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Development of a photoelectrocatalytic method to improve the efficiency of *E. coli* removal

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ABSTRACT

The photoelectrocatalytic technology has attracted significant attention for effectively eliminating organic matter and microbiological pollutants in the environment, owing to its remarkable efficiency and low power consumption. The major goal of this research is to develop and determine the optimal conditions that will facilitate the photoelectrocatalytic technique's enhancement of *E. coli* eradication. The $WO_3/BiVO_4$ photoanode was fabricated on a conductive glass substrate using the automatic dip coating process, employing a layer-by-layer deposition method. Subsequently, the $WO_3/BiVO_4$ photoanode was calcinated at 550 °C for 60 minutes. The produced $WO_3/BiVO_4$ electrodes were employed as working electrodes to investigate and determine the optimal parameters for enhancing the eradication of *E. coli* process. The primary factors investigated in this study were the concentration of KCl electrolyte solution and the applied potential. These parameters were examined to identify the best circumstances that would result in the highest efficiency for the degradation of *E. coli* in a photoelectrochemical system. The study also aimed to comprehend the catalytic mechanism implicated in eliminating *E. coli* by implementing three different processes: photocatalysis, electrocatalysis, and photoelectrocatalysis. We discovered that the key factors directly influencing *E. coli* eradication effectiveness under the photoelectrocatalytic process were applied potential and electrolyte solution concentration. The optimum conditions eliminated 99.99% of *E. coli* in 150 minutes with an initial concentration of 10^6 CFU/ml, an electrolyte concentration of 0.01 M KCl, and an applied potential of 2.0 V. The study confirmed photoelectrocatalytic cells' efficacy in removing microorganisms and recommended their application in a wider range of wastewater treatment systems.

Keywords: Photoelectrocatalytic cell, *E. coli* removal, $WO_3/BiVO_4$ photoanode

INTRODUCTION

Contaminated wastewater has serious consequences for human and animal health due to the presence of bacteria and microbes [1, 2]. *Escherichia coli* (*E. coli*) is one of the most prevalent antibiotic-resistant pathogens. Therefore, it is necessary to develop effluent treatment methods [3-5]. Photoelectrocatalytic (PEC) technique, an advanced electrochemical technique that catalyzes the reaction with light and applied potential, has attracted great interest in applications for microbial elimination [4, 6-9]. The development of PEC technique can be achieved by finding optimum conditions for the highest efficiency in removing target substances or improving the reaction process [10, 11]. This research aims to study factors affecting the efficiency of eliminating *E. coli* using PEC techniques. We have investigated the factors that affect the efficacy of *E. coli* degradation, including electrolyte solution concentration, electric potential, and the catalytic mechanism at the electrode surface. We investigated and determined the optimal conditions for PEC cells to maximize the efficacy of *E. coli* elimination and confirm the mechanism responsible for the acceleration of elimination at the electrode surface. This research holds significant potential and carries implications for the advancement of novel technologies aimed at eradicating microorganisms in water and for further enhancing water treatment systems.

MATERIALS AND METHODS

FTO/WO_3/$BiVO_4$ electrode preparation

This study prepares the electrodes with an automatic dip coating machine, as displayed in figure 1. Fluorine-doped tin oxide (FTO) conductive glass was used as a support material for the fabrication of

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semiconductor thin films by ultrasonic cleaning in various solutions for 10 minutes, as detailed as follows: solution of detergent, 3 M sodium hydroxide solution, ethanol solution, and deionized water. The FTO conductive glass was immersed in a 0.1 M WO_3 solution, dried at 150 °C for five minutes, and then calcined at 500 °C for thirty minutes. Then, using an automatic dip coater, the FTO/WO_3 electrode was immersed in 0.05 M $BiVO_4$ solution. Then, they are dried at 150 °C for 5 minutes and calcined at 550 °C for 60 minutes. In the experimental setup, FTO/WO_3/$BiVO_4$ electrodes are employed as an anode electrode to further examine the effectiveness of *E. coli* eradication.

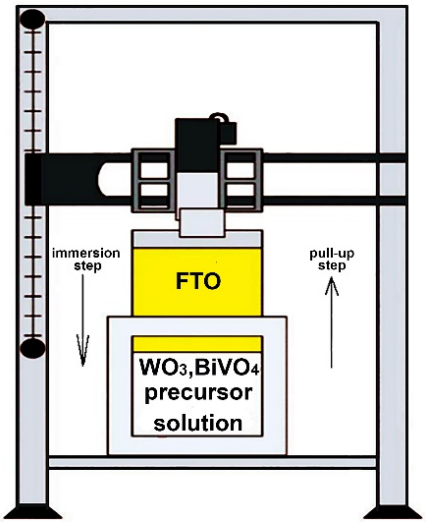

Figure 1 The schematic dip coating machine for FTO/WO_3/$BiVO_4$ electrode fabrication process.

E. coli removal efficiency study

The FTO/WO_3/$BiVO_4$ electrode was employed to investigate the effectiveness of eradicating *E. coli* by utilizing the PEC approach. The concentrations of KCl electrolyte solutions at 0, 0.01, 0.025, 0.05, 0.075, and 0.1 M and applied potentials of 0, 0.5, 1.0, 1.5, 2.0, and 2.5 V were studied to determine the optimal conditions for the highest PEC *E. coli* degradation efficiency. In addition, the catalytic mechanism for the eradication of *E. coli* was investigated using Photocatalytic (PC), Electrocatalytic (EC), Photoelectrocatalytic (PEC), and control containing approximately 10^6 CFU/ml of *E. coli*. The concentration of *E. coli* was examined utilizing the spread plate technique at different time intervals. Subsequently, the concentration and percentage degradation were determined and represented by Equation (1) [12] and Equation (2), respectively.

$$\text{CFU/ml} = \frac{\text{Total number of colonies obtain x dilution factor}}{\text{Volume of specimen used}} \quad (1)$$

$$\textit{E. coli}\ \text{degradation}\ (\%) = \frac{A_0 - A_t \times 100}{A_0} \quad (2)$$

where A_0 represents the quantity of *E. coli* present at the beginning of the elimination process in CFU/ml. A_t represents the quantity of remaining *E. coli* at the time in CFU/ml.

RESULTS AND DISCUSSIONS

Optimization of PEC cell for E. coli degradation

The experimental results demonstrated a direct relationship between an increase in applied voltage within the range of 0 to 2.5 V and the subsequent enhancement in the efficiency of *E. coli* removal, as demonstrated in figure 2.

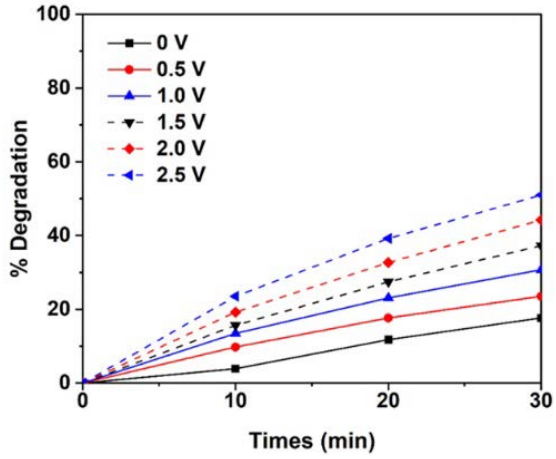

Figure 2 The effect of applied potential on *E. coli* degradation efficiency.

This phenomenon occurs due to the acceleration of charge transfer and the subsequent increase in electron flow from the anode to the cathode when the applied voltage is raised in the positive direction. Increasing the positive potential also increases the amount of positively charged (hole; h^+) at the anode, which supports a high oxidizing efficiency for the degradation of *E. coli* [13]. However, the electrode life may be compromised if the system voltage is too high.

Figure 3 illustrates the effect of varying concentrations of potassium chloride (KCl) electrolyte solution on eradicating *E. coli*. The efficacy of eradicating *E. coli* is directly proportional to the concentration of electrolytes within the range of 0 to 0.1 M. The results that were noticed through the investigation of KCl concentration. Increasing electrolyte concentration facilitates enhanced electrical conductivity and charge transfer between the electrodes, hence promoting electrode surface reactions that contribute to more effective degradation of *E. coli* [14]. Increasing the concentration of KCl can also result in the dissociation of chloride ions in the solution. The chloride ion present in the electrolyte undergoes a constant reaction with the hydroxyl radical, resulting in the formation of chlorine compounds that exhibit exceptional efficacy in eradicating *E. coli*.

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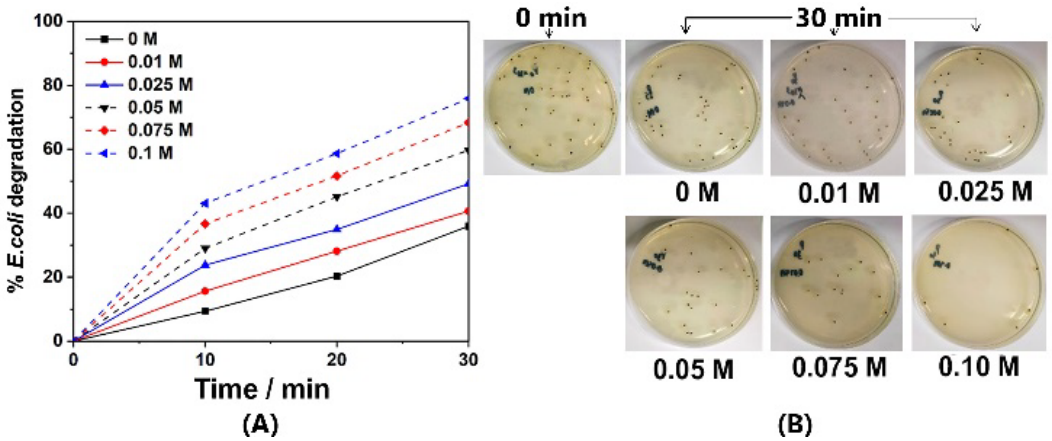

Figure 3 The effect of KCl electrolyte concentration on the efficacy of *E. coli* degradation, where (A) the percentage of *E. coli* degradation and (B) the photo of *E. coli* depending on KCl concentration and time degradation.

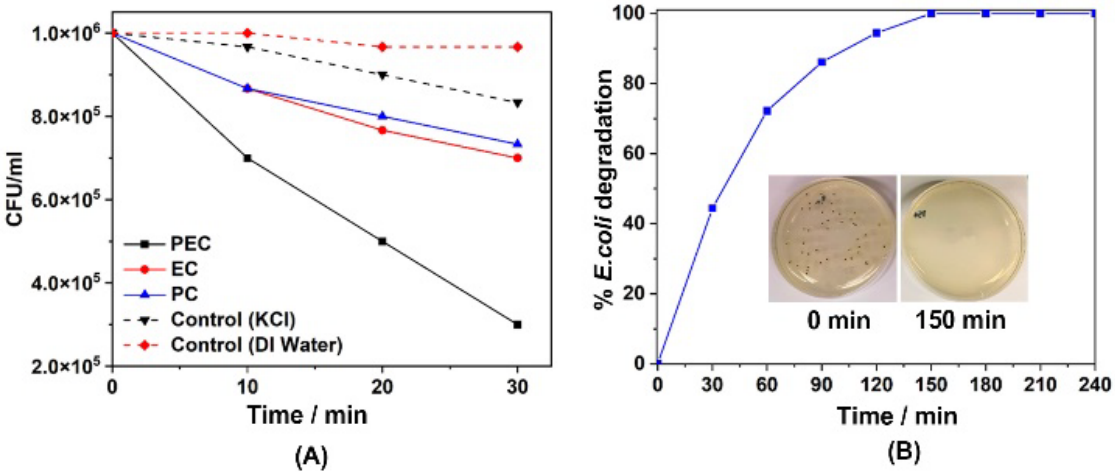

Figure 4 The catalytic mechanism in *E. coli* degradation was investigated; (A) reduction of *E. coli* in CFU/ml and (B) *E. coli* degradation efficiency under optimal conditions and inset of photographs of *E. coli* analysis.

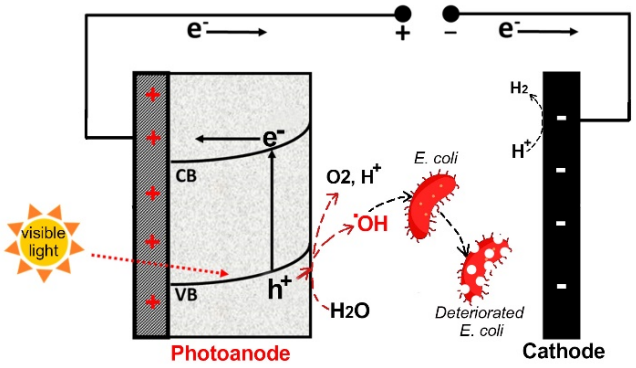

Figure 5 Diagrammatic representation of the process at the semiconductor anode surface for PEC-based removal of *E. coli*.

Catalytic mechanism and E. coli degradation efficiency

Figure 4 demonstrates that the PEC catalytic mechanism has the highest *E. coli* degradation efficiency of 70% within 60 minutes, followed by the EC, PC catalytic mechanism, and solution-based potassium chloride control system, which eliminated only 20%, 26%, and 17%, respectively. In contrast, controls that do not utilise potassium chloride electrolyte solution can only remove a small amount. In addition, figure 4B demonstrated that *E. coli* at a concentration of 10^6 CFU/ml could be eliminated 100% in 150 minutes using a potassium chloride electrolyte concentration of 0.01 M and an applied potential of 2 V with the PEC technique.

Figure 5 exhibits the schematic diagram illustrating the reaction occurring at the surface of the semiconductor anode, specifically in the presence of photo-acceleration and electric potential. When the valence band (VB) layer of a semiconductor is stimulated by light within a certain range, it leads to the dissociation of electrons (e^-) from the VB to the conduction band (CB) [9, 15], while simultaneously creating a positively charged vacancy (hole; h^+) inside the VB. The VB layer has a notable propensity to facilitate water oxidation at the surface of the semiconductor electrode $WO_3/BiVO_4$, generating hydroxyl radicals (^oOH). This generated ^oOH has powerful oxidizing capabilities, efficiently oxidizing *E. coli* in aqueous solution [16]. In addition, controlling the positive potential at the anode electrode

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may accelerate transport and induce e^- flow at the CB layer from the anode electrode to the cathode electrode to convert H^+ to H_2 [17]. This procedure can potentially mitigate the recombination occurrence of electron-hole pairs (e^- and h^+) and enhance the PEC characteristics for eliminating *E. coli*.

CONCLUSION

This research successfully applied a $WO_3/BiVO_4$ photoanode to eliminate *E. coli* under a photoelectrocatalytic process effectively. The electrolyte concentration and potential applied factors in the *E. coli* elimination system were investigated. We demonstrated that when the proposed PEC approach was optimized with a 0.01 M potassium chloride electrolyte solution and an applied voltage of 2 V, *E. coli* at 10^6 CFU/mL was 100% eliminated in 150 minutes. This study generates information for developing a new alternative technique for treating wastewater contaminated with microorganisms and other organic waste.

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