APPLICATION OF ELECTROCOAGULATION FOR WATER CONDITIONING

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Abstract: *Water conditioning is a method of removing altering minerals, chemicals and contaminants from a water source and it is carried out on facilities equipped with the corresponding electro-mechanical equipment. Although efficient, conventional processes typically use several complex devices connected to a single functional unit, which are often expensive to maintain and occupy large areas. Therefore, the aim of this paper is to present the electrocoagulation (EC) method as an alternative to conventional water conditioning processes. The examples of previous studies of the EC process application is presented in this paper. The focus of the paper is to investigate the influence of the certain operational parameters such as pH, temperature, electrode material, etc., on the efficiency of pollutant removal such as Escherichia coli and elevated concentrations of iron, arsenic, manganese, ammonia and others. Further, an economic analysis is made, which, from an economic point of view, shows when it is feasible to use the EC in the conditioning process. Furthermore, a case study of electrocoagulation process for Total Nitrogen (TN) removal is presented. According to results, 69.7 % of TN was removed with aluminum electrodes after 240 minutes. For this case, total operating costs were 7.60 €/m*^{3}.
Keywords: *electrocoagulation, water conditioning, operative parameters, total nitrogen*

Received: 27.06.2019. / Accepted: 18.11.2019.
Published online: 09.12.2019.

Original scientific paper

1. INTRODUCTION

Access to clean drinking water is a basic human right. According to the Council Directive 98/83/EC (Official Journal L 330) on the quality of water intended for human consumption, drinking water shall be wholesome and clean if it is free from any micro-organisms, parasites and substances which in numbers or concentrations constitute a potential danger to human health, whereby must meet minimum requirements regarding microbiological and chemical properties, radioactivity and physical properties.

In case of unsatisfactory values of certain parameters established by the Council Directive 98/83/EC (Official Journal L 330), appropriate activities shall be undertaken to ensure the quality of the water and, where necessary, to prohibit or limit the use of the water. Water conditioning is a method of removing altering minerals, chemicals and contaminants from a water source and it is carried out on facilities equipped with the corresponding electro-mechanical equipment (Vuković 2017).

According to (Croatian Institute of Public Health 2018) in 69 % of water supply areas water is not processed before the distribution to the consumers. In the remaining 31 % of water supply areas, the water is treated, dominating the filtration process, and a combination of aeration and filtration, a combination of filtration, coagulation, flocculation and precipitation, and manganese and/or iron removal are also used (**Figure 1**). Although efficient, conventional processes typically use several complex devices connected to a single functional unit, which are often expensive to maintain and occupy large areas. Therefore, the aim of this paper is to present the electrocoagulation (EC) method as an alternative to conventional water conditioning processes.

Furthermore, (Croatian Institute of Public Health 2018) analyzed the quality of water for human consumption and stated that the most common cause of its malfunction is the presence of *Escherichia coli (E. coli)* and total coliforms, as well as the elevated concentrations of iron, arsenic, manganese, ammonia, color, smell and turbidity. Therefore, the focus of the paper is to investigate the influence of the certain operational parameters such as pH, temperature, electrode material, etc., on the efficiency of removing the mentioned contaminants.

An economic analysis is also made, which, from an economic point of view, shows when it is feasible to use EC in the conditioning process.

Additionally, the influence on total nitrogen removal efficiency was evaluated for some of the process parameters on a full-scale EC unit. For this specific case, operating costs are also determined.

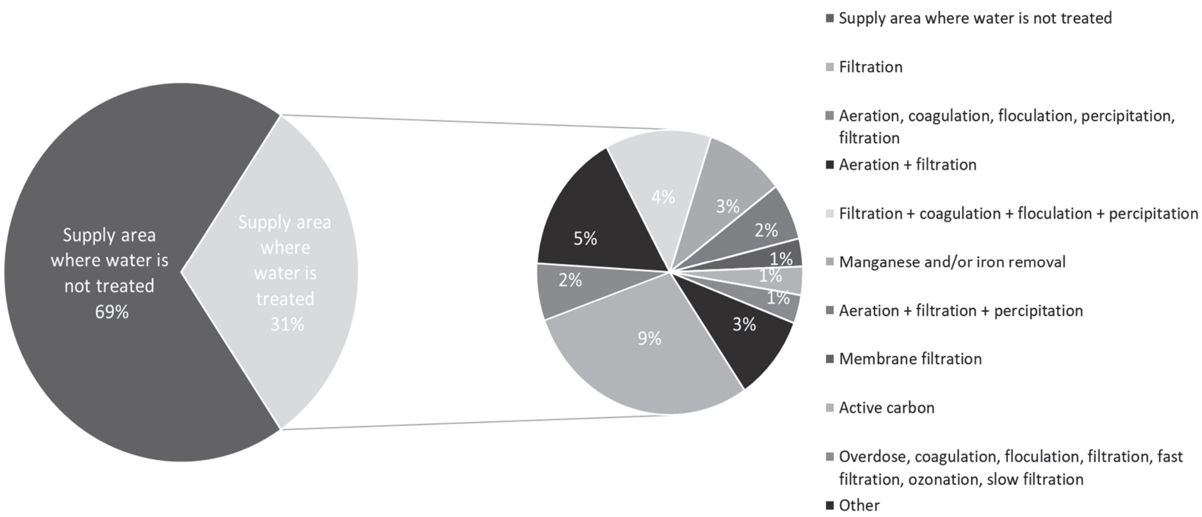

Figure 1. *Water conditioning technologies (Croatian Institute of Public Health, 2018)*

2. THEORY OF ELECTROCOAGULATION

The EC process, **Figure 2**, combines the benefits of coagulation, flotation or precipitation and electrochemistry (Moussa 2017). It includes coagulation and precipitation of pollutants (suspended solids and solutes) from the wastewater by the use of electricity and sacrificing electrodes for the "in situ" coagulant production (Gardić 2007). In the EC reactor, the wastewater flows between electrodes while the direct current is applied to them. Electrodes are usually made of metal, mostly iron (Fe) or aluminum (Al), because these materials are cheap, available, non-toxic and proven effective. The choice of electrode material and the arrangement of electrodes depend on the wastewater contamination and the required effluent quality. Usually, aluminum is used for the drinking water treatment and iron for the wastewater treatment (Chen and Hung 2007; Shammas et al. 2010; Kuokkanen et al. 2013; Hakizimana 2017).

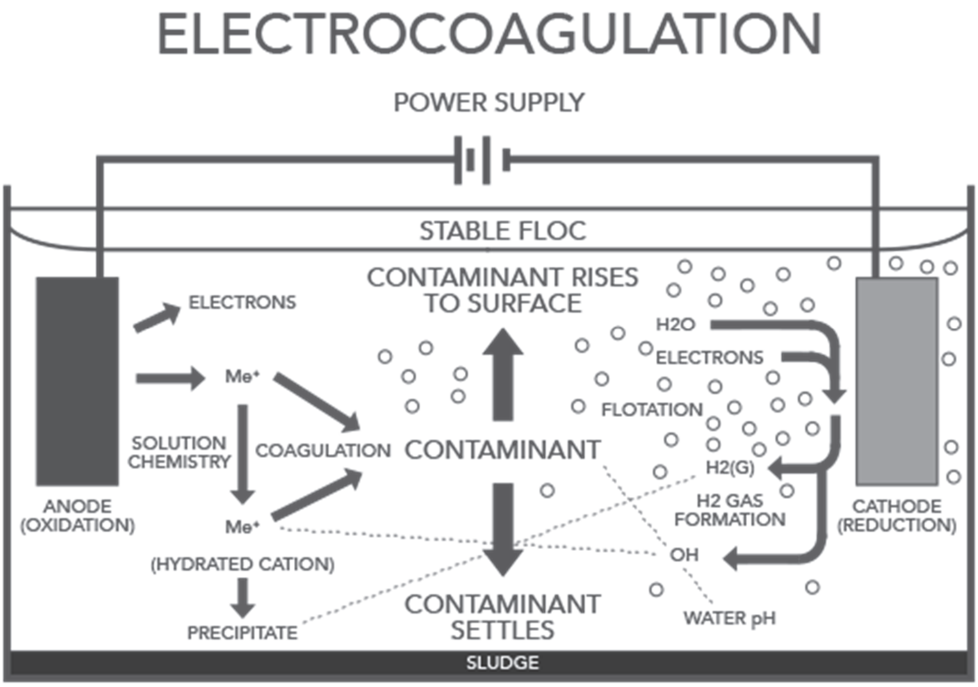

Figure 2. *Electrocoagulation process*
(https://www.crs-reprocessing.com/en/crs-solutions/electrocoagulation/, Accessed March 11 2019)

According to the **Equation 1**, when the current is passed through metal electrode, the metal (M) from the anode is oxidized to its cations (M^{n+}). Simultaneously, water is reduced to hydrogen gas and the hydroxide ion (OH^-) on the cathode (**Equation 2**) (Kabdaşli et al. 2012):

$$M \rightarrow M^{n+} + ne^- \quad (1)$$

$$2H_2O(l) + 2e^- \rightarrow H_2(g) + 2OH^-\ (aq) \quad (2)$$

By forming monomeric and polymeric hydroxides, metal cations (M^{n+}) destabilize colloidal particles, i.e. trap colloidal particles and create flocs which can be easily removed from water by sedimentation or flotation (Kabdaşli et al. 2012; Pirkarami & Olya 2017).

Some of the advantages of the EC process are: effluent contains less total dissolved solids compared to the other chemical processes, easy maintenance of the device, more efficient and faster degradation of organic matter compared to chemical coagulation, larger and more stable flocs are formed than those produced by chemical coagulation, it is not necessary to control the pH of the water, except in extreme cases, no chemicals are required, reduces residue, it can process multiple pollutants which can easily be removed, operating costs are much lower compared to most conventional technologies, the device is smaller and simpler than the coagulation device so it can be used as decentralized process and if the solar panels are used, the device can be used as a batch process in rural areas that don't have access to the electricity for processing the smaller quantities of wastewater (Vepsäläinen, 2012; Kuokkanen et al. 2013; Marriaga-Cabarales & Machuca-Martínez 2014; Hakizimana et al. 2017).

However, some of the EC disadvantages are: in some countries, the use of electricity may be expensive, possible passivation of anode due to the oxygen presence and the deposition on the cathodes (can be overcomed by switching the electrode poles), the electrodes need to be regularly replaced which increases the maintenance costs, the high conductivity of the wastewater is required, the high concentrations of iron and aluminum need to be removed from the effluent, in some cases, the gelatinous hydroxides may be dissolved in water, it is not effective for the removal of the soluble substances such as sugars, organic acids, solvents, phenols, alcohol and similar (Vepsäläinen 2012; Kuokkanen et al. 2013; Marriaga-Cabarales & Machuca-Martínez 2014; Hakizimana et al. 2017).

3. APPLICATION OF ELECTROCOAGULATION FOR WATER CONDITIONING

Water health parameters for human consumption are determined by the Ordinance on conformity parameters, analytical methods, monitoring and drinking water safety plans, and keeping register of legal entities which provide public water supply (Official Gazette 125/17). The aim of monitoring these parameters is to protect human health from the adverse impact of any contamination of water intended for human consumption and to ensure its health. According to the (Official Gazette 125/17), drinking water should be free of color, taste and smell. Maximum permissible concentrations of E. coli, iron, arsenic, manganese, ammonia, color, smell and turbidity, according to the (Official Gazette 125/17), are shown in **Table 1**.

Table 1. Chemical and indicator parameters

PARAMETER	UNIT	MAXIMUM PERMISSIBLE CONCENTRATIONS
Ammonia	mg/l	0.50
Arsenic	μg/l	10
Escherichia coli	*E. coli*/100 ml	0
Iron	μg/l	200
Manganese	μg/l	50
Color	mg/PtCo	20
Smell	/	-
Turbidity	NTU	4

3.1. Removal of E. coli

Microbiological contamination of water can be effectively counteracted by disinfection measures. Disinfection is the last stage of water preparation for the purpose of eliminating or decreasing the number of microorganisms in it. Mostly, chemical disinfection uses chlorine, chlorine dioxide or ozone. However, water disinfection is, most often, just one step in the water treatment and is often combined with other chemical processes (Andrija Stampar Teaching Institute of Public Health 2017). The examples in **Table 2** show that EC has very high efficiency of *E. coli* removal. It can be noticed that 30 min is enough for complete *E. coli* removal with Al electrodes.

Table 2. Recent applications of EC for *E. coli* removal

VOLUME TREATED	OPTIMAL OPERATIVE PARAMETERS	% REMOVAL	OPERATING COSTS [€/m^3]	REFERENCE
0.5 l	Al electrodes; Electrode distance: 5 cm; 12 V; treatment time 30 min	100	-	Ghernaout et al. (2008)
1 l	Al electrodes; Electrode distance: 2 cm; 30 V; 22 A; treatment time: 30 min	99.8	-	Ricordel et al. (2014)

3.2. Iron removal

The presence of iron in drinking water is not directly harmful to human health, but problems with discoloration, turbidity and unpleasant taste occur (Doggaz et al. 2018). There are several methods for removal of iron from drinking water, but aeration and separation are the most common methods (Gosh 2007). Some of the recent applications of EC for iron removal are shown in **Table 3**. Most commonly Al electrodes or their combinations with other materials are used. Also, the increase of current density influences the treatment efficiency and shortens the electrolysis time.

Table 3. Recent applications of EC for iron removal

VOLUME TREATED	OPTIMAL OPERATIVE PARAMETERS	% REMOVAL	OPERATING COSTS [€/m^3]	REFERENCE
3 l	Al electrodes; Electrode distance: 0.5 cm; treatment time 35 min	99.2	5.35	Gosh et al. (2007)
1 l	Mn anode, Fe cathode; 6 A/m^2; treatment time: 60 min	98.4	-	Vasudevan et al. (2009a)
1 l	Al anode, SS cathode; 6 A/m^2; treatment time: 60 min; pH 6.5	98.8	-	Vasudevan et al. (2009b)
2.2 l	Al electrodes; Electrode distance: 0.5 cm; 15 A/m^2; treatment time: 20 min; pH 6	98.5	0.20	Hashim et al. (2017)

3.3. Arsenic removal

Among several investigated technologies for removal of arsenic from drinking water, most common are ion-exchange, precipitation, coagulation/adsorption and membrane treatment systems. Although these processes produce high quality water, they require expensive resins, replaceable adsorption media and chemicals (Alferness 2016). It can be noticed that EC is a very efficient method for arsenic removal (**Table 4**). Total arsenic removal can be achieved under less than 30 min with Fe electrodes at an inter-distance of about 1 cm. Applied voltage should be around 12-15 V.

Table 4. Recent applications of EC for arsenic removal

VOLUME TREATED	OPTIMAL OPERATIVE PARAMETERS	% REMOVAL	OPERATING COSTS [€/m^3]	REFERENCE
1 l	Fe electrodes; Electrode distance: 2 cm; 12 V; treatment time 30 min; pH 7	100	-	Wan et al. (2011)
1.4 l	Fe electrodes; Electrode distance: 0.5 cm; 0.54 mA/cm^2; treatment time: 30 min; pH 4	99.5	-	Can et al. (2014)
20 l	Al electrodes; Electrode distance: 0.55 cm; 5.5 mA/cm^2; treatment time: 15 min	92.2	-	Flores et al. (2014)
10 l	Fe and Al electrodes; Electrode distance: 1 cm; 6 A; 15 V; treatment time: 20 min	100	-	Oreščanin et al. (2014)
13.2 l	SS electrodes; Electrode distance: 2.2 cm; 6 V; treatment time: 60 min	96.7	0.47	Alferness et al. (2016)

3.4. Manganese removal

The presence of manganese and other metals in drinking water may be responsible for its coloration. Conventional methods for removing manganese include chemical precipitation, coagulation, flotation, ion-exchange, oxidation/filtration, adsorption and membrane filtration (Alvarez-Bastida et al. 2018). Some of the recent applications of EC for manganese removal are shown in **Table 5**. Manganese removal efficiency varies from 50-100 %, depending on the treatment time and applied voltage. It can be assumed that the optimal operative parameters are Fe electrodes, electrode distance of 2 cm, 90 min of treatment time, pH 7 and current density of 15 mA/cm^2. Also, the addition of supporting electrolyte, such as SO_4^{2-}, helps to increase the removal efficiency. More research on manganese removal needs to be done, and until then, it is suggested to combine it with other water treatments.

Table 5. Recent applications of EC for manganese removal

VOLUME TREATED	OPTIMAL OPERATIVE PARAMETERS	% REMOVAL	OPERATING COSTS [€/m^3]	REFERENCE
0.5 l	Fe electrodes; Electrode distance: 2 cm; 2 A; treatment time 90 min; pH 6	99	-	Gatsios et al. (2015)
0.5 l	Fe electrodes; Electrode distance: 2 cm; 10 mA/cm^2; treatment time: 60 min; pH 7	50	-	Xu et al. (2017)
0.5 l	Fe electrodes; Electrode distance: 2 cm; 15 mA/cm^2; treatment time: 120 min; pH 7; addition of electrolyte: 25 mmol/l SO_4^{2-}	85.5	-	Xu et al. (2018)

3.5. Ammonia removal

Ammonia in wastewater can originate from many sources such as fertilizer manufacturing, food processing, landfill leachate, agriculture, slaughterhouses and tanneries. Ammonia is considered as one of the most toxigenic contaminants, and high ammonia concentrations can cause eutrophication of rivers and lakes, thus disrupting the ecological balance. Till now, the main ammonia removal processes involved: air stripping, biological nitrification, denitrification, chemical treatment and selective ion exchange method (Desai et al. 2016). However, these methods are limited because of their cost, low efficiency and the use of toxic chemicals (Aoudj et al. 2017). Therefore, EC seems as an interesting solution for ammonia removal, and some results are shown in **Table 6**. According to results, EC is not effective for ammonia removal. The removal efficiency is less than 50 % in all the mentioned research. Since it has been shown that EC is not effective enough to meet the standards determined by the Ordinance on conformity parameters, analytical methods, monitoring and drinking water safety plans, and keeping register of legal entities which provide public water supply (Official Gazette 125/17), it is suggested that EC needs to be combined with other water treatment processes.

Table 6. Recent applications of EC for ammonia removal

VOLUME TREATED	OPTIMAL OPERATIVE PARAMETERS	% REMOVAL	OPERATING COSTS [€/m^3]	REFERENCE
1.8 l	Al electrodes; 3 A; treatment time 60 min; pH 8	24	-	Son et al. (2017)
2 l	Al electrodes; Electrode distance: 2 cm; 1.5 A; 15 V; treatment time: 90 min	47	-	Desai et al. (2016)
90 l	Al electrodes; 150 A/m^2; treatment time: 120 min; pH 7	36	1.95	Lončar et al. (2019)

3.6. Color removal

Colored water is not suitable nor for drinking nor for many industrial purposes such as food industry or cloth washing. There are two types of color in water, true and apparent color. True color is the result of soluble substances that cannot be isolated by filtration, and apparent color is the result of suspended solids and colloid particles that can be separated by filtration (Malakootian and Fatehizadeh 2010). It can be noticed that EC has high color removal efficiency (97 %), but the choice of optimal operative parameters differs (**Table 7**). Therefore, more research on color removal by EC needs to be done.

Table 7. Recent applications of EC for color removal

VOLUME TREATED	OPTIMAL OPERATIVE PARAMETERS	% REMOVAL	OPERATING COSTS [€/m^3]	REFERENCE
1.5 l	Al electrodes; 300 A/m^2; treatment time 120 min; pH 5.2	97.2	-	Kara et al. (2013)
3 l	Fe electrodes; Electrode distance: 5 cm; 2.07 mA/cm^2; treatment time: 45 min; pH 7.6	97	0.26	Khansorthong and Hunsom (2016)

3.7. Turbidity removal

Water turbidity is caused by suspended solids and colloidal particles of clay, sludge, fine organic matter, microorganisms and other. The precipitation of particles depends on their density and size. Particles with higher density precipitate due to gravity and smaller particles, especially ones whose density is similar as water density, such as bacteria and colloidal particles, don't precipitate, but remain suspended in water and need to generate larger flocs. Conventional treatments for turbidity removal have several disadvantages, such as the use of large amounts of chemicals and generating large amounts of sludge which causes disposal problems and the loss of water (Gulić 2003). It has been shown that EC is good for removing water turbidity (more than 95 %) and some results of previous research are shown in **Table 8**. According to previous research, optimal operative parameters very much differ. Since all mentioned types of the electrode material have high removal efficiency, their price can be a deciding factor. Suggested optimal operative parameters are Al electrodes, 2 cm of electrode distance, 20 V voltage and 40 min of treatment time.

Table 8. Recent applications of EC for turbidity removal

VOLUME TREATED	OPTIMAL OPERATIVE PARAMETERS	% REMOVAL	OPERATING COSTS [€/m^3]	REFERENCE
2 l	Fe electrodes; Electrode distance: 5 cm; 5.62 mA/cm^2; treatment time 40 min; pH 5.2	95	-	Ni'am et al. (2007)
3 l	Al electrodes; Electrode distance: 2 cm; 20 V; treatment time: 10 min	97	-	Rahmani (2008)
1 l	SS electrodes; Electrode distance: 2 cm; 20 V; treatment time 180 min; pH 7	98	-	Adapureddy and Goel (2012)
3 l	Al electrodes; Electrode distance: 2 cm; 30 V; treatment time 25 min; pH 7	95	0.12	Moosavirad (2017)

4. ECONOMY ANALYSIS AND THE COMPARISON OF REMOVAL EFFICIENCY

Economy analysis, along with the removal efficiency analysis, plays an important role in the selection of optimal water treatment. The economic factors that can influence this choice are chemicals, coagulation resins, membranes, electricity, work, maintenance, etc. Only few papers analyzed the cost of EC, and most of the experiments were conducted on small batch units (Vepsäläinen 2012). Therefore, only rough estimation and approximate cost comparison of EC and similar water treatments can be given. Operative costs of some previous research are shown in **Tables 3-8**, and cost and efficiency comparisons between EC and several water treatments are given in **Figures 3-6**.

The first comparison is for turbidity and **Figure 3** shows that EC has higher removal efficiency than the combination of ultrafiltration/coagulation by 10 %. Also, operative costs of EC are lower than for the combination of ultrafiltration/coagulation by 0.20 €/m^3 (**Figure 4**). Therefore, it is justified to use EC for removal of turbidity.

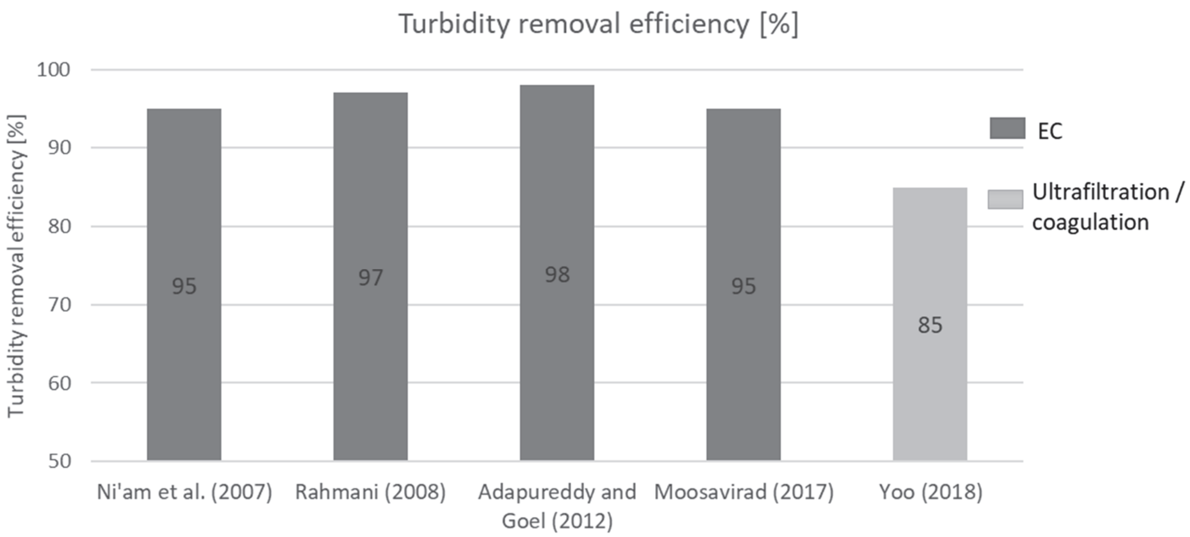

Figure 3. *Comparison of turbidity removal efficiency*

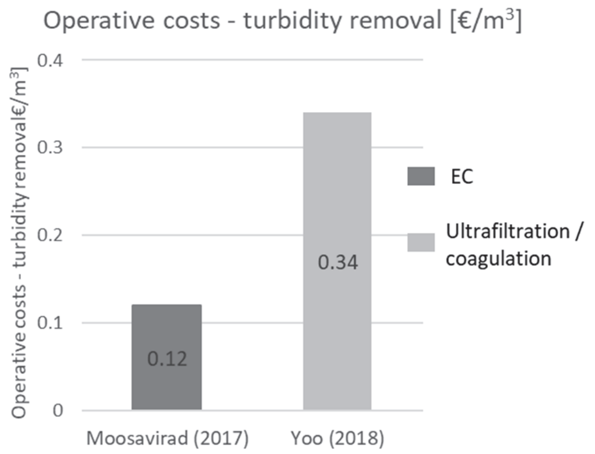

Figure 4. *Comparison of operative costs for turbidity removal*

The second comparison is for arsenic removal efficiency. Reverse osmosis and EC process both have very high removal efficiencies and similar operative costs (**Figure 5** and **Figure 6**). But since more research was carried out on it, the advantage is given to reverse osmosis. It is noted that all studies were conducted on units with similar capacity.

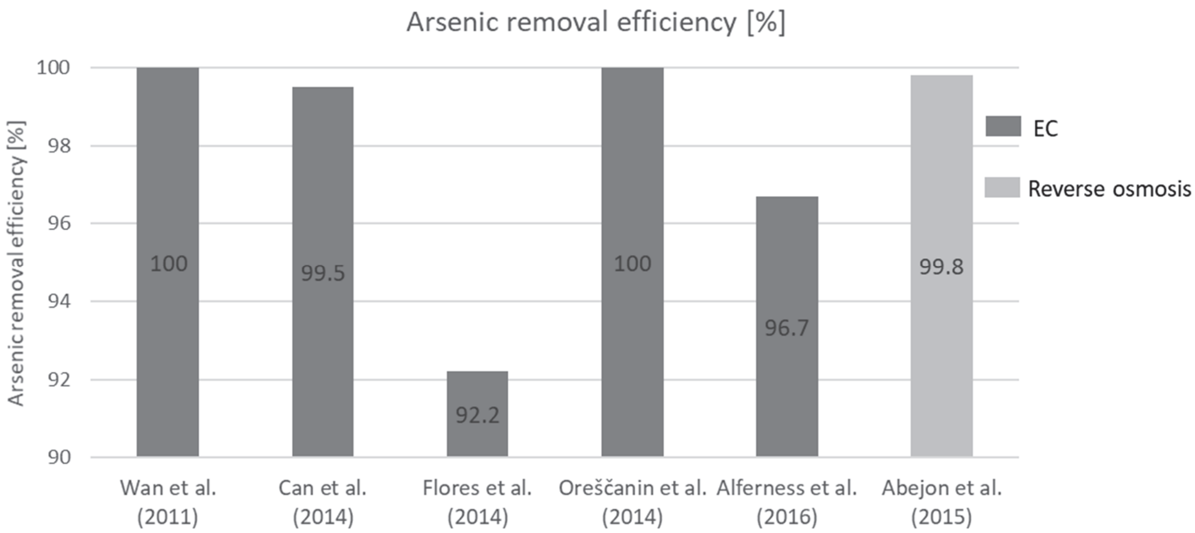

Figure 5. *Comparison of arsenic removal efficiency*

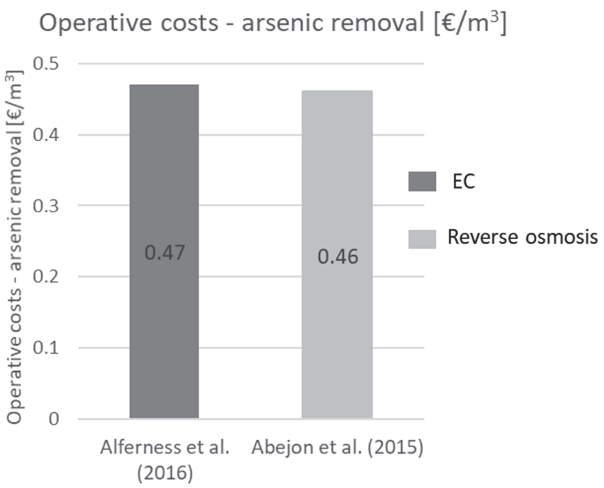

Figure 6. *Comparison of operative costs for arsenic removal*

4.1. Removal of Total Nitrogen on a full-scale electrocoagulation reactor

A case study with Al electrodes was performed on a full-scale batch EC unit made from stainless steel, **Figure 7**. The unit has two rectangular chambers (tanks), whose dimensions are 0.80 m × 0.55 m × 1.10 m. In the study, 90 L of water was used as the operating volume. The first tank is used for EC process, from which water can circulate (by pump) between two rectangular Fe, Al or SS electrode plates, while the second one is used as a settling tank. The total surface of Al electrodes was 0.063 m^2 and the electrode distance was 0.5 cm.

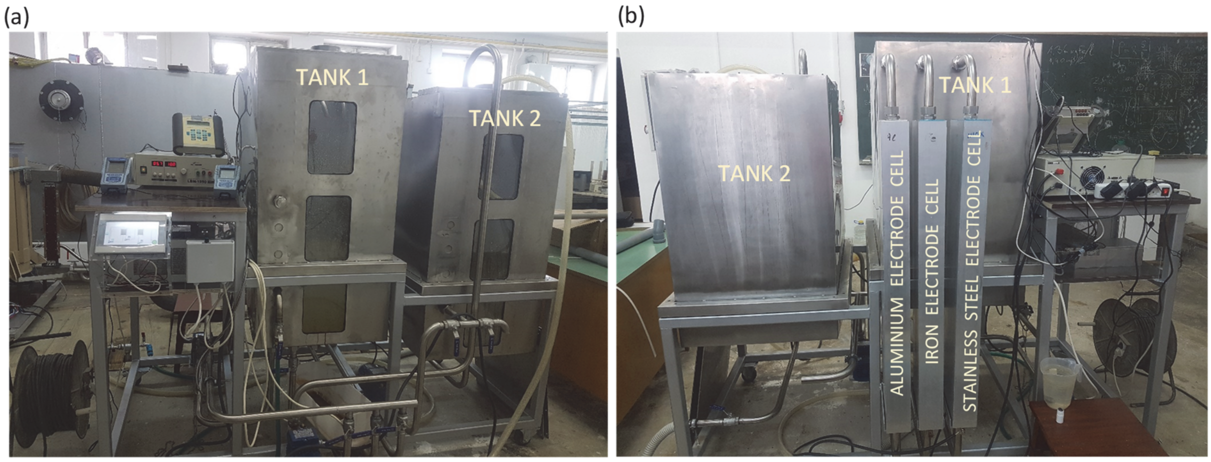

Figure 7. *(a) The front and (b) the back of the EC unit*

In the tank 1 (**Figure 7**), 100 mL of 25 % NH_3 solution was mixed with 90 L of drinking water from public water supply system in order to obtain Total Nitrogen (TN) concentration just over 200 mg/L. Also, 180 g of NaCl was added in order to increase the solution conductivity and obtain its concentration around 2 g/L. Everything was mixed for several minutes. The initial pH was 9.8 and total treatment time was 240 min. Water samples were taken before the beginning of the treatment, at every 60 min, and at the end of the process. TN concentrations were measured with NANOCOLOR 500D (by Eutech) Test 0-88 (TNb 220). Flow (Q = 0.03 L/s) was measured with ultrasonic water meter FLUXUS F601 (by Flexim), and the current was maintained approximately constant at 12 A by the MC Power LBN-1990 lab power supply. After 240 min, TN was decreased to 63 mg/L, corresponding to removal efficiency of 69.7 % (**Figure 8, Table 9**).

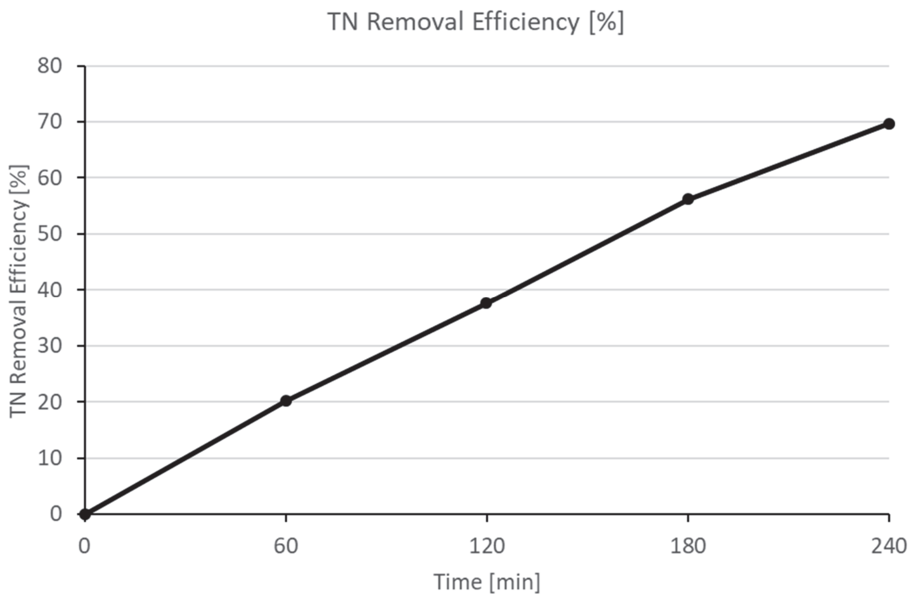

Figure 8. *TN removal efficiency*

According to **Figure 8**, a linear trend of TN removal can be noticed, approximately 33 mg/L per hour. In comparison with some previous results (Posavčić et al., 2018), it is assumed that flow significantly affects the removal efficiency and is better that the water runs slower through the electrodes because there is more time for the formation of Al hydroxides.

Further, the operating costs are determined for this case (**Table 9**). An assessment of the operational costs of the EC process is given regarding energy cost, consisting of electricity and pump (power) costs, and material (electrodes) cost according to **Equation 3**:

$$\text{Operating costs} = a\ (C_{electricity} + C_{pump}) + \ b\ C_{material} \qquad (3)$$

where: $C_{electricty}$ is electrical energy cost of 1 m^3 of treated water (kWh/m^3), C_{pump} is the energy cost of the pump for 1 m^3 of treated water (kWh/m^3) and $C_{material}$ (kg Al/m^3) is the cost of the electrode material used in 1 m^3 of the treated water. *a* is the average electricity price of 0.13 €/kWh (according to the national tariff models), and *b* is the average market price of aluminum given as 1.54 €/kg. After 240 min, total operational costs were 7.60 €/m^3.

Table 9. TN removal efficiency (%) and operating costs for energy (electricity and pump) and material (electrode)

TIME [min]	TN [mg/L]	% REMOVAL	$C_{electricity}$ [kWh/m^3]	C_{pump} [kWh/m^3]	$C_{electrode}$ [kg/m^3]	OPERATING COSTS [€/m^3]
0	208	-	-	-	-	-
60	166	20.19	8.00	5.56	0.09	1.90
120	130	37.50	16.00	11.11	0.18	3.80
180	91	56.25	24.00	16.67	0.27	5.70
240	63	69.70	32.00	22.22	0.36	7.60

5. CONCLUSION

According to previous research, EC is suitable for removal of *E. coli*, iron, arsenic, color and turbidity. However, in cases with manganese and ammonia, it has been shown that EC is not effective enough to meet the standards determined by the Ordinance on conformity parameters, analytical methods, monitoring and drinking water safety plans, and keeping register of legal entities which provide public water supply (Official Gazette 125/17) and needs to be combined with other water treatment processes.

Generally, most of the previous research were conducted on "small-scale" units, i.e. small capacity devices (up to 10 L), where received results (operative costs and operative parameters), are not applicable in real conditions. Therefore, in order to obtain more credible results, more research on pilot devices need to be done.

In this paper, a case study is also presented. Observed linear change indicates that the EC process for TN removal, with specified reactor setup, can be modelled with a simple linear rule with the average removal rate of 33 mg/L·h for 0.03 L/s flow rate.

Linear change can be attributed to the approximately constant current that was maintained at 12 A. Also, what was not previously reported is that during the study the current was constantly slowly decreasing. Approximately every 15 minutes current dropped for 1 A. In order to keep the current constant, the voltage was increased accordingly.

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