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A photoelectrochemical device for water splitting using oligoaniline-crosslinked $[\text{Ru}(\text{bpy})_2(\text{bpyCONHArNH}_2)]^{+2}$ dye/ IrO_2 nanoparticle array on TiO_2 photonic crystal modified electrode

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ABSTRACT

This article describes the construction of photoelectrochemical cell system splitting water into hydrogen and oxygen using UV–vis light under constant applied voltage. Oligoaniline-crosslinked 2-(4-aminobenzyl)malonic acid functionalized $\text{IrO}_2 \cdot n\text{H}_2\text{O}$ nanoparticles and visible light absorbing dye, $[\text{Ru}(\text{bpy})_2(\text{bpyCONHArNH}_2)]^{+2}$ arrays on titanium dioxide (TiO_2) photonic crystals modified electrodes were used as photoanode, and nanostructures based on bonding of Pt nanoparticles by using electropolymerization on poly 4-(2,5-di(thiophene-2-yl)-1H-pyrrol-1-yl)benzenamine P(SNS-NH₂) conducting polymer modified gold electrode acted as cathode. Each component in anode and cathode of the system was characterized successfully using the methods related. Some optimization studies such as the molar concentration ratio of $[\text{Ru}(\text{bpy})_2(\text{bpyCONHArNH}_2)]^{+2}$ dye to $\text{IrO}_2 \cdot n\text{H}_2\text{O}$ nanoparticles, the optimum cycle number of each components and thickness of TiO_2 film were performed in order to investigate the system performance. Furthermore, the photocurrent generation capacity of the photoanode against oxygen resulting and UV stability experiments of photoanode were also investigated. After obtained all necessary informations and improvements of the system, the cell was constructed, and corresponding hydrogen gas evolution from water splitting was calculated as 1.25×10^{-8} mol/cm² by using a gas chromatography (GC). The cell generated a photocurrent with a quantum yield of 3.5%.

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Introduction

Semiconductor-based photocatalytic water splitting using solar irradiation is recognized as one of the ideal approaches to a clean and renewable energy source [1–7]. Photo-electrochemical water splitting was discovered by Fujishima and Honda in 1972 for the first time [8]. In their system, a titanium dioxide (TiO_2) photoanode was exposed to ultraviolet light (UV), producing oxygen and hydrogen at the anode and cathode, respectively. Since then, many groups have attempted to modify this system using visible instead of UV light, and many of these systems share common components [9–12]. A simple way of studying visible light-driven water splitting is to use a dye-sensitized semiconductor oxide material in a photo-electrochemical cell with separate anodic and cathodic compartments to avoid the mixing of H_2 and O_2 . Four important components are required to build a light-induced electrochemical system for fuel generation: i) an efficient light-harvesting material with a high molar extinction coefficient (ϵ) of approximately $90,000 \text{ L mol}^{-1} \text{ cm}^{-1}$ for natural chlorophylls; ii) a molecular donor–acceptor pair for effective charge separation; iii) a robust and efficient water oxidation catalyst; and iv) a transparent conducting electrode surface or a semiconducting material [13–16]. If a semiconductor is used as a light absorber, molecular donor–acceptor pairs are not necessary for charge separation because both light absorption and charge separation are performed by the semiconductor. However, the chemical architecture of a synergetic blend of a water oxidation complex with a light-harvesting unit to trigger high-efficiency photo-electrochemical water oxidation is the major challenge to the use of semiconductors as light absorbers [17]. Similarly, fast charge recombination in most suitable semiconductors occurs much faster than the redox reactions of water, resulting in poor quantum yields [3,18,19]. To meet these challenges, increasing research efforts have now been put into the design and development of new photocatalytic materials (or composites) as well as the novel photocatalytic mechanisms that may lead to further improvement of photoconversion efficiency.

The first example of a functional system for visible-light-driven water oxidation with molecular components was developed by Mallouk and coworkers [20]. In this system, a nanoparticulate anatase TiO_2 surface is sensitized with a heteroleptic ruthenium–polypyridyl chromophore bound through a phosphonate anchor. An IrO_2 nanoparticle catalyst is bound to a ruthenium chromophore through a malonate linkage. The system produces oxygen when illuminated with visible light under a small bias voltage. However, the quantum yield is reportedly 0.9%, and the coulombic efficiency of oxygen production is roughly 20% by Mallouk and coworkers. The relatively poor performance of this system is attributed to a much faster recombination of electrons from the TiO_2 conduction band and the oxidized dye than from the electron transfer between the iridium catalyst and the oxidized dye. Although this system is inefficient, it represents a proof-of-concept that water splitting can be performed using a sensitizer-based photosystem. The low quantum efficiency can be understood in terms of three problems that can be addressed by design at the molecular level. The simplest of

these is the needed to make catalyst particles that are connected to only one sensitizer molecule, in order that each sensitizer can bind to both TiO_2 and $\text{IrO}_2 \cdot n\text{H}_2\text{O}$. A second problem is the needed to slow down the back electron transfer reaction, for example, by changing the distance between redox partners, and a third is to speed up the electron transfer from Ir(IV) to Ru(II) [15,21]. To overcome these problems, extensive research efforts are directed toward photo-electrochemical applications of functionalized photoanode/cathode electrodes with novel ligand or anchoring groups and to the design of solar cells, especially because the charge ejection processes lead to low photocurrent yields or even to the elimination of photocurrent. For enhancing the light-electrical energy conversion yield, charge separation of the electron–hole species should be assisted. To this end, coupling relay units to semiconductors associated with electrodes leads to charge separation and enhanced photocurrent generation by trapping the conduction-band electrons in the relay sites and effectively transferring electrons to the electrode. Alternatively, the electrochemical crosslinking of semiconductor nanoparticle monolayers onto electrodes by charge-carrying oligoaniline units has been suggested as a means to facilitate charge separation and to enhance photocurrent generation [22].

In the present study, a photoelectrochemical solar cell was designed using a photoanode with the oligoaniline cross-linked bridging units to serve as both sensitizers ($\text{Ru}(\text{bpy})_2(\text{bpyCONHArNH}_2)^{+2}$) and molecular bridges connecting the catalyzer (2-(4-aminobenzyl)malonic acid functionalized $\text{IrO}_2 \cdot n\text{H}_2\text{O}$ nanoparticles) to a metal oxide semiconductor (TiO_2), and using a photocathode including gold electrode coated with Poly(SNS- NH_2) functionalized platinum nanoparticles (Fig. 1). The incorporation of oligoaniline crosslinked bridging units into this device enhances the photocurrent in visible light as well as the quantum yield and efficiency of oxygen/hydrogen production. This is achieved due to strong attachment to the TiO_2 surface, efficient charge separation, and suitable oxidation potential by providing a path for the effective trapping of the conduction-band electrons and their transport to the electrode.

Experimental

Deposition of TiO_2 thin films on PMMA nanofibers by hot filament chemical vapor deposition

Titanium dioxide thin films were deposited conformally over electrospun polymethyl methacrylate (PMMA) fibers using hot filament chemical vapor deposition method. Nanotubular TiO_2 structure was clearly identified via SEM (scanning electron microscope). It was found that this structure was ideal for good photocatalytic activity because of its high surface area per unit volume ratio. FTIR (Fourier Transform Infrared Spectroscopy) and XPS (X-ray Photoelectron Microscopy) results show the formation of stoichiometric TiO_2 , and the crystalline form of the final nanotubes was found to be anatase (1 0 1) after XRD analysis. Details deposition procedures of TiO_2 film can be found in our reference [23].

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