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Photocatalytic water splitting to produce hydrogen using multi-junction solar cell with different deposited thin films

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ABSTRACT

Multi-junction solar cell (MJSC) was integrated into an H-type reactor system to carry out water-splitting reaction without external circuit and bias. In the H-type reactor, hydrogen and oxygen could be evolved separately. Different kinds of materials were deposited on the bottom cell to prevent corrosion. These materials include silver, platinum, and iron oxide layers which were prepared via an electron beam-induced deposition (EBID) method. Under visible-light irradiation, hydrogen production was achieved by using the MJSC; however, only a small amount of oxygen was evolved. The photogenerated holes may oxidize the photoelectrode instead of water, meaning that Ge subcell or the deposited thin film was oxidized during the photoreaction. The thin films ranked from highest hydrogen evolution to lowest are: Pt > bare Ge > Fe₃O₄ > Ag. The photoelectrode deposited with metal thin film showed better activity due to its higher conductivity, except Ag. The poor performance of photoelectrode with silver thin film was due to the formation of silver oxide during photoreaction which may trap electrons, lowering the amount of electrons available to carry out water reduction. The chemical bias for H-type reactor system was eliminated by using sodium sulfate as the electrolyte. The ratio of evolved hydrogen to oxygen was near 2:1, suggesting that real water-splitting reaction can be achieved by using the MJSC incorporated H-type reactor system without chemical bias.

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1. Introduction

Due to the problems of energy shortage and global warming, alternative energies have been developed to reduce the use of fossil fuels. Alternative energies are renewable and low in carbon emission. Some examples are wind energy, hydro energy, and solar energy. Among these clean energies, solar energy is very promising because it is free, easily accessible, and inexhaustible. However, solar energy is intermittent, meaning that it is only available in the presence of sunlight. Therefore, a medium to store solar energy is required.

Although solar energy is an ultimate energy resource, its application is limited by season, location, and time. Therefore, most researchers attempt to convert solar energy to another kind of energy form, such as chemical energy. Hydrogen is the best candidate for solar energy storage because it is energy efficient, carbon free, and abundant in nature. Solar hydrogen production can be realized via thermochemical water splitting [1], photobiological water splitting [2,3], and photoelectrochemical water splitting [4,5]. For thermochemical water splitting, large-scale

solar concentrator system and appropriate heat-resisting materials are essential, which makes this technique often costly. On the other hand, photobiological water splitting has the disadvantages of low hydrogen yield, and long reaction time. Furthermore, additional process is needed to separate the generated hydrogen. Therefore, photoelectrochemical water splitting seems to be a better technique to produce hydrogen via solar energy. Comparing with thermochemical and photobiological water-splitting techniques, photoelectrochemical water splitting has the advantages of reasonable solar-to-hydrogen efficiency, low process cost, separate hydrogen and oxygen evolution upon reaction, and small system for household application.

A photoelectrochemical water-splitting system mainly comprises of a reactor, and a photoelectrode which is made of photoactive semiconductor materials. The working principle of photoelectrochemical water splitting is simple. First, electrons and holes are generated when photoelectrode is irradiated by light with energy greater than the bandgap of the material. These electrons and holes will reduce and oxidize the chemical species on the surface of the photoelectrode, respectively. If a counter electrode, such as Pt electrode, and an external circuit are present in the system, electrons can be transferred to counter electrode via external circuit to conduct reduction reaction. An example of photoelectrochemical water splitting was first demonstrated by

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Fujishima and Honda in 1972 [6]. In many water-splitting studies [7,8], TiO_2 was chosen as the material for making photoelectrode because it is stable, non-corrosive, environmental-friendly, abundant, and low cost. However, reaction involving TiO_2 has showed low photoactivity under solar-light irradiation mainly due to the recombination of photogenerated electrons and holes, the backward reaction of hydrogen and oxygen to form water, and the inability of TiO_2 to utilize visible light. In order to harvest more visible light, which contributes about 50% of sunlight, several modifications of TiO_2 , such as ion doping [4,9,10] and dye sensitization [11–13], have been demonstrated. Despite modification of TiO_2 can improve the absorption of visible light, the efficiency of hydrogen production is still low. Therefore, many researches were focused on the development of other light-response materials to improve hydrogen yield [14–16]. III–V group materials [17–19] and multi-junction solar cells [20,21] were considered as potential photoelectrodes to enhance water-splitting efficiency. Among these materials, the multi-junction solar cell (MJSC), which contains InGaP, GaAs, and Ge sub-cells, could convert solar energy to electricity with high efficiency [22,23]. Therefore, we attempt to utilize modified MJSC to perform water-splitting reaction using H-type reactor system [24] in which separate hydrogen and oxygen evolution can be realized.

2. Experimental

2.1. Characterization of MJSCs

The multi-junction solar cells (MJSCs) used in this study were provided by the High Concentration Photovoltaic R and D Project in the Institute of Nuclear Energy Research. The MJSC was comprised of InGaP, GaAs, and Ge subcells, as shown in Fig. 1, and is sometimes referred to as the triple-junction solar cell. The tunnel diode between two subcells provides a low electrical resistance bridge for connecting two p–n junctions. Since the structure of the MJSC is consisted of three n-on-p junctions, the cathode (top cell), in theory, can perform water reduction to generate hydrogen while the anode (bottom cell) can perform water oxidation to generate oxygen.

In general, solar cells are used to convert sunlight into electricity. To efficiently collect the photogenerated electrons, front and back metal contact will be deposited on the top and bottom subcell, respectively. In our MJSC, patterned front metal contact made up of Ni/Ge/Au/Ag/Au layers was deposited on the top side of InGaP subcell while back metal contact of Au/Ag/Au layers was deposited on the bottom side of Ge subcell. Fig. 2 shows the structure of the MJSC with metal contacts. The photovoltaic efficiencies of the MJSCs were measured in order

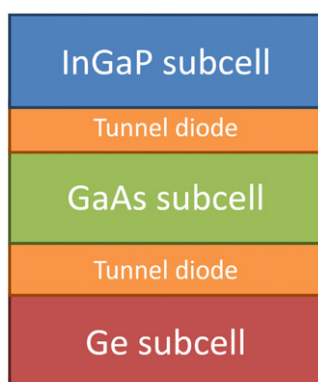


Fig. 1. Structure of a InGaP/GaAs/Ge triple-junction solar cell.

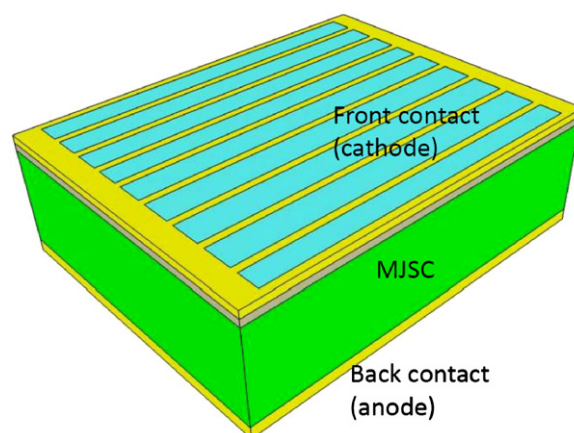


Fig. 2. Structure of MJSC photoelectrode with front and back metal contact.

to characterize their performance. Keithley 2602A sourcemeter system was employed to provide voltages between 0 and 2.5 V at a scan rate of 0.05 V/s and to record the photocurrent generated under light irradiation. The light source used to perform photovoltaic efficiency measurement was AM 1.5 G with an intensity of 100 mW/cm^2 . The light was produced from Xe and halogen lamp equipped with suitable filters (WACOM corp.) In order to analyze the external quantum efficiency of each subcell, spectral response was measured by the Solar Cells Spectral Response Measurement System (Enli Technology Co., Ltd.) equipped with xenon arc lamp and halogen lamp as the light source. With the help of monochromator, the incident light can be adjusted to a specific spectral wavelength within the range 350–1750 nm. To measure the photocurrent of each subcell, current generated from the subcell of interest under specific spectral band irradiation was collected while bias laser was applied to excite the other two subcells. The quantum efficiency was then calculated from the ratio of generated electrons (from the photocurrent) to incident photons (from the light intensity).

2.2. Thin-film preparation and hydrogen production of the MJSC

In this study, the front contact of MJSC without modification was used to collect photogenerated electrons and to provide the site for hydrogen evolution via water reduction. On the other hand, different thin films, including metal (silver and platinum) or oxide (iron oxide) layers, were deposited on the back contact of the MJSC to provide the site for oxygen generation and to prevent the corrosion of the bottom subcell. All of the thin films were prepared by the electron beam-induced deposition (EBID) method. During the deposition, the MJSC was placed above the granular target, such as Ag, Pt, and Fe_3O_4 , with the bottom subcell facing the target. The thickness of thin films was well-controlled at around 300 nm by using a quartz crystal monitor. An additional photoelectrode with thicker Pt film of 500 nm was prepared for comparison.

For characterization, iron oxide (Fe_3O_4) layer was studied by X-ray diffraction (XRD) to reveal its crystallinity. X-ray diffraction pattern of Fe_3O_4 was obtained with a Bruker-D8-ADVANCE X-ray diffractometer in the diffraction angle (2θ) between 20 and 80° , using Cu $K\alpha$ radiation as the source. The surface morphology and chemical composition of the thin film were revealed by scanning electron microscope-energy dispersive spectrometry (SEM-EDS JSM-7600F, JEOL corp. and X-MAX80, OXFORD corp.) under the magnification of $100,000\times$.

To carry out water-splitting reaction, the photoelectrode (with the size of $2 \times 1 \text{ cm}^2$), which comprised front contact/MJSC/thin film (FC/MJSC/TF), was fixed by a clamp and assembled in the

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