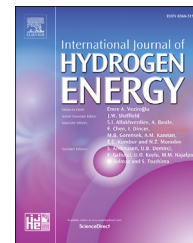




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Design of cocatalyst loading position for photocatalytic water splitting into hydrogen in electrolyte solutions

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ABSTRACT

Photocatalytic water splitting into hydrogen is a very attractive and desirable technology to realize sustainable and renewable green energy conversion. Up to now, many research results have confirmed that cocatalyst such as Pt is essential for a high efficiency photocatalytic H₂ evolution system. In a traditional view, the cocatalyst should be closely combined with the photocatalyst to achieve a high H₂ or O₂ photo-productive rate. In this work, an unusual point has been put forward that the suitable loading position of cocatalyst Pt for film-type TiO₂ catalyst is on the bare Fluorine-doped tin oxide (FTO) substrate instead of on the surface of TiO₂ in the electrolyte solutions. Especially, in acidic electrolyte, the hydrogen production rate of this new designed catalyst with Pt loaded on FTO (TiO₂-Pt/FTO) reaches 2.4 times that of the common catalyst with Pt loaded on TiO₂ (Pt/TiO₂-FTO). According to the experiment results, it is supposed that this loading way of cocatalyst on the substrate can construct a self-bias photocatalytic electrochemical cell system, drive electrolyte ions' movement directionally, and obtain high photocatalytic H₂ production efficiency. The universality of this innovation has been verified by CdS and CdS@TiO₂ film-type catalysts. This study provides a new guide in exploring high-efficiency film-type photocatalytic system for water splitting into hydrogen in the electrolyte solution.

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Introduction

Photocatalytic water splitting for H₂ production using semiconductor materials is one of the ideal methods to solve global

energy and environmental problems. In order to improve the photocatalytic performance, considerable efforts have been devoted to developing efficient photocatalytic system for water splitting, such as adjusting the construction and morphology of the semiconductors, adding cocatalysts and

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selecting suitable sacrificial agent [1–4]. Among various photocatalytic materials, in recent years, one-dimensional (1D) nanoarray structured film-type photocatalysts have aroused great attraction for the development of both direct solar-to-hydrogen (DSTH) and indirect photoelectrochemical (PEC) devices [5–7]. Especially, TiO_2 and CdS nanoarrays film photocatalysts were two of the most extensively studied catalysts so far, because of the high stability and good availability for TiO_2 under UV light irradiation, and high photocatalytic activity for CdS under visible light irradiation [8–10]. However, both TiO_2 and CdS have also suffered their instinctive drawbacks, such as the limited electron-hole separation rate for TiO_2 and evidently serious photocorrosion for CdS, which extremely restrict their practical applications. To overcome these problems, the highly effective cocatalysts and suitable sacrificial agents are indispensable in a DSTH system [11]. Recently, people have sparked a huge surge in the developing of various cocatalysts, such as transition metal sulfides, carbides and phosphides [12]. According to the common sense, these cocatalysts are typical directly loaded on the surface of TiO_2 or CdS photocatalysts to achieve a high photocatalytic performance [12–15].

However, very recently, we found that the photocatalytic H_2 production rate of CdS/ TiO_2 -FTO was significantly enhanced by using a Pt-wire connected with FTO substrate rather than Pt nanoparticles (NPs) loaded on the surface of CdS/ TiO_2 [16]. Therefore, it is proposed that the loading position of cocatalyst may be an important factor affecting the photocatalytic activity for film-type photocatalyst, but there are few reports so far to reveal this issue. At the same time, we also realized that such a Pt-wire connected CdS/ TiO_2 monolithic photocatalyst system constituted a simple self-bias photoelectrochemical cell (SPEC) [17–19], which is desirable to realize the energy saving hydrogen generation under solar light irradiation without external electric powers [20–22]. As is well known, the electrolyte solution plays a key role in a PEC cell and as well as SPEC system, and the transfer process of proton or hydroxide has large effect on water splitting reactions in acidic or alkaline electrolyte solutions [23–25]. However, to the best of our knowledge, few studies have reported the effects of cocatalyst loading position and the electrolyte species on the photocatalytic hydrogen evolution for film-type photocatalysts. Herein, we synthesized branched TiO_2 nanorod arrays (TiO_2 NRs) on FTO substrate and used it as a representational film-type photocatalyst. The typical Pt NP, which had the highest efficiency for decreasing the H^+/H_2 reduction potential and nearly fell at the apex of the “volcano” plots for various of recorded metal materials [26], was selectively loaded on the photocatalyst film by a controlled ion beam sputtering deposition (IBSD) method. Hereby, two typical DSTH and SPEC photocatalyst systems were synthesized, i.e. Pt NPs selectively loaded on photocatalyst (Pt/ TiO_2 -FTO) for DSTH system or on FTO substrate (TiO_2 -Pt/FTO) for SPEC system, as shown in Fig. 1A and B. Different electrolyte aqueous solutions were employed to further understand the influence of cocatalyst loading position on photocatalytic water splitting into hydrogen.

The photocatalytic hydrogen production rates indicate that the optimum loading position of Pt NPs for film-type TiO_2 catalyst is on the surface of TiO_2 in pure water or water-

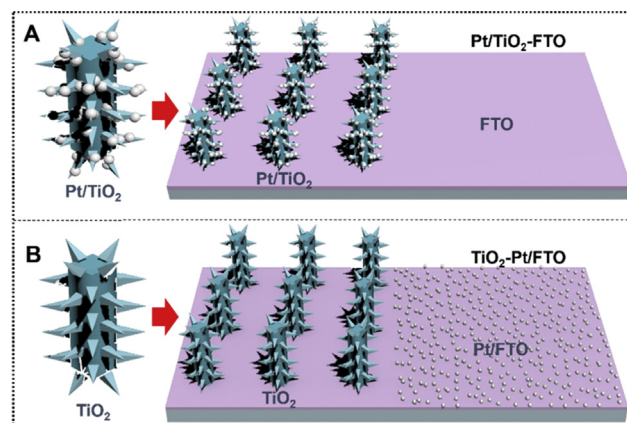


Fig. 1 – Schematic view of the designed Pt/ TiO_2 -FTO (A) and TiO_2 -Pt/FTO (B) films.

methanol systems. However, in the electrolyte reaction solutions, an unusual viewpoint has been put forward that the most suitable loading position of Pt cocatalyst is on the bare conductive substrate (FTO) instead of on the surface of TiO_2 . To further verify the universality of this novel viewpoint, CdS and CdS@ TiO_2 film-type catalysts have also been designed and prepared. The results demonstrate that this innovation is very favorable for CdS-based photocatalysts using electrolyte solution (Na_2S) as stabilizing agent and sacrificial agent. This study provides a new train of thought on designing high-efficiency SPEC systems for water splitting into hydrogen in the electrolyte solutions such as seawater, which has a great practical applications potential [27].

Experimental

Synthesis of branched TiO_2 nanorod arrays

TiO_2 NRs was synthesized according to the previously reported method with slight modifications [28]. In a typical synthesis, a piece of $3 \times 6 \text{ cm}^2$ FTO substrate, which had been ultrasonically cleaned in acetone, deionized water and alcohol, respectively, was placed into autoclave at an angle against the wall of Teflon-liner with the conductive side facing down. Then, a mixed solution containing 15 mL of aqueous HCl (38%), 15 mL of deionized water and 400 μL of titanium butoxide (97% Aldrich) was carefully injected into the above 100 mL autoclave and kept the liquid level to about half height of the glass. A hydrothermal synthesis was conducted at 170 $^\circ\text{C}$ for 7 h. And after the autoclave was cooled down to room temperature, the half-growth TiO_2 NRs on FTO glass was taken out and rinsed with deionized water and alcohol for several times. The obtained TiO_2 NRs was then branched in a mixed solution of 30 mL H_2O , 300 μL HCl and 300 μL TiCl_3 at 70 $^\circ\text{C}$ for 1 h. The finally synthesized sample was simply noted as TiO_2 -FTO.

Synthesis of CdS-FTO nanorod arrays

The CdS NRs were synthesized on FTO substrate with a similar condition as Li et al. reported [29]. The difference is that CdS

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