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Full Length Article

Enhancement of g-C_3N_4 cathode for inactivation of marine microorganisms in $ZnWO_4$ photocatalytic system

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ABSTRACT

Generated from photoelectrocatalytic (PEC) reaction systems, hydroxyl radicals can be applied for the treatment of ballast water contaminated with various microorganisms. However, main cathodic and hydrogen evolution reactions have no effect on the inactivation efficiency, which reduces the overall reaction efficiency. Inactivation efficiency can be improved by inducing the oxygen reduction reactions (ORRs) on a cathode. Oxygen can be reduced to H_2O_2 under $2e^-$ reactions with ORR catalysts, and $^{\cdot}OH$ can be produced by H_2O_2 under ultraviolet (UV) irradiation, which is beneficial to microorganism deactivation. This work investigated the contributions of ORR in a graphene-like carbon nitride (g-C_3N_4) to the PEC activities of a $ZnWO_4$ anode. Results indicated that microorganisms were deactivated completely after 18 min under 0.7 V bias potential when ORR was used on the g-C_3N_4 cathode, and thus approximately one of the total time was conserved. ORR on the g-C_3N_4 cathode plays a key role in PEC inactivation under low voltage potential. The g-C_3N_4 ORR in seawater involves two electron reactions and generates H_2O_2, which results in high inactivation efficiency under UV irradiation. This work shows that the search for an efficient $2e^-$ reaction ORR catalyst is of considerable importance in improving ballast water treatment.

1. Introduction

Microorganisms may spread to various parts of the world through ships and subsequently cause significant damage to ecosystems [1–3]. Hence, efficient and environment-friendly strategies should be established in ocean ships for microorganism inactivation. The deactivation of hydroxyl radical ($^{\cdot}OH$) in microorganisms is considered a promising method because it has a strong oxidation capability and can effectively damage the cell walls and DNA of microorganisms without causing environmental pollution [4–10]. $^{\cdot}OH$ can be generated in photocatalytic reaction systems [11–14]. Several studies indicated that photocatalysts exhibit excellent sterilization performance [15–20]. Therefore, photocatalytic materials can be applied to ocean pollution treatment.

PEC technology is efficient and thus widely use in deactivating microorganisms [21]. Understanding photoelectrocatalysis and seawater electrolysis that occur simultaneously is important. In this process, the main cathodic reaction is hydrogen evolution, which has no effect on inactivation efficiency.

In photoelectrocatalysis, reduction reactions transfer from the conduction band of photocatalysts to the cathodes along the direction of electron movement. In theory, the enhancement of cathodic ORR results in the improvement of PEC efficiency. Oxygen can be reduced to $^{\cdot}HO_2^-$, OH^-, H_2O_2, and $^{\cdot}OH$, which are beneficial for microorganism deactivation. Photoelectrocatalysis requires the use of a cathode ORR catalyst to promote ORR [22,23]. Several works indicated that graphene-like carbon nitride (g-C_3N_4) exhibits ORR activities. Oxygen can be reduced to H_2O_2 on a g-C_3N_4 surface because its ORR mechanism involves two-electron reactions and $^{\cdot}OH$ can be produce by H_2O_2 under UV irradiation, which is beneficial for microorganism deactivation [24].

$ZnWO_4$ is an ultraviolet photocatalyst with excellent performance, but its application is limited by its low light absorption and by the easy recombination of photogenerated charge carriers. In previous studies, $ZnWO_4$ exhibited high efficiency as an electrode for the photoelectric synergistic inactivation of *Chlorella* [31]. Thus, we used $ZnWO_4$ as photoanode to treat simulated ballast water. We selected Ti and g-C_3N_4 electrodes as cathodes to test whether an ORR catalyst improves deactivation efficiency. Then, different cathodic reactions in the cathode materials were determined through a comprehensive electrochemical test. The cathodic ORR mechanism that enhanced the photoelectrocatalysis process was discussed.

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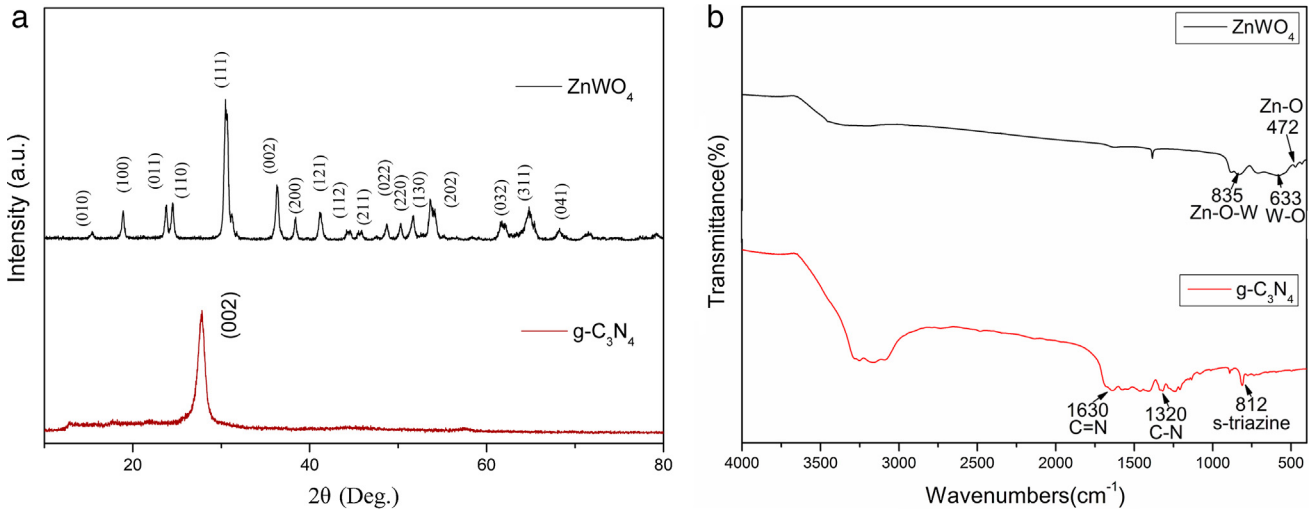

Fig. 1. XRD (a) and FT-IR (b) of $ZnWO_4$ and g-C_3N_4.

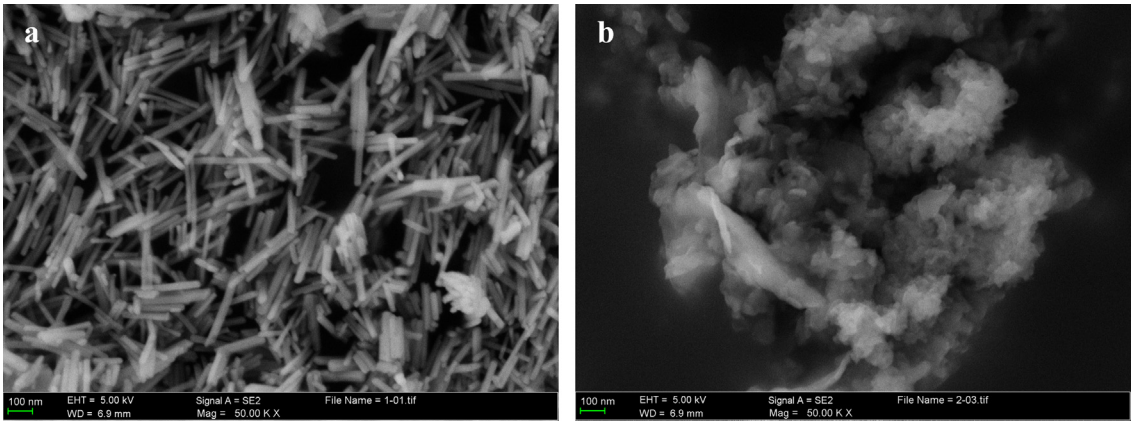

Fig. 2. FE-SEM images of $ZnWO_4$ (a) and g-C_3N_4 (b).

2. Experimental

2.1. Preparation of $ZnWO_4$ and g-C_3N_4

$ZnWO_4$ was synthesized through a hydrothermal method. First, 2.974 g of $Zn(NO_3)_2{\cdot}6H_2O$ and 3.298 g of $Na_2WO_4{\cdot}2H_2O$ were mixed in 80 mL ultrapure water. Then, the solution pH was adjusted to 9 by using a $NH_3{\cdot}H_2O$ solution. After adding an ultrasonic cleaner, the slurry was transferred to a reaction kettle heated at 180 °C for 24 h. Finally, the obtained samples were washed with ultrapure water and dried at 80 °C for 4 h [25]. g-C_3N_4 was prepared using sintering method. About 2 g of melamine was sintered at 550 °C for 4 h [26].

2.2. Preparation of the $ZnWO_4$ and g-C_3N_4 electrodes

$ZnWO_4$ and g-C_3N_4 electrodes were prepared by a simple brush-painting method. For the preparation of the $ZnWO_4$ electrode, 0.1 g of $ZnWO_4$ and 5 mg of XC72 were dispersed adequately in a mixture containing 9 mL of ultra-pure water and 1 mL of Nafion. After sufficient mixing, 2 mL of the suspension was painted on the ITO surface (60 mm × 20 mm). The electrodes were then dried in air for 24 h.

For the g-C_3N_4 electrode, 4 mL of concentrated HNO_3 was added to 100 mg of g-C_3N_4. Then, 2 mL of the completely mixed g-C_3N_4 and HNO_3 slurry was brushed on Ti foll. We finally coated the Ti and g-C_3N_4 cathodes with an insulating adhesive (KE45W) to ensure effective working areas. The effective electrode area was 12 cm^2.

2.3. Electrochemical test

All electrochemical tests were recorded in VMP3 electrochemistry at Princeton with three electrode cells. The cyclic voltammetry (CV) had a potential range of −1.2 V to 0.2 V and a scan rate of 10 mV/s. Liner sweep voltammetry (LSV) is from 0 V to −1.75 V and scan speed is 10 mV/s.

2.4. Calculation of electron transfer number

The electron transfer numbers in seawater were determined by rotating the disk electrode. The rotation rates were 400, 900, 1600, and 2500 rpm [27,28]. All electrochemical tests were conducted in oxygen-saturated seawater. LSV curves were obtained from 0 V to −0.8 V at a scan rate of 10 mV/s. The electron transfer numbers for ORR on g-C_3N_4 catalysts were calculated by Koutecky Levich equation, as follows [29]:

$$\frac{1}{J} = \frac{1}{JL} + \frac{1}{JK} = \frac{1}{B\omega^{1/2}} + \frac{1}{JK} \quad (1)$$

$$B = 0.2nFC_0(D_0)^{2/3}v^{1/6} \quad (2)$$

where the current density J is a polarization curve, J_L and J_K are the diffused limit current density and kinetic energy current density, respectively, Student: omega is the rotation rate, F is the Faraday constant (96485C/mol), C_0 and D_0 are the volumetric concentration and diffusion coefficient of O_2 in seawater (0.938×10^{-3} mol/L, 2.75×10^{-5} cm^2/s); and V = 0.00956 cm^2/s [30].

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