



Extraordinary catalysis induced by titanium foil cathode plasma for degradation of water pollutant

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HIGHLIGHTS

- In situ hybrid induced photocatalytic method was proposed.
- The degradation rates are dependent on the electrical conduction of cathodes.
- Titanium-based metal foil plays bifunctional roles in the rapid degradation.
- The Au film improves the electrical conductivity and also produces many Au NPs.
- The degradation rate improved up to one order of magnitude.

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ABSTRACT

The present paper reports a rapid and cost-effective bifunctional approach to the degradation of organic pollutants in the aqueous solution. This in situ hybrid induced photocatalytic method involves the advanced oxidation process, and photocatalytic process induced by ultraviolet radiated from the plasma discharge to improve the degradation efficiency. This powerful plasma allows the organic molecules to be cleaved either in the plasma zone or on the plasma/solution interface through hydrogen abstraction and electron transfer. Four parallel metal foil electrodes (i.e. Ta, Cu, Ti and Au coated Ti), used as cathodes in the two-electrode system, were evaluated in terms of their degradable performance to organic pollutants. It was found that the degradation rates are dependent on the electrical conduction of metal cathodes. During the discharge process, the Ti-based foil produces TiO₂ particles, which then act as catalyst in the electrolyte and perform the photocatalytic process along with the plasma discharge process to degrade organic pollutants. It is of particular interest that gold nanoparticles, generated from Au coated Ti foil film during electrode discharging, are less than 5 nm in size and further enhance the TiO₂ photocatalytic activity. In fact, this bifunctional plasma discharge process to the degradation of water pollutant provides an insight into more applications such as chemical conversion, water purification and dust pollution.

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1. Introduction

Titanium dioxide (TiO₂) as a photocatalyst for the destruction of organic pollutants has received great attention owing to its high redox potential, photo-induced hydrophilicity and chemical stability (Bian et al., 2014; Zeng et al., 2016; Hinojosa et al., 2017). However, the large electronic bandgap of pure TiO₂ (3.0–3.2 eV), which restricts its optical absorption light wavelength to less than

390 nm and thus affects the formation of excited state conduction band (e^-) and valence band hole (h^+) pairs in the photocatalytic process (Akpan and Hameed, 2009; Liao et al., 2012; Dette et al., 2014). As such, many methods have been used to improve the visible light responsive of TiO_2 , such as nonmetal/metal doping, semiconductor coupling and surface deposition of noble metals (Pang and Abdullah, 2012; Ayati et al., 2014; Lu et al., 2015; Biswas et al., 2018; Jia et al., 2018). For example, the titanium oxide loaded with mesoporous materials MCM-48 allows the removal of 87–92% of rhodamine B (Rh B) in a higher concentration range from 10^{-4} M to 5×10^{-4} M with a removal apparent rate constant k_{app} up to $4.55 \times 10^{-3} \text{ min}^{-1}$ (Rasalingam et al., 2015). However, the rapid recombination of photo-generated electron-hole pairs in TiO_2 nanoparticles (NPs) gives rise to a low quantum efficiency and poor photocatalytic activity.

Recently, electrical discharge plasma in liquid phase added with TiO_2 powder, that combines the advanced oxidation processes (AOPs) of liquid plasma discharge (LPD) and TiO_2 photocatalytic function, has been used to degrade organic pollutants (Wang et al., 2011; He et al., 2014; Li et al., 2016; Duan et al., 2018). LPD is a powerful discharge process where the plasma is generated in the interface of the electrode and the electrolytic solution, as demonstrated to initiate a variety of chemical and physical effects (Cheng et al., 2016). These include ultraviolet radiation, overpressure shock waves, high electric field and, in particular, formation of chemically various active species, such as $\cdot OH$ radicals, $\cdot H$ radicals, H_2O_2 , and hydrated electrons, which are generated either in the plasma zone or along the plasma-solution interface (Muhammad and Salman, 2001; B. R. Locke et al., 2006; Sen and Singh, 2016). In the LPD process, a large amount of $\cdot OH$ radicals whose oxidative potential is only lower than fluorine, enable the transformation of the majority of the organic compounds into CO_2 and H_2O (Cheng et al., 2007; Sen, 2017). Especially in high conductive electrolyte, the LPD also initiates intensive radiation in a wide range of wavelength (200–1000 nm) (Allagui et al., 2016; Bruggeman et al., 2016). Thus, utilizing ultraviolet radiation in the discharge process, conventional combination technique of LPD with TiO_2 photocatalysis can be an impactful approach to improve the overall energy efficiency and degradation performance.

Nevertheless, almost all researches have been carried out by using the anodic electrode with high voltage to generate plasma with the addition of TiO_2 powder in the electrolyte. The emission characteristics of the radiation generated by LPD were found strongly influenced by the electrolyte conductivity. The increasing of electrolyte conductivity brings about the radiation with higher intense and broader emission spectral lines. Meanwhile, intense of the radiation wavelength reduces with the increase of the distance between added TiO_2 powders and plasma discharge center. In this study, we developed a novel bifunctional method that uses titanium foil as cathode. Upon the supply of the sufficient high voltage, the cathode emits plasma in the electrolyte. On the other hand, the dissolution of the cathode results in the formation of TiO_2 particles. Plasma carries high energy and implements the oxidation process allowing the damage of organic molecules in the solution. In the meantime, some of organic molecules are also decomposed simultaneously by the photocatalysis induced by TiO_2 that is irradiated by plasma. The both processes work together leading to the rapid degradation of the organic environment. In the present paper, four different cathode foils (tantalum, copper, titanium and titanium-coated gold) were selected as cathode to compare with the conventional combination method of plasma discharge in addition to the TiO_2 powders in terms of their degradation performance to organic pollutants at both high and low concentrations. Effect of the conductivity of cathode on the degradation rate was also investigated and discussed. The liquid chromatography-

mass spectrometry (LC-MS) and gas chromatography-mass spectrometry (GC-MS) were used to clarify the possible degradation mechanism of the catalysis-assisted plasma induced degradation method.

2. Experimental section

2.1. Liquid plasma discharge

The degradation was carried out in a two-electrode system, where a 99.9% purity titanium foil (Good Fellow Cambridge Limited Company) with thickness of 0.15 mm was used as the cathode and a platinum foil (30 mm \times 10 mm \times 1 mm, 99.9% purity) as the anode in the 1 M NaCl electrolyte solution. A schematic diagram of this system is shown in Fig. 1. Three parallel metal foils (i.e. tantalum Ta, copper Cu, Ti foil and gold coated titanium Ti–Au) were chosen and used as cathode in the two-electrode system to compare with the combination method of adding the commercial P25 TiO_2 powders (Sigma, 99% purity) in terms of their degradable performance to rhodamine B. The cathode and anode are connected to a low voltage DC power supply. When the voltage exceeds the threshold value, a large number of gas bubbles are produced over the surface of the cathode forming a gaseous film that insulates the electrolyte solution leading to the generation of liquid plasma discharge at the gas-solid interface in the solution. The value of threshold voltage is dependent on the electrolyte. In the present study, NaCl mixed with Rh B solution was used. During the degradation process, water bath was employed to control the temperature of electrolyte. Magnetic stirring at 300 rpm was also maintained to ensure the uniform concentration of Rh B (Sigma) in the solution.

2.2. Material characterization

Upon the completion of liquid plasma process, the obtained products were collected with the help of centrifugation followed by washing for several times with deionized water to remove the NaCl salt. After drying for 10 h in oven, The crystallinity and phases of the samples were characterized using a thin film X-ray diffractometer (XRD, XRD-6000, Shimadzu, Japan), which employs Cu $K\alpha$ radiation ($\lambda = 0.154059 \text{ nm}$) under 2θ mode with X-ray grazing angle of 1.5° . All samples were placed at the stage height of 7.3 mm and scanned at the rate of 1° min^{-1} within the range $20^\circ \leq 2\theta \leq 80^\circ$. The morphology and internal structure of the nanocomposites were further examined by using field emission scanning electron microscope (FESEM, JEOL JSM-7600F) and high-resolution transmission electron microscope (HR-TEM, JEOL JEM-2100F) at an

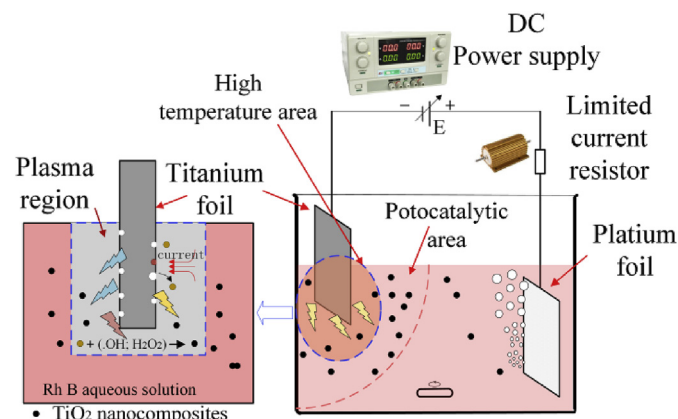


Fig. 1. Schematic diagram of the two electrode system.

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