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Electrocoagulation for Treatment of Tunnel Wash Water: A Proof of Concept

Elektrokoagulering som reinsemetode for tunellvaskevatn: Eit dugleiksprov

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Abstract

There are over 1000 road tunnels in Norway. Inside the tunnels, particles from wear and tear of asphalt, tires, brakes, vehicle bodies, and more will accumulate. The tunnels are washed 2-12 times every year, in part to flush out these particles, and the result is a wash water with high levels of pollutant concentrations and in turn adverse effects for aquatic ecosystems.

The environmental impact of tunnel wash water (TWW) and possible treatment solutions was one of the central aspects of the research project Norwegian Road Water (NORWAT), funded by the Norwegian Public Roads Administration. A report from this project explored possible mobile treatment systems, and suggested electrocoagulation (EC) as a possible treatment process, but the method has not previously been tried on TWW.

EC is a chemical treatment process in which electric current flows through metal electrodes submerged in the raw water, dissolving metal into the water. The dissolved metal forms metal hydroxide flocs, which interacts with colloid and particles in the water, creating easily separable flocs. At the same time, hydrogen gas is produced. The gas bubbles attach to flocs, which makes them rise and creates a sludge layer on top of the water.

EC was tested in a bench-scale reactor with aluminum electrodes, using a response-surface central composite design to test a range of treatment times of 6-35 minutes and current densities of 15-89 A/m^2. Metal removal rates of > 90 % for all metals except aluminum was achieved on all configurations. No settings for optimizing simultaneous removal of all metals were found.

Effluent aluminum concentrations spanned 9 000 to 83 000 µg/l, from an initial concentration of 48 000 µg/l. Sludge production also varied a lot, from 10-63 %. Lowest sludge production came from experiment runs with short duration, low current density, or a combination of these. This coincides with the lowest effluent aluminum concentrations.

The study suggests that 10-15 minutes' treatment time is ideal, because only small removal rate increases were achieved after this point for nickel, while other removal rates did not increase, and sludge production and effluent aluminum concentration increased at longer durations. EC can therefore treat TWW in about half the time needed by other fast methods studied, namely chemical precipitation using iron, aluminum, and lime based coagulants, with comparable treatment efficiencies.

The experiments done in this study are a proof of concept for EC treatment of TWW. Further studies need to convert this process to a flow-through reactor. If good results are achieved in a flow-through reactor, a scaled-up version of the system must be tested, before EC treatment of TWW can be deemed suitable for deployment.

Samandrag

Det norske vegnettet har over 1000 tunellar. Inni desse akkumulerast partiklar frå slitasje av m.a. karosseri, dekk, bremser og asfalt. Tunellane vaskast 2-12 gongar kvart år, dels for å skylje ut alle desse partiklane, og resultatet er eit vaskevatn med svært høge forureiningskonsentrasjonar, og ei negativ påverking på livet i fjordar og vassdrag.

Tunellvaskevatn (TVV) sin miljøpåverknad og teknikkar for å reinse dette vatnet var eit av fokusområda til forskingsprosjektet Norwegian Road Water (NORWAT), i regi av Statens Vegvesen. Ein av rapportane frå dette prosjektet var ein litteraturstudie av moglege mobile reinseløysingar for TVV. Her blei elektrokoagulering (EK) foreslått som eit alternativ, men EK har tidlegare ikkje blitt forsøkt brukt til reinsing av TVV.

EK er ein kjemisk reinsingsmetode kor elektrisk straum ledast gjennom elektrodar i vatnet som skal reinsast, og løyser opp metall frå anodane i vatnet. Det oppløyste metallet oksiderast til metallhydroksid. Metallet dannar så flokkar, som kjem i kontakt med partiklar og kolloidar i vatnet, og går saman til større partiklar, som er lettare å separere ut frå vatnet. På same tid dannast det hydrogengass på katodane. Gassboblene festar seg til fnokkane, og får dei til å flyte opp og danne eit slamlag på vassoverflata.

EK blei testa i ein labskalareaktor med aluminiumselektrodar. Forsøket blei designa ved å nytte respons-overflate-metodologi. Reinsingsmetoden blei testa med forskjellige oppsett, frå 6-35 minuttar og strøymingstettleikar på 15-89 A/m^2. Reinsingsgrad for metall blir målt til > 90 % for alle metall utanom aluminium, ved alle konfigurasjonar. Ingen innstillingar blei funne som ga optimal fjerning av alle metall samtidig.

Utløpskonsentrasjonen for aluminium var på 9 000 til 83 000 µg/l, frå ein startkonsentrasjon på 48 000 µg/l. Slamvolumet varierte også veldig, frå 10 til 63 % av vassvolumet etter reinsing. Lågast slamproduksjon blei observert for dei forsøka med kort tid, låg spenningstettleik eller ein kombinasjon av desse. Dette fell saman med dei forsøka som ga lågast utløpskonsentrasjon av aluminium.

Studien visar at ei opphaldstid på 10-15 minuttar i EK-reaktoren latar til å vere ideell. Berre små aukingar i reinsingsgradar for nikkel vart observert for lengre opphaldstider, medan reinsingsgradar for andre metall ikkje vart betre med lengre tid. Slamvolumet og aluminiumskonsentrasjon auka ved lengre opphaldstider. EK viste seg å kunne reinse TVV på om lag halvparten av tida tidlegare studerte metodar har brukt. Kjemisk felling med jern-, aluminium- og kalkbaserte fellingsmiddel trong om lag ein halvtime på reinsing og separering, og hadde samanliknbare reinsegradar.

Denne studien er eit dugleiksprov for EK til TVV-reinsing. Det er bruk for meir forsking for å teste denne metoden i ein gjennomstrøymingsreaktor. Om dette synar gode resultat, må også forsøk på oppskalering gjerast, før ein kan fastslå at teknologien kan nyttast til reinsing av TVV.

Preface

This thesis is written at the Faculty of Science and Technology at the Norwegian University of Life Sciences (NMBU) during the spring of 2017, ending my master's program in Water and Environmental Technology. My supervisor has been Arve Heistad.

This thesis was supposed to be about tunnel wash water, but not electrocoagulation, as I had never heard about electrocoagulation. I read about it in a Norwegian Public Roads Management report. It sounded promising. Hence this subject.

I want to thank Arve Heistad for inspiration throughout my five years of studies, for giving me freedom to go out on a limb for my thesis subject, and for guidance on my work. Thanks also go out to Fasil Ejigu Eregno for helping me with theoretical experimental design, and Melesse Eshetu Moges for help with practical experimental design.

A big thank you to Aurora, for support, laughter and patience.

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Glossary

Coagulant	A chemical substance added to raw water to destabilize colloidal suspensions
Colloid	A particle < 1 µm that does not settle in a fluid
Current density	Electrical current divided by the anode surface it flows though, measured in Ampere/m^2
Floc	Aggregated colloids forming an easier separable particle
NORWAT	A research program by the Norwegian Public Road Authorities aimed at reducing environmental impact from road water runoff
Pollutant	A substance that causes adverse effects in the environment it is introduced into
Precipitation	Separating a dispersed substance from the water it is suspended in
Raw water	Water that is to be treated, but has not been treated yet
Response-surface	An experimental design methodology aimed at correlating input parameters with output parameters by varying multiple input parameters at a time
Solubility	The degree to which a substance can be dissolved in water
Turbidity	A measure of the haziness of a fluid, caused by particles suspended in the fluid

List of abbreviations

AADT	Annual average daily traffic
AA-EQS	Annual average environment quality standard
BP-P	Bipolar-parallel
CCD	Central composite design
EC	Electrocoagulation
EQS	Environment quality standard
MAC-EQS	Maximum admissible concentration environment quality standard
MP-P	Monopolar-parallel
MP-S	Monopolar-serial
NPRA	Norwegian Public Road Authorities
PAH	Polycyclic aromatic hydrocarbons
SS	Suspended solids, a measure of particulate matter in suspension in water
TWW	Tunnel wash water

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1 Introduction

Road runoff pollutants has increasingly attracted attention the last decades, due to the lack of treatment of organic pollutants and metals present in this water. Increasing road traffic has increased the pollutant load, and increased scrutiny of road runoff disposal.

A subsection of road runoff that has gotten less attention is the water from tunnels washing events. In Norway, there are more than 1000 tunnels, and these are washed 2-12 times each year, depending on annual average daily traffic (AADT). During washes, accumulated pollutants in the tunnel are discharged with the wash water, together with detergents used for washing. This results in an effluent with very high concentrations of metals like copper, zinc and lead, and various polycyclic aromatic hydrocarbons (PAHs) and nitro-PAHs (Petersen et al. 2016).

There has been some research into treatment of tunnel wash water (TWW). Several sedimentation ponds have been built in conjunction with tunnels, but Meland et al. (2010a) showed that runoff from such a pond has clearly elevated levels of pollutants after a tunnel wash event. Paruch and Roseth (2008) looked at the use of organic sorbent materials for the treatment of TWW, while others have tried chemically enhanced sedimentation (Hallberg et al. 2014) and chemical precipitation with flocculation with different flocculants (Garshol et al. 2016a; Nersten 2016). In a literature review of possible mobile treatment solutions for TWW, Garshol et al. (2016b) suggest, among other solutions, electrocoagulation (EC) as a treatment option that has been proposed by contacted contractors, but has so far not been tested on TWW.

1.1 Research objectives

The goal of this study was to test the feasibility of electrocoagulation treatment of TWW, and assess its suitability for use in a mobile facility. Hence, the following research objectives were examined:

- Treatment efficiency for metal pollutants
- Optimal treatment duration and current density
- Minimizing sludge production
- A brief look at a possible upscaling of the treatment system, with regards to space requirements compared to treatment capacity (l/h/m^3)

1.2 Tunnel wash water

In Norway, there are numerous tunnels with a varying degree of annual average daily traffic (AADT). Like on normal roads, there is a production of potentially toxic particles in tunnels, from tear and wear of asphalt and car parts, combustion products, oil residues and road salt. Unlike other roads, the pollutants inside tunnels are not regularly flushed away by rainwater, and will accumulate on the walls, ceiling, asphalt, and technical installations. The accumulation of particles decreases visibility because of dusty air, worsens the esthetical experience of the tunnel, gives a harmful work environment for technical staff, and might lower the lifespan of technical equipment in the tunnel (NPRA 2014). To mitigate these effects, the Norwegian Public Road Authorities (NPRA) has set guidelines for the frequency of different kinds of tunnel washes, depending on the AADT, as outlined in Table 1.

The technical wash is for signs, lights, emergency stations, and other equipment, as well as the roadway and road shoulder. Full wash also includes collecting dust and other particles before washing, emptying of gully pots and wash of walls and ceiling. Half wash includes everything in a full wash, except emptying gully pots and washing the tunnel ceiling.

Before the washing procedure starts, as well as during and after washing, particles on the road surface are collected by a sweeping vehicle. The particles collected in this way accounts for 20-80 % of the pollutants produced annually (Roseth & Meland 2006).

Table 1: Tunnel wash frequencies for Norwegian road tunnels, adapted from (NPRA 2014)

Traffic volume AADT/tunnel lane	Full wash	Additionally: Half wash	Additionally: Technical wash
0 - 300	Every 5 years	---	1 in years w/o full wash
301 - 4000	1/year	---	1/year
4001 - 8000	1/year	1/year	2/year
8001 - 12000	1/year	2/year	3/year
12001 - 15000	2/year	3/year	5/year
15001 -	2/year	4/year	6/year

Water consumption during a tunnel wash varies greatly. Type of wash, tunnel length, number of lanes and nozzle type will all factor into how much water is consumed for washing a tunnel. Torp and Meland (2013) reports of up to 350 % increased water consumption when low-pressure nozzles are used, compared to high-pressure nozzles.

In addition to water, detergent is often used in the washing process. The soaps used must comply with EU regulations, which demands a biodegradability of 60 % within 28 days (European Parliament 2004), and are usually mixed to a concentration of 0.2-0.5 % (Roseth & Meland 2006). Since the regulations regarding detergents are very little specific as to what kind of chemicals might be applied, the different detergents that are used, and potentially might be used, vary greatly in composition (Roseth & Søvik 2015). The effect detergents have on treatment processes is not well explored, but Aasum (2013) found an increased mobility for some heavy metals, after adding the detergent TK601 Clean Extra Autovask, to TWW from a washing event where detergent had not been used.

The pollutant load in TWW will vary greatly, depending on many factors. Time between washing events, type of wash, traffic load, the amount of particles collected by sweeping vehicles, water use during wash, season and more, all affect the quality of the wash water. Earlier studies have shown a great variation in measured pollutant concentrations, even from the same tunnel. Looking at zinc concentration from TWW collected at the Nordby tunnel, Nersten (2016) found a concentration of 500 µg Zn/l, Petersen et al. (2016) found from 9 to 3290 µg Zn/l, while Garshol et al. (2016a) found at most 44 000 µg Zn/l, though there is good reason to doubt the last finding, as discussed in their report. A compilation of studies done in Stockholm, showed a range of 2 200-19 800 µg Zn/l, while a Swedish study of a washing event in a Stockholm tunnel, spanning two nights, showed a range of 632 – 6290 µg Zn/l, with an average of 2694 µg Zn/l (Byman 2012). These studies show similar patterns for other pollutants, with concentrations spanning several orders of magnitudes, as shown in Table 2.

Table 2: Envionmental quality standards for metals in fresh water, concentrations of metals found in previous studies of TWW, and theoretical metal concentrations after dilution in a river recipient

Metal		AA-EQS	MAC-EQS	TWW metal concentration range	Concentration after 10X dilution in a river recipient
Arsenic	[µg/l]	0.5	8.5	0.25[1] – 37.3[2]	0.03 – 3.73
Cadmium	[µg/l]	0.08[4]	0.45[4]	0.1[1]-133[1]	0.01-13.3
Chromium	[µg/l]	3.4	3.4	5.6[1] -970[3]	0.56 -97
Copper	[µg/l]	7.8	7.8	27.2[1]-3300[3]	2.7-330
Nickel	[µg/l]	4	34	1[1]-187[2]	0.1-18.7
Lead	[µg/l]	1.3	14	0.05[1]-2000[3]	0.005-200
Zinc	[µg/l]	11	11	9[1]-19800[3]	0.9-1980

[1] Petersen et al. (2016)

[2] Byman (2012) – own results

[3] Byman (2012) – compilation of previous TWW studies from Stockholm

[4] Limits for soft water

TWW contains a variety of different pollutants, as detailed in Table 3. Most prominent among them are a wide range of metals, different polycyclic aromatic hydrocarbons (PAHs) and de-icing salts. In addition to this, the wash water often contains oil particles and detergents (Meland 2010).

Metals originate from wear and tear of car parts, tunnel surfaces and technical equipment in tunnels. Different metals are associated with a variety of health risks in humans as well as fish, with exposure causing for example genotoxicity (Arsenic), impaired kidney function (Cadmium), phytotoxicity and anemia (Zinc), and high lethality (Copper) (Gautam et al. 2015). One metal can appear as many different species with varying toxicity, depending on factors like pH, water temperature and other particles present. Being elements, metals can't be degraded, and will accumulate in sediments or organisms.

PAHs is a group of organic compounds found to have many toxicological effects, including deformities, genotoxicity, carcinogenicity, and narcotic effects (Logan 2007). While they can be formed in nature, the PAHs found in TWW are pyrogenic or petrogenic in origin, stemming from incomplete combustion of organic matter or from petroleum products, respectively. The varying structures, sizes, and chemical properties of PAHs affect their toxicity, and external factors like presence of other chemicals and light will also change the interaction between PAHs and aquatic life (Logan 2007).

While many of the individual pollutants have been thoroughly scrutinized for health and environmental risks, discharge of TWW itself has also been linked to detrimental effects in aquatic environments. TWW has been found to cause acute toxic effects in fish, like oxidative stress (Meland et al. 2010b) and a range of molecular changes in liver tissue (Meland et al. 2011), as well as long-term effects, showing a reduced growth for a brown trout population, downstream of a TWW discharge (Meland et al. 2010a).

Based on the EU directive regarding priority pollutants (European Parliament 2013), Norway implemented concentration limits for specified chemicals in water bodies into the regulation of water management in 2015 (Vannforskriften 2006). There are limits defined for 33 chemicals, and 12 more to be enforced by the end of 2018. For each chemical, there are four limits defined, annual average environment quality standard (AA-EQS) and maximum admissible concentration (MAC-EQS), each of which are defined for both freshwater and coastal water. There is also a system for classifying the ecological status of water bodies, with threshold values for pollutants, dividing the water bodies into one of five classes, from 1 – Background levels, to 5 – Very poor quality (Miljødirektoratet 2016). Level 1 and 2 are considered good, while classes 3, 4 and 5 are considered unacceptable,

Table 3: Contaminants in tunnel wash water and their sources, adapted from (Meland 2010)

Source	Contaminant
Brakes	Ba, Cu, Fe, Mo, Na, Ni, Pb, Sb
Tires	Al, Zn, Ca, Cd, Co, Cu, Mn, Pb, W, hydrocarbons, PAH (pyrene, fluoranthene, benzo(ghi)perylene)
Catalytic converters	Pt, Pd, Rh
Vehicle body	Cr, Fe, Zn (steel)
Combustion	Ag, Ba, Cd, Cr, Co, Mo, Ni, V, Sb, Sr, Zn, PAH (naphthalene), MTBE, BTEX
Oil and petrolium spills, dripping, used lubricant oil	PAH (LMM)
Road surface (asphalt, bitumen)	Al, Ca, Fe, K, Mg, Na, Pb, Si, Sr, Ti, PAH (mix of HMM and petrogenic LMM, chrysene)
De-icing and dust suppression	Ca, Mg, Na, Cl, ferro-cyanide (anticaking agent)
Road equipment (e.g. crash barriers, traffic signs etc.)	Zn (galvanized steel)
Detergents used in tunnel wash	Tensides

Abbrevations: BTEX=benzene, toluene, ethylbenzene and xylenes, HMM=high molecular mass species, LMM=low molecular mass species, MTBE=methyl tert-butyl ether, all other are elements

as measures should be taken to reduce the pollutant load. The limits between level 2 and 3, and 3 and 4, are set at AA-EQS and MAC-EQS respectively. For a list of metals and their class limits, see Appendix A.

At the point of discharge, a zone of mixing occurs, where the pollutants are gradually diluted. There are guidelines for what is an acceptable mixing zone before the water phase reaches the EQSs set for the water body. This depends on the hydromorphological and dynamic conditions in the recipient, the recipient's usage, and other local conditions (Ranneklev et al. 2013). As a rule of thumb, it is possible to apply an expected dilution factor, D, though for any real case scenario, the actual conditions of the recipient and the point of discharge must be considered. The expected dilution factors are given to be 2-10, for rivers, 5-20 for lakes, and 10-50 for marine recipients (Ranneklev et al. 2013). The AA-EQS for zinc in freshwater is 11 µg Zn/l (for zinc the AA-EQS is the same as the MAC-EQS), and thus the limit for acceptable concentrations in a freshwater recipient. Applying a dilution of 10 (assuming a river recipient with good mixing) to the range of zinc concentrations found in previous TWW studies, the diluted concentration is in the range 0.9-1980 µg Zn/l, implying a need for treatment for most concentrations. Diluted concentrations for other metals are found in Table 2.

Though these guidelines exist, other limits are used when granting discharge permits. The County Governor, the government authority in charge of discharge permits for TWW in Norway, has set limits for suspended solids (SS) < 100 mg/l, oil concentration < 5 mg/l and pH between 6 and 8.5, for TWW effluent to vulnerable recipients (Haraldsen 2017). These limits are only for new discharge permit applications, so older tunnels operate without the same restrictions.

A survey of tunnels in Eastern Norway showed that only 78 % of the 74 tunnels had TWW treatment facilities, and most of the existing facilities were simple sedimentation tanks or ponds (Torp & Meland 2013). Though sedimentation provides some treatment and reduces the peak concentrations of pollutants, there are still elevated effluent concentration of metals and PAHs after a sedimentation pond. Many pollutants exceed their set EQS's, and adverse effects on aquatic life has been observed downstream of a sedimentation pond outlet (Meland et al. 2010a).

Previous studies have tried various methods for treating TWW. Paruch and Roseth (2008) tested organic sorbents in two different experiments, one with one month treatment time and one with one day treatment time. Treatment using sedimentation was tried by Hallberg et al. (2014), in an experiment in a Stockholm tunnel. The treatment method consisted of settling TWW in a clarifier for 24 hours, with and without adding the aluminum based chemical flocculent PAX, before the clarifier. Several studies have been conducted the last years, funded by the Norwegian Public Roads Management's project NORWAT. The most thorough study of TWW treatment was conducted by Garshol et al. (2016a) on behalf of NPRA. A range of treatment methods were tested: long or short pre-settling plus chemical precipitation, chemical precipitation and pH adjustment, aerobic, semi-aerobic or anaerobic biological treatment plus chemical precipitation, and anaerobic biological treatment plus filtration. The chemical precipitation in this study was done using the iron based chemical PIX, and the aluminum based chemicals PAX and SAX. Complementing these experiments, Nersten (2016) treated TWW using lime for a chemical treatment process with sedimentation. The best results for each study are presented in Table 4, along with the duration of the treatments.

The treatment solutions with long residence times could be implemented either by collecting TWW and bringing it to a centralized treatment plant, or by building a separate treatment facility for each tunnel. For faster treatments, a mobile treatment solution would be feasible, which can be deployed to each tunnel as they are being washed. This solution would probably have higher operating costs, but far lower investment costs. A mobile solution would also be easier to implement in existing tunnels, where there might be space constraints for building treatment facilities, or construction work can be of hindrance to road traffic.

In addition to their extensive research on TWW treatment methods, Garshol et al. (2016b) wrote a literature review, exploring possible mobile treatment solutions for TWW. Their study highlighted a lack of knowledge about the range of pollutant concentrations, flow rates and washing frequencies, even for a single tunnel. The authors were also in contact with contractors to find possible solutions for a mobile treatment system. A range

Table 4: Metal effluent concentrations and removal rates in previous studies of TWW treatment. The best results from each study are presented. Studies with both fast and slow treatment setups, the best results for both are presented. All chemical treatments used sedimentation for solids separation.

Author(s)		**Paruch and Roseth (2008)**	**Paruch and Roseth (2008)**	**Hallberg et al. (2014)**	**Garshol et al. (2016a)**	**Garshol et al. (2016a)**	**Nersten (2016)**
Method		Organic sorbents	Organic sorbents	Chemical precipitation (PAX) + settling	Anaerobic biological + chemical precipitation (PIX)	pH adjustment (pH 12) + chemical precipitation (PIX)	Chemical precipitation (lime + Mg)
Treatment time		1 month	1 day	24 hours	3 weeks	Coagulation + 30 min settling	8 min coagulation + 21 min settling
Copper	**[µg/l]**	< 6.0	39.1	-	6.6	20	13
	[%]	88.6	83.6	-	99.85[1]	94.28	91.88
Nickel	**[µg/l]**	< 4.0	29.9	< 8	20	9,5	3.9
	[%]	97.7	17.6	-	96.9[1]	71.2	79.47
Zinc	**[µg/l]**	17.6	208	< 60	12	19	< 0.13
	[%]	98.8	23.8	-	99.97[1]	99.2	99.96
Aluminum	**[µg/l]**	74	1200	-	-	-	-
	[%]	99.7	70.4	-	-	-	-

[1] Extremely high raw water concentrations might make these removal rates incomparable to other studies

of different systems were proposed, but the communication also revealed little interest in the topic, as the market for these treatment solutions was deemed to be undeveloped. One of the proposed treatment methods was electrocoagulation, but the report notes that the method has not previously been used for TWW treatment, and would need to be tested to assess its suitability.

1.3 Electrocoagulation

Electrocoagulation (EC) for water treatment purposes was patented over a century ago (Dieterich 1906). Since then it has been applied for treating drinking water and wastewater from textile, diary, and electroplating factories (Chen 2004).

In an EC unit, an electrical current is applied through a set of metal electrodes, most often made from aluminum or iron, submerged in the raw water to be treated. Metal ions are dissolved from the anode by the current, while hydrogen gas is produced at the cathode. After dissolving in the raw water, the metal ions react with the water, creating metal hydroxides, which again react with dissolved pollutants and colloids in the wastewater, much in the same way traditional chemical precipitation works. For aluminum electrodes, the chemical reactions at the anode are described by the following equations (Chen 2004):

$$Al \rightarrow Al^{3+} + 3e^{-} \tag{1}$$

$$Al^{3+} + 3OH^{-} \rightarrow Al(OH)_3 \text{ (alkaline conditions)} \tag{2}$$

$$Al^{3+} + 3H_2O \rightarrow Al(OH)_3 + 3H^{+} \text{ (acidic conditions)} \tag{3}$$

At sufficiently high potentials, oxygen evolution might also take place, possibly reducing the efficiency of the electrochemical cell (Shalaby et al. 2014):

$$2H_2O \rightarrow O_2 + 4H^+ + 4e^- \text{- acidic conditons} \tag{4}$$

As the anodes are oxidized by the electrochemical reactions, oxygen is reduced at the cathodes, and hydrogen gas is produced (Gu et al. 2009):

$$O_2 + 4H^+ + 4e^- \rightarrow 2H_2O \tag{5}$$

$$2H_2O + 2e^- \rightarrow H_2 + 2OH^- \tag{6}$$

$$2H^+ + 2e^- \rightarrow H_2 \tag{7}$$

These reactions cause a reduced pH at the anodes, and an increased pH at the cathodes. In the proximity of the anodes, the pH is lowered by the aluminum hydrolysis and potentially by oxygen evolution. Around the cathodes, alkaline conditions are caused by the processes described in equations (5)-(7).

Hydrolyzed aluminum ions from the anodes will interact with colloids and suspended particles in the water, according to the same principles as the ones that govern normal chemical coagulation, using addition of metal salts to the wastewater. Though the full chemical processes are not well understood, the dominating actions follow these categories:

1. Adsorption and charge neutralization
2. Adsorption and interparticle bridging
3. Enmeshment in sweep floc

Adsorption and charge neutralization happens when metal hydroxides are adsorbed on colloids in the water to be treated. With the particles charge now neutralized, it will no longer resist collision with other particles, and may aggregate to larger particles. When the metal hydroxides form chained polymer species, there will be several adsorption sites available for particles to attach to, and the metal hydroxides "bridges" together several colloids to form flocs. At higher metal ion concentrations, the dissolved metal will precipitate and form large metal hydroxide flocs, that will enmesh colloidal particles within them, thus separating the colloids from the water phase (Metcalf & Eddy 2014).

The solubility and speciation of aluminum in water, as well as the metals to be removed from the water, are pH dependent. Figure 2 shows that the total aluminum dissolved in the water is lowest around pH 6, while Figure 1 shows that hydroxides of other metals have their lowest solubility in the pH range of 9-11, depending on the type of metal. This pH dependent solubility of metals should play a role in their removal in an environment with a sharp pH gradient, as in an EC reactor. Even so, how these effects play out is not discussed in studies looking at metal removal using EC and/or the effect of bulk pH on EC treatment (Bazrafshan et al. 2015; Cañizares et al. 2009; Un & Ocal 2015). It is possible that when the metals enter areas of least solubility, they precipitate and take less soluble forms by interacting with coagulants, and so the metal removal is higher than what could be expected considering only the bulk pH, but this remains speculation.

For EC reactors with more than two electrodes, there are three common arrangements for the electrodes: monopolar-serial (MP-S), monopolar-parallel (MP-P) and bipolar-parallel (BP-P), shown in Figure 3. In an MP-P setup, every second electrode is connected by wire in parallel to the same pole on the DC power source, and the other ones are connected to the other pole. For a MP-S setup, the outer electrodes are each connected to one

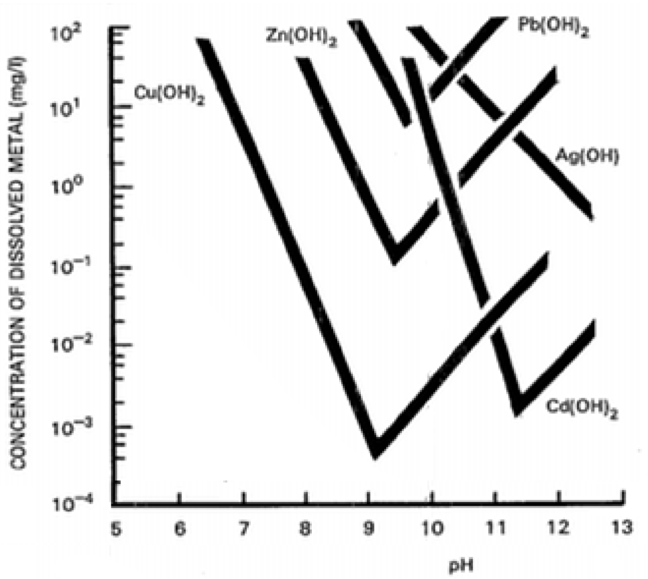

Figure 1: Solubility of metal hydroxides as a function of pH, adapted from (EPA 1980).

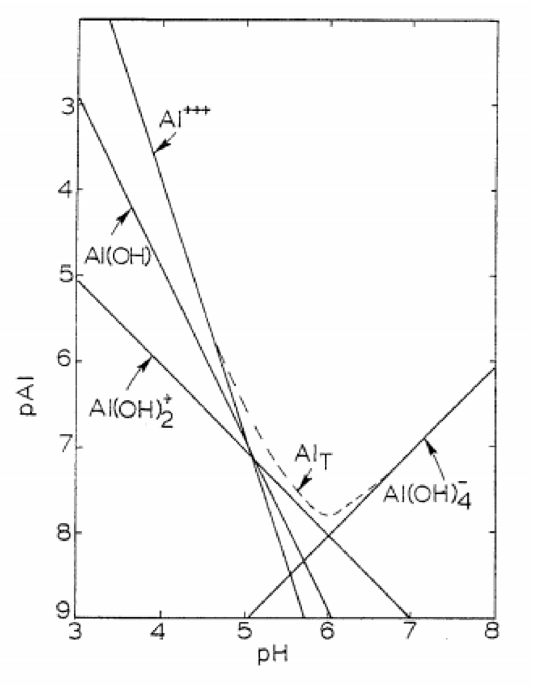

Figure 2: Solubility of Al species in water as a function of pH, (Driscoll & Schecher 1990). Al_T = Total Aluminum

of the poles on the power source, while the inner electrodes are connected to each other by a wire. The last setup is BP-P, in which the outer electrodes are connected to the power source, as is the case with MP-S, but the inner electrodes are only connected to each other and the outer electrodes through the water being treated. These inner electrodes will then become bipolar, with the two sides of the electrode having opposite polarity. The MP-P setup has been found to be the most cost-effective of these (Demirci et al. 2015).

The current density, *I*, is one of the most common operating parameters evaluated when testing EC performance. Even so, there is confusion over how to define it. In a review by Chen (2004) the current density is defined as total current being applied by the power source, divided by the effective *electrode* area, while Kuokkanen et al.'s (2013) review divides the current by effective *anode* area. Using only anode or cathode area is more meaningful, as the same current flows through both anode and cathode, and the latter definition will therefore be used here. The effective anode area will be the submerged area of the anode where the current flows through (Kuokkanen et al. 2013). The current density will not be evenly distributed throughout the anode surface, so the current density used will in effect be an average current density. The area on the outer anode, not facing a cathode, has very little flow current flowing through it, as visualized in the primary potential distributions of Vázquez et al.

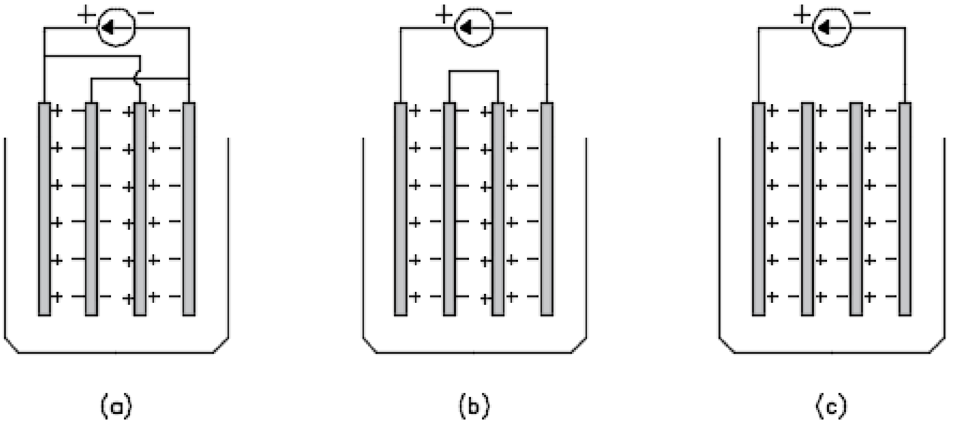

Figure 3: Common electrode configurations in EC. (a) Monopolar-parallel (b) Monopolar-serial (c) Bipolar-parallel.

(2012). This implies a low current through this surface, but the actual current will depend on conductivity and other design features.

The dissolution of anode material is theoretically described by Faraday's law of electrolysis, which gives a relationship between the current passing through an electrode, and the dissolved material. Mollah et al. (2004) presents this in the terms of current density and material dissolved per anode surface:

$$w = \frac{itM}{nF} \tag{8}$$

where w is the quantity of electrode material dissolved (grams of metal*cm^{-2}), i is the current density (A*cm^{-2}), t is the time in seconds, M is the molar mass of the electrode material in g/mol, n is the number of electrons in the oxidation/reduction reaction and F is Faraday's constant (96,500 C mol$^{-1)}$. When using an aluminum electrode: M = 26.981539 g/mol and n = 3, so that

$$w \approx it * 9.3 * 10^{-5}\mathrm{gC}^{-1} \tag{9}$$

This is the theoretical mass dissolved from the anodes by the current, or Faradic dose. Several studies have found that Faraday's law of electrolysis underestimates the amount of aluminum dissolved. Gu et al. (2009) found it likely that the extra dissolution was caused by a reduced pH at the anodes and increased pH at the cathodes, which leads to oxidation of the anodes and a dissolving of the oxide layer on the cathode surfaces, and in turn the underlying aluminum. How large these effects are, compared to the Faradic dose, varies from study to study, and is probably affected by factors such as the electrode characteristics and the mixing in the electrocoagulation cell (Gu et al. 2009).

As current is applied through the electrodes, the corresponding potential, and in turn, the power consumption, are governed by the Ohm's law. Using a term for surface resistance to generalize for an undefined electrode surface, the Ohmic resistance in electrocoagulation is defined by Gu et al. (2009):

$$\Omega = \frac{\text{electrode spacing (cm)}}{\text{solution conductivity (S/cm)}}$$

This shows that reducing the electrode spacing and/or increasing the solution's conductivity, will, for a given current density, lower the potential and power consumption. If the conductivity in the water to be treated is low, electrolytes like NaCl can be added to increase the conductivity (Chen 2004).

As the conductivity is intrinsically linked to the total dissolved solids, and the pollutant load in the TWW is expected to be high, addition of electrolytes is probably not needed for treating TWW, assuming a reasonable electrode spacing.

The electrode spacing is as important as the conductivity in determining the Ohmic resistance in the EC process. Closer electrode spacing lowers the resistance, and makes space for more electrodes per volume, increasing the active electrode area. The electrode thickness and the electrode spacing will determine the specific electrode area (electrode area divided by waste water and electrode volume). When operating a batch experiment, water needs to be circulated from between the electrodes to the rest of the reactor, so the dissolved aluminum particles can come into contact with the pollutants. A smaller electrode spacing would need a higher stirring speed for this to happen. In a review of experiments, Kuokkanen et al. (2013) found that most experimental batch setups use an electrode spacing in the range of 5-20 mm.

The pH of the water can affect dissolution of the electrodes, speciation of the metal dissolved from the anode, and how these species interact with the pollutants in the TWW. Different pH values will also affect the solubility of metals in the water. As discussed previously, the pH close to the electrodes is affected by the reactions taking place there, and the corrosion of the electrodes is therefore mainly caused by the local pH, not the pH of the bulk solution. Corrosion caused by bulk pH in a system without current running through the electrodes, accelerates at pH less than 2 or greater than 11, but is low and stable within this range (Gu et al. 2009).

Stirring speed in a batch experiment will change the distribution of the metals dissolved from the anode, and how much time they have to hydrolyze before coming into contact with colloids in the wastewater. The low or high pH environments around the electrodes are also reduced in size by increased stirring. If the stirring speed is too high, the agitation might break the flocs, and resuspend sludge that has floated to the top of the reactor. While studying the influence of stirring speeds on cadmium removal in industrial wastewater, Khaled et al. (2015) found the optimal stirring speed at 300 rpm, while Khandegar and Saroha (2012) found an optimum at 500 rpm, for COD removal in distillery wastewater.

The presence of detergents in TWW, as used in some washing events, might interact with the electrocoagulation performance. Detergents have been observed to increase mobility of metals (Aasum 2013), and to interfere with alum floc formation in conventional chemical precipitation (Smith et al. 1956), but their effect in an EC reactor is unknown. Several studies have been conducted with detergent present in the wastewater, primarily those looking at EC for treatment of laundry wastewater (Ge et al. 2004; Janpoor et al. 2011; Wang et al. 2009). Of these, only Wang et al. reports which detergent is used, a linear alkylbenzene (anionic) based detergent. Takdastan et al. (2017) used car wash water in their study, but, as with the studies on laundry water, the main focus was COD and/or phosphate removal, as well as removal of detergents, not metal removal. One study looked at removal of metals from truck wash water (Gomes et al. 2016). It is assumed that the truck wash water contains surfactants from detergents, but the presence of these in the sampled water and any effect they may have had on treatment efficiencies is not discussed.

The gas generated at the cathodes during EC creates a flotation process. The gas bubbles attach to particles in the waste water, decreasing the buoyancy of the flocs, causing them to rise to the surface. As sludge accumulates on top of the reactor, an easily separable foam is created. This creates a possibility for building a more compact, efficient reactor, where coagulation and separation happens in the same chamber, if the electrochemical reactor is built with this purpose in mind. The size distribution of the bubbles created is affected by factors like pH, current density, electrode material, and electrode design, and determines the effectiveness of the flotation (Chen 2004).

1.4 Response-surface models

A response-surface model is a statistical tool used in experimental design, to determine optimal values for several input parameters at once, generating a surface model showing how the output changes with different combinations of input variables. For two input variables, the relationship can be described as:

$$y = f(x_1, x_2) + \epsilon \tag{10}$$

where *y* is the output, x_1 and x_2 are the input variables, ε is the noise or error observed in the response and $f(x_1,x_2)$ represents a response surface (Venter et al. 1996).

Using a central composite design (CCD), the response sensitivity to two or more input variables are measured simultaneously. For each input variable, a middle value is chosen, as well as an upper and lower bound, at an equal distance from the middle value. These values can be coded to the steps -1, 0, 1. An experiment designed using CCD experiments on the center point, as well as the combination of extreme values of the input variable, called factorial points. This is contrasted with another block of points, axial points, at a distance α from the center point, along the x_1 and x_2 axes. For this setup, α is usually set to $\sqrt{2}$, which along with three center runs per block reduces the variance, and keeps the variance stable over the region that is experimented on. See Figure 4 for a visual representation of the test points.

This design of experiments is more efficient than the more standard one-factor-at-a-time experiments, because fewer observations are needed, the interaction between different input variables can be estimated systematically, and it gives response data on a larger range of inputs variables (Czitrom 1999).

When evaluating a statistical model, the term R^2 is often used. R^2 is a measure, from 0 to 1, of how well a regression model fits the data points it is based on, and will always increase with more input variables. When using more than one input variable, the term adjusted R^2 (R^2-adj) is used instead, which compensates for the extra input variables (Montgomery & Morrison 1973). Predicted R^2 (R^2-pred) is a term for evaluating a model, by removing one observation at a time, and testing how well the model will predict that observation, based on the other observations (Colton & Bower 2002).

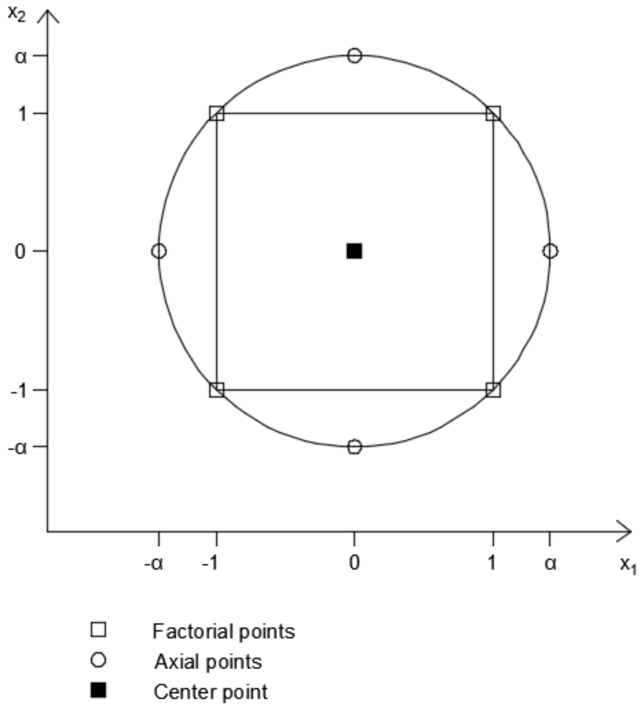

Figure 4: Test runs in a central composite design, with coded units. Each point marked corresponds to an experiment run, with different combinations of input variables. Multiple runs are usually conducted on the center point.

2 Materials and methods

2.1 *Tunnel wash water*

TWW was collected at the Nordby Tunnel, during a half wash event for the northbound lane, on the night of February 16. The Nordby tunnel[1] is 3.9 km long, with two lanes going in each direction, and an AADT of 37 068 (NPRA 2017). TWW is usually collected through the tunnel's storm water pipes, which lead to a small pumping station outside the tunnel. From here the TWW is normally pumped to an open sedimentation pond, which discharges into the Årungen river.

The northbound tunnel lane was cleaned with high pressure nozzles, without detergents, using 40 m^3 water over 6 hours. Prior to collecting TWW, the sump was flushed with tap water while the resident pumps were running, to remove old sludge. TWW then accumulated as the wash progressed, and after reaching a suitable amount, a pump was lowered into the sump, and TWW was pumped into three plastic containers, totaling approximately 200 liters.

The water was transferred to a cylindrical steel tank, which had previously been used to store TWW from the same tunnel. The tank was rinsed with water and scrubbed with a broom, before the new TWW was transferred to it. The collected water was stored at 12 °C.

The TWW was stirred up using a shovel before sample retrieval to get a representative and homogenous sample. The water was collected in a 10-liter plastic can, which was refilled for the second block of experiments. The water was stored overnight at 4 °C, before the experiments, to lower the temperature to what is expected to be encountered for a winter tunnel wash.

2.2 *Electrocoagulation tests*

Electrocoagulation was tested in a batch process. The EC reactor consisted of four aluminum electrodes, made from EN-AW 5005 aluminum (Astrup, Oslo, Norway), measuring 13 x 6 x 0.3 cm. The electrodes were put in a PVC lid with slits cut for the electrodes, with a set spacing of 1 cm, and put in on top of a 1000 ml beaker. A DC power source, GW GPS-3030 (Taipei, Taiwan) with adjustable voltage and current was wired to the aluminum electrodes in a monopolar-parallel arrangement. See Figure 5 for a schematic of the experimental setup.

After intense shaking of the storage cans, the beaker was filled with TWW, approximately 1050 ml. One sample was retrieved for metal analysis (approximately 120 ml), 2 samples for suspended solids (30 ml each) and 2 samples for turbidity, using a 10-ml pipette. The pH was measured in the beaker. For two runs, the conductivity was also measured. The magnetic stirrer was used to keep the wash water homogenous while extracting samples. After the samples were taken, the TWW volume was reduced to approximately 800 ml, and the lid with electrodes inserted was put on top of the beaker, with the TWW covering 7.5 cm of the electrodes' height, giving a total anode surface of 192.6 cm^2. The stirrer, operating at approximately 300 RPM was used to keep the water circulating throughout the experiments.

After connecting the wires, the DC power source was turned on and the timer started. The current was adjusted according to the current wanted for the experiment. The polarity in the EC cell was changed every 3 minutes to minimize scaling on the anode surfaces. After the stirrer and current was switched off, the sample was left to clarify for 4-10 minutes. The clarification duration was judged by observing the flotation behavior in the EC cell. When there was no more visible accumulation and/or compacting in the sludge layer, the samples of clarified water were collected.

Samples of the treated water were collected from the bottom, because the sludge accumulated on top. This was done by using a syringe and a pipe going to the bottom of the beaker. In the first five runs, a 100-ml syringe was used, and connected directly to a PVC pipe inserted through a hole in the lid. The pipe was in place through the

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[1] Coordinates, EUREF 89 UTM 33N:260317 6626871 to 262675 6629904

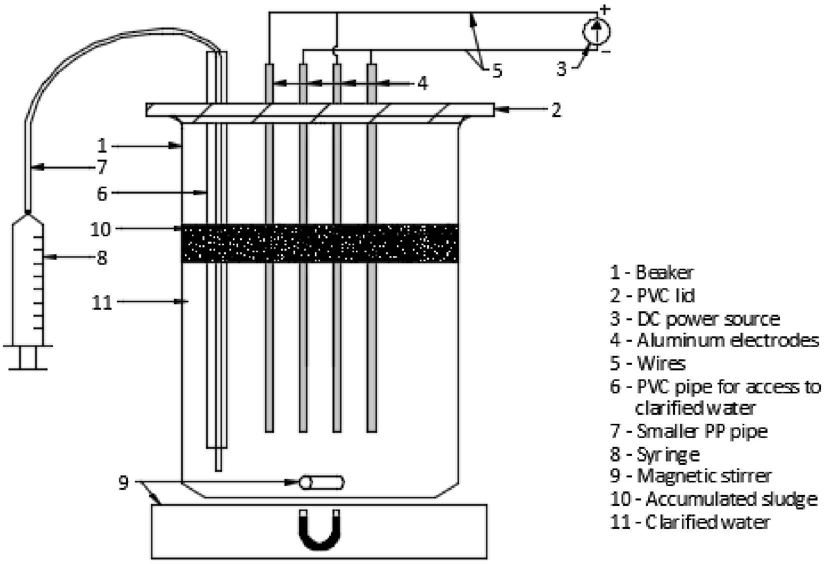

Figure 5: Experiment setup. Aluminum dissolved from the anodes and coagulated pollutants in the TWW. Gas created at the cathodes attached to these flocs, and rose to form a sludge layer on top of the water. Samples were retrieved using a syringe connected to the bottom of the beaker.

whole experiment, to avoid any disturbances. After the fifth run, the syringe broke and was replaced by a smaller, 60-ml syringe, which was connected to a smaller hose and inserted into the larger PVC pipe, after the treatment was finished.

In two experiments where high sludge production was observed, a sample of the gray, bottom part of the sludge was collected, and analyzed for metal content.

pH was measured using a ProfiLine pH 3110 pH meter, and conductivity using a ProfiLine Cond 3210 Conductivity meter (WTW, Weilheim, Germany). For turbidity measurements, a 2100N IS Turbidimeter (Hach Company, Loveland CO, USA) was used. Samples for metal analyses were sent to Eurofins Norge avd. Kristiansand for ICP-MS analysis**,** and analyzed for arsenic, mercury, lead, cadmium, copper, chromium, nickel, zinc, and aluminum. The analyses were done according to EN ISO 17292-2. Suspended solids (SS) were measured by filtering a sample through a glass fiber filter, pore size 0.45µm (Merck Millipore, Darmstadt, Germany), and measuring the increase in filter weight, after drying the filter for minimum 2 hours at 105 °C.

Before repeating the experiment, the equipment was washed and the electrodes were polished with sandpaper and rinsed with water, to remove any scaling formed on the metal.

The beaker, hoses and syringes used were washed in a 5% HCl solution between runs, to remove any metal residuals.

Each experiment was filmed, using a Sony α33 DLSR camera (Tokyo, Japan).

2.3 *EC treatment of deionized water*

After testing EC on TWW, an additional run on deionized water was conducted. NaCl was added to increase the conductivity of the water up to 12 mS/cm, about the same conductivity as in the TWW. The procedure for treatment of deionized water followed that of TWW, except for an increased volume of water (100 ml) filtered for SS prior to EC treatment. The sludge produced was also sampled and analyzed for metal concentrations, using the same procedures as for the TWW.

Table 5: Time and current configurations for experiment runs on TWW.

Standard order	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**	**12**	**13**	**14**
Run order	**6**	**7**	**3**	**2**	**4**	**1**	**5**	**10**	**11**	**14**	**8**	**12**	**9**	**13**
Time [m]	10	30	10	30	20	20	20	5.9	34	20	20	20	20	20
Current [A]	0.5	0.5	1.5	1.5	1.0	1.0	1.0	1.0	1.0	0.3	1.7	1.0	1.0	1.0
Current density [A/m^2]	26	26	78	78	52	52	52	52	52	15	89	52	52	52
Block	B1	B1	B1	B1	B1	B1	B1	B2	B2	B2	B2	B2	B2	B2

2.4 Experimental design

To find the optimal settings for the two input parameters, electrocoagulation time and current, the tests were designed utilizing a response-surface model. The circumscribed central-composite design, was used, with 4 factorial points, 4 star points ($\alpha = \sqrt{2}$) and 3 center points for each block, varying time around 20 ± 10 minutes and current 1 ± 0.5 A, corresponding to 52 ± 26 A/m^2, giving the test design listed in Table 5. The experimental design was set up using the Stat - DOE – Response Surface function in Minitab, and the run order within each block was randomized, but the blocks were done sequentially. When referring to runs, run order is only referred to if specified, if not standard order is meant when numbering experiment runs. The results were analyzed using Minitab. The results were analyzed testing different combinations of time, current density, these parameters squared and 2-way interaction between them, to find the optimal model for a response-surface for each output parameter.

3 Results

Overall, the treatment showed a great reduction in turbidity, suspended solids, and metals in the treated TWW, with a moderate pH increase (from 8.2 to 8.8-9.6). For all experiment results, see Appendix B. Time-lapse collages is presented for two runs, run 8 in Figure 6 and run 9 in Figure 7 and Figure 8.

Surprisingly poor performance for SS and turbidity removal was recorded for run 5, and upon reviewing the video of the experiment, disturbance in the sludge layer was observed, before retrieving these samples. These results were therefore omitted in analysis of the response-surface model.

During the treatment, a sludge layer would form on top of the water, usually forming two to three distinct layers by the end of the clarifying step. On top, there was a brown-black foamy layer. On the bottom a thicker, white-gray layer, which varied greatly in thickness. Between these two layers, there would sometimes form a thin, white sludge layer with larger bubbles. No sludge settled at the bottom of the beaker after treatment.

The tunnel wash water characteristics are described in Table 7**.** These are based on samples taken for each run of the untreated water. When referring to percentage removal or reduction in metals, turbidity and SS, the pre-treatment value measured for each run was used to calculate the removal rate, not the mean value.

Treatment efficiencies are listed in Table 6. For parameters where a response-surface model could be established, the range of results is listed. See Appendix C for response-surface model details. The other parameters are assumed to have a normal distribution. The mean removal rates and standard deviations were calculated using Minitab. See Appendix D for details.

The voltage of the electrical circuit was not rigorously observed, but for all experiments the start voltage ranged from 1-3 V, and the voltage would often decrease slightly through the experiment run.

Table 7: Characteristics for untreated TWW collected at the Nordby tunnel February 16

	pH	**Conductivity [mS/cm]**	**SS [mg/l]**	**Turbidity [NTU]**	**Copper [µg/l]**	**Nickel [µg/l]**	**Zinc [µg/l]**	**Aluminum [µg/l]**
Samples	*14*	*2*	*28*	*28*	*14*	*14*	*14*	*14*
Mean	8.24	11.86	898.2	416	716	1243	2529	47571
Standard Deviation	0.02	-	42.7	8.5	79.7	93.8	246	4309
Minimum	8.22	11.86	820	388	570	1100	2200	42000
Maximum	8.30	11.86	1087	434	840	1400	2900	54000

Table 6: Treatment efficiencies for EC of TWW

	SS	**Turbidty**	**Copper**	**Nickel**	**Zinc**	**Aluminum**
Removal rate [%]	90.6[2]	97.4[2]	98.3[2]	91.7-99.0[1]	98.2[2]	-84.4-79.5[1]
Standard deviation	2.6	0.82	0.72	-	1.9	-

[1] Range of removal rates listed. Removal rate depends on treatment time and current. See response-surface model for details.

[2] No reliable response-surface model could be made. Removal rates calculated assuming normal distribution.

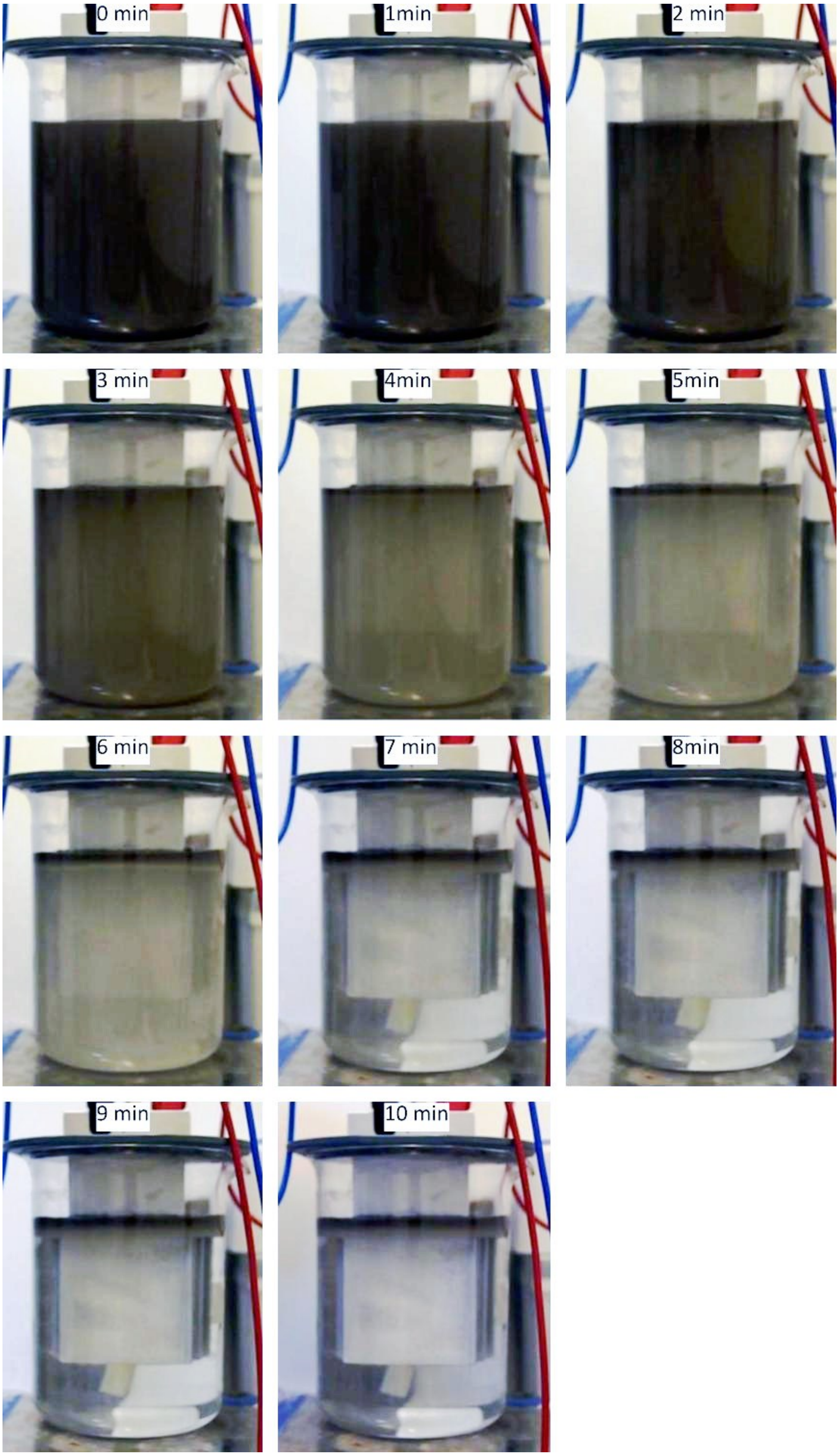

Figure 6: Time-lapse of run 8, short duration, medium current density. The duration of the EC was 5.9 min, plus 4 min flotation. A thin, gray layer can be seen in the bottom part of the sludge.

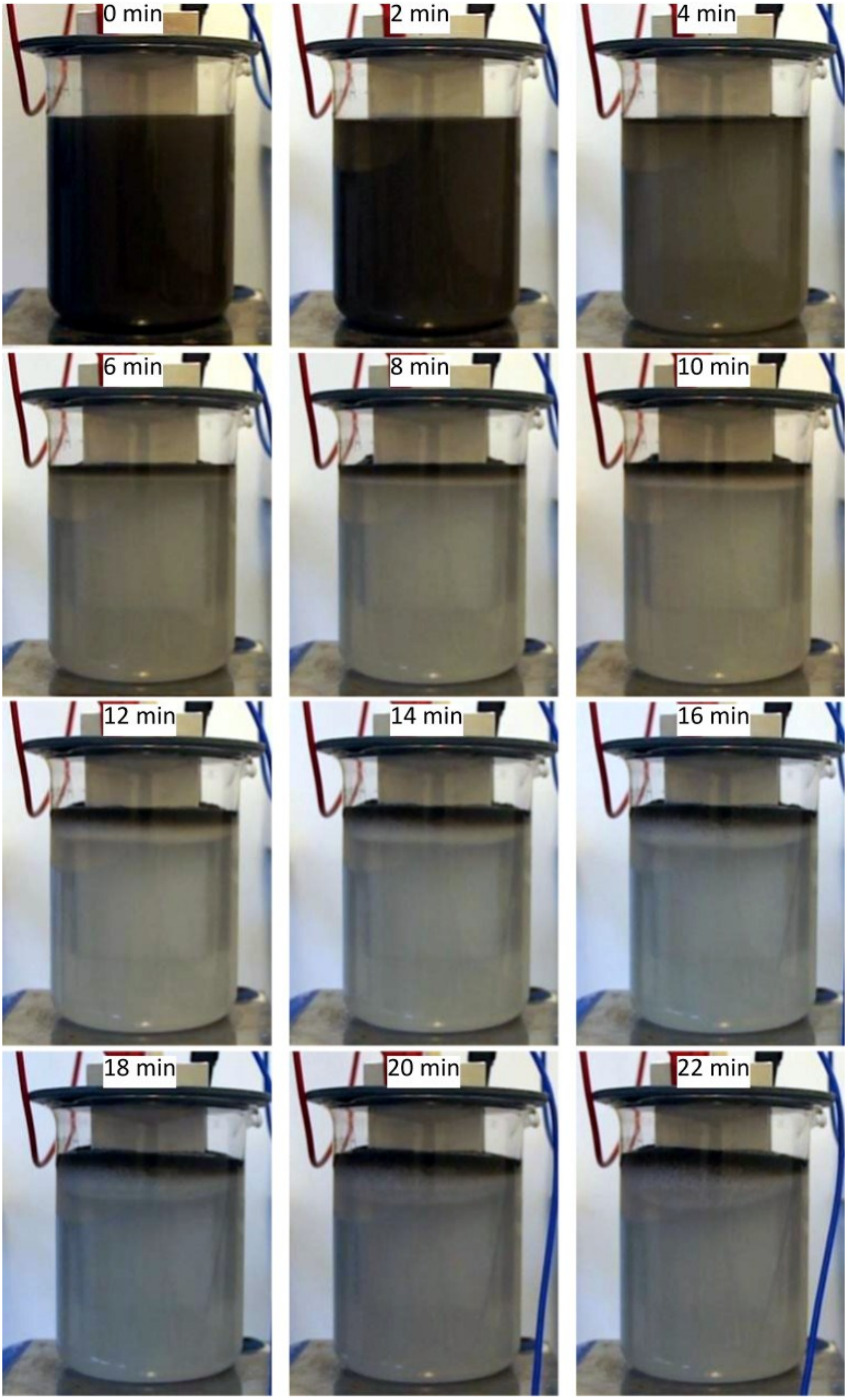

Figure 7: Time-lapse of run 9, long duration, medium current density, part 1. The duration of the EC was 34 min, plus 10 min flotation. A thick, gray layer can be seen in the bottom part of the sludge.

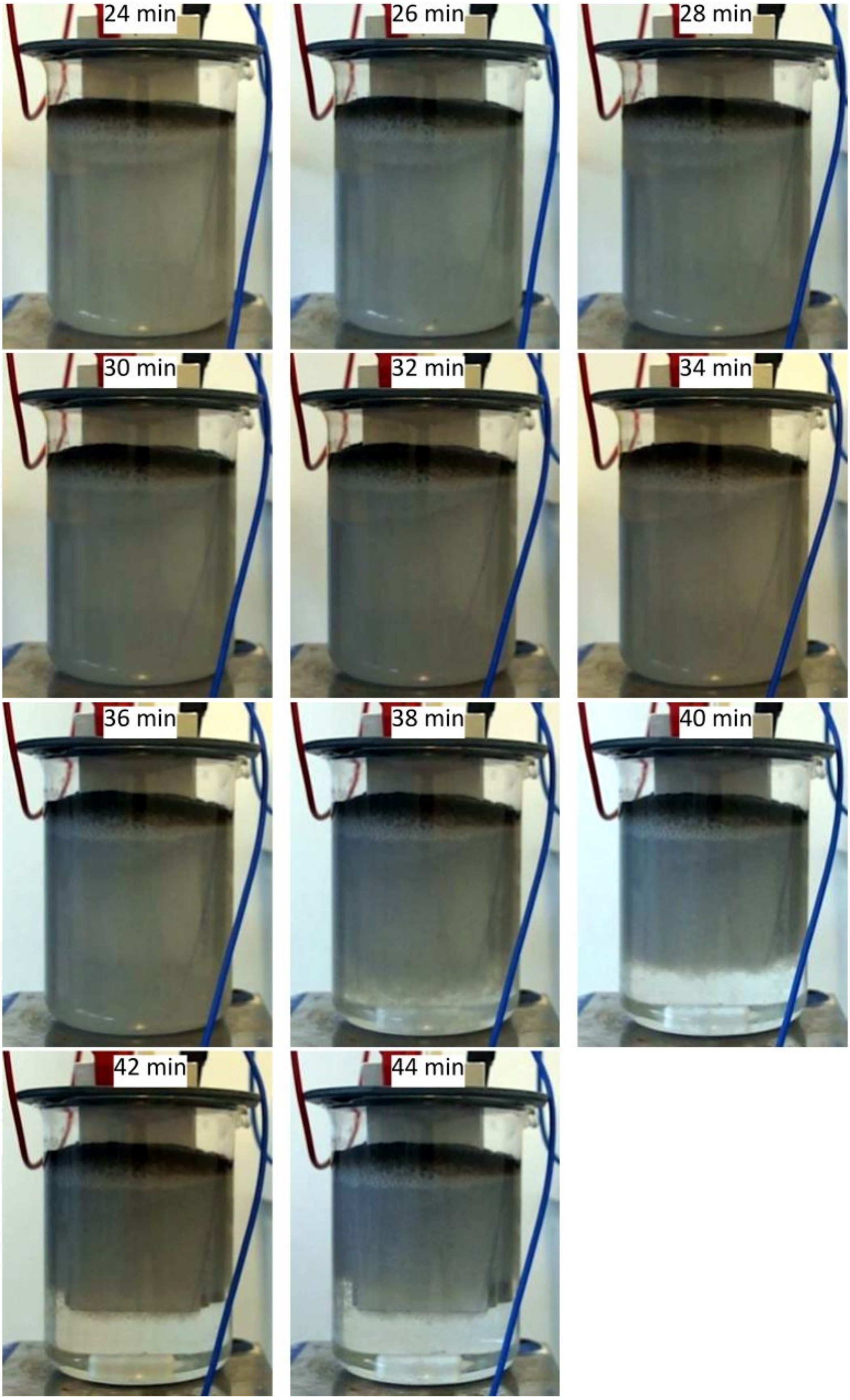

Figure 8: Time-lapse of run 9, long duration, medium current density, part 2. The duration of the EC was 34 min, plus 10 min flotation. A thick, gray layer can be seen in the bottom part of the sludge.

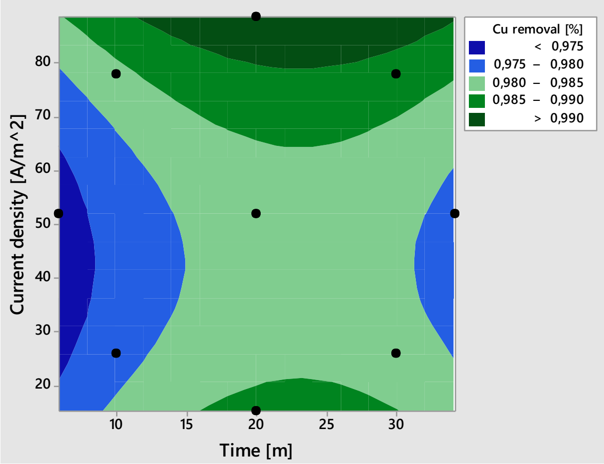

Figure 9: Contour plot of copper versus current density and time

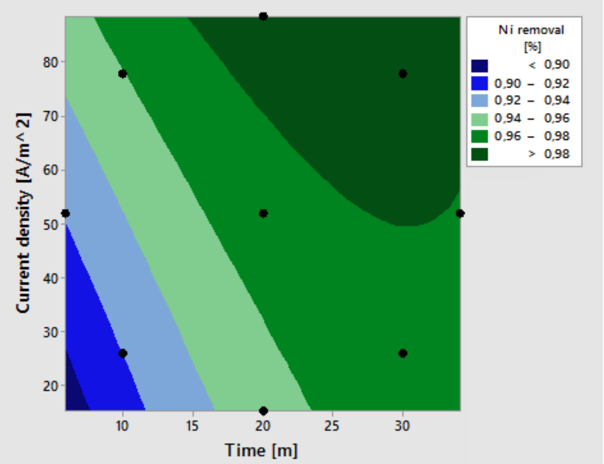

Figure 10: Contour plot of nickel versus current density and time

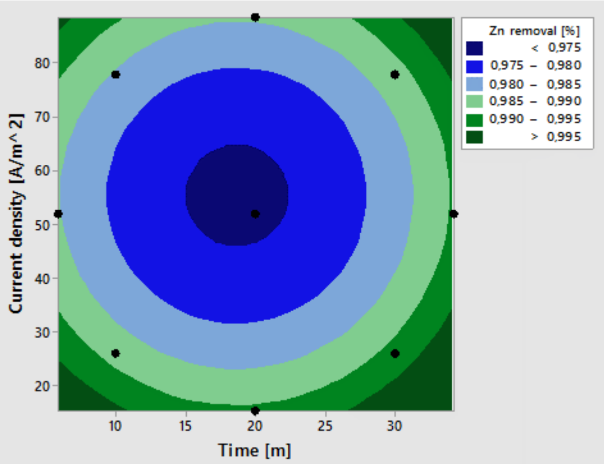

Figure 11: Contour plot of zinc versus current density and time

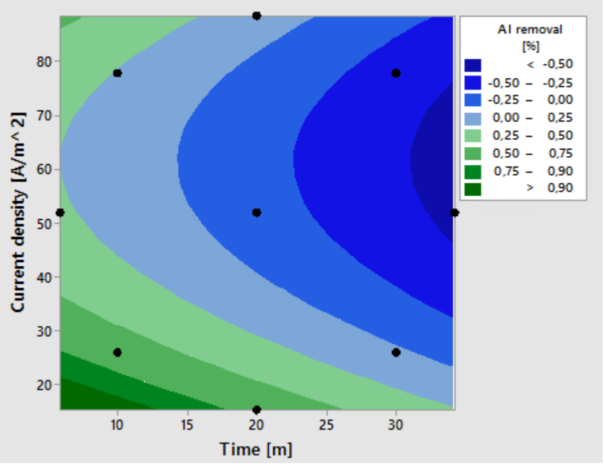

Figure 12: Contour plot of aluminum versus current density and time

3.1 Metal removal

3.1.1 Copper

The range of copper removal observed was 97.1-99.5 %, with a range of effluent concentrations of 4-22 µg Cu/l. The best treatment efficiency was observed for run 4, long duration, high current, and the lowest removal on a center run, though another center run recorded up to 99.1 % copper removal. A contour plot of the response is featured in Figure 9, implying best treatment around 20 minutes, and worse at longer or shorter durations. For current the picture is reversed, with increased treatment efficiency at current densities higher and lower than the middle value. This model has a low adjusted R^2 at 13.63 % and a p-value of 0.316, implying that the model is not suitable for explaining copper removal.

3.1.2 Nickel

The range of nickel removal observed was 91.6-99.0 %. The effluent concentrations ranged from 13 to 96 µg Zn/l. Worst performance was recorded for the run with medium length and low current, while best performance came with long time and high current. The contour plot for nickel removal, Figure 10, shows a trend of increasing nickel removal with higher current and lower time. The adjusted R^2 for the nickel removal is 90.76 % and the predicted R^2 fifteen percent lower than that, at 74.41 %. The p-value is 0.000.

3.1.3 *Zinc*

The range of zinc removal observed was 93.8-99.7 %. The highest effluent concentrations were recorded for run 5 and 7, both center runs, with 150 and 160 µg Zn/l. All other runs recorded 56 µg Zn/l or less. Other center runs recorded 6.7-38 µg Zn/l. The lowest effluent concentration recorded was 6.7 µg Zn/l, coinciding with the best removal rate. Figure 11 shows the contour plot for zinc, indicating worst treatment on center runs, and better treatment for any increase or decrease of duration and/or current density. The adjusted R^2 for this model runs as low as 27.45 %, and a predicted R^2 of 0. The model's p-value is 0.186, implying an insignificant correlation.

3.1.4 *Aluminum*

Aluminum had a high initial value concentration in the water, and after treatment the concentration was at best 79.5 % decreased, at worst 84.4 % increased. Lowest concentration came from the low current, short time run, with 22 000 µg Al/l and 52.2 % removal, and the low current, medium time run, with 9 000 µg Al/l and 79.5 % removal. The runs with highest effluent concentration was run 5, a standard run, and run 9, a medium current, long duration run, both with 83 000 µg Al/l. The latter had the highest increase of aluminum concentration. The lowest effluent concentration and best removal rate was on the medium time, low current run, with 9 000 µg Al/l. The contour plot in Figure 12, shows lowest aluminum concentrations for low current and low duration and increased concentrations with up to medium current density, and for increased treatment duration. Adjusted R^2 for this response surface is 64.98 %, and the predicted R^2 is 41.97 %. The p-value for this model Is 0.008, and the model can be considered significant.

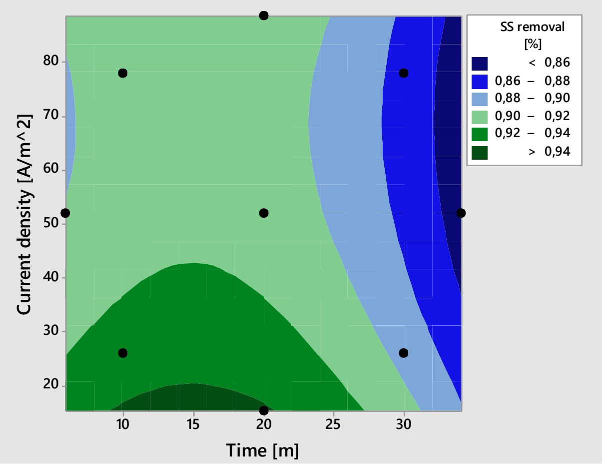

Figure 13: Contour plot of SS removal versus current density and time.

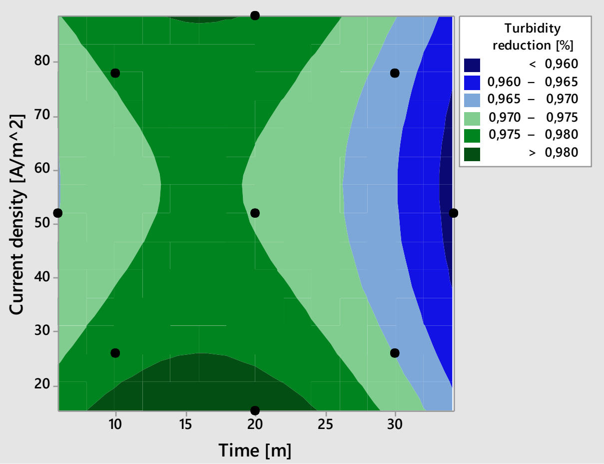

Figure 14: Contour plot of turbidity reduction versus current density and time.

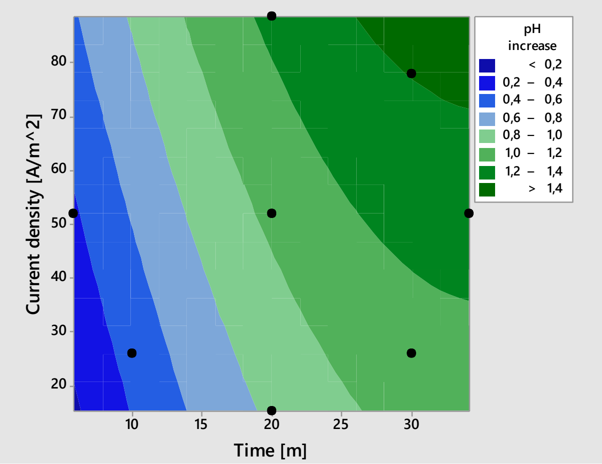

Figure 15: Contour plot of pH increase versus current density and time

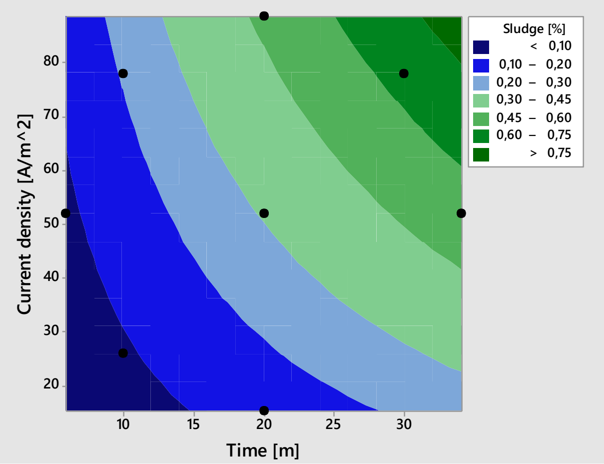

Figure 16: Contour plot of percentage sludge volume versus current density and time

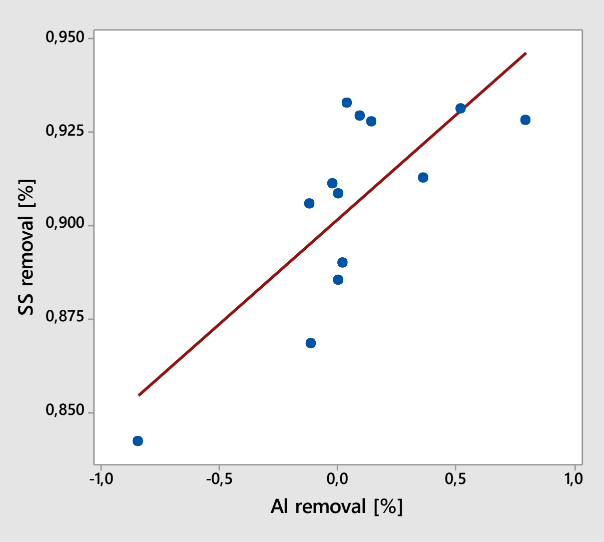

Figure 17: Linear correlation between SS removal rate and aluminum removal rate. Pearson correlation = 0.774, p-value = 0.002

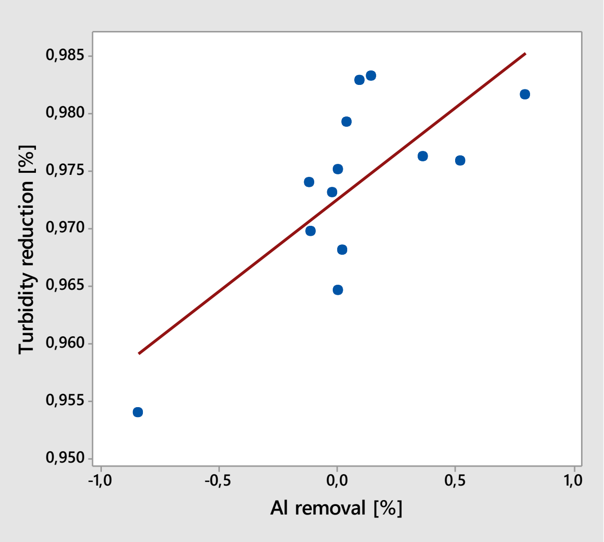

Figure 18: Linear correlation between turbidity reduction and aluminum removal rate. Pearson correlation = 0.742, p-value = 0.004

3.2 SS and turbidity

All SS values were based on two samples, but because of procedural errors, SS after treatment in run 4 was based on only one sample. Run 5 was omitted because of sampling error.

The range of SS removal recorded is 84.2-93.3 %, excluding the 63.3 % from run 5. SS removal shows a pattern similar to that of aluminum removal, with best results for low current density and medium time, and worsening with increased current and duration, with time being more important, as Figure 13 shows. With a p-value of 0.047, the SS model is significant, but the R^2 does not give much confidence to the modelled surface, with an adjusted R^2 of 56.10 %, and a predicted R^2 of 0.

As with SS, the results from run 5 is omitted in the analysis. Two effluent turbidity values, from run 7 and 11, are, because of experimental error, only based on one sample each, as opposed to the regular two samples.

The turbidity removal ranges from 95.4-98.3 %, with effluent turbidities of 6.88 to 19.0 NTU. The contour plot in Figure 14 shows the same response pattern as for SS, but the R^2 reflects an even lower suitability of this model to predict outputs, with an adjusted R^2 of 42.84 % and a predicted R^2 of 0. The p-value of this model is 0.106, implying no significant correlation.

A correlation analysis was performed in Minitab, using the function Stat - Basic statistics – Correlation, comparing SS removal, turbidity reduction, and copper, nickel, zinc, and aluminum removal to each other, using both a Pearson correlation and a Spearman correlation. The results are listed in Appendix E.

No significant correlation (p=0.05) was found between SS or turbidity and copper, nickel or zinc. Significant correlations were found between both SS and aluminum removal, and turbidity and aluminum removal, as visualized in Figure 17 and Figure 18.

3.3 pH increase

Because of procedural errors, several data points for pH is missing, resulting in three runs, 1, 3, and 5, missing pH change values. Even so, pH shows a clear trend of increasing with longer treatment duration and current density, as shown in Figure 15. The range of pH increases was from 0.33 to 1.34, with effluent pH being 8.58-

9.60. Highest pH was measured for the long duration, high current run, while the largest increase in pH was recorded for the medium duration, high current run. The lowest pH increase was measured after the short duration, medium current run. The adjusted R^2 is 91.97% for the surface model, while the predicted R^2 is 75.90%, and the p-value is < 0.000.

3.4 Sludge volume

Using the movies taken of each experiment, the thickness of the sludge layer was estimated. Some runs did not have usable pictures of the final sludge formation, because of technical problems with the camera. For most of these, supplemental pictures from a Sony Xperia XZ Compact phone camera were used, at some cost of accuracy, since these pictures were taken from another angle. Run 5 did not have any usable picture.

Sludge volume as a percentage of TWW volume after treatment ranged from 10-63 %, the lowest sludge volume coming from both run 1 (short duration, low current) and run 14 (short duration, medium current). The highest sludge volume came from the long duration, high current run. Figure 16 shows a trend of increased sludge volume for higher currents and higher treatment times. This model gives an adjusted R^2 of 93.06 %, and a predicted R^2 of 91.31 %, indicating a robust model for predicting sludge volume based on a run's given duration and current density. The p-value is < 0.000.

3.4.1 Gray sludge components

The gray bottom layer formed during EC was hypothesized to consist mainly of aluminum hydroxide from the anodes, with the bulk of non-aluminum pollutants in the top, brown-black layer of the sludge. Sludge layers are shown in Figure 19. This hypothesis was tested in two ways. First samples of the gray sludge were taken from two of the runs generating most gray sludge. Secondly, an additional run was conducted on deionized water to visually observe the sludge produced from EC of deionized water, and to measure the sludge's metal components, and compare this to the gray sludge in the normal runs.

Samples were collected from three runs. Run 9, long treatment duration at medium current density, and run 11, medium treatment duration at high current density. The third run was the run conducted on deionized water. Figure 20 shows the metal concentrations for untreated water, treated water, and the gray part of the sludge.

From these numbers, we can see that for the TWW runs, the copper, nickel and zinc concentrations are all lower in the gray sludge, than in the untreated TWW, but higher than in the treated water. For aluminum, the picture is radically different, with extremely high aluminum concentrations in the sludge.

The deionized water had very low concentrations of all metals before treatment, but some metals increased in concentration through the treatment, most pronounced for aluminum.

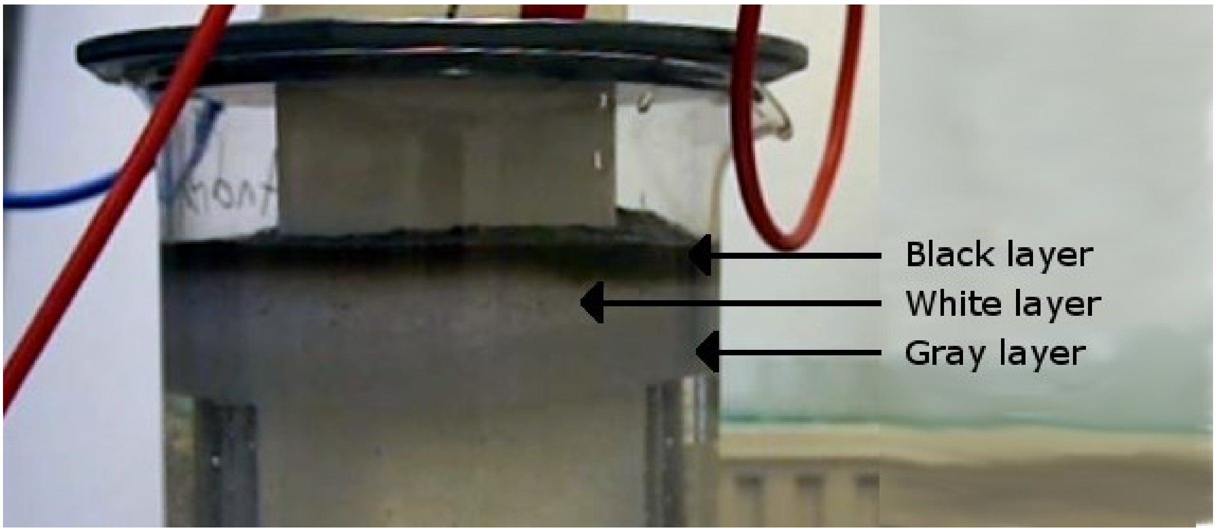

Figure 19: Sludge layers after electrocoagulation.

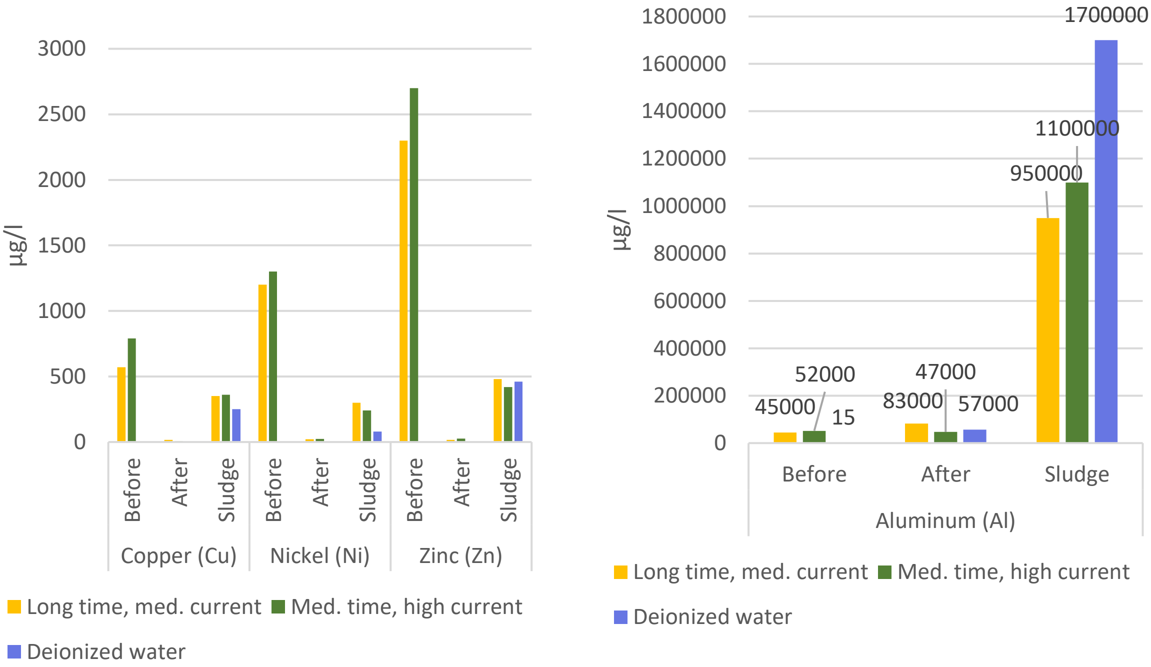

Figure 20: Metal concentrations in TWW before and after treatment, and in the gray part of the sludge. Aluminum separated because of scale.

4 Discussion

Some bias in treatment efficiency might be present between experiments with run order 1-5 and 6-14, because of the change of syringe for retrieving samples. The new, smaller syringe disturbed the sludge less, because the syringe was not connected directly to the PVC pipe inserted into the lid. This effect should be more pronounced for SS and turbidity, as the water for metal samples was connected first, when the sludge was less disturbed, and then water for other samples. After studying residuals, no such bias was noticeable.

Some systematical underperformance for metal removal might be expected to be recorded, because some TWW would accumulate in the PVC pipe during treatment, and stayed there throughout the experiment. This might be extracted together with the samples, though these particles wouldn't be present if not for the PVC pipe. Some flocs would also accumulate on the beaker walls, and be extracted with the clarified water samples, though these looked more likely to be aluminum hydroxide flocs.

4.1 Metal removal

Though 9 metals were analyzed for, only 4 of these, Ni, Cu, Zn, and Al will be used in the following discussion. Initially, Pb was supposed to be used instead of Ni, but because 12 out of 14 post-treatment values were reported as < 1.6 µg/l, this was not a suitable parameter to detect differences in the treatment setups.

There is no uniform picture formed of optimal input variables for removal of all metals. Zinc has worst treatment performance in the center of the explored input area, nickel has improved removal with increased time and current, while treatment of copper seems to be best at medium time, around 20 minutes, and better at higher or lower current densities.

Given the high p-value for the zinc and copper models, the response surfaces should be discarded and not used for analysis. The treatment results, though they cannot be correlated with treatment duration and current density, are still valid, showing > 93.8 % removal for zinc and > 97.1% for copper.

The effluent copper concentration was at best low enough for environmental condition class II (n=3), and all other samples would fall within this class with a dilution factor of 3. Zn also had three runs with concentrations good enough for class II. All other runs would need a dilution factor of 5 to reach class II, except the two highest concentrations, which needed a dilution factor of 20.

Nickel removal seems to be well described by the model, based on the low p-value and predicted R^2 of 74.41 %. One reason for most consistencies in the nickel removal, compared to other metals, might be the aluminum alloy used for the electrodes. Aluminum type EN AW-5005 contains 0.2 % copper and 0.25 % zinc, but no nickel. As aluminum dissolves from the anodes, some copper and zinc will also dissolve, while nickel will be confined to the top layer of the sludge, as no nickel is added through the EC process. The lowest effluent nickel concentration needs a dilution factor of 4 to be placed in class II, while the highest concentration needs to be diluted 50 times.

The aluminum concentrations stayed put, increased drastically, or reduced after treatment. The runs with lowest effluent concentration, and highest removal rates, are the three runs with the combination of lowest treatment duration and/or lowest current density. Even with these runs, the effluent concentrations of 9 000, 22 000 and 28 000 µg Al/l are very high.

The adjusted R^2 of the aluminum response-surface is 41.97 %, meaning that a bit less than half the variation in output aluminum concentration can be explained by the treatment duration and current density. Some variation in determining the effluent concentration might result from aluminum flocs attached to the beaker wall, or accumulated in the pipe for clarified water access, which might have come into the clarified water samples. The first part of the sample was always discarded, but there might still have been some overestimation of aluminum concentration because of this.

There is no EQS for aluminum, as there are for many other metals, and it therefore falls to the local authority to set the limit of the allowed discharge concentration. Even though there is no national limit, considering the known toxicity of aluminum, the effluent concentration should always be minimized, implying a short duration and/or low current run.

A reason for the high effluent aluminum concentrations might be that the EC and the mixing was turned off at the same time. This would leave the newest dissolved aluminum with insufficient time to hydrolyze and form easily separable sludge. Leaving the stirring on a short while after turning of the electrical current, could possibly have helped reduce aluminum concentrations, and should be tested in future studies.

Comparing the metal removal in this study to removal rates from other treatment methods suitable for mobile solutions listed in Table 4, shows that EC is as good as the other treatment methods. Copper removal is better than for lime precipitation, and comparable to PIX precipitation. Zinc removal using EC is also comparable to removal rates using PIX; but better zinc removal is achieved using lime. Considerably better nickel removal is achieved for EC than the other methods. EC treatment achieves these results in half the time used for PIX or lime precipitation, which could make it more suitable for a mobile system. Aluminum removal was not measured in the other studies, but would probably be higher than for EC, considering the high effluent concentrations encountered in this study.

4.2 SS and turbidity

The good correlation found between SS and turbidity reduction and aluminum removal, looks good, but does not hold up to closer scrutiny. Extrapolating the regression line, even just little, to lower SS and turbidity reduction would get very high outlet aluminum concentrations, something that could make sense if this is caused by more aluminum hydroxide in the clarified water, but not if the water is simply less treated. Removing one outlier in the left region of the figure and three in the right area would give a very different, much steeper regression line.

The failure to establish a plausible correlation between SS and turbidity and any of the metals, probably stems from little variation in the results for these parameters. Most results were around 90 % for SS and > 95 % for turbidity, while metals had mostly > 95 % removal. Since the variation in removal rates is low for all these parameters, the error associated with each value are more pronounced than the differences between these values, which makes it difficult to establish a correlation.

4.3 pH

All the runs had an effluent pH > 8.5, exceeding the limits set for treated TWW by the County Governor of Oslo and Akershus. As the lowest values were achieved for short durations and/or lowest current densities, these settings should be considered if low pH is a priority.

4.4 Sludge production and composition

No sludge settled at the bottom of the reactor after treatment. This might indicate that the method for retrieving TWW from the storage tank, and the long residence time in the tank before the experiments, did not give a representative sample, discriminating against larger, heavier particles. Whether this was reflected in the pollutant concentration, or affected the efficiency of the EC treatment, is not known.

The results for percent sludge volume after treatment are very unsure estimates. The method used was developed post hoc, after suspecting a pattern of increased sludge production with longer treatment duration and/or higher current density. The sludge thickness was measured with the beaker height as comparison. Different angles of the pictures taken, one run missing an estimate, and occasional low contrast between the sludge bottom and the electrode background of the picture, are all obvious weaknesses of this analysis. It is also reasonable to assume that the sludge quantities are overestimated, because of the gas content of the sludge. Stirring the sludge, releasing the gas trapped in it, would probably reduce the sludge volume.

Despite these weaknesses, the measurements produce a compelling correlation of increased sludge production with longer treatment duration and increased current. Comparing with the experiment on deionized water, we see that sludge is produced, even with no particles or colloids present. The sludge from this run was analyzed, showing extremely high concentrations of aluminum, both in itself and compared to other metals. This confirms the hypothesis that the gray sludge consists mainly of aluminum hydroxide flocs. The longer the treatment time, the more aluminum is dissolved from the anodes, forming more aluminum hydroxide sludge. Too long treatment time is not only inefficient as the increased treatment time decreases the surface loading capacity of an EC treatment, but also because it results in unnecessary power consumption, electrode wear and increased sludge production. As the metal removal of other metals than aluminum was good for all settings tested, and no clear pattern for optimizing simultaneous removal of all metals emerged, sludge production might be the most important factor for optimizing the process. Properly tuning the sludge production might also be a key to reducing the effluent aluminum concentration, as the best effluent aluminum concentration coincides with low sludge production. This suggests that the treatment time should not be higher than 10 minutes, and could possibly be as low as 6 minutes.

The sludge production might turn out to be a cheap and effective process control. The production of gray sludge can be observed visually, which will indicate a too long EC residence time, and measures can be taken to reduce the treatment time.

4.5 Space requirements

For this treatment to be applicable for a mobile treatment solution, it needs to be scalable and take up sufficiently small space. The first of those will not be touched upon here, and will have to be a subject of further studies, but assuming upscaling is not an issue, the latter can be superficially considered.

Assuming the same time to volume ratio as for a 10-minute run in the EC unit, with 4 minutes' flotation, and converting it to a flow through reactor, gives 800 ml / 14 min = 0.95 ml/s for this size of a reactor. The volume of water and electrodes, the EC reactor volume, is about 850 ml. The treatment rate per m^3 EC reactor can then be calculated to be 1.1 $l/s/m^3$.

The washing event the TWW was gathered from used 40 m^3 water in 6 hours, giving an average water consumption of 1.85 l/s. From other washing events, it has been observed that less than 80 % of the water used for washing ends up in the storm water system (Garshol et al. 2016a). The uncollected water goes to wetting surfaces and evaporation, and would therefore be more pronounced early in the washing event, only reducing the load at the start of the washing event. The treatment capacity can therefore not be reduced accordingly. From these calculations, an EC treatment unit for 2.2 l/s would take up approximately 2 m^3, excluding housing and all support units such as power supply, pumps, sludge handling, etc. This would provide more capacity than needed for this washing event, but the calculations assume an even load over the whole washing event, and higher capacity is probably needed, depending on flow variations and buffering capabilities. These calculations are also based on a washing event using high-pressure nozzles. For low-pressure nozzles, the water consumption would be considerable higher, increasing the required treatment capacity.

4.6 Sludge dewatering potential

One of the stated research goals was dewatering potential of the sludge produced. This was attempted to measure by decanting all the clarified water from the bottom, transferring the sludge to a narrow cylinder and watching the rate of settling. This was not successful, because the reactor design and method for extracting the clarified water failed to preserve the integrity of the sludge, mixing it with clarified water. Further study is therefore needed to determine this aspect of the EC treatment of TWW, though it seems that the most important part of sludge management is minimizing the sludge production.

4.7 Design considerations

The next step on the way to a full-scale EC solution, would be to try EC treatment of TWW in a flow-through reactor. Such a setup should be designed for a residence time of about 10 to 15 minutes. One of the biggest design challenges is to make a flow-through reactor without dead-zones, with sufficient mixing to allow all pollutants to come into contact with aluminum hydroxide flocs, but at the same time allow for flotation. The design should allow for sludge to be continually scraped off from the top of the reactor, but without creating areas where water can short-circuit through the reactor.

4.8 Application of TWW treatment

When deciding whether to implement new treatment methods for any wastewater, including TWW, a cost-benefit analysis must be made. TWW has very high concentrations of pollutants, but the pollutant production per length of road will be the same if the road is open or in a tunnel. When considering TWW impact in the recipient, both acute effects from extremely high pollutant levels in the TWW, and local chronic effects from long term exposure to elevated pollutant concentration and accumulation of pollutants in biota and sediments should be considered, as these has been documented.

Concern might also be raised for the long-term accumulation of non-degradable pollutants, such as metals, at some place further downstream, where pollutants will settle. As road runoff in general usually is not treated, and normal roads have the same pollutant production as tunnel roads, TWW only provides a small fraction of total pollutant load to the recipient. Treatment of TWW will therefore not contribute much to reducing total pollutant load, and this concern should be of less importance, though such treatment would go a long way in mitigating acute toxic effects.

There are no mobile TWW treatment solutions on the market today, probably because there is no demand for such a solution, and therefore no incentive to develop it. Several methods have been tested the last years, and many of them, including the one featured in this study, show promising results in terms of treatment efficiencies, even for potentially mobile systems. For these studies to translate into actual treatment systems, more legislation demanding TWW treatment is needed, and follow-up on existing treatment solutions that have proven inefficient. As most TWW is discharged without any treatment today, introducing treatment demands could potentially have a great effect, even for moderate treatment standards. A system of carefully planned, gradually increasing treatment standards would allow for a period of research and development for contractors, to make and improve designs, resulting in development of treatment systems and more stringent standards working in tandem.

5 Conclusion

High rates of metal removal from TWW show that electrocoagulation is a robust and suitable technology for this purpose, even though no conclusive picture of optimal settings can be derived from this study.

The exception is aluminum, for which the effluent concentrations are very high. This study indicates that shorter time and lower current density is optimal for minimizing effluent aluminum. Using these settings also reduces the sludge volume significantly, and increases the current loading of the system, increasing the viability of this process as a mobile treatment solution.

Using a current density of 52 A/m^2 with a treatment time of 10 minutes gives good results, low sludge volume, moderate pH increase and high treatment capacity. These settings, with the same electrode spacing (1 cm), would be a good starting point for further studies.

The natural next step would be testing a flow-through reactor, further exploring the feasibility of EC for treating TWW. This reactor should facilitate EC and removal of flotated sludge simultaneously. The first objectives should be to try to replicate the treatment efficiencies for a batch process in a flow-through reactor, and to find ways of reducing effluent aluminum concentrations. If a flow-through reactor with good sludge separation is built, sludge dewatering potential could also be tested.

As this study only focused on metal removal, more research is needed to test the efficacy of removing PAHs, oil and other organic pollutants. The TWW used in this study did not contain detergents. How the presence of detergents affects EC, and the removal rate of detergents from the TWW using EC should also be looked into.

The main reason for treating TWW is to reduce the toxicity of the effluent for organisms in the recipient. Toxicity tests should therefore also be a part of further studies.

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Appendix A – Environmental quality standards for freshwater recipients

Table A-1: Threshold values for metals for different classes of environmental condition in freshwater recipients. All threshold values in µg/l. Table modified from (Miljødirektoratet 2016).

	Class I Background	**Class II** Good AA-MQS	**Class III** Moderate MAC-EQS	**Class IV** Poor	**Class V** Very poor
Cadmium[1] (soft water)	0.003	0.08	0.45	4.5	> 4.5
Lead[1]	0.02	1.2	14	57	> 57
Nickel[1]	0.5	4	34	67	> 67
Copper[2]	0.3	7.8	7.8	15.6	> 15.6
Zinc[2]	1.5	11	11	60	> 60
Arsen[2]	0.15	0.5	8.5	85	> 85
Krom[2]	0.1	3.4	3.4	3.4	> 3.4

[1] EU priority substances

[2] Water region specific substances

Appendix B – Experiment results

Standard Order			1	2	3	4	5	6	7	8	9	10	11	12	13	14	Treatment run on deionized water	15
Run			6	7	3	2	4	1	5	10	11	14	8	12	9	13		15
Time		[m]	10	30	10	30	20	20	20	5.9	34.1	20	20	20	20	20		20
Current		[A]	0.5	0.5	1.5	1.5	1	1	1	1	1	0.29	1.71	1	1	1		1
Current density		[A/m^2]	25.96	25.96	77.88	77.88	51.92	51.92	51.92	51.92	51.92	15.21	88.63	51.92	51.92	51.92		51.9
Flotation time		[m]	4	4	🟥	10	6	🟥	4	4	10	4	6	4	4	4		8
pH	Before		8.23	8.24	8.24	8.27	8.24	8.23	8.25	8.25	8.22	8.30	8.24	8.25	8.24	8.22		6.50
	After		8.77	9.36	🟥	9.60	🟥	9.26	🟥	8.58	9.56	8.99	9.45	9.33	9.38	9.31		9.69
	Difference		**0.54**	**1.12**	🟥	**1.33**	🟥	**1.03**	🟥	**0.33**	**1.34**	**0.69**	**1.21**	**1.08**	**1.14**	**1.09**		**3.19**
SS	Before	SS [mg/l]	876.7	900.0	908.3	895.0	893.3	881.7	900.0	880.0	893.3	930.0	895.0	878.3	890.0	853.3		26.5
	After	SS [mg/l]	60.3	80.0	100.0	118.0	328.0	83.0	103.3	77.0	141.0	67.0	63.5	63.5	81.5	57.5		76.0
	Removal	efficiency	**0.931**	**0.911**	**0.890**	**0.868**	**0.633**	**0.906**	**0.885**	**0.913**	**0.842**	**0.928**	**0.929**	**0.928**	**0.908**	**0.933**		**-1.87**
Turbidity	Before	[NTU]	418	416	414	413	418	404	422	412	412	431	417	412	416	416		0.9
	After	[NTU]	10.1	11.2	13.2	12.5	102	10.5	14.9	9.76	19.0	7.92	7.1	6.88	10.4	8.64		3.97
	Removal	efficiency	**0.976**	**0.973**	**0.968**	**0.970**	**0.757**	**0.974**	**0.965**	**0.976**	**0.954**	**0.982**	**0.983**	**0.983**	**0.975**	**0.979**		**-3.41**
Sludge		[%]	**0.10**	🟥	**0.21**	**0.63**	**0.37**	**0.27**	**0.24**	**0.10**	**0.57**	**0.13**	**0.48**	**0.34**	**0.25**	**0.33**		**0.45**
Copper (Cu)	Before	[µg/l]	680	790	640	780	750	630	760	690	570	840	790	640	680	790		2.8
	After	[µg/l]	16	11	11	4	22	8.9	14	17	16	12	7.1	11	12	6.8		4.5
	Removal	efficiency	**0.976**	**0.986**	**0.983**	**0.995**	**0.971**	**0.986**	**0.982**	**0.975**	**0.972**	**0.986**	**0.991**	**0.983**	**0.982**	**0.991**		**-0.61**
	Sludge	[µg/l]									350		360					250
Nickel (Ni)	Before	[µg/l]	1200	1300	1100	1300	1200	1100	1200	1200	1200	1400	1300	1200	1300	1400		0.50
	After	[µg/l]	100	44	39	13	49	35	36	96	21	62	24	27	32	33		1.5
	Removal	efficiency	**0.917**	**0.966**	**0.965**	**0.990**	**0.959**	**0.968**	**0.970**	**0.920**	**0.983**	**0.956**	**0.982**	**0.978**	**0.975**	**0.976**		**-2.0**
	Sludge	[µg/l]									300		240					79
Zinc (Zn)	Before	[µg/l]	2300	2800	2200	2700	2600	2200	2600	2500	2300	2900	2700	2200	2600	2800		2.0
	After	[µg/l]	38	42	56	30	160	38	150	16	17	8.7	27	6.7	11	29		4.7
	Removal	efficiency	**0.983**	**0.985**	**0.975**	**0.989**	**0.938**	**0.983**	**0.942**	**0.994**	**0.993**	**0.997**	**0.990**	**0.997**	**0.996**	**0.990**		**-1.4**
	Sludge	[µg/l]									480		420					460
Aluminum (Al)	Before	[µg/l]	46000	54000	45000	53000	52000	43000	52000	44000	45000	44000	52000	42000	44000	50000		15
	After	[µg/l]	22000	55000	44000	59000	83000	48000	52000	28000	83000	9000	47000	36000	44000	48000		57000
	Removal	efficiency	**0.522**	**-0.019**	**0.022**	**-0.113**	**-0.596**	**-0.116**	**0.000**	**0.364**	**-0.844**	**0.795**	**0.096**	**0.143**	**0.000**	**0.040**		**-3799**
	Sludge	[µg/l]									950000		1100000					1700000
Arsen (As)	Before	[µg/l]	14	14	12	13	14	11	14	14	12	16	13	15	14	17		0.30
	After	[µg/l]	1.6	1.6	1.6	1.6	1.6	1.6	1.6	0.65	0.28	0.6	1.6	0.48	0.47	1.6		0.33
	Removal	efficiency	**0.886**	**0.886**	**0.867**	**0.877**	**0.886**	**0.855**	**0.886**	**0.954**	**0.977**	**0.963**	**0.877**	**0.968**	**0.966**	**0.906**		**-0.10**
	Sludge	[µg/l]									2.0		1.6					0.59
Lead (Pb)	Before	[µg/l]	37	42	34	42	43	34	41	33	26	44	41	30	32	39		0.2
	After	[µg/l]	1.6	1.6	1.6	1.6	1.6	1.6	1.6	1.6	1.6	0.8	1.6	0.34	1.6	1.6		0.20
	Removal	efficiency	**0.957**	**0.962**	**0.953**	**0.962**	**0.963**	**0.953**	**0.961**	**0.952**	**0.938**	**0.982**	**0.961**	**0.989**	**0.950**	**0.959**		**0**
	Sludge	[µg/l]									21		26					16
Cadmium (Cd)	Before	[µg/l]	11	13	11	12	13	11	12	11	9.8	14	12	11	11	13		0.036
	After	[µg/l]	0.32	0.065	0.080	0.080	0.20	0.080	0.032	0.23	0.073	0.19	0.080	0.800	0.081	0.080		2.0
	Removal	efficiency	**0.971**	**0.995**	**0.993**	**0.993**	**0.985**	**0.993**	**0.997**	**0.979**	**0.993**	**0.986**	**0.993**	**0.927**	**0.993**	**0.994**		**-55**
	Sludge	[µg/l]									1.7		1.3					0.32
Chromium (Cr)	Before	[µg/l]	2200	2500	2100	2500	2400	2000	2400	2400	2200	2700	2500	2300	2000	2300		0.50
	After	[µg/l]	20	22	17	7.5	36	16	14	16	22	9.9	11	8.7	14	11		6.2
	Removal	efficiency	**0.991**	**0.991**	**0.992**	**0.997**	**0.985**	**0.992**	**0.994**	**0.993**	**0.990**	**0.996**	**0.996**	**0.996**	**0.993**	**0.995**		**-11**
	Sludge	[µg/l]									650		580					570

Concentration below LOQ, or removal efficiency based on data below LOQ

Based on one sample, instead of two

🟥 Data missing

Appendix C – Statistics printouts for response-surface models

Response Surface Regression: Cu removal [%] versus Blocks; Time [m]; Current density [A/m^2]

```
Analysis of Variance

Source                                               DF    Adj SS    Adj MS  F-Value  P-Value
Model                                                 5  0,000318  0,000064     1,41    0,316
  Blocks                                              1  0,000000  0,000000     0,01    0,931
  Linear                                              2  0,000109  0,000054     1,21    0,348
    Time [m]                                          1  0,000035  0,000035     0,78    0,402
    Current density [A/m^2]                           1  0,000074  0,000074     1,63    0,237
  Square                                              2  0,000218  0,000109     2,42    0,151
    Time [m]*Time [m]                                 1  0,000085  0,000085     1,88    0,207
    Current density [A/m^2]*Current density [A/m^2]   1  0,000117  0,000117     2,59    0,146
Error                                                 8  0,000360  0,000045
  Lack-of-Fit                                         4  0,000186  0,000046     1,06    0,477
  Pure Error                                          4  0,000175  0,000044
Total                                                13  0,000678


Model Summary

        S    R-sq  R-sq(adj)  R-sq(pred)
0,0067123  46,85%     13,63%       0,00%


Coded Coefficients

Term                                                Effect       Coef   SE Coef  T-Value
Constant                                                      0,99219   0,00953   104,14
Blocks
  1                                                          -0,00016   0,00179    -0,09
Time [m]                                           0,00420    0,00210   0,00237     0,89
Current density [A/m^2]                          -0,000492  -0,000246  0,000192    -1,28
Time [m]*Time [m]                                 -0,00678   -0,00339   0,00247    -1,37
Current density [A/m^2]*Current density [A/m^2]   0,000003   0,000001  0,000001     1,61

Term                                             P-Value    VIF
Constant                                           0,000
Blocks
  1                                                0,931   1,00
Time [m]                                           0,402   1,00
Current density [A/m^2]                            0,237  17,69
Time [m]*Time [m]                                  0,207   1,01
Current density [A/m^2]*Current density [A/m^2]    0,146  17,70


Regression Equation in Uncoded Units

Cu removal [%] = 0,9749 + 0,00157 Time [m] - 0,000504 Current density [A/m^2]
                 - 0,000034 Time [m]*Time [m]
                 + 0,000006 Current density [A/m^2]*Current density [A/m^2]

Equation averaged over blocks.
```

Response Surface Regression: Ni removal [%] versus Blocks; Time [m]; Current density [A/m^2]

```
Analysis of Variance

Source                              DF    Adj SS    Adj MS  F-Value  P-Value
Model                                5  0,005767  0,001153    26,54    0,000
  Blocks                             1  0,000085  0,000085     1,95    0,200
  Linear                             2  0,002419  0,001209    27,83    0,000
    Time [m]                         1  0,000954  0,000954    21,95    0,002
    Current density [A/m^2]          1  0,001465  0,001465    33,71    0,000
  Square                             1  0,000740  0,000740    17,02    0,003
    Time [m]*Time [m]                1  0,000740  0,000740    17,02    0,003
  2-Way Interaction                  1  0,000145  0,000145     3,33    0,106
    Time [m]*Current density [A/m^2]  1  0,000145  0,000145     3,33    0,106
Error                                8  0,000348  0,000043
  Lack-of-Fit                        4  0,000278  0,000070     4,00    0,104
  Pure Error                         4  0,000070  0,000017
Total                               13  0,006115


Model Summary

        S    R-sq  R-sq(adj)  R-sq(pred)
0,0065918  94,32%     90,76%      74,41%


Coded Coefficients
```

```
Term                              Effect       Coef   SE Coef  T-Value  P-Value   VIF
Constant                                    0,94371   0,00509   185,37    0,000
Blocks
  1                                        -0,00246   0,00176    -1,40    0,200  1,00
Time [m]                         0,06443    0,03221   0,00688     4,69    0,002  8,70
Current density [A/m^2]         0,000521   0,000261  0,000045     5,81    0,000  1,00
Time [m]*Time [m]               -0,01996   -0,00998   0,00242    -4,13    0,003  1,00
Time [m]*Current density [A/m^2]  -0,000232  -0,000116  0,000064    -1,82    0,106  8,70


Regression Equation in Uncoded Units

Ni removal [%] = 0,8384 + 0,00724 Time [m] + 0,000985 Current density [A/m^2]
                 - 0,000100 Time [m]*Time [m] - 0,000023 Time [m]*Current density [A/m^2]

Equation averaged over blocks.


Fits and Diagnostics for Unusual Observations

     Ni removal
Obs         [%]      Fit     Resid  Std Resid
  8     0,98154  0,99185  -0,01031      -2,08  R

R  Large residual
```

Response Surface Regression: Zn removal [%] versus Blocks; Time [m]; Current density [A/m^2]

```
Analysis of Variance

Source                                             DF    Adj SS    Adj MS  F-Value  P-Value
Model                                               5  0,002542  0,000508     1,98    0,186
  Blocks                                            1  0,001832  0,001832     7,15    0,028
  Linear                                            2  0,000417  0,000208     0,81    0,477
    Time [m]                                        1  0,000026  0,000026     0,10    0,758
    Current density [A/m^2]                         1  0,000391  0,000391     1,52    0,252
  Square                                            2  0,000656  0,000328     1,28    0,329
    Time [m]*Time [m]                               1  0,000343  0,000343     1,34    0,281
    Current density [A/m^2]*Current density [A/m^2]  1  0,000363  0,000363     1,42    0,268
Error                                               8  0,002051  0,000256
  Lack-of-Fit                                       4  0,000817  0,000204     0,66    0,650
  Pure Error                                        4  0,001233  0,000308
Total                                              13  0,004593


Model Summary

        S    R-sq  R-sq(adj)  R-sq(pred)
0,0160104  55,35%     27,45%       0,00%


Coded Coefficients

Term                                               Effect       Coef   SE Coef  T-Value
Constant                                                      1,0050    0,0227    44,22
Blocks
  1                                                         -0,01144   0,00428    -2,67
Time [m]                                          0,00362    0,00181   0,00566     0,32
Current density [A/m^2]                         -0,001133  -0,000566  0,000459    -1,23
Time [m]*Time [m]                                 0,01363    0,00682   0,00589     1,16
Current density [A/m^2]*Current density [A/m^2]   0,000005   0,000003  0,000002     1,19

Term                                            P-Value    VIF
Constant                                          0,000
Blocks
  1                                               0,028   1,00
Time [m]                                          0,758   1,00
Current density [A/m^2]                           0,252  17,69
Time [m]*Time [m]                                 0,281   1,01
Current density [A/m^2]*Current density [A/m^2]    0,268  17,70


Regression Equation in Uncoded Units

Zn removal [%] = 1,0298 - 0,00255 Time [m] - 0,001154 Current density [A/m^2]
                 + 0,000068 Time [m]*Time [m]
                 + 0,000010 Current density [A/m^2]*Current density [A/m^2]
```

Response Surface Regression: Al removal [%] versus Blocks; Time [m]; Current density [A/m^2]

```
Analysis of Variance

Source                                                DF   Adj SS   Adj MS  F-Value  P-Value
Model                                                  4  1,63474  0,40869     7,03    0,008
  Blocks                                               1  0,05687  0,05687     0,98    0,348
  Linear                                               2  1,44393  0,72197    12,42    0,003
    Time [m]                                           1  0,71053  0,71053    12,22    0,007
    Current density [A/m^2]                            1  0,73340  0,73340    12,62    0,006
  Square                                               1  0,55428  0,55428     9,53    0,013
    Current density [A/m^2]*Current density [A/m^2]    1  0,55428  0,55428     9,53    0,013
Error                                                  9  0,52321  0,05813
  Lack-of-Fit                                          5  0,31261  0,06252     1,19    0,446
  Pure Error                                           4  0,21060  0,05265
Total                                                 13  2,15795


Model Summary

       S    R-sq  R-sq(adj)  R-sq(pred)
0,241111  75,75%     64,98%      41,97%


Coded Coefficients

Term                                               Effect      Coef   SE Coef  T-Value
Constant                                                      1,305     0,341     3,83
Blocks
  1                                                         -0,0637    0,0644    -0,99
Time [m]                                          -0,5960   -0,2980    0,0852    -3,50
Current density [A/m^2]                          -0,04894  -0,02447   0,00689    -3,55
Current density [A/m^2]*Current density [A/m^2]  0,000203  0,000101  0,000033     3,09

Term                                             P-Value    VIF
Constant                                           0,004
Blocks
  1                                                0,348   1,00
Time [m]                                           0,007   1,00
Current density [A/m^2]                            0,006  17,59
Current density [A/m^2]*Current density [A/m^2]    0,013  17,59


Regression Equation in Uncoded Units

Al removal [%] = 1,950 - 0,02980 Time [m] - 0,0497 Current density [A/m^2]
                 + 0,000406 Current density [A/m^2]*Current density [A/m^2]

Equation averaged over blocks.
```

Response Surface Regression: SS removal [%] versus Blocks; Time [m]; Current density [A/m^2]

```
Method

Rows unused  1


Analysis of Variance

Source                                                DF    Adj SS    Adj MS  F-Value  P-Value
Model                                                  5  0,006767  0,001353     4,07    0,047
  Blocks                                               1  0,000461  0,000461     1,38    0,278
  Linear                                               2  0,003258  0,001629     4,89    0,047
    Time [m]                                           1  0,002498  0,002498     7,50    0,029
    Current density [A/m^2]                            1  0,000760  0,000760     2,28    0,174
  Square                                               2  0,002877  0,001438     4,32    0,060
    Time [m]*Time [m]                                  1  0,002101  0,002101     6,31    0,040
    Current density [A/m^2]*Current density [A/m^2]    1  0,000459  0,000459     1,38    0,279
Error                                                  7  0,002330  0,000333
  Lack-of-Fit                                          4  0,001791  0,000448     2,49    0,239
  Pure Error                                           3  0,000539  0,000180
Total                                                 12  0,009097


Model Summary

        S    R-sq  R-sq(adj)  R-sq(pred)
0,0182437  74,39%     56,10%       0,00%
```

```
Coded Coefficients

Term                                                Effect       Coef   SE Coef  T-Value
Constant                                                       0,9624    0,0261    36,88
Blocks
  1                                                          -0,00600   0,00510    -1,18
Time [m]                                          -0,03534   -0,01767   0,00645    -2,74
Current density [A/m^2]                          -0,001630  -0,000815  0,000539    -1,51
Time [m]*Time [m]                                 -0,03485   -0,01742   0,00694    -2,51
Current density [A/m^2]*Current density [A/m^2]   0,000006   0,000003  0,000003     1,17

Term                                             P-Value    VIF
Constant                                           0,000
Blocks
  1                                                0,278   1,01
Time [m]                                           0,029   1,00
Current density [A/m^2]                            0,174  18,81
Time [m]*Time [m]                                  0,040   1,02
Current density [A/m^2]*Current density [A/m^2]    0,279  18,84


Regression Equation in Uncoded Units

SS removal [%] = 0,9297 + 0,00520 Time [m] - 0,00165 Current density [A/m^2]
                 - 0,000174 Time [m]*Time [m]
                 + 0,000012 Current density [A/m^2]*Current density [A/m^2]

Equation averaged over blocks.
```

Response Surface Regression: Turbidity reduct versus Blocks; Time [m]; Current density

```
Method

Rows unused  1


Analysis of Variance

Source                                               DF    Adj SS    Adj MS  F-Value  P-Value
Model                                                 5  0,000535  0,000107     2,80    0,106
  Blocks                                              1  0,000078  0,000078     2,03    0,197
  Linear                                              2  0,000215  0,000107     2,81    0,127
    Time [m]                                          1  0,000132  0,000132     3,45    0,105
    Current density [A/m^2]                           1  0,000083  0,000083     2,16    0,185
  Square                                              2  0,000307  0,000154     4,02    0,069
    Time [m]*Time [m]                                 1  0,000196  0,000196     5,12    0,058
    Current density [A/m^2]*Current density [A/m^2]   1  0,000073  0,000073     1,92    0,208
Error                                                 7  0,000268  0,000038
  Lack-of-Fit                                         4  0,000191  0,000048     1,86    0,318
  Pure Error                                          3  0,000077  0,000026
Total                                                12  0,000802


Model Summary

        S    R-sq  R-sq(adj)  R-sq(pred)
0,0061827  66,65%     42,84%       0,00%


Coded Coefficients

Term                                                Effect       Coef   SE Coef  T-Value
Constant                                                      0,98959   0,00884   111,90
Blocks
  1                                                          -0,00246   0,00173    -1,43
Time [m]                                          -0,00813   -0,00406   0,00219    -1,86
Current density [A/m^2]                          -0,000537  -0,000269  0,000183    -1,47
Time [m]*Time [m]                                 -0,01064   -0,00532   0,00235    -2,26
Current density [A/m^2]*Current density [A/m^2]   0,000002   0,000001  0,000001     1,39

Term                                             P-Value    VIF
Constant                                           0,000
Blocks
  1                                                0,197   1,01
Time [m]                                           0,105   1,00
Current density [A/m^2]                            0,185  18,81
Time [m]*Time [m]                                  0,058   1,02
Current density [A/m^2]*Current density [A/m^2]    0,208  18,84
```

```
Regression Equation in Uncoded Units

Turbidity reduction [%] = 0,9770 + 0,001722 Time [m] - 0,000547 Current density [A/m^2]
                          - 0,000053 Time [m]*Time [m]
                          + 0,000005 Current density [A/m^2]*Current density [A/m^2]

Equation averaged over blocks.


Fits and Diagnostics for Unusual Observations

     Turbidity
     reduction
Obs        [%]      Fit     Resid  Std Resid
 11    0,95395  0,96086  -0,00691      -2,05  R

R  Large residual
```

Response Surface Regression: pH increase versus Blocks; Time [m]; Current density [A/m^2]

```
Method

Rows unused  3


Analysis of Variance

Source                       DF   Adj SS    Adj MS  F-Value  P-Value
Model                         4  1,02197  0,255491    29,64    0,000
  Blocks                      1  0,00004  0,000040     0,00    0,948
  Linear                      2  0,96194  0,480968    55,79    0,000
    Time [m]                  1  0,67664  0,676637    78,49    0,000
    Current density [A/m^2]   1  0,13759  0,137588    15,96    0,007
  Square                      1  0,06734  0,067335     7,81    0,031
    Time [m]*Time [m]         1  0,06734  0,067335     7,81    0,031
Error                         6  0,05173  0,008621
  Lack-of-Fit                 4  0,04966  0,012415    12,01    0,078
  Pure Error                  2  0,00207  0,001033
Total                        10  1,07369


Model Summary

        S    R-sq  R-sq(adj)  R-sq(pred)
0,0928490  95,18%     91,97%      75,90%


Coded Coefficients

Term                       Effect      Coef   SE Coef  T-Value  P-Value   VIF
Constant                             0,7540    0,0771     9,78    0,000
Blocks
  1                                 -0,0020    0,0301    -0,07    0,948  1,07
Time [m]                   0,6450    0,3225    0,0364     8,86    0,000  1,06
Current density [A/m^2]  0,005605  0,002802  0,000701     3,99    0,007  1,06
Time [m]*Time [m]         -0,2045   -0,1022    0,0366    -2,79    0,031  1,02


Regression Equation in Uncoded Units

pH increase = -0,306 + 0,0731 Time [m] + 0,00560 Current density [A/m^2]
              - 0,001022 Time [m]*Time [m]

Equation averaged over blocks.


Fits and Diagnostics for Unusual Observations

           pH
Obs  increase     Fit    Resid  Std Resid
 14    0,6900  0,8356  -0,1456      -2,25  R

R  Large residual
```

Response Surface Regression: Sludge [%] versus Blocks; Time [m]; Current density [A/m^2]

```
Method

Rows unused  1


Analysis of Variance

Source                               DF    Adj SS    Adj MS  F-Value  P-Value
Model                                 4  0,328866  0,082217    41,25    0,000
  Blocks                              1  0,000498  0,000498     0,25    0,631
  Linear                              2  0,103801  0,051900    26,04    0,000
    Time [m]                          1  0,000781  0,000781     0,39    0,549
    Current density [A/m^2]           1  0,084007  0,084007    42,15    0,000
  2-Way Interaction                   1  0,008480  0,008480     4,25    0,073
    Time [m]*Current density [A/m^2]  1  0,008480  0,008480     4,25    0,073
Error                                 8  0,015945  0,001993
  Lack-of-Fit                         4  0,001801  0,000450     0,13    0,965
  Pure Error                          4  0,014145  0,003536
Total                                12  0,344812


Model Summary

        S    R-sq  R-sq(adj)  R-sq(pred)
0,0446451  95,38%     93,06%      91,31%


Coded Coefficients

Term                                Effect      Coef   SE Coef  T-Value  P-Value    VIF
Constant                                      0,0741    0,0407     1,82    0,107
Blocks
  1                                          -0,0065    0,0131    -0,50    0,631   1,11
Time [m]                            0,0819    0,0409    0,0654     0,63    0,549  14,85
Current density [A/m^2]           0,004586  0,002293  0,000353     6,49    0,000   1,17
Time [m]*Current density [A/m^2]  0,002312  0,001156  0,000560     2,06    0,073  14,35


Regression Equation in Uncoded Units

Sludge [%] = -0,008 + 0,00386 Time [m] - 0,00004 Current density [A/m^2]
             + 0,000231 Time [m]*Current density [A/m^2]

Equation averaged over blocks.
```

Appendix D – Statistics printouts for mean removal rates

Descriptive Statistics: Zn removal [; SS removal [; Turbidity reduction

```
                         Total
Variable                 Count   N  N*     Mean  SE Mean    StDev  Minimum  Maximum    Range
Zn removal [%]              14  14   0  0,98221  0,00502  0,01880  0,93846  0,99700  0,05854
SS removal [%]              14  13   1  0,90554  0,00764  0,02753  0,84216  0,93262  0,09045
Turbidity reduction [%]     14  13   1  0,97370  0,00227  0,00818  0,95395  0,98329  0,02934
```

Appendix E – Statistics printouts for correlation models

Correlation: SS removal [; Turbidity re; Cu removal [; Ni removal [; Zn removal [; ...

```
                    SS removal [%]  Turbidity reduct    Cu removal [%]    Ni removal [%]     Zn removal [%]
Turbidity reduct             0,925
                             0,000

Cu removal [%]               0,256             0,451
                             0,398             0,122

Ni removal [%]              -0,388            -0,193             0,507
                             0,190             0,527             0,064

Zn removal [%]               0,256             0,416             0,357             0,045
                             0,398             0,157             0,210             0,877

Al removal [%]               0,774             0,742             0,316            -0,518          0,361
                             0,002             0,004             0,271             0,058          0,204

Cell Contents: Pearson correlation
               P-Value
```

Spearman Rho: SS removal [; Turbidity re; Cu removal [; Ni removal [; Zn removal [; ...

```
                    SS removal [%]  Turbidity reduct    Cu removal [%]    Ni removal [%]     Zn removal [%]
Turbidity reduct             0,846
                             0,000

Cu removal [%]               0,223             0,270
                             0,464             0,373

Ni removal [%]              -0,346            -0,104             0,438
                             0,247             0,734             0,117

Zn removal [%]               0,308             0,610             0,134             0,209
                             0,306             0,027             0,647             0,474

Al removal [%]               0,773             0,740             0,099            -0,436            -0,473
                             0,002             0,004             0,736             0,119             0,088
Cell Contents: Spearman rho
               P-Value
```

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