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Photon-Assisted Electrodeposition of <0001>-n-ZnO/<111>-p-Cu_2O Photovoltaic Devices with TiO_2 Intermediate Layer

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Abstract. The <111>-Cu_2O/<0001>-ZnO photovoltaic (PV) device has been constructed by a electrodeposition f Cu_2O layer followed by a photon-assisted electrochemical reaction in aqueous solutions, and the effect of the insertion of the TiO_2 layer prepared by a sol-gel technique on the photovoltaic performance was investigated. The structural, optical, and electrical characterizations were carried out with XRD, FE-SEM, UV-Vis-NIR spectrophotometer, and solar simulator. The performance of AZO/<0001>-ZnO/TiO_2/<111>-Cu_2O PV-devices changed depending on the preparation condition for the TiO_2 layer, and the short-circuit current density of 4.86 $mAcm^{-2}$ has been obtained for the PV device prepared under optimized condition.

Introduction

A photovoltaic device composed of p-Cu_2O and n-ZnO semiconductors has received broad attention as a candidate of the next generation thin film solar cell, because of the nontoxicity, abundance, theoretical conversion efficiency of around 18% [1], and an absorption coefficient higher than a single crystalline Si [2]. The Cu_2O layers for the photovoltaic application have been prepared by several techniques such as a thermal oxidation of a metallic Cu sheet [3], RF magnetron sputtering [4], and electrodeposition [5]. The ZnO layers have been prepared by electrodeposition in an aqueous solution containing either zinc nitrate [6] or zinc chloride [7] as well as gas-phase deposition techniques such as sputtering, molecular beam epitaxy, and laser ablation techniques. The electrodeposition process in aqueous solutions is a well-known technique due to several advantages such as low-fabrication cost, low temperature, ambient pressure processing, controllable film thickness, and possible large scale deposition, as demonstrated for the $CuIn_xGa_{1-x}Se_2$ solar cell production. The conversion efficiency, however, was limited at 1.28% for the randomly oriented Cu_2O/ZnO heterojunction diode prepared only by electrodeposition [8]. The quality of the Cu_2O layer was improved by using an electrochemical heteroepitaxial growth [9], and a photon-assisted electrochemical growth of the ZnO layer without the reduction of Cu_2O to metallic Cu was developed [10]. Here, we prepare <0001>ZnO/<111>Cu_2O PV device by the heteroepitaxial and photon-assisted electrochemical process, and the effects of the insertion of a TiO_2 layer prepared by a sol-gel process were investigated by the structural, optical, and electrical characterizations.

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Experimental Procedures

The Cu_2O layer was deposited potentiostatically at −0.5 V referenced to Ag/AgCl electrode on the Au(111)/Si(100) substrate at an electric charge of 1.7 coulomb cm^{-2} with a potentiostat in an alkaline aqueous solution containing a 0.4 M copper(II) acetate monohydrate, and 3 M lactic acid at 328 K. The solution was prepared with reagent grade chemicals and de-ionized (DI) water, and KOH was added for the pH adjustment to 12.5. A Pt plate was used as the counter electrode. The TiO_2 layer was deposited on the Cu_2O layer by using spin coating technique at 5000 rpm for 15 sec with 3% and 5% of TiO_2 sol at substrate temperature of 65˚C. The ZnO layer was stacked on the TiO_2 layer at a potential of −0.06 V referenced to the Ag/AgCl for the deposition time of 30 min in a simple aqueous solution containing 0.08 M zinc nitrate hydrate at 336 K with light irradiation. The 200-nm-thick ZnO:Al layer was deposited using radio frequency (rf) magnetron sputtering with AZO target. The substrate temperature was 25˚C, pressure chamber of 1.0 Pa, and the rf power of 100 W. Structural, optical, and electrical characterizations were performed by XRD, FE-SEM, UV-Vis-NIR spectrophotometer, and Keithley 2400 source meter under illumination with 100 $mWcm^{-2}$ power.

Results and Discussion

Figure 1 shows X-ray diffraction patterns of ZnO/Cu_2O heterojunction structure before and after inserting the TiO_2 sol concentration of 3% and 5%. Three diffraction peaks assigned as the ZnO(0002) [11], Cu_2O(111) [12], and Au(111) planes could be observed on the XRD pattern before and after inserting the TiO_2. No obvious peaks identified as the TiO_2 could be observed on the XRD patterns. The ZnO peak intensity decreased as the TiO_2 sol concentration increased.

Figure 2 shows FE-SEM images of the ZnO layers before and after stacking the TiO_2 layer with sol concentrations of 3% and 5%. The Cu_2O layers deposited on the Au(111) layer were composed of aggregates of hexagonal columnar grains grown in direction perpendicular to the Au(111) substrate surface, and the thickness was estimated to be about 2.4 μm. The hexagonal columnar ZnO grains with the width of approximately 150 nm grew in the direction normal to the surface and hexagonal facets corresponding to the (0001) plane was observed on the surface image without the TiO_2 layer. The TiO_2 layer thickness was estimated to be about 100 nm at 5% TiO_2 sol and decreased at 3% TiO_2 sol. For the TiO_2 layer prepared at 5%, ZnO grains with the width of approximately 90 nm were grown in the direction normal to the surface, and some pores could be observed between the ZnO grains. The width changed to 110 nm at 3% as shown in Fig. 2 (b). The thickness of the ZnO layer was estimated to be around 200 nm.

Figure 3 shows the absorption spectra for ZnO/Cu_2O PV devices after depositing TiO_2 layer with 3% and 5% TiO_2 sol. The Cu_2O film showed an absorption edge at 600 nm, and the interference fringe pattern was observed at a wavelength less than 600 nm, indicating the formation of Cu_2O layer with the characteristic bandgap energy of 2.1 eV.

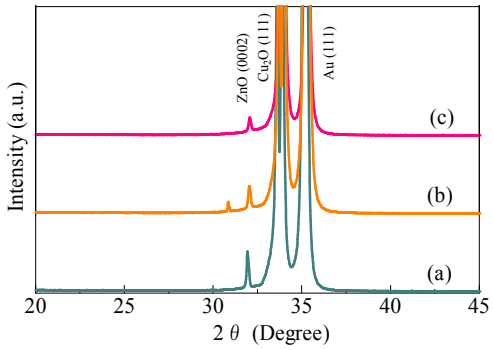

Fig. 1 XRD patterns for ZnO/Cu_2O PV devices on Au substrate before (a) and after inserting TiO_2 layers at 3% (b) and 5% sol (c).

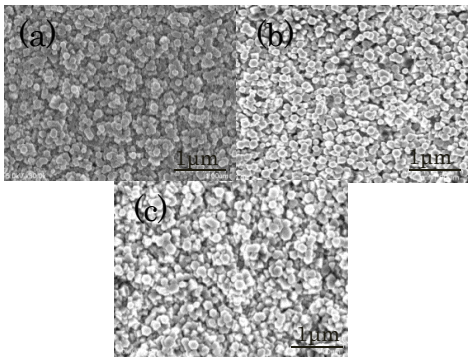

Fig. 2 FESEM images of ZnO prepared without (a) and with TiO_2 layers with 3% (b) and 5 % sol (c).

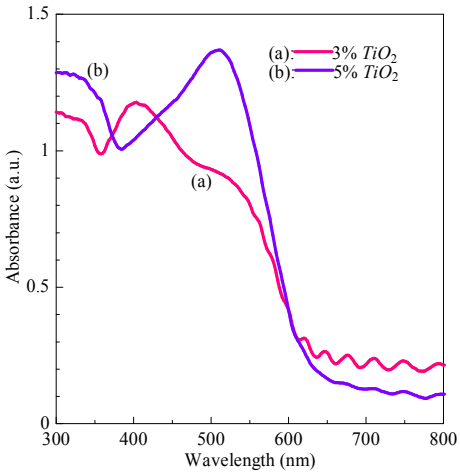

Fig. 3 Absorption spectra for ZnO/Cu_2O PV devices with inserting the TiO_2 layer of 3% (a) and 5% sol (b).

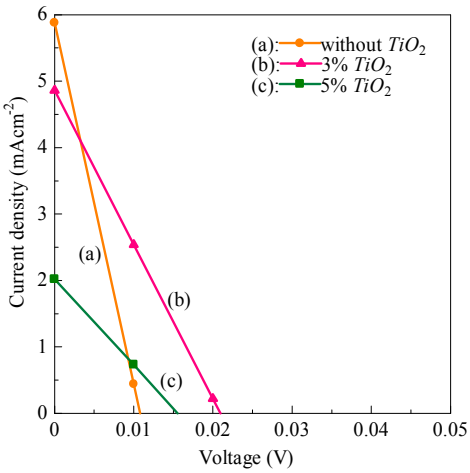

Fig. 4 J-V curve of the $AZO/ZnO/Cu_2O$ PV cells without (a) and with inserting the TiO_2 layer of 3% (b) and 5% sol (c).

An absorption edge was also observed at a wavelength around 380 nm originated from the ZnO layer in addition to that for the Cu_2O layer. An absorption edge assigned as the TiO_2 layer also observed at a wavelength around 350 nm corresponding to the characteristic bandgap energy of 3.5 eV.

To demonstrate the effect of the TiO_2 buffer layer on ZnO/Cu_2O PV cell performance, we fabricate ZnO/Cu_2O PV cells by inserting 3% and 5% TiO_2 layer. Figure 4 shows the current density-voltage curves for the $AZO/ZnO/Cu_2O$ PV cells with and without TiO_2 layers under AM 1.5G illumination (100 $mWcm^{-2}$). The PV device without TiO_2 exhibited a conversion efficiency of 0.004 % with 0.01 V open current voltage (Voc) and 5.87 $mAcm^{-2}$ short-circuit current (Jsc). The performance of $AZO/ZnO/TiO_2/Cu_2O$ PV devices strongly depended on TiO_2 sol concentration closely relating to the TiO_2 layer thickness and the resultant ZnO layer. The $AZO/ZnO/TiO_2/Cu_2O$ PV device prepared at 3% TiO_2 sol showed an improved conversion efficiency of 0.03 %, although the Jsc decreased slightly. The low Voc suggested the electrical shorting thorough the pores between the deposited ZnO grains. Further improvement of the quality including the homogeneity and energy state is indispensable to raise the photovoltaic performance.

Conclusion

The effects of the inserted TiO_2 layer at the electrodeposited ZnO/Cu_2O PV devices on the photovoltaic performance were investigated by structural, optical, and electrical characterizations. The insertion of the TiO_2 layer by a sol-gel process resulted in the improvement of the photovoltaic performance to 0.03% with Voc of 0.02 V, Jsc of 4.86 $mAcm^{-2}$, and FF of 0.25.

Acknowledgments

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