Oxidation and volatilization of bromide by electrolysis in drinking water

David Eugene Kimbrough, Lina Boulos and Sirikarn Surawanvijit

ABSTRACT

To reduce the concentrations of brominated disinfection by-products, a process is presented here which removes bromide from a widely used surface water source, the California State Water Project (SWP). The process consists of oxidizing bromide to bromine and volatilizing the bromine. SWP water was passed through this unit under various conditions and the bromide was oxidized and volatilized under a variety of conditions. Five different reactor bodies with seventeen different configurations were tested. The reactors differed in the depth of the anode, in the distance between the anodes and, in the surface area of the anodes. Each reactor had SWP water pumped through the reactor at three or more different flows and at four or more different currents, producing 267 experimental conditions. Both reaction rates and removal efficiency increased with increasing current and were generally higher in the shallower reactors. The highest reaction rates were observed in the shallowest reactors and highest flow rates but the greatest efficiency was achieved is in a slightly deeper reactor at lower flows. This appears to have been the effect of the shallowest reactor having the smallest surface area that was easily saturated and but being closest to atmosphere, allowing the most rapid volatilization of bromine.

Key words | bromide, disinfection by-products, drinking water, electrolysis, haloacetic acids, trihalomethanes

David Eugene Kimbrough (corresponding author)
Castaic Lake Water Agency,
27234 Bouquet Canyon Road,
Santa Clarita, CA 91350-2173, USA
E-mail: *dkimbrough@clwa.org*
Present address: City of Pasadena
Water and Power Department,
150 S. Los Robles, Suite 200,
Pasadena, CA 91101,
USA
E-mail: *dkimbrough@cityofpasadena.net*

Lina Boulos
L Boulos Consulting Inc,
26653 Oak Terrace Place,
Valencia, CA 91381,
USA

Sirikarn Surawanvijit
University of California,
School of Engineering,
Department of Chemical Engineering,
5531 Boelter Hall,
Los Angeles, CA 90095-1592,
USA

INTRODUCTION

A key process for making water potable is disinfection, which has historically meant the addition of oxidizing chemicals such as chlorine or ozone. While this process has revolutionized public health protection from pathogenic organisms, these chemical oxidants can give rise to disinfection by-products (DBPs) such as bromate, trihalomethanes (THMs) and haloacetic acids (HAAs) (Morris *et al.* 1992; Pegram *et al.* 1997; Klotz & Pyrch 1998; Kargalioglu *et al.* 2002; Plewa *et al.* 2002, 2003). Chlorine reacts with natural organic matter to form chlorinated DBPs. If bromide is present in the water, then brominated DBPs will be also formed; under the right conditions they will be the dominant species. Brominated DBPs are thought to pose a greater threat to public health than species without bromine (Kargalioglu *et al.* 2002). As Kargalioglu *et al.* note:

'quantitative structure function analysis of the brominated vs. the chlorinated analogs of the haloacetic acids showed that brominated acetic acids had a higher mutagenic potency.'

Lowering the concentration of bromide in source water could greatly reduce the concentrations of DPBs in general and the brominated species in particular. However, there is currently no known technology that can lower the concentration of bromide in drinking water in a cost-effective manner.

Recently, however, a new technology has emerged which has offered the potential of changing this situation. Electrolysis of water has been shown to oxidize bromide to bromine and water to oxygen gas and hydrogen ions, resulting in the volatilization of bromine and the decrease

of the concentration of bromide. This in turn has been shown to reduce the concentration of brominated DBPs (Kimbrough & Suffet 2002, 2006; Kimbrough 2007; Boulos *et al.* 2008). While it has been shown that this process is capable of lowering the concentration of both bromide and DBPs, it remains an open question whether it can be applied in a practical and cost-effective manner to drinking water treatment. Toward this end, a research project to make such a determination was developed and funded collaboratively by the Water Research Foundation and the Castaic Lake Water Agency (Tailored Collaboration 4216). This paper presents some of the results of this study as they apply to the design of a practical electrolytic reactor.

OXIDATION AND VOLATILIZATION BY ELECTROLYSIS

During the electrolysis of water, there are two separate but parallel oxidation reactions that take place; one is the oxidation of bromide to bromine and the other is the oxidation of water into hydrogen ions and oxygen gas.
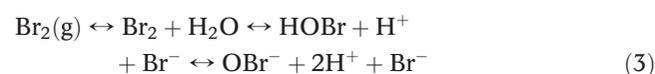
$$2Br^- \leftrightarrow Br_2 + 2e^- \tag{1}$$

($E_0 = 1.09$ volts) (all standard potentials are versus a saturated hydrogen electrode (SHE) from *CRC Handbook*, Weast 1978).

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

($E_0 = 1.23$ v)

If bromine in water is hydrolysed, it can exist in one of three states: molecular bromine (Br_2), hypobromous acid (HOBr) or as hypobromite ion (OBr^-).

$$Br_2(g) \leftrightarrow Br_2 + H_2O \leftrightarrow HOBr + H^+ \\ + Br^- \leftrightarrow OBr^- + 2H^+ + Br^- \tag{3}$$

Chloride is also oxidized to chlorine which reacts to form bromine chloride (BrCl); however, this is usually a minor component (Margerum & Huff Hartz 2002). The equilibrium constant for the hydrolysis of molecular bromine at 25 °C has been measured to be between 5.2 and 7.2×10^{-9} M^{-2} (Radford 1966; Downs & Adams 1973) at pH 7 (pKa = 8.7) (Radford 1966). The hydrolysis constant is highly pH dependent; low pH is more favourable to Br_2 (Downs & Adams 1973) which dominates at pH less than 3.5 (Johnson & Sun 1975). This is fortunate as the pH is quite low around an anode during electrolysis of water owing to the release of hydrogen ions (Boulos *et al.* 2008). In addition, since the Henry's Law constant for bromine (7.6×10^{-1} M/atm [Dean 1992]) is quite a bit more favourable to volatilization than hypobromous acid (6.1×10^3 M/atm [Frenzel *et al.* 1998]), the concomitant release of large quantities of very fine oxygen bubbles during electrolysis allows this potential for volatilization to be realized.

The oxidation of bromide to bromine is expected to follow the equation electrolytic reaction rates,

$$i = nFkc_{Br^-} \tag{4}$$

where i is the current in A, n is number of electrons transferred, F is the Faraday constant (96,485 C/mol), k is the rate constant, and c_{Br^-} is the concentration of bromide. Since the same equation would apply to the production of oxygen gas and hydrogen ions, the combined reaction rate for both oxidation and volatilization should be proportional to the applied current.

EXPERIMENTAL SECTION

Analytical techniques

1. Bromide ions were measured by ion chromatography (DIONEX DX-500) using USEPA Method 300.0 (USEPA 1993). This method cannot measure bromine, hypobromous acid or hypobromite.
2. Bromine (Br_2, HOBr or OBr^-) was measured by collecting electrolysed samples in duplicate and adding excess sodium thiosulfate (STS) to one of the samples. The STS reduced any bromine, hypobromous acid, bromine chloride or hypobromite that was present to bromide. The bromide was measured in both samples by ion chromatography (above) which can measure bromide but not bromine, so the difference between the bromide with and

without STS was the bromine. Samples for bromide and chloride were collected in 100 ml polypropylene bottles.

3. THMs were analysed using gas chromatography combined with a mass spectrometer (Shimadzu GC-17A and QP5000) using USEPA Method 524.2 (USEPA 1995). Samples were collected in zero-headspace vials preserved with STS.

4. THM formation potential (THMFP) was measured by collecting a sample from the effluent of the reactor and in a 1-l amber glass bottle, adding sufficient ascorbic acid to reduce all measurable free available chlorine (FAC) formed during electrolysis. Then the bottle was sampled for THMs and sufficient sodium hypochlorite was added to produce a concentration of 20 mg l^{-1} FAC. The bottle and sample were then incubated in the dark at room temperature for seven days. A sample from this bottle was then collected and analysed for THMs as described above.

Water source

For most of these experiments, water from the California State Water Project (SWP) was used. The SWP is a system of dams, conveyances and pumping stations spanning 1,000 km (600 miles) and supplies drinking water to 20 million Californians. The SWP has four termini, one of which is Castaic Lake in northern Los Angeles County, in southern California. The water delivered by the SWP has historically had high bromide concentrations (100 to 400 $\mu g\,l^{-1}$), high TOC concentrations (2 to 9 mg l^{-1}), and a potential to form high concentrations of DBPs, particularly of the brominated species. The experiments described in this paper were conducted between August and December 2009 when the bromide concentrations in Castaic Lake were between 260 and 280 $\mu g\,l^{-1}$ and the TOC was between 1.55 and 2.5 mg l^{-1}.

Reactors

Five electrolytic up-flow reactor bodies were constructed in a clear acrylic plastic body and polyvinyl chloride (PVC) base. All five had a square shape with an internal cross-sectional dimension of 10 cm by 10 cm. Water entered the reactor through an orifice on the side of the base and passed through plenum beneath the reactor. The water then moves up

through a diffusion plate (PVC) with 16 orifices equally spaced across the cross-sectional area. Each orifice had an inverted funnel shape; a 0.08 cm length was very narrow with parallel walls and the remainder opened up into a wider 'V' shaped area with the walls at 90° from each other. Each reactor had a different sized orifice, 0.02, 0.03, 0.05 and 0.07 cm as measured at the narrow, parallel walled portion of the orifice. A hydraulic model indicated that the diffusion plate created sufficient mixing to produce approximately plug flow within the plenum below the anodes.

The water then flowed up between two 316 l stainless steel plate cathodes mounted on opposite walls of the reactors. Between these cathodes, anode plates were arrayed in parallel to each other but perpendicular to the cathodes (Figure 1). The titanium anode plates were coated with a proprietary coating of RuO_2 and TiO_2 (DE NORA TECH, Chardon, Ohio) (Grotheer 1998). A titanium bus rod with similar coatings was placed on top of the anodes and held in place by holes in the top of the reactor body. The cathodes and bus rod were connected to an Agilent 6674 A DC power source (Hewlett-Packard, Palo Alto, California). The five different reactor bodies used differed in the depth of the walls and electrodes. The walls and electrodes were 1.2 cm, 2 cm, 5 cm or 10 cm deep from the diffusion plate to the bottom of the V-notch weir.

Four of the reactors held a maximum of 57 anodes at 1 mm apart from each other. The anodes were removable so the number of anodes and the distance between them was adjustable. One reactor (#17) did not have anode plates but rather a single 10 cm × 10 cm sheet of titanium mesh 0.06 cm thick. The mesh sheet was laid in a reactor body of 1 cm depth. The mesh was laid flat with the edges parallel to the cathodes so the water would flow up through a depth of 0.06 cm. Around the top of all reactors was a weir with 'V' notches which allowed the water to flow evenly out of the reactor with minimal time into a collection trough which brought all of the effluent together into a common exit.

Experimental design

Each of the four reactor bodies with plate anodes was arranged into four different configurations with either 57 anodes 0.1 cm apart, 29 anodes 0.2 cm apart, 12 anodes 0.5 cm, or six anodes 1.0 cm apart, which along with the
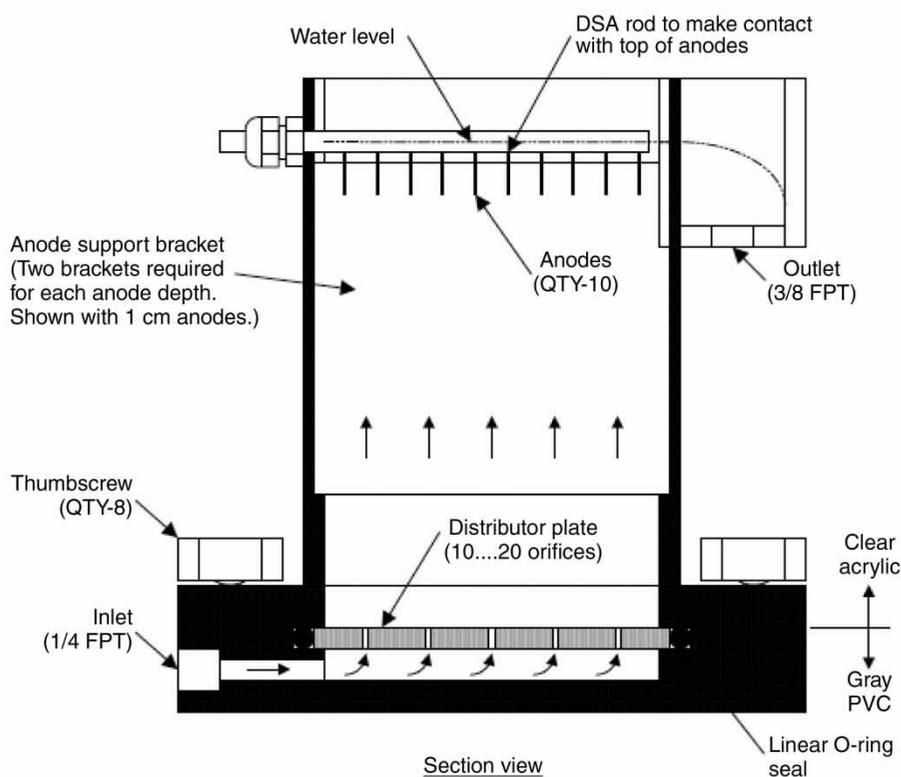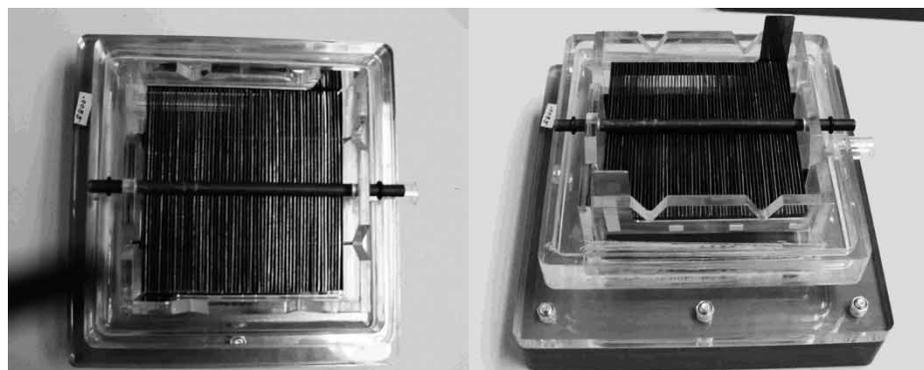
Figure 1 | 1.2 cm deep reactor.

mesh reactor, produced 17 different reactor configurations (reactor 1 is shown in Figure 1). Each of these configurations had Castaic Lake water flowing at several different flow/velocity/hydraulic detection time (HDT) conditions and each flow condition had several different applied currents producing 267 different experimental conditions. For each experiment condition, the amount of bromide, bromine and FAC was measured. The amount of power consumed was calculated by multiplying the current by the potential by the HDT and was reported in units of watt-minutes

(W-m). Additionally, after the results from these 17 reactors was examined, an 18th 'hybrid' reactor was assembled using the shallowest reactor body, with 29 1-cm deep anodes, 2 mm apart, with the same mesh as used in the #17 reactor placed on top of plates. SWP water was fed into this hybrid reactor at 95 ml min^{-1} with a contact time of 0.9 min with applied currents ranging from 0 to 8 A.

Bromide removal was measured by collecting the unelectrolysed influent and adding STS and collecting the electrolysed effluent and adding STS; the difference was the

Table 1 │ Trihalomethane formation potential results from reactor 3

Current (A)	Potential (V)	Cl$_2$ mg l^{-1}	Br$^-$ (µg l^{-1})	Br removal (%)	THM initial (µg l^{-1})	THM final (µg l^{-1})	THM drop (%)	THMFP drop (%)
0	0	0	250	0	29	276	0	0
0.1	4.5	0.3	220	12	25	253	8	8
0.2	6.7	1.1	190	24	37	253	12	8
0.5	11	1.6	150	40	30	227	18	20
1.0	17	4.0	140	44	32	204	26	30

amount removed. However, it is possible the bromine had been incorporated into either organic DBPs such as THMs or bromate. In order to test this hypothesis reactor 3 was used (1.2 cm deep, 12 anodes, 5 mm gaps) to determine whether the THM formation potential was reduced by the process of electrolysis. If the bromide had been removed, the THM formation potential should be reduced proportionally. A fixed flow of 100 ml min^{-1} was fed into reactor 3 and a current of either 0, 0.1, 0.2, 0.5 or 1.0 A was applied. The bromide, bromine and chlorine concentrations were determined. Samples were collected for THMs at the effluent for an instantaneous determination and THMFP sample was also collected as described above (see Table 1).

RESULTS AND DISCUSSION

Table S1 (see supplementary online material: http://www.iwaponline.com/jws/060/0041.pdf) shows all of the results for each of the 272 experimental conditions. Table S2 (see supplementary online material: http://www.iwaponline.com/jws/060/0041.pdf) provides operational variables for all experimental conditions such as anodic surface area, effective volumes, hydraulic retention times and current density. Table S1 has the results from the THMFP experiment with reactor 3. The results shown on Table S1 indicate with successively higher currents, up to 44% removal. In parallel the THMFP also declined in proportion to the current applied and the amount of bromide removed, up to 33%. In previous research it has been shown that this electrolytic process does not form bromate (Boulos *et al.* 2008); therefore, it can be concluded that the bromide is being volatilized and not merely removed from solution by incorporation into organic DBPs.

Figures 2–5 present the curves that were generated for each of the 17 reactor configurations grouped according to the depth of the reactor with the different flow conditions combined plotting the percentage removal versus the applied current. As can be seen, for each reactor depth, there was a general pattern of increasing bromide removal as the current is increased, although there is a wide range of removal at any given current. However all of the reactors are more or less in the same range; there is little, if any, difference between reactors of the same depth with different distances or gaps between the anodes and the number of anodes at a given current. For example in Figure 2, the four 1.2 cm plate reactors (1–4) all removed 38% to 55% when 1 A of current is applied. As can be seen in Figure 6, most of that variability in percentage removal is due to differences in flow. For example, all plate reactors had removals ranging from 42 to 54% when the flow was 100 ml per minute and the current was 1 A.

Similar patterns can been in Figures 3–5. In Figure 3 for example, there appear to be three parallel lines, each with a tightly grouped set of results from each of the four reactor conditions (i.e. different gaps). These parallel lines represent different flow condtions and can be seen more clearly in Figure 6. It would seem that greater flow rates result in higher reaction rates, at least under the study condtions. This makes sense as the anodes are being presented with more bromide per minute and thus can oxidize more of it per minute.

To examine the impact that the depth of the anodes had on the percentage removal, Figure 7 was prepared showing the results from reactors 1, 5, 9, 13, 17 and 18 with all flow conditions combined, each having a different depth but containing the same number of anodes and an equal gap between anodes (except for reactors 17 and 18). The three deeper reactors all had about the same reaction rates for
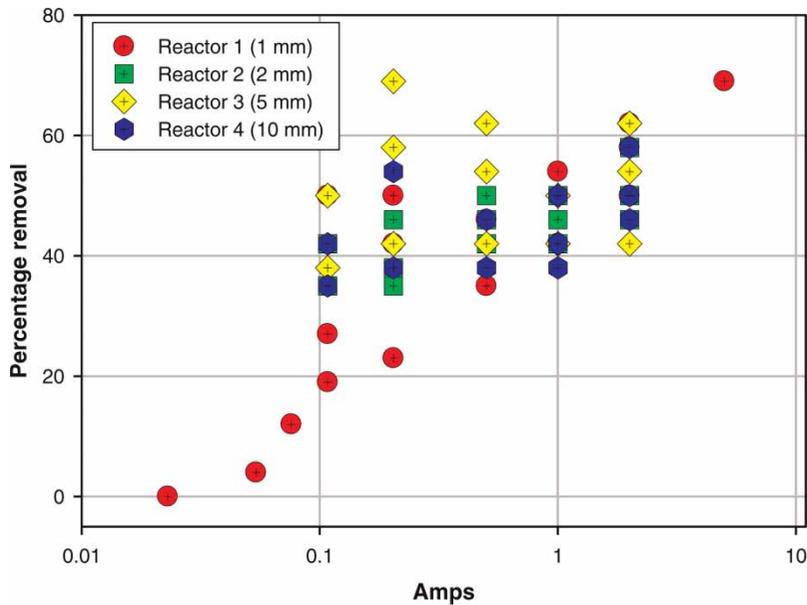
Figure 2 | Percentage removal of bromide as a function of current: reactors 1–4 (1.2 cm depth) with flows combined.
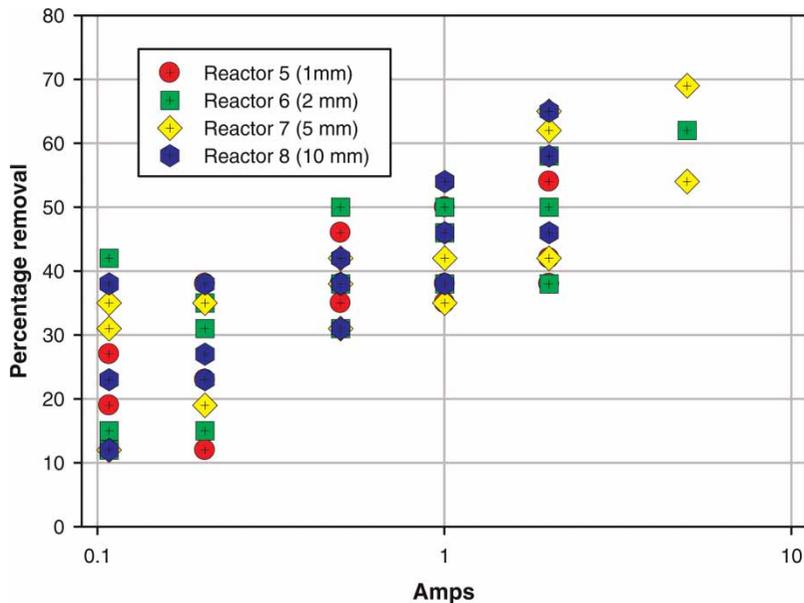

Figure 3 | Percentage removal of bromide as a function of current: reactors 5–8 (2 cm depth) with flows combined.

any given current. However, the shallower reactors showed dramatically higher reaction rates; the 1.2 cm deep reactor had noticeably higher rates than the other plate reactors and the hybrid. The mesh reactor exhibited much higher percentage removals at lower currents. At 0.02 A, the mesh reactor removed much more bromide than reactor 1. As the current increased, however, the mesh reactor removed

a lower and lower percentage of the bromide. At 2 A, the mesh reactor was removing as much bromide as the deeper plate reactors and less than the 1.2 cm deep reactor (reactor 1). This might suggest that the mesh reactor had significantly less anodic surface area than the plate reactors. This could mean that, while the mesh reactor had higher reaction rates, the limited surface became saturated and
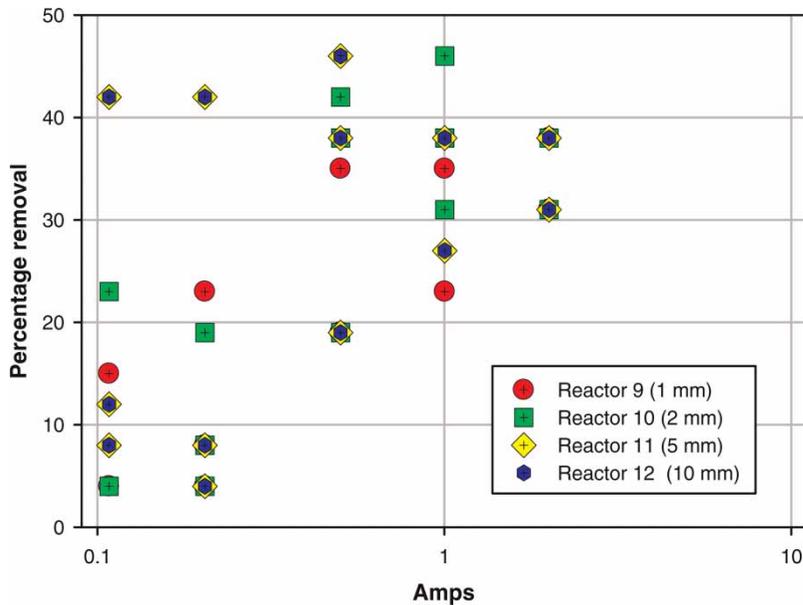
Figure 4 │ Percentage removal of bromide as a function of current: reactors 9–12 (5 cm depth) with flows combined.
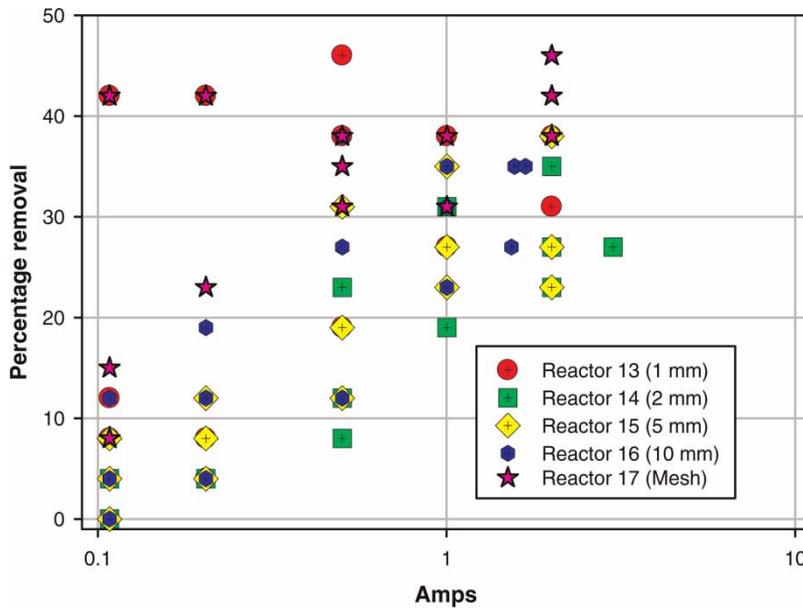

Figure 5 │ Percentage removal of bromide as a function of current: reactors 13–17 (10 cm depth and mesh) with flows combined.

could not remove proportionately more bromide as the current was increased. Of particular note, the hybrid reactor, number 18, removed all measurable bromide at 8 A.

To test this hypothesis, the data was re-plotted with *apparent* reaction rate on the Y axis and current on the X axis (Figure 8). It must be emphasized that these are apparent reaction rates because it was not possible to perform effective tracer studies in such small reactors. Also,

the hydraulics were most likely impacted by the formation of gas during electrolysis. Gas formation, particularly at the cathode, produced additional lift and buoyancy to water closer the cathode as opposed to that coming up through the centre. The gas formation also imparted mixing energy into the upflowing water. Since the amount of gas formation was proportional to the applied current, different mixing characteristics were achieved at each
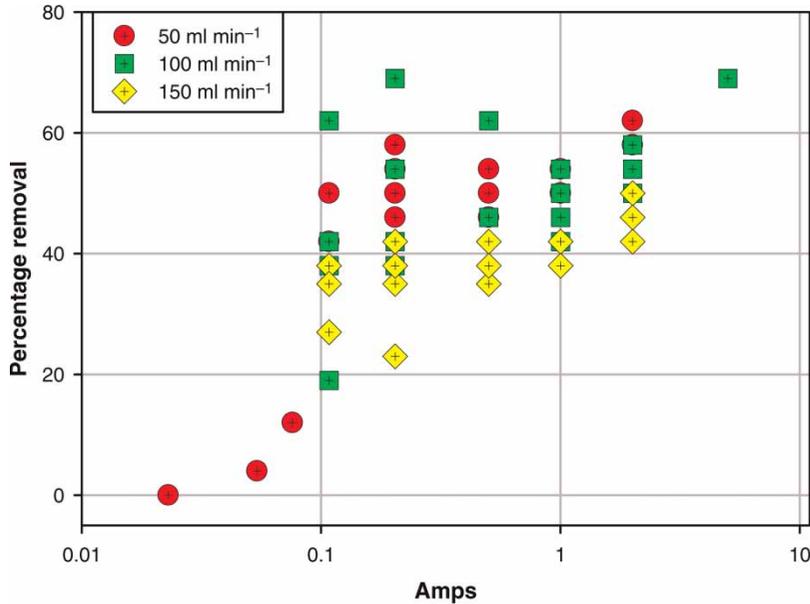
Figure 6 | Percentage removal of bromide as a function of flow and current: reactors 1–4.
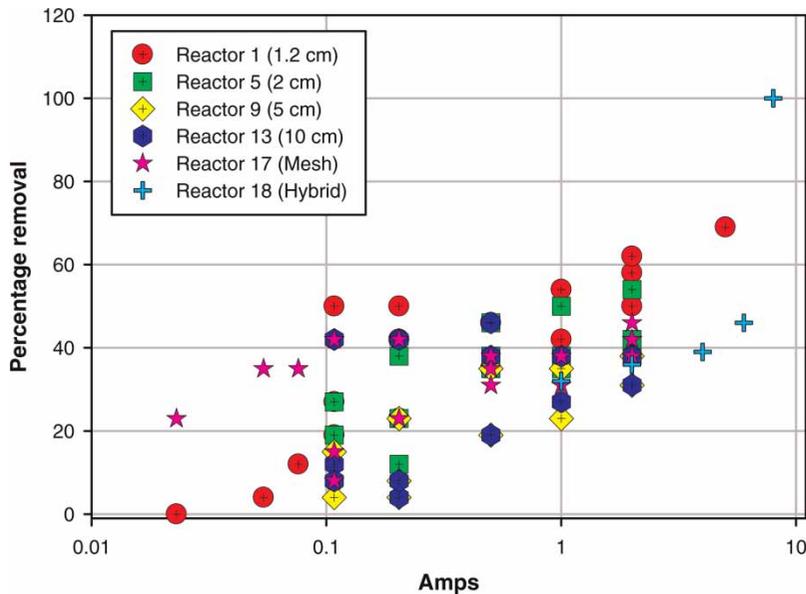

Figure 7 | Percentage removal of bromide as a function of current and anode depth: reactors 1, 5, 9, 13, 17, 18 with flows combined.

different current setting. With all of those limitations noted, Figure 8 shows the apparent reaction rates for reactors 1, 5, 9, 13, 17 and 18. At every measured applied current, the mesh reactor had a much higher apparent reaction rate and reactor 1, the shallowest plate reactor at 1.2 cm, had higher apparent reaction rates than the other plate reactors. The hybrid reactor's apparent reaction rates were not as high as either the mesh reactor or reactor 1.

The results seem to be somewhat contradictory. The mesh reactor has the highest apparent reaction rates at all currents but when the results are expressed as percentage removal, the mesh reactor is more effective only at very low currents. At higher currents, the shallower plate reactors have greater percentage removals. The hybrid reactor has apparent reaction rates that are no different from any of the plate reactors but has the highest percentage removal
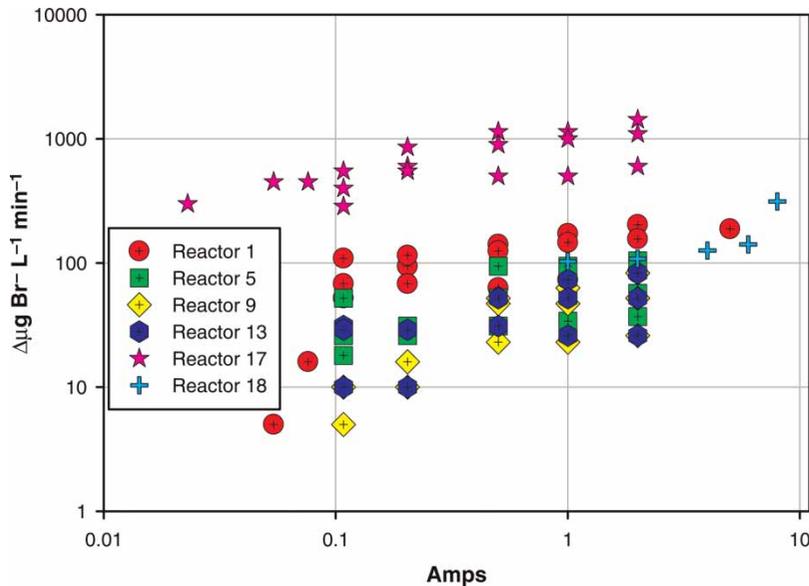
Figure 8 │ Reaction rate as a function of current and anode depth: reactors 1, 5, 9, 13, 17, 18 with flows combined.

at high currents. The hypothesis that appears to best explain these phenomena is that the reaction is two step; oxidation of bromide to bromine is one, and the volatilization of bromine is a second. The second, volatilization step works best with the shallower reactors. What might be occurring is that the deeper reactors allow re-solubilization of the bromine from the gas phase back into the aqueous phase as the gas bubble moves up through water to the surface. The 1.2 cm deep plate reactor allows this re-solubilization to occur less than the other plate reactors, and the mesh reactor, being only 0.06 cm deep, allows even less.

However, the mesh reactor had considerably smaller surface area than the plate reactors. This means that the available surface area of the mesh can be saturated much more readily than the plate reactors. So at low currents when the surface area is not saturated, the greater reaction rates of the mesh reactor results in more removal of bromide than the plate reactors. At higher currents, even though it has lower reaction rates, the 1.2 cm plate reactor can remove more bromide because of the greater reactive surface area. The hybrid reactor was designed to balance these two features; however, it does not seem to do so except at the highest currents (8 A). Part of this is due to the fact that the hybrid reactor had the largest surface area interface between the plates and the bus rod, producing the least resistance, more current was able to flow. However, there appears to be some

threshold effect with perhaps a second reaction occurring at much higher currents (above 6 A in this study) as the results are clearly non-linear. The nature of this threshold and other possible reactions is at this time unknown.

CONCLUSIONS AND IMPLICATIONS FOR FULL-SCALE TREATMENT PLANTS

If the results from this study are applicable to larger scale applications, it would seem that the optimum reactor has the maximum surface area of anodes to provide the greatest capacity to oxidize and volatilize, which are as shallow as possible to maximize the removal of bromine and minimize its reabsorption into the aqueous phase. A variable to be considered would be how much bromide needs to be removed. Bromide does not react in linear fashion with disinfectants and TOC; below a certain concentration, it is not incorporated into DBPs. This means there is a 'floor effect' below which further removal of bromide provides no additional benefits to lowering DBP concentrations. So if only a small amount of bromide needs to be removed, a mesh style reactor may be what is needed, while if a larger amount is needed, a shallow plate reactor may be more appropriate, and the hybrid may be better for other conditions. Obviously, further research is needed but the preliminary

results of this study would indicate that electrolytic oxidation and volatilization may be a potential practical technique for the removal of bromide from drinking water sources.

ACKNOWLEDGEMENTS

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First received 26 July 2010; accepted in revised form 5 December 2010