the technological scheme);
- Possibility of concentration and extraction of valuable products;
- Formation of easily settleable and dewaterable flocks;

- Mineralization of organic pollution and disinfection effect (Wang, Hung, & Shamma, 2011).

Thus, the combination of EC/MBR methods of water treatment could be very advanced and highly promising. Therefore, the main objectives of this study were to investigate the influence of the main parameters of EC (current density, pH, wastewater composition) on treatment efficiency and membrane fouling, determination of optimal parameters for the combined process of EC/Membrane purification and comparison of electrocoagulation and conventional coagulation as a treatment step before membrane.

# 1. Literature review

## 1.1 Wastewater treatment principles

The wastewater treatment process includes the removal of contaminants, compounds, and particles from the water. Physical, chemical, and biological processes are used to purify water with the subsequent possibility of its safe return to the environment without negative consequence. There are decentralized wastewater treatment systems, which implies water purification close to the source of water or “on-site” (using septic tanks, biofilters, etc.), as well as centralized, which means transportation of water through pipes and pumping stations to municipal wastewater treatment plants (Vickers, Thompson, & Kelkar, 1995).

Because of increased water stress in many regions of the world, fast population growth and other water-related issues, wastewater recycling, and reuse of treated water are becoming more and more critical. The worldwide freshwater contamination is increasing with thousands of industrial and natural chemical compounds, which is one of the key environmental problems, humanity facing. Although most of these compounds are present at low concentrations, many of them raise considerable toxicological concerns, particularly when present as components of complex compounds (Schwarzenbach et al., 2006).

For prevention health problems and meet treatment requirements, there is a simple chain of sanitation service (Figure 1). The main point of this connection is wastewater treatment. This stage is one of the most important, as it precedes the last step, namely the release of the final result into the environment (Com, 2018).



Figure 1. Sanitation service chain

In wastewater treatment, three main steps require special attention and development to improve the process of water purification - primary treatment, secondary treatment, and tertiary (Figure 2).

Primary wastewater treatment means the separation of suspended matter from water using physical processes such as sedimentation, filtration. The basic principle is the passage of suspended solids through grit/screens/sand removal (preliminary treatment), thereby separating of contaminants from the water followed by settling in sedimentation chambers (Peterson, 2001).

Primary treatment is an important step in the wastewater treatment process, as it helps to prevent coarse and other undesirable contaminants from entering subsequent stages of treatment, thereby reducing the risk of malfunctions, clogging and reduction in overall removal efficiency.

The processes that relate to the secondary method of wastewater treatment are based on biological and sedimentation principles. The basis of the process is the microbial consumption of organic pollutants, followed by their conversion into carbon dioxide and energy with the potential for the reproduction of their growth and quantity. An example of this wastewater treatment step is

the use of activated sludge technology, trickling filters, biofilters, fixed-film, IFAS (integrated fixed-film activated sludge process) (Nathanson & Ambulkar, 2019).

The tertiary method of WW treatment usually includes chemical and biological methods of removing nutrients (P, N). By chemical precipitation, it is possible to remove nutrients which play a vital role in the development and growth of plants. For example, phosphorus and nitrogen-containing compounds need to be removed because they are a key factor of algae growth, which turns to the eutrophication process. Ammonium affects oxygen consumption when converted to nitrates, becoming a toxic substance, in case of further contact with water organisms. Examples of tertiary WW treatment are coagulation/flocculation, membranes for the advanced treatment process, RO, adsorption (Mareddy, 2017).

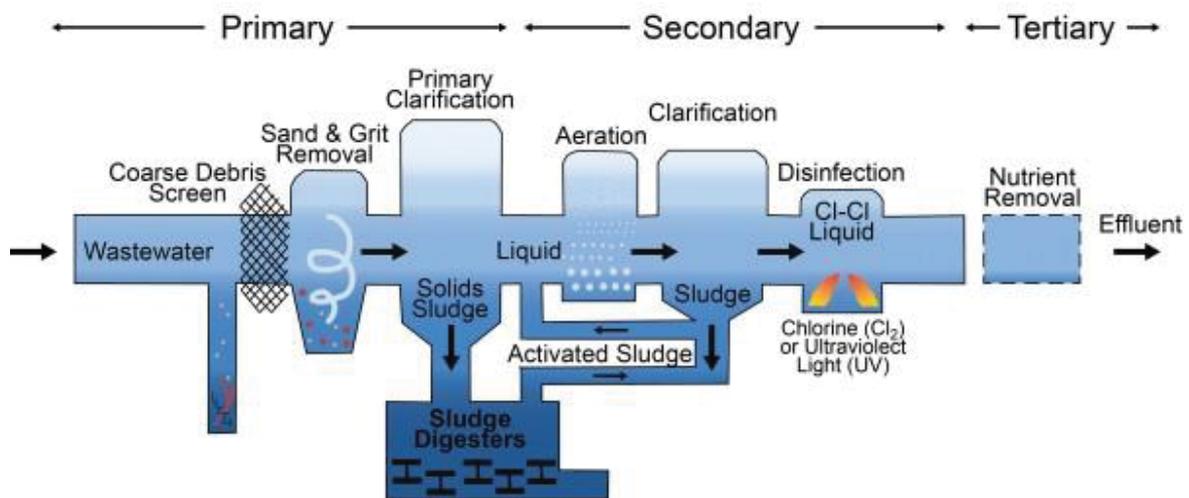


Figure 2. Wastewater treatment process (Center of Sustainable Systems, 2018)

The average removal efficiency of each step of wastewater treatment is demonstrated in Table 1 (Aljabali & Tratschin, 2018).

Table 1. Removal efficiency at different steps of wastewater treatment processes

Parameter	Primary treatment Removal efficiency, %	Secondary treatment Removal efficiency, %	Tertiary treatment Removal efficiency, %
Suspended solids	50-80	80-90	>90
BOD	20-40	70-90	>90
Phosphates	<10	30	>90
Nitrates	<10	30	70
E.Coli	1 log	2-3 log	3-5 log

For European countries, it is imperative to follow the EU regulations\_(*Council Directive of 21 May 1991 concerning UWWT*, 1991), which are established for maintaining stable concentrations of various pollutants in the water and ensure the reduction of possible adverse effects on human health and the environment.

Table 2. EU regulations/requirements for discharges from urban wastewater treatment plants

Parameter	Concentration
Biochemical oxygen demand (BOD)	25 mg/l O <sub>2</sub>
Chemical oxygen demand	125 mg/l O <sub>2</sub>
Total suspended solids	35 mg/l
Total phosphorus	2 mg/l
Total nitrogen	15 mg/l

Table 3, presented below, listed the advantages and disadvantages of the main methods that are used in wastewater treatment for a better understanding of the positive and negative aspects of each step.

Table 3. Advantages and disadvantages of the most widely-used wastewater treatment methods

Wastewater treatment method	Advantages	Disadvantages
Coagulation/flocculation	<ul style="list-style-type: none"> <li>● Reduction of required settling time;</li> <li>● The high removal efficiency of particles, phosphates and many protozoa, bacteria and viruses;</li> <li>● The high removal efficiency of colour and turbidity;</li> <li>● Possible to automatize this process;</li> <li>● Many investigations of new types of reagents.</li> </ul> (Robinson, 2015)	<ul style="list-style-type: none"> <li>● Precise dosing required;</li> <li>● Frequent monitoring of process;</li> <li>● Many factors which have an influence on the process efficiency, such as pH, temperature, a dose of coagulant, set process parameters (time of mixing and settling), properties of initial water;</li> <li>● Residual components in treated water;</li> <li>● Because of need in optimization and control - not the best choice for supplies with small</li> </ul>

		capacity (Colombet, 2011).
Membrane bioreactor	<ul style="list-style-type: none"> <li>● System compaction (decreasing of the necessary area);</li> <li>● High removal efficiency (SS, COD, BOD, nutrients);</li> <li>● Optimized maintenance. (Khan et al., 2018)</li> </ul>	<ul style="list-style-type: none"> <li>● Membrane fouling;</li> <li>● Economic aspects (replacement of membrane, cleaning, maintenance cost, energy consumption);</li> <li>● Needs in aeration;</li> <li>● Complicated process control. (Goswami et al., 2018)</li> </ul>
Electrocoagulation	<ul style="list-style-type: none"> <li>● Significantly small needed area;</li> <li>● Simple operation;</li> <li>● No need for chemicals;</li> <li>● Low sludge production (also, sludge is easy to settle and has good properties for dewatering)</li> <li>● Small required process time;</li> <li>● Could be used for a broad spectrum of wastewater types;</li> <li>● Decreased amount of residual metals in treated water. (Siringi, Home, Chacha, &amp; Koehn, 2012)</li> </ul>	<ul style="list-style-type: none"> <li>● Energy consumption (high cost);</li> <li>● pH control needed;</li> <li>● Regular replacement of electrodes. (Friedrich, Rodriguez, Stopić, &amp; Friedrich, 2007)</li> </ul>
Conventional Activated Sludge process	<ul style="list-style-type: none"> <li>● Good removal and treatment efficiency;</li> <li>● Compact system (no need of big area);</li> <li>● Cost;</li> <li>● Easy to operate (Kiss, Vatai, &amp; Bakassy-Molnar, 2003)</li> </ul>	<ul style="list-style-type: none"> <li>● High dependence on inlet sewage properties;</li> <li>● Cost of operation and maintenance;</li> <li>● Control of sludge activity;</li> <li>● Need in sludge disposal;</li> <li>● High dependence on temperature. (Hendricks, 2011)</li> </ul>
IFAS	<ul style="list-style-type: none"> <li>● High removal efficiency;</li> <li>● Reduction of the needed</li> </ul>	<ul style="list-style-type: none"> <li>● High construction costs;</li> <li>● Need in oxygen supply and its control;</li> </ul>

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|---|---|
| <ul style="list-style-type: none"> <li>area;</li> <li>● Stable process;</li> <li>● Increased nitrification recover speed;</li> <li>● Less sludge output;</li> <li>● Improved nitrification and denitrification processes;</li> <li>● Increased treatment system capacity</li> </ul> <p>(Johnson, McQuarrie, &amp; Shaw, 2012)</p> | <ul style="list-style-type: none"> <li>● Energy consumption;</li> <li>● Control on a biomass thickness;</li> <li>● Recommended making a prediction related to sludge age, thickness parameter, kinetic rates.</li> </ul> <p>(Brentwood, 2019)</p> |
|---|---|
- 

## 1.2 Biological treatment by IFAS process

The technology of using activated sludge during wastewater treatment is a very expedient solution at the stage of biological treatment. This method is based on the removal of pollutants by using bacterial biomass suspension. At this stage of treatment, organic carbon can be highly removed, and nutrient elements such as N and P to a lesser extent. There are systems in which biomass growth occurs on the surface, thereby forming a biofilm (MBBR, IFAS, trickling filter) (Gernaey & Sin, 2013).

To ensure an effective treatment step by using activated sludge technology, it's required to maintain the oxygen supply, monitor the sludge age, and also the installation of an additional tank – clarifier needed as well (Pell & Wörman, 2008).

Integrated Fixed-film Activated Sludge means the combination of carrier-fixed biofilms with an activated sludge system, using a smaller area due to the addition of special media (surface for microbial grows) to aeration tanks, thereby preventing the need of additional tanks. The selected media can affect the growth and amount of biomass in the system; therefore, the temperature of the system, energy, and intensity of mixing, loading, wastewater characteristics, dissolved oxygen concentration parameters should be monitored and controlled.

The volume occupied by media usually is 30-60 percent of the total volume of the aeration tank.

In terms of loading, the plastic media can be chosen (for example HydroxylPac Media), sponge media type (Linpor Media), string media system (Ringlace Media), but it is important to mention, that the selected area should provide an appropriate surface to ensure proper biological growth. For the present research work, the plastic rectangular beads were chosen for carrying an experiment (Gernaey & Sin, 2008).

### 1.3 MBR

Nowadays, technological innovations and improved techniques are inherent in biological wastewater treatment. Mainly, it is necessary to highlight the application and implementation of membrane bioreactor (MBR) at water treatment plants (H.-D. Park, Chang, & Lee, 2015).

MBR is a successful and efficient combination of biologically active sludge and membrane filtration, demonstrating a numerous advantage over the conventional activated sludge systems. The quality of MBR purification helps for many water treatment plants to reach the established regulations for the quality of wastewater treatment, without additional foot-print. The main goal of membranes implementation is to divide and separate biomass part from treated/clean water (Falizi et al., 2018).

At the stage of membrane filtration, it is necessary to have an understanding of the state of water at different stages, for example, incoming water is called feeding, or just a raw source of water entering the treatment; water that has passed through membrane filtration is called permeate (the final stage of wastewater treatment using membrane separation); the part of the water that lingers in the membrane is called concentrate.

One of the most important parameters for monitoring the condition and efficiency of the membrane is the transmembrane pressure, which represents the pressure difference observed at the stage of water entry into the purification system, or the pressure value, which is presented at the permeate stage (Dickhout et al., 2017).

An essential indicator of the feasibility of using a membrane, as well as the benefits of using membrane technologies, is the capacitive ability of the filtration element, which implies the passage and the possibility of cleaning water flow. The ability of the purified water to pass through the membrane is called permeability. This parameter depends on many factors, such as the type and material of the membrane, the composition of the water supplied for treatment, and temperature conditions, etc.

The most widely known variants of membrane purification are the microfiltration process (working size range 100–1000 nm), ultrafiltration (5–100 nm), nanofiltration (1–5 nm), and the reverse osmosis process (0.1–1 nm), these methods produce permeate and concentrate. Also, the processes of MBR include electro dialysis and electro-deionization (Radjenovic et al., 2008).

### 1.4 Ceramic membranes

The most important component in the method of water purification using membrane technologies, in fact, is a membrane. The membrane is a specific barrier that passes through the identified components of liquid or gaseous mixtures, thereby improving the condition of the incoming water, reduces the number of undesirable impurities, thereby reducing the possible risk and consequences of subsequently released water (Gitis & Rothenberg, 2016).

Usually, membranes made from synthetic organic polymers (such as polysulfone, polyvinylidene fluoride, polyacrylonitrile, polyvinyl chloride, etc.) or inorganic materials (examples are ceramics or metals). Size of pores depends on conditions by which was done the

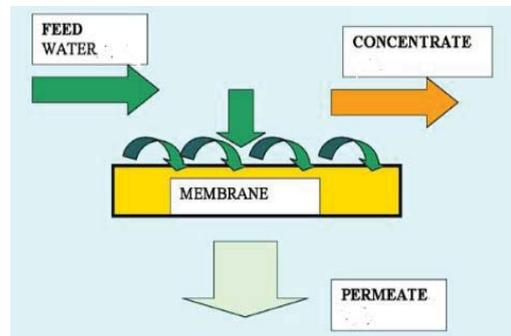


Figure 3. The basic principle of membrane filtration (Falizi et al., 2018)

membrane formation. The main difference of membrane technologies from conventional filtration is that contaminations from water do not accumulate in the membrane but remains in liquid form at its surface. Possible methods for cleaning and removing those particles are reversed flow (backwashing) or lateral flushing, ultrasound (S. H. Park, Park, Lim, & Kim, 2015).

The advantages of using ceramic membranes based on their strength; they can be used to treat water, in which pollutants contain coarse suspended particles, oil, or other mechanical suspensions. The surface and general properties of the membranes allow the use of strong chemical cleaning at different temperature ranges since this type of membrane is resistant to the above conditions. One of the following advantages, which distinguish the ceramic type of the membrane over others is the absence of the requirement to be always in water since the membrane is able to function even after removing it from the cleaning tank and restarting the cleaning process. With this type of membrane, it is possible to achieve a high degree of purification from suspended matter (up to 98% of removal efficiency). There is a possibility of processing the materials of which the membrane is made, thereby increasing the level of positive environmental impact by recycling materials. Ceramic membranes have a long service life (up to 20 years, in some cases), which reduces the cost of the constant replacement of cleaning elements. The above points prove the advantages of ceramic membranes over many other types, due to their cost-effectiveness, efficiency and ability to adapt to the specified conditions (S. J. Lee, Dilaver, Park, & Kim, 2013).

There is a list of disadvantages of ceramic membranes, such as the fragility of this product, negligence in the use and operation can lead to element failure. The properties of this kind of membranes are affected by abrupt changes in temperature by more than 30°C, thereby requiring control that minimizes this risk for the cleaning element. Freezing is also not desirable.

It is important to determine how hydrophobic or hydrophilic the membrane is since the operating flux rate depends on this characteristic (higher hydrophilicity means a higher value of this indicator) (Porcelli & Judd, 2010).

In general, the choice of ceramic membranes due to the high degree of purification from pollutants, as well as an effective way to pathogens and log removal is efficient and has many benefits (AMTA, 2018).

In our research project, flat sheet SiC (silica carbide) microfiltration membranes with 0.1 µm pore size were used. In our case, the chosen membrane surface refers to the very hydrophilic type (“CEMBRANE - new generation ceramic membranes,” n.d.).

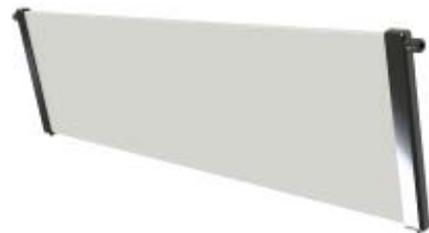


Figure 4. Flat sheet SiC microfiltration membranes with 0.1 µm pore size

## 1.5 Membrane fouling

One of the main disadvantages of membranes is blocking ability. Reduction of permeate flux, as well as an increase in pressure during membrane filtration, means a decrease in the treatment efficiency, in the form of membrane fouling (Falizi et al., 2018).

The main factors that cause this problem are the characteristics of the selected membrane (membrane type, material, pore size, etc.); conditions of service and use (mode, aeration factor, hydraulic retention time, the temperature at which the treatment is carried out, the ratio and the amount of organic matter and nutrients in the system); as well as the properties and characteristics

of the feed water for treatment (size of flocks and particles, pH value, concentration of mixed liquor suspended solids, sludge viscosity, etc.) (Bernardes, 2014).

Membrane fouling results in different forms at different stages, such as pore narrowing, pore-clogging and the last one and most problematic - layer (biocake) formation (Figure 5) (Iorhemen, Hamza, & Tay, 2016).

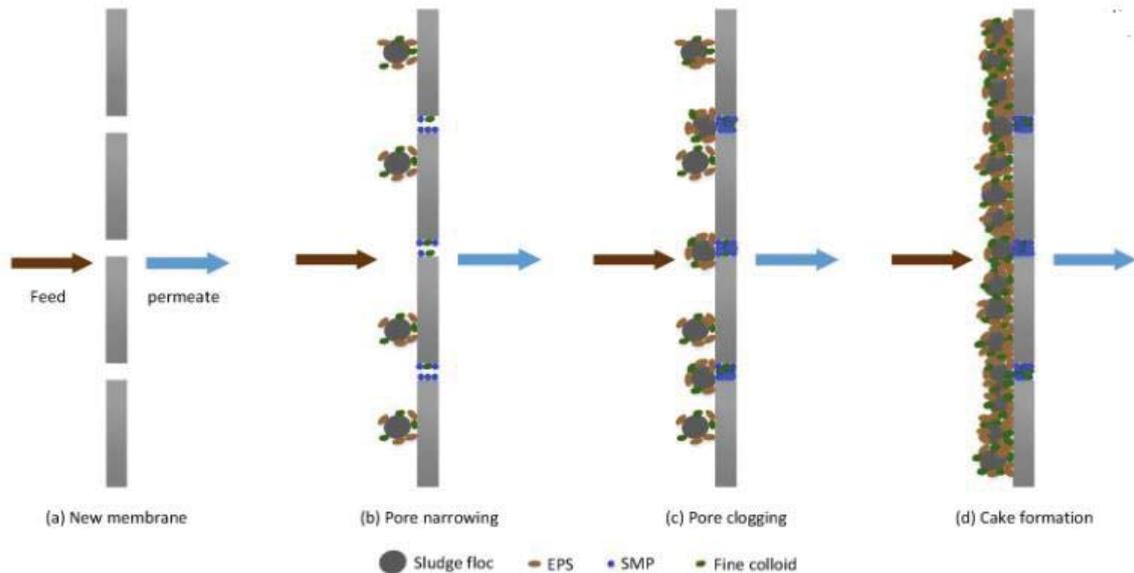


Figure 5. Membrane fouling demonstration

There are several types of membrane fouling by some biological and chemical properties, such as:

- Biofouling, which is caused by the growth and increase in the number of microorganisms on the surface of the membrane element and requires special attention in the process of washing or cleaning, as there is a risk of a critical increase in the amount of unwanted surface layer (present fouling type could be caused by increasing of bacteria amount and its growth, number and formation of microorganisms);
- Colloidal fouling. It is the result of film accumulation on the surface of membrane element (fouling could be caused by organic colloids, colloidal hydroxides/silicates/silicic acid);
- Scaling (inorganic fouling). It is caused by the formation of a specific crystallized coating on the membrane (the coating can be the following compounds:  $\text{CaSO}_4$ ,  $\text{SiO}_2$ ,  $\text{Mg(OH)}_2$ , etc.);
- Organic fouling, which caused by the presence of large amounts of organic pollutants (NOM). For the most part, these components are present in surface waters and are less common in groundwater (Chang, Lee, & Lee, 2019).

There is a need to prevent membrane fouling, due to the fact that it has influences on the decrease of efficiency, the capacity of the membrane element and increase of power consumption. Reduction of the filtration cycle, area and capacity of the membrane negatively affects the degree of pollutants removal and requires special attention and the development/search for solutions to reduce the risk of membrane fouling (Goswami et al., 2018).

There is a list of methods that contribute to the reduction of membrane fouling, as well as a positive effect on the degree of water purification in general, such as:

- the addition of coagulant to the water treatment system allows for the integration of smaller ones and the formation of larger compounds that increase the filtration capacity of the membrane element, reducing the risk of clogging with fine elements;
- adding an adsorbent that contributes to a larger surface to adsorb the components (for example, dissolved organic polymers) in water/wastewater. Effective is the use of powdered activated carbon, which reduces the risk of both organic and biofouling.
- the use of aerobic granulation. This method involves the addition of granular biomass to the membrane bioreactor, thereby introducing the possibility of interaction of microelements without the intervention of biocarriers (self-immobilization).
- the proposed method is the use of aeration together with granulated materials, the result of which is the continuation of the stage of mechanical water purification/wastewater. This method allows to increase the life of the membrane stage, as well as to achieve a higher permeate flow.
- the use and the addition of special bacteria that reduce the number and volume of bioformations on the membrane, as well as increase the throughput of the membrane element (Iorhemen et al., 2016).

An alternative method of coagulation is the use of electrocoagulation using an artificial coagulant, by dissolving metal in water (aluminium, iron, etc.). This method allows to reduce the likelihood and degree of contamination and clogging of the membrane, thereby positively affecting the service life and operation of the element.

## 1.6 Coagulation process

One of the most common and well-known methods for treating both drinking and wastewater is the coagulation process. The coagulation process has been well known since the 1900s, as it is a highly efficient and cost-effective treatment method. Most often, coagulation occurs in combination with other water purification processes. This combination allows to improve the effluent quality result (Jiang, 2015).

This key purification process implies the integration of small, destabilized particles together into larger and more substantial ones, thus facilitating the process of separating and extracting these aggregates from water by pre-sedimentation (Figure 6) (López-Maldonado, Oropeza-Guzman, Jurado-Baizaval, & Ochoa-Terán, 2014).

Substances that are introduced into water for the subsequent enlargement of particles are called coagulants, which are represented by inorganic coagulants, which are represented by metal salt solutions (most often aluminium and iron, but recently the efficiency of using titanium and zirconium is studied); organic (synthesized monomers of Al and Fe based coagulants; cationic or anionic polymers) (Kasih, 2014) and natural (such as chitosan, starches, alginate, etc.) (Kumar, Othman, & Asharuddin, 2016).

The coagulation process is influenced by several factors, such as temperature, pH, molecular weight and polymer charge density, the type of selected coagulant, as well as its dose, mixing conditions: speed, time, and sedimentation factors (Samer, 2015). Coagulation of water contaminations is the process of enlargement of the smallest colloidal and dispersed particles,

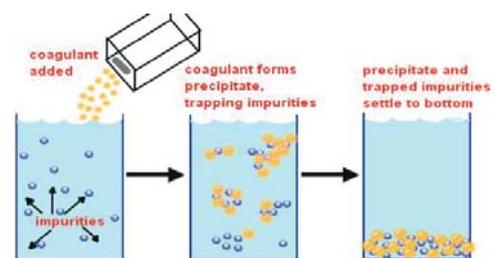


Figure 6. Basic process view of the coagulation process

which occurs as a result of their mutual sticking under the action of molecular gravity forces. Coagulation ends with the formation and separation of aggregates-flocks from water.

Colloidal and finely dispersed impurities of water have a certain aggregative stability, due to the presence of hydrate cover or a double electric layer around the particles. By heating or frizzling conditions, when electrolytes are added to water when a magnetic field is applied, the aggregative stability of impurities is disturbed (Kim, Park, & Novak, 2011).

Hydrophilic and hydrophobic impurities can be suspended in water.

Hydrophilic pollutants is mainly represented by organic substances, and hydrophobic one by particles of silt, clay, powdered grains, etc. A significant amount of water is retained on the surface of hydrophilic impurities in the form of a hydration shell, due to the presence of polar surface groups, such as  $-\text{OH}-$ ,  $-\text{COOH}-$ , etc. They keep the hydration shell around the particle. Being in thermal Brownian motion, the hydrophilic particles diffuse together with the hydration shell. Hydrophilic particles usually carry small electric charges and, as a rule, do not coagulate under the action of electrolytes (Forster, 2003).

Hydrophobic impurities are almost devoid of hydrated shells but have a double electric layer and carry significant electrical charges. Visibility or reduction of the electric charge of the particles leads to the violation of aggregative stability and coagulation of hydrophobic impurities.

Typically, the hydrophobic particles have a crystalline structure and a large specific surface on which present in water ions are adsorbed (Forster, 2003).

It is characteristic that all particles of a given substance collect ions of the same sign predominantly, reducing the free surface of the energy of hydrophobic particles. The ions are adjacent directly to the nucleus form a surface-nuclear layer (adsorption layer). Since an electric charge is formed at the boundary of the adsorption layer, a diffusion layer is created around the core with the adsorption layer (granule) from counterions that compensate for the charge of the granule. In general, a nucleus with adsorption and diffuse layers is called a micelle.

In the state of rest, the micelle is electrically neutral, since the charge of the granule is neutralized by counterions of the diffuse layer. Being in the state of Brownian motion, the counterions of the diffuse layer of the micelles lag behind, break away from the granule, and the particle acquires an electric charge (Samer, 2015).

Thus, particles with the same charges in the interaction repel each other. Along with this, between the colloidal impurities of water, there are molecular forces of mutual attraction, called Van der Waals forces, which act at a small distance between the interacting particles and decrease with an increase in the distance between them, and when the particles approach each other attraction increase. At the same time, initially, the repulsive force prevails over the force of attraction. However, if the particles, moving with high speed, overcome the indicated “force barrier”, then the forces of attraction become predominant and such particles merge and become larger (Petzet et al., 2012).

The magnitude of the “force barrier” is characterized by the electrokinetic potential or  $\zeta$ -potential. Optimally, when the electrokinetic potential is zero. This state of the colloidal system is called isoelectric, and the pH value corresponding to it is the isoelectric point of the system (pH).

Most of the colloidal particles of natural waters have a negative charge in the granule. Its neutralization, the destruction of the diffusion layer, and the hydration shell is achieved by introducing an electrolyte and sols with oppositely charged particles (C. S. Lee, Robinson, & Chong, 2014).

Iron and aluminium salts are widely-used as electrolytes. Coagulation can be achieved by adding a monovalent cation; however, the required amount of salt will be about 1000 times higher than the salt of the trivalent cation. When the electrolyte is introduced into water, the concentration

of anti-ions increases and a significant part of them go from the diffuse layer to the adsorption layer, which leads to a decrease in the thickness of the diffuse layer. The transition of counterions from the diffuse layer to the adsorption layer causes the charge of the granule to be neutralized and lowers the value of the electrokinetic potential of the particle.

Adding coagulant to water initially causes the formation of flocks (in the form of chains) from a single reagent (Jiang, Ho, & Li, 2015).

In turn, the coagulant chains may adhere to the surface of large impurities. Also, the almost insoluble hydroxides of iron and aluminium precipitate and, by adsorbing, insoluble particles of the suspension (sludge, plankton cells, plant residues, etc.) are carried along with them.

Iron and aluminium hydroxides are good sorbents. They adsorb bacteria, humic substances, and some dissolved compounds, for example, heavy metal ions, on the surface of their particles (Tebbutt, 2013).

## 1.7 Electrocoagulation step

Electrochemical wastewater treatment is a very promising technics which does not require the addition of a chemical. Unfortunately, electrocoagulation has received very little scientific attention even though it has great potential to avoid the disadvantages of conventional coagulation and got high commercialization, especially in the last 30 years (Mollah, Schennach, Parga, & Cocke, 2001).

Electrocoagulation wastewater treatment is the process of passing water through the interelectrode space of the electrolyzer, on the electrodes of which a specific potential difference is applied. At the same time, such phenomena as water electrolysis, particle polarization, electrophoresis, redox processes, and their combination can occur in the electrolyzer, which helps to reduce deposition of foulants at the membrane surface. The process can be carried out by using both soluble (iron, aluminium) and insoluble electrodes (titanium, lead oxide and so on) (An, Huang, Yao, & Zhao, 2017).

The choice of electrode material is based on the aggregative stability of particles of contamination. With a low content of the colloidal phase and low aggregative stability, it is advisable to use insoluble electrodes. For highly stable contaminants that require significant doses of coagulant for treatment, it is better to use soluble electrodes.

For instance, usually for the purification of industrial wastewater with high concentrations of contaminants, EC is performed by using soluble steel or aluminium anode.

Figure 7 represents the basic mechanisms of wastewater treatment by EC. As agreed by many authors, theoretically, the process of removing contaminants from wastewater by using electrocoagulation consists of three main stages (Hashim et al., 2019):

- 1) Electrolytic oxidation of the sacrificial electrode followed by the formation of a coagulant;
- 2) Destabilization of the emulsion, contaminants:
  - a. Compression of the diffuse double layer around the charged particles through the interaction with ions formed as a result of the sacrificial electrode corrosion;
  - b. Charge neutralization of the ionic species in water medium takes place, caused by the counterions formed in the process of the electrochemical dissolution of the anode. Van der Waals forces of attraction become stronger than the electrostatic forces of repulsion between particles due to counterions influence, which leads to the process of coagulation. Ideally, the net charge should be equal to zero;

c. Bridging and entrapping of colloidal particles as a result of flock formation with farther settling because of the increased density (another mechanism is flotation due to the  $H_2\uparrow$  gas formation).

3) Aggregation and flocs formation of destabilized contaminants.

The more in-depth details of each these steps require additional investigations.

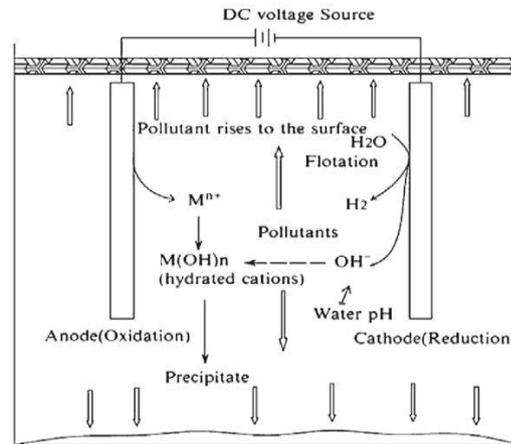
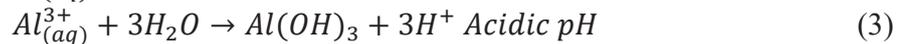


Figure 7. The mechanism of wastewater treatment by EC

The mechanism of electrocoagulation process depends on the aqueous environment and other parameters such as conductivity, concentrations of chemical constituencies, pH, particle size, etc. The following equations represent the process which is taking place at the Al cathode and anode during the electrocoagulation (Hashim et al., 2019):

Anode reactions:



Under the influence of direct current, the anode starts to corrode with the release of aluminium cations and  $Al(OH)^{2+}$  at lower pH with the subsequent hydrolysis process into aluminium hydroxide and finally can be polymerized to polymeric hydroxides  $Al_n(OH)_{3n}$ . According to pE-pH equilibrium diagram, the formation of the different charged form of polymeric hydroxo  $Al^{3+}$  species occurs under appropriate conditions (Figure 8). These multimeric, gelatinous hydroxo cationic complexes have a high surface area and very effective in pollutants removal by adsorption (charge neutralization) and enmeshment in formed flocks which make Al more suitable in comparison with other electrode materials (Hashim et al., 2019).

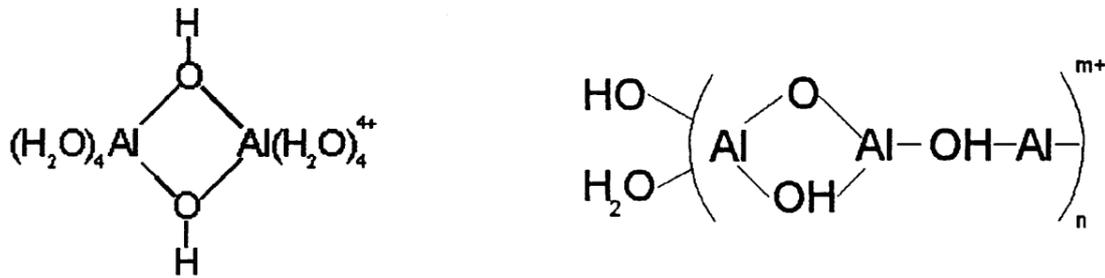


Figure 8. Structures of dimeric and polymeric  $Al^{3+}$  hydroxo complexes

Cathode reactions:



At the cathode, the process of electrode passivation and water reduction to hydrogen gas ( $H_2$ ) and the hydroxyl ion ( $OH^-$ ) taking place. The formation of this gas could help to separate the particles from aqueous media through the adhesion of bubbles to light solids and make them more buoyant with subsequent flotation. Moreover, the presence of hydroxyl radicals in the solution contributes to the oxidation of some contaminants (Tian et al., 2018).

If phosphates are also present in solution, it could be precipitated by the following reaction:



Due to the high neutralizing capacity of Al complexes, they are very effective for organic materials removal. In addition, the wide surface area creates the opportunity for adsorbing and capturing of soluble organic pollutants and colloidal particles, which can be easier for treatment by the membrane filtration process.

The electrocoagulation process is influenced by the material of the electrodes, the distance between them, the speed of wastewater mixing, its temperature and composition, voltage, and current density. With an increase in the concentration of suspended solids of more than 100 mg/l, the efficiency of electrocoagulation decreases. The reduction of the distance between the electrodes leads to a drop in the energy consumption for anodic dissolution of the metal. The theoretical energy consumption for the dissolving of 1 g of iron is 2.9 W·h and for 1 g of aluminium - 12 W·h respectively. The process is recommended to be carried out with a current density of not more than 10 A/m<sup>2</sup> and a distance between electrodes of no more than 20 mm (Barrera-Díaz, Roa-Morales, Balderas Hernández, Fernandez-Marchante, & Rodrigo, 2014).

Theoretically, the process of electrocoagulation has several advantages that make this type of wastewater (or mixed liquor) treatment highly efficient and promising, especially in combination with membrane cleaning (Naje, Chelliapan, Zakaria, Ajeel, & Alaba, 2017). The benefits in terms of the combined process of EC/MBR include:

- The sludge after electro-coagulation treatment mainly consists of metal oxides/hydroxides, which make it more settleable and dewaterable. Also, the amount of produced sludge is significantly lower in comparison to conventional coagulation;
- Faster filtration of EC flocs, due to a bigger size, less water content, higher stability and acid-resistance in comparison to chemical flocs;

- Efficient removal of small colloidal particles, due to the facilitation of coagulation by an electric field;
- Combination of two processes of particles removal: flotation and sedimentation;
- Easier control and less maintenance due to simple coagulant dosing and absence of moving parts respectively;
- No problems with secondary pollution and neutralizing of chemical excess, as it could be with conventional coagulation;
- Small footprint (Figure 9) (Sardari, Fyfe, Lincicome, & Ranil Wickramasinghe, 2018).

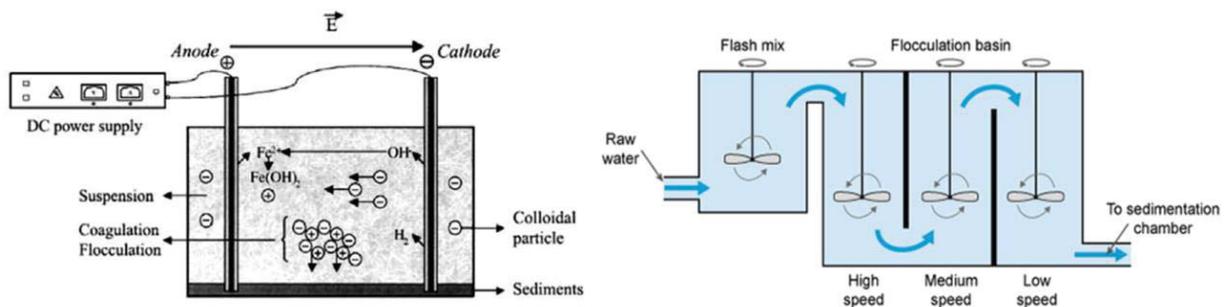


Figure 9. Demonstration of EC and conventional coagulation schemes

Therefore, the objective of this studies was to estimate the efficiency of electrochemical modification of mixed liquor after biological treatment (IFAS process) in terms of membrane fouling mitigation, thus extending membrane filtration cycle, phosphorous removal, and other standard parameters in municipal wastewater treatment. Another aim of this research was the establishment of the optimal parameters for electrochemical treatment with using both model medium-soft WW and real municipal wastewater. Based on the foregoing it can be concluded, that by the combination of up to date technologies, such as IFAS, EC, and MBR it is possible to achieve a high-quality effluent, which corresponds not only to modern standards (requirements) of quality of wastewater treatment but also to the near future (stricter) requirements (Devlin et al., 2018).

## 2. Materials and methods

The technological scheme of the experimental installation of municipal wastewater treatment is presented in figure 10. The source of real municipal wastewater is a sewage system in Ås commune. Initially, the wastewater passed through a mechanical filter, after which it flows into the equalization tank. After this step, water enters the biological treatment. Biological purification is represented as IFAS reactor, it is an integration of active sludge and fixed film systems. There are no anaerobic zones in the system, therefore, denitrification does not occur. The output of water after a biological tank is divided into 3 streams. The first flow (I) is the modification of mixed liquor by an inorganic coagulant, the second corresponds to purification by natural coagulant (II) and the third to electrocoagulation (III), respectively (Figure 10).

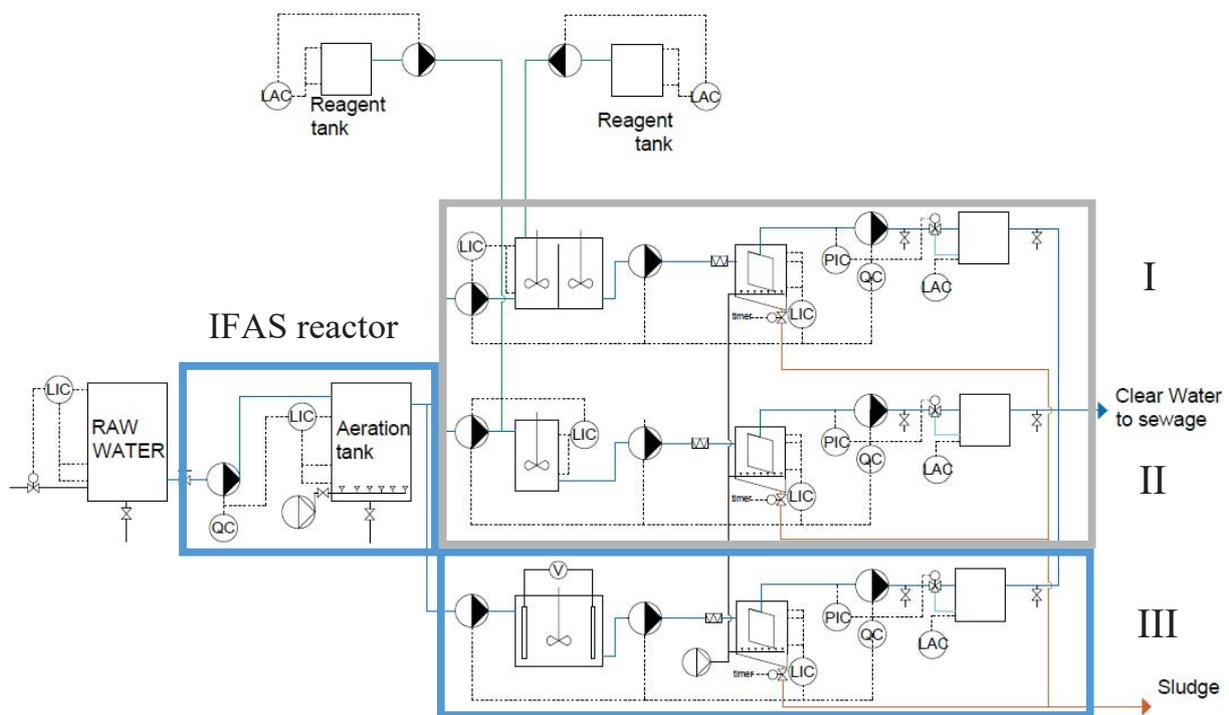


Figure 10. Technological scheme of municipal WW treatment

The installation for EC which was specially constructed for this project is represented at Figure 11. The EC cell itself consists of 3 Al tubes which are cathodes and anodes at the same time (Figure 11 a). The barrels 1 and 3 are electrically connected. Current is flowing between barrel 2 and 1-3. In this case, the wall thickness of the barrel 2 should be higher than barrels 1 and 3. The internal and external walls of the barrel 2 are working layers. At the upper shelf, there is a direct current power supply, and through the special program, it is possible to establish a current, periods of cathodes/anodes changing and check the energy consumption. Under this shelf, there is the flow controller (Figure 11 b).

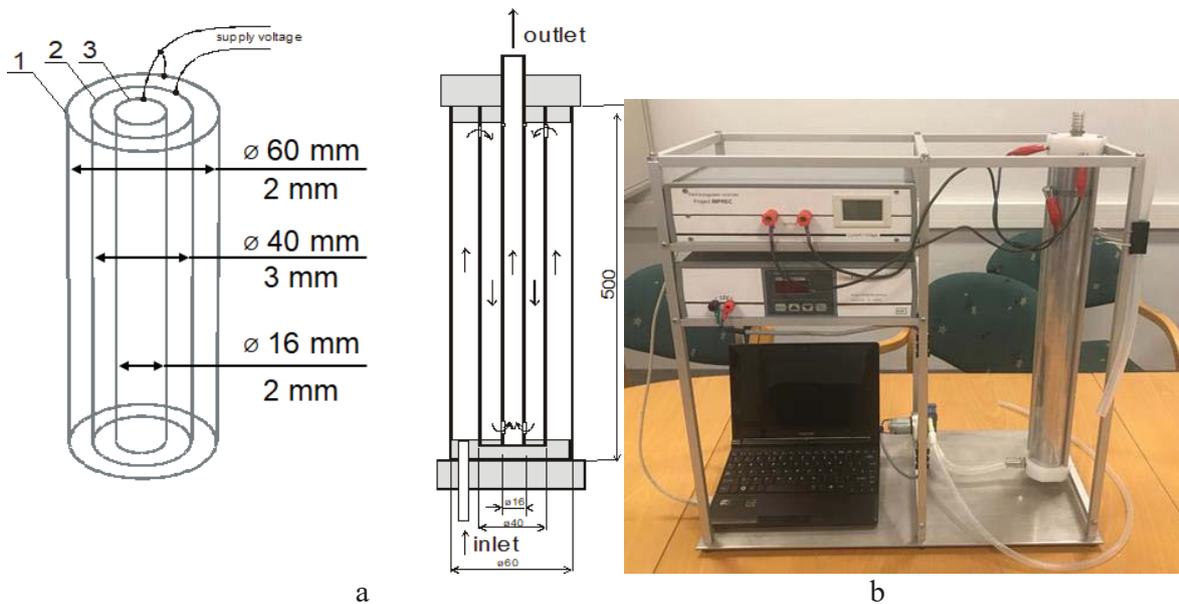


Figure 11. a - schematic view of EC cell, where 1- Al barrel 60 mm diameter, 2 mm wall thickness, 2 – Al barrel 40 mm diameter, 3 mm wall thickness, 3 – Al barrel, 16 mm diameter, 2 mm wall thickness.; b - real view of EC unit installation

Advantages of the cylindrical EC unit are:

- Very simple construction, much simpler than the comb arranged electrodes.
- Easy replacing of the worn electrodes.
- The electrical connection of the electrodes much easier than in the comb arrangement.
- Ratio volume of the EC unit to the electrode area is advantageous.
- The capacity of the system can be easily expanded by increasing the length of the barrels or by application more EC unit, connected in series and/or parallel.

## 2.1 Calculation of Aluminium doses

The Electrocoagulation process was conducted at constant current, the voltage was registered continuously every second. By the programmable power supply, the polarity of electrodes was changed every 256 seconds, since this time contributes to the best cathode cleaning and minimization of the polarization loss, according to the previous studies. The coagulant dose was calculated theoretically by Faraday's law:

$$m = k \cdot i \cdot t$$

where:

k – the electrochemical equivalent of Al, g/(A·s);

i-current, A;

t-time, s.

$$k = \frac{M(Al)}{Q \cdot z(Al)} = \frac{27}{(96500 \cdot 3)} = 9,3 \cdot 10^{-5} \text{ g/(A} \cdot \text{s)}$$

where:

M(Al) – a molar mass of Al;  
Q – Faraday constant;  
z – the equivalent of Al.

The Aluminium dose is proportional to the time of electrolysis. In order to get the same dose of Al at different flows (time of EC process), the different current was applied respectively. The volume of treated water is 1.5 L. The volume of EC unit is 1 L approximately, subsequently, the volume of sample for analysis is 0.5 L.

Example of calculation (Smoczyński et al., 2017)

If to apply a current of 0,6 A during the time of 1 hour, the mass of the Al which will be dissolved during the EC process can be calculated by the next equation:

$$m = k \cdot i \cdot t = 9,3 \cdot 10^{-5} \cdot 0.6 \cdot 3600 = 0.2 \text{ g of Al}$$

If to consider that the volume of wastewater which goes for the treatment is 1.5 L with the flow of 6 l/h (electrocoagulation process duration is 15 min in this case), then the mass of Al in one litre of WW is equal to:

$$m(\text{Al}) = 0.2 \text{ g}(\text{Al}) \cdot \frac{1000}{1.5 \text{ (L)} \cdot 4} = 33.57 \frac{\text{mg}(\text{Al})}{\text{L}}$$

Considering the duration of the treatment process at different flows and the desired mass of Al the current which should be applied was calculated.

The current density parameter (J), which is usually used by electrochemists as an expression of the coagulant dosage can be expressed as the ratio of the applied current to the active surface area of the anode. The active anode area of the EC unit is 0.12 m<sup>2</sup>:

$$J = \frac{I}{A} = \frac{0.6}{0.12} = 5 \frac{\text{A}}{\text{m}^2}$$

where:

J – current density A/m<sup>2</sup>;  
I – current, A;  
A – anode area, m<sup>2</sup>.

The Faradic yield or current efficiency is a very important parameter for the estimation of electrode dissolution, which can be determined by the relation of electrode weight loss during the experiment to theoretically consumed weight (according to Faraday's Law). Based on some EC studies, where the Al was used as a sacrificial electrode and the current efficiencies vary from ≈100% to more than ≈ 300%, that is why the measurements of Al has primary importance. (Omwene & Kobya, 2018).

## 2.2 Parameters of investigation and initial conditions

In present research work, synthetic wastewater was used for preliminary analyze of EC unit principles for the determination of optimal dose of mg Al/L with the following removal efficiency. The detailed description of model wastewater receipt is given in the paper of Hallvard Ødegaard,

Joachim Fettig and Harsha Chandima Ratnaweera (Ødegaard, Fettig, & Ratnaweera, 1990) and presented in Table 4.

Table 4. Composition of model WW

Components	Medium concentration (soft)
NaHCO <sub>3</sub> (mg/l)	60
NaCl (mg/l)	400
NH <sub>4</sub> Cl (mg/l)	100
K <sub>2</sub> HPO <sub>4</sub> (mg/l) (Required concentration of PO <sub>4</sub> <sup>3-</sup> = 5 mg/l)	28
Na-salt of Humic acid (mg/l)	5
Dry milk (mg/l)	300
Potato starch (mg/l)	60
Bentonite (mg/l) (Required concentration of TSS =100 mg/l)	45

After the static preliminary experiment, there was a set with real wastewater in dynamic regime.

Main properties of this water after biological tank were determined (Table 5):

Table 5. Parameters of water after the biological tank

Parameter	Value
pH	6.4
Turbidity, NTU	422
PO <sub>4</sub> <sup>3-</sup> , mg/l	9.43
TSS, mg/l	810
TOD, mg/l	728
Zeta potential, mV	-14.1

### 2.2.1 Turbidity

One of the most problematic parameters in measurement is turbidity. This parameter displays the optical properties of water, which may be affected due to the following factors:

- chemical pollution;
- bacterial particles;
- Some natural components (sand, silt and others);
- other coloured contaminants.

A high value of this parameter can lead to disruption of the condition of pipes, taps, and other regulators.

It is very important to know the turbidity index at the initial stage of purification, as well as after each subsequent stage. These measurements are important at the sewage treatment plant both low and high load/capacity.

There is a direct dependence on the amount and concentration of suspended substances in water and turbidity values. In this study, turbidity was measured in the initial water, as well as at all subsequent stages in unfiltered samples. All results presented in nephelometric turbidity units (NTU). In present research work was used turbidimeter by Hach 2100N Turbidity Meter (*Turbidity meter*, n.d.).

### 2.2.2 TSS

Determining the amount and concentration of suspended solids in water is very important since information about this parameter can prevent disruptions in the operation of the system as a whole and reduce possible harm to the state and health of the environment. Usually, this parameter is expressed in mg/L. The complete procedure for determining the amount of suspended solids in water is as follows: - pre-dry the filter to constant weight; - weigh the filter and record its mass; - filter 50 ml of the sample; - put in the oven at 120 degrees Celsius and keep there until a stable mass is obtained; - the next step is to weigh the filter after the drying process; - the last step is the process of converting the concentration of suspended particles in water (Kiepper, 2016). The present procedure is recommended to repeat twice, make a duplicate for more precise results. In this work, we used glass microfiber filters GF/C<sup>TM</sup>ø47 mm.

### 2.2.3 pH

Such a parameter as pH is important, as it has a direct impact on the cleaning efficiency at the chemical stage, biological and on the whole process. Depending on the pH value, it is necessary to take precise measures in this area, such as adding certain chemicals, for example. Measurement of this parameter should be implemented at the initial stage of determining the initial parameters of water, as well as at all subsequent stages, in order to monitor and control the health of the treatment process (Theobald, 2016).

In present research work was used next pH meter: WTW ProfLine pH meter 3110.

### 2.2.4 OP

In this research work, was measured ortho-phosphate concentration. The concentration of orthophosphate ions in water samples was measured according to ISO 6678:2004(E) standard (International Organization for Standardization, 2004).

Measurements of ortho-phosphates were done by using laboratory analyzer Systea EasyChem, which has principle operation and methods (Analysis, 2009).

For the preparation of working reagents needed:

- For R1 (Working Reagent):

Molybdate stock (ammonium molybdate  $(\text{NH}_4)_6\text{Mo}_7\text{O}_{24}\cdot 4\text{H}_2\text{O}$  + DI water), Sulfuric acid (5N) stock (sulfuric acid (conc.)  $\text{H}_2\text{SO}_4$  + DI water), Antimony stock (antimony potassium tartrate + DI water);

- For R2 (Ascorbic Acid Solution):

Ascorbic acid, DI water;

All reagents (R1 and R2 solutions) were transferred to EasyChem Reagent Containers.

All solutions have own expiry date, so it is really necessary to mark the date of preparation on each of reagents and follow the instructions about using and hazard for every one of them (Analysis, 2009).

### 2.2.5 TOD

Organic pollution and impurities are difficult to use analytical measurement methods, thus there are a number of parameters that help to predict, measure and calculate this element (COD, BOD, TOD and TOC).

One of the possible options for monitoring and measuring the content of organic matter in wastewater is to measure the TOD parameter, which represents the total oxygen consumption.

The principle of measuring the TOD parameter is similar to the measurement of the COD parameter, since it implies the oxidation of all organic components and particles, with the further possibility of determining and calculating the required oxygen consumption (Genthe & Pliner, 2017).

For present measurement was used: Quick COS lab by LAR company.

### 2.2.6 Z-potential

The zeta potential of the sample determines whether the particles in the liquid system tend to coagulate or not. Therefore, with the measurement of zeta potential in relation to other variables, the repulsion between charged particles is indicated and therefore the aggregative stability of the colloidal system can be quantified (Instruments, 2018).

Z-potential of the particles was measured with a zeta-potential analyzer (Z-sizer Nano 2S, Malvern, UK), for surface charge assessment.

### 2.2.7 Aluminium

In the process of wastewater treatment using reagents (coagulants) and elements containing aluminium salts as part of both the traditional method of coagulation and the method of electrocoagulation. An important point is the measurement of residual aluminium in sludge formed after the process, and in purified water (L. Lee, Wang, Guo, Hu, & Ong, 2015).

The aluminium ions contained in the water do not explicitly belong to substances with a pronounced toxic effect, but since the solutions of aluminium salts are highly stable, they have a detrimental effect on human and animal organisms, with their gradual accumulation.

Measurements of aluminium were done by using laboratory analyzer Systea EasyChem, which has principle operation and methods. (Analysis, 2009).

For the preparation of working reagents needed:

- For R1 (Acid):

Sulfuric acid (conc.)  $H_2SO_4$ , ascorbic acid  $C_6H_8O_6$  and distilled water;

- For R2 (Buffer):

Sodium acetate trihydrate  $CH_3COONa \times 3H_2O$ , acetic acid (conc.) and distilled water;

- For R3 (Working colour reagent):

Stock E.C.R (eriochrome cyanine R  $C_{23}H_{15}Na_3O_9S$  + acetic acid (conc.)  $CH_3COOH$  + DI water), distilled water.

It was very important to mark the date, follow the instructions/guidelines and keep all those reagents refrigerated at  $+4^\circ C$  (Analysis, 2009).

During the experiment, different approaches for Al digestion were applied (concentrated Nitric acid, combination of Nitric acid and Hydrochloric acid), but the results of measurements made by spectroscopic are not representative. According to literature review, spectroscopic methods cannot be directly determined by UV/visible light spectrophotometry (Sposito, 1996).

### 2.3 The process of membrane filtration and cleaning

The ceramic membrane type (SiC) was used in during cleaning process. During this step, the dead-end filtration is used at constant pressure.

After passing the water through the electrocoagulation stage, the next step is cleaning with membrane using. The volume of the cell in which the membrane is immersed is 4 litres. Water enters this cell after the EC process, where there is a need for aeration in the cell since this is one of the options for reducing the membrane fouling, as well as preventing concentration polarization. After the membrane stage, the treated wastewater enters the permeate collection tank, from which a small part of the sample is taken for analysis of all the main parameters described above, and the rest is returned to the membrane tank, which is an indicator of the recycling process. After these two stages of water cleaning, the treated wastewater is discharged from the membrane chamber, but with a preliminary sampling of the supernatant for analyzing its quality. The washing process of the membrane consists of the reverse passage (backwashing) of sodium hypochlorite in a volume of 1.5 litres through the membrane, followed by filling the rest volume of the reactor in which the membrane is installed with tap water (approximately 2.5 litres), and the membrane relaxation time is 1 hour. After all these steps, the membrane is considered ready for subsequent experiments.

### 3. Results and discussions

The overall research was divided into 4 experiments, starting from the investigation of EC cell removal efficiency and finishing with a combination of Membrane treatment with electrocoagulation.

Since the installation was new and unused the first step of our investigation was the establishment of main parameters influence the treatment efficiency, such as:

- The dose of Al;
- pH;
- Flow;
- Influence of recirculation.

For this purpose, medium-soft model wastewater was used, with the concentration of Al 5mg/L and TSS 100 mg/L. At the first attempts we tried to use a very low current, to get the dosages of Al comparable to conventional coagulation, but the efficiency of treatment was almost no noticeable. Most likely, this is due to the low sensitivity of the power supply at low current values or because of the relatively big distance between the electrodes. The results of these attempts are not included.

#### 3.1 Experiment 1. Investigation of initial pH and current density influence on the efficiency of EC treatment (model WW)

To investigate the influence of pH on the treatment efficiency, the following conditions were met:

- Constant flow (6 L/h);
- Different current, which means a different concentration of Aluminium (Table 6);
- The volume of treated water 1.5 L;
- Duration of treatment 15 minutes.

Table 6. The theoretical dose of Al vs applied current (current density) for the experiment with model WW

Dose, mg-Al/L	22.38	44.77	67.15	89.53	111.92
Current, A (flow 3 L/h) (current density, A/m <sup>2</sup> )	0.2 (1.67)	0.4 (3.33)	0.6 (5)	0.8 (6.67)	1 (8.33)
Current, A (flow 6 L/h) (current density, A/m <sup>2</sup> )	0.4 (3.33)	0.8 (6.67)	1.2 (10)	1.6 (13.33)	2 (16.67)
Current, A (flow 9 L/h) (current density, A/m <sup>2</sup> )	0.6 (5)	1.2 (10)	1.8 (15)	2.4 (20)	3 (25)
Current, A (flow 12 L/h) (current density, A/m <sup>2</sup> )	0.8 (6.67)	1.6 (13.33)	2.4 (20)	3.2 (26.67)	4 (33.33)

The treatment efficiency at these graphs is represented as a function of Al dosage at different pH.

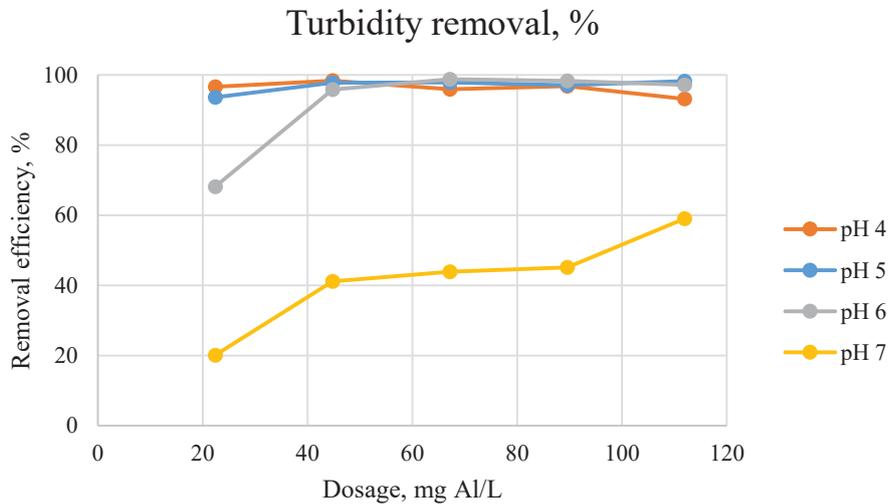


Figure 12. The dependence of turbidity removal efficiency on the dosage of Al at different pH

From Figure 12, it is clear, that at the optimal pH 4, the high removal efficiency can be achieved even at the low dosages of Al (current densities), and controversially at pH 7, which is not optimal, the removal efficiency is low, even at the higher doses (maximum 60%). The same tendency is maintained for the removal efficiency of total suspended solids (at the highest dose, it does not exceed 65%). EC is very efficient in terms of phosphorus removal, even though the pH 7 is not optimal according to other graphs it is possible to achieve more than 97% removal at the highest dosage.

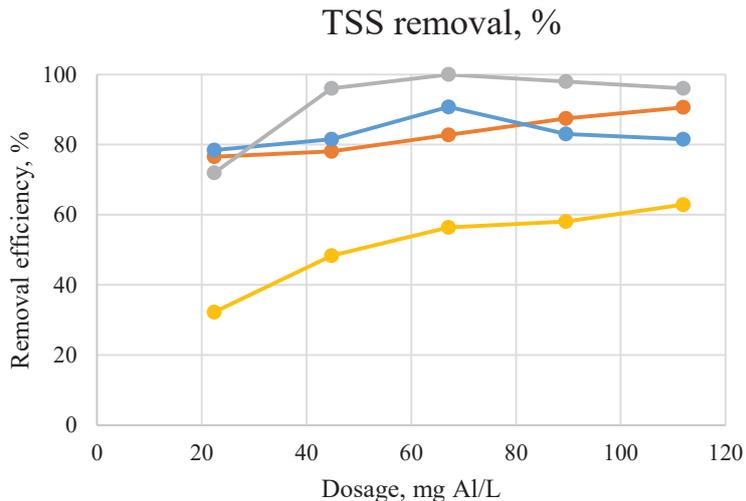


Figure 13. The dependence of TSS removal efficiency on the dosage of Al at different pH

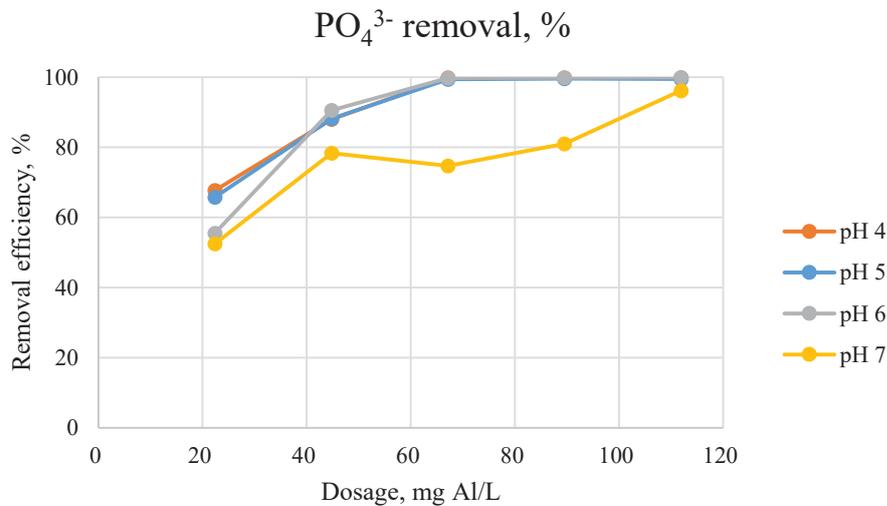


Figure 14. The dependence of OP removal efficiency on the dosage of Al at different pH

### 3.1.1 Influence of current density on treatment efficiency

In electrocoagulation treatment one of the primary importance parameters is current density. First of all, current density determines the dosage of coagulant, the rate of bubbles formation, floc size their strength and growth speed, in order, these parameters influence the efficiency of water/wastewater treatment. In its turn, the rate of bubbles formation ( $H_2\uparrow$  gas formation) have an influence on the particles removal by flotation, mixing condition (hydrodynamics in EC reactor) and subsequently coagulant distribution (aluminium, aluminium-hydroxy species). From Figures 12, 13, 14 it can be seen, that with the increasing of current density and subsequently dose of Al, the removal efficiency of all the parameters increases. With the increase of current density, the dissolution rate of sacrificial Al anode becomes higher due to Faraday's law (Attour et al., 2014).

In order to obtain a high efficiency of phosphorus removal, the optimal dose of coagulant should be applied. According to the equation, the theoretical molar ratio between Al:P = 1:1, however in practice it is higher. As it has been reported by other researches, the main reason for this is competing of reactions for particles and phosphorus removal. Based on the presented graphs, at low doses the removal of TSS (starts from 76% at pH 4) is higher than orthophosphates (67% respectively), that may indicate that particles removal is predominant in EC process. However, at the dosage of 67 mg-Al/l the removal of  $PO_4^{3-}$  increase up to 99% at pH 4,5 and 6, while at the same dosage the TSS removal shows the high result, only at pH 6.

One of the problems connected with using of Al as an electrode material is its coating with a passive oxide film, especially at low current densities (from 2 to 6  $A/m^2$ ), which is also proved by other studies (Hashim et al., 2019). This could be another reason for poor treatment efficiency at low doses. At the medium range of current density (around 10  $A/m^2$ ) this problem become less significant, and at the higher current density (more than 15  $A/m^2$ ) this passive oxide film disappears, due to intensive  $OH^-$  anions production which is accompanied by electrode corrosion (An et al., 2017).

In the framework of this study, to prevent the passivation of electrodes, a polarity change was performed, with a periodicity of 256 seconds, which was established as the optimal time in

previous studies. The change in polarity contributes to the self-cleaning of the electrode through the progression of the electrolytic decomposition of water with the subsequent release of hydrogen (Smoczyński et al., 2017).

Another problem with very high current densities, which was observed during the experiment is higher buoyant of sludge and problem with sedimentation. The explanation for this phenomenon could be very small bubbles of H<sub>2</sub>, which can be entrapped by sludge, due to this reason sludge can float at the surface of water, even after complete sedimentation of particles in water body, this caused the problems with water sampling for analysis (Attour et al., 2014).

### 3.1.2 Influence of initial pH on treatment efficiency

Initial pH is very important and one of the most sensitive parameters, which have an influence on the treatment efficiency, through the control of Al hydroxide speciation. Figures 12, 13 and 14 represent the impact of this parameter on the removal efficiency of turbidity, TSS and orthophosphates respectively in the initial pH range of 4.0-7.0, at the constant flow and different Al dosages. It can be clearly seen from these graphs, that with the increase of acidity of initial water the removal efficiencies are also increased. In almost all the cases the pH rises with the dosage of Al, which can be explained by cathodic water reduction with the release of OH<sup>-</sup> and chemical dissolution of electrodes. As it is explained in literature, at the pH less than 3.5 the dominant in solution is Al<sup>3+</sup> ion and at above pH values – polymeric and monomeric aluminum-hydroxyl species, for instance: Al(OH)<sup>2+</sup>, Al(OH)<sub>2</sub><sup>+</sup>, Al<sub>2</sub>(OH)<sub>2</sub><sup>4+</sup>, Al<sub>6</sub>(OH)<sub>15</sub><sup>3+</sup>, Al<sub>7</sub>(OH)<sub>17</sub><sup>4+</sup>, Al<sub>8</sub>(OH)<sub>20</sub><sup>4+</sup>, Al<sub>13</sub>O<sub>4</sub>(OH)<sub>12</sub><sup>7+</sup>, Al<sub>13</sub>(OH)<sub>34</sub><sup>5+</sup> etc., with general form Al<sub>x</sub>(OH)<sub>y</sub>(H<sub>2</sub>O)<sub>n</sub><sup>(3x-y)+</sup>. By the reactions of complex precipitation, these species converts to amorphous aluminium hydroxide (Al(OH)<sub>3(s)</sub>). In the pH range from 4 to 7 this positively charged gelatinous polyhydroxo-complexes are effective flocculants and effectively can remove pollutants by enmeshment in a precipitate and by adsorption through charge neutralization mechanisms. At the pH value higher than 10, the Al(OH)<sub>3(s)</sub> dissolves and form soluble Al(OH)<sub>4</sub><sup>-</sup> ions, which influence the pollutants removal efficiency (Mollah et al., 2001).

The pH 7 shows the worst results, which can be explained by higher solubility of aluminium hydroxide species since the pH is increasing during the electrocoagulation treatment. The lowest solubility is observed at an approximate pH of 6.3 and equal to 0.03 mg-Al/L. In addition, the positively charged hydroxide species in the pH range 4-7, enhancing adsorption of contamination, including anionic phosphate species (HPO<sub>4</sub><sup>2-</sup>, H<sub>2</sub>PO<sub>4</sub><sup>-</sup>, etc.) by electrostatic attraction and ligand exchange. At higher pH, the phosphorus adsorption ability is becoming worse, due to the sol surface charge decrease, and vice versa, the lower pH values contribute to release of hydroxyl anions and subsequently promote the anion adsorption. Furthermore, at the pH range, 4-10 Al-complexes promotes the formation of insoluble compounds with PO<sub>4</sub><sup>3-</sup>, such as AlPO<sub>4(s)</sub>, Al<sub>1.4</sub>PO<sub>4</sub>(OH)<sub>1.2(s)</sub>, Al<sub>3</sub>(OH)<sub>3</sub>(PO<sub>4</sub>)<sub>2(s)</sub>. Generally, it is considered that there are two main mechanisms of P removal (Tian et al., 2018):

- Precipitation at pH < 6.5;
- Adsorption at pH > 6.5.

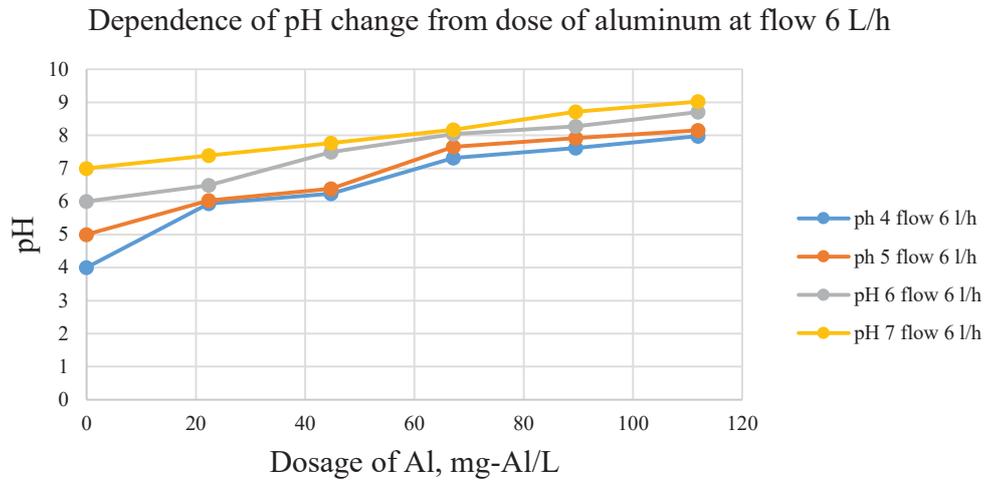


Figure 15. Dependence of pH change with the increasing of Al dose at the constant flow

Another reason is that the decreasing of pH is leading to easier destabilization of the colloidal system, which can be proved by Z-potential measurements. The graphs also show insignificant differences between the results at pH 4, 5 and 6 at the constant flow of 6 L/h, science the final pH (after EC) does not achieve the critical value (pH 10).

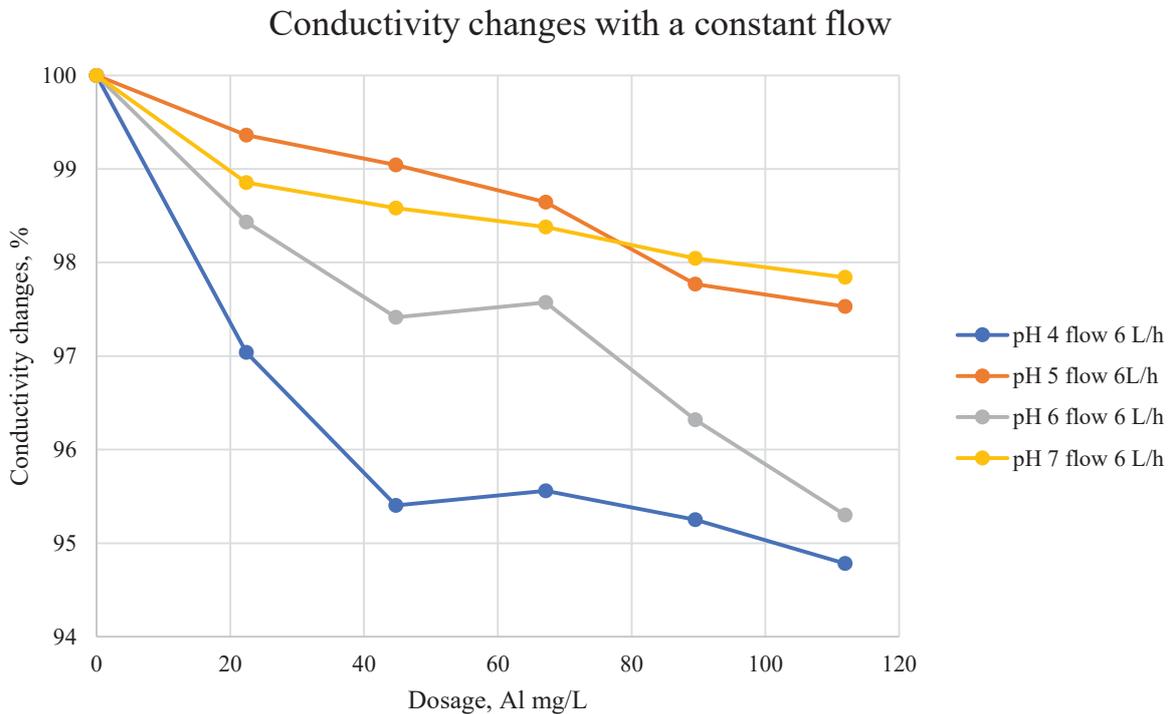


Figure 16. Dependence of conductivity change with the increasing of Al dose at the constant flow

### 3.2 Experiment 2. Investigation of flow influence on the efficiency of EC treatment (model WW)

To investigate the influence of flow on the treatment efficiency, the following conditions were met:

- constant pH of initial WW - 4;
- different current, which means a different concentration of Aluminium (Table 6);
- The volume of treated water 1.5 L.

#### 3.2.1 Influence of WW flow on treatment efficiency

Consider that with the different flow, water pass through the reactor for a different time, the dosage (current density) was adjusted to get an equal condition in terms of Al dosage (Table 6). Based on that, the different flow represents the mixing conditions in the EC reactor and residence time of wastewater.

Low removal efficiency at lower doses of flow 3 L/h can be explained by the low accuracy of the power supply at the current less than 0.4-0.5 A, or/and electrode passivation. For example, if to establish the current at 0.4 A, the real current is in the range of 0.2-0.3 A. That is why it is recommended not to use the current less than 0.5-0.6 A for this particular power supply. the low doses it is better to use the higher flow and applied current. The best results were demonstrated at flow 12 L/h, more than 90% for all the parameters at the lowest dosage.

The explanation of the low accuracy of the power supply could be the combination of significant distance between electrodes and not sufficient conductivity of initial wastewater. In addition, the electrode passivation with oxide film is responsible for poor treatment. But since the design of cylindrical EC reactors is not adjustable, it is recommended to check this assumption in further research, by increasing of initial conductivity of wastewater (which also accelerate the corrosion of sacrificial electrode).

Proceeding from the foregoing it can be concluded, that high purification rates at the flow 12 L/h is first of all connected to the high applied current, which helps to avoid the problems with passivation and possible problems with sufficient accuracy of the power supply at low doses. The second one is the appropriate mixing regime in EC reactor, which accompanied a better distribution of coagulant.

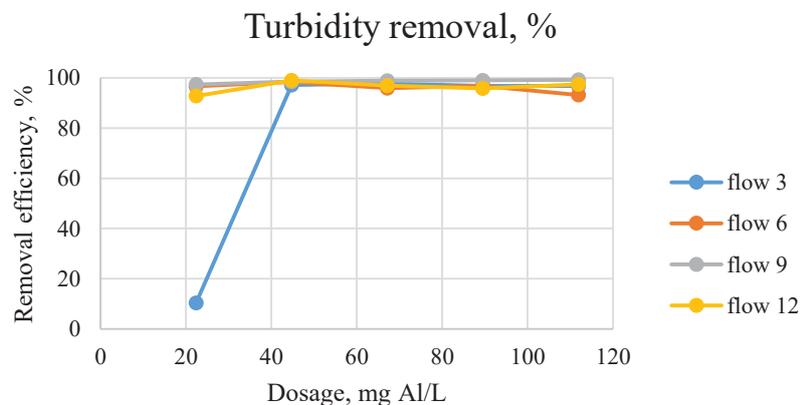


Figure 17. The dependence of turbidity removal efficiency on the dosage of Al at the different flow of WW

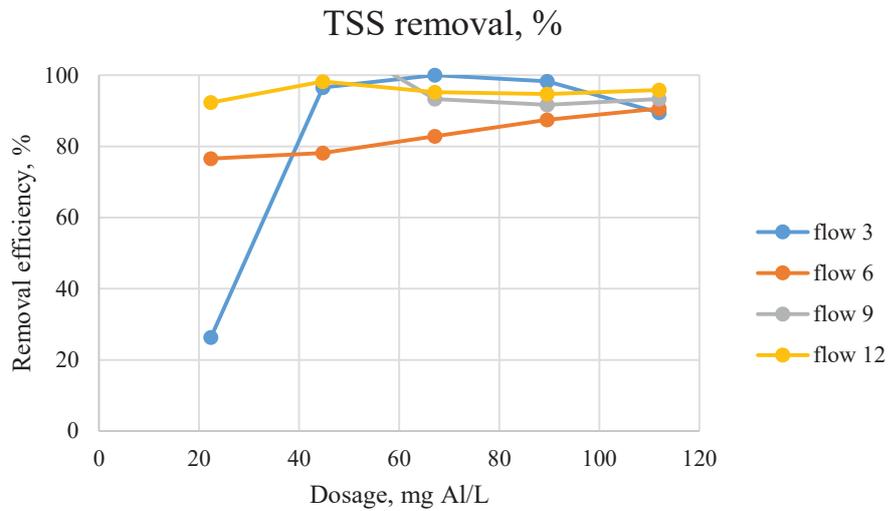


Figure 18. The dependence of TSS removal efficiency on the dosage of Al at the different flow of WW

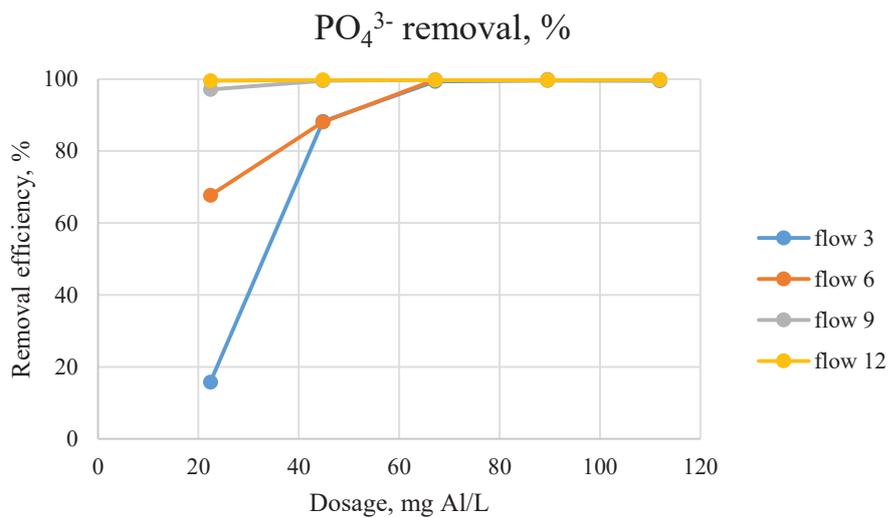


Figure 19. The dependence of orthophosphates removal efficiency on the dosage of Al at the different flow of WW

Proceeding from the obtained data for the removal efficiency at different flows, it can be concluded that there are no big discrepancies in results, which means that this parameter is not in primary importance, at least for the selected flows and doses of Al, that is why for the experiment with real municipal wastewater it was decided to use flow 6 L/h, since the overall wastewater treatment installation was created respectively to this value and biological treatment is able to handle this flow.

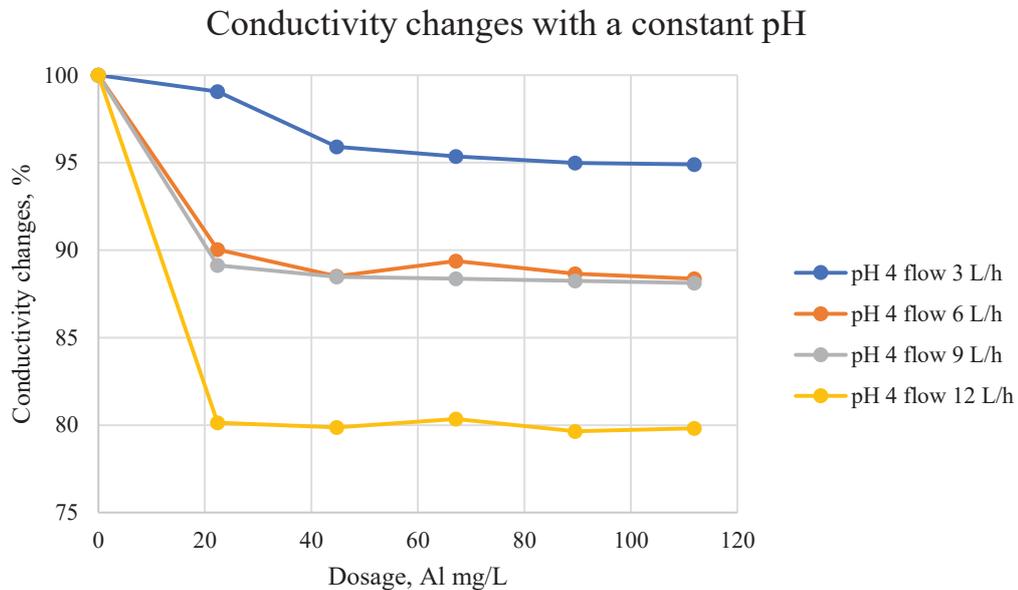


Figure 20. Dependence of conductivity change with the increasing of Al dose at different flows

### 3.3 Experiment 3. Investigation of flow influence on the efficiency of EC treatment (recycle test with model WW)

The idea of WW recycle was based on the assumption, that with the slow flow, and passing of the water through the EC reactor, it is possible to obtain the flocs on the first run and significantly increase them by additional coagulation at the second cycle, which theoretically can reduce the sedimentation time.

Initial conditions:

- pH 4;
- The volume of treated water – 1.5 L;
- Number of cycles - 2;

In order to obtain the same dosage of Al at the twice bigger time, the applied current was decreased twice. Due to the lack of accuracy of the current source, at low current values, the dose of Al was significantly lower than planned, that is why the efficiency of the purification is significantly lower compared to the treatment process without the recycle. It was decided not to use flow 3L/h, because of very low Al dosages, which should be applied in case of recirculation, due to the low accuracy of power supply, which lead to insufficient WW treatment.

Dependencies, which are shown on the three following graphs are a prime example of phenomena explained in the previous two experiments, and once more prove the theoretical and practical assumptions.

The dosages of Al on x-axes are theoretically calculated, and most probably they are not coincided with real ones, due to the electrode passivation, and low accuracy or Power supply.

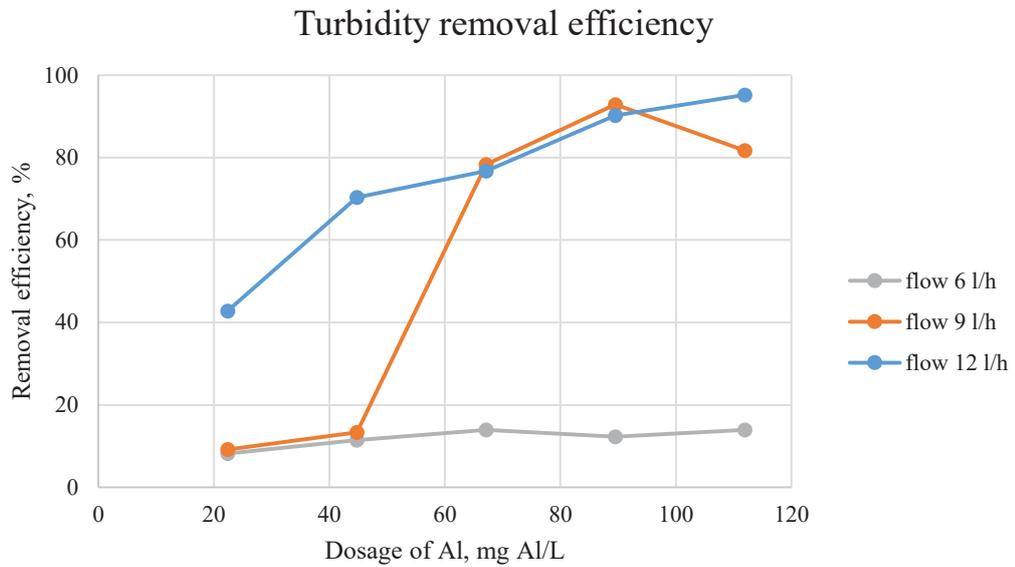


Figure 21. The dependence of turbidity removal efficiency on the dosage of Al at the different flow of WW at recycling test (constant pH = 4)

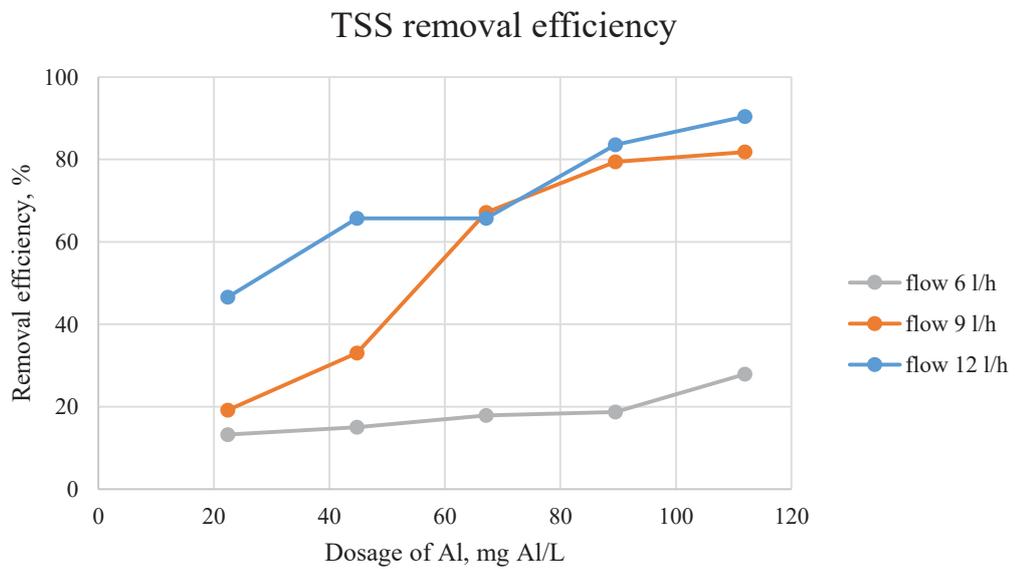


Figure 22. The dependence of total suspended solids removal efficiency on the dosage of Al at the different flow of WW at recycling test (constant pH = 4)

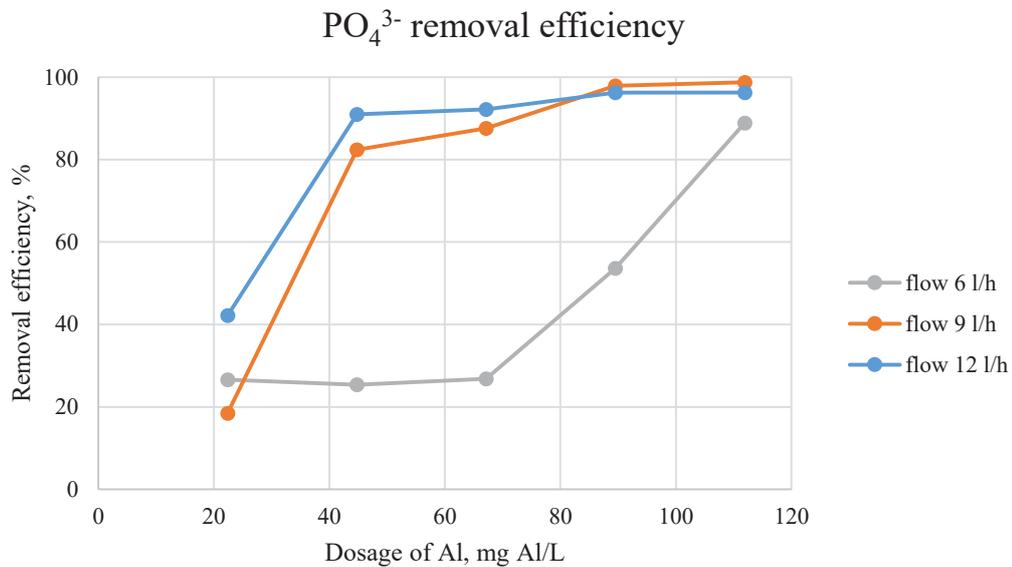


Figure 23. The dependence of orthophosphates removal efficiency on the dosage of Al at the different flow of WW at recycling test (constant pH = 4)

As we can see from previous graphs, the flow 6 L/h shows worse results in comparison with other flows (9 L/h and 12 L/h), the removal efficiency of turbidity and total suspended solids did not reach 20% and 40% respectively and removal efficiency of orthophosphates is the lowest even with highest dosage of Al (less than 90%).

The flow 12 L/h demonstrated the highest results of treatment efficiency, the main reason for this is the high enough applied current. For example, the first current at the recirculation test was 0.4 A (lower result than they should be) and the second one is 0.8 A, which is higher than the threshold of sensitivity of power supply unit, that is why the difference between the removal efficiency at first dosage and the second one is so high.

Based on these graphs, one can conclude that it is not recommended to organize the recirculation of WW, with an application of low current density. In order to clearly check this parameter influence, the flow of WW should be higher, with following increasing in current density. However, these conditions raise another issue related to the strength of the received flocs, will they be stable under such a hydrodynamic regime?

### 3.4 Experiment 4. Test with real wastewater

The purpose of the experiment with real wastewater was to confirm the effectiveness of the selected doses, and the hydrodynamic regime that was used in studies with model wastewater.

For the experiments with real WW, the flow of 6 L/h was used since it was investigated that this parameter has not a big influence on the removal efficiency and the overall unit was modelled for this flow. In this experiment, the differences between doses were decreased, since the range of optimal dose is in between 40-70 mg-Al/L, depends on the parameters.

Table 7. The theoretical dose of Al vs applied current for the experiment with real WW

Dose, mg-Al/L	0.6	0.9	1.2	1.5	1.8	2.4
Current, A (flow 6 L/h)	22.38	33.58	44.7	55.96	67.15	89.53

To investigate the influence of pH on the treatment efficiency of real WW, the following conditions were met:

- constant flow (6 L/h);
- different current, which means a different concentration of Aluminium (Table 6);
- The volume of treated water 1.5 L;
- Duration of treatment 15 minutes.

*\*It should be mentioned, that values on y-axis started not from -0- value, for graphs with turbidity and TSS removal it was started from 90% and for phosphates it was started from 60% for demonstration of results more clearly and precisely.*

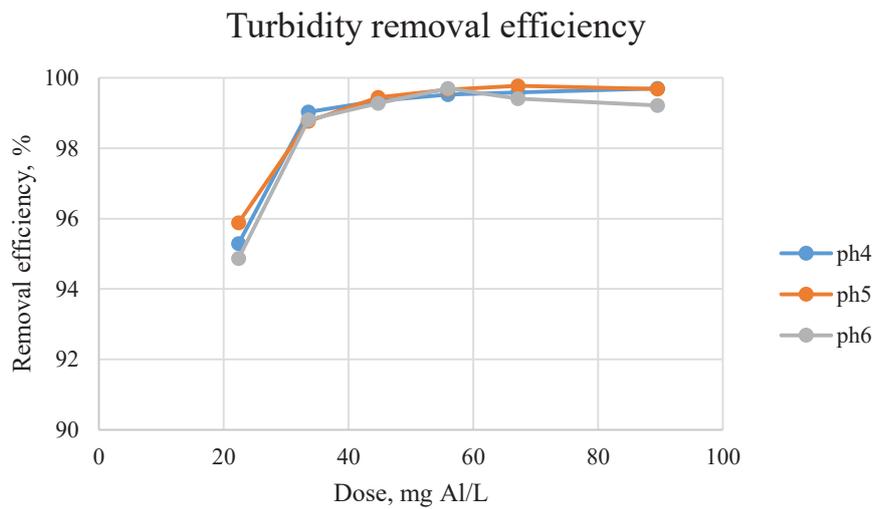


Figure 24. Dependence of turbidity removal efficiency on pH at constant flow (6 l/h)

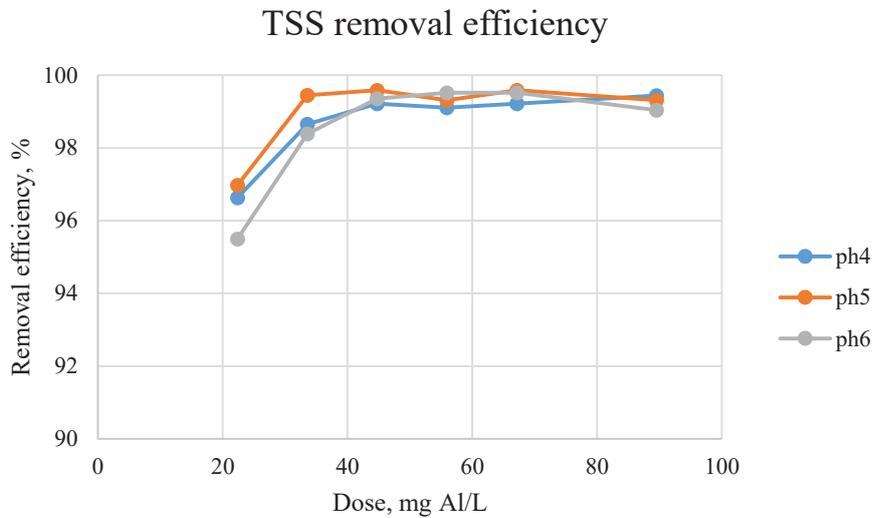


Figure 25. Dependence of TSS removal efficiency on pH at constant flow (6 L/h)

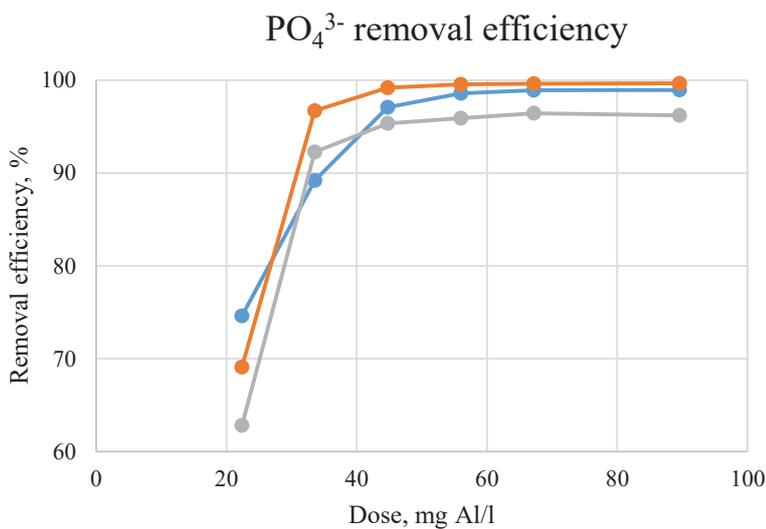


Figure 26. Dependence of orthophosphates removal efficiency on pH at constant flow (6 L/h)

By analyzing the graphs shown, one can come to the conclusion that the treatment efficiency of wastewater, which represented by the parameters such as (orthophosphates, TSS and turbidity) has become higher in comparison with model water, and, moreover, the optimum dose has become lower. This phenomenon may be due to an increase in the effective area of the anode, due to corrosion and dissolution of the electrode. After each analysis, the electrocoagulation cell was disabled, cleaned with a brush and surfactant, and washed with distilled water. However, it is recommended to polish the electrode with subsequent rinsing with a solution of nitric acid (1:1) and distilled water for the representativeness of the results. In our case, the EC unit worked in the real mode of sewage water treatment.

Due to these graphs, the optimal dose is in the range of 30-50 mg-Al/L, according to this fact, it was decided to use this range for the test with membrane treatment. Since the initial pH of WW after the biological treatment is 5.5 - 6, there is no strict requirement to adjust the pH prior to EC treatment process.

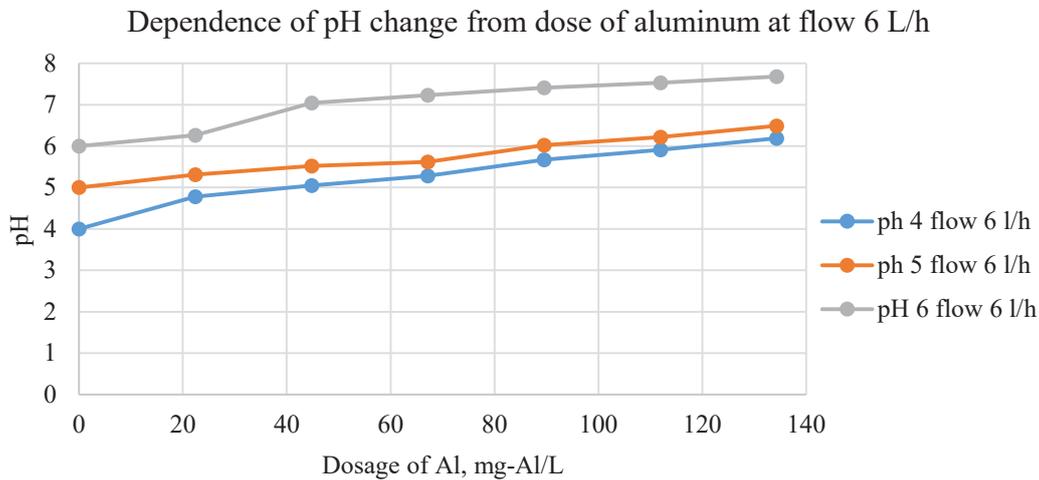


Figure 27. Dependence of initial pH change from the dose of Al at the constant flow

Figure 27 shows the dependence of the change in the initial pH value from the dose of Aluminum. pH tends to increase with the doses of Al, regardless of the initial one. This phenomenon is explained in detail in Experiment 1. In the case of wastewater, the rate of pH growth is significantly lower in comparison with model water. This indicates that WW has a higher buffer capacity, which positively reflects the efficiency of cleaning, and reduces the effect of the pH parameter. This allows carrying on the electrocoagulation at higher initial pH values than 4.

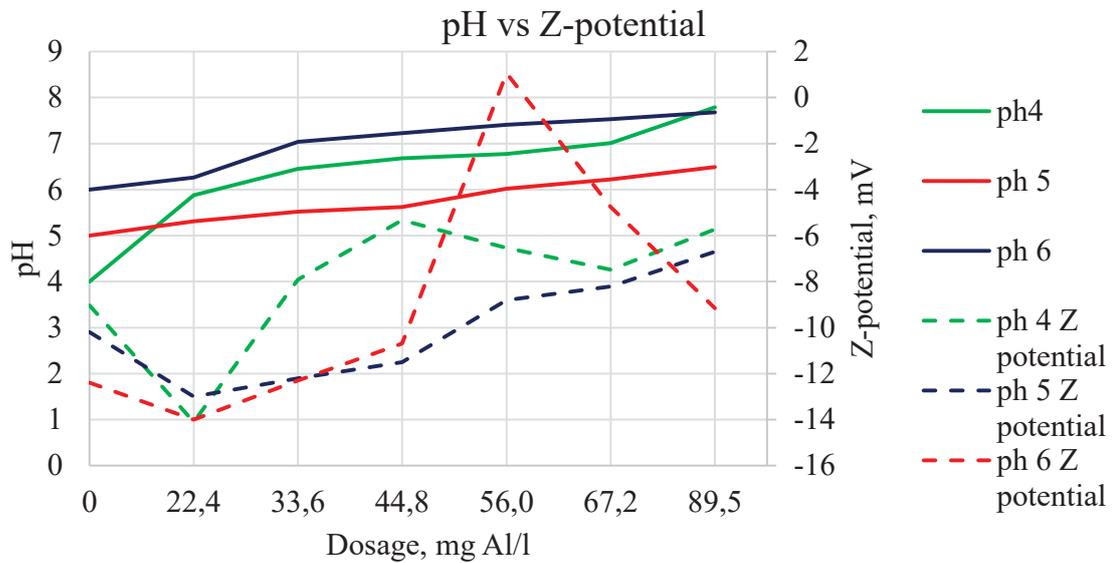


Figure 28. Dependence of pH and  $\zeta$ -potential on the dosage of Al at constant flow (6 L/h)

The  $\zeta$ -potential parameter characterizes the properties of double-layer and represents the stability of the colloidal system.  $\zeta$ -potential depends on the dosage of Al and subsequently aluminum hydroxide species since it is the coagulant (destabilization agent) of the colloidal system. At the same time the  $\zeta$ -potential also depends on the pH and because of the different simultaneous impact of these two parameters, the values of  $\zeta$ -potential is not linearly increasing but can decrease as well. Figure 28 shows the relation between the electrokinetic potential ( $\zeta$ -potential) and the pH of the wastewater. All measurements of  $\zeta$ -potential were made in triplicate. As it can be admitted, with the increasing of pH, electrokinetic potential decrease. With the addition of alkali particles tend to obtain a negative charge, and vice versa with acid addition, the charge will be neutralized, and in case of additional dosing of  $H^+$  ions the charge will be positive (Friedrich et al., 2007). In the pH range of wastewater from 4 to 5, the stability of the colloidal dispersion in wastewater without the addition of coagulant is the lowest, according to Figure 28.

The optimal dose which shows the high removal efficiency in almost all the cases is observed at the range 45-56 mg-Al/L (theoretical value). Electrokinetic potential can prove this fact. According to Figure 29, the  $\zeta$ -potential values are also significantly increased (from -14 mV to almost -6 mV). After the optimal range, these values are insignificantly increasing (except pH 5), but the treatment efficiency almost remained the same. For this investigation, the same municipal wastewater was used, adjustments of pH were done with NaOH and  $H_2SO_4$  respectively.

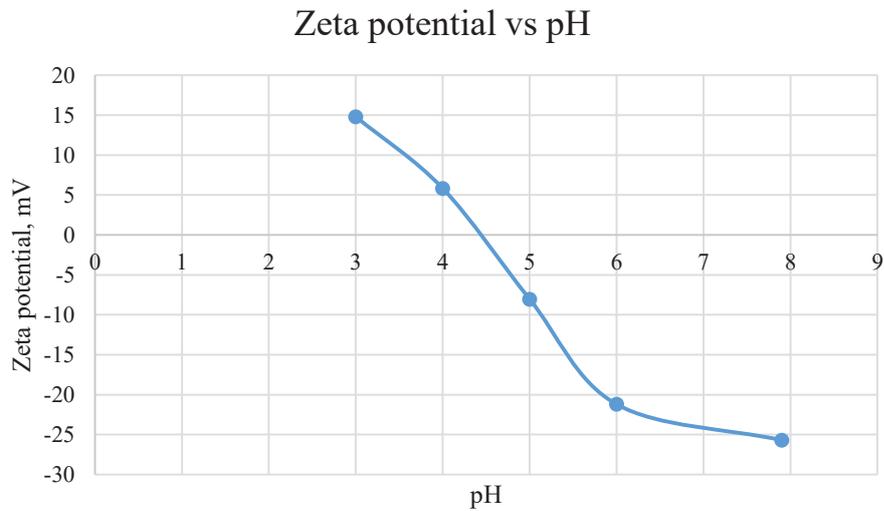


Figure 29. The relation between pH and Z-potential

The further step of our investigation was the work with the combination of biological-electrochemical-membrane treatment, to find out the optimal dose for the membrane fouling prevention (so-called recycle test) and other “wastewater treatment” parameters since the optimal dose does not always coincide with established at jar-tests.

### 3.5 Experiment 5. Combined treatment of real municipal WW

To estimate the optimal dose more precisely, it was decided to reduce the difference between the applied current to 0.1 A in the range of most probable finding of optimum. The theoretical doses of Al are presented in Table 6.

In order to check the efficiency of water purification at both stages, electrocoagulation and membrane filtration, the analysis of the main parameters was carried out in permeate and in the supernatant.

Table 8. The theoretical dose of Al vs applied current for the experiment with real WW + membrane filtration

Current, A (flow 6 l/h)	0.5	0.6	0.7	0.8	0.9	1	1.2	1.5	1.8
Current density, A/m <sup>2</sup>	4.17	5	5.83	6.67	7.5	8.33	10	12.5	15
Dose, mg-Al/l	27.98	33.58	39.17	44.7	50.36	55.96	67.15	83.94	100.72

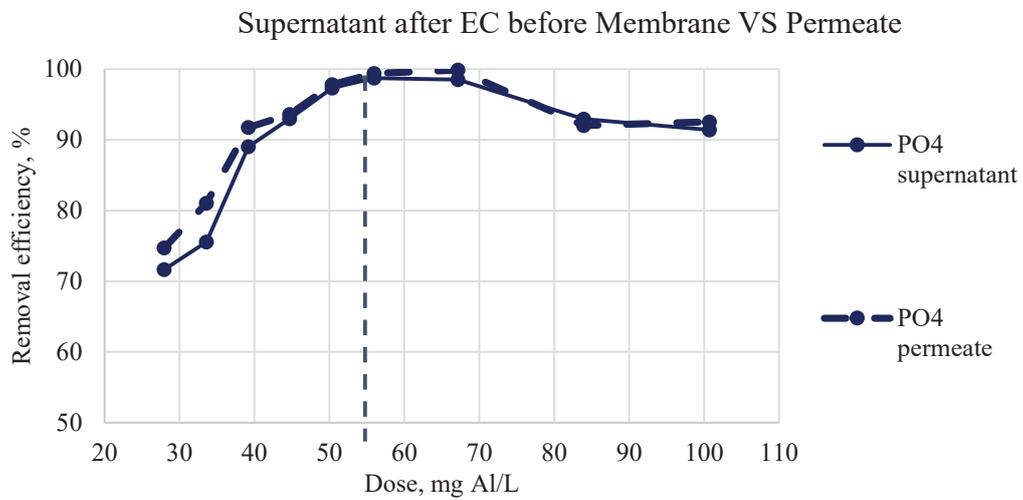


Figure 30. The relation between the removal efficiency of  $PO_4^{3-}$  and a dose of Al

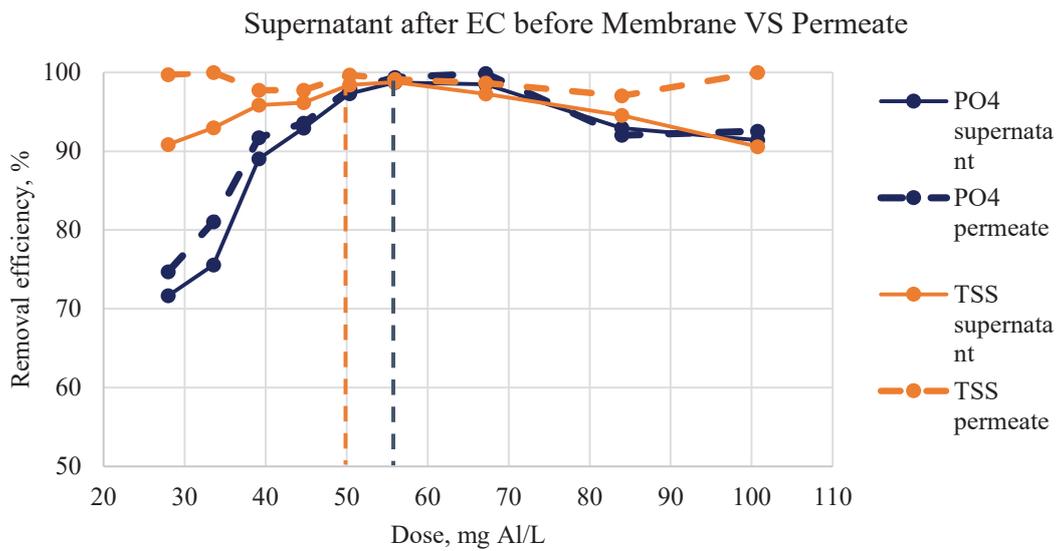


Figure 31. The relation between removal efficiency of  $PO_4^{3-}$ , TSS and dose of Al

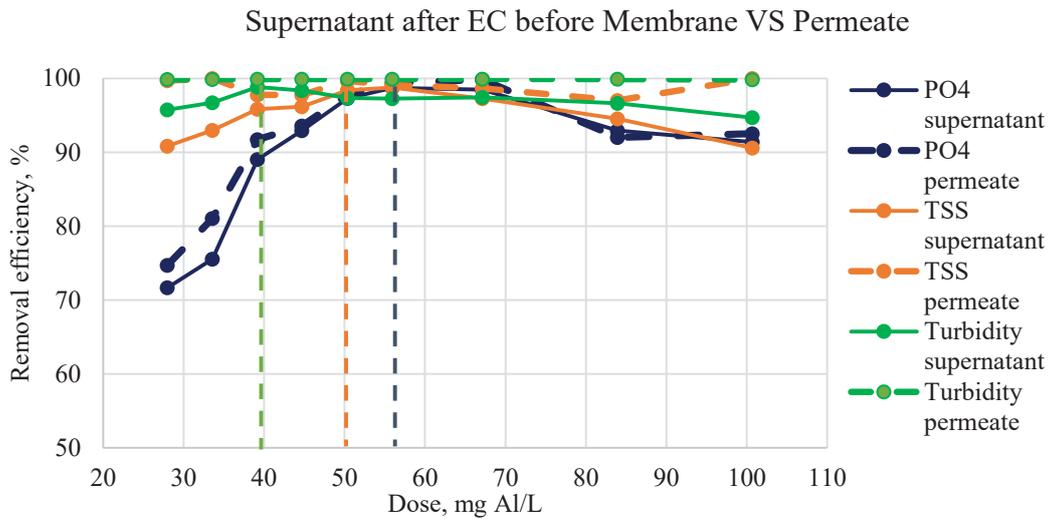


Figure 32. The relation between removal efficiency of  $PO_4^{3-}$ , TSS, Turbidity and dose of Al

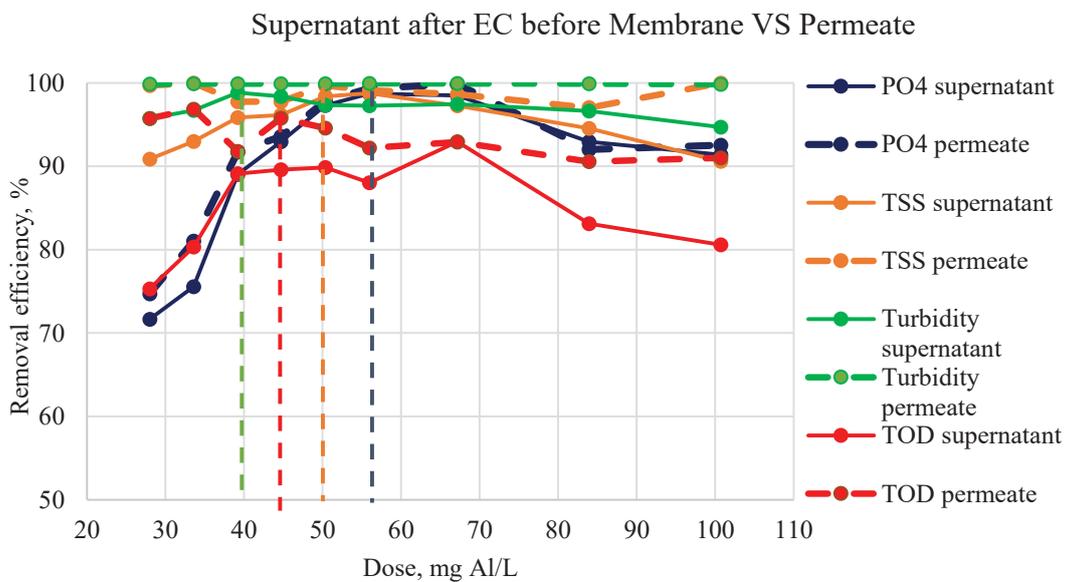


Figure 33. The relation between removal efficiency of  $PO_4^{3-}$ , TSS, Turbidity, TOD and dose of Al

From the graphs shown above (Figures 30-33), it is possible to summarize the values of the optimal doses for different measured parameters. These graphs show the results for the supernatant, which represents the removal efficiency after the electrocoagulation step and before the membrane module, while the values obtained for the permeate mean removal rates after the membrane element and the final quality of the water after all the stages.

As we can see from Figure 30, the optimal coagulant dose for high removal efficiency (up to 99%) of soluble type of phosphates is 55.96 mg Al/L. This dose is quite the same for supernatant and permeate, because of low dissolved phosphates treatment efficiency at membrane step. Based on Figure 31, we can see that the optimal coagulant dose for TSS removal is 50.36 mg Al/L, and

in both cases – supernatant and permeate it is possible to get more than 99% removal. For the achievement of turbidity removal for more than 99.9% we can see from Figure 32 the needed dose is 39.17 mg Al/L. The dose of Al which equals to 44.7 mg Al/L is an optimal in case of TOD removal for more than 89% in the supernatant (after EC step), and more than 95% in permeate (after EC + membrane step) (Figure 33).

So, it is possible to summarize, that the highest removal efficiency for all parameters such as OP, TSS, TOD and Turbidity can be achieved by the usage of coagulant in range of 39.17 – 55.96 mg Al/L. The electrocoagulation treatment is very efficient technic, and even without additional membrane filtration can produce a high-quality effluent. The small differences between the results, which are presented at Figure 33, prove that fact (except TOD). In a real case of the wastewater treatment plant, the main reason to install the additional membrane filtration is the reducing of the footprint by removing of large sedimentation tank.

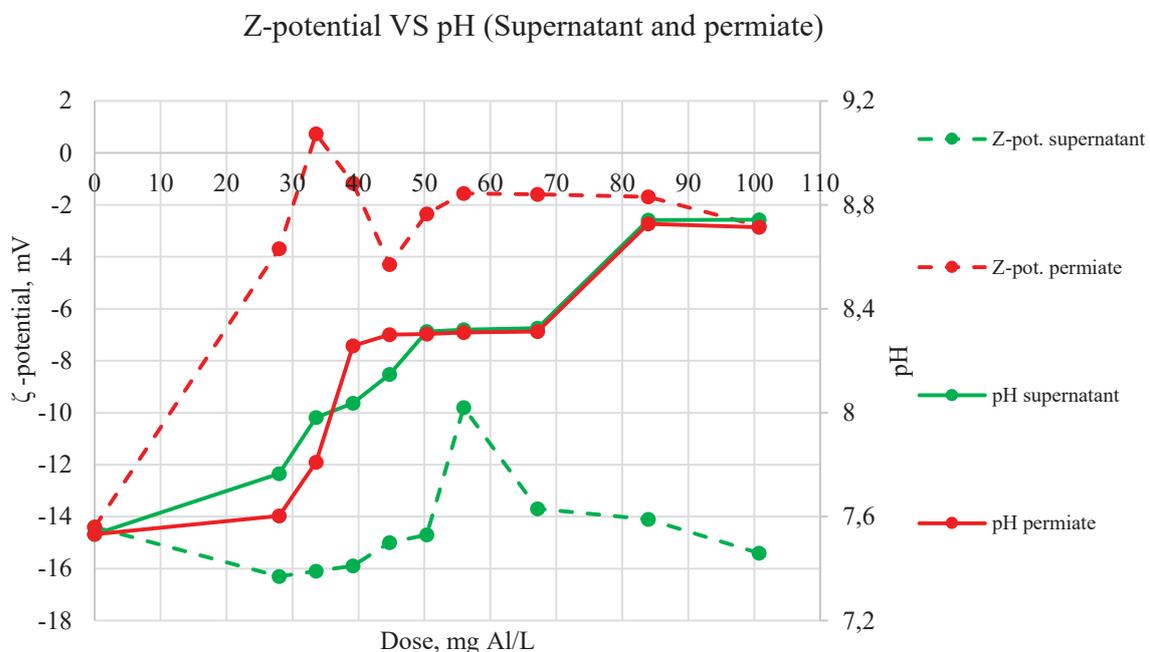


Figure 34. Dependence of pH and  $\zeta$  -potential on the dosage of Al at supernatant and permeate

Figure 34 shows us the change in the value of the zeta potential depending on the change in the pH of the wastewater. As described earlier, this relationship is unconditional. The main point on this graph, which requires attention, is the difference in  $\zeta$  -potential for the supernatant and permeate. This significant difference can be explained by the ability of fine particles removal from the water after the electrocoagulation process by the membrane filtration. It can be seen, that from the very beginning the  $\zeta$  -potential values for the permeate are close to the isoelectric point at which the surface charge of the water is equal to zero, which indicates low stability of colloidal dispersion. Even thou, the  $\zeta$  -potential values for the supernatant are high, the treatment efficiency remains at a high level. The slight increase in electrokinetic value can be observed at the dosage of 55.96 mg-

Al/L, which is optimal for the suspended solids removal. After this point, it decreases again, which is reflected at TSS removal quality. The same tendency was noticed for the TSS removal efficiency in permeate at dose 33.58 mg-Al/l where it was achieved the absolute maximum – 100%. The  $\zeta$ -potential value at this point is very close to zero (+0.7 mV).

### 3.5.1 Total recycle test

For the Total recycle test, the following conditions were met:

- Varying current (Al doses)
- The volume of treated water  $\approx$  4.0 L
- Duration of membrane filtration 60 minutes with recirculation speed 6 L/h

Samples of initial WW, water after biological treatment, supernatant after EC, permeate after membrane were taken and analyzed.

The efficiency of a combination of EC treatment/membrane filtration was estimated by the filtration cycle length or total retention time (TRT). First, all the obtained curves were analyzed and by the prior estimation, it was decided to use the range of initial  $TMP_{in} - 1,5 \cdot TMP_{in}$  (transmembrane pressure) in order to avoid the use of very short intervals and very long extrapolations. By using a linear trend equation, it is possible to define the:

$$y = -0.0084x - 9.6948$$

where:

x- time, s;

y – pressure, kPa.

In the initial period of time (x=0), the pressure is equal to:

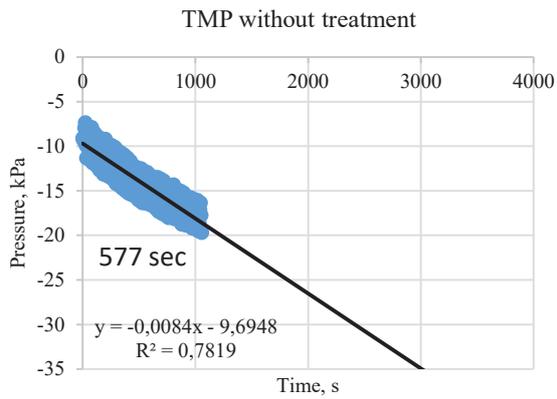
$$y = -9.6948$$

If the initial pressure was increased in 1.5 times, then time is equal to:

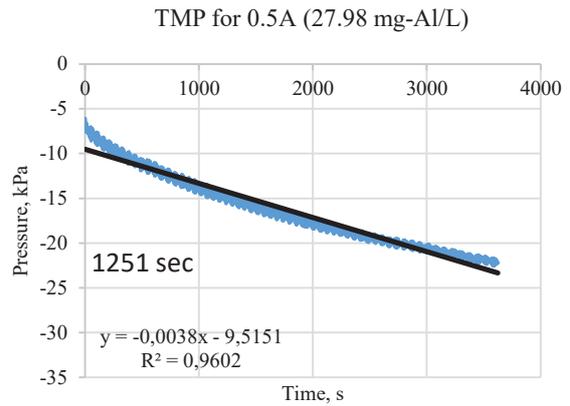
$$\begin{aligned} y &= -0.0084x - 9.6948 \\ -14.5422 &= -0.0084x - 9.6948 \\ x &= 577 \text{ s} \end{aligned}$$

The pressure value was automatically recorded at a frequency of 12 times per minute. As can be seen from the graphs below, the recorded data does not look like a straight line, but as some line with fluctuations on the segment. This phenomenon can be explained by the work of a peristaltic pump. At the moment when the silicon tube is clamped with the bearings of the pump, the pressure is higher than when the tube is in the intermediate position between the two roller bearings, but not completely clamped (the period of partial relaxation). In addition, these fluctuations reduce the correlation coefficient without visible for the reasons (Naje et al., 2017).

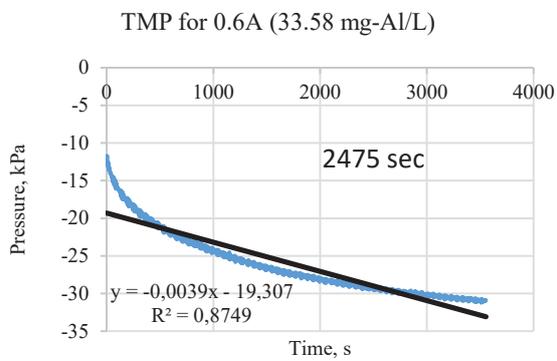
Another phenomenon observed on some of the graphs is the higher rate of transmembrane pressure decreasing over a short initial period of time, followed by normalization, which can be explained by the faster clogging of the small pores of the membrane, at the beginning of the filtration process.



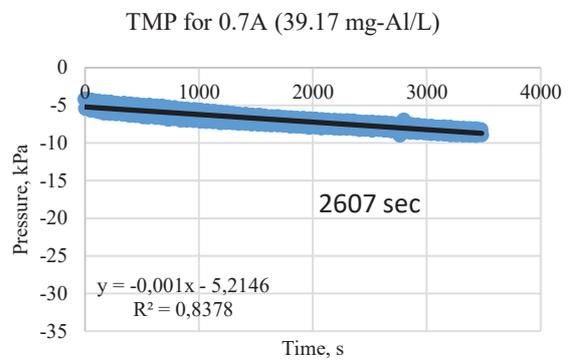
a



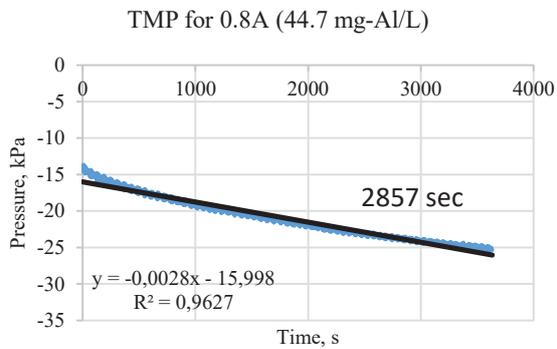
b



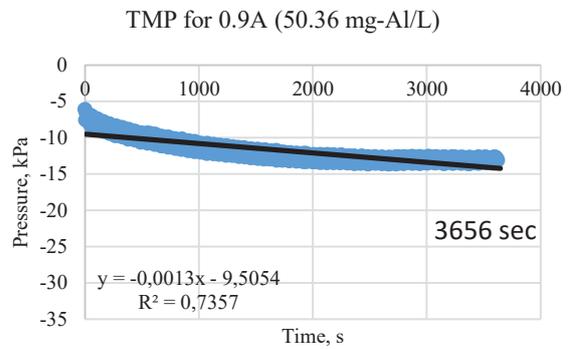
c



d



e



f

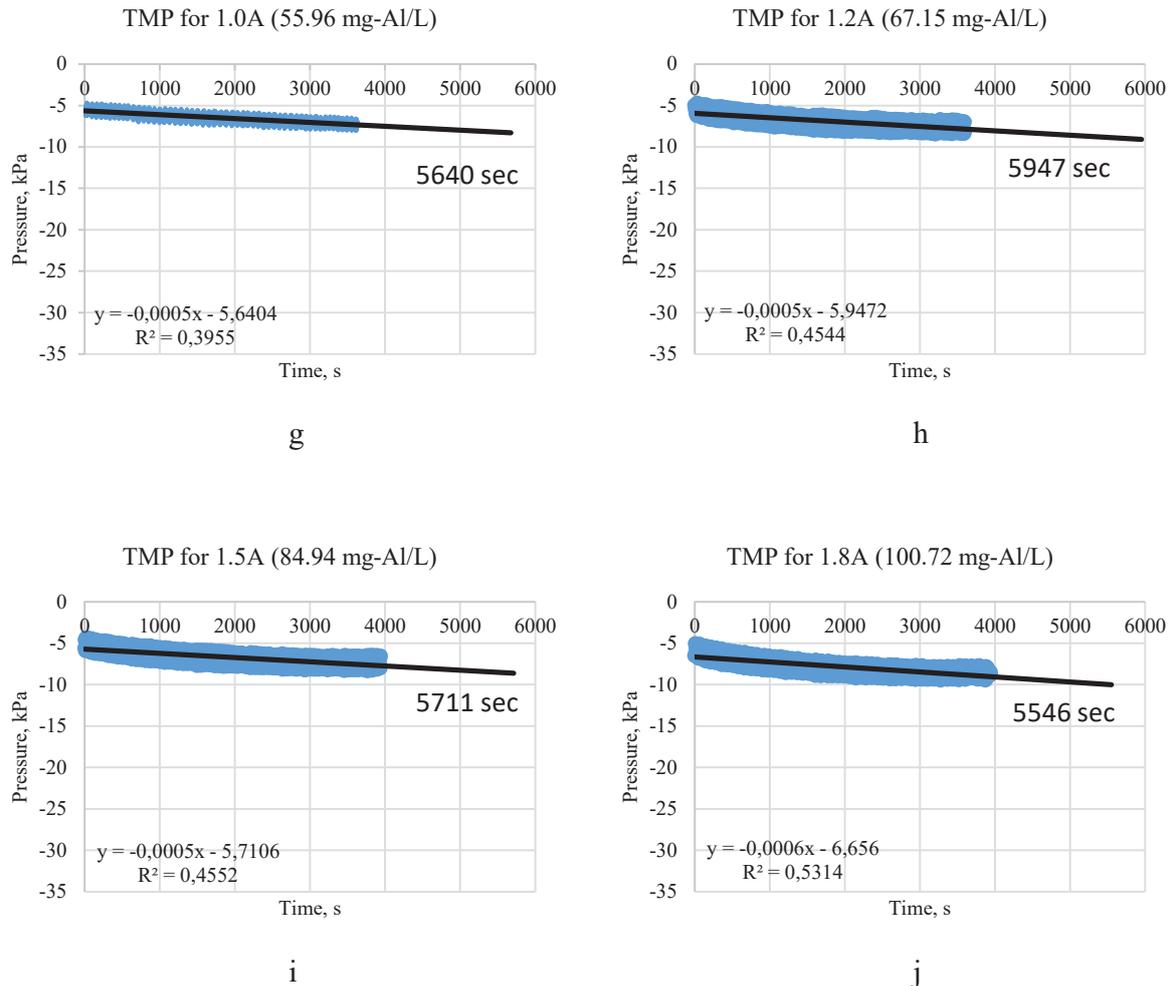


Figure 35. (a-j) Transmembrane pressure change within time in treated water obtained at different dosages of Al

The figure below shows a change in the filtration length in the range of  $TMP_{in}-1.5TMP_{in}$  from the dose of aluminium used for water purification (modifications of the mixer liquor). As can be seen from the graph, with an increase in the dose of Aluminium, the filtration time increases, which indicates a positive effect of the electrocoagulation process on the membrane fouling. The optimal dose of Al for fouling mitigation is 67.15 mg-Al/L or current density – 10 A/m<sup>2</sup>. Even thou, the dose of 55.96 mg-Al/L show very high results in terms of the combined process since this dose was appropriate for PO<sub>4</sub><sup>3-</sup>, Turbidity, TSS, TOD efficient removal and significantly extend the filtration cycle.

Usually, the positive effect of electrocoagulation on membrane filtration is explained by three mechanisms:

- the dissolving of a metal cation from the anode into a solution accompanied by its hydrolysis and the formation of monomeric and polymeric hydroxo-complexes of metal (in our case, aluminium), due to this process, the neutralization (destabilization) of negatively charged colloids and the sorption of the suspended and dissolved components take place;
- control of foulant deposition at the membrane surface;

- chemical oxidation of bound organic polymers and biopolymers.

It has been reported, that during the electrochemical modification of mixed liquor process, electric field modify the physicochemical properties of flocks: the inorganic suspended solids form higher density flocks in comparison to activated sludge flocks, which make them more stable, improve a mixed liquor stability (because of the smaller content of bounded water), and reduce a membrane fouling. Additionally, the formation of hydroxyl radical ( $\cdot\text{OH}$ ), which occur during electrocoagulation, act as strong oxidant and enhance the removal efficiency of organic compounds by breaking down the proteins and carbohydrates into compounds with lower molecular weight, thus reduce the bio- and organic fouling.

The flat sheet ceramic membrane, which was used in experiments is made from silicon carbide (SiC). This membrane has the hydrophilic nature and mixed liquor flocs are hydrophobic which to high extend reduce their interaction.

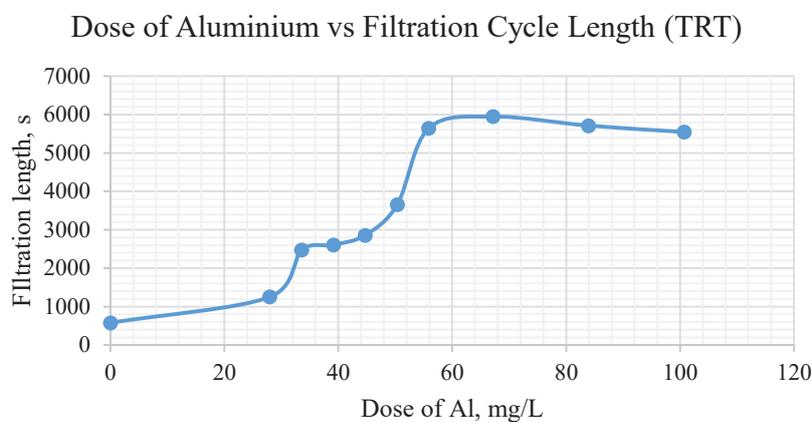
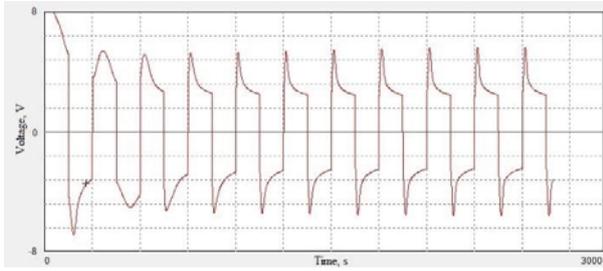


Figure 36. Dependence of filtration length in range of  $\text{TMP}_{\text{in}}-1.5\text{TMP}_{\text{in}}$  on the dose of Al, which was applied for water purification

### 3.5.2 Dependence of voltage change within time

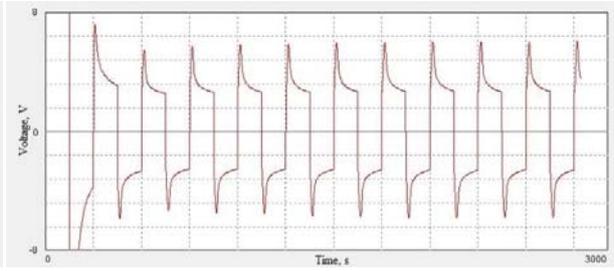
The graphs depicted below (Figure 37) reflect the change in voltage over time. The data presented for the study of the combination of electrocoagulation and membrane filtration were recorded in the charts in a special program for power supply. At the beginning of the process, the voltage reached 12 and more volts (not shown on the graph) after which it stabilized. This can be explained by the fact that at the beginning of the process, the reactor is not completely filled with water, which greatly increases the electrical resistance of the medium. The periodic change in the sign of the curves on the graphs reflects the change in the polarity on the electrodes. With an increase in the value of the current, the value of the voltage increases respectively, which contributes to accelerating the dissolution of the cathode. The approximate duration of the process is 40-45 minutes, to fill the reactor where the membrane is installed.

Voltage vs time (0.5 A)



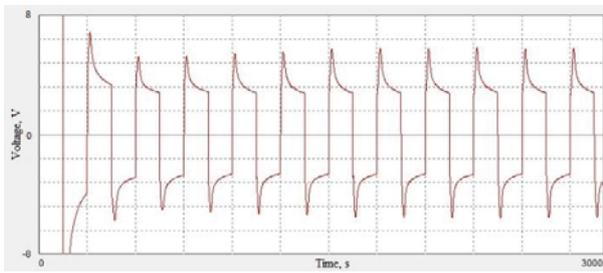
a

Voltage vs time (0.6 A)



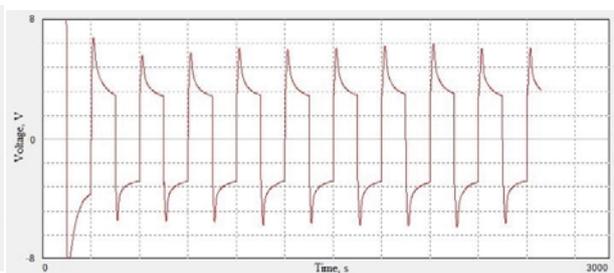
b

Voltage vs time (0.7 A)



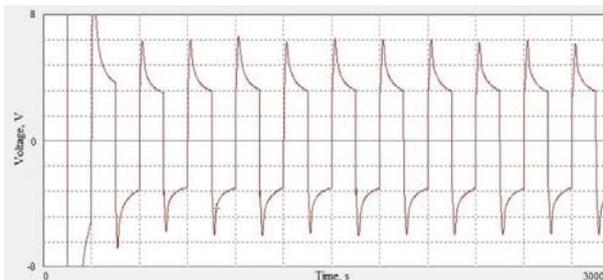
c

Voltage vs time (0.8 A)



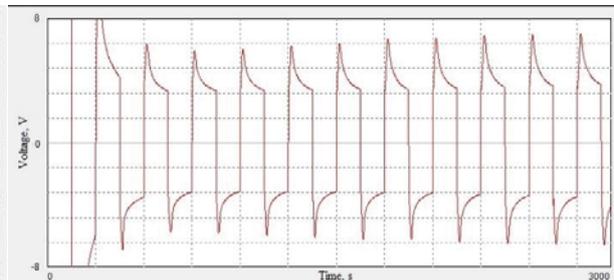
d

Voltage vs time (0.9 A)



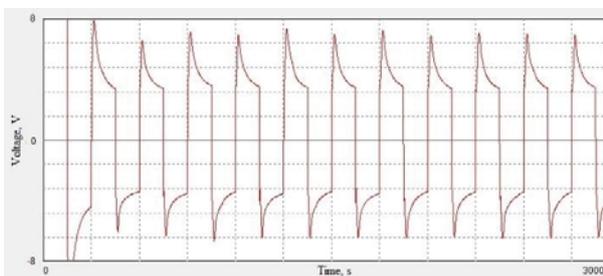
e

Voltage vs time (1.0 A)



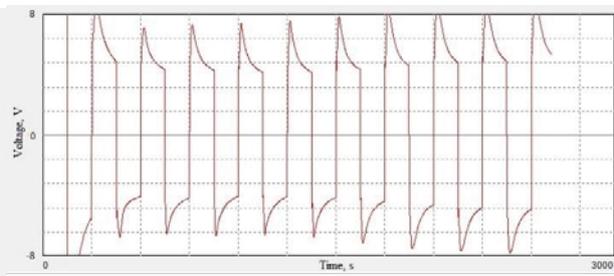
f

Voltage vs time (1.2 A)



g

Voltage vs time (1.5 A)



h

Voltage vs time (1.8 A)

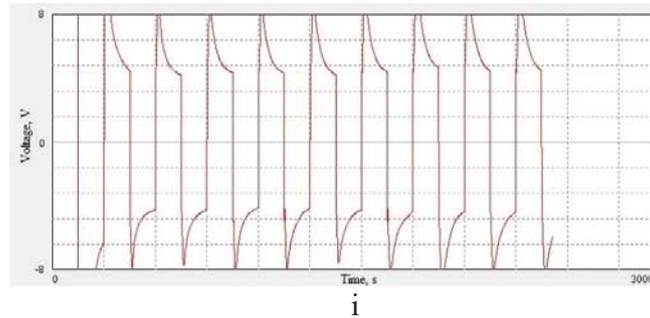
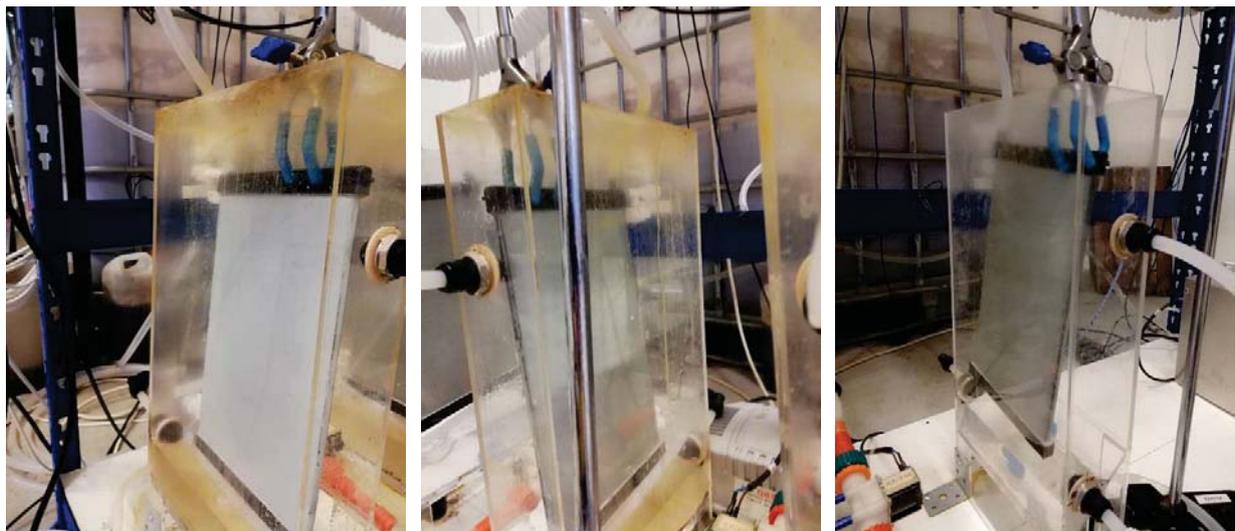


Figure 37. (a-i) Voltage change within time during electrocoagulation process at different dosages of Al

The photographs below (Figure 38) represent the appearance of the reactors in which the membranes were located after all the performed experiments. As can be seen, the reactor in which the water passed the electrocoagulation treatment is much less polluted than reactors that were filled with water, which was purified by inorganic coagulant and natural coagulant respectively.



Inorganic coagulant

Natural coagulant

Electrocoagulation

Figure 38. Comparison of the reactors appearance after the recycle test

In Figure 39, the state of the electrode material after a series of electrocoagulation experiments is demonstrated. The images include all three components, which clearly show that the aluminium material was not smooth due to the constant presence in the water, and as a result of corrosion processes. Aluminium, although it is an active metal, is distinguished by fairly good corrosion properties.

The corrosion resistance of aluminium electrodes depends on a number of factors, such as pH, purity of the metal, corrosive environment, composition and components of the wastewater passing through the installation, process temperature values, etc.



Figure 39. Appearance of electrodes after all the tests

### 3.6 Energy consumption analysis

To assess the economic feasibility of using the electrocoagulation process using aluminium as the electrode material (in our case), it is necessary to pay attention to the calculation and final price of cleaning 1 m<sup>3</sup> of wastewater under different conditions. There are formulas which were used below, as well as prices relevant to our research work, also a table with the final calculations of the cost of the process (Table 9).

For calculation and determination of *electrical energy* (kWh/m<sup>3</sup>) and *Al-electrode consumption* during coagulation process with using of EC method, the next equations should be used (Omwene & Koby, 2018):

$$C_{energy} = \frac{i \times t_{EC} \times U}{v}$$

where:

- $i$  – applied current (A);
- $t_{EC}$  – time of electrode consumption (h or sec);
- $U$  – cell voltage (V);
- $v$  – volume of WW in EC unit (m<sup>3</sup>);

$$C_{electrode} = \frac{i \times t_{EC} \times M_{Al}}{z_{Al} \times F \times v}$$

- $M_{Al}$  – molecular weight of Aluminum (26.98 g/mol);
- $z_{Al}$  – transferred electrodes (3);

- $F$  – Faraday’s constant (96.487 C/mol).

The experiment was done for 30 minutes (1800 sec) per each run. It is possible to calculate the cell voltage for each current density respectively.

For achievement of the highest removal efficiency (92-99.9%) of OP with 55.96 mg Al/L (applied current 1 A) it is needed 2.64 kWh/m<sup>3</sup>; of TSS with 50.37 mg Al/L (applied current 0.9 A) it is needed 2.29 kWh/m<sup>3</sup>; of turbidity with 39.17 mg Al/L (applied current 0.7 A) it is needed 1.5 kWh/m<sup>3</sup>; of TOD with 67.15 mg Al/L (applied current 1.2 A) it is needed 3.04 kWh/m<sup>3</sup>; of optimal TMP value with 55.96 mg Al/L (applied current 1.2 A) it is needed 3.04 kWh/m<sup>3</sup>.

Also, the *cost of the operating part*, which includes material cost and electricity cost was calculated with the next equation:

$$\text{Operating cost} \left( \frac{\text{NOK}}{\text{m}^3} \right) = \alpha \times C_{\text{energy}} + \beta \times C_{\text{electrode}}$$

where:

- $\alpha$  – electrical energy cost, in our case it is 55.2 øre/kWh (“SSB,” 2019)
- $\beta$  – cost of the Al electrode, in our case we took the maximum dosage of Al from optimal dosages for all parameters (67.15 mg Al/L, which means 0.067 kg Al/m<sup>3</sup>) and calculated cost of electrode consumption based on price for kg Al, which equal to 1.78 Euro = 17.39 NOK.

Usually, it is also needed to add the cost of each used chemical reagent in an EC step (NaOH, H<sub>2</sub>SO<sub>4</sub>, etc.), but in our case, we didn’t use chemicals for pH adjustment or something else, so there is no need in using of this option.

Table 9. Energy consumption, electrode consumption and OC of EC step

Current, A	Current density, A/m <sup>2</sup>	Time, sec	Cell voltage, V	C <sub>energy</sub> , kWh/m <sup>3</sup>	C <sub>electrode</sub> , kg/m <sup>3</sup>	Operating cost, NOK/m <sup>3</sup>
0.5	4.17	1800	3.65	0.98	7.77	9.61
0.6	5		3.93	1.28	9.32	11.59
0.7	5.83		4.03	1.50	10.87	13.53
0.8	6.67		4.12	1.73	12.43	15.47
0.9	7.5		4.61	2.29	13.98	17.59
1.0	8.33		4.79	2.64	15.53	<b>19.60</b>
1.2	10		4.91	3.04	18.64	23.45
1.5	12.5		6.06	4.52	23.30	29.71
1.8	15		6.36	5.80	27.96	35.86

As we can see from the Table 9, the constant time of electrocoagulation run and increasing of current density values are lead to increasing of cell voltage values as well, it means the increase

of energy consumption, electrode consumption and operating cost in general also. The performance of EC process should be noted, because of high treatment efficiency (removal of OP, TSS, Turbidity, etc.) with the eventually acceptable price of the operating cost, as well as other consumption costs.

## Conclusions

In current research work, the effect of electrochemical treatment of mixed liquor in submerged ceramic membrane bioreactor has been investigated.

In experiment 1, 2 and 3 the influence of operating parameters was checked and explained, such as:

- Current density (dose of Al);
- pH (4 - 7);
- Flow (3L/h, 6L/h, 9 L/h, 12 L/h);
- Recirculation through the EC unit (2 cycles, with recirculation flow 6 L/h, 9 L/h, 12 L/h).

In the fourth experiment with real wastewater, the optimal parameters, which were established in the experiments with medium-soft model WW, were validated. The following conditions were selected for the combined treatment process, according to obtained results:

- pH 6, due to higher buffer capacity of municipal WW after biological treatment;
- The flow of 6 L/h, because of negligible difference in treatment efficiency in comparison to the other two flows (9 L/h, 12 L/h);
- Current density in a broad range with small differences between dosages of Al.

At the optimal conditions it is possible to achieve the high removal efficiency of  $\text{PO}_4$  (up to 99%); TSS (up to 98%); Turbidity (up to 98.5%); TOD (up to 70%), which make this process very perspective in terms of WW treatment. An excellent treatment efficiency of WW also were proves by other researches (Attour et al., 2014) (Franco, Lee, Arbelaez, Cohen, & Kim, 2017) (An et al., 2017) (Hashim et al., 2019). Electrochemical treatment of mixed liquor in submerged ceramic membrane bioreactor demonstrated a very high results in extension of the membrane filtration cycle at the optimal dose of Al (67.15 mg-Al/L or current density – 10 A/m<sup>2</sup>) it is possible to expand the filtration cycle time in 10.3 times in the range of  $\text{TMP}_{\text{in}} - 1.5 \text{ TMP}_{\text{in}}$ . However, it is recommended to use the dose of 55.96 mg-Al/L, which show very high results in terms of the combined process since this dose was appropriate for  $\text{PO}_4^{3-}$ , Turbidity, TSS, TOD efficient removal and extend the filtration cycle up to 10 times (filtration length is 5640s).

Usually, the EC treatment is associated with high energy consumption and costs for sacrificial electrode change. In this experiment, we calculated the operational costs, which include these two parameters (energy and electrode costs). It was established, that for the optimal dose of Al (55.96 mg-Al/L), which show a very high level of WW treatment, the operating cost is 19.6 NOK per cubic meter of sewage water, which was calculated in the economic section. The resulting price of water treatment approximately corresponds to the price obtained in other studies (Omwene & Koby, 2018).

Preceding from the foregoing, it can be concluded that the combined IFAS/EC/MBR process of wastewater treatment is highly effective, modern, and very close to water circularity and reuse, at least for technical purpose. Obtained water quality meet all the EU requirements for municipal wastewater. The EC treatment itself is an excellent alternative to conventional coagulation and could be even more suitable in particular conditions.

For the future investigation, it is recommended to check the treatment efficiency of Iron electrodes in this type of EC unit. Investigate the influence of polarity change. Compare the theoretically calculated dose with really obtained. It is also interesting to study the characteristics of EC sludge such as Hydrophobicity, CST, MLSS and compare them with the sludge obtained by conventional coagulation, to clearly define the influence of this treatment on membrane fouling mitigation.

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