Ex-situ Electrochemical Disinfection with the PbO₂ Anode

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This research investigated application of tap water prepared by the electrochemical method with the PbO₂ electrode in air-conditioner ex-situ disinfection. Influences of different factors on disinfection effects were analyzed. Results show chloride concentration, electro-conductivity, voltage and bacteria species affected disinfection performance conspicuously but pH and the time after electrolysis presented little effects on it. Total bacteria elimination with the method was quick and could be achieved, under specific conditions, in 50 s and 60 s for Escherichia coli (E. coli) and Staphylococcus aureus (SA), respectively. In addition, erosion was not observed on air conditioner surface. This method was found to possess the anti bacteria re-growth ability after disinfection. Overall energy consumption was evaluated as 1.336 kWh·m⁻³ and considered economic in application. This method presents a promising approach for air-conditioner disinfection.

Keywords: ex-situ electrochemical disinfection; air conditioner disinfection; tap water; lead dioxide anode; active chlorine

1. INTRODUCTION

Air-conditioner disinfection has attracted much attention recently [1, 2]. Bacteria bred in air-conditioners severely threaten human health [3]. Possible methods are divided into three categories: 1) chemical means such as air-conditioner cleaning agents and ozone, 2) physical-chemical means including photo-catalysis, and 3) physical means containing ultraviolet irradiation [4]. However, when applied in air-conditioner disinfection these methods are accompanied by some technical problems as following. Bacteria in corners and gaps of heat exchangers are difficult to inactivate. The bacteria elimination process should be quick, durable and inactivate a wide range of bacteria without corrosion
on metal and plastic surface. Some methods are not equipped with bacteriostasis after disinfection but temperature and moisture in the air-conditioner are suitable for bacteria recovery after bacteria-elimination. In addition, the bacteria-inactivation system should be cost-efficient and easily operated for practical application. Air-conditioning cleaning agents have defects of erosion and toxicity [5]. Half-life period of ozone is short and it is very unstable [6]. Ultraviolet disinfection needs long time and endangers human [7]. Action range of photo-catalysis is relatively small and is related with high costs [8]. Based on defects above, a novel method denominated as ex-situ electrochemical disinfection with tap water and the PbO$_2$ anode, which can satisfy demands of air-conditioner disinfection, is presented in the research.

The electrochemical method has been raised as an important alternative for chlorine disinfection [4]. Compared with addition of chlorine products, electrochemical disinfection presents conspicuous advantages including its low toxicity to environment, low operation costs and high capacity to inactivate a wide range of microorganisms [4, 9, 10]. One of commonly used electrochemical methods is electro-chlorination [11]. Active chlorine [12, 13] including Cl$_2$, HOCl, OCl$^-$ and ClO$_2$ are considered as main disinfectants inactivating microorganisms in electrochlorination [14]. Literatures report that microorganisms could be inactivated effectively in chloride-containing water by electro-chlorination [11, 15-18]. However, over high concentration of chloride can result in corrosion and production of excessive harmful by-products, which is not suitable for air-conditioner disinfection and public health [4]. Tap water contains relatively low chloride contents so that it might cause little erosion on metal and plastic and be suitable as electrolyte for electrochemical disinfection in the air-conditioner. In addition, tap water is cost-efficient, which could make application more economic.

Electrochemical methods are traditionally adopted for in-situ disinfection [4, 19]. However, in-situ methods can only be adopted to disinfect bacteria in water or fluid but can’t be applied to inactivate microorganisms on solid surface, in corners or in intervals, for example, in the air-conditioner. Based on these defects above, the ex-situ electrochemical disinfection was proposed in this article against the traditional way. In the ex-situ process, procedure sequences are arranged as electrolysis, electrolyte transfer and the washing on surface of heat exchangers.

For ex-situ air-conditioner disinfection, appropriate electrodes with high electrochemical activity producing active chlorine, long service life and low costs are essential [11]. Popular anode materials for electrochemical disinfection are dimensionally stabilized anodes (DSA), the Pt electrode and boron-doped diamond (BDD) electrodes [11, 13, 19-22]. Compared with Pt and BDD electrodes, DSA electrodes are considered to be easy-prepared, cost-efficient and active-chlorine-productive [11, 19, 20, 23]. Besides, harmful byproducts like chlorate and perchlorate are not generated at DSA [24]. Among those reported DSA electrodes, the PbO$_2$ electrode is regarded as the optimum electrode for its high chemical stability in corrosive media, relatively high electrochemical activity, long life-span and low price compared with noble metal electrodes [23, 25, 26]. Therefore, the PbO$_2$ electrode might be suitable as the experimental anode in ex-situ air-conditioner disinfection.

The objective of this research was to study performance of ex-situ electrochemical disinfection with tap water and the PbO$_2$ electrode in the air conditioner. Chloride concentration, electroconductivity, pH, voltage, the time after electrolysis, bacteria species and complete bacteria-
elimination time were considered. Besides, erosion on air conditioner surface, bacteria re-growth after disinfection and energy consumption were also investigated.

2. MATERIALS AND METHODS

2.1. Electrodes

The PbO$_2$ electrode was adopted as the anode. The PbO$_2$ layer was formed on the titanium sheet by the co-deposition method previously described [23, 27]. The coated substrate area was \~6 cm$^2$ with a catalysis loading of \~1.10 mg cm$^{-2}$. The Cu electrode was adopted as the cathode which possessed same dimension with the anode. Both electrodes were sealed and rinsed by sonication in de-ionized water before use.

2.2. Reagents

All reagents were of analytical grade. Sodium chloride, sodium sulfate, sodium hyposulfite, absolute ethyl alcohol, extractum carnis, peptone and powdered agar were purchased from Sinopharm Chemical Reagent Co. Ltd. De-ionized water was obtained from a super-pure water preparation system (EPED-40TF, China). Tap water (initial chloride concentration of 4.36-15.74 mg/L, initial electro-conductivity of 125-1750 μS/cm, initial pH of 7.5-8.0) was obtained from the distribution system in Xi’an Jiaotong University and sterilized before experiments by ultraviolet radiation. Physiological saline was achieved with de-ionized water and sodium chloride. The colorimetric test kit used for measuring active chlorine was purchased HKM Co. Ltd.

2.3. Bacteria

In this research Escherichia coli (EA; ATCC8739) and Staphylococcus aureus (SA; ATCC6538) were employed as indicator bacteria because they can represent gram-negative bacteria and gram-positive bacteria in the air-conditioner, respectively. Preparation of bacteria was based on the literature [9, 28, 29]. Bacteria were first cultured at 37 °C for 16 h from stock suspension in 50 mL of nutrient broth in the constant temperature oscillator (SHA-C, Changzhou Guohua Co., China). Nutrient broth included extractum carni 3.0 g, sodium chloride 5.0 g and peptone 10.0 g in 1 dm$^3$ de-ionized water, which was corrected to pH 7.0 by the precise pH meter (PHS-3C, INESA Scientific Instrument Co., China) and autoclaved at 121 °C for 20 min in the vertical high pressure steam sterilizer (YX-280A, Shanghai Sanshen Co., China). Bacteria were harvested by centrifugation at 4000 rpm for 5 min (TD52, Hunan Kaida Co., China) and washed three times with sterile physiological saline before re-suspension in sterile physiological saline. Turbidity of acquired bacteria suspension was measured at 600 nm by the ultraviolet-visible spectrophotometer (Agilent 8453, Agilent Co., USA) to adjust initial population to approximately 1×10$^8$ CFU/mL. Before experiments concentration of bacteria suspension was diluted from 1×10$^8$ CFU/mL to 2 × 10$^3$ CFU/mL.
2.4. Equipments

The experimental installation consisted of the electrolytic part and the washing-disinfection part (Fig.1). In the electrolytic part, a 1000 mL beaker was adopted as the electrolytic cell and electrode distance was set as 1 cm. The magnetic stirrer (DCG-C, Gongyi Yingyuyuhua Co., China) was set under the beaker. Electricity was supplied by a direct current power source. In the washing-disinfection part, a pipe with the diameter of 0.8 cm was adopted to transfer water. The peristaltic pump (YZ1515X, Baoding LongerPump Co., China) revolution was set 30 rpm and the flux through the pipe was 2.1 mL/s accordingly. Several holes were penetrated on the pipe for washing. Multi layer aluminum sheets acquired from heat-exchangers were adopted as bacteria supporters, which were used to simulate practical conditions of the air-conditioner. All supporters were used after sterilizing and set vertical when being washed. All experiments were conducted after ultraviolet treatment and on the sterile experiment platform (C7-1D, Tianjin Taisite Co., China).

![Figure 1. Schematic view of experimental set-up (A: Electrolyte B: Electrolytic cell C: Electrodes D: Pump E: Pipe with penetrated holes F: Multi layer supporter)](image)

2.5. Procedures

Factors including chloride concentration, electro-conductivity, pH, voltage, the time after electrolysis were firstly studied in this research. Then washing time for total bacteria-elimination and effects of bacteria species were discussed. Finally the erosion accelerated experiments and bacteria regrowth experiments were conducted. Quantities of active chlorine were measured at the same time. Active chlorine was quantified by N,N-diethyl-p-phenylenediamine (DPD) using a colorimetric test kit (DPD WATER TEST HKM; limit of detection LOD=100 μg/L Cl2).

Prior to experiments, bacteria should be attached on supporters. 1 mL of the \(2 \times 10^3\) CFU/mL bacteria suspension was injected in intervals of supporters. Electrolysis was conducted when moisture on surface evaporated, after which electrolyte was pumped out through the pipe to wash supporters. When washing was finished, supporters were immersed in sterile physiological saline and shaken to ensure all bacteria on supporters diffused into liquid. Excess Na2S2O3 was added to eliminate residual disinfectants in acquired suspension which was sampled for plate spreading. Three replicates of sampled 0.10 mL suspension were spread on three agar medium plates grown at 37 °C for 24 h for
counts in the water jacket incubator (GHP-9080, Shanghai Yiheng Co., China). Agar medium plates included extractum carni 3.0 g, sodium chloride 5.0 g, peptone 10.0 g and powdered agar 18.0 g in 1 dm$^3$ de-ionized water, which were corrected to pH 7.0 and autoclaved at 121 °C for 20 min.

Blank groups were made in this research. Three replicates of 0.1 mL diluted $2 \times 10^3$ CFU/mL bacteria suspension was directly spread on three agar medium plates grown at 37 °C for 24 h for calculation of elimination rates. This group was named as the blank group. Elimination rates were calculated by the formula:

$$\text{ER} = \frac{N_x}{N_0}$$  \hspace{1cm} (1)

Where ER is the elimination rate, $N_x$ is the bacteria count in washing experiments, $N_0$ is the bacteria count in the blank group.

In the erosion study, accelerated erosion experiments were conducted on aluminum sheets and ABS (Acrylonitrile Butadiene Styrene) plastic sheets taken from the air-conditioner. Generally, aluminum and ABS are important components of air conditioners. They were immersed in 100 mg/L NaCl solution for electrolysis. Experimental duration was 168 hours and surface of them was observed by the optical microscope (Axio Lab.A1 M, Beijing Precise Instrument Co., China) before and after experiments.

Bacteria re-growth experiments were carried out after bacteria on supporters were completely eliminated by ex-situ electrochemical disinfection. Supporters were then laid under the air out-let of the air-conditioner. Experimental duration was 10.5 hours. Same amounts of dust were sampled from surface every 1.5 hours and were spread on agar medium plates grown at 37 °C for 24 h for counts. Before experiments, the same amount of sample bacteria was obtained for calculation of bacteria re-growth percentage:

$$\text{BRP} = \frac{N^*_x}{N^*_0}$$  \hspace{1cm} (2)

Where BRP is bacteria re-growth percentage, $N^*_x$ is bacteria counts in experiments, $N^*_0$ is bacteria counts before complete bacteria-elimination.

All experiments in this article were repeated three times so that all results in this research were reproducible and convincible. Errors in this study were presented by error bars in figures.

3. RESULTS AND DISCUSSION

3.1. Effects of chloride concentration

Effects of chloride concentration on disinfection are shown in Fig.2a. NaCl in this research was regarded as indicator substance for chloride in tap water and the experimental chloride range (0.5-15 mg/L) was close to the chloride content range of tap water in Xi’an Jiaotong University. Results show that the bacteria elimination rate increases from below 60% to over 90% for both kinds of bacteria with the NaCl concentration of 0.5-15 mg/L (Fig.2a). In Fig.2b, production of active chlorine also increases with NaCl concentration. By comparison of Fig.2a and Fig.2b, increasing trends of the elimination rate and the active chlorine concentration were very similar. Active chlorine is thought to be the substance
that could disrupt bacteria membrane by strong oxidation [4]. Active chlorine can be generated by reactions as follows [4, 19]:

\[
\begin{align*}
2\text{Cl}^- & \rightarrow \text{Cl}_2 + 2\text{e}^- \\
\text{Cl}_2 + \text{H}_2\text{O} & \rightarrow \text{HClO} + \text{HCl} \\
\text{HClO} & \leftrightarrow \text{ClO}^- + \text{H}^+ \\
\text{HClO} & \rightarrow 0 + \text{Cl}^- + \text{H}^+ \\
\text{ClO}^- & \rightarrow 0 + \text{Cl}^- 
\end{align*}
\]

In this experiment, higher chloride concentration provided more chloride ions and enhanced electrolytic reactions above so that more active chlorine was produced [30]. Effects of reactive oxygen species (ROS) could be neglected because of its rather short life span compared with the water-transferring time from the electrolytic cell to washing. Effects of direct contacts of bacteria with electrode could also be ignored because the disinfection part was separated from the electrolytic part. Thus reasons for the rising elimination rate could be attributed to the growing production of active chlorine in the study. Therefore, the increase of chloride had facilitating effects on the bacteria elimination rate for both bacteria and the total bacteria elimination could be achieved by simply increasing chloride concentration.

![Graph A](image1)

**Figure 2.** a) Effects of chloride concentration on disinfection (Washing time: 5 s); b) Variations of active chlorine with NaCl concentration. (Electrolytic time: 10 min; Electrolyte: NaCl solution; Voltage: 20 V; Electrolytic volume: 1 L)
3.2. Electro-conductivity

Effects of electro-conductivity on disinfection are presented in Fig. 3a. Electro-conductivity was set from 125 to 1250 μS/cm. The electro-conductivity range in this experiment was close to that of tap water in Xi’an Jiaotong University. Electro-conductivity was measured and determined by the conductivity meter (DDSJ-308A, Shanghai Leici., China).

Figure 3. a) Effects of electro-conductivity on disinfection (Washing time: 5 s); b) Variations of active chlorine with electro-conductivity. (Electrolytic time: 10 min; the electrolyte consisted of 5mg/L NaCl and Na₂SO₄ and electro-conductivity was adjusted by Na₂SO₄; Voltage: 20V; Electrolytic volume: 1 L)
It is observed both bacteria elimination and active chlorine increase greatly with electrodemand conductivity of 125-1250 μS/cm (Fig.3a). For both kinds of bacteria, the total bacteria elimination could be achieved at 1000 μS/cm. In Fig.3b, production of active chlorine also increases with electrodemand conductivity. Alike with Fig.2, the increasing trends of the elimination rate and active chlorine production in Fig.3 were also similar. It is considered higher electro-conductivity gave rise to higher applied current density. An increase of current density influenced mass transfer, kinetics of charge transfer [30], which means Cl⁻ were transported and discharged better and production of active chlorine was accelerated and its amount was increased [31]. In the same way, effects of ROS and direct contacts with electrodes could be neglected so that active is the key factor in this experiment. Therefore, the increase of electro-conductivity facilitated the bacteria elimination rate for both bacteria and the total bacteria elimination could be achieved by simply increasing electro-conductivity.

3.3. Effects of pH

Effects of pH on disinfection are shown in Fig.4a. It is observed that for E. coli the bacteria elimination rate stabilizes at about 83% and for SA the rate stabilizes at about 87% as pH varies (Fig.4a). According to (3)-(7), hypochlorous acid and hypochlorite anions formed a pH-dependent equilibrium [19]. Under acid conditions the most predominant active chlorine species is the powerful disinfectant hypochlorous acid, as opposed to the less powerful hypochlorite ions which dominate under alkaline conditions [32]. Fig.4b shows active chlorine under acid conditions was higher than that under alkaline conditions. Thus the activation should be more severe in acidic pH but experimental results show bacteria-elimination rate changed less and some other mechanisms should counteract the effects above. The pH variation in a single solution was not considered as counteraction effects. The pH values were measured right after electrolysis. Results show little change between the measurements before and after electrolysis, which was caused by relatively equivalent amounts of hydroxyl anions and hydrions formed on both electrode surfaces [18] in spite of oxidation of some amounts of chloride on the anode. Hence compositions of formed active species would not be changed by pH variation in a single solution. The adaptive faculty of both bacteria to various pH values also was not thought to play a role because 7.5 is the premium value for reproduction of both bacteria and their survival pH range is close to the experimental pH range. One possible explanation for the question is the relatively low amount of chloride in tap water. With the same amount of chloride, compositions of the produced active chlorine were not changed obviously, which is shown in Fig.4b. Another possible explanation is the influence of scouring, which might dilute effects of active chlorine and thus effects of pH. Therefore, pH had little effects on the bacteria elimination rate in the study.
Figure 4. a) Effects of pH on disinfection (Washing time: 5 s); b) Variations of active chlorine with pH. (Electrolytic time: 10 min; pH was adjusted by adding HCl or NaOH; Electrolyte: tap water; Voltage: 20 V; Electrolytic volume: 1 L)
3.4. Effects of voltage

Effects of voltage on disinfection are presented in Fig.5a. Voltage was set from 5 to 25 V. It is observed for E. coli the bacteria elimination rate increases from 80 % to 87 % and for SA the rate...
increases from 86% to 92% (Fig.5a). In Fig.5b, the active chlorine production also increases with voltage. In this experiment, the increased voltage resulted in increased current density and higher electrical charge [33]. Accordingly, reactions in equations (3)-(7) were intensified and more amount active chlorine was produced [30, 31]. However, in Fig.5a, as the voltage increases, the increasing trend of the elimination rate becomes slower, which is similar to the trend of active production in Fig.5b. This was caused by the fixed chloride amount in electrolyte. The maximal active chlorine production was determined by the chloride amount. As the active chlorine production approaches its limitation, its increasing curve will be smoother, so is the increasing curve of the elimination rate. Therefore, the increase of voltage could facilitate the bacteria elimination but the total elimination could not be achieved by simply increasing voltage.

3.5. Effects of the time after electrolysis

Effects of the time after electrolysis are shown in Fig.6a. Prior to washing experiments, electrolyte solution was placed in static condition after electrolysis. The static time was set as 1, 2, 5, 10 and 15 minutes, respectively. It is observed that the elimination rate for E. coli stabilizes at about 80% and that for SA stabilizes at about 87% as the time after electrolysis increases (Fig.6a). These phenomena could be attributed to the relative long life of active chlorine [4, 30], which could be proved in Fig.6b. In Fig.6b, the concentration of the formed active chlorine shows little change after electrolysis. It is indicated that active chlorine still possessed the same strong ability to disinfect microorganisms after the static time. Therefore, the time after electrolysis had little effects on bacteria elimination and the ex-situ electrochemical method was proved to be equipped with durable disinfection ability, which is a great advantage over other disinfection methods.
3.6. Washing time needed for total elimination and effects of bacteria species

Washing time needed for the total elimination of E. coli and SA is presented in Fig. 7. In the experiments, washing time was set from 3 to 70 s. Results show for both bacteria the elimination rate increases with washing time (Fig. 7). The total elimination for E. coli was achieved at 60 s and that for SA was achieved at 50 s. In other words, elimination effects on SA were more effective than those on E. coli in these experiments. Theoretically, SA is considered to be wrapped up with thicker and compacter cell wall compared with E. coli [34, 35]. In experiments, thicker cell wall could make bacteria less vulnerable to produced strong oxidants-active chlorine so that the membrane integrity of E. coli was easier to be disrupted and accordingly massive cell kills and lyses were brought. Seemingly more efficient E. coli elimination should be achieved. However, it is against the experimental outcomes and thus some mechanisms should counteract the phenomena. Surface characteristics of bacteria were considered to play a big counteraction role. Surface of E. coli is full of flagella, whereas SA presents the absence of those on its surface [34, 35]. Accordingly, E. coli could control its motion through flagella and possesses stronger adhesion onto supporters than SA, which could make E. coli less likely to be scoured away compared with SA. Therefore, bacteria species had great effects on the bacteria elimination rate.

Compared with traditional in-situ electrochemical disinfection, bacteria elimination with the ex-situ electrochemical method is quicker because not only oxidation of active chlorine functions in the process but also washing effects take an important role in eliminating bacteria. Besides, two advantages could be raised from this research section. Firstly, this method offers more effective
disinfection because it could eliminate a wide range of microorganisms in a short time. Second, this method could eliminate bacteria in corners and intervals where other methods could not function.

![Figure 7](image)

**Figure 7.** Effects of Washing time on disinfection for E. coli and SA (Electrolytic time: 10 min, Electrolyte: tap water, Voltage: 20 V, Electrolytic volume: 1 L)

3.6. Erosion study

Surface conditions of aluminum and ABS plastic before and after accelerated erosion experiments were illustrated in Fig.8. Pitting corrosion is not observed on aluminum surface, which could be mainly attributed to pH neutral state of tap water [36]. When aluminum was in the pH neutral environment, a compact layer of Al₂O₃·3H₂O would be formed on its surface preventing further contacts between active aluminum and surroundings [37]. Besides, the amount of chloride in this experimental section was low that it could not dissolve the oxide film [36, 37]. Also from Fig.8, erosion is also not observed on ABS plastic surface, which could be attributed to its strong anti-erosion ability [38]. Thus it could be concluded electrolyzed tap water would not damage the aluminum and plastic surface for a long time.

![Surface conditions](image)
3.7. Bacteria re-growth after disinfection

Bacteria re-growth on supporters after disinfection is shown in Fig.9. It is observed that bacteria grew very slow after complete bacteria-elimination. In 10.5 hours the bacteria recovery percentage increased from 0% to 27%. One reason for the outcome is some amount of active chlorine remained on supporters, which had durable antibacterial effects [4, 30]. Another reason might be that formed active chlorine inhibited the feigning death conditions of bacteria. Therefore, the ex-situ electrochemical disinfection is proved to possess anti bacterial re-growth ability compared with other methods.

3.8. Energy consumption

Main costs of the ex-situ electrochemical disinfection system in the air conditioner are energy consumption of the process. Energy needed during the treatment was calculated by the following equation [33]:

![Figure 9. Bacteria re-growth on supporters after disinfection](image)
Where $E_{\text{cell}}$ is cell voltage (volt), $I$ is applied current (ampere), $T$ is treatment time (h) and $V$ is volume of treated water (m$^3$).

In the study for complete bacteria-elimination, cell voltage was 20 V, current was 0.04 A, treatment time was 0.167 h and electrolytic volume was 0.1 m$^3$. As a result, energy consumption for the study was 1.336 kWh·m$^{-3}$. Compared with other air conditioner disinfection methods, the result is economically practical.

4. CONCLUSION

This paper presents findings of ex-situ electrochemical disinfection with tap water and the PbO$_2$ electrode in the air conditioner. Results show chloride concentration, electro-conductivity, voltage and bacteria species affected disinfection performance conspicuously but pH and the time after electrolysis presented little effects on it. Total bacteria elimination was quick and could be achieved, under specific conditions, in 50 s and 60 s for Escherichia coli and Staphylococcus aureus, respectively. In addition, erosion was not observed on aluminum and ABS plastic surface. This method was found to possess the anti bacteria re-growth ability after disinfection. The overall energy consumption was evaluated as 1.336 kWh·m$^{-3}$ and considered economical in this study. Compared with other methods, this method provides several major advantages from the results. First, the ex-situ electrochemical system adapts to various types of tap water which is the electrolyte material in the study. Second, energy consumption and basic materials of the method is economically cost-efficient. Third, disinfection effects in the process are quick, durable and could eliminate a wide range of bacteria in corners and intervals. Fourth, erosion and bacteria re-growth, which could cause air conditioner failure and increasing costs, could be well inhibited. Finally, the whole system could be easily operated. Therefore, this method provides a promising approach for air condition disinfection.

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