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Non-Vacuum Based Preparation of Heterojunction Thin Film Layers for Photovoltaic Application

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Abstract

Earth abundant and non-toxic materials were chosen for the fabrication of thin film solar cell using simple and widely applicable deposition techniques. Cu_2ZnSnS_4 (CZTS) thin films was used as a p-type absorber layer in the device and TiO_2 was used as the n-type layer. A p-n junction was formed with TiO_2 and CZTS layers prepared by sol-gel spin coating and spray pyrolysis technique respectively. The device was constructed on a FTO substrate and Al was used as the back electrode with the device structure Glass/FTO/TiO_2/CZTS/Al. The structural, optical and optoelectronic response of the device was studied using Glancing Incidence angle X-ray Diffraction (GIXRD), UV-Visible spectrum and I-V measurements. The GIXRD analysis revealed the polycrystalline nature of the material and the structural formation of each layer. The fabricated device was analyzed to study with photocurrent response. The device showed an open circuit voltage (Voc) of 647 mV and a short circuit current density (Jsc) of 4.5 nA/cm^2 upon illumination. The UV-Visible measurement after depositing each layer shows the variation in transmittance after each layer deposition.

Keywords: Thin films, Cu_2ZnSnS_4, p-n junction, Photocurrent response.

1. Introduction

Cu_2ZnSnS_4 based thin film photovoltaic devices are widely studied in the recent years because of its abundant elemental availability, non-toxic nature and less processing cost [1]. The material possesses an optimum bandgap of 1.45 eV with a high absorption coefficient of 10^4 cm^{-1} which is suitable for p-type absorber layer in thin film solar cells [2]. In the same way TiO_2 is an optimum material to act as an n-type layer which has a wide bandgap of 3.3 eV [3].

The studies of the p-n junction formation with these two materials are of wide interest in photovoltaic research due to its simple and easy preparation using chemical deposition techniques such as spray pyrolysis [4, 5], sol-gel [6], chemical bath deposition [7] and dip coating technique [8]. The photovoltaic device fabricated with this material has a high throughput and is an upcoming development in this field of research [9-12].

In this present work, the fabrication of cost efficient and eco-friendly thin film photovoltaic device is fabricated using versatile chemical deposition techniques with the superstrate device structure FTO/TiO_2/CZTS/Al.

2. Experimental Procedure

The device was fabricated in a layer by layer manner with a superstrate structure. The first Glass/FTO was purchased from Sigma Aldrich with 13Ω/cm^2 resistance. The n-type TiO_2 layer was deposited on top of FTO substrates using sol-gel spin coating technique. The sol-gel solution was prepared by taking Titanium tetra isopropoxide as precursor material for titanium in 10 ml of iso-propyl alcohol solvent. The gelation time took about 2 h. The sol-gel

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solution was deposited at 4000 rpm for 30 s using spin coating technique and the coating was repeated again to improve the film thickness. The next p-type CZTS layer was deposited over the n-layer using chemical spray pyrolysis technique. The precursor solution was prepared with copper (II) chloride dihydrate ($CuCl_2.2H_2O$), Zinc (II) chloride dihydrate ($ZnCl_2.2H_2O$), Tin (II) chloride dihydrate ($SnCl_2.2H_2O$) and thiourea (CH_4N_2S) in 2-methoxyethanl solvent. The solution was sprayed at a substrate temperature of 350˚C with a constant pressure of 1.2 kg/cm^2 and a continuous solution flow rate of 4 ml/min was maintained. The back electrode Aluminum was deposited using thermal evaporation.

The fabricated device with the structure FTO/TiO_2/CZTS/Al was characterized by Glancing Incidence X-ray diffraction (GIXRD, XPERT-PRO diffractometer) equipped with Cu-K_α (λ = 1.54060 Å, 45 kV, 40 mA with a step size of 0.02 degree and scan range from 10˚ to 90˚). The optical properties were studied using UV-Visible spectroscopy (Perkin Elmer Lambda 576) for a wavelength region of 300-1100 nm. The photocurrent measurement was carried out for the device using Keithley (source meter 2450) with a Xenon lamp illumination of 100 mW/cm^2 with a voltage range from -1 V to 1 V.

3. Results and Discussion

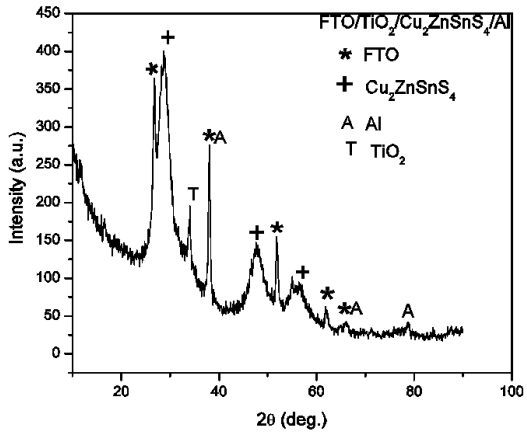

Figure 1. GIXRD pattern of the multilayered device

The GIXRD pattern of Figure 1 shows the structural formation of the multi-layered device. The major peaks at 2θ = 28.5˚ (112), 47.5˚ (220) and 56.3˚ (312) corresponds to the Cu_2ZnSnS_4 product formation with the kesterite phase and tetragonal crystal structure (JCPDS no. 26-0575) [13]. The 2θ peaks at 25.3˚, 25.7˚ and 36.2˚ corresponding to the planes (210), (111) and (102) respectively shows the Brookite phase formation of the TiO_2 layer with orthorhombic crystal structure (JCPDS no.65-2448) [14]. The Al and FTO peaks are also present in the figure at 2θ = 38.2˚, 52.6˚, 62.8˚, 65.4˚, 78.4˚ [15].

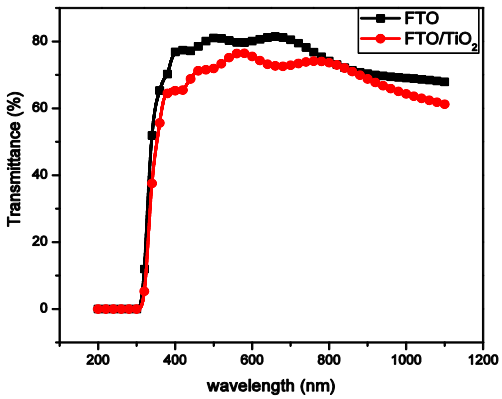

Figure 2. Transmittance spectrum of FTO and FTO/TiO_2 layers

Figure 2 shows the transmittance spectrum of FTO substrate and TiO_2 deposited FTO substrate. The decrease in transmittance value from 80% to 75% for FTO to FTO/TiO_2 is clearly observed from the graph. In the device the transmittance varies from the transparent conducting layer (FTO) to n-type window layer (TiO_2) in this manner. The transmittance has to be more for the TCO and window layer in order for the photons to reach the absorber layer.

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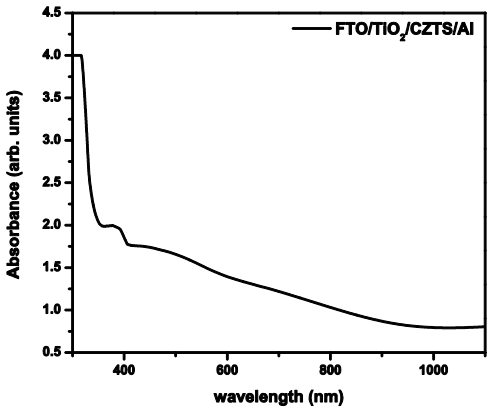

Figure 3. Absorbance spectrum of the device

The absorbance spectrum of the device is shown in Figure 3. From the figure the multiple cut-off wavelengths of different layers can be observed. The cut-off wavelength around 360 nm, 409 nm and 860 nm corresponds to FTO, TiO_2 and CZTS layers respectively. The band gap values are calculated to be 3.44 eV, 3.03 eV and 1.44 eV for FTO, TiO2 and CZTS layers respectively. The TCO and n-type layer has a wide bandgap while the absorber layer has a narrow bandgap. The bandgap values obtained are optimum values of the corresponding material.

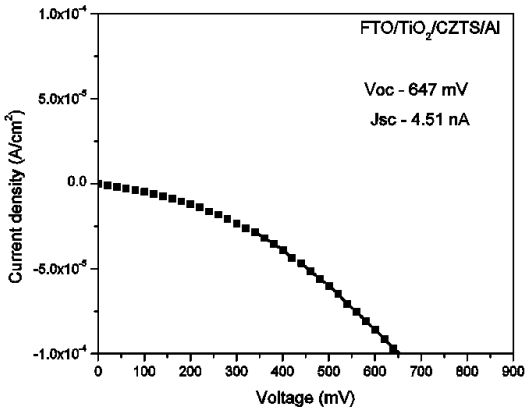

Figure 4. Photocurrent (I-V) response of the device

The fabricated device photocurrent (I-V) response is shown in Figure 4. It shows a very less development of current. The short circuit current density (Jsc) is observed as 4.51 nA and the open circuit voltage (Voc) is measured to be 647 mV. The light current response alone is shown in the figure. The dark current response is not shown here which is of negligible order.

4. Conclusions

Superstrate structured heterojunction device was fabricated using non-vacuum based techniques such as spray pyrolysis and so-gel spin coating techniques. The deposited layers of TiO_2 and CZTS materials are earth abundant and environment friendly. The structural formation of the device was confirmed from the GIXRD pattern. The optical response of each layer and the optical and opto-electric response of the device were studied. It showed a photovoltaic response in the fabricated device.

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