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Near-complete charge separation in tailored $BiVO_4$-based heterostructure photoanodes toward artificial leaf

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ABSTRACT

As an artificial leaf, a tandem device for zero-bias solar water splitting is a capable solution for practical hydrogen production. Despite a promise, poor charge transport of $BiVO_4$ hampers photoelectrochemical performances under front-side illumination, which is a hindrance to the tandem system. Herein, we design a new photoanode comprising nanoporous $BiVO_4$ and SnO_2 nanorods focused on the charge separation via structural and interfacial engineering. $BiVO_4/SnO_2$ photoanode exhibits not only remarkable charge separation efficiency of 97% but also, by loading NiFe as a co-catalyst for water oxidation, high photocurrent density of 5.61 mA cm^{-2} at 1.23 V versus the reversible hydrogen electrode under front-side 1 sun illumination. Consequently, a tandem cell comprising $NiFe/BiVO_4/SnO_2$ photoanode and perovskite/Si tandem solar cell generates an operating photocurrent density of 5.90 mA cm^{-2} with a solar-to-hydrogen conversion efficiency of 7.3% in zero-bias. This work would be a significant step to develop spontaneous solar hydrogen production.

1. Introduction

Photoelectrochemical (PEC) water splitting, which directly converts sunlight into hydrogen fuel, is one of the most sustainable solutions to stockpile solar energy. However, low solar-to-hydrogen (STH) conversion efficiency compared to its potential, has been a barrier to the commercialization, unlike a highly efficient photovoltaic (PV) system. The photovoltage generated by a single photoanode is insufficient to reach a required voltage (1.23 V + overpotential) for the water splitting reaction, resulting in a nonspontaneous reaction requiring external bias [1,2]. In this single absorber system, a trade-off between photovoltage generation and light absorption according to the bandgap limits improving PEC performances [3,4]. Thus, a tandem system consisting of dual absorbers with an optimal bandgap combination has been widely studied to maximize light harvesting by separating each absorption wavelength range [5–10]. A PEC-PV tandem system, especially, is favorable for spontaneous PEC water splitting in zero-bias due to the high STH conversion efficiency in terms of photovoltage production [2]. In this system, high photocurrent density in water oxidation of front photoanode is key to maximize the efficiency of the entire cell.

$BiVO_4$, which is one of the most promising photoanode materials, has a suitable band gap and band edge position for water oxidation, delivering a high theoretical photocurrent density of 7.5 mA cm^{-2} and STH conversion efficiency of 9.2% [11–13]. These PEC characteristics, however, markedly diminish under front-side illumination, and this issue hinders the implementation of PEC-PV tandem cell that requires high front PEC performance. Such a drawback is caused by the slow electron transport limiting the photocurrent generation of $BiVO_4$ [14,15]. Namely, the short carrier diffusion length (~70 nm) of $BiVO_4$ inhibits charge separation [16]. To enhance the charge separation efficiency of $BiVO_4$, two representative strategies are used; the first one is the nanostructuring of $BiVO_4$ [11,17,18]. Kim et al. fabricated a nanoporous $BiVO_4$ photoanode with charge separation efficiency of 90% [19], and Kuang et al. found an optimal average diameter (≈120 nm) of $BiVO_4$ to maximize PEC properties under front-side illumination [15]. The distance between the electrode inside and the electrolyte interface was structurally engineered as much the carrier diffusion length to make efficient charge transport without a reduction of light absorption. The

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