



Inhibition of bromate formation during drinking water treatment by adapting ozonation to electro-peroxone process



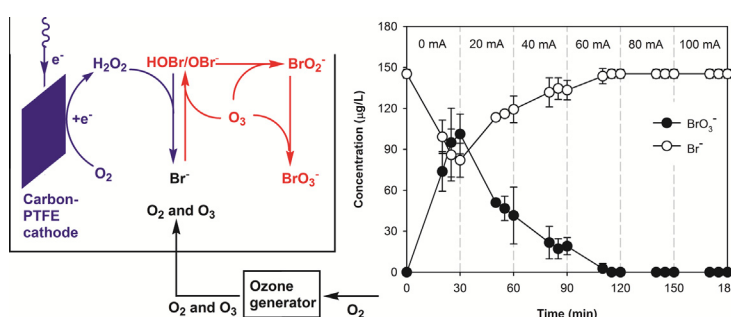
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HIGHLIGHTS

- E-peroxone couples ozonation with electrolysis to drive peroxone reaction electrochemically.
- H_2O_2 is electro-generated at a carbon-based cathode using O_2 in sparged O_2 and O_3 gas mixture.
- The in situ generated H_2O_2 rapidly reduces HOBr to Br^- , inhibiting BrO_3^- formation.
- Adaption of ozonation to E-peroxone enhances mineralization of organic contaminants.
- E-peroxone offers a simple way to improve existing ozonation systems for drinking water treatment.

GRAPHICAL ABSTRACT



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ABSTRACT

This study investigated bromate formation during drinking water treatment by conventional ozonation and electro-peroxone (E-peroxone) processes. The E-peroxone process combined conventional ozonation with electrolysis processes and used a carbon-based cathode to electrochemically convert O_2 in the sparged ozone generator effluent (O_2 and O_3 gas mixture) to H_2O_2 , which can then react with sparged O_3 to yield $\cdot\text{OH}$ (i.e., the peroxone reaction). Results show that ozonation of bromide-containing water (Br^- of $150 \mu\text{g/L}$) produced significant amounts of bromate (e.g., $\sim 60\text{--}120 \mu\text{g/L}$) in the effluent. In contrast, by applying a small current (e.g., $60\text{--}100 \text{ mA}$) to electrochemically generate H_2O_2 from sparged O_2 , the E-peroxone process can decrease bromate to $<10 \mu\text{g/L}$ and significantly increase natural organic matter (NOM) removal. The results indicate that the E-peroxone process may provide a simple and effective way to improve existing ozonation process for controlling bromate formation and removing NOM during drinking water treatment.

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1. Introduction

Ozone (O_3) is widely used in drinking water treatment as disinfectant and oxidant. However, ozonation of bromide-containing

water produces bromate (BrO_3^-) [1,2], which is a potential human carcinogen and pose significant threat to human health [3]. For example, it has been reported that long-term exposure to bromate may cause tumor development in different organs, renal failure, kidney impairment, hearing loss, etc. [3–5]. Due to the severely harmful effects of bromate on human health, stringent regulations have been set for the maximum bromate concentration in drinking waters, e.g., $10 \mu\text{g/L}$, in many countries [1].

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During ozonation, bromate is formed from a multistep oxidation of bromide by either O_3 , $\cdot OH$, or their combination [1,2,6]. Once formed, bromate is difficult to remove by conventional drinking water treatment processes (e.g., biologically active carbon (BAC) filtration and UV irradiation) that usually follow ozonation [1,7]. It is therefore desirable if bromate formation can be minimized during ozonation.

To prevent excessive bromate formation during ozonation, several strategies have been tested in drinking water treatment, for example, pH depression, ammonia addition, H_2O_2 addition, ferrate addition, and catalytic ozonation [1,8–13]. These strategies are capable of decreasing bromate formation to varied extents via different reaction mechanisms [1,9–12,14–16]. In particular, addition of H_2O_2 during ozonation (i.e., the so-called “peroxone process”) has been considered an attractive treatment option because it can simultaneously inhibit bromate formation and enhance pollutant degradation during drinking water treatment [2,6,7,11,17,18]. These improvements are mainly because (a) H_2O_2 can rapidly reduce $HBrO/BrO^-$ (which is a key intermediate for bromate formation) back to Br^- (Eq. (1), $k = 7.6 \times 10^8 \text{ M}^{-1} \text{ s}^{-1}$), thus decreasing bromate formation [1,2,17], and (b) H_2O_2 can react with O_3 (Eq. (2), $k = 2.8 \times 10^6 \text{ M}^{-1} \text{ s}^{-1}$) to yield $\cdot OH$, which is a much stronger oxidant than O_3 and can oxidize most organic solutes very rapidly [17,19]. Addition of H_2O_2 can thus generally improve the process performance of ozonation in terms of both bromate formation and pollutant degradation [2,11,17,18].



While the conventional peroxone process may offer a viable way to control bromate formation during drinking water treatment, it requires the addition of external H_2O_2 , which is an expensive chemical. Moreover, transportation, storage, and handling of high concentration H_2O_2 solution are dangerous due to its high reactivity. These problems have considerably limited the practical application of conventional peroxone process in water treatment [20,21]. To overcome the limitations associated with conventional peroxone process, we have recently developed an electro-peroxone (E-peroxone) process that can drive the peroxone reaction using in situ electro-generated H_2O_2 [22]. The E-peroxone process simply combines conventional ozonation with electrolysis processes and uses a carbon-based cathode to electrochemically produce H_2O_2 from O_2 (Eq. (3)) in the sparged ozone generator effluent (which is an O_2 and O_3 gas mixture). Thus, by utilizing the O_2 that is the predominant part (usually >90–95% V/V) of the sparged O_2 and O_3 gas, but would otherwise be wasted in conventional ozonation processes, the E-peroxone process can electrochemically produce H_2O_2 in situ at controllable rates [22]. We thus expected that the E-peroxone process may be able to provide a convenient, economical, and safe alternative to conventional peroxone process for controlling bromate formation during drinking water treatment.



To test this hypothesis, we investigated bromate formation during conventional ozonation and E-peroxone treatment of bromide-containing water. The effects of O_3 dosage, applied current, and the presence of natural organic matter (NOM) on bromate formation were evaluated. The results suggest that by simply adapting conventional ozonation to the E-peroxone system, the process performance in terms of bromate inhibition and NOM mineralization can be significantly improved.

2. Materials and methods

2.1. Sample collection and preparation

To investigate bromate formation during ozonation and E-peroxone treatment, two water samples were used: a synthetic water prepared in laboratory and a surface water collected from a lake in the suburban area of Beijing. The synthetic water was freshly prepared before each test by adding certain amounts of $CaCl_2$, $MgSO_4$, $NaHCO_3$, and KBr (analytical grade) to 40 L of deionized water; this composition was intended to simulate typical drinking water quality in China [23]. In some cases, small amounts of humic acid (Sigma–Aldrich) was also added into the synthetic water to obtain a dissolved organic carbon (DOC) level of $\sim 3 \text{ mg/L}$, so as to evaluate the effect of NOM on bromate formation. The surface water collected from the lake had a low original Br^- concentration of $\sim 25 \text{ }\mu\text{g/L}$. To better evaluate bromate formation during ozonation and E-peroxone process, appropriate amounts of KBr was added to the surface water to increase the Br^- concentration to $\sim 150 \text{ }\mu\text{g/L}$ before commencing the treatment. The main water quality parameters of the synthetic and surface water are summarized in Table 1.

2.2. Ozonation and E-peroxone treatment

The experimental setup of ozonation and E-peroxone treatment is shown in Fig. 1. The water was continuously fed into the bottom of an acrylic column (6 cm i.d., 1 m height) at a constant flow rate of 8.5 L/h, which resulted in a hydraulic retention time of 20 min in the reactor. The effluent was discharged at the top of the reactor. Electro-generation of H_2O_2 from O_2 was evaluated by sparging pure O_2 into the reactor during electrolysis (the ozone generator was off). For ozonation treatment, an ozone generator (EXT120-T, Ozone services, Canada) was used to produce O_3 from pure O_2 gas (99.9%) while the DC power for H_2O_2 production was off. The ozone generator effluent (O_2 and O_3 gas mixture) was sparged into the bottom of the reactor at a constant flow rate of 80 mL/min. After 30 min of ozonation, the treatment was changed into the E-peroxone process by turning on the DC power to produce H_2O_2 , while the ozone dose was kept at the same level as ozonation alone. The anode was a RuO_2/IrO_2 coated Ti plate (5 cm \times 4 cm \times 0.2 cm) and the cathode was a carbon–polytetrafluorethylene (carbon–PTFE) electrode (5 cm \times 6 cm \times 0.2 cm) prepared with Vulcan XC-72 carbon powder (Cabot Corp., USA), PTFE dispersion, and anhydrous alcohol following the procedure described elsewhere [24]. The distance between the anode and cathode was $\sim 0.4 \text{ cm}$.

Table 1
Main water quality parameters of the synthetic and surface water used in this study.

Constituent	Synthetic water	Surface water
DOC (mg/L)	0 ^a	6.1
Br^- ($\mu\text{g/L}$)	150	150 ^b
pH	8.1	8.3
Alkalinity (mg/L as $CaCO_3$)	75	116
Conductivity ($\mu\text{S/cm}$)	417	378
Ca^{2+} (mg/L)	29.0	42.1
Mg^{2+} (mg/L)	10.1	27.1
Na^+ (mg/L)	34.5	13.7
HCO_3^- (mg/L)	91.5	142
NO_3^- (mg/L)	n/a	1.66
Cl^- (mg/L)	51.4	19.1
SO_4^{2-} (mg/L)	40.6	50.9
F^- (mg/L)	n/a	0.32

^a Small amount of humic acid was added to increase DOC to $\sim 3 \text{ mg/L}$ in some tests.

^b Original Br^- concentration was $\sim 25 \text{ }\mu\text{g/L}$; appropriate amounts of KBr was added to increase Br^- to $150 \text{ }\mu\text{g/L}$ before ozonation and E-peroxone treatment.

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