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Electrochemical/granular Activated Carbon Hybrid System for Drinking Water Disinfection at Flow Conditions

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The electrochemical disinfection has gained much attention due to its simplicity, environmental sustainability and effective in-situ free chlorine generation. Since chloride present at low concentration in tap water, it has become possible to use electrolyzed tap water for disinfection. In the developing countries, the disinfection of tap water is a necessity to reduce the risks of the growth and transmission of pathogenic microorganisms in drinking water distribution systems. In this study, the electrochemical disinfection device EDI-001 equipped with titanium oxide (TiO_{2-x}) ceramic electrodes combined with granular activated carbon (GAC) columns was examined for disinfection of drinking water at flow conditions. The GAC columns were used for removing the chlorine residuals from tap water to reach health authority standards. The results have shown that common microorganisms from tap water was completely disinfected within 15 minutes and total chlorine concentration level was decreased over 50% by using electrochemical/granular activated carbon hybrid system.

1. Introduction

Around 2.1 billion people world-wide do not have access to microbiologically safe drinking water, and about 500 000 people die of diarrhoea each year (WHO, 2018). In hospitals tap water is considered as one of the sources for secondary infections (Nakajima et al., 2004). Drinking water disinfection or inactivation of pathogenic microorganisms is an essential step to minimize infection risks and decrease the incidence of waterborne diseases such as diarrhea, dysentery, cholera, typhoid and polio (Cabral et al., 2010). The ability of these organisms to demonstrate the resistance to antimicrobial agents after chlorination (Shi et al., 2013) and the ability of some of them to reproduce and grow in water distribution systems (Szewzyk et al., 2000) have resulted in the need to upgrade the technologies and find more novel approaches.

Chlorine-based disinfection as one of the most widely applied technologies still faces several minuses like, unpleasant taste and odour, ineffectiveness against resistant microorganisms, such as *Giardia* and *Cryptosporidium* (Razzolini et al., 2016), and potential formation of hazardous disinfection by-products, such as trihalomethanes and haloacetic acids, when chlorine ions react with natural organic compounds (Bongiovani et al., 2015; Xue et al., 2017).

In recent years, electrochemical disinfection has been regarded as one of the alternatives to traditional chlorination due to its high effectiveness and environmentally sustainability - it can be performed without the use of chlorine chemicals. The inactivation efficiency of electrochemical disinfection system is dependent on many parameters, such as electrode material, cell configuration, electrolyte composition, current density, electrolyte flow rate and type of water to be disinfected (Kerwick et al., 2005). Recently, many different electrode materials, such as Pt, Pt-Nb, IrO_2, boron doped diamond (BDD), Si/BDD, Ti/RuO_2, have been applied to the electrochemical disinfection process (Martinez-Huilte and Brillas, 2008). Previous studies have demonstrated that electrochemical disinfection by direct electrolysis is effective in killing a wide spectrum of microorganisms from bacteria (Monasterio et al., 2014) to viruses and algae (Drees et al., 2003; Mascia et al., 2013), even the bacterial spores (Mezule et al., 2014). However, the overall design of the system must be simple in use and rapid in production of disinfected water.

Currently a wide range of commercial/development stage systems have been described and tested (Kraft, 2008; Martinez-Huilte and Brillas, 2008; Denisova et al., 2017), however, there is no such system that

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simultaneously complies with high effectivity, rapid treatment (flow conditions), simplicity and economic sustainability.

In this study an automated disinfection device EDI-001 applicable for flow condition and equipped with titanium oxide (TiO_{2-x}) based ceramic electrodes was used for determination of drinking water electrochemical disinfection efficacy. Titanium oxide-based ceramic electrodes TiO_{2-x} have been selected due to their high resistance to corrosion and low absorption coefficient, and they present good efficiencies in the production of active disinfectant from low chloride concentration solutions (Reimanis et al., 2011). In the effort to neutralise residual chlorine concentration and to reach health authority standards, an additional two granular activated carbon (GAC) columns were added after the electrochemical disinfection system.

2. Materials and methods

2.1 Electrochemical/granular activated carbon hybrid system configuration

The electrochemical disinfection was performed at flow conditions in a specially made electrolytic system EDI-001 (Riga, Latvia) containing the rectangular cell of 1.2 L volume and equipped with 24 titanium oxide (TiO_{2-x}) ceramic electrodes with an area 12.1 cm^2 (total area: 290.4 cm^2). The EDI-001 device was combined with two GAC filter columns to achieve consumer acceptable drinking water quality parameters. The experimental electrochemical/ granular activated carbon hybrid system used in this study is schematically shown in the Figure 1. In order to investigate the effect of total chlorine concentration removal efficiency, water samples were taken: (a) before and (b) after GAC filter columns.

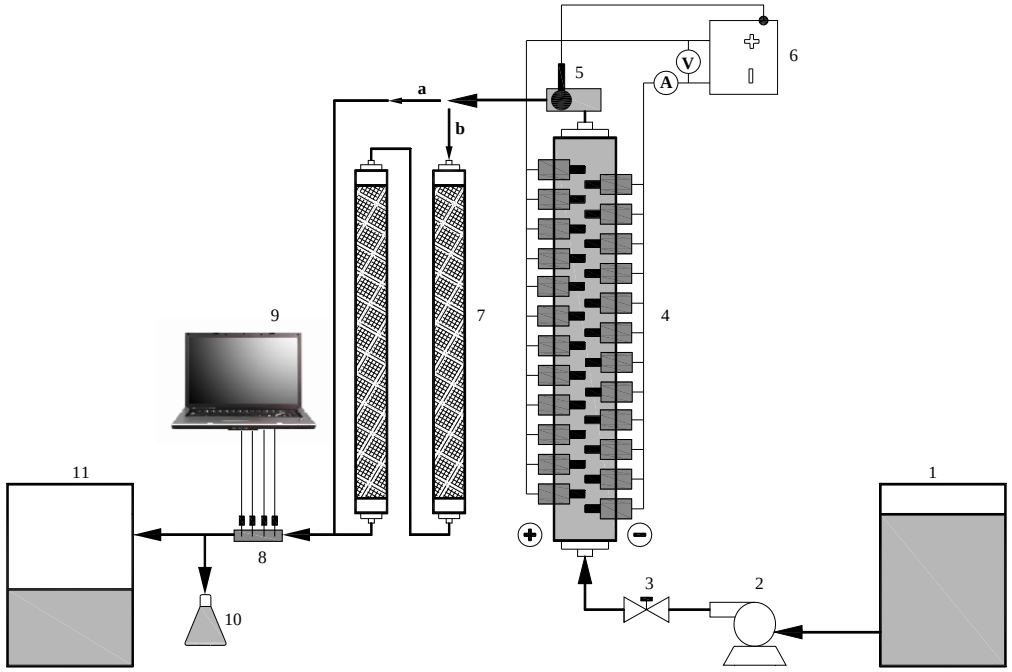

Figure 1: The schematic diagram of experimental setup: 1 – contaminated drinking water; 2 – peristaltic pump; 3 – pressure regulator (max - 1.5 atm); 4 – electrochemical cell; 5 - total chlorine sensor; 6 – control unit; 7 - GAC packed bed columns; 8 – measuring cell; 9 – PC; 10 – samples collection in flow; 11 – samples collection in storage tank. Tap water electrochemical disinfection experiments applied: (a) without GAC columns and (b) with two GAC columns.

2.2 Granular activated carbon

A 0.4 – 1.4 mm mesh-sized GAC delivered from bituminous coal (Chemviron) was used for the studies with hybrid system. For drinking water electrochemical disinfection experiments at flow conditions, 460 g of GAC were packed in two PVC columns of 35.7 cm length and 5.2 cm of inner diameter. Two stainless sieves with a mesh size of 220 μm were placed at the top and the bottom of the PVC column in order to avoid the washing out of filter material. Before each experiment, the GAC filter material was washed with sterile distilled water using continuous flow to remove brown-coloured water.

2.3 Microorganisms and water quality parameters

This study was focused on tap water bacteria which are commonly found in drinking water distribution system and are more resistant to minimal residual chlorine concentration (Shi et al., 2013). The hybrid system was connected to public drinking water distribution system in the city of Riga, Latvia with water quality parameters complying with all local and national regulations (Table 1.) (Rigas udens, 2017).

Table 1: The drinking water quality parameters in Riga, Latvia.

Parameter	Value
pH	7.6
Turbidity	0.3 NTU
Conductivity	0.28 mS/ cm
Chloride	8 – 100 mg/L Cl^-
Sodium	4 – 84 mg/L
Iron	0.06 mg/L
Hardness	1.6 – 2.2 mmol/L

2.4 Disinfection tests

Before each experiment, EDI-001 was pre-treated with sodium hypochlorite solution (2 mg/L as free chlorine) for 10 minutes, and then washed several times with sterile distilled water until the free chlorine concentration in the water fell to zero. The disinfection experiments were carried out using tap water of Riga city.
The tap water with a known concentration of natural bacteria (1-4 x 10^4 CFU/mL) was pumped through the disinfection system (30.9 A/m^2, 25.1 V) and two PVC columns packed with GAC filter material. Total chlorine concentration was set at 1 mg/L Cl^-. All experiments were performed at 300 mL/min, at room temperature (20°C ± 2°C) for 15 minutes. Samples were collected before electrolysis (0h) and after 5 min, 10 min and 15 minutes of treatment before and after GAC. Immediately after sampling, residual chlorine was neutralized with equal or excess volume of 0.02 M sodium thiosulfate ($Na_2S_2O_3$).
Conductivity, pH, temperature and redox potential (ORP) was measured online using the AMR WinControl online monitoring system (ALMEMO® 5690-1M, conductivity, pH, temperature and redox potential sensor). All data were transferred to the personal computer immediately. The total chlorine concentrations in the water was measured online by CP2.1MA10 chlorine sensor where values are displayed on display control unit. Additionally, the concentration of total chlorine species was determined using N, N-diethyl-p-phenylenediamine (DPD) colorimetric method, according to the detailed procedure described by LVS EN ISO 7393-1:2001.

2.5 Bacterial sample analysis

To evaluate the disinfection efficiency, the number of viable tap water bacterial cells inactivation was determined by the plate count technique: 0.1 mL aliquot of the sample, its decimal dilution or membrane filter was inoculated on R2A agar plate (Oxoid Ltd., UK) and incubated for 5 days at 25 ˚C. Detection limit was set to 10 CFU/L. Each sample was plated in triplicates. The results were expressed as colony forming units (CFU) per milliliter. Log reduction was calculated using the following equation (Eq.1):

$$\text{Log reduction} = \log\left(\frac{N}{N_0}\right) \quad (1)$$

Where N_0 is the initial concentration of cultivable microorganisms from public tap water distribution system, N is the concentration of cultivable microorganisms from public tap water distribution system after electrochemical disinfection at time t.

3. Results and discussion

The efficiency of tap water disinfection with electrochemical/granular activated carbon hybrid system at low current density (30.9 A/m^2) under flow conditions was investigated. In addition to disinfection tests, pH, temperature, conductivity and RedOx changes of tap water were constantly monitored. During the treatment process no significant changes of water parameters was detected. The average pH values before and after treatment were 7.6 ± 0.1, temperature - 20.8 ± 1.4 °C, conductivity - 0.27 ± 0.01 µS/cm and RedOx potential values were 80 ± 34 mV.
The analyses of log reduction in cultivable microorganisms treated with EDI-001 device showed that during the first 5 minutes of electrochemical disinfection a mere 0.4-log reduction of tap water bacteria was observed (Figure 2). However, further 10-15 minutes of treatment demonstrated more than 2-4 log decrease in

cultivable tap water bacteria cell, indicating on formation of active disinfectant. In the previous studies (Denisova et al., 2017), contaminated pre-filtered tap water with suspensions of *E. coli* (10^4 CFU/mL) were completely disinfected (more than 5-log reduction) by EDI-001 device after 10 minutes of treatment. This difference between disinfection efficiency of indigenous microorganisms and faecal indicator organism can be explained by various survival capabilities and adaptation to unfavourable environmental conditions.

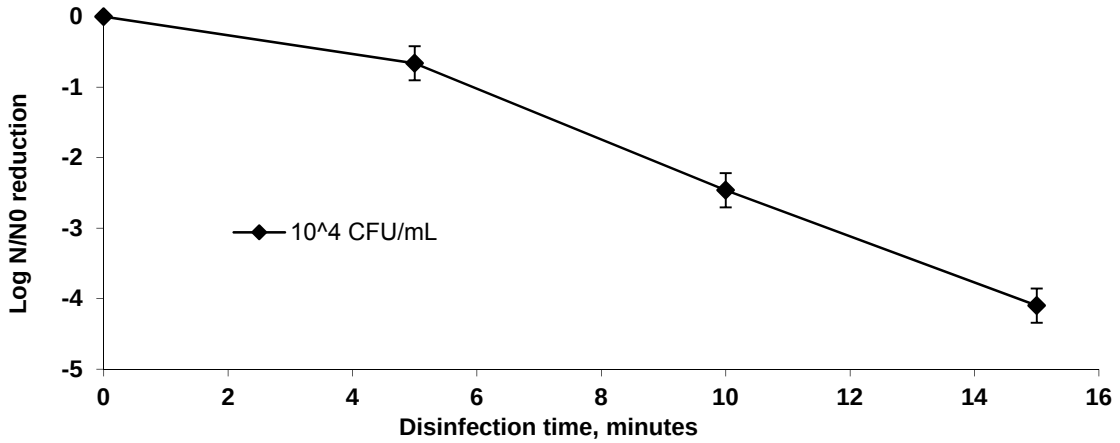

Figure 2. The effect of electrochemical disinfection (30.9 A/m^2) to reduce the number of cultivable microorganisms commonly present in tap water distribution system. The data represent the average values of three experiments.

About 60% of the cultivable bacteria present in the tap water was removed during the first 5 minutes of electrochemical disinfection (Figure 2) when the highest the total chlorine concentration was detected. At the same time effective removal needs at least 10 – 15 min which is more than reported before (Denisova et al., 2017). As shown in Figure 3, the total chlorine concentration was reached rapidly by electrochemical hybrid system, within first 5 minutes of operation the total chlorine concentration was 1.36 ± 0.2 mg/L Cl^-. In an effort to reach health authority standards according to the recommended free chlorine residual of 0.2-0.5 mg/L in drinking water distribution systems (WHO, 2008; Madzivhandila and Chirwa, 2017), two GAC filter columns were added after EDI-001 (Figure 1). The effect of potential electrochemical generation of total chlorine and its removal efficiency by GAC columns during electrochemical disinfection at constant current density (30.9 A/m^2), was obtained at set the total chlorine production of 1 mg/L Cl^-.

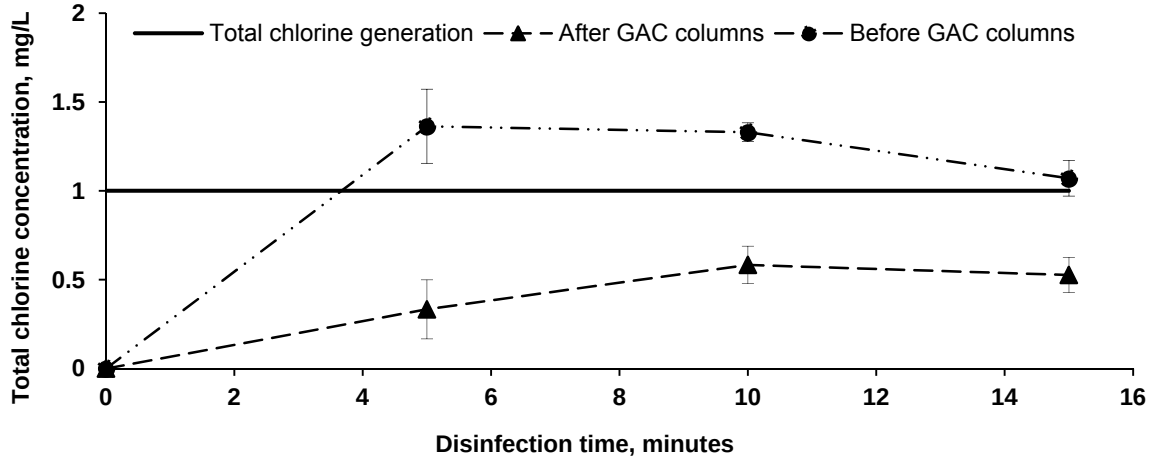

Figure 3. The potential electrochemical generation of total chlorine by EDI-001 device and its removal by two granular activated carbon (GAC) columns during electrochemical disinfection. Total chlorine concentration was set at 1 mg/L Cl. The data represent the average values of three experiments.

As expected, the total chlorine concentration decreased after two GAC filter columns (Figure 3). The obtained results showed that GAC columns can be used in the drinking water treatment to remove high chlorine concentration after electrochemical disinfection. During the electrochemical disinfection, the total chlorine concentration level was decreased over 50% by using electrochemical/granular activated carbon hybrid system. After 5 minutes of operation before the GAC columns, the total chlorine concentration was 1.36 ± 0.2 mg/L Cl^-, but after GAC filters – the total chlorine concentration decreased to 0.33 ± 0.2 mg/L Cl^-. Further the generation of total chlorine species was decreased by EDI-001 from 1.36 ± 0.2 mg/L Cl^- to 1.33 ± 0.1 mg/L Cl^- and to 1.07 ± 0.1 mg/L Cl^- before the GAC after 10 and 15 minutes of treatment, respectively. After GAC filter columns, the total chlorine concentration decreased from 1.33 ± 0.1 mg/L Cl^- to 0.58 ± 0.1 mg/L Cl^- and from 1.07 ± 0.1 mg/L Cl^- to 0.53 ± 0.1 mg/L Cl^- after 10 and 15 minutes of treatment, respectively.
The obtained results showed that the health authority standards can be reached with electrochemical/granular activated carbon hybrid system even at flow condition. However, as reported by Vacca et al. (2011) the real flow pattern in drinking water distribution systems are not under ideal conditions. Normally in water distribution systems presents dead or stagnant zones, bypass flows, and recirculation zones that can affect the efficiency of the electrochemical drinking water disinfection. Further research is needed in order to investigate the potential single-pass flow under non-ideal conditions (Abokifa et al., 2016; Vacca et al., 2011). At the same time, the results showed that bacteria cells adsorbed on GAC surface have a tendency for survival. As the result, attached bacteria cells can accumulate and growth on GAC (Nishijima et al., 1997). Thus, other potential filter materials like ceramic (Man et al., 2018) or polythersulfone/titania nanocomposite membrane (Rahmati et al., 2017) should be evaluated.

4. Conclusions

The efficiency of electrochemical/granular activated carbon hybrid system for the disinfection of indigenous water microorganisms was demonstrated. The complete disinfection (4 log reduction) of contaminated tap water was achieved within 15 minutes by using the EDI-001. GAC filter columns resulted in reduction of the total chlorine concentration from 1.36 ± 0.21 to 0.53 ± 0.10 mg/L Cl^-. Based on the experimental results, it can be concluded that the electrochemical/granular activated carbon hybrid system can be applied for disinfection purposes. However, further investigations are needed to improve GAC filter material properties for the clean and safe treatment of drinking water.

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