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# Disinfection performance of coal-based carbon membrane coupled with electrochemical oxidation process for *Vibrio cholerae* in seawater

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ABSTRACT

A treatment system combining the coal-based carbon membrane with electrochemical oxidation process was designed for the enhanced disinfection performance. The effects of various parameters including electric voltage, *Vibrio cholerae* density, rotate speed, electrical conductivity, and electrode distance on the disinfection performance were carried out. The results showed the disinfection performance of the carbon membrane was significantly improved by coupling with electrochemical oxidation process and 2.0 V electric voltage was recommended. High *Vibrio cholerae* density usually increased the load of the treatment system, resulting in low permeate flux and disinfection efficiency. High rotate speed prompted the permeation of *Vibrio cholerae* cells, which inevitably made some of *Vibrio cholerae* cells penetrate though the carbon membrane quickly without being treated. High conductivity favored the electrochemical disinfection due to the generation of more total residual chlorine (TRC). Reducing electrode distance intensified electrochemical reactions and produced a large amount of gas bubbles, hindering water permeate.

*Keywords:* Carbon membrane; Electric voltage; Disinfection; *Vibrio cholerae* cells

## 1. Introduction

Marine pathogenicmicrobes including *Vibrio cholerae* etc. are potential risk sources to aquaculture systems because the accumulation of metabolites of marine microbes will poison the surrounding environment and cause the great losses to aquaculture industries [1–4]. In recent years, various disinfection technologies such as ultraviolet irradiation, chlorination, ozone, hydrogen peroxide, chemical biocides and electrochemical sterilization have been proposed for seawater disinfection during the past few decades [5–10]. Among them, electrochemical disinfection is becoming increasingly important for seawater disinfection because of many benefits including no additional chemicals, high efficiency, and simple operation [11–13].

The selection of anodic materials plays a crucial role in electrochemical disinfection processes. Patermarakis and Fountoukidis earlier used Ti as an anode to electrochemically treat natural water with highly contaminated with coliforms [14]. Subsequently, various electrodes such as plastic electrode consisted of graphite and silicone rubber [15], diamond electrode [16,17], $IrO_2$ electrode [18], and carbon electrode [19–21] are developed rapidly. Among these anodic materials, carbon materials have many great potential as anodic materials due to low cost, corrosion resisting, and excellent electrochemical properties. Matsunaga et al. adopted carbon-cloth electrode to build an electrochemical reactor for *Escherichia coli K-12* disinfection [19]. Saha and Gupta used graphite electrode to design an electro-chlorinator reactor for drinking water disinfection [20]. Shang et al. developed a pyrolytic graphite electrode modified with

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multi-walled carbon nanotubes to construct an electrochemical disinfection system [21]. These works reveal that carbon materials are good candidate materials to act as electrodes for electrochemical sterilization.

Porous carbon membranes fabricated from various carbonaceous materials have received great attention in many industrial areas [22]. In the past several years, we successfully fabricated the tubular microfiltration carbon membrane derived from coal, which not only demonstrated considerable separation performance, but also possessed good electrical conductivity [23,24]. Based on this idea, we constructed an electrochemical disinfection system combining the carbon membrane with electrochemical oxidation process. The effects of main operating conditions including electric voltage, *Vibrio cholerae* density, rotate speed, conductivity, and electrode distance on the disinfection performance were studied in detail.

## 2. Materials and methods

### 2.1. Materials

The carbon membranes (average pore size: 0.382 μm; porosity: 49.56%; outside diameter: 8.5 mm; inner diameter: 0.5 mm) derived from coal were fabricated according to our published work [23]. The morphologies of the carbon membrane were obtained by a Quanta450 scanning electron microscope (FEZ Company, USA). Ti plate (length: 45 cm; thickness: 0.01 mm; purity: 99.99%) was obtained from Qingyuan metal materials co., Ltd, China.

### 2.2. Treatment of Vibrio cholerae

*Vibrio cholerae* cells (Shanghailuweico., Ltd, China) were firstly cultured in alkaline peptone water broth (APB) at 37°C for 20 h, and then, they were collected by centrifugation and washed three times by saline water. Subsequently, the obtained *Vibrio cholerae* pellet was disperse in the sterilized seawater to obtain the solution with different initial cell density.

The experimental schematic of the treatment system was presented in Fig. 1. The conductivity carbon membrane as the anode was put in the center of the treatment system, and Ti plate as the cathode surrounds the carbon membrane. The initial density of *Vibrio cholerae* cells was measured with a XB-K-25 hemocytometer (Shanghai Qiujing biochemical reagents & instrument co., Ltd., China) under a Model Eclipse E200MV R optical microscope (Nikon, Japan). The *Vibrio cholerae* density after disinfection treatment was obtained by the method of live bacteria counting. Residual chlorine was measured using a HACHDR6000 UV–Vis spectrometer (HACH Inc., USA) according to N,N-diethyl-p-phenylenediamine (DPD)colorimetric method [25–27].

## 3. Results and discussion

### 3.1. Effect of electric voltage

Disinfection performance greatly depended on the electric voltage exerted on the treatment system. As shown in Fig. 2, when no electric voltage was adopted, a low permeate flux was observed during the whole process, and the final flux only reached 162.19 L/m$^2$·h·bar. As for disinfection efficiency, it was also only 70.16% after 6 h treatment (as shown in Table 1), which implied that the *Vibrio cholerae* removal from seawater only by the carbon membrane was not satisfied because the width of *Vibrio cholerae* (about 0.40 μm) was very close to the pore size of the carbon membrane. The SEM images (Figs. 3a and c) demonstrated that slight membrane fouling phenomenon was found, imply-

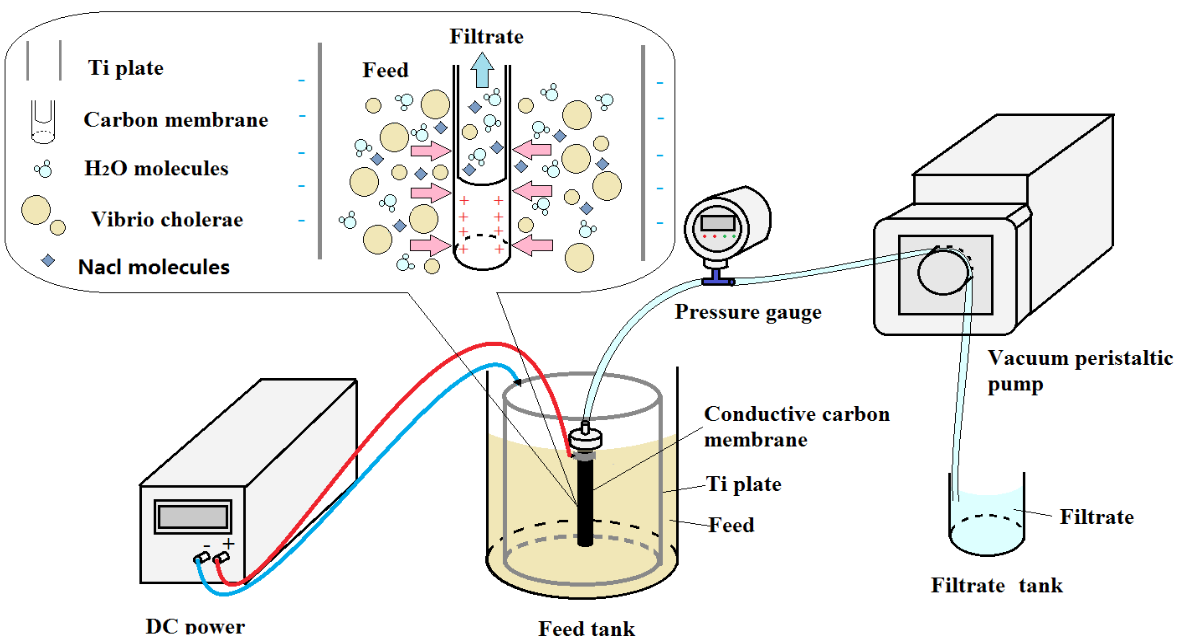


Fig. 1. Flow schematic diagram of carbon membrane coupling with an electric voltage.

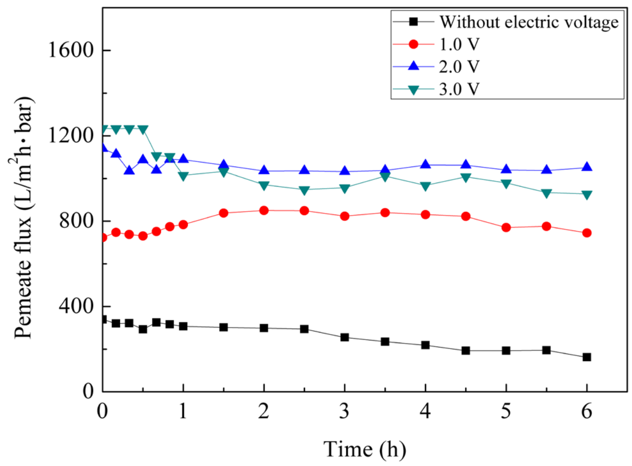


Fig. 2. Effect of electric voltage on permeate flux for $5.00 \times 10^4$ CFU/mL of initial *Vibrio cholerae* density at 7.5 rpm of rotate speed, 48.62 ms/cm of electrical conductivity and 7.5 cm of electrode distance.

Table 1
Effect of electric voltage on disinfection efficiency at 7.5 r/min of rotate speed, 48.62 ms/cm of electrical conductivity and 7.5 cm of electrode distance

| Electric voltage (V) | Initial *Vibrio cholerae* (CFU/mL) | Final *Vibrio cholerae* (CFU/mL) | TRC (mg/L) | Disinfection efficiency (%) |
|---|---|---|---|---|
| Without electric voltage | $5.00 \times 10^4$ | $3.53 \times 10^4$ | – | 70.6 |
| 1.0 V | $5.00 \times 10^4$ | 0 | 0.01 | 100 |
| 2.0 V | $5.00 \times 10^4$ | 0 | 0.03 | 100 |
| 3.0 V | $5.00 \times 10^4$ | 0 | 0.05 | 100 |

ing *Vibrio cholerae* cells could give rise to membrane fouling by adhering to membrane surface or penetrating into membrane pores.

When 1.0 V electric voltage was applied, the permeate flux and disinfection efficiency got obvious improvement owing to the generation of total residual chlorine (TRC), which played an important role on improving permeate flux and disinfection efficiency [Eqs. (1)–(4)] [28–30]. Increasing electric voltage to 2.0 V, the final permeate flux and disinfection efficiency were kept at 1050.74 L/m²·h·bar and 100%, respectively. From SEM images (Figs. 3b and d), no obvious foulant was observed, suggesting that the phenomenon of membrane fouling were effectively depressed due to electrochemical oxidation process.

$$2Cl^- - 2e^- \rightarrow Cl_2 \quad (1)$$

$$2H_2O + 2e^- \rightarrow H_2 + 2OH^- \quad (2)$$

$$Cl_2 + 2NaOH \rightleftharpoons NaCl + NaClO + H_2O \quad (3)$$

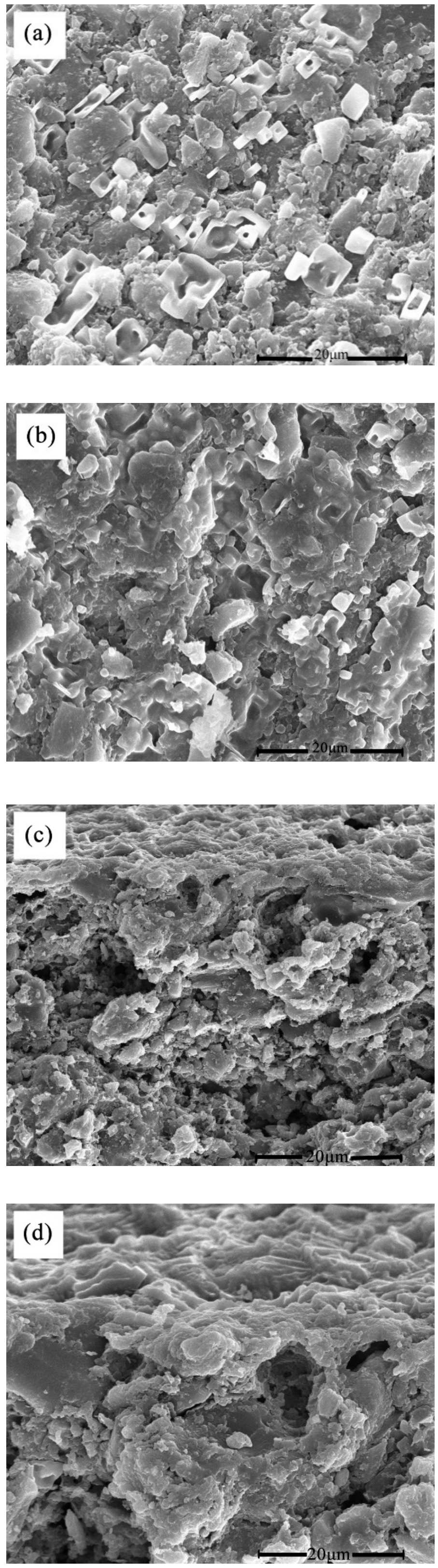


Fig. 3 SEM images from carbon membrane: (a) surface, without the electric voltage; (b) surface, with 2.0 V electric voltage; (c) cross-section, without the electric voltage; (d) cross-section, with 2.0 V electric voltage.

$$NaClO + H_2O \rightleftharpoons HClO + NaOH \quad (4)$$

While further improving electric voltage to 3.0 V, the disinfection efficiency was still kept at 100%, while the final permeate flux decreased to 927.48 L/$m^2$·h·bar instead. The phenomenon can be explained by the fact that the side reactions became more dramatic at a higher electric voltage, which produced a lot of gas bubbles, forming a big obstacle for permeation [31,32].

### 3.2. Effect of initial cell density

Fig. 4 presents the variation of permeate flux with treatment time at various *Vibrio cholerae* density. Apparently, increasing *Vibrio cholerae* density would result in the decrease of final permeate flux (from 1170.39 L/$m^2$h·bar to 86.28 L/$m^2$h·bar), implying higher *Vibrio cholerae* density tended to produce heavier membrane fouling. Similarly, at low *Vibrio cholerae* density, the disinfection efficiency of *Vibrio cholerae* cells could reach 100% (as shown in Table 2). However, when the *Vibrio cholerae* density was greater than $5.04 \times 10^5$ CFU/mL, the treatment system could not disinfect all *Vibrio cholerae* cells, and some of the *Vibrio cholerae* cells may penetrate through the membrane pores without being killed [33]. Therefore, it was not appropriate to efficiently disinfect the solution with high *Vibrio cholerae* density.

### 3.3 Effect of rotate speed

Fig. 5 provides the effect of rotate speed on permeate flux. With the increase of the rotate speed, the final permeate flux demonstrated an increasing tendency(from 1050.74 L/$m^2$·h·bar to 1699.41 L/$m^2$·h·bar) because of larger driving force at higher rotate speed. As for removal efficiency, when the rotate speed was set at 7.5 rpm, 100% disinfection efficiency could be achieved (as shown in Table 3). This was ascribed to that the system had enough time to disinfect *Vibrio cholerae* cells at a low rotate speed before penetrating though the membrane pores. In other words, the TRC produced by seawater electrolysis was sufficient to disinfect the *Vibrio cholerae* cells. However, further improving the rotate speed to 15.0 rpm, 20.0 rpm and 25.0 rpm, the disinfection efficiency decreased to 99.98%, 99.95% to 99.83%, respectively. The reason was probably related that the high rotate speed accelerated the permeation of *Vibrio cholerae* cells, which inevitably made some of *Vibrio cholerae* cells penetrate through the carbon membrane quickly before

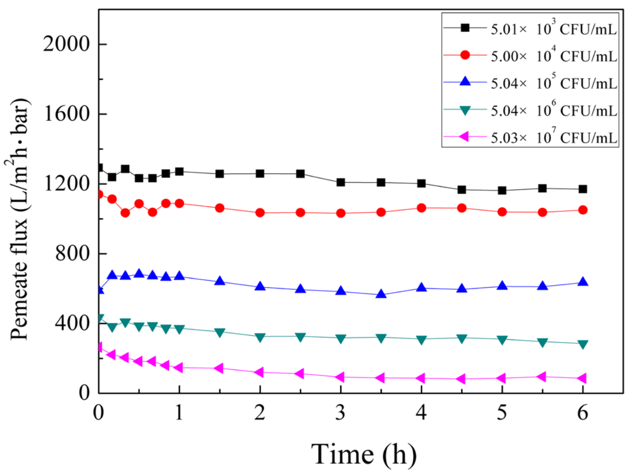


Fig. 4. Effect of initial *Vibrio cholerae* density on permeate flux at 2.0 V of electric voltage, 7.5 rpm of rotate speed, 48.62 ms/cm of electrical conductivity and 7.5 cm of electrode distance.

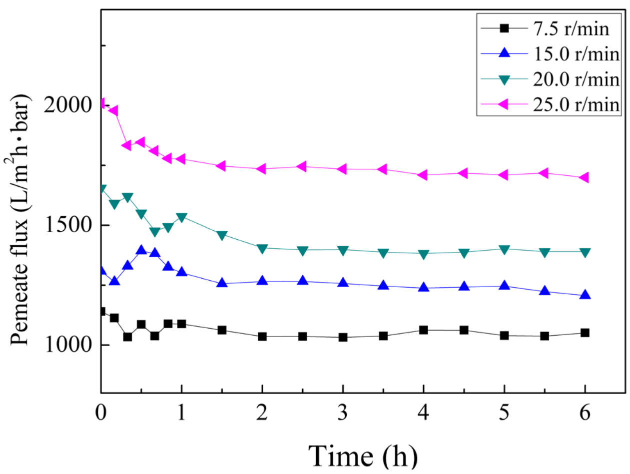


Fig. 5. Effect of rotate speed on permeate flux for $5.00 \times 10^4$ CFU/mL of initial *Vibrio cholerae* density at 2.0 V of electric voltage, 48.62 ms/cm of electrical conductivity and 7.5 cm of electrode distance.

Table 2
Effect of initial *Vibrio cholerae* density on disinfection efficiency at 2.0 V of electric voltage, 7.5 rpm of rotate speed, 48.62 ms/cm of electrical conductivity and 7.5 cm of electrode distance

| Initial *Vibrio cholerae* (CFU/mL) | Final *Vibrio cholerae* (CFU/mL) | TRC (mg/L) | Disinfection efficiency (%) |
|---|---|---|---|
| $5.01 \times 10^3$ | 0 | 0.03 | 100 |
| $5.00 \times 10^4$ | 0 | 0.03 | 100 |
| $5.04 \times 10^5$ | $1.01 \times 10^2$ | 0.03 | 99.98 |
| $5.04 \times 10^6$ | $1.97 \times 10^4$ | 0.03 | 99.61 |
| $5.03 \times 10^7$ | $1.41 \times 10^6$ | 0.03 | 97.20 |

Table 3
Effect of rotate speed on disinfection efficiency at 2.0 V of electric voltage, 48.62 ms/cm of electrical conductivity and 7.5 cm of electrode distance

| Rotate speed (r/min) | Initial *Vibrio cholerae* (CFU/mL) | Final *Vibrio cholerae* (CFU/mL) | TRC (mg/L) | Disinfection efficiency (%) |
|---|---|---|---|---|
| 7.5 | $5.00 \times 10^4$ | 0 | 0.030 | 100 |
| 15.0 | $5.00 \times 10^4$ | 10 | 0.025 | 99.98 |
| 20.0 | $5.00 \times 10^4$ | 25 | 0.025 | 99.95 |
| 25.0 | $5.00 \times 10^4$ | 85 | 0.020 | 99.83 |

being killed. Meanwhile, the high rotate speed also resulted in the reduction of TRC in the system, which decreased the disinfection ability of the system. Therefore, the high rotate speed would result in low disinfection efficiency [34].

### *3.4. Effect of electrical conductivity*

Fig. 6 and Table 4 shows the influence of electrical conductivity on permeate flux and disinfection efficiency. The electrical conductivities of seawater (0.02 ms/cm, 18.52 ms/cm, 33.40 ms/cm and 48.62 ms/cm) were adjusted by mixed seawater with distilled water at the ratios of 0:1, 1:2, 2:1 and 1:0, respectively. Clearly, low electrical conductivity (0.02 ms/cm) gave rise to low permeate flux and disinfection efficiency (650.66 L/m$^2$·h·bar and 80.70%). Increasing the electrical conductivity to 18.52 ms/cm, the permeate flux and disinfection efficiency achieved significant improvement (861.47 L/m$^2$·h·bar and 100%). When the electrical conductivity was adjusted to 48.62 ms/cm, the permeate flux and disinfection efficiency reached 1050.74 L/m$^2$·h·bar and 100%. The reason might be ascribed to that high conductivity was benefit to electrochemical disinfection reactions, which produced more TRC (as shown in Table 4) to sterilize *Vibrio choleraee* cells [35,36].

### *3.5. Effect of electrode distance*

Fig. 7 and Table 5 demonstrate the influence of electrode distance on the disinfection performance of the treatment system. With the increase of electrode distance from 3.5 cm to 7.5 cm, the final permeate flux also increased (from 231.42 L/m$^2$·h·bar to 1050.74 L/m$^2$·h·bar). This result could be explained by that small electrode distance intensified electrochemical reactions and produced a large amount of bubbles, hindering water permeate [37,38]. Although TRC produced in the treatment system slightly decreased with the increase of electrode distance, 100% disinfection efficiency was observed during the whole treatment process.

## 4. Conclusions

In order to effectively sterilize *Vibrio cholerae* cells in seawater, a treatment system combining membrane separation with electrochemical disinfection was constructed. In this work, we found that the electric voltage had an obviously positive influence on electrochemical disinfection performance. However, when the electric voltage was increased above 2.0 V, the final permeate flux started to decrease

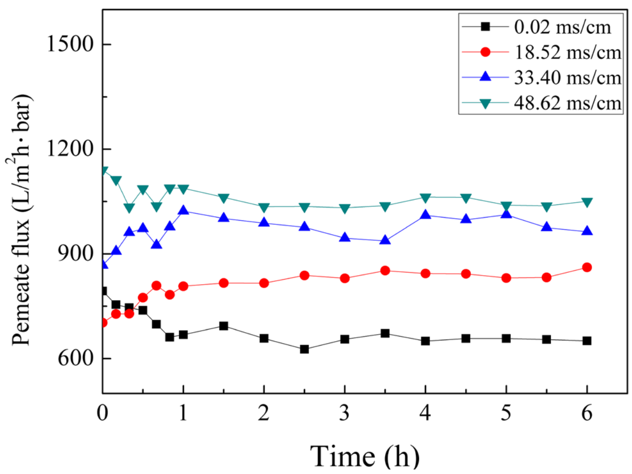


Fig. 6. Effect of electrical conductivity on permeate flux for 5.00 × 10$^4$ CFU/mL of initial *Vibrio cholerae* density at 2.0 V of electric voltage, 7.5 rpm of rotate speed and 7.5 cm of electrode distance.

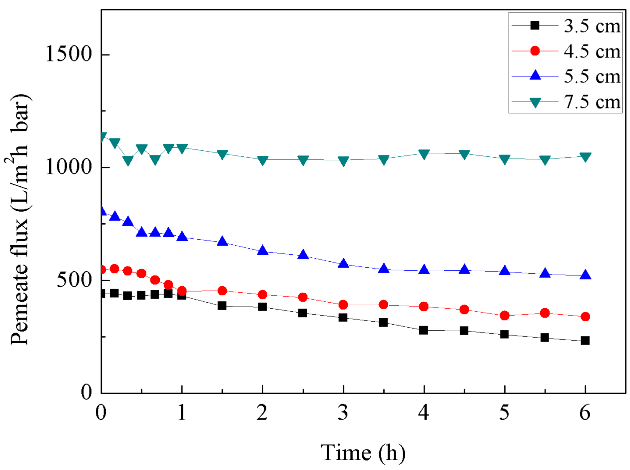


Fig. 7. Effect of electrode distance on permeate flux for 5.00 × 10$^4$ CFU/mL of initial *Vibrio cholerae* density at 2.0 V of electric voltage, 48.62 ms/cm of electrical conductivity and 7.5 rpm of rotate speed.

Table 4
Effect of electrical conductivity on disinfection efficiency at 2.0 V of electric voltage, 7.5 rpm of rotate speed and 7.5 cm of electrode distance

| Electrical conductivity (ms/cm) | Initial *Vibrio cholerae* (CFU/mL) | Final *Vibrio cholerae* (CFU/mL) | TRC (mg/L) | Disinfection efficiency (%) |
|---|---|---|---|---|
| 0.02 | 5.00 × 10$^4$ | 9.65 × 10$^3$ | 0.00 | 80.7 |
| 18.52 | 5.00 × 10$^4$ | 0 | 0.01 | 100 |
| 33.40 | 5.00 × 10$^4$ | 0 | 0.03 | 100 |
| 48.62 | 5.00 × 10$^4$ | 0 | 0.05 | 100 |

Table 5
Effect of electrode distance on disinfection efficiency at 2.0 V of electric voltage, 48.62 ms/cm of electrical conductivity and 7.5 rpm of rotate speed

| Electrode distance (cm) | Initial *Vibrio cholerae* (CFU/mL) | Final *Vibrio cholerae* (CFU/mL) | TRC (mg/L) | Disinfection efficiency (%) |
|---|---|---|---|---|
| 3.5 | 5.00 × 10$^4$ | 0 | 0.05 | 100 |
| 5.5 | 5.00 × 10$^4$ | 0 | 0.03 | 100 |
| 6.5 | 5.00 × 10$^4$ | 0 | 0.04 | 100 |
| 7.5 | 5.00 × 10$^4$ | 0 | 0.03 | 100 |

owing to various side reactions. High *Vibrio cholerae* density usually led to low final permeate flux because it was beyond the disinfection capability of the system itself. High rotate speed prompted the permeation of *Vibrio cholerae* cells, while it inevitably made some of *Vibrio cholerae* cells penetrate through the carbon membrane quickly without being killed. High conductivity was benefit to electrochemical disinfection due to the generation of more TRC. The permeate flux displayed a decreasing trend with the reduction of electrode distance because drastic electrochemical reactions built a big obstacle for the permeate flow.

### Acknowledgement

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