Graded Bandgap Device Architecture of Perovskite Solar Cells

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ABSTRACT

This study purposes graded bandgap design for lead-free perovskite solar cells which aim to maximize the solar spectrum with good output current and and better power conversion efficiency (PCE) by improving the solar cell archtecture. Titanium dioxide (TiO_2) was used as electron transport layer (ETL) and Spiro-OMeTAD was used as hole transport layer (HTL) due to its facile implementation and high performance in electronic device. Lead-free bismuth oxyiodide (BiOI) was chosen to replace conventional lead-halide perovskite absorber layer. BiOI has iso-electronic properties to lead-halide perovskite with high efficient light absorption, high thermal stability and photocatalic activity, excellent photo-generated charge carrier. The variation of ioidine concentration in BiOI estabishes bandgap tuning and conductivity type of the layer BiOI films. The increse of iodine concentration would reduce band gaps and induce the change of semiconductor behavior from n-type to p-type. In this strategy, the absorbance component consists of three BiOI perovskite layer with different concentration of iodine that form n- and p- type homojuctions. BiOI with half concentration of iodine (BIOI 0.5) is first perovskite layer, then the second perovskite layer is BiOI with same concentration of iodine (BIOI 1.0) and the third perovskite layer is BiOI with double concentration of iodine (BIOI 2.0). This configuration produces cells with desirable perforamance that effectively absorb the photons in almost all parts of the solar spectrum. Both open circuit voltage (V_{oc}) (940 mV) and fill factors (~58%) for the best cells have shown drastic improvement over single active layer device and the short current densities (J_{sc}) measured are in the range (20-30) $mAcm^{-2}$. The effects of quasi-electric fields, caused by the band-gap variation of the active semiconductor, upon the illumination current density and open-circuit voltage of a solar cell will be discussed.

Keywords: *Graded Bandgap, Perovskite Solar Cells, Bismuth Oxyiodide*

INTRODUCTION

Perovskite solar cell (PSC) shows great potential for photovoltaic applications due to its excellent solar cell performance and atmospheric stability. PSCs are traditionally fabricated in single bandgap device structure where an active layer (Pb-halide perovskite) is deposited in between of hole-transport layer (HTL) and electron-transport layer (ETL). PCE conventional device structure has surpasses 20% [1-2]. However, Pb-halide PSCs raising a serious environmental concern for large-scale development and has brought controversial within the scientific community, since Pb is listed among the chemical of major public health concern by the World Health Organization and its use is restricted under several legislations worldwide [3].

Multiple studies have documented on finding lead-free materials to replace the Pb in perovskite solar cells without negatively affecting their performance. Nevertheless, the lead-free PSCs demonstrates low PCE with less than 5% because of small output current and instability. Tin (Sn) halide perovskites can be considered as an alternative material to Pb-halide perovskites, but their low current and unsteadiness impede the substitution of Sn-halide for Pb-halide perovskite [4]. Study on Indium (In) also has been made, but the commercial prospective has been severely restricted since In-halide perovskites is volatile against reduction–oxidation circumstance [4]. Research on graded bandgap perovskite has been developed by changing halide anion concentration and bandgap tuning of the perovskite absorber layer that deposited in between gallium nitride (GaN) as electron transport layer and graphene aerogel as a hole transport layer [5]. This device structure produces stable and reproducible solar cells but the PCE is still low. Another work on multi-junction tandem cell also has been done using tunable bandgap of methylammonium-lead-halide integrated with crystalline silicon and copper indium gallium selenide (CIGS), but it necessitates interconnection between the perovskite sub-cells and complicated electrical coupling, which causes electron-hole recombination centers [6]. Here, we proposed, highly-efficiency graded bandgap lead-free perovskite solar cells by fabricating triple layer lead-free perovskite devices.

Here, a bismuth oxyiodide (BiOI) PSCs with a graded bandgap was explored and growth using successive ionic layer adsorption and reaction (SILAR) with dip coating technique to enhance the device performance. BiOI was acted as photo-active layers which performed to absorb light in the solar cell device of this invention. The active layers consist of three layers of BiOI with different concentration ratios of iodine to bismuth. The concentration ratio of iodin to bismuth of the first photo-active layer increased in a substantially manner (twice) from the first photo-active layer to the second photo-active layer and a concentration ratio of iodin to bismuth of the second photo-active layer increased in a substantially manner (twice) from the second photo-active layer to the third photo-active layer. The aim is to gradually convert the material layer from n-type, i-type and p-type forming an overall graded bandgap n-i-p- type solar cell structure. Each of the deposited layer should be closely monitored in order to perform a well distributed and uniformly textured layers between the wet film and the substrate. Under optimum conditions, the proposed device structure could produce large currents and maximizes the absorption of solar radiation as well as minimize thermalization effect in the solar cell device.

The elecron transport layer (ETL) and hole transport layer (HTL) commonly included in PSCs fabrication to improve the charge collection in the device. Any researcher reported that the ETL and HTL layers could enhance the movement of the charge to the external circuit and reduce the probablity of charge recombination. The 3D device for this graded bandgap PSCs is shown in Figure 1.

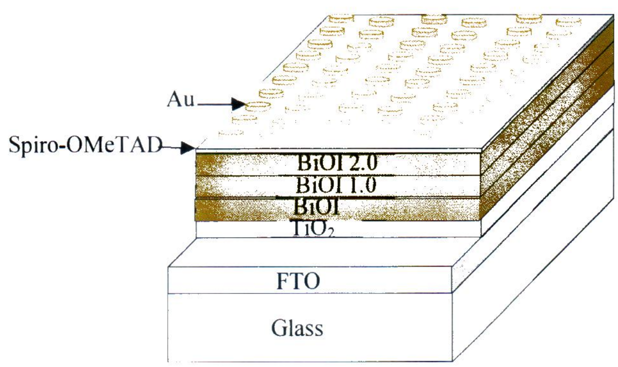

Figure 1: Schemetic diagram of a 3-D view of graded bandgap BiOI perovskite solar cells in this invention.

METHODOLOGY

BiOI thin film was deposited using SILAR technique. In order to start the synthesize solution, two beakers, with each contained 50 ml of deionized water were prepared. The first beaker is added with 0.5 M of bismuth (III) nitrate ($Bi(NO_3)$) while another beaker is added 0.5 M potassium iodide (KI). Both solutions were stirred for 30 minutes. The third beaker contained deionized water was prepared as the rinse bath. Glass/indium doped tin oxide (ITO)TO substrate was used as substrate in this work. It was clean thoroughly before the coating process. The coating process started with initially dipped the glass/ITO into the first beaker for 10 s and straight dipped to the second beaker for chemical reaction for 20 s. Finally, the glass/ITO was dipped in the third beaker and rinsed to complete one cycle dipping process. During the dipping process, the substrate was positioned inclined in the beaker. The dipping cycle process was repeated for 10 times to obtained a decent layer that comprises a first BiOI layer with half concentration ratio of iodine to bismuth (I : Bi = 0.5 : 1.0). The procedure was repeated for the second active layer comprises of BiOI layer with equal concentration ratio of iodine to bismuth (I : Bi = 1.0 : 1.0) as well as for the third active layer with double concentration ratio of iodine to bismuth (I : Bi = 2.0 : 1.0). After deposited these three layers, it will be annealed at 350°C for 20 minutes to achieve the optimum electronic characteristics of the materials.

The ETL layer, TiO_2 was grown using spin coating tecnique from synthesized solgel of titanium (iV) isopropoxide (TTIP), ethanol and acetic acid in deionized water [6]. The coated TiO_2 is about 100 nm. The thickness of ETL is controled by the speed of the rotation per minute (rpm) during the spin coating, the quantity of the material drop, the concentration and viscocity of the prepared sol gel. In this invention we used four drops of the synthesize sol-gel, spin coated at 4000 rpm for 30 s. The coated layer was pre heat in air at 150°C prioi to the deposition of multi-photoactive layer (BiOI) above it.

HTL in this invention is spiro-OMEtad layer deposited from the synthesized solution of spiro-OMEtad in chlorobenzene, 4-tert-butylprydine and lithium bis(trifluoromethanesolfonyl) imide using spin coating technique. The coated TiO_2 is about 100 nm, controlling the thickness by spin coated at 3500 rpm for 30 s.The coated spiro-OMEtad was leave in air for 24 hours before proceed the next layer (gold (Au) electrode) in the solar cell device fabrication.

The fabricated graded bandgap device was measured using I-V measurement under a solar spectrum simulator with AM 1.5 illuminations. The performance of this device has been compared with conventional PSCs device with single layer.

RESULTS AND DISCUSSIONS

J-V characteristic for measurement under light of graded bandgap BiOI PSCs is shown in Figure 2. High effiiency graded bandgap perovskite solar cells with very large current density, J_{sc} (28.6 mA cm^{-2}) and open circuit voltage, V_{oc} (0.64 V) are observed. BiOI is an intrinsic semiconductor which has equal number of holes and electrons. According to this analyses, it is revealed that the presence of iodine would reduce the band gaps, make the band potentials shift upwards and and induce the change of this semiconductor (BiOI) behavior from *-n* type to *-p* type. The decreasing mount of ioidine concentration in BiOI could slightly increased the bandgap of BiOI, reduces hole carriers and makes electron the majority carriers, thus make it more to n type conductivity[7]. The device with grading design started with n-type and slightly larger bandgap than the subsequent layers bennefitted to convert infrared (IR) photons into useful charge carriers using the effective slopes available through the device structure. This design make use of the naturally occuring defect levels to create more charge carrier using the impurity PV effect using infrared (IR) photon to generate charge carriers with same level of defect level.

Vice versa, the increase of iodine concentration would reduce band gaps, makes hole the majority carriers and make it more to p-type semiconductor. The active layer with three kind of BiOI layers formed with different concentration of iodine that form n-, i and p- type homojuctions, producing graded bandgap structure in PSCs as shown in Figure 3. The device architecture with wide bandgap in the front and three perovskite layers that formed p-i-n- graded bandgap structure has created a strong built-in electric field that enhance the seperation of the charge carriers and speeds up the photo-generated electron to the bottom electrode (ITO) and transfers holes to the top electrode (Au) with little or no recombination, result in better J_{sc} and higher V_{oc}, consequently higher efficiency through multi active layer in graded bandgap PSCs structure

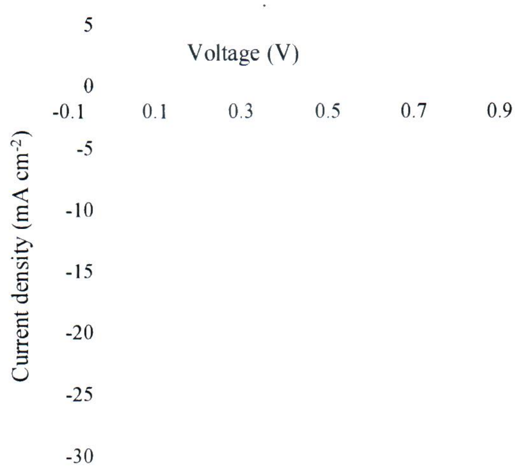

Figure 2: J-V measurement for PSC with graded bandgap BiOI active layer.

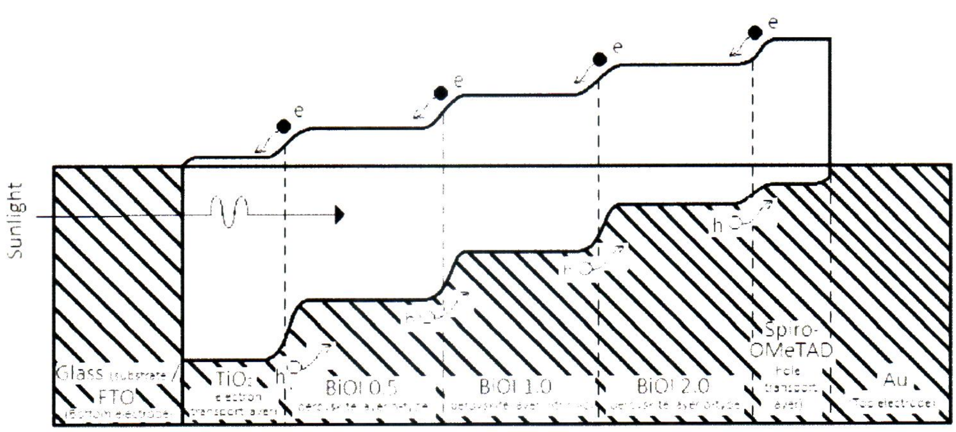

Figure 3: The sketch of the energy band diagram for proposed multi active layer graded bandgap lead-free perovskite solar cells.

We also have measured the efficiency for monolayer BiOI but the device show low J_{sc} (14.1 mA cm^{-2}) and V_{oc} (0.39 V) with only 1.89% efficiency as shown in Figure 4. This proven that the device without graded bandgap structure (single active layer) exhibit low performance and a rapid photocurrent decrease. The result also justifies the device with graded bandgap structure exhibit high performance (triple the monolayer) and a stable photocurrent.

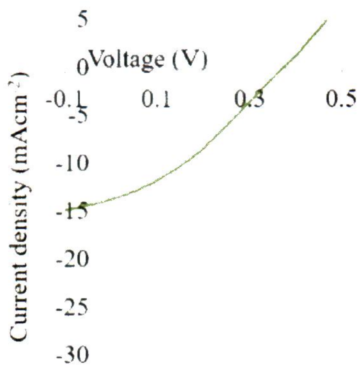

Figure 4: J-V measurement for PSC with single BiOI active layer.

CONCLUSION AND RECOMMENDATIONS

This work study the multi active layer graded bandgap by tuning the bandgap and conductivity type of BiOI films, which acted as photovoltaic (PV) active layer in PSCs. Theoretically, this configuration could improve better performance from the previous (single active layer) design since with this wide bandgap layer at the front, the electromagnetic (EM) spectrum of light (photons) would be effectively absorbed and improve the seperation of the charge carriers to the external circuit. From this work, it is evident that the device with graded bandgap structure exhibit high performance (J_{sc}, V_{oc} and efficiency) compared to the device with single layer. Therefore, the findings suggested that the PSCs device can be improved through device architecture of active layer, where this particular design has maximized the absorption of solar radiation as well as minimized thermalization effect in the solar cell device.

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