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Laboratory and pilot plant studies on electrochemical pretreatment of drinking water

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ABSTRACT

Human health risks associated with drinking water chlorination disinfection have been identified, and the development of new disinfecting methods to minimize the risks with less energy and chemicals is the direction of water treatment industry. In this study, an electrochemical system was investigated for drinking water treatment in an undivided electrochemical reactor using Ti/RuO_2–IrO_2 anode and graphite felt cathode. The reactor configuration was designed in such a way that the angle of the electrodes was adjustable between 0° and 90°. It was found that the laboratory reactor (660 × 620 mm size) with electrodes positioned between 30° and 65° provided satisfactory results at a treatment capacity of 250 L/h water. A pilot plant test was conducted for treatment of Yellow River water using four laboratory-scale reactors in parallel, and the electrochemical treatment was compared with chemical disinfection using sodium hypochlorite, ozone, and potassium permanganate in the pilot plant facility. The results demonstrate that the electrochemical treatment is superior to chemical disinfection using sodium hypochlorite, ozone, and potassium permanganate in terms of removal of taste and odor and formation of disinfection byproducts.

Keywords: Drinking water treatment; Electrochemical disinfection; Chemical disinfection; Hydrogen peroxide

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1. Introduction

Water disinfection and purification are essential to protect human health and life, and the world has been facing a great pressure on clean and safe water supply due to climate change, increased environmental pollution, population, and urbanization [1–3]. Chlorination disinfection has been widely employed for drinking water treatment [4]. However, the discovery of chlorination disinfection byproducts (DBPs) such as trihalomethanes (THMs) and haloacetic acids (HAAs) and their links with increased rates of miscarriages and cancers since the late 1970s has raised a great concern regarding the safety risk of chlorination disinfection [5]. As a result, the public health risk associated with chlorination disinfection has prompted development of alternative technologies using other chemical

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disinfectants such as ozone, hydrogen peroxide, potassium permanganate, chloramine, ClO_2, physical means (UV radiation, ultrasonication, microwave), and physical–chemical means, for example, photocatalysis with titanium dioxide [2,6]. Of these disinfection techniques, ozone and UV radiation have gained acceptance in water treatment processes. However, these technologies have also been acknowledged with problems of producing different classes of DBPs, high cost in operation and maintenance, and/or less effectiveness [7]. Advanced oxidation processes (AOPs) using UV/H_2O_2, Fe^{2+}/H_2O_2, and O_3/H_2O_2 are effective to eliminate ozone recalcitrant contaminants in natural waters by increasing the concentration of hydroxyl radical ($^{\cdot}OH$) [8,9]. AOPs use a large quantity of chemicals and are mainly applied to wastewater treatment [10,11]. To date, chlorine is still the dominant water disinfectant [12,13].

Problems with water are expected to grow worse in the coming decades, and one of the solutions is the development of robust new methods of disinfecting and decontaminating water at lower cost and with less energy, chemicals dosing, and impact on the environment [2]. In this aspect, electrochemical disinfection and decontamination system is a promising alternative [14]. The electrochemical reactions involve the anodic oxidation of water to produce oxygen and cathodic reduction of oxygen to form hydrogen peroxide (H_2O_2) [7,15]. Hydrogen peroxide is a powerful oxidizing agent preventing bacterial growth in water. During the electrolysis, anodic oxidation of H_2O_2 can also take place and triggers electrogeneration of intermediate active species and radicals such as $^{\cdot}OH$, $O_2^{\cdot}$, $^{\cdot}O$, and $HO_2^{\cdot}$ [16,17]:

$$\text{Anode: } H_2O - 2e \rightarrow 1/2O_2 + 2H^+ \quad (1)$$

$$\text{Cathode: } O_2 + 2H^+ + 2e \rightarrow H_2O_2 \quad (2)$$

$$\text{Overall reaction: } H_2O + 1/2O_2 \rightarrow H_2O_2 \quad (3)$$

$$H_2O_2 \rightarrow HO_2^{\cdot} + H^+ + e^- \quad (4)$$

$$HO_2^{\cdot} \rightarrow O_2 + H^+ + e^- \quad (5)$$

$$H_2O_2 \rightarrow O_2 + 2H^+ + 2e^- \quad (6)$$

$$O_2 + e^- \rightarrow O_2^{\cdot -} \quad (7)$$

$$O_2^{\cdot -} + H^+ \rightarrow HO_2^{\cdot} \quad (8)$$

$$O_2^{\cdot -} + H_2O_2 + H^+ \rightarrow O_2 + {}^{\cdot}OH + H_2O \quad (9)$$

These intermediate active species and radicals are strong non-selective oxidants and have high abilities in inactivating a wide variety of microbes and virus and in degrading a large number of organic pollutants [18,19]. As compared to other chemical disinfection methods, *in situ* generation of oxidants in the electrochemical systems avoids transport, storage, and dosage of chemicals and disinfectants and the disinfecting effect may be adjusted in accordance with on-site demand [6,7]. In addition, the electrochemical disinfection method is cost-effective in small scales and is particularly attractive in remote and small communities [20]. These are distinct advantages of the electrochemical disinfection method [6,20]. Nonetheless, the debate has been undergoing as to whether the electrochemical system can replace the chlorination for drinking water disinfection and purification [6,20].

Poor solubility of O_2 in water, slow reaction rate of oxygen reduction (reaction (2)), and side reactions (e.g., reactions (10) and (11)) are the major factors limiting the current efficiency in oxygen electroreduction.

$$O_2 + 4H^+ + 4e^- \rightarrow 2H_2O \quad E^\circ = 1.229V \quad (10)$$

$$H_2O_2 + 2H^+ + 2e^- \rightarrow 2H_2O \quad E^\circ = 1.77V \quad (11)$$

The electrocatalytic materials and structure of both anode and cathode electrodes play a critical role. Numerous studies have been carried out on enhancing catalytic activity and selectivity of the two-electron reduction of oxygen on the surface of electrode through modification using polyaniline, anthraquinone, anthraquinone derivative, porphyrin, and noble metal [21,22]. These approaches are effective, but the long-term stability in water treatment industry is a concern [23]. To increase the utilization efficiency of oxygen, gas diffusion electrode (GDE) has been extensively investigated for production of H_2O_2 [24,25]. Such electrodes are useful for kinetics studies and produce high concentrations of H_2O_2 [25–27]. However, these systems require high concentrations of electrolytes (e.g. 0.1 M K_2SO_4 + 0.1 M H_2SO_4) and therefore are not applicable in drinking water treatment industry. Of numerous electrode materials investigated, a combination of Ti/RuO_2 anode and graphite cathode appears to be a good choice for drinking water treatment [6,20]. Drogui et al. reported a cylindrical electrolysis cell in which Ti/RuO_2 anode and carbon felt cathode were separated by an insulating diaphragm for drinking water treatment without adding chemicals and electrolytes [28]. However, it was difficult for such a configuration to obtain a high throughput in a cost-effective manner due to high flow resistance.

Treatment cost, effectiveness, efficiency, and throughput are the major factors to be considered for use of the electrochemical systems in drinking water treatment [7]. In considering these factors, we injected oxygen gas into a regular and undivided electrochemical reactor and investigated the angle of the electrodes between 0° and 90° to make a maximum contact of oxygen to produce H_2O_2. Ti/RuO_2–IrO_2 material and graphite felt were employed as the anode and cathode material, respectively. A treatment capacity of 250 L/h water was demonstrated in laboratory, and then, a pilot plant testing (1,000 L/h) was carried out for treating the water from Yellow River. Water quality parameters investigated include taste and odor, turbidity, COM_{Mn}, color, pH, $CHCl_3$, and BrO_3^-.

2. Experimental

2.1. Chemicals and materials

Graphite felt was obtained from Shanghai Qijie Carbon Material Co. Ltd (Shanghai, China). The felt thickness was 3 mm, and its specific area was 900 m^2/g. Ti/RuO_2–IrO_2 and Ti/RuO_2 were supplied by Northwest Institute for Nonferrous Metal Research (Shanxi, China). Humic acid and H_2O_2 (30%) were purchased from Beijing Chemicals and used as received.

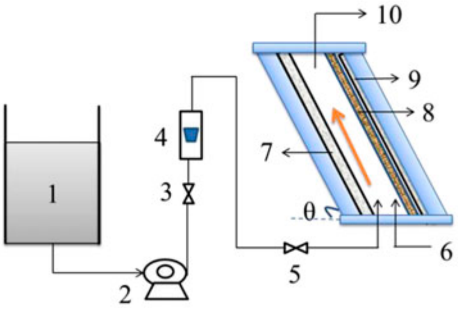


Fig. 1. Experimental setup of flow cell: (1) reservoir, (2) pump, (3) and (5) valves, (4) rotometer, (6) O_2 inlet, (7) Ti/RuO_2–IrO_2 anode, (8) graphite felt cathode, (9) current collector (Ti/RuO_2), (10) effluent. The distance between the anode and cathode was 4.4 mm, and the size of electrodes was 500 mm × 500 mm (the size of the cell was 660 mm × 620 mm; 170 holes with an internal diameter of 1 mm drilled on the bottom of the cell for oxygen aeration). The cell was placed on an adjustable frame to adjust the cell angle (θ) between 0° and 90°.

2.2. Experimental setup

Fig. 1 shows the schematic diagram of electrochemical system for drinking water treatment in the laboratory. Graphite felt cathode and Ti/RuO_2–IrO_2 anode were employed in the electrochemical cell. A Ti/RuO_2 plate was attached to the graphite felt cathode to collect the current. The flow rate of oxygen aeration was controlled to ensure that individual bubble was visible (the diameter of oxygen gas bubbles was approximately 3 mm by visual estimation). To investigate the effect of electrode position, the electrochemical cell was fixed on a frame to adjust the angle of the electrode (θ) between 0° (horizontal flow mode, the anode was underneath the cathode) and 90° (vertical flow mode).

2.3. Current efficiency and energy consumption calculation

Current efficiency (η) of hydrogen peroxide production was calculated using the following equation:

$$\eta = \frac{nFC_{H_2O_2}V}{M\int_0^t Idt} \times 100\% \tag{12}$$

where n is the stoichiometric number of transferred electrons (n = 2), F is the Faraday constant (96,485 C/mol), $C_{H_2O_2}$ is H_2O_2 concentration in a bulk solution (mg/L), and V is the electrolyte volume (L), M is the molecular weight of hydrogen peroxide (34 g/mol), I is the current (mA), and t is the electrolysis time (s). Energy consumption (Φ) for the production of 1 kg H_2O_2 (kWh/kg H_2O_2) was calculated using the Eq. (13):

$$\Phi = \frac{nFE}{3600M\eta} \tag{13}$$

and energy consumption (ψ) for the treatment of 1 m^3 water (kWh/m^3 water) was calculated using Eq. (14):

$$\psi = \frac{EIt}{3600000} \tag{14}$$

where E is the cell voltage (V), η is current efficiency (%), I is the current (A), and t is the electrolytic time (s).

2.4. Pilot plant testing

The pilot plant testing was carried out for treatment of Yellow River water in Zhengzhou, Henan

Province, China. The pilot plant process consisted of five units: electrochemical pre-oxidation, coagulation/flocculation, sedimentation, sand filtration, and activated carbon filtration (Scheme 1). The pilot plant electrochemical reactor consisted of four laboratory-scale reactors in parallel. Yellow River water was used as raw water in the pilot plant testing, and chemical and electrochemical pre-oxidation were compared by replacing the electrochemical reactor with a chemical dosing tank. The chemicals used were NaClO, O_3, and $KMnO_4$. Water quality parameters investigated were taste and odor, turbidity, COD_{Mn}, $CHCl_3$, BrO_3^- and pH. The flow rate of water and oxygen aeration was set at 1,000 L/h and 400 L/h, respectively.

2.5. Analytical methods

Hydrogen peroxide concentrations were determined by the titanic sulfate $[Ti(SO_4)_2]$ method [29] in which a spectrophotometer (UV-2,550, Shimadzu, Japan) was used to measure the light absorbance of the Ti^{4+}-H_2O_2 orange complex at 430 nm. Color, taste, and odor were determined according to GB/T 5750.4–2006 Drinking Water Sanitation Standardized Testing Method. Sample pH and turbidity were determined using pH meter Model miniLab IQ 125 and HACH portable turbidimeter Model 2100P, respectively. The concentration of humic acid was determined using by a UV spectrophotometer (UV-2450, Shimadzu, Japan) at a 253 nm. COD_{Mn} values were determined by potassium permanganate reflux method [30].

3. Results and discussion

3.1. Comparison of graphite plate and graphite felt cathodes

The material of cathode is an important factor for the electrogeneration of hydrogen peroxide, and graphite has been extensively used for cathode. Sudoh et al. reported that graphite felt had high catalytic activity for electrogeneration of hydrogen peroxide in alkaline solutions [31]. Gyenge et al. investigated the performance of graphite felt and reticulated vitreous carbon (RVC) for the electrosynthesis of hydrogen peroxide in acidic solutions and found that the graphite felt had higher current efficiency and mass transfer coefficient [32]. In this study, we employed an undivided electrochemical reactor to disinfect water flowing between the two electrodes (see Fig. 1). To compare the performance of graphite plate and felt in producing hydrogen peroxide, the cyclic voltammogram was investigated using tap water as the electrolyte and the results are shown in Fig. 2. The peak current with O_2 aeration was 5 times higher when no O_2 was aerated, and the graphite felt had a significantly higher current density than the graphite plate under the same potential. The current density sharply increased from –0.5 to –3.2 mA/cm^2 in a potential range of –0.2 and –0.55 V and then remained unchanged to –0.75 V. With a further increase of the potential, hydrogen gas evolution started. The scanning rate of cyclic voltammogram was 5 mV/s and thus, Fig. 2 is a steady-state polarization. Therefore, the current density should be maintained between –0.5 and –3.2 mA/cm^2 for electrosynthesis of H_2O_2 in a two-electrode electrolytic system.

Fig. 3 shows H_2O_2 concentration and current efficiency in a small electrochemical reactor using graphite plate and felt electrode (150 × 150 mm) in a current density range between 0.5 and 3.2 mA/cm^2. Clearly, graphite felt demonstrates better performance than graphite plate, and this is because the felt has higher specific surface area and macroporous structure of the material provides numerous active reaction sites for oxygen adsorption. The H_2O_2 concentration increased with current density and started to decrease beyond a current density of 2 mA/cm^2. This is likely due to increased tendency of hydrogen peroxide decomposition at higher potential. When the current density increased to 3.3 mA/cm^2, the current efficiency of the graphite felt decreased to 25.9%. The oxygen mass transfer coefficient obtained in this study is 3.72×10^{-4} m/s, 10 times higher than the value of 2.4×10^{-5} m/s reported by Gyenge et al. [32].

3.2. Effect of electrochemical reactor's structure

There are two types of electrochemical reactor for water treatment by electrosynthesis of hydrogen peroxide from water and O_2. One is that the electrodes are vertically placed and water flows along the electrodes (parallel-flow mode) [33], and the other one is that water flows across the electrode mesh (cross-flow mode) [28]. The factors for designing the reactor for water treatment include current efficiency, electricity consumption, utilization efficiency of oxygen, throughput, and cost. The parallel-flow mode with vertical electrode has a low efficiency of O_2 utilization, thus increasing the cost of operation. The cross-flow mode can maximize the use of O_2, but its flow resistance is also large, thus decreasing the throughput of the treatment process.

To improve efficiency of O_2 utilization and throughput, the effect of reactor configuration was investigated by adjusting the angle of the electrode from 0˚ (horizontal flow) to 90˚ (vertical flow), and the results are shown in Fig. 4. The H_2O_2 concentration increased with increasing the electrode's angle from 0˚

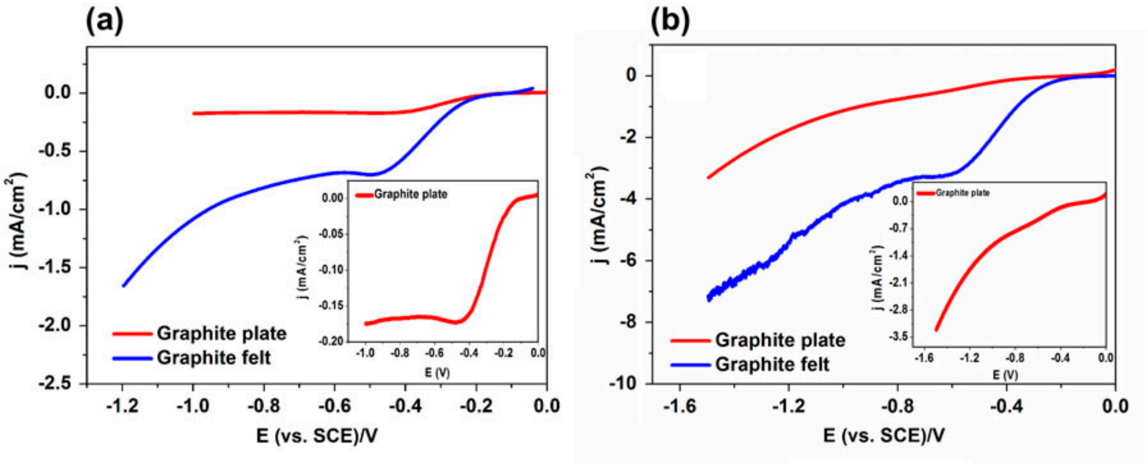


Fig. 2. Cyclic voltammogram for graphite plate and graphite felt, (a) without oxygen aeration and (b) with oxygen aeration. The volume of electrolytic cell, 20 cm^3; surface area of the graphite electrode, 0.5 cm^2; electrolyte, O_2-saturated tap water; a plastic tube of 3 mm internal diameter was placed in the vicinity of the graphite electrode to inject oxygen at a flow rate of 80 mL/min; scanning rate = 5 mV/s; 25℃.

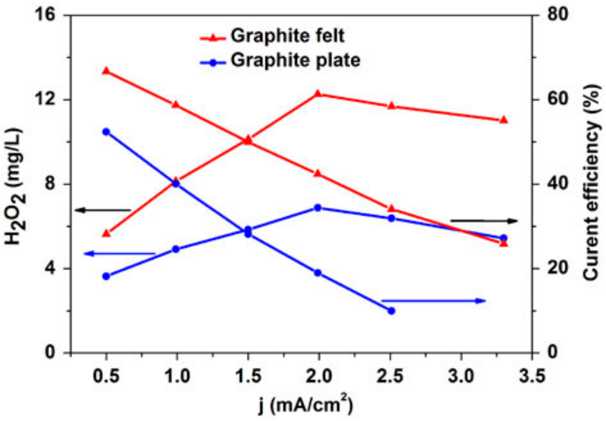


Fig. 3. Effect of carbon-based material on the H_2O_2 electrogeneration. Conditions: tap water, water flow rate 10 L/h, oxygen flow rate 8 L/h, 25℃, electrode angle 30°, cell size 150 × 150 mm (electrode size 148 × 150 mm).

and reached a maximum value at around 40° (Fig. 4(a)). The current efficiency had a similar trend. The electrogenerated H_2O_2 is obtained by reduction of O_2 at the cathode, and therefore, a continuous steady supply of O_2 at a certain concentration is important to maintain a high efficiency. Due to the low solubility of O_2 in water and decomposition (oxidation) of H_2O_2 at anode, the retention time of O_2 within the cell is important. The conventional cell with vertical electrodes (90°) does not provide a sufficient retention time and good contact between O_2 and the cathode. It was found that the anodic oxidation rate of H_2O_2 increased when the electrodes were positioned at less than 30° and the electrode position between 30° and 65° provided satisfactory performance in this work. To avoid the anodic oxidation in the undivided cell, Drogui et al. reported the use of insulating porous diaphragm in a cross-flow cell (water and O_2 flowed sequentially across the anode mesh, the diaphragm, and the carbon felt cathode) for electrogeneration of H_2O_2 for drinking water treatment [28]. However, this type of cell design creates a large resistance for water and oxygen flow and is of limited use in industrial applications.

3.3. Effect of water flow rate

The flow rate of water is an important parameter for electrogeneration of H_2O_2. Fig. 5 shows that current efficiency increases while H_2O_2 concentration decreases with increasing water flow rate. For cost-effectiveness of the process, high water flow rates and current efficiencies are recommended. It was reported that 5 mg/L H_2O_2 was sufficient to degrade typical organic pollutants and inactivate harmful cyanobacteria in raw drinking water [34,35]. In this study, a H_2O_2 concentration of 5.25 mg/L and a current efficiency of 36.5% were obtained at 250 L/h water and 120 L/h O_2.

3.4. Effect of oxygen and air aeration rates

Fig. 6 shows H_2O_2 concentration as a function of air and pure O_2 aeration rates. Without aeration, the soluble O_2 in water from air and electrogenerated O_2

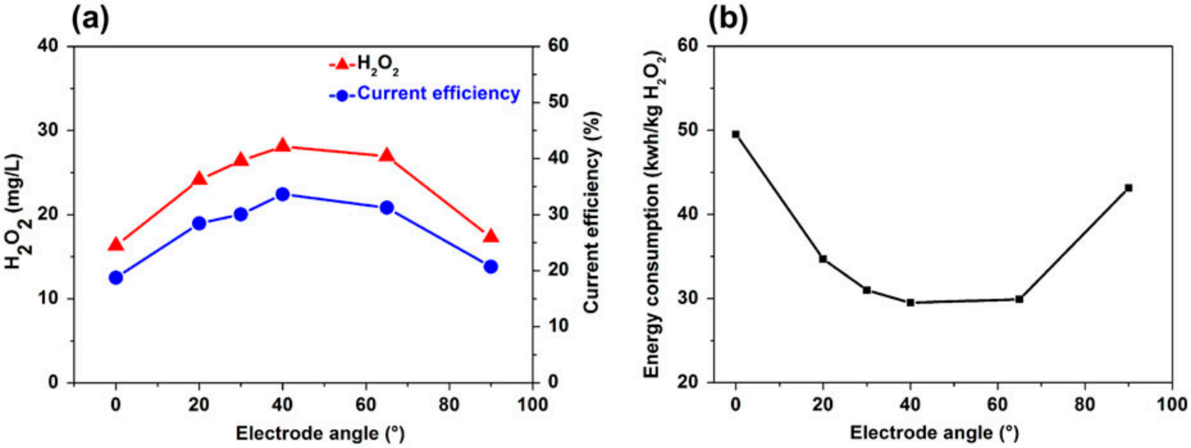


Fig. 4. Effect of electrode angle on (a) hydrogen peroxide concentration and current efficiency; and (b) energy consumption. Conditions: tap water, current density 2 mA/cm^2, water flow rate 36 L/h, O_2 flow rate 200 L/h.

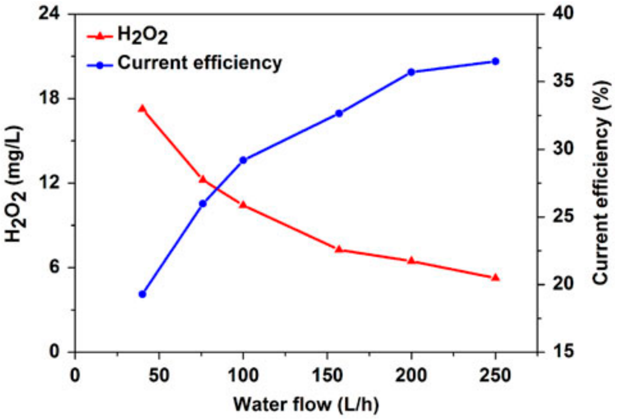


Fig. 5. Effect of water flow rate on the H_2O_2 electrogeneration. Conditions: tap water, current density 2 mA/cm^2, O_2 flow rate 120 L/h, electrode angle 40˚.

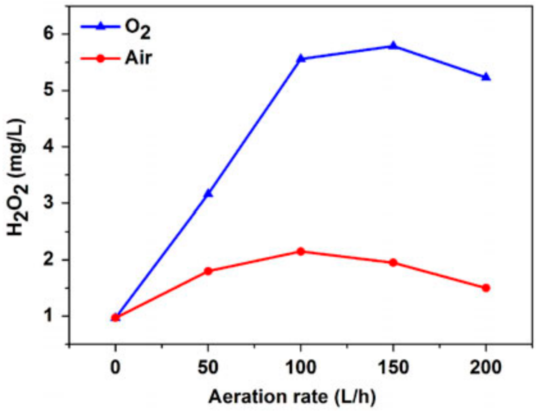


Fig. 6. Effect of gas flow rates on H_2O_2 concentration. Conditions: tap water, current density 2 mA/cm^2, water flow rate 250 L/h, electrode angle 40˚.

are the source of O_2 for cathodic reduction. In this case, the concentration of H_2O_2 produced was low (about 1 mg/L). When oxygen gas was aerated, H_2O_2 concentration increased linearly with increasing the aeration rate up to 100 L/h O_2 and remained almost unchanged between 100 and 200 L/h O_2. The O_2 aeration was likely to increase the concentration of O_2 in water and the diffusion and transfer rate of O_2 to the surface of the cathode and therefore enhanced the formation of H_2O_2. However, higher aeration rates can also increase the population of gas bubbles in water (i.e., the resistance of the solution). The cell voltage was 4.98 V at the O_2 aeration rate of 100 L/h and increased to 5.66 V at 200 L/h. Under higher cell voltage, O_2 is further reduced to water via a four-electron reduction pathway and H_2O_2 is reduced to water (reactions (10) and (11)). It was noted that the H_2O_2 concentration was significantly lower when the aeration was carried out with air. Therefore, a 100 L/h O_2 aeration rate was employed in subsequent experiments, and the gas-to-liquid ratio was 0.4 for a water flow of 250 L/h. This ratio was much lower than the ratio of 1.2–5 reported in the literature [36,37].

3.5. *Effect of current density*

The effect of current density on H_2O_2 concentration and current efficiency is shown in Fig. 7. The concentration of H_2O_2 was 3 mg/L at a current density of 1 mA/cm^2 and increased to a maximum value of 5.55 mg/L at 2 mA/cm^2. The current efficiency was 45% at 1 mA/cm^2 and decreased with increasing the

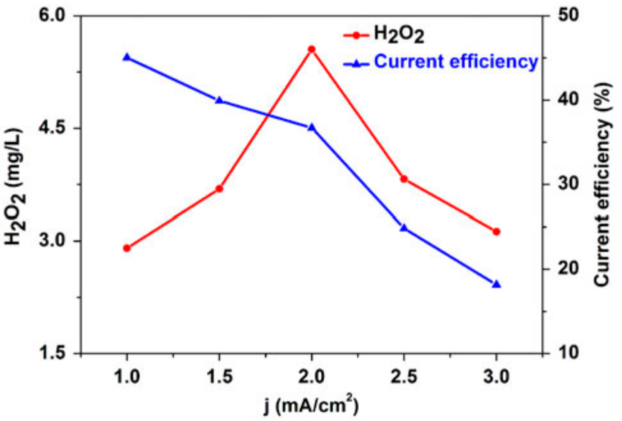


Fig. 7. Effect of current density on H_2O_2 electro generation. Conditions: tap water, water flow rate 250 L/h, O_2 flow rate 100 L/h, electrode angle 40˚.

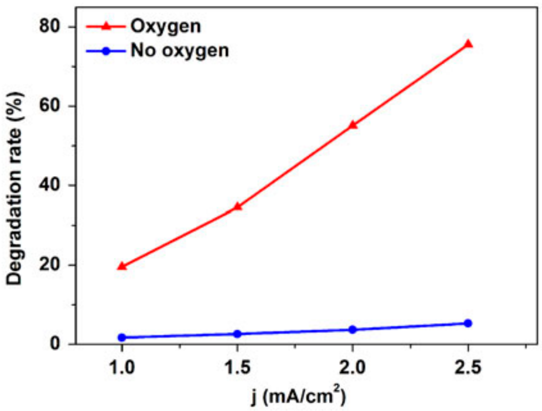


Fig. 8. Effect of current density on the degradation of humic acid. Conditions: humic acid concentration, 20 mg/L; water flow rate, 12 L/h; O_2 flow rate, 10 L/h; electrode angle, 40˚; cell size, 150 × 150 mm.

current density. At 2 mA/cm^2, the current efficiency was 36.7%. Drogui et al. obtained a current efficiency of 21% at 11.3 mA/cm^2 in electrochemical disinfection of municipal sewage [28]. The decrease of H_2O_2 concentration above 2 mA/cm^2 was due to O_2 reduction to water (reaction (10)), H_2O_2 reduction (reaction (11)) and H_2O_2 oxidation at the anode (reactions (4)–(6)). The electricity consumption for H_2O_2 production was 21 kWh/kg under the condition of 2 mA/cm^2, 100 L/h O_2 aeration rate, and 250 L/h water flow rate. Badellino et al. obtained the electricity consumption of 34.7 kWh/kg for production of H_2O_2 by electroreduction of oxygen using RVC cathode, 0.3 M K_2SO_4 electrolyte solution (pH = 10), and a N424 Nafion membrane [36].

3.6. Degradation of humic acid

Humic substances, which mainly consist of humic acids, are the major precursor of DBPs (trihalomethane) with the chlorination disinfection process. These DBPs have been identified to be carcinogen [38,39]. Removal of humic acid prior to chlorination has proved to be an effective measure to eliminate or minimize the formation of chlorinated DBPs [40]. It was found that the degradation rate of humic acid increased with the current density and 20–80% of 20 mg/L humic acid was degraded at the current density of 1–2.5 mA/cm^2 after 10 min when the system was aerated with oxygen (Fig. 8). By contrast, the degradation of the same concentration of humic acid by dosing H_2O_2 at 20 mg/L was 12.4%. The higher degradation by electrochemcial treatment was due to a combination of H_2O_2 oxidation, intermediate active species ($^{\cdot}OH$, $O_2^{\cdot-}$, $HO_2^{\cdot-}$, see reactions (7)–(9)), and direct anodic oxidation. This is a typical advantage of the electrochemical water treatment as compared to the external dosed H_2O_2 water treatment. The intermediate active species are strong non-selective oxidants and react with organic pollutants quickly [18].

3.7. Formation of DBPs

The formation of DBP $CHCl_3$ was compared between the chlorination and electrochemical treatment for water samples from Sanjiadian reservoir in Beijing. This reservoir is a secondary source of drinking water for Beijing. The water sample was dosed with 1.5–5 mg/L NaClO, and then after 30 min of the dosing, the concentration of $CHCl_3$ was determined to be 67.9–113.1 μg/L. In comparison, $CHCl_3$ in the sample was less than 6 μg/L by electrochemical treatment at 0.5–2 mA/cm^2 current density.

3.8. Disinfection

The raw water was treated by the proposed electrochemical reactor at a current density of 2 mA/cm^2. A different sample of the raw water was also dosed with commercial H_2O_2 at 10 mg/L. Then, these treated water samples were incubated for 48 h. The total number of bacteria was counted to be 106 and 2,400 for electrochemical treatment and commercial H_2O_2 dosing treatment, respectively (Fig. 9). The commercial H_2O_2 treatment and the electrochemical treatment were also compared for other parameters of water quality (Table 1). The electrochemical treatment is superior in terms of odor, turbidity, UV_{254} compounds, color, and disinfection.

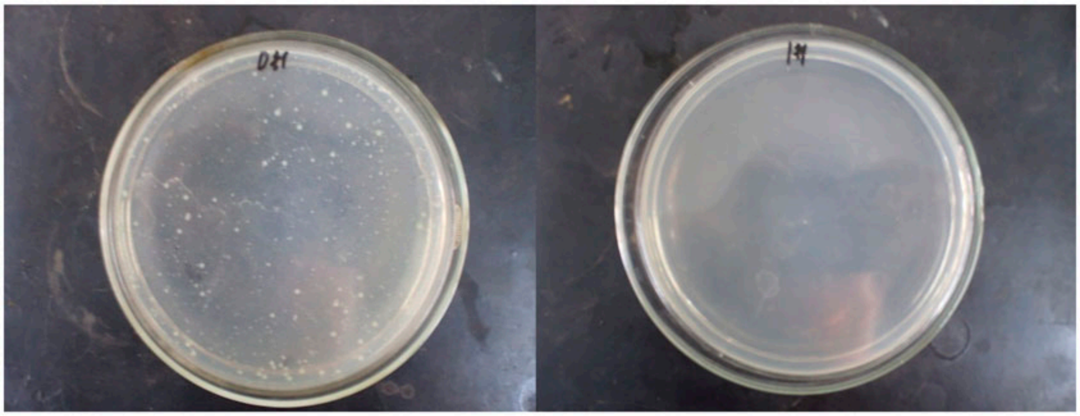


Fig. 9. Comparison of total number of bacteria of (0#) H_2O_2 oxidation; (1#) electrochemical oxidation. Total number of bacteria: 0# 2,400; 1# 106.

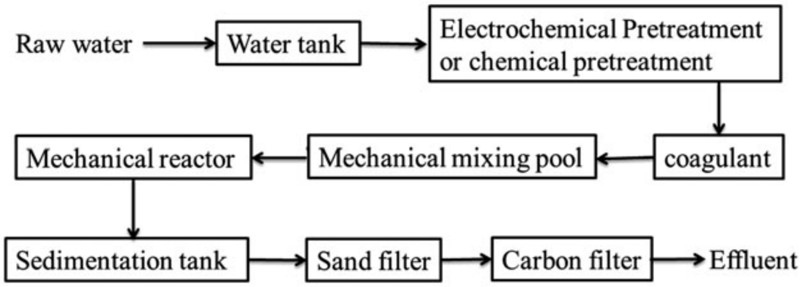
Table 1
Comparison of water quality between chemical treatment (H_2O_2 dose) and electrochemical treatment

Water quality parameter	Raw water	Chemical treatment with H_2O_2 dose H_2O_2 concentration (mg/L)				Electrochemical treatment Current density (mA/cm^2)			
		2	4	6	8	1	1.5	2	2.5
Taste and odor (Grade)	3	3	3	3	3	2	1	0–1	0–1
Turbidity (NTU)	6.44	4.96	4.7	4.5	4.25	1.35	1.22	1.02	0.74
UV_{254} (cm^{-1})	0.112	0.11	0.108	0.105	0.102	0.071	0.064	0.059	0.054
Number of bacteria*	>1,000	>1,000	>1,000	>1,000	>1,000	125	98	68	46
Color (mg Pt/L)	25	24.1	23.2	22.1	19.7	17	15	12.5	7.5

*Incubated for 48 hrs at 25℃.

3.9. Pilot plant test

The pilot plant test was carried out for treatment of Yellow River water in Zhengzhou (Henan Province) at a water flow rate of 1,000 L/h. The pilot plant process is illustrated in Scheme 1. The chemical disinfection using NaClO, O_3, and $KMnO_4$ was also conducted in the pilot plant test, and the results are shown in Table 2. The chlorination with NaClO resulted in the formation of $CHCl_3$ at 108 μg/L, while the ozone pretreatment produced BrO_3^- of 0.025 mg/L, which is greater than the drinking water standard (0.01 mg/L) by 2.5 times. Clearly, the electrochemical treatment avoided the formation of $CHCl_3$ and BrO_3^- and had excellent removal of taste and odor as compared to chemical treatment using NaClO, O_3, and $KMnO_4$. It was found that the electrochemical



Scheme 1. Schematic diagram of the pilot plant setup.

Table 2
Water quality comparison of chemical and electrochemical pretreatment in the pilot plant test

Water quality parameter	Raw water[a]	Electrochemical or chemical pretreatment				Effluent water				Drinking water standard
		Electro[b]	NaClO[c]	O_3[d]	$KMnO_4$[e]	Electro	NaClO	O_3	$KMnO_4$	
Taste and odor (Grade)	3	1	3	3	3	0	2	1	2	0
Turbidity (NTU)	10.52	8.73	9.47	9.62	12.46	0.19	0.27	0.14	0.14	3
COD_{Mn} (mg/L)	2.78	2.00	2.37	2.22	2.71	0.89	1.23	1.22	1.52	3
Color (mg Pt/L)	12.5	7.5	12.5	9.38	12.50	<5	<5	<5	<5	15
pH	8.22	7.85	8.15	8.21	8.19	7.81	8.07	8.04	8.10	6.5–8.5
$CHCl_3$ (μg/L)	1.81	1.29	107.95	ND	ND	<0.3	57	ND	ND	60
BrO_3^- (mg/L)	<0.004	<0.004	ND	0.025	ND	<0.004	ND	0.007	ND	0.01

ND—Not determined.

[a] The water contained 0.25 mg/L Br.

[b] The current density of the electrochemical treatment was 1.5 mA/cm.

[c] The concentration of NaClO was tested between 0.45 and 2 mg/L, and the data were the result of 1.45 mg/L NaClO.

[d] The retention time was 15 min, and O_3 concentration was 0.5–2 mg/L (the data were the result of 1.36 mg/L O_3).

[e] The concentration of $KMnO_4$ was 0.2–7 mg/L, and the data were the result of 2.32 mg/L $KMnO_4$.

Table 3
Electrolysis energy consumption of pre-oxidation

Current (A)	Current density (mA/cm^2)	Cell voltage (V)	Energy consumption (kWh/m^3)
1.25	0.5	11.35	0.014
2.50	1.0	13.17	0.033
3.75	1.5	16.35	0.061
5.00	2.0	18.7	0.093
6.25	2.5	21.3	0.133

performance declined gradually during the water treatment and the major cause was fouling of electrodes (especially for cathode). Kerwick et al. reported that reversing the polarity of the electrodes was effective to clean the fouling of electrodes in electrochemical drinking water treatment [41]. In this study, we reversed the polarity of the electrodes after 5–7 d operation at a current density of 3.2 mA/cm^2 for 5–10 min and found that the reversed polarity of the electrodes was able to recover the performance to the initial stage of the reactor after 30 d continuous operation.

The electricity consumption for the treatment of 1,000 L/h water is shown in Table 3 and can be readily adjusted by controlling the current of the electrolysis in accordance with the quality of water to be treated. Theoretically, oxygen consumption was estimated to be 4 L per tone of the Yellow River water treated. However, the actual use of oxygen was 400 L/ton water. Therefore, the utilization efficiency of oxygen is 1%. Oxygen may be collected at the top of the reactor and re-injected to the reactor to reduce the operation cost of the process.

4. Conclusions

Laboratory and pilot plant studies carried out in this work have demonstrated that electrochemical water treatment technology offers distinct advantages of no added chemicals, better removal of taste and odor and organic pollutants, no formation of DBPs, and other comparable water quality parameters for the Yellow River water as compared to conventional chemical disinfection methods (chlorination, ozonation, and $KMnO_4$). Electrochemical disinfection and decontamination system for water treatment is proved to be a green and sustainable process. For the electrochemical technology to replace conventional chlorination in drinking water industry, further evidence regarding inactivating a wide variety of microbes and viruses and long-term performance of the process is warranted.

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