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Microbial electrolytic disinfection process for highly efficient *Escherichia coli* inactivation

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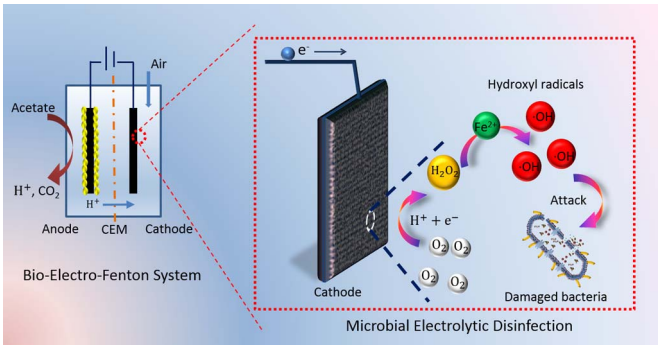
HIGHLIGHTS

- An innovative microbial electrolytic disinfection process for water purification.
- H_2O_2 was in situ synthesized in cathode to trigger Fenton reaction.
- Identified key factors affecting the *E. coli* inactivation in the system.
- Cell membrane damage as potential mechanism of *E. coli* inactivation.

GRAPHICAL ABSTRACT



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ABSTRACT

Water quality deterioration caused by a wide variety of recalcitrant organics and pathogenic microorganisms has become a serious concern worldwide. Bio-electro-Fenton systems have been considered as cost-effective and highly efficient water treatment platform technology. While it has been extensively studied for recalcitrant organics removal, its application potential towards water disinfection (e.g., inactivation of pathogens) is still unknown. This study investigated the inactivation of *Escherichia coli* in a microbial electrolysis cell based bio-electro-Fenton system (renamed as microbial electrolytic-Fenton cell) with the aim to broad the application of microbial electrochemistry. Results showed that a 4-log reduction of *Escherichia coli* (10^7 to hundreds CFU/mL) was achieved with an external applied voltage of 0.2 V, 0.3 mM Fe^{2+} and cathodic pH of 3.0. However, non-notable inactivation was observed in the control experiments without external voltage or Fe^{2+} dose. The disinfection effect was enhanced when cathode air flow rate increased from 7 to 41 mL/min and was also in proportion to the increase of Fe^{2+} concentration from 0.15 to 0.45 mmol/mL. Fatal cell membrane destruction by ·OH was identified as one potential mechanism for disinfection. This study successfully demonstrated the feasibility of bio-electro-Fenton process for pathogens inactivation, which offers insight for the future development of sustainable, efficient, and cost-effective biological water treatment technology.

1. Introduction

Water contamination by pathogenic microorganisms may cause serious diseases to human beings, which makes sanitation a priority issue worldwide in recent years [1]. The increasing pressure on calling for pure water flows (e.g., drinking water) leads to the research of more

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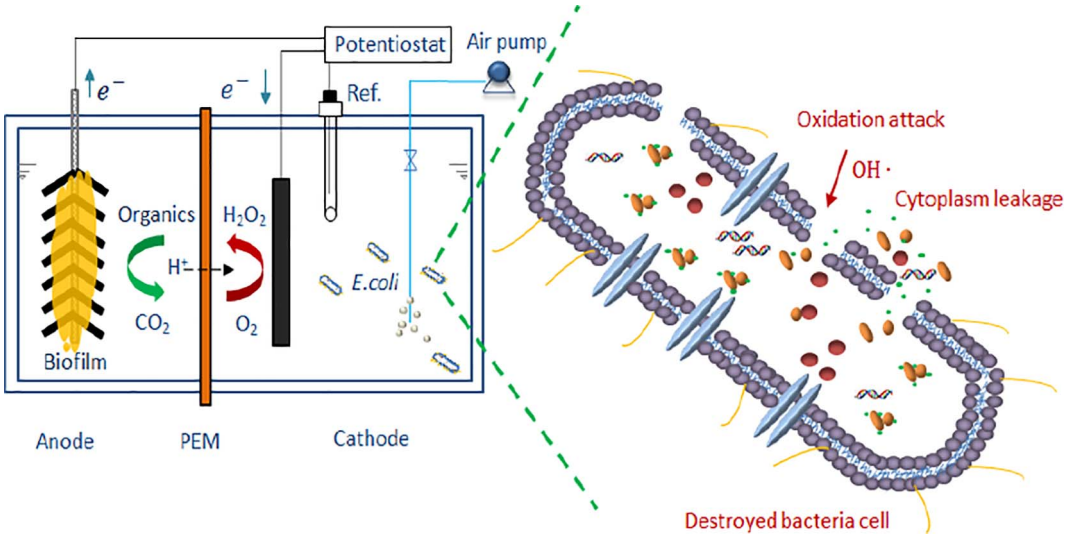

Fig. 1. Schematic illustration of disinfection in Bio-electro-Fenton System.

efficient and sustainable technology for biohazards inactivation. Several disinfection processes (e.g., chlorination, ozonation and photocatalysis) have been developed in the past years with extended prospects. However, most of the technologies suffer from inherent drawbacks such as cost, energy-intensive, toxic byproducts or low efficiency [2,3], thus it is of importance to develop an innovative technology that could address these limitations.

Among all the chemical disinfection approaches, Fenton reaction, mainly benefiting from the powerful oxidation capability of hydroxyl radical derivatives produced from hydrogen peroxide and catalysts (e.g., iron (II)), has been proven highly effective on disinfection process [4,5]. This disinfectant presents disinfection efficacy in a relative wide spectrum of microbes including but not limited to bacteria, viruses and spores, by launching an attack on cell membrane, nucleic acid molecules, etc. [6]. Though promising, the technology is still facing several challenges. Firstly, H_2O_2 as the main reactant of Fenton reaction is mainly produced from complex anthraquinone oxidation process [7]. Secondly, the supply of H_2O_2 is also limited by safety risks of transportation and storage so as to be restrained in many fields.

Recently, bio-electro-Fenton systems, which integrate Fenton reaction into various bio-electro-chemical systems (BES), have been widely studied in the field of bio-refractory organic wastewater treatment [8–11] due to their unique merits in cost-effective and efficient in-situ H_2O_2 generation [12]. The first generation of bio-electro-Fenton systems is based a typical BES termed microbial fuel cell (MFC) [13–15]. In this system, oxygen is reduced to H_2O_2 through a two-electron reduction pathway (Eq. (1)) at the surface of cathode by receiving the electrons transferred from the bacterial oxidation of organics in anode [16,17]. As a result, electricity is generated and the H_2O_2 produced in-situ in the cathode reacts with Fe^{2+} to form ·OH, a very reactive compound effective for water purification (Eq. (2)) [9,18]. Latterly, it was found that the treatment capacity can be boosted by supplying small amounts of electricity (0.2–1.0 V) to the system, due to the improved H_2O_2 production [19]. In lights of the advantages in H_2O_2 production and utilization, the bio-electro-Fenton systems have been applied to treat a broad spectrum of recalcitrant organic pollutants, including azo days, aniline, pharmaceuticals, p-nitrophenol and so on [10,11]. However, water disinfection which could be another important application of the bio-electro-Fenton technology has never been explored.

$$O_2 + 2H^+ + 2e^- \rightarrow H_2O_2 \quad (1)$$

$$H_2O_2 + Fe^{2+} \rightarrow \cdot OH + Fe^{3+} + OH^- \quad (2)$$

Herein, an innovative application of the bio-electro-Fenton system for the inactivation of pathogenic microorganisms in water (*E. coli* as a model), named microbial electrolytic disinfection (MED) process, was proposed and investigated. In this process, the bacteria in the anode of MFC assisted with small input voltage (0.1–0.4 V) will first oxidize organics and transfer the electrons to the cathode for H_2O_2 synthesis (Eq. (1)), which will react with Fe^{2+} to form ·OH (Eq. (2)). The ·OH could oxidize organic pollutants and inactivate pathogens. To date, bio-electro-Fenton systems have never been applied for water disinfection. The objective of this work is to elucidate and demonstrate the feasibility of the MED process as simple, cost-effective, efficient and reliable approach for water disinfection. The performance of the system was evaluated in terms of cell inactivation effects, electron flows, and hydrogen peroxide generation/utilization. The effect of different operating parameters (e.g. cathodic aeration rate, external applied voltage and Fe^{2+} concentrations) on the disinfection effect was also evaluated. The outcomes are expected to offer an efficient, cost-effective and environment-friendly platform technology for water disinfection, and expand the application of microbial electrochemical system.

2. Materials and methods

2.1. Reactor setup and operation

Dual-chamber reactor with 100 mL total volume and 80 mL working volume of each chamber was used for this study (Fig. 1). The anode electrode was carbon brush (5.9 cm in diameter, 6.9 cm in length, Mill-Rose, USA) on which the exoelectrogenic biofilm was first enriched in MFC mode for about one month using domestic wastewater as organic sources. An auxiliary glass bottle containing necessary nutrients solution was connected with anode chamber. Recirculation of the solution through anode was adopted to ensure efficient mass transfer and maintain the sufficient fuels for biofilm. The cathode was made of graphite plate (4 cm × 3.5 cm × 0.4 cm) and was boiled in 3 M H_2SO_4 for 10 min prior to use. A cation exchange membrane (CEM) (CMI 7000, membrane international, NJ) was used to separate two chambers. An Ag/AgCl reference electrode (+0.197 V vs SHE) was inserted into cathode chamber, close to the graphite plate (all potentials presented in this article were presented versus this electrode).

In the biofilm enrichment stage, the anode and cathode chambers were respectively fed with domestic wastewater and 50 mM potassium ferricyanide. The domestic wastewater was collected from primary clarifier (Lyngby Wastewater Treatment Plant, Copenhagen, Denmark) which was amended by 1 g/L acetate prior to use. The characteristics of