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Recent developments in aqueous phase bromate (BrO_3^-) species treatment using adsorption, reduction, and ion-exchange processes

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ABSTRACT

Recently, bromate (BrO_3^-) presence in the drinking water sources and its health-related concerns (e.g., carcinogenic) has initiated a lot of attention both within the regulatory bodies across the globe (such as WHO, US EPA, EU, etc.) and also within the scholarly community that look for efficient bromate removal techniques to meet the desired bromate water quality standards. Bromate species is typically formed during disinfection–ozonation of bromide containing water. Considering its carcinogenic and mutagenic threat toward human health, several recent research studies have investigated different processes for efficient bromate removal from the aqueous phase under a varying set of conditions and also report the optimum process variables. In this review paper, we have presented and summarized findings from the respective recent work on bromate removal techniques and have also discussed the effect of various process variables on bromate removal efficiency. The adsorption process is noted to be the most common and widely studied technique. Some of the adsorbent materials used included activated carbon, both unmodified and modified. The respective adsorbent samples are noted to have varying specific surface area, pore size, and surface morphology. In most studies, the effect of pH, initial bromate concentration, contact time, temperature, adsorbent dose, mixing speed, and coexisting ions was investigated to get optimum bromate removal. The adsorption equilibria and kinetics were mainly predicted by Langmuir/Freundlich isotherms and pseudo-second-order models, respectively. Also, the Donnan dialysis and electrodialysis ion-exchange processes removed bromate ions to acceptable concentrations. Bromate reduction can also be achieved using advanced reduction processes where ultraviolet light has been used as an activating agent and mainly sulfite as the reducing agent. Furthermore, chemical reduction, electrochemical reduction, and bio-reduction-based bromate removal processes are also discussed. The reduction-based processes indicated the formation of different reduced species including bromine. The ferrous-based reduction processes are noted to be very effective and several variations of ferrous-based technologies including adsorption and reduction processes are presented and discussed. In summary, this review work indicates a very positive development in the respective area of study, that is, bromate removal from the concerned aqueous streams, and hopefully will also serve as a focal point for further scientific innovations and endeavour for a better human health.

Keywords: Bromate; Adsorption; Activated carbon; Reduction; Ion exchange; Bio-reduction

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1. Introduction

Recently, a great concern has been shown regarding the formation of disinfection by-products (DBPs) during drinking water treatment due to their potential health concerns including carcinogenic nature of some of the DBPs. Though chlorine-based DBPs are known, however, new research has shown that bromide-based DBPs are also of concern. One such compound, that is, bromate (BrO_3^-) has really caught the attention of regulators in many advanced countries with its drinking water limit up to 10 μg/L [1–5]. Considering this a great interest has been shown in investigating different processes and techniques that can effectively remove bromate from respective water streams under a varying set of conditions. Several recent studies have explored the use of adsorption, ion-exchange, and several reduction-based technologies for bromate removal from the aqueous phase. The present work summarizes the respective findings from some of the recent advances on bromate removal and presents the respective process efficiencies (under a varying set of process variables) using several traditional and advanced systems. Hopefully, this work will help as a valuable review source for further advancement in this important area of study that is of great public health concern.

2. Recent advances in aqueous phase bromate (BrO_3-) treatment

2.1. Bromate removal using adsorption

Adsorption is a widely used and an effective water treatment process that has been successfully employed to remove several contaminants of concern to acceptable levels [6]. Though granular activated carbon (GAC) or powder activated carbon (PAC) produced from a variety of carbon-based sources are common adsorbents nevertheless several other carbon-based advanced materials and aluminum- or iron-based material have also been used as adsorbents [7–12]. Furthermore, the removal of bromate from water using activated carbon under a varying set of process parameters like pH, initial bromate concentration, adsorbent dose, temperature, contact time, and coexisting anions have also been investigated by several researchers (Table 1). Some of these studies have also focussed on the use of modified forms of adsorbents that would typically introduce cationic surface sites to enhance the bromate adsorption capacity of the parent adsorbent material (Table 1). The modifiers reported include surfactants [6,13], metallic impregnation [14,15], nano zero-valent iron [7,16–18], ferrous sulfate [19], HNO_3 [20,21], NaOH [20,21], NH_3 [20], HCl [19], isopropyl alcohol [22], and H_2O_2 [21]. Though bromate concentration in typical water intake sources is generally <1 mg/L [14,23–25] nevertheless bromate amount in respective treatment studies covered a broad range (Table 1). Furthermore, technologies that incorporate iron-based materials and ion exchange [26–33] have also been used for bromate removal. Several studies on bromate removal indicate it to be exothermic [11,24,30] with pseudo-second-order kinetics [2,6,9,10,17,22,24,30]. Surface modifications of respective parent materials also reveal details that are important for efficient bromate removal. For example, several researchers have investigated the use of cationic surfactants to introduce positive surface groups onto activated carbon surface for enhanced bromate removal. The use of activated carbon modified with cetylpyridinium chloride (CPC) and having a specific surface area of 392 m^2/g (Table 1) yielded approximately 80%–85% bromate removal within a wide pH range of 4–9 and with adsorption capacity of 17 mg/g and chemisorption noted to be the dominant bromate removal mechanism [6]. In another study, bromate removal was investigated using PAC modified with three types of cationic surfactants, that is, CPC, hexadecyltrimethylammonium chloride (CTAC), and hexadecyltrimethylammonium bromide (CTAB); both CPC and CTAC modified activated carbon indicated enhanced efficiency with CTAC modified PAC showing tripling in bromate adsorption compared with unmodified PAC [13]. However, unlike the other findings [6] the pH effect was more specific with pH below 7 yielding better bromate removal. Such a difference in effect of pH on process efficiency as noted in above studies [6,13] could be explained by differences in pH_{zpc} between the two surfactant modified activated carbon samples with one having low pH_{zpc} showing more profound pH depended effect. Hence, pH control should be considered as an important process variable to optimize bromate's adsorption-based removal. Like cationic surfactants several cationic metal species have also been used for bromate removal. However, their role is typically more broad and varying such as catalyst, surface modifier, redox agent, ion exchange, or adsorbent. In such systems at first the metal species typically initiates an interaction between the metal-support surface and bromate species. In one study, bromate removal using silver-activated carbon showed better bromate removal performance than the unmodified carbon specifically at lower pH [15]. Similarly, the use of bimetallic catalysts Pd (1%) and Cu (2%) supported on activated carbon yielded >95% bromate removal via reduction to bromine [14]. Furthermore, several studies have also reported the use of zero-valent iron (ZVI) loaded onto supports such as activated carbon for efficient bromate removal. The main mechanisms in such systems involve reduction of bromate species and adsorption both onto activated carbon and Fe precipitates that result from simultaneous oxidation of ZVI. For example, use of nanoscale zero-valent iron (nZVI) immobilized onto activated carbon sample is reported to yield near complete bromate removal [7]. However, such processes are very pH dependent with pH between 3 and 9 noted to yield efficient bromate removal. The dominant explanation for such pH dependent process efficiency is ZVI releasing Fe species and its oxidation to Fe^{2+} that follows electron gain by the bromate species. A further addition to such process could be Fe^{2+} oxidation to Fe^{3+} species that is thermodynamically even more favorable over ZVI/Fe^{2+} couple. As Fe^{2+} will have a higher presence at lower pH values and the Fe^{2+}/Fe^{3+} redox path is also thermodynamically more favorable, could possibly result in an overall enhanced bromate reduction at lower pH, followed by its enhanced removal because of adsorption onto produced Fe-based precipitates. Hence, a combination of abovementioned simultaneous reactions specifically at low pH values will initiate enhanced bromate removal. A further support for such bromate removal mechanisms comes from another set of studies from Xu et al. [2,17]. The use of only ordered mesoporous carbon (OMC) [2] caused about 85% bromate removal from aqueous phase with bromate removal capacity

Table 1
Summary of different adsorbent materials, treatment parameters, and adsorption capacity for bromate removal

Adsorbent details			Treatment parameters								Adsorption capacity, mg/g	Model[b]	Ref.
Adsorbent	Modifier[a]	Specific surface area (m^2/g)	$C_i BrO_3$ (mg/L)	pH	Optimum pH	Dosage (g/L)	Temperature (K)	Shaking speed (rpm)	Contact Time (h)	Coanions			
Activated carbon	CPC	392	20	3–11	4–9	2.00	293, 303, 313	200	16	–	17	L	[6]
Commercial activated carbon	Pb and Cu	870	10	NA	NA	2.22	NA	700	0.5	–	NA	NA	[14]
Commercial powder activated carbon	nZVI	97.5	10	3–10	3–9	0.2–0.5	288, 298, 308	200	0.05–0.33	–	433.9	NA	[7]
Coal-based GAC	Iron	488	6.3–37.8	1.5–12	6–8	1.00	NA	200	50	–	21.42	L	[26]
Ordered mesoporous carbon (OMC)	nZVI	118	1–10	6	NA	0.35	288, 298, 308	NA	0.017–2	–	31.82	L	[27]
Ordered mesoporous carbon (OMC)	None	1061	0.1–10	3–11	6–10	0.01–0.07	288, 298, 308	150	0.083–12	Cl, NO_3, CO_3, SO_4, PO_4	17.58	F	[2]
Carbon nanotubes (CNTs)	Iron	NA	0.1–2.5	3–11	3	0.1–2.5	298	150	1–48	NO_3, Cl	0.35	NA	[28]
Raw carbon nanotube	None	NA	0.1–2.5	3–11	3	0.1–2.5	298	150	1–48	NO_3, Cl	0.32	NA	
Powdered activated carbon	None	822.7	1–100	3–11	3–7	2.00	298, 308, 333	NA	0.25–12	–	NA	L	[13]
Powdered activated carbon	CPC	608.9	1–101	3–11	3–7	2.00	298, 308, 333	NA	0.25–12	–	NA	L	
Powdered activated carbon	CTAC	571.3	1–102	3–11	3–7	2.00	298, 308, 333	NA	0.25–12	–	NA	L	

(Continued)

Table 1 *(Continued)*

Adsorbent details			Treatment parameters										
Adsorbent	Modifier[a]	Specific surface area (m^2/g)	$C_i BrO_3$ (mg/L)	pH	Optimum pH	Dosage (g/L)	Temperature (K)	Shaking speed (rpm)	Contact Time (h)	Coanions	Adsorption capacity, mg/g	Model[b]	Ref.
Powdered activated carbon	CTAB	600.1	1–103	3–11	3–7	2.00	298, 308, 333	NA	0.25–12	–	NA	L	
Granular activated carbon	None	975	1–10	6.5	NA	NA	298, 303, 313, 333	NA	0–1	–	NA	NA	[5]
Coal-based activated carbon	None	798	0.05	NA	NA	NA	298	180	2	–	1.47	T	[20]
Coal-based activated carbon	HNO_3	672	0.05	NA	NA	NA	298	180	2	–	0.88	T	
Coal-based activated carbon	NaOH	709	0.05	NA	NA	NA	298	180	2	–	1.1	T	
Coal-based activated carbon	NH_3	867	0.05	NA	NA	NA	298	180	2	–	1.54	T	
Date seed carbon	$Fe(SO_4).7H_2O$	NR	0.051	7	NA	10.00	298	250	0.25–24	–	NA	NA	[19]
Granular activated charcoal	HCl	NR	0.051	7	NA	10.00	298	250	0.25–24	–	NA	NA	
Nano-crystalline akaganeite (β-FeOOH)-coated quartz sand (CACQS)	None	1.83	0.6–10	3–12	3–10	20–60	288, 298, 308	150	0.017–4	–	0.038	L	[10]
Nano-Al_2O_3	None	NR	5–10	2–12	6	0.5	298	–	2	–	~6	L	[9]
Calcined Zn–Al	None	NR	0.1–1	4.5–11.5	7.0	0.8	293–323	–	6	Cl, F, PO_4, NO_3, SO_4, ClO_4	NA	F	[24]

(Continued)

Table 1 *(Continued)*

Adsorbent details			Treatment parameters								Adsorption capacity, mg/g	Model[b]	Ref.
Adsorbent	Modifier[a]	Specific surface area (m^2/g)	$C_i BrO_3$ (mg/L)	pH	Optimum pH	Dosage (g/L)	Temperature (K)	Shaking speed (rpm)	Contact Time (h)	Coanions			
Granular ferric hydroxide	None	250–350	10–50	4–8	NA	10	283, 298, 318	–	24	Cl, F, PO_4, NO_3, CO_3, SO_4	16.5	L	[32]
Corncobs	IPA	NR	20–120	2–10	8–9	1.00	303	250	1.5	Cl	101	F	[22]
Wood-based carbon	None	828	0.1–0.5	2–10	2–6	1.00	293	120	24	–	<0.1	ID	[29]
Fruit-based carbon	None	1,102	0.1–0.5	2–10	2–6	1.00	293	120	24	–	~0.27	ID	
Coal-based carbon	None	688.8	0.1–0.5	2–10	2–6	1.00	293	120	24	–	~0.4	ID	
Wood-based carbon	N_2	841.1	0.1–0.5	2–10	2–6	1.00	293	120	24	–	<0.1	ID	
Fruit-based carbon	N_2	1,270	0.1–0.5	2–10	2–6	1.00	293	120	24	–	~0.3	ID	
Coal-based carbon	N_2	773.1	0.1–0.5	2–10	2–6	1.00	293	120	24	–	~0.45	ID	
Virgin granular activated carbon	None	NR	0.1–1	5–10	9.46	1.50	298	180	0.17–20	Cl, F, Br, PO_4, NO_3, CO_3, SO_4	NA	NA	[15]
Activated carbon	Silver	NR	0.1–1	5–11	9.46	1.50	298	180	0.17–20	Cl, F, Br, PO_4, NO_3, CO_3, SO_4	0.63–3.32	NA	
Coconut shell-based activated carbon	None	950	0.2	7.5	NA	NA	298	120	24	–	2.5	L	[23]
Bituminous coal-based activated carbon	None	950	0.21	7.5	NA	NA	298	120	24	–	1.5	L	
Wood-based activated carbon	None	980	0.2	7.5	NA	NA	298	120	24	–	6.5	L	

(Continued)

Table 1 (*Continued*)

Adsorbent details			Treatment parameters								Adsorption capacity, mg/g	Model[b]	Ref.
Adsorbent	Modifier[a]	Specific surface area (m^2/g)	C_iBrO_3 (mg/L)	pH	Optimum pH	Dosage (g/L)	Temperature (K)	Shaking speed (rpm)	Contact Time (h)	Coanions			
Virgin activated carbon	None	654	60	6.0 ± 0.02	NA	1.00	298	200	48	NO_3, SO_4, Cl, NOM	5	F	[21]
Activated carbon	Thermal	643	60	6.0 ± 0.02	NA	1.00	298	200	48	NO_3, SO_4, Cl, NOM	7.5	F	
Activated carbon	HNO_3	489	60	6.0±0.02	NA	1.00	298	200	48	NO_3, SO_4, Cl, NOM	4	F	
Activated carbon	H_2O_2	554	60	6.0±0.02	NA	1.00	298	200	48	NO_3, SO_4, Cl, NOM	5.5	F	
Activated carbon	NaOH	548	60	6.0±0.02	NA	1.00	298	200	48	NO_3, SO_4, Cl, NOM	3.8	F	

[a] Cetylpyridinium chloride (CPC), hexadecyltrimethylammonium chloride (CTAC), hexadecyltrimethylammonium bromide (CTAB), isopropyl alcohol (IPA), nano zero-valent iron (nZVI).

[b] Langmuir (L), Freundlich (F), Toth (T), intra-particle diffusion (ID), not applicable (NA), not reported (NR).

of 17.6 mg/g and no bromide formation that conforms to systems having no reducing agent such as ZVI species; this is also indicative of adsorption as the dominant bromate removal mechanism. This argument is augmented by the other study in which OMC impregnated with nZVI [17] yielded dominant reduction to bromide as confirmed by mass balance analysis (90% bromide formation) and comparatively higher bromate removal capacity of 31 mg/g. This confirmed that the introduction of nZVI in OMC at microlevel sets the thermodynamics in favor of faster electron transfer hence resulting into the reduction of bromate species. A comparison between the unmodified and modified OMC bromate removal data also indicated increased kinetics in case of latter, which shows that modified OMC yields dual advantage of bromate reduction to bromide in a comparatively short duration thus requiring a smaller reactor size for similar bromate treatment requirement and is therefore economically more feasible. Hence, nZVI impregnated systems look to be promising for efficient bromate removal. Similarly, other iron-based treatment systems also report higher bromate removal. For example, the use of both raw and Fe (1%) modified carbon nanotubes (CNTs) showed the latter to yield higher efficiency that was attributed to greater and increased bromate attraction because of iron species on the CNT surface [27]. The respective Fe-based CNT samples were also noted to be more efficient at acidic pH like the other aforementioned modified systems [13,15]. On the other hand the application of nano-Al_2O_3 for bromate removal [9] showed that both lower and higher pH result in decreased bromate removal efficiency with pH 6 reported to be the optimum. At very acidic pH values, a higher concentration of H^+ ions in the near vicinity of amphoteric metal oxide surface may restrict bromate approach to the surface whereas at elevated pH values this could be due to (1) competition for the active sites by the OH^- ions and (2) electrostatic repulsion of anionic bromate species by the negatively charged nano-Al_2O_3 surface. This indicates that the amphoteric nature of adsorbent including its pH_{zpc} value plays an important role in bromate removal and therefore should be considered as an important process parameter. Another study that investigated the application of nanocrystalline akaganeite (beta-FeOOH)-coated quartz sand (CACQS) for bromate removal noted maximum adsorption capacity at pH 3 and 10 [10]. Also, the use of nano-iron hydroxide impregnated granular activated carbon (Fe–GAC) at several Fe to carbon ratios 0.6%–1.2% (w/w) has shown optimum pH for bromate removal between 6 and 8 [26]. Both bromate reduction to bromide along with adsorption were reported to be the dominant bromate removal mechanisms. Hence, the aforementioned discussion indicates that the use of modifiers (for parent adsorbents) along with careful adjustment of process pH yields effective bromate removal.

The application of acid or base treated surfaces especially activated carbon has also been studied, and several groups explored such surfaces for bromate removal as well [19,20,21]. Respective studies reveal several important factors that relate the bromate removal efficiency to specific surface properties of adsorbents. For coal-based activated carbon [20] the following bromate adsorption capacity trend was noted: ammonia-treated carbon (1.54 mg/g) > untreated carbon (1.47 mg/g) > sodium hydroxide-treated carbon (1.1 mg/g) >

nitric acid-treated carbon (0.88 mg/g). It is also interesting to note that the abovementioned bromate adsorption trend for different modified activated carbon samples also followed the respective specific surface area values (Table 1), that is, ammonia-treated carbon (867 m^2/g) > untreated carbon (798 m^2/g) > sodium hydroxide-treated carbon (709 m^2/g) > nitric acid-treated carbon (672 m^2/g). This also indirectly indicates the significance of surface sites and their role in bromate adsorption. A comparatively better bromate removal performance as noted for the ammonia-modified activated carbon was suggested to result from electrostatic interactions at lower pH values along with adsorption. Nevertheless, it is also important to note that the untreated carbon yields better performance than the nitric acid and sodium hydroxide treated carbon samples [20] and similar observations have been made by others as well [21] indicating that the introduction of moieties such as nitrate or hydroxyl ions could also potentially cause excessive presence of anionic species in the near surface region of adsorbent thus resulting in reduced bromate removal. A preferential nitrate removal (compared with bromate) has also been noted for bio-membrane-based ion-exchange processes [32]. The same has been noted for nitrate/CNT systems as well though the use of HCl modified CNT showed increased bromate removal [27]. Furthermore, the use of HCl modified GAC also indicated 99% bromate removal with both adsorption and reduction reported to be the main bromate removal mechanisms [19]; a reasonable bromate mass transfer from the bulk liquid to bulk solid phase even at low initial bromate concentration indicated higher affinity of bromate for appropriately modified carbon-based material surfaces.

Removal of bromate using several other materials including agricultural wastes has also been investigated showing effective results [19,22,24,28]. The use of powdered activated carbon produced from several carbonaceous materials including wood, coal, and fruit materials indicated that the fruit-based activated carbon having the highest specific surface area and pore volume also yielded maximum bromate adsorption capacity [28]. A higher pore volume helps to enhance interparticle diffusion thus resulting in higher bromate removal with some bromate reduction as well. In one study [19] date seeds modified with ferrous sulfate yielded nearly 70% bromate removal with both adsorption and reduction reported to be the main bromate removal mechanisms. The use of unmodified and modified corncobs was also investigated for bromate removal [22]; the chemically modified corncobs with 80% isopropyl alcohol yielded higher adsorption than the other modified corncob samples using *n*-butyl alcohol, citric acid, and hydrochloric acid. The adsorption data fitted well to the Freundlich model with maximum adsorption capacity of 101 mg/g. The authors report ion exchange and adsorption as the main mechanisms involved in the removal of bromate, whereas the adsorption kinetics was well described by pseudo-second-order model. Also, use of calcined Zn–Al layered double hydroxides yielded 98% bromate removal at neutral pH with the reaction noted to be exothermic [24].

The effect of initial bromate concentration onto its removal shows varying trends depending on the adsorbent type and modifications of adsorbent. For example, the use of unmodified GAC showed decreased bromate removal with an increase in its initial aqueous phase concentration [5]. On the other hand modified activated carbon and CNT samples showed increased bromate removal with increased initial bromate concentration that could result from bromate ions' higher mass transfer rate at elevated concentrations [6,27]. Similarly, the use of granular ferric hydroxide (GFH) for bromate removal also showed that an increase in the initial bromate concentration had a positive influence on the bromate removal indicating increased mass transfer across the liquid–solid interfacial zone [30]. Higher bromate removal and bromate sorption on GFH was explained using surface diffusion and pore diffusion mechanisms. Higher concentration of bromate in case of modified surfaces specifically those having Fe species are expected to go through a cyclic process during which the bromate accepts electron and is reduced to bromide with further adsorption onto the produced iron precipitates and the parent adsorbent, which thus keeps driving the mass transfer from bulk liquid to bulk solid phase, resulting in increased bromate removal [7,17].

In summary, the above literature review from recent bromate adsorption-based removal studies indicates that the aqueous phase bromate species can be removed under a varying set of conditions and using a variety of both unmodified and modified materials along with a careful adjustment of process variables. We now report bromate removal findings using other processes including the ion-exchange process and reduction-based techniques, as provided in the following sections.

2.2. Bromate removal using ion-exchange process

The ion-exchange process that is widely used for water treatment specifically for the removal of ionic species is an environmentally friendly technique considering that the ion-exchange resin that can be regenerated and reused [29,31]. To that end several resins-based ion-exchange materials and also inorganic ion exchangers have been used [3,29,31,34,35] with process kinetics reported as pseudo-second-order type [8,31]. Furthermore, the selectivity of specific ion-exchange resins for specific ionic species also renders this process very efficient for the removal of target pollutants. For example, it has been reported that the anion exchange materials can bring bromate levels down to 10 μg/L or even less [3,32], whereas the use of an ion-exchange material for pre-treatment also caused reduced bromate formation in the downstream processes [33,36]. Other details from respective ion-exchange systems are summarized in Table 2. Several studies report successful removal of bromate under competitive conditions as well [3,8,29,32]. Application of an inorganic ion exchange (Mg–Al hydrous oxide-based) for arsenate, fluoride, bromate, bromide, selenite, and borate removal under competitive conditions yielded bromate removal capacity of 90 mg/g at pH 7 [3]. Also, the use of De-Acidite FF-IP resin yielded greater than 98% bromate removal in 10 min with process noted to be more efficient within acidic pH range of 4 and 7 [29]. The presence of competing anions reduced the bromate removal efficiency to 85%. Another study that used Amberlite IRA-400 for bromate removal showed an enhanced bromate removal with increasing pH (from 2 to 6.5) and increasing temperature (from 20°C to 45°C, endothermic process) [31]. Similar was noted in the case of magnetic ion

Table 2
Summary of different ion-exchange systems for bromate removal along with the treatment parameters

Ion-exchange types	Treatment conditions					Removal efficiency (%)	Coexisting ions	Reference
	$C_i BrO_3$ (mg/L)	Salt concentration, mM	Number of cells	pH	Time (h)			
Amberlite IRA-400 (OH− form)	0.2–1	NA	NA	2–7	1	62.8–84	NA	[31]
Ion-exchange membranes (Donnan dialysis) Selemion AMV membranes	0.1	100	20	NA	NA	93	HCO_3, Cl, SO_4	[37]
Ion-exchange membranes (Donnan dialysis) Neosepta ACS membranes	0.1	100	20	NA	NA	97	HCO_3, Cl, SO_4	
Ion-exchange membranes (electrodialysis) AMX/CMX membranes	0.1	100	15	NA	NA	96	HCO_3, Cl, SO_4	
Ion-exchange membranes (electrodialysis) ACS/CMX membranes	0.1	100	15	NA	NA	92	HCO_3, Cl, SO_4	
Selemion AMV membranes	0.05–0.2	50–300	20	NA	3	90	HCO_3, NO_3	[39]
Neosepta AFN membrane	0.05–0.2	50–300	20	NA	3	70	HCO_3, NO_3	
Macroporous anion exchange (D201-Cl) resin	0.2–0.6	NA	NA	2.5–13.0	0.17–10	90.7	NO_3	[34]
Double-layered hydrous oxides of Mg–Al ion-exchange	20–500	NA	NA	7	0.05	90[a]	F, AsO_4, Br, SeO_4, SeO_3, BO_4	[3]
Selemion AMV membranes	0.2	200	20	5.6	3.5	94	HCO_3, SO_4	[38]
Neosepta ACS membrane	0.2	100	20	5.6	2.5	90	HCO_3, SO_4	
Neosepta ACS membrane	0.2	100	NA	7.4	NA	96	NA	[32]
Magnetic ion-exchange resin (MIEX)	5–10	NA	NA	6–9	2	85	Cl, SO_4, HCO_3	[8]
De-Acidite FF-IP anion exchange resin	1	NA	NA	2–10	0.17–2	>98	NO_3, CO_3, SO_4, Cl, F, PO_4	[29]

[a] mg [Br]/g.

exchange resin that yielded high bromate removal at pH 6–7 and higher temperature (endothermic) though the presence of anions like chloride, bicarbonate, and sulfate had a negative impact on bromate removal due to competitive exchange with the resin surface [8]. Zhong et al. [34] who investigated the use of a macroporous Cl-type anion exchange resin to remove bromate along with different design of experimental statistical approaches report significant bromate removal at optimal conditions of resin dosage 1.08 g/L and pH 8.9 in the presence of other anions. Successful application of Donnan dialysis (DD) ion-exchange membranes for the bromate removal has also been reported with more than 94% removal [32,37–39]. Some details from these studies are also summarized in Table 2. In one study, the use of DD membranes with total surface area of 0.140 m^2 removed about 97% of the initial bromate [37]. Also, Wiśniewski and Kabsch-Korbutowicz [39] reported bromate removal efficiency of 90% using a DD membrane at low salt concentration. The application of DD ion-exchange membrane with biofilm for bromate removal has also been reported, though at a slower rate [32]. The specific bromate bio-reduction factor (mg-BrO_3/$g_{cell\ dry\ weight}$.h) was noted to be within 0.03–0.1 thus needing longer hydraulic retention times for near complete bromate removal via bio-reduction pathway. The above summary indicates that bromate can be successfully removed using different types of ion-exchange-based systems under varying conditions, though a fine control of process variables such as pH, may be required.

2.3. Bromate removal using reduction-based processes

The application of several reduction-based processes that include advanced UV lamp systems, iron-based systems, bio-reduction, and electrochemical techniques, has been reported for bromate removal. In that regard use of UV lamps incorporating reducing agent such as sulfite or photocatalyst TiO_2 for bromate reduction-based removal is discussed here and summarized in Table 3. The use of UV lamp irradiated systems with sulfite as a reducing agent has shown successful bromate reduction to bromine/bromide species [40–42]. The reaction kinetics in respective systems was noted to be the pseudo-first-order type, which also well described bromate removal under a varying set of conditions [40–42]. Also, UV lamps with wavelength lower than 254 nm were noted to be more efficient than those with wavelength higher than 254 nm [40,41] and in another study, a medium-pressure UV lamp was noted to be more efficient compared with low-pressure UV lamp [42]. Furthermore, increasing sulfite concentration was also noted to have a positive impact on bromate removal [40,42] especially for the low-pressure UV lamp [42]. The main mechanisms during such bromate removal studies included direct photolysis and reaction with radicals including the sulfite anion radicals, aqueous electrons, and hydrogen atom radicals. However, the presence of specific ions may affect bromate removal efficiency because of synergistic effects. Kishimoto et al. [41] noted that the presence of bicarbonate and nitrate ions decreased bromate removal whereas sulfate and phosphate had no effect on bromate removal, which is similar to elsewhere noted insignificant effect of coexisting ions on bromate reduction [43]. Such negative effects of specific ions onto bromate removal could result from competition for the aqueous electrons. Also, the application of photocatalyst-based reduction process has shown 70%–75% bromate reduction to bromide at neutral pH using electrode coated with either titanium oxide gel or titanium foil and irradiated with a UV lamp [43]. The above systems indicate successful application of UV-irradiated systems for bromate removal via the reduction pathway. In continuation the use of iron-based systems for chemical reduction of bromate has also been investigated as summarized in Table 4. The respective iron-based systems

Table 3
Summary of different advanced reduction systems for bromate removal along with the treatment parameters

Wave length (nm)	Power (W)	Reducing agent	pH	Optimum pH	C_i BrO_3 (mg/L)	Coexisting ions	Time (h)	Pseudo-first-order rate (min^{-1})	Mechanism	Reference
222	NA	Sulfite	3–11	7–9	0.352	NA	3–4	0.265–0.303	Photolysis, sulfite radicals, and aqueous electron	[40]
254	36	Sulfite	3–11	7–9	0.352	NA	3–4	0.013–0.017		
172	20	Sulfite	3–11	3–7	0.1–1	SO_4, PO_4, NH_4, HCO_3, NO_3	1	0.0241	Photolysis, sulfite radicals, and aqueous electron	[41]
254	20	Sulfite	3–11	3–7	0.1–1	SO_4, PO_4, NH_4, HCO_3, NO_4	1	0.0059		
254	8	Sulfite	4–11	9–11	0.25–1	NA	2–2.5	0.017	Direct photolysis	[42]
200–600	150	Sulfite	4–11	7–11	0.25–1	NA	2–2.5	0.051		
200–900	500	Ti/TiO_2	4–8	Neutral	0.25	NO_3, Ca, Mg, Cl	1.25	NA	Photoelectrocatalytic	[43]

Table 4
Summary of different chemical and electrochemical reduction systems for bromate removal along with the treatment parameters

Electrode type	Potential (V)	Electrolyte	Reducing agent	pH	Optimum pH	C_i BrO_3 (mg/L)	Time (h)	Removal efficiency (%)	Pseudo-first- order rate (min^{-1})	Reduction mechanisms	Ref.
Activated carbon fiber (ACF) and Mg–Ca–Al(NO_3) hydrotalcite (MACF electrode)	0.5–2.0	NA	NA	6.54–6.60	NA	2–10		76	NA	Electro-adoption	[53]
Pd-rGO modified granular activated carbon (Pd-rGO/GAC) particles	0.9 mA/cm^2	NA	Pd	NA	NA	0.02	0.33	66–72	NA	Electrochemical	[49]
Reduced graphene oxide/carbon fiber paper (rGO/CFP) composites	(–)0.5–(–)2.0	NA	–	2–8	NA	1	1	99	0.105	Indirect mechanism	[50]
Pd modified carbon fiber	(–)0.5–(–)2.5	0.1–10 mM Na_2SO_4	Pd	2–10	NA	0.085	2	87	0.016–0.031	Indirect mechanism	[51]
Tin cathode	(–)1.6–(–)2.0	2 M NaCl	NA	NA	NA	12,800	3	75.60	0.00048	Electrochemical	[47]
Layered copper mesh cathode	(–)1.5–0.0	0.1 M K_2SO_4	NA	7	Neutral	0.098	0.06	66.7	0.13	Electroreduction	[48]
Boron-doped diamond electrode	(–)2–5.0	0.1 M Na_2SO_4	NA	2–9.5	4.2	20	2	90	0.007–0.025	Electroreduction	[54]
Polyaniline-modified electrode	(–)0.80–0.80	0.1 mM Na_2SO_4	NA	5–9	7.0	1,280	0.42	99	NA	Electroreduction	[55]
Activated carbon felt electrodes	1.0–10.0	5×10^{-3} mol/L H_2SO_4	NA	2–10	2.2–3.4	12.8	0.0025	90–98	NA	Electrochemical	[56]
$CeO_{2-}ZrO_2$	NA	NA	Pd	5.6	NA	52.5	0.83	>99.9	NA	Reduction	[50]
None	None	None	Sulfite	5.5–7	5.5–7.0	0.1	50.4–98.4	99	0.0044-0.035	Chemical	[46]
None	None	None	Iron(II)	5.5–8	8	0.05–0.1	0.03–9.93	99	0.00072–0.58	Chemical	
None	None	None	ZVI[a]	3–11	3.5–7	0.1	0.5	>99	0.3248	Chemical	[57]
None	None	None	Zn–Fe(II)–Al	3–12.7	5.0–11.0	49–197	0.5	100	–	Chemical	[44]
None	None	None									
None	None	None	NPZVI[a]	3.5–10.5	5–7	0.05–0.2	1	97	–	Chemical	[16]
None	None	None	$FeSO_4$	4–10	6–8	0.025	2	66	–	Chemical	[45]

[a] Zero-valent iron (ZVI), nanoparticle zero-valent iron (NPZVI).

incorporated the use of Zn–Fe(II)–Al layered double hydroxides [44], ferrous species [45,46], and nano zero-valent iron (nZVI) particles [16]. The application of Zn–Fe(II)–Al layered double hydroxides under different experimental conditions yielded near complete bromate removal at neutral pH [44]. The positive charge on the Zn–Fe(II)–Al surface initiated bromate adsorption within the interlayers of Zn–Fe(II)–Al followed by former's reduction. This system is simpler with essentially no other chemical addition and possibility of Zn–Fe(II)–Al to be immobilized onto inert surfaces thus resulting into reduced head loss and in turn energy savings for real life applications. On the other hand, several aqueous phase systems have also demonstrated bromate removal. For example, removal of bromate using $FeSO_4$ report good removal at approximately neutral pH [45,46] and lower dissolved oxygen (DO) [45]. At high DO values, the competitive reduction of oxygen will minimize bromate removal. On the other hand, the application of nano zero-valent iron (nZVI) particles for the reduction of bromate under several process variables has shown the reduction efficiency to increase with an increase in nZVI, reaction temperature, and mixing rate with pseudo-first-order kinetics [16]. The above discussion shows that iron-based reducing systems are very efficient and a careful control of process conditions can yield efficient bromate reduction-based removal under wide ranging system conditions. Also, the reduction-based removal of bromate employing several electrochemical processes has shown promising results. The respective systems report the use of both traditional metal electrodes including tin and copper [47,48] and application of novel modified electrodes [49–53]; in case of latter bromate removal was initiated via factors such as produced atomic H, higher specific surface area, positive surface charge, etc., [49–53] whereas other studies also report positive effect of process variables such as increasing bromate concentration, increasing electrical potential, neutral/acidic pH, etc., on to bromate removal using the respective electrochemical processes [48,53,54–56]. Nevertheless, some specific factors were also noted to negatively affect the process efficiency. For example, during the use of palladium-reduced graphene oxide modified carbon paper (Pd-rGO/C) cathode and Pd-rGO modified granular activated carbon (Pd-rGO/GAC) particles, the bromate removal was inhibited by the presence of dissolved organic matter and precipitates from Ca^{2+} and Mg^{2+} ions [49]. Also, in another study that used a boron-doped diamond electrode the presence of sulfate and chloride ions caused lowering in bromate removal due to their competitive adsorption onto electrode surface [54]. Similarly, the use of modified activated carbon fiber using Mg–Ca–Al (NO_3) and unmodified activated carbon fiber electrodes showed that the presence of coexisting ions caused decreased bromate removal [53]. In such systems, physical sorption by electrodes and intra-particle diffusion is followed by bromate reduction and hence any competing species cause decreased bromate removal because of competitive attachment of other ions on the modified electrode surface. On the other hand, during catalytic hydrogenation over $Pd/CeO_2–ZrO_2$ catalyst, near complete bromate reduction was observed with a positive charge on the catalyst surface favoring bromate reduction [52]. Also, the use of polyaniline modified electrode at the optimum pH of 7 (and Na_2SO_4 electrolyte solution) yielded significant bromate reduction to bromine resulting due to efficient transfer of electrons from the nitrogen atoms on the polyaniline chains [55]. The use of an activated carbon-based electrode also resulted in significant amount of bromate reduced to bromide [56]. The above discussion shows that the electrochemical processes can be successfully used to remove bromate with its effective reduction to bromide/bromine species. Furthermore, studies on removal of bromate using bio-reduction process have also been reported [58–60] and respective findings are also summarized in Table 5. Bio-reduction of bromate employing sulfur-based autotrophic and methanol-based heterotrophic microorganisms yielded near complete reduction to bromide [58]. In another study for bromate reduction by anaerobic sulfate-reducing bacteria, both *Clostridium* and *Citrobacter* genera were noted during bromate reduction in the presence of sulfate while only *Clostridium* genus was noted in the absence of sulfate [59]. In summary, the above review outlines several reduction-based systems that could be successfully used for bromate removal from the aqueous phase.

3. Conclusions

Currently, there is a growing concern on regulating the level of bromate in drinking water (both tap and bottled water) due to its adverse health effects. The issue becomes more of a concern considering the difficulty of bromate control during conventional water treatment. Hence, active research in the bromate removal area is needed with numerous advances already reported in the literature. In this review paper, we have presented and summarized findings from the respective recent work on bromate removal techniques and

Table 5
Summary of some bio-reduction systems for bromate removal along with the treatment parameters

Type of bacteria	C_i BrO_3 (mg/L)	pH	Temp (°C)	Time (d)	Removal efficiency (%)	Reference
Sulfur-based autotrophic	0.1 and 0.5	NA	30	NA	100	[58]
Methanol-based heterotrophic	0.1 and 0.5	NA	30	NA	100	
Sulfate reducing bacteria (in the presence of sulfur)	5.12	7	21 ± 1	3	96	[59]
Sulfate reducing bacteria (in the absence of sulfur)	5.12	7	21 ± 1	3	90	

have discussed the effect of various process variables on bromate removal efficiency. The adsorption process was noted to be the most common and widely studied technique. Some of the adsorbent materials used also included activated carbon, both unmodified and modified. The respective adsorbent samples were noted to have the varying specific surface area, pore size, and surface morphology. In most studies, the effect of pH, initial bromate concentration, contact time, temperature, adsorbent dose, mixing speed, and coexisting ions was investigated to get optimum bromate removal. The adsorption equilibria and kinetics were mainly predicted by Langmuir/Freundlich and pseudo-second-order models, respectively. Also, the DD and electrodialysis ion-exchange processes removed bromate ions to acceptable concentrations. Furthermore, bromate reduction can also be achieved using advanced reduction processes where ultraviolet has been used as an activating agent and mainly sulfite as the reducing agent. Furthermore, chemical reduction, electrochemical reduction, and bio-reduction-based bromate removal processes were also discussed. Hopefully, the present review work will help to initiate further interest and research studies on a very important water quality issue, that is, bromate removal.

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