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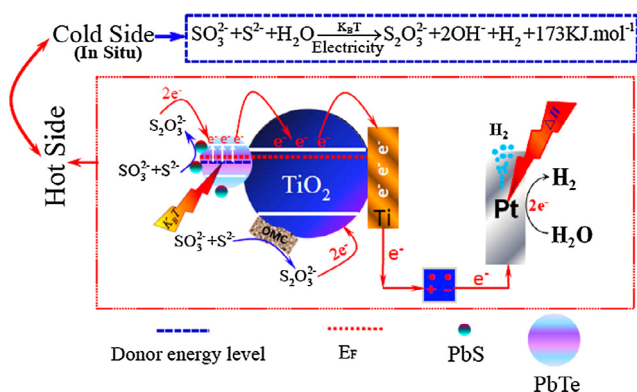
Enhanced hydrogen production of PbTe-PbS/TNAs electrodes modified with ordered mesoporous carbon



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GRAPHICAL ABSTRACT



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ABSTRACT

PbTe-PbS/ TiO_2 nanotube arrays (PbTe-PbS/TNAs) were synthesized by the successive ionic layer adsorption and reaction (SILAR) followed by linear sweep voltammetry (LSV). Using Nafion as a binder, ordered mesoporous carbon was cast on these materials to generate the modified electrodes OMC/PbTe-PbS/TNAs. It was demonstrated that the electrode modification with OMC could enhance the charge transfer between the electrode surface and the electrolyte solution, improve the energy band bending of the electrode/electrolyte interface, increase the active electrochemical surface area of the electrode, and reduce the overpotential of the electrode reactions. Under ambient conditions, the short circuit current density (37.84 mA cm^{-2}) and the active electrochemical surface area (29 mF cm^{-2}) of the OMC/PbTe-PbS/TNAs electrode were 27.49% and 36.79% higher than that of PbTe-PbS/TNAs (29.68 mA cm^{-2} and 21.2 mF cm^{-2}), respectively. A particularly important feature of the OMC modification is that the hot electron extraction capability of the PbTe-PbS/TNAs electrode remained in the new system to provide rapid enhancement of short circuit current density upon increasing temperature. The OMC/PbTe-PbS/TNAs electrode registered a hydrogen generation rate of $11 \text{ mL cm}^{-2} \text{ h}^{-1}$, with an energy efficiency of 98.79% and a heat efficiency of 43.03% under cell voltage of 1.0 V at 55 °C.

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1. Introduction

Hydrogen is a clean renewable energy source and has been pursued as one of the promising alternatives to fossil fuels due to increasing global energy demands and climate change [1,2].

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Splitting water via electrochemical reactions is considered to be the most promising approach towards hydrogen production. Efficient water splitting requires high active electrocatalysts to lower the overpotentials of the hydrogen evolution reaction (HER) at the cathode and the oxygen evolution reaction (OER) at the anode, eventually in order to facilitate the hydrogen production [3]. Precious metals (Pt, Pd, Ir, Ru, etc.) provide an excellent electrocatalytic surface that demonstrates low overpotentials, exemplified by an anodic electrocatalyst with the formula $\text{Ir}_x\text{Ru}_y\text{Ta}_z\text{O}_2$ registering an electric consumption of $42 \text{ kW h kg}^{-1}\text{H}_2$ and an energy efficiency of 94% [4]. However, the high cost and scarcity of the precious metals hinder the application in industry. Thus, it is highly expected to develop non-precious metal catalysts that can utilize low value energy such as the low-temperature waste heat under the premise of both low materials cost and reduced energy input.

A narrow band gap II–VI semiconductor, PbTe has an fcc cubic crystallographic structure with a lattice constant of 0.646 nm, constituted of Pb^{2+} and Te^{2-} connected by metallic bonds. PbTe semiconductors are well known for their symmetric energy band structure, a narrow band gap of 0.31 eV, a large Bohr exciton radius of 46 nm, good conductivity of $28.29\text{--}99.50 \text{ S cm}^{-1}$, and large electron and hole transport rates of 1730 and $780 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$. PbTe semiconductors are regarded highly promising in applications such as thermoelectric conversion [5], far infrared detection (3–30 μm) [6], and photoelectro-chemical devices [7]. More recently, a novel system that may greatly expand the application of PbTe semiconductors has been proposed [8]. This design can generate a novel pathway to produce chemical energy from low quality waste heat. In the study, we prepared PbTe-PbS/TiO₂ nanotube arrays (PbTe-PbS/TNAs) by using the successive ionic layer adsorption and reaction (SILAR) followed by linear sweep voltammetry (LSV). At 70 °C and cell voltage of 1.0 V, the system registered an energy efficiency of 88.5% and a heat efficiency of 49.9%. Ordered mesoporous carbon (OMC), since its discovery in 1999 [9], has attracted a great deal of attention owing to its distinguished properties of extremely well-ordered pore structure, high specific pore volume, high specific surface area, large adsorption capacities, low electron-transfer resistance, and chemical inertness. These advantages make it suitable for applications in adsorption [10], chemical sensors [11], catalyst carriers [12], energy storage [13] and new functional composite materials [14]. As reported, OMC CMK-3 offered faster electron-transfer kinetics than glassy carbon, fullerenes, or even CNTs, resulting in increasing the intensity of voltammetric peaks and lowering the overpotentials [15]. The OMC modified glass carbon electrode (OMCs/GCE) can remarkably enhance electrocatalytic activity towards the oxidation of ractopamine with a great increase of peak current [16], and reduce the oxidation overpotential and improve the current response of phenols to some extent [17]. Because of the good absorptivity, conductivity, electron storage/releasing capacity of OMC, this family of materials can effectively improve the adsorption and reaction of chemical species inside the mesopores, enhance the charge transfer efficiency of the key reaction elemental steps, thus greatly increasing reaction rates. To further promote the catalytic reaction kinetics and improve the energy efficiency in the current work, we apply OMC as the charge transport layer and electrochemical reaction interface of the PbTe-PbS/TNAs electrode, and drive the anodic reaction ($\text{SO}_3^{2-} + \text{S}^{2-} \rightarrow \text{S}_2\text{O}_3^{2-} + 2\text{e}^-$) proceeding both at the OMC surface and the PbTe-PbS nanocrystal surface. The results show that OMC modification can reduce the resistance of the electrode process, improve the energy band bending of the electrode/electrolyte interface, increase the active electrochemical surface area of the electrode, enhance the charge transfer between the electrode surface and the electrolyte solution and charge transport

within the OMC phases, decrease the polarization of the electrode, as well as enhance short circuit current density upon increasing temperature of PbTe/PbS/TNAs electrode. The overall results are the enhanced electrocatalytic activities towards hydrogen production.

2. Experimental

2.1. OMC/PbTe-PbS/TNAs preparation

The preparation method and procedure details of PbTe-PbS/TNAs can be found in our published work [18,19]. The OMC was first dealt with hydrophilic pretreatment prior to deposition. 0.1 g CMK-3 was added to H_2O_2 (10%), stirred at 60 °C for 2 h. After cooled to room temperature, the sample was centrifugated three times and decanted of the supernatant. The solid was placed in an oven (60 °C) before following experimental procedures. 5.0 mg hydrophilic OMC was added to 100 μL Nafion solution (5%) mixed in 1 mL ethanol, and ultrasonicated for 30 min to obtain the OMC/Nafion dispersion. The dispersion was applied to the PbTe-PbS/TNAs electrode by using a micro-syringe with an amount of 5–15 $\mu\text{L cm}^{-2}$. The electrode was then dried at 60 °C for 2 h and labeled OMC/PbTe-PbS/TNAs. For comparison, a PbTe-PbS/TNAs electrode was modified with Nafion only and labeled Nafion/PbTe-PbS/TNAs, and a PbTe/TNAs electrode modified with OMC/Nafion without the linear sweep voltammetry sulfuration was labeled OMC/PbTe/TNAs.

2.2. Characterization method

X-ray diffraction (XRD) analysis was carried out on a Philips XPert pro MPD to investigate the crystalline phases of the samples. Scanning electron microscopy (SEM) was performed on a JSM-7500F to provide the surface morphology and chemical composition of the samples. Fourier transform infrared spectrophotometry was applied on a Nicolet 6700 spectrometer to examine the functional groups and contents of the samples. Raman spectrometry was conducted on a LabRAM HR spectrometer to analyze the sample surfaces.

2.3. Electrochemical measurements

A CHI660E electrochemical work station was used for all electrochemical measurements, including linear sweep voltammetry (LSV), electrochemical impedance spectroscopy (EIS), Tafel curves, Mott-Schottky curves. The three electrode system was constituted of a sample working electrode, a platinum counter electrode, and an SCE reference electrode. The electrolyte was $0.5 \text{ mol L}^{-1} \text{ Na}_2\text{SO}_3 + 0.5 \text{ mol L}^{-1} \text{ Na}_2\text{S}$ solution. During LSV, the starting and ending voltages were -0.5 V and $+0.5 \text{ V}$, respectively, with a scanning rate of 10 mV s^{-1} . The EIS measurement was conducted at a voltage of the open circuit voltage of each sample in the frequency range of $0.01\text{--}10^5 \text{ Hz}$. The Tafel analysis was performed in the potential range of open circuit voltage $\pm 0.15 \text{ V}$ with a scanning rate of 0.05 mV s^{-1} . Unless specified, all electrochemical measurements were carried out at 25 °C.

2.4. Electrochemical hydrogen production

The hydrogen production experiments were carried out at a direct current potentiostat in a two-electrode cell. The sample was the anode and a Pt foil the cathode, with an inter-electrode distance of 10 cm under cell voltages from 1.0 V to 1.5 V at a fixed temperature of 55 °C. The other conditions were the same as the LSV test. The volume of produced H_2 gas was measured via water

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