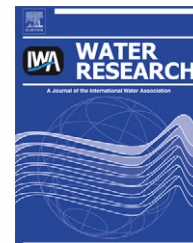




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Reductive electrochemical remediation of emerging and regulated disinfection byproducts

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ABSTRACT

Long-term exposure to low concentrations of disinfection byproducts (DBPs) in drinking water has been associated with increased human-health risks of bladder cancer and adverse reproductive outcomes. In this study, we investigated electrochemical reduction utilizing a resin-impregnated graphite cathode for the degradation of 17 DBPs (i.e. halo-methanes, haloacetonitriles, halopropanones, chloral hydrate and trichloronitromethane) at low $\mu\text{g L}^{-1}$ concentration levels. The reduction experiments were potentiostatically controlled at cathode potentials -700 , -800 and -900 mV vs Standard Hydrogen Electrode (SHE) during 24 h. At the lowest potential applied (i.e. -900 mV vs SHE), the disappearance of DBPs from the solution after 24 h of reduction was $>70\%$, except for chloroform (32%), 1,1-dichloropropanone (48%), and chloral hydrate (31%). Due to the participation of several removal mechanisms (e.g. electrochemical reduction, adsorption, volatilization and/or hydrolysis) it was not possible to distinguish the removal efficiencies of electrochemical reduction of individual compounds. Adsorption of the more hydrophilic DBPs (i.e. haloacetonitriles, chloral hydrate, and 1,1-dichloropropanone) onto the electrode seems to be affected by the cathode polarization, as the removals observed in the open circuit experiments were significantly higher than the ones obtained in electrochemical reduction under the same conditions. The overall efficiency of reduction was estimated based on the analyses of the released Cl^- , Br^- and I^- ions. Nearly complete C–I bond cleavage was achieved at all three potentials applied, and from the theoretically predicted release of I^- ions, calculated based on the removed DBPs, 86 ± 9 to $92 \pm 1\%$ was measured in the catholyte solution at -700 to -900 mV vs SHE. Debromination efficiencies obtained were 74 ± 3 , 79 ± 6 and $68 \pm 4\%$ at -700 , -800 and -900 mV vs SHE, while for C–Cl bond cleavage the obtained values were 69 ± 1 , 72 ± 1 and $76 \pm 4\%$, respectively. Nevertheless, dechlorination efficiencies are to be considered as approximate, since an increase in Cl^- concentration was observed in the open circuit experiments due to the hydrolysis of some of the chlorine-containing DBPs. Although the Coulombic efficiencies for DBPs dehalogenation were only 1.9 ± 0.3 (-900 mV vs SHE) $-4.1 \pm 0.2\%$ (-700 mV vs SHE), relatively low energy consumption of the process was observed, estimated at 72 ± 2 Wh m^{-3} at -900 mV vs SHE for the concentration range of DBPs in this study (i.e. 65.3 – 129.7 $\mu\text{g L}^{-1}$). The study demonstrated that reductive electrochemical treatment has the potential to be a modern remediation technology for the removal of low concentrations of halogenated DBPs in water.

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1. Introduction

The widespread treatment and distribution of public drinking water is one of the greatest achievements of the twentieth century. However, formation of potentially hazardous disinfection byproducts (DBPs) by the reaction of disinfectants with natural organic matter (NOM) and/or halide ions present in source water is an undesired side effect of any disinfection process. More than 600 DBPs have been identified by now, yet only a small number of them are regulated for the quality of drinking water. Currently, the focus of scientific community is on the so-called emerging DBPs – among them nitrogen-containing DBPs as well as brominated and iodinated DBPs, which are suspected to be more toxic, carcinogenic and mutagenic to humans than their chlorinated analogs (Komaki et al., 2009). For example, among trihalomethanes (THMs) iodinated THMs are of special concern, as they were reported to be highly cytotoxic in mammalian cells (Richardson et al., 2008). As potable water treatment is the last line of defence against contamination of water prior to consumption, it is necessary to ensure that the finished water has very low levels of DBPs and any other contaminants hazardous to humans.

There are several strategies for DBP control in drinking water treatment, including: i) removal of organic precursors, ii) minimization of chlorine contact time, iii) use of alternative disinfectants (e.g. chloramines), and iv) removal of formed DBPs. Many water utilities opt to move the point of chlorination further downstream in the treatment process or use chloramines instead of free chlorine. However, this may lead to an incomplete microbial inactivation of finished water (Ahuja, 2009), as well as the formation of other, unknown hazardous byproducts. Moreover, switching from chlorine to chloramines can have other unexpected consequences such as the release of lead in pipes of the water distribution system (Liu et al., 2009; Sedlak and von Gunten, 2011). Strategies aiming at removal of DBP precursors such as enhanced coagulation, adsorption, ion exchange or advanced oxidation are expensive and have variable removal efficiencies. Moreover, oxidative processes can generate a series of unknown intermediates (Sedlak and von Gunten, 2011; Karanfil et al., 2008). Removal of the formed DBPs by air stripping is inefficient for non-volatile DBPs, while activated carbon treatment has high environmental footprint and short effective life of carbon. Many waterworks employ conventional treatment (coagulation, sedimentation, filtration and chlorine disinfection) that is ineffective for the removal of both DBP precursors and formed DBPs (Chang et al., 2001).

In considering alternative approaches for DBP control, researchers have reported the degradation of haloacetic acids (HAAs) and THMs on a laboratory scale by employing different reducing agents such as sodium sulphite (Croue and Reckhow, 1989) and zero-valent iron (ZVI) (Hozalski et al., 2001). In recent years there has been a revived interest in environmental applications of electrochemical processes, which are chemical-free, electricity driven and easily scalable to a full-scale continuous treatment. For example, Sonoyama et al. (1997, 2003) reported an efficient electrochemical dehalogenation of several regulated THMs using metal electrodes, while Korshin and Jensen (2001) demonstrated the reduction

of chloro- and bromo-HAAs at copper and gold electrodes. Nevertheless, little is known about the performance of electrochemical reduction of other DBPs in dilute aqueous solutions. Electrochemical reduction is a low-cost process which proceeds at ambient temperature and pressure, and does not generate secondary pollution. It could be easily applied as a continuous treatment using a flow-through reactor. Furthermore, DBPs present at low concentrations could be efficiently decomposed using electrodes with a large surface area (e.g. granular electrodes). The compact reactor size, minimized formation of byproducts and low maintenance requirements of the electrochemical reduction process make it highly attractive as point of use drinking water treatment.

In this study, we used a resin-impregnated graphite cathode to reduce electrochemically 17 DBPs, among which were regulated THMs and non-regulated iodine-containing THMs (I-THMs), haloacetonitriles (HANs), chloropropanones and others. Table 1 summarizes the physico-chemical properties of the investigated DBPs, including their octanol-water partition coefficients ($\log K_{OW}$), Henry constants (K_H) and hydrolysis constants (k_{HYD}). The resin-impregnated graphite cathode used is characterized by its low cost, mechanical stability, reasonably high overpotential for hydrogen evolution and high surface area that enhances the adsorption of DBPs and thus their subsequent reduction (Schmal et al., 1986). Furthermore, it can be deployed in various configurations and unlike metals that can corrode, graphite is inert. Previously, graphite electrodes have been investigated for electrochemical reduction of chlorinated contaminants such as pentachlorophenol (Szebenyi-Gori et al., 1999) and trichloroethylene (Al-Abed and Fang, 2006; Fang and Al-Abed, 2007). The main objective of this study was to demonstrate the feasibility of electrochemical reduction on a graphite cathode for dehalogenation of a number of regulated and emerging DBPs, of which several had not been studied previously in any similar process.

2. Materials and methods

2.1. Chemicals

Analytical standards for chloroform (TCM), bromodichloromethane (BDCM), dibromochloromethane (DBCM) and bromoform (TBM) were purchased from Sigma Aldrich as THMs calibration mix of $100 \mu\text{g mL}^{-1}$ each in methanol. Dichloroacetonitrile (DCAN), trichloroacetonitrile (TCAN), bromochloroacetonitrile (BCAN), dibromoacetonitrile (DBAN), 1,1-dichloropropanone (1,1-DCP), 1,1,1-trichloropropanone (1,1,1-TCP), and trichloronitromethane (TCNM) were purchased from Sigma Aldrich as EPA 551B Halogenated Volatiles Mix at $2000 \mu\text{g mL}^{-1}$ each in acetone. Chloral hydrate (CH) was purchased as individual standard to Sigma Aldrich at $1000 \mu\text{g mL}^{-1}$ in acetonitrile. Dichloriodomethane (DCIM), chlorodiiodomethane (CDIM), bromochloriodomethane (BCIM), dibromiodomethane (DBIM) and bromodiiodomethane (BDIM) were purchased as pure compounds from Orchid Cellmark (Canada). Analytical grade methyl tert-butyl ether (MtBE), 99.9% was purchased from Sigma Aldrich as

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