APPLICATION OF ELECTROCOAGULATION FOR DISINFECTION AND TREATMENT OF WATER CONTAMINATED WITH ARSENIC AND CHROMIUM

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Abstract: *Local water supply systems pose the greatest risk from a public health perspective because water is delivered to consumers without any treatment and often even without disinfection. The most common causes of contamination are microbiological contamination and elevated concentrations of heavy metals. Such water must be treated to be safe for human consumption. The most common water disinfection process used in drinking water treatment is disinfection with chlorine-based compounds. However, the by-products of the reaction of chlorine and organic matter are chlorinated hydrocarbons and nitrosamines, which can be extremely carcinogenic. The accumulation of these by-products along with the accumulation of heavy metals in the human body increases the risk of adverse effects on human health. Electrocoagulation (EC) is an alternative method of raw water treatment. Electrocoagulation uses an electric field and sacrificial electrodes to generate coagulants that separate, aggregate, and precipitate contaminants from the water. This method has proven successful in removing heavy metals from water, but also as a disinfection method. The objective of this study was to use EC to remove arsenic and chromium and to disinfect raw water enriched with Escherichia coli. The process took place in a batch reactor with 2 electrodes (1 anode and 1 cathode) at a voltage of 62 V. The efficiency of the process as a function of electrode material (Fe, Al, Cu and graphite) and treatment time was studied. With the exception of graphite, all electrodes showed 100% efficiency in the removal of As and Cr, while the disinfection efficiency varied.*
Keywords: *water treatment, electrocoagulation, heavy metals, disinfection*

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1. INTRODUCTION

According to *The report on the health suitability of water for human consumption in the Republic of Croatia for 2020* by the Croatian Institute of Public Health, there is still a significant percentage of local water supply users in Croatia. Local water supply consists of small water distribution networks, which are mainly taken care of by groups of citizens or local communities. The supply of water to the population through local waterworks does not comply with the regulations in the field of water management and represents a risk from a public health point of view, because water is delivered to consumers without any treatment, even without disinfection. Contaminated drinking water can be the source of trace metal intake and pathogen infections (Daud et al. 2017). Consumption of undisinfected water can cause various waterborne diseases like diarrhea, cholera, typhoid, or, in extreme cases, even death (Hashim et al. 2020; Martínez-Huitle & Brillas 2008; Zhou et al. 2020). Coliform microorganisms, including *Escherichia coli,* are most common contaminants and can be used as an appropriate indicator of water quality (Solmaz et al. 2018). While heavy metals in trace concentrations are necessary for normal metabolism, growth and development of the organisms, exposure to high concentrations can have adverse effects on human health (Yu et al. 2021). For example, chromium has been linked to allergic dermatitis, nausea, vomiting and alopecia, while arsenic can cause acute poisoning, fatigue, dermatitis and circulation issues. Both are systematic toxins and carcinogens (Carolin et al. 2017; Joseph et al. 2019; Sharma & Bhattacharya 2017; Shrestha et al. 2021). To ensure safe human consumption, drinking water from local water supplies should be treated before distribution.

Two of the most important purposes of disinfection is removal or inactivation of microorganisms in raw water supply and ensuring the residual effect in the distribution network (Martínez-Huitle & Brillas 2008). Chlorination is widely used disinfection method based on the introduction of chlorine gas, sodium hypochlorite or calcium hypochlorite into the treated water. Generated hypochlorous acid and hypochlorite ions oxidize bacteria and viruses present in the water (Licht et al. 2021; Pichel et al. 2019). Chlorine treatment is highly effective in inactivating microorganisms and provides residual protection against microbial recontamination in the distribution system (Sadiq & Rodriguez, 2004). However, the biggest downside of this treatment is the formation of disinfection by-products, some of which are considered carcinogenic and mutagenic (Naddeo et al., 2014; Rook 1974). Other disinfection methods such as UV or ozone disinfection, while effective, do not provide residual protection.

The electrocoagulation process (EC) could be a promising alternative for both disinfection and removal of heavy metals from drinking water. This electrochemical process is based on the "in situ" production of coagulants by passing electric current through sacrificial electrodes (Posavčić et al. 2019). When the current is passed through a metal electrode, metal cations are released from the anode. At the same time, water is reduced to hydrogen and hydroxide ions at the cathode (Kabdaşlı et al. 2012). As a result, metal hydroxides are formed, leading to destabilisation of colloidal particles and formation of flocs that can be easily removed. Several mechanisms of microorganism inactivation have been proposed for electrochemical disinfection: oxidative stress, destruction of the cell wall by electrochemically generated oxidants, permeabilization of cell membranes by the applied electric current, electrochemical oxidation of vital cell components, and electrosorption of negatively charged microorganisms on the anode surface (Bubalo et al. 2020; Ghernaout et al. 2019a). In addition to the direct effect of the electric field and the electrochemical formation of oxidant species, the EC disinfection process is also characterised by the physical elimination of microorganisms via sweep coagulation and the entrapment of microorganisms in the formed flocs and their settling (Ghernaout et al. 2019b). Some of the advantages of the electrocoagulation method include simultaneous removal of multiple pollutants, simplicity, easy automation and integration with other treatments, no need for added chemicals and reduced sludge generation (Alattabi et al. 2017; Betancor-Abreu et al. 2019; Cataldo Hernández et al. 2012; Doggaz et al. 2019; Hashim et al. 2017; Myllymäki et al. 2018; Yazici Guvenc et al. 2019).

In this study, the efficiency of the EC process for the bacterial inactivation and removal of arsenic and chromium from raw water was investigated using various electrode material combination.

2. MATERIALS AND METHODS

The experimental setup of the EC cell is shown in **Figure 1**. The experiments were performed in a Plexiglas batch reactor with a magnetic stirrer placed underneath. The total volume of the EC cell was 3 litres and the volume of treated water (solution) for each run was 2.5 litres. Two plate electrodes were placed in the cell and connected to the laboratory power supply. The active surface area or total immersed surface area of each electrode was 119 cm^2 and the distance between the electrodes was 1 cm. The electrodes were made of iron, aluminium, copper and graphite, and different anode-cathode combinations were tested. All experiments were performed at constant temperature and constant applied voltage. The pH of the treated water was not changed, and no additional electrolytes were added. As a model solution, raw surface water from a nearby lake was used and spiked with arsenic and chromium standard solution. Between each run, the electrodes were disinfected using 70% ethanol solution and thoroughly rinsed with distilled water.

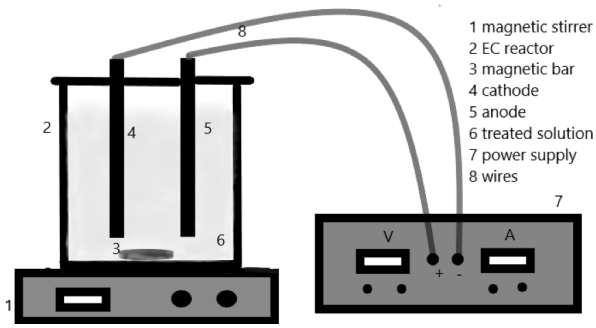

Figure 1. Experimental setup of EC process

2.1. Disinfection experiments

The disinfection experiments were performed using raw water rich in coliform bacteria with CFU (Colony Forming Units) in the range of 10^4 to 10^5. In this study, we focused on *E. coli* and total coliforms. Two parameters were studied: the applied voltage and the electrode material used. Four pairs of electrodes with cathode and anode made of the same material (Al-Al, Cu-Cu, Fe-Fe, C-C) were tested at two voltage levels of 25 and 62 V. The total treatment time was 15 minutes, and samples were taken at the beginning of the experiment, after three minutes, after 8 minutes, and at the end of each run. An automated microbiological analyser TECTA B4 from Pathogen Detection Systems was used for microbial analysis and quantification of bacteria present in the samples (**Figure 2**). Appropriate cartridges for E. coli and total coliforms were used for sample collection and analysis. The operation of this device is based on the reaction of specific enzymes, characteristic for each bacterial species, with the substrate in the cartridge, resulting in fluorescent compounds. The intensity of the fluorescence is proportional to the number of bacteria in the sample. Results are obtained in less than 24 hours and are expressed as CFU per 100 ml.

Figure 2. TECTA B4

2.2. Heavy metals removal experiments

For the heavy metal removal experiments, water from the same lake as used in the disinfection experiments was used and spiked with arsenic and chromium solutions. The resulting initial concentrations of arsenic and chromium were 0.15 and 0.7 ppm, respectively. The experiments were conducted at a maximum voltage of 62 V because higher efficiencies were obtained in the disinfection experiments when a higher voltage was applied. To better understand the effects of electrode material on the efficiency of the heavy metal removal process, different combinations of anode and cathode were used. Graphite anodes were not used as they are nonmetal and ineffective for heavy metal removal. The total treatment time was 3 minutes. Samples were taken at the beginning and the end of every run and analysed by using the ICP-OES technique.

3. RESULTS AND DISCUSSION

3.1. Efficiency of the EC process for bacterial inactivation

The experimental results presented in **Table 1** show that the EC is an effective disinfection treatment and removes over 99% of bacteria in 8 minutes at an applied voltage of 62 V for all electrode pairs. Results for both E. coli and total coliforms showed similar trends and efficiencies. Although the efficiency of the process decreased with decreasing voltage, especially in the case of the copper electrodes, over 90% of the bacteria were removed at the end of the process. In accordance to some other studies (Cotillas et al. 2020; Zarei et al. 2018), it can be concluded that, under the same experimental conditions, a higher potential difference and longer treatment time lead to better disinfection results.

Table 1. The efficiencies (%) of the bacterial inactivation depending on the electrode material, treatment time and applied voltage

	25 V				62 V				
t/min	**Al/Al**	**Fe/Fe**	**Cu/Cu**	**C/C**	**Al/Al**	**Fe/Fe**	**Cu/Cu**	**C/C**	
3	87.42	56.22	48.56	40.12	92.81	99.53	99.99	97.66	***E. coli***
8	98.02	70.14	86.41	95.61	99.76	99.99	100	99.95	
15	99.66	94.45	95.41	99.56	99.88	100	100	99.98	
3	93.62	77.32	3.54	42.59	86.84	99.71	99.91	97.97	**Total coliforms**
8	99.38	89.58	67.39	95.40	99.08	99.97	99.99	99.95	
15	99.55	96.62	97.11	99.54	99.70	100	100	99.98	

Complete inactivation of *E. coli* was achieved with iron electrodes after 15 minutes and with copper electrodes after 8 minutes. At both voltage levels, the tendency was observed for most bacteria to die in the first three minutes, after which the mortality rate decreases. Log kill values for *E. coli* as a function of treatment time are shown in **Figures 3** and **4**. For bacteria, common removal values for chlorine and ozone based disinfection range from 2 to 6 log, while UV processes typically result in 2 to 4 log (Bicudo et al. 2021). These values are comparable to those achieved by EC disinfection, suggesting that EC is a promising alternative to conventional water disinfection processes. Among other advantages of electrochemical disinfection, such as simplicity and effectiveness, it should be noted that there is no risk of microorganisms developing resistance to the effects of the electric current (Feng et al. 2004). However, to better understand and optimize the EC disinfection process, further research is needed on the inactivation mechanism of microorganisms at the subcellular level and on the characterization of the electrogenerated oxidants that cause cell inactivation (Martínez-Huitle & Brillas, 2021).

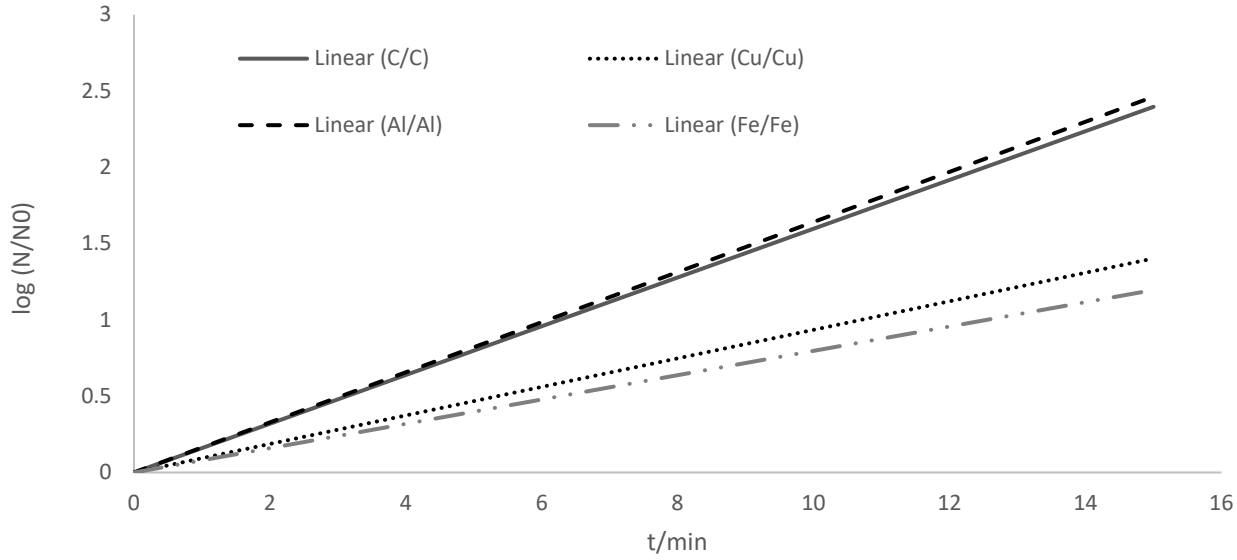

Figure 3. E. coli log kill values for the lower current density experiments

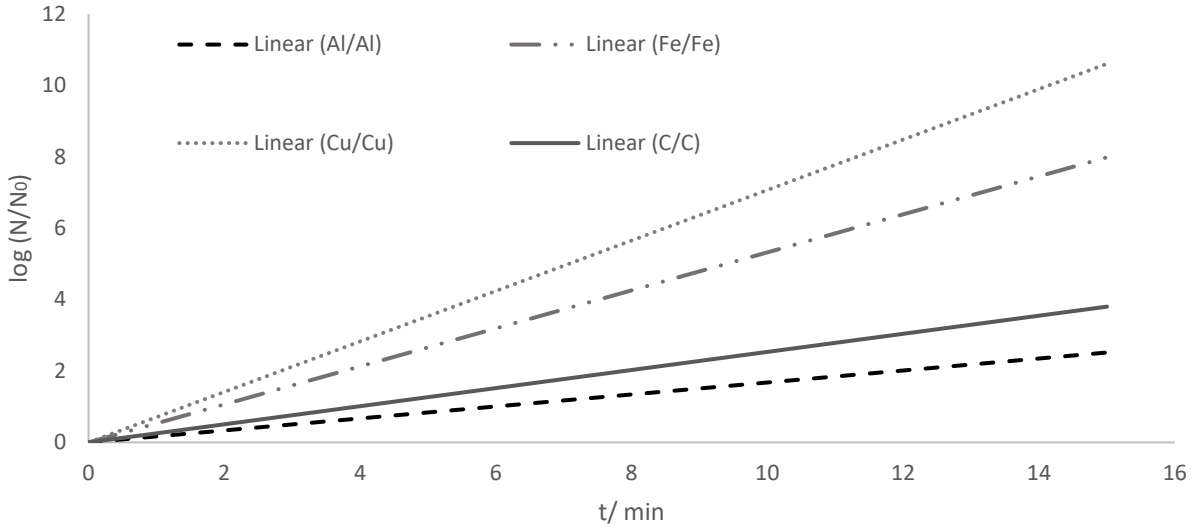

Figure 4. E. coli log kill values for the higher current density experiments

3.1. Efficiency of the EC process for As and Cr removal

Since the disinfection effect of our process was better at higher voltage, experiments for the removal of As and Cr were performed only at 62 V. To investigate the influence of the electrode material, we used different anode and cathode combinations. As graphite electrodes do not produce cations and are consequently ineffective for the removal of heavy metal ions, they were used only as cathodes. In the total treatment time of 3 minutes, chromium

was almost completely or totally removed regardless of the electrode material used. As shown in **Figure 5**, the efficiency of arsenic removal is slightly lower, especially when aluminium anodes are used. The residual concentrations of arsenic and chromium in water after treatment are listed in **Table 2**, and it can be seen that they are far below the maximum concentrations (50 μg/L for Cr and 10 μg/L for As) specified in the Drinking Water Ordinance of the Republic of Croatia.

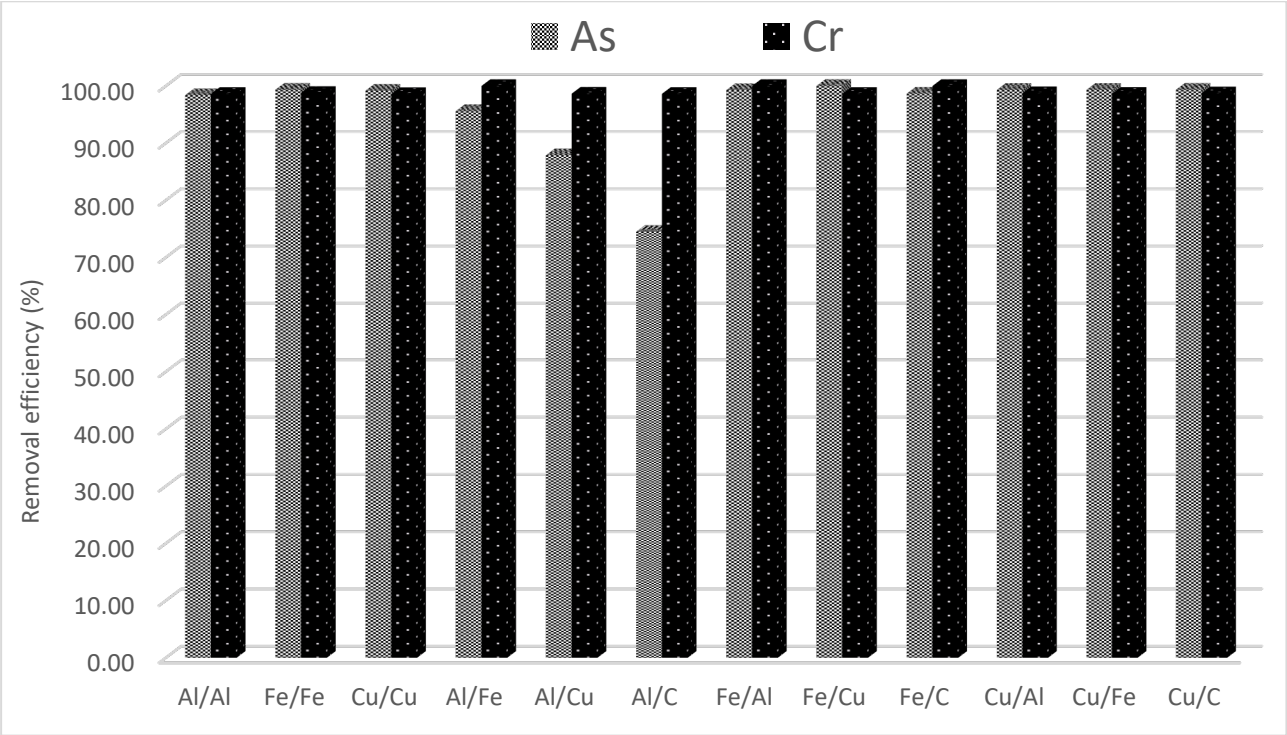

Figure 5. Efficiencies of chromium and arsenic removal after 3 minutes of treatment

Table 2. Residual concentrations of arsenic and chromium in the water after the treatment

Anode	Cathode	As	Cr
Al	Al	0.002	0.001
Fe	Fe	0.001	0.001
Cu	Cu	0.001	0.001
Al	Fe	0.006	0
Al	Cu	0.017	0.001
Al	C	0.036	0.001
Fe	Al	0.001	0
Fe	Cu	0	0.001
Fe	C	0.002	0
Cu	Al	0.001	0.001
Cu	Fe	0.001	0.001
Cu	C	0.001	0.001

Although the process was effective in removing As and Cr, it was interesting to see how effective it would be in removing ions of a heavy metal that occurs as a potential impurity in electrode materials, such as manganese in iron electrodes. Experiments were conducted using the method described for the removal of As and Cr. The initial concentration of manganese in the treated water was about 0.07 ppm. The change in manganese concentration at the end of the process depending on the electrode material is shown in **Figure 6**. As expected, in the case of iron anodes manganese concentration increases after the treatment. The largest increase in manganese concentration is observed for the combination of iron anode and copper cathode. These results show that great attention must be

paid to the selection of electrodes for drinking water treatment, their composition and possible impurities that could enter the water.

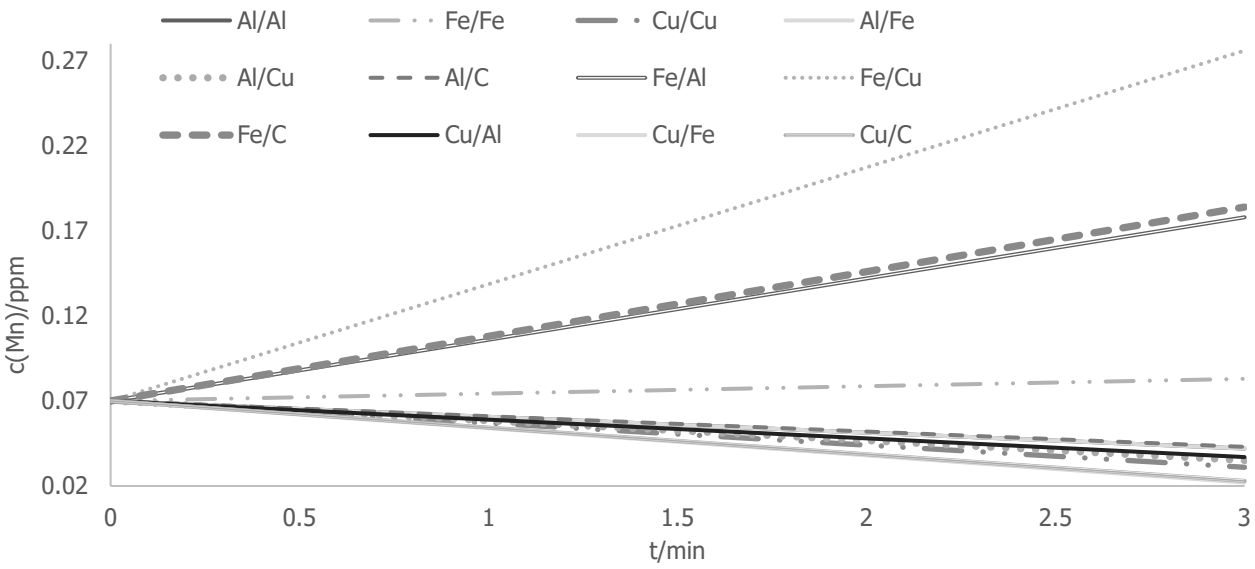

Figure 6. The change in manganese concentration during the EC process with different electrodes

4. CONCLUSION

This work has shown that electrocoagulation, unconventional chemical-addition free technology, is an effective method for the treatment of microbiologically and chemically contaminated water. Copper electrodes were found to be most effective for disinfection, as it took 8 minutes to completely inactivate E. coli. In general, the results showed that removal of E. coli and total coliforms was more efficient at higher current densities and longer treatment time. Similar results were obtained for the removal of heavy metals. Both arsenic and chromium were successfully removed by electrocoagulation, and the concentrations in the effluent were mostly below the regulatory maximum concentrations after 3 minutes of treatment. In addition, the purity of the electrode material can have a significant impact on the results, but also on the cost of the process itself. In general, there is a need for further research, especially on a large scale, and it is worth exploring the possibility of combining electrocoagulation with other water treatment technologies.

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Authors contribution: Licht – investigation, writing – original draft; Halkijević – project administration, writing – review&editing; Posavčić – validation; Nakić - supervision

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