



Elucidating the effects of different photoanode materials on electricity generation and dye degradation in a sustainable hybrid system of photocatalytic fuel cell and peroxi-coagulation process

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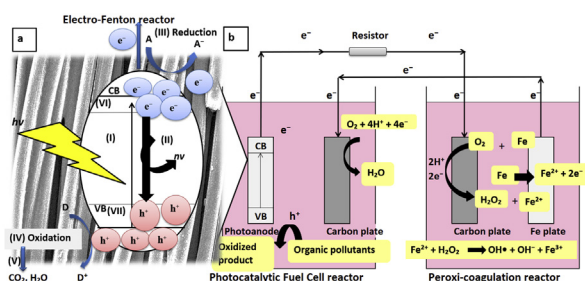
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HIGHLIGHTS

- Amaranth degradation at PFC was affected by different photoanode materials.
- Influence of photoanode on degradation at peroxi-coagulation reactor was insignificant.
- ZnO/CC generated higher electricity compared to TiO₂/CC and α-Fe₂O₃/CC in the system.
- Fast mineralization of intermediate compounds occurred at PFC except for α-Fe₂O₃/CC.

GRAPHICAL ABSTRACT



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ABSTRACT

The hybrid system of photocatalytic fuel cell – peroxi-coagulation (PFC-PC) is a sustainable and green technology to degrade organic pollutants and generate electricity simultaneously. In this study, three different types of photocatalysts: TiO₂, ZnO and α-Fe₂O₃ were immobilized respectively on carbon cloth (CC), and applied as photoanodes in the photocatalytic fuel cell of this hybrid system. Photocatalytic fuel cell was employed to drive a peroxi-coagulation process by generating the external voltage accompanying with degrading organic pollutants under UV light irradiation. The degradation efficiency of Amaranth dye and power output in the hybrid system of PFC-PC were evaluated by applying different photoanode materials fabricated in this study. In addition, the effect of light on the photocurrent of three different photoanode materials was investigated. In the absence of light, the reduction of photocurrent percentage was found to be 69.7%, 17.3% and 93.2% in TiO₂/CC, ZnO/CC and α-Fe₂O₃/CC photoanodes, respectively. A maximum power density (1.17 mWcm⁻²) and degradation of dye (93.8%) at PFC reactor were achieved by using ZnO/CC as photoanode. However, the different photoanode materials at PFC showed insignificant difference in dye degradation trend in the PC reactor. Meanwhile, the degradation trend of Amaranth at PFC reactor was influenced by the recombination rate, electron mobility and band gap energy of photocatalyst among different photoanode materials.

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1. Introduction

The peroxi-coagulation (PC) process is an emerging technology

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of electrochemical advanced oxidation processes (EAOPs) which are being successfully applied in the wastewater remediation. This process involves the reaction of electrogenerated H_2O_2 with Fe^{2+} , which leads to the hydroxyl radical formation (Brillas et al., 2009; Ren et al., 2018; Yazdanbakhsh et al., 2015). It promotes more benefits in term of risks and costs for chemical storages and transportation compared to the chemical Fenton process due to the on-site production of H_2O_2 (Boye et al., 2003; Brillas et al., 2009; Zhang et al., 2015). The sacrificial anode such as iron plate is utilized in the PC process to promote the continuous regeneration of Fe^{2+} which also can minimize the sludge production (Brillas et al., 2009; Feng et al., 2010; Ghanbari and Moradi, 2015). However, the power consumption has limited their application as it mainly contributes to the high operation cost (Feng et al., 2010; Zhang et al., 2015; Zhao et al., 2017b). Therefore, an effort has been made by developing a photocatalytic fuel cell-electro-Fenton (PFC-PC) system to degrade organic pollutants and generate electricity simultaneously. PFC has been employed to drive an PC process by generating the external voltage accompanying with simultaneously degrading organic pollutants under the light irradiation (Nordin et al., 2017b, 2017a).

In recent years, PFC has drawn great attention as an emerging photoelectrocatalytic (PEC) technology which contributes to the treatment of wastewater and electricity generation (Sui et al., 2017). The production of photogenerated electron (e^-) and holes (h^+) occur at the photocatalytic sites of photoanode during the light irradiation in the PFC system (Xia et al., 2016a; Zeng et al., 2018; Zhao et al., 2017a). The electrons that are released from the photoanode through the degradation of organic pollutant are moved to the cathode to generate the electricity in the system. Meanwhile, the holes react with the organic pollutant to produce the oxidized product (Xia et al., 2016a; Xie and Ouyang, 2017). However, the intrinsic semiconductor limitations is a major challenge in the PFC as the photocatalytic activity of semiconductor photoanode plays an important role in the PFC performance (Xie et al., 2017; Zhao et al., 2017b).

During the past few decades, titanium dioxide (TiO_2) has attracted great interest as photocatalyst among semiconductors and became the first material that has been used as a photoanode material in PFC owing to its outstanding stability and active features (Antoniadou and Lianos, 2010; Lianos, 2011; Liu et al., 2012, 2011; Zeng et al., 2018). TiO_2 is a semiconductor which absorbs only 4% of the solar light irradiation because its higher band gap energy (3.0–3.2 eV) (Wang et al., 2014). Therefore, the usage of TiO_2 is more favorable and efficient under the UV light irradiation compared to solar light irradiation (Ong et al., 2018). Zinc oxide (ZnO) is one of the n-type semiconductors with a wide band gap of 3.2 eV. It became an alternatives photocatalysts to TiO_2 as it possesses similar band gap energy. Nevertheless, ZnO can absorb the higher solar spectrum compared to TiO_2 (Habibi and Rahmati, 2015). The inexpensive and non-toxic material with a variety of nanostructure ZnO has attracted many attentions among the metal oxide semiconductor photocatalysts (Habibi and Karimi, 2014; Habibi and Rahmati, 2015). However, the rapid recombination rate of photogenerated electrons and holes become the greatest limitation of ZnO as a photocatalyst which perturbs the photo-degradation reaction (Ong et al., 2018).

Hematite ($\alpha\text{-Fe}_2\text{O}_3$) is a cheapest n-type semiconductor with a relatively small band gap energy (2.0–2.2 eV) (Mallick and Dash, 2013; Mishra and Chun, 2015). $\alpha\text{-Fe}_2\text{O}_3$ is a promising semiconductor for photocatalytic process as the absorption of the photon by $\alpha\text{-Fe}_2\text{O}_3$ up to 600 nm which starts in the near-infrared spectrum and absorbs approximately 40% of the solar energy (Tahir et al., 2009). Nevertheless, the limiting factors of $\alpha\text{-Fe}_2\text{O}_3$ such as short diffusion lengths of holes is within the range of 2.0–4.0 nm, excessive carrier recombination, poor charge transport

property and electrical conductivity can affect the performance of photocatalytic activity (Mishra and Chun, 2015; Xia et al., 2016b).

In general, the photocatalytic activity depends on the band gap energy and recombination of photogenerated carriers in the photocatalyst materials to produce hydroxyl radicals and electricity generation (Ong et al., 2018; Zhao et al., 2012). Thus, the photoanode material with different band gap energy, recombination rate and electron mobility properties play an important role in this hybrid system which can influence the fuel cell performance in term of dye removal efficiency and electricity generation. Besides, no report on the comparative study of TiO_2 , ZnO and $\alpha\text{-Fe}_2\text{O}_3$ photocatalyst immobilized on carbon cloth photoanode using this PFC-PC system is available so far. The purpose of this study is to evaluate the effects of different photoanode materials on the hybrid system performances.

2. Materials and methods

2.1. Immobilization of the TiO_2/CC , ZnO/CC and $\alpha\text{-Fe}_2\text{O}_3/\text{CC}$ photoanodes

The dimensions (5.0 cm \times 3.0 cm) of carbon cloth (CC) from Ballard Material Products (AvCarb[®]1071 HBC) with nominal thickness 350 μm was used as substrate to prepare the TiO_2/CC , ZnO/CC and $\alpha\text{-Fe}_2\text{O}_3/\text{CC}$ photoanodes through an immobilization method. The CC was cleaned by sonication sequentially in deionized water and ethanol for 30 min. Then, it was dried at 60 °C. The commercially available powder Aerioxide TiO_2 P25 from Evonik Industries AG, Germany, ZnO powder from HmbG Chemicals and $\alpha\text{-Fe}_2\text{O}_3$ powder from Sigma Aldrich were used as photocatalyst in this study. The amount of 2.00 g of photocatalyst powder was added in 30 mL of deionized water to prepare the suspension of the photocatalyst. The cleaned CC was immersed into photocatalyst suspension and was sonicated by using 40 kHz ultrasonic bath with digital temperature control (Model CH-01BM from Ultrasonic Cleaner, China) at 60 °C for 30 min. Then, the CC immobilized with photocatalyst was dried in the oven at 60 °C for 2 h. This process was repeated 3 times before the photocatalyst immobilized CC was annealed in furnace at 300 °C for 2 h. To remove any poorly attached photocatalyst powder, the immobilized TiO_2/CC , ZnO/CC and $\alpha\text{-Fe}_2\text{O}_3/\text{CC}$ photoanodes were immersed in deionized water for a few seconds before it was applied in the PFC reactor as the photoanode.

2.2. Characterization of the TiO_2/CC , ZnO/CC and $\alpha\text{-Fe}_2\text{O}_3/\text{CC}$ photoanodes

All the photoanodes were characterized by using X-ray diffractometer (XRD) from Bruker (Model: D2 Phaser benchtop) with LYNXEYE 1D-detector. The $\text{Cu-K}\alpha$ radiation was used in this XRD analysis at room temperature. The diffraction patterns were recorded using a step size of 0.02° and scan rate of 6° per min with scan range 2θ (20° to 80°). The surface morphology of the immobilized TiO_2/CC , ZnO/CC and $\alpha\text{-Fe}_2\text{O}_3/\text{CC}$ photoanodes was characterized by using scanning electron microscopy (SEM) (Model: JSM 6460 LA from JEOL, Japan).

2.3. Reactors setup and operation

The schematic diagram of PFC-PC system is shown in Fig. SM-1. Two beakers of 0.5 L were used to setup the PFC and PC reactors. Carbon plate with thickness of 3.0 mm was applied as cathode in both reactors. The Fe^{2+} was generated by using 99% purity of iron plate with thickness 1.2 mm as the anode at the PC reactor. All the electrodes were prepared by using the dimensions

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