

FABRICATION AND CHARACTERIZATION OF ZnTiO₃ BASED PEROVSKITE SOLAR CELL ON ITO SUBSTRATE

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Abstract

Firstly ZnTiO₃; Zinc titanate powder was successfully prepared by a solid state reaction method. After that ZnTiO₃ perovskite solar cell was fabricated by titanium dioxide (TiO₂) layer as the electron transporter layer (ETL) and CuO as the hole transport layer (HTL). Indium doped Tin oxide (ITO) was used as a substrate. In this research, fabrication of ZTO based perovskite solar cell was investigated. From J-V characteristics, efficiency (η) and fill factor were evaluated under illuminated condition. The conversion efficiency 2.12% of ZTO perovskite solar cell was found. By using fabrication and characterization, this structure ZTO based perovskite solar cell was useful in solar cell application.

Keywords: ZnTiO₃ perovskite, solid state reaction, efficiency (η) and fill factor

Introduction

Most of the renewable energy from wind, micro-hydro, tidal, geothermal, biomass, and solar are converted into electrical energy. Solar technologies use the sun to provide heat, light, electricity, etc for domestic and industrial applications. The best conversion efficiency of most commercially available solar cells is in the range 10-20% [Green, Martin A, 1982]. Perovskites are emerging as a new generation of photovoltaic materials. The transition metal oxide such as TiO₂, their perovskite structures ZnTiO₃, is widely used in optical and microelectronic applications because of their excellent mechanical, optical and insulating properties. Up to now, perovskite films have been formed by film deposition approaches such as spin-coating, sequential deposition of the inorganic and organic precursor, and co-evaporation of the precursors [Burschka, J et al 2013- Liu, M et al 2013]. There are three compounds that exist in the ZnO-TiO₂ system: ZnTiO₃ with a hexagonal limonite structure (h-ZnTiO₃), Zn₂TiO₄ with a cubic spinel crystal structure, and Zn₂Ti₃O₈ with a cubic defect spinel structure [Bartram, S.F et al 1961]. Zn₂Ti₃O₈ has been observed as a low-temperature form of h- ZnTiO₃ that exists at T < 820 °C [Steinike, U et al 1997], and is only produced based on the Zn₂TiO₄ phase [Kim, H.T et al 2001]. It is known that h- ZnTiO₃ decomposes into Zn₂TiO₄ and rutile TiO₂ at T > 945 °C [Dulin, F.H et al 1960], However, perovskite crystallizes very rapidly, so it is very challenging to form uniform, continuous, and leakage-free perovskite films by solution methods [Liu, M et al 2013] . In this study, fabrication of the ZnTiO₃ perovskite solar cell was carried out by spin coating method.

Experimental Procedure

Powder preparation for Zinc Titanate (ZnTiO₃)

In this research, ZnO and TiO₂ were chosen as the starting materials. Firstly ZnO and TiO₂ powder with stoichiometry (ZnO: TiO₂= 1:1 molar ratio) were mixed. Ethanol was added to

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the mixture powders. Zinc titanate (ZnTiO_3 ; ZT) powders were prepared by a solid state reaction method. And then the mixture solution stirred with the magnetic stirrer and milled with the ball milling machine for 12 hr. After that the powders were dried and sieved to form uniform particles size. And then ZT powders were heated at various calcination temperatures, ranging from 500 °C to 850 °C for 2 h at a heating /cooling rate of 5 °C/min. After the sample was annealed at 850 °C, ZnTiO_3 crystals were successfully obtained. The phase formation was identified by X-ray diffraction (XRD). Similarly, CuO and TiO_2 were checked to observe successfully forms. After that, ZnTiO_3 (850 °C), TiO_2 and CuO were chosen for further investigation.

Perovskite Solar Cell Fabrication

Perovskite solar cells were fabricated in two major architectures, p-i-n and n-i-p type. We fabricated perovskite solar cells with a n-i-p type. The solar cell device consisted of layers of conductive material, titanium dioxide (TiO_2), perovskite zinc titanate (ZnTiO_3) and Copper (II) Oxide (CuO) as in Figure 1. The first layer was an electrically conductive indium doped tin oxide (ITO). The next layer was the electron transport layer (ETL) for which we used titanium dioxide (TiO_2). The ETL was responsible for extracting the excited electrons out of the perovskite material and transporting them to the electrically conductive layer. After that this substrate was annealed at 160°C for 30 minutes. Next was the light absorbing perovskite layer where electrons and holes were generated. At that time, the annealing temperature 200°C was used to form perovskite layer deposition. After the perovskite, we deposited a hole transport layer (HTL) and for this layer, we used CuO, which collects and transports the holes to the anode. After that, this substrate is then annealed at 110°C for 30 minutes. In this method, each layer was coated by using the spin-coater. The substrate is rotated at a very high speed (between 1500-3000 rpm), while the precursor solution is dropped onto the rotating substrate in each step. Finally silver back contact was used as the electrode for the device.

Result and discussion

XRD Analysis

X-ray powder diffraction is a rapid analytical technique used for phase identification of crystalline material and can provide information on unit cell dimensions. Based on the results of XRD, calcinations were carried out from 500 °C to 850 °C for 2 h. After annealed from 500 °C to 850 °C, in the temperature at 500 °C and 600 °C the precursor of ZnO and TiO_2 were detected. A high purity of the cubic perovskite phase was discovered in powders annealed at 700 °C, 800 °C and 850 °C. The comparison of XRD pattern for ZnTiO_3 powders at 500 °C, 600 °C, 700 °C, 800 °C and 850 °C is shown in Figure 2. XRD spectrums of TiO_2 and CuO were found in Figure 3(a) and 3(b). Some crystallographic information about the powder ZnTiO_3 , TiO_2 and CuO were collected and listed in Tables 1.1~ 1.3. These tables also gave the crystallite size and average crystallite size of powders. Five most intense diffracted peaks are observed on XRD spectrum. While the upper site of XRD spectrum indicated the observed XRD spectrum and the lower site showed the JCPDS (Joint Committee on powder Diffraction Standards) Library file. All identification peaks were well matched with those of JCPDS library file.

SEM analysis

The microstructure of ZnTiO_3 powders were observed by SEM. The grain sizes were measured by using well known bar code system. Figures 4(a) ~ (c) are SEM images of ZnTiO_3 , TiO_2 and CuO . According to SEM images, the grains were uniformly distributed on the experimental area of SEM microphotograph. The shape of grain was examined to be circular shape. The average grain sizes were estimated to be 364 nm at 850 °C for ZnTiO_3 . Some porosity was found on all SEM images and they were porous structure.

UV-Vis Analysis

Figure 5 (a), 6 (a), 7 (a) show the absorption spectrums of ZnTiO_3 , TiO_2 and CuO film. The optical transmittance range and transparency cut off wavelength of the crystal are important factors for optical applications. Figures 5 (b), 6 (b) and 7 (b) show the optical properties of ZnTiO_3 , TiO_2 and CuO . The optical band gap (E_g) was evaluated from the absorption spectrum and the optical absorption coefficient (α) near the absorption edge. The optical band gap and absorption edge of ZnTiO_3 film are presented in Table 1.4.

J-V Analysis

To examine the PV behavior of ZnTiO_3 perovskite solar cell fabricated, J-V characteristics were measured under illumination. J- V curve showed the photovoltaic behavior as in Figures 8. It meant the open-circuit voltage and short-circuits current (V_{oc} - J_{sc}) characteristics. From J-V characteristics curve, the current at maximum power point (J_m) and the voltage at maximum power point (V_m) were measured. Short-circuit current, Maximum Current, Open-circuit voltage, Maximum Voltage, Fill factor and Efficiency of ZnTiO_3 Perovskite Solar Cells were measured and calculated at Table 1.5.

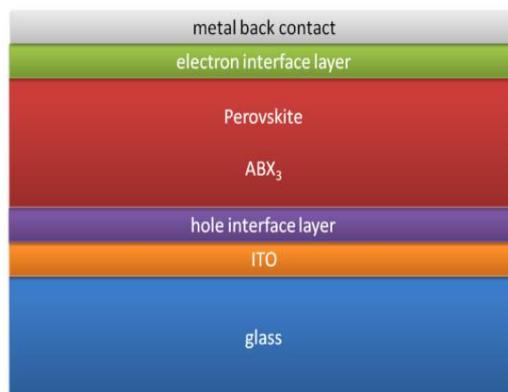


Figure 1 Generic structure of a standard (non-inverted) perovskite solar cell

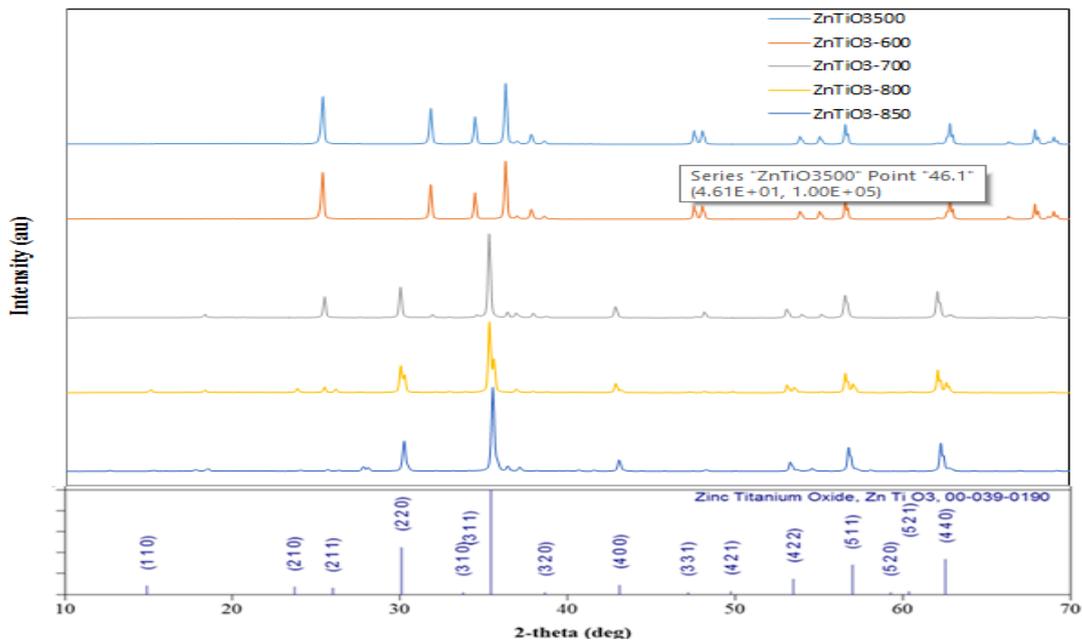


Figure 2 Comparison XRD patterns of ZnTiO₃ powders at 500 °C, 600 °C, 700 °C, 800 °C 850 °C for 2 h

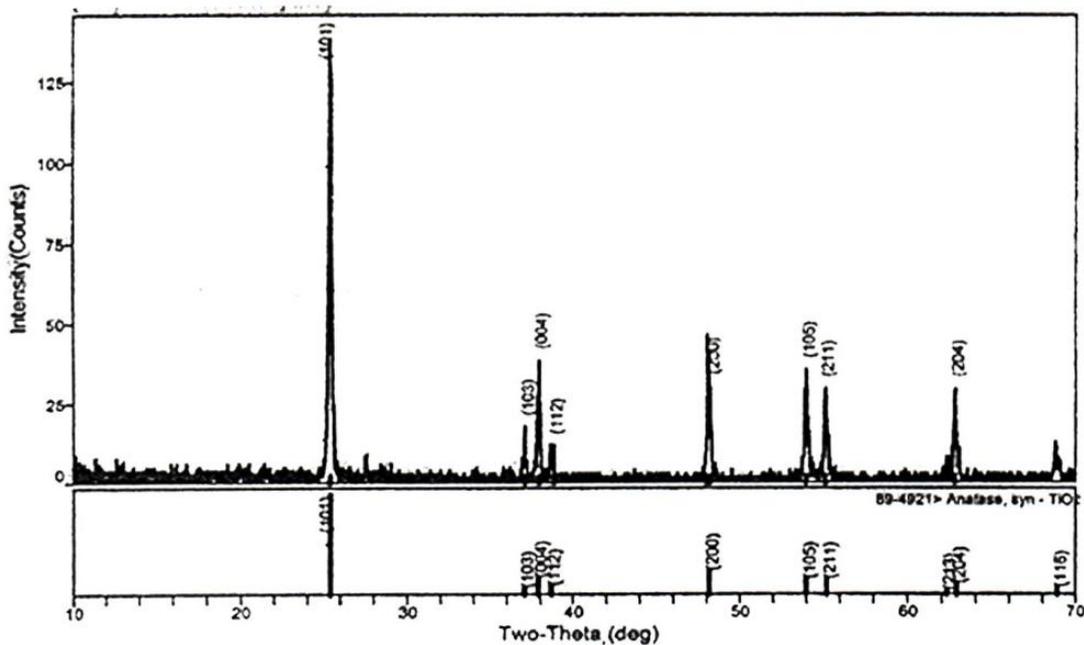


Figure 3 (a) XRD spectrum of TiO₂ powder

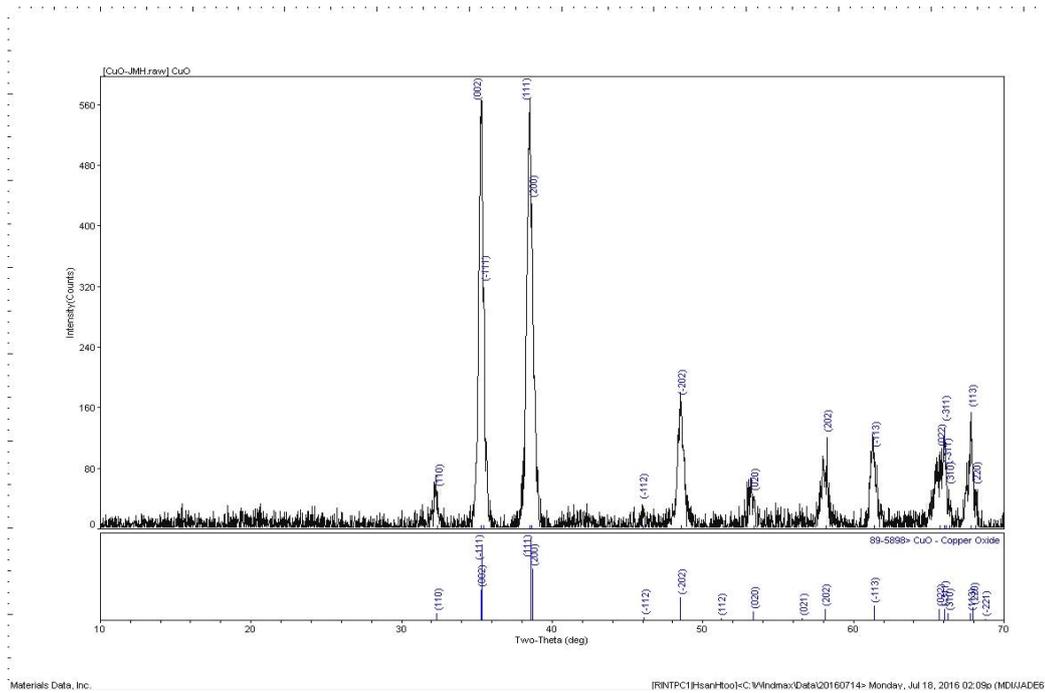


Figure 3(b) XRD spectrum of CuO powder

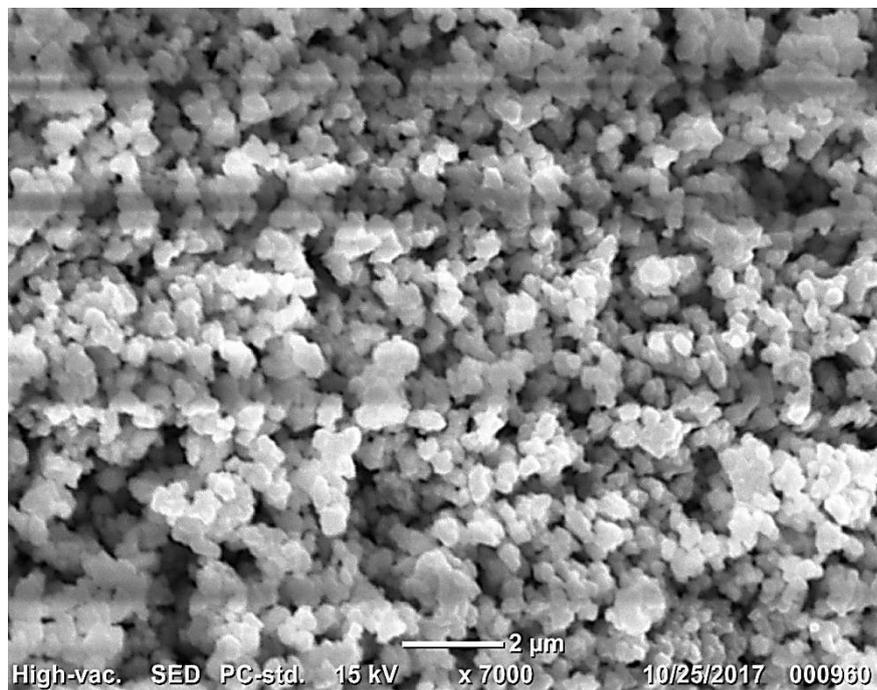


Figure 4 (a) SEM image of ZnTiO₃ powder at 850°C for 2 h

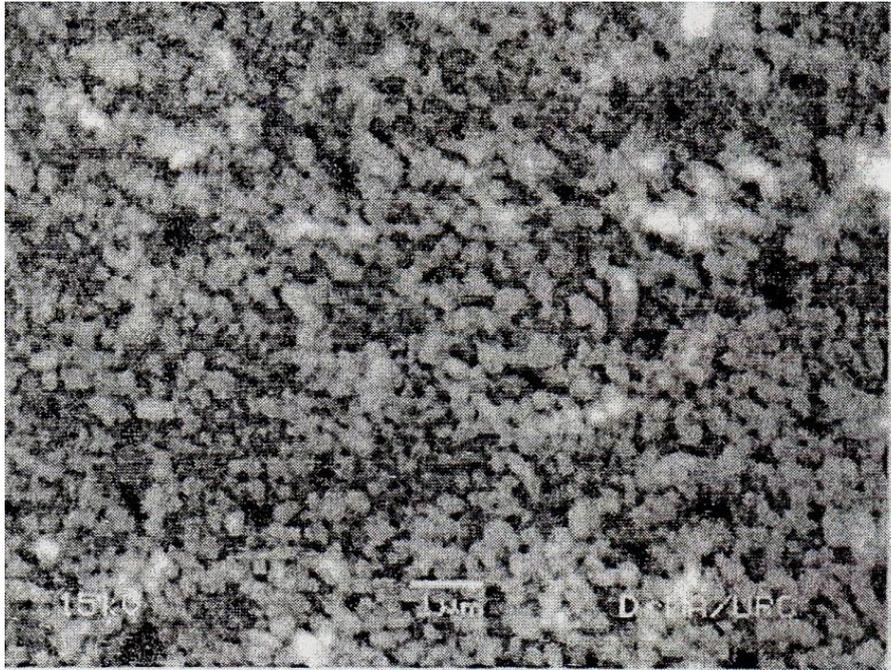


Figure 4(b) SEM image of TiO₂ powder

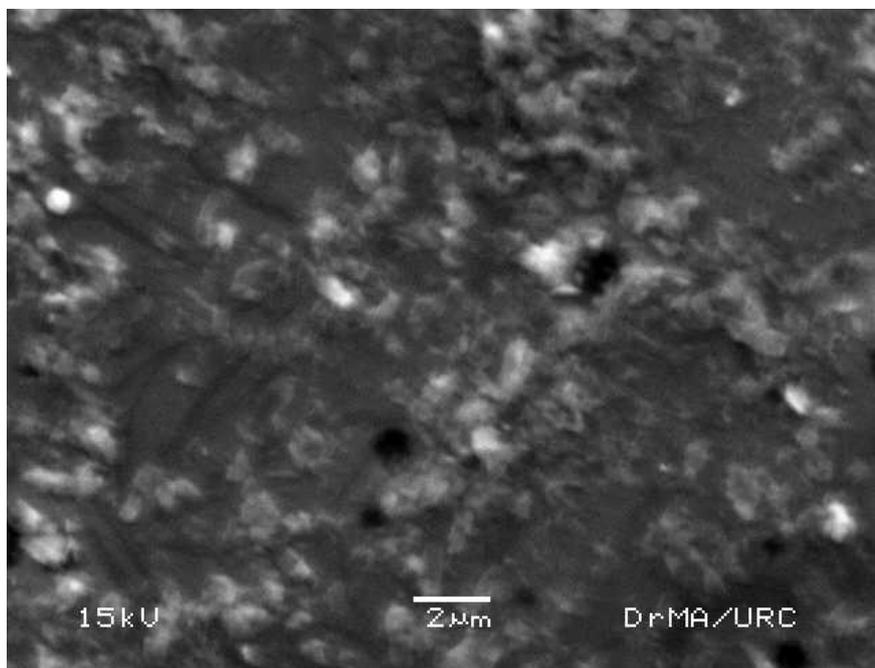


Figure 4(c) SEM image of CuO powder

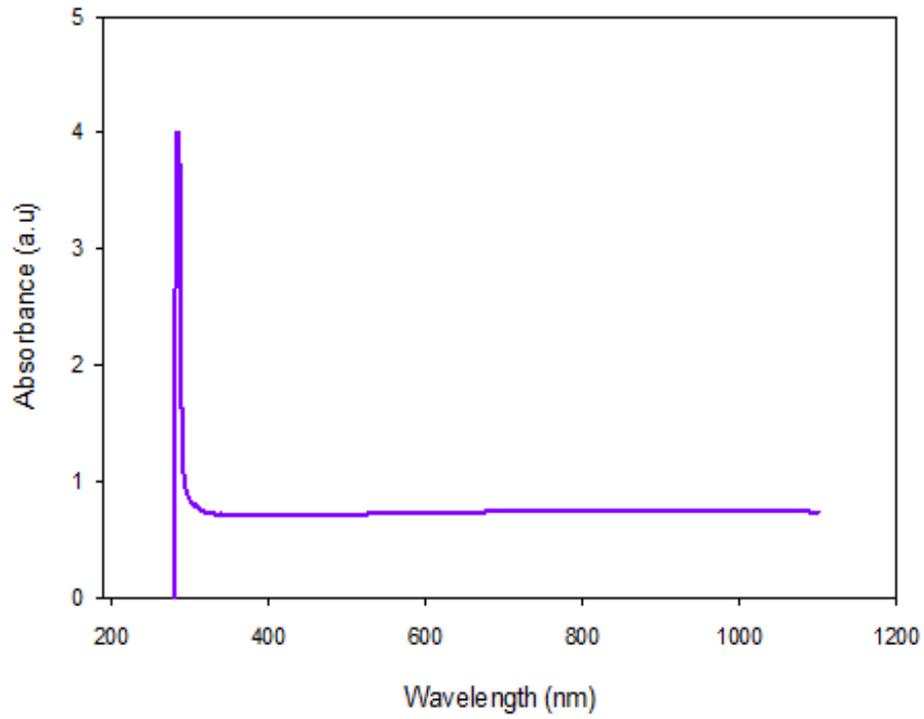


Figure 5 (a) The absorption spectrums of ZnTiO₃

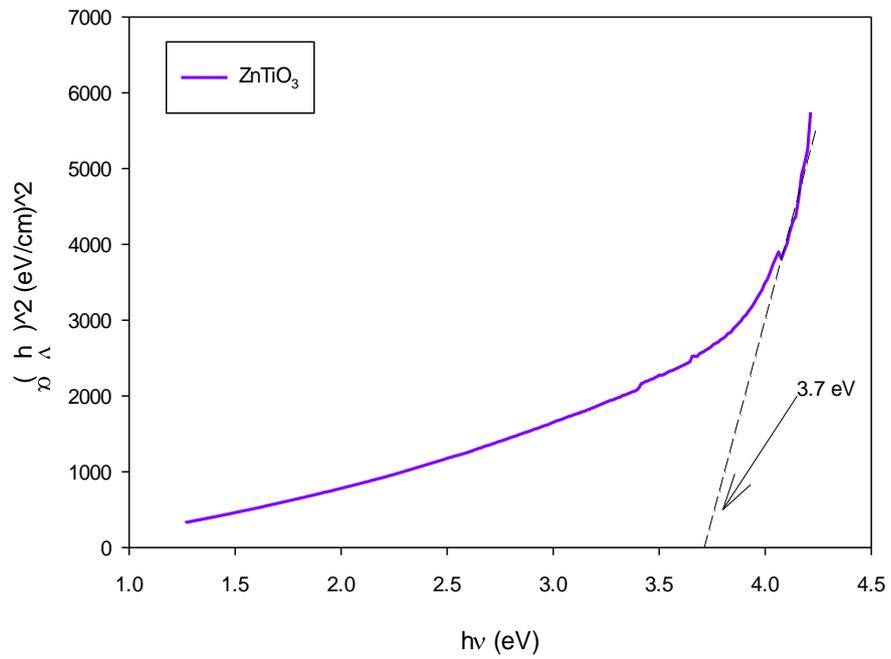


Figure 5 (b) Optical energy band gap of ZnTiO₃

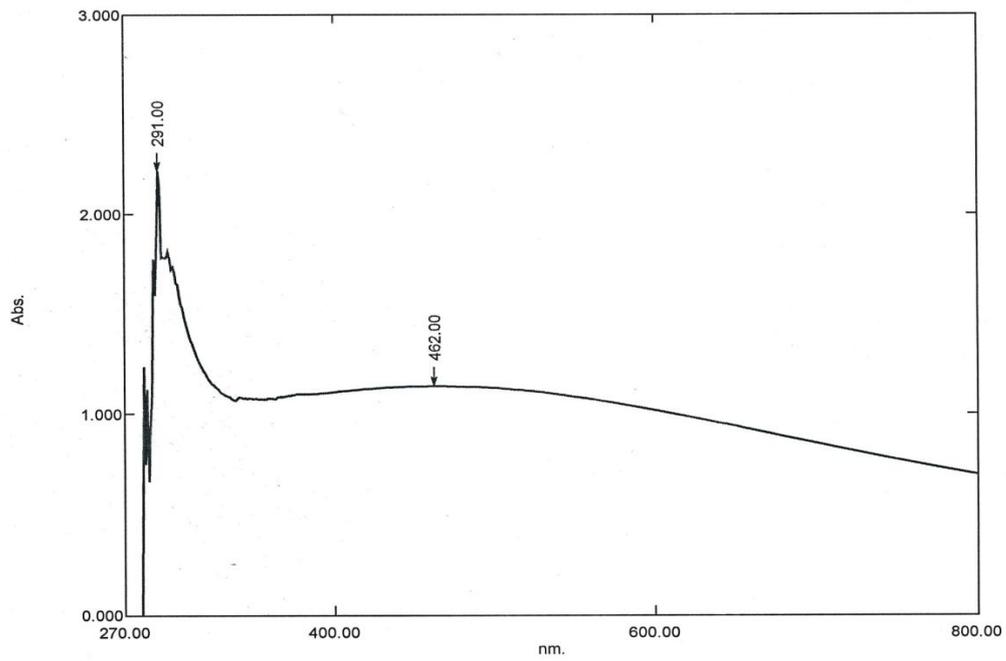


Figure 6 (a) The absorption spectrums of TiO₂

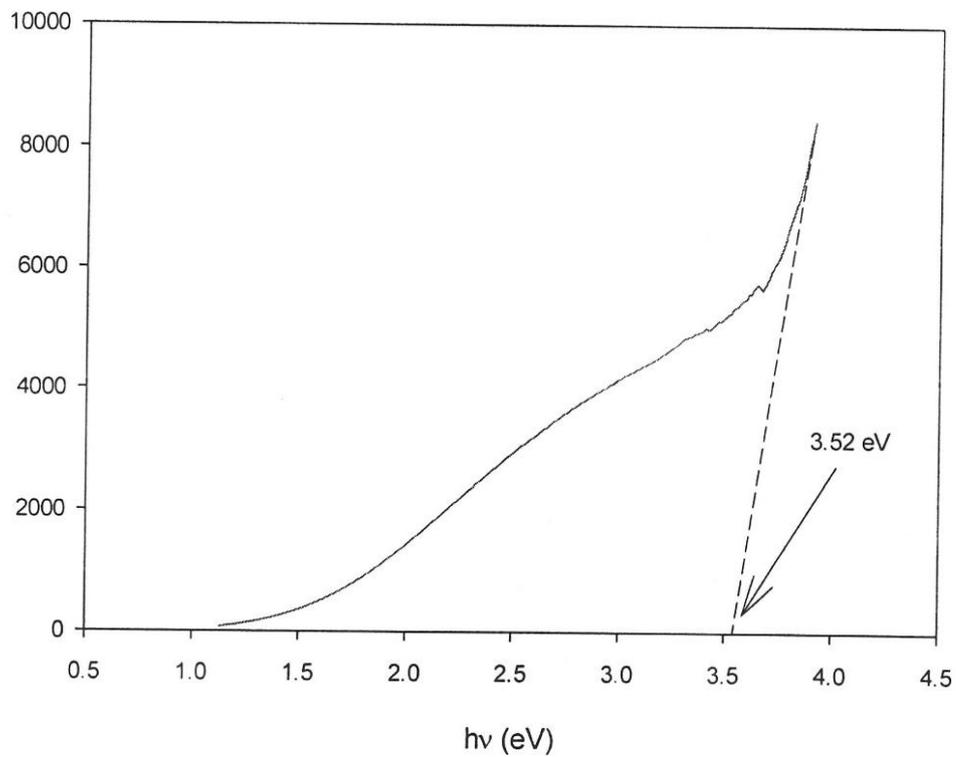


Figure 6 (b) Optical energy band gap of TiO₂

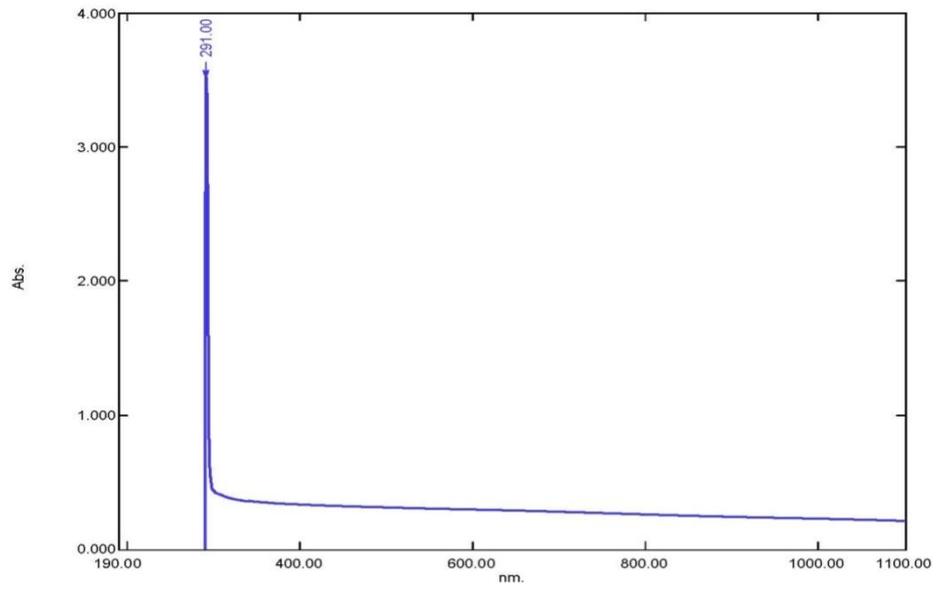


Figure 7 (a) The absorption spectrums of CuO

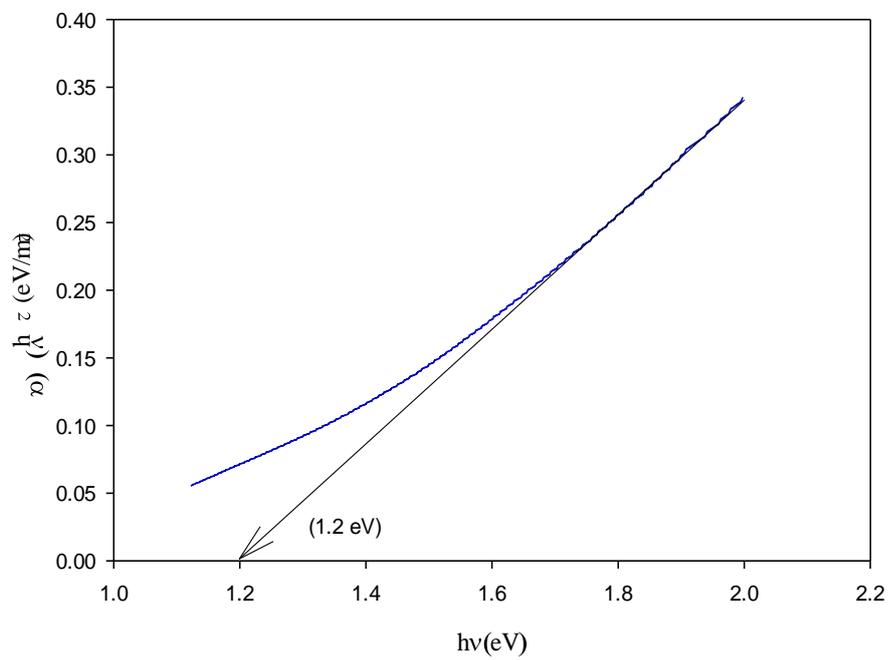


Figure 7 (b) Optical energy band gap of CuO

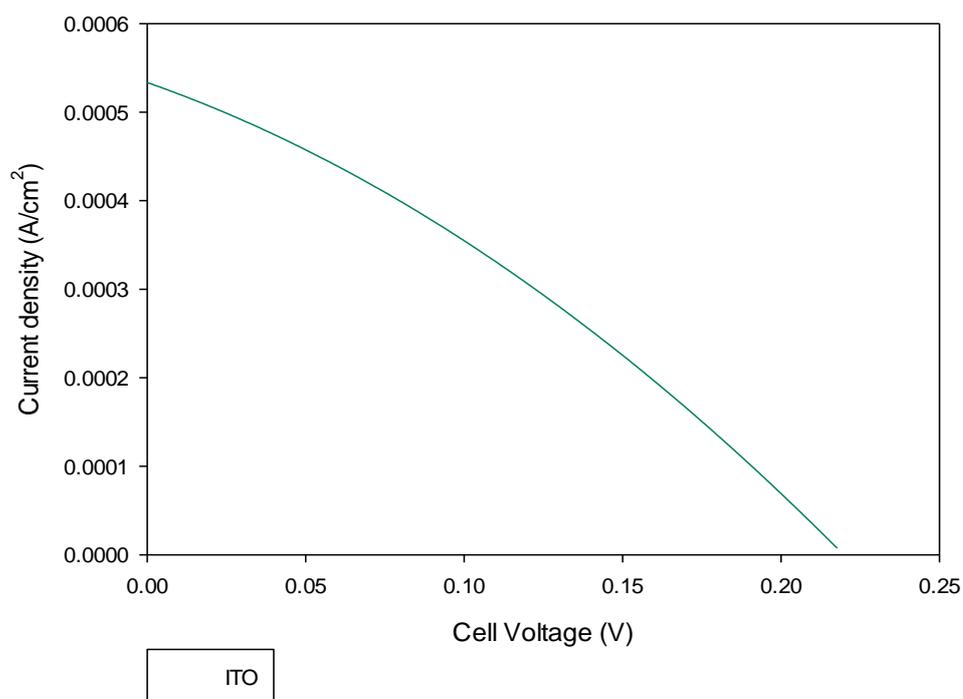


Figure 8 Current density – voltage curve for ZnTiO₃ perovskite solar cell with ITO (Indian doped tin oxide) substrate

Table 1.1 Crystallite size of ZnTiO₃ powder at 850°C for 2 h

No	Peak	2 θ	FWHM (deg)	Crystallite size (nm)
1	220	30.175	0.179	45.91
2	311	35.4721	0.166	50.18
3	422	53.699	0.18	49.41
4	511	57.214	0.193	46.83
5	440	62.795	0.288	32.23
Average crystallite size				44.91

Table 1.2 Crystallite size of TiO₂

No.	Peaks	2 θ	FWHM (deg)	Crystallite size (nm)
1	101	25.355	0.207	39.340
2	103	37.013	0.181	46.293
3	0 04	37.846	0.200	42.000
4	112	38.643	0.260	32.370
5	200	48.143	0.211	41.250
6	105	53.973	0.236	37.760
7	211	55.184	0.212	42.270
8	204	62.810	0.243	38.290
Average crystallite size				39.949

Table 1.3 Crystallite size of CuO powder

No	Peak	2 θ	FWHM (deg)	Crystallite size (nm)
1	111	38.513	0.339	24.79
2	002	35.281	0.320	26.02
3	110	32.400	0.056	14.75
4	113	67.804	0.218	43.85
5	202	58.195	0.161	56.40
Average crystallite size				33.16

Table 1.4 Optical band gap energy and Absorption edge

Sample	Optical band gap (eV)	Absorption edge (nm)
ZnTiO ₃	3.7	284
CuO	1.2	291
TiO ₂	3.52	291,462

Table 1.5 Short-circuit current, Maximum Current ,Open-circuit voltage, Maximum Voltage, Fill factor and Efficiency of ZnTiO₃ Perovskite Solar Cells

Solar Cell	J _{sc} (A)	J _{max} (A)	V _{oc} (V)	V _{max} (V)	Fill factor (%)	Efficiency (%)
ITO	5.34E-04	3.35E-4	0.2172	0.1065	30.8	2.12

Conclusion

The perovskite structure of ZnTiO₃ was formed at 700 °C, 800 °C and 850 °C. As the detail analysis of the phase by XRD, ZnTiO₃ was successfully formed at 700 °C, 800 °C and 850 °C. SEM analysis of ZnTiO₃ showed advantageous microstructure with the appropriate pores, materials ratio and applications in the electronic industry as catalysts and color pigments. From the SEM results, it was found that all SEM images were porous structures. Small TiO₂ particles were formed on glass substrate. As a result of UV-vis spectroscopic measurement, the optical band-gap of TiO₂, CuO and ZnTiO₃ were examined according their wavelengths. Therefore, light energy can absorb easily. From J-V characteristics of ZnTiO₃ perovskite solar cell with ITO (Indian doped tin oxide) substrate showed the photovoltaic behaviour. The efficiency of ZnTiO₃ perovskite solar cell with ITO substrate is 2.12 %.The efficiency of ZnTiO₃ 2.12% is quite satisfied and credible compared with other photovoltaic applications (in Nature Energy,2019). The result of fill factor (30.8%) showed that ZnTiO₃ perovskite solar cell with ITO substrate exhibited a promising application in the preparation of perovskite solar cell.

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