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ORIGINAL RESEARCH ARTICLE

Fabrication and Characterization of Multijunction Solar Cell of *Cu/Cu₂O/ZnO/FTO* Via Spray Pyrolysis Technique

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ABSTRACT
Photovoltaic cells made of silicon and germanium, which are currently in use, eliminate fuel transport and storage issues but are hindered by the current cost of the materials and fabrication. The spray pyrolysis technique was employed to fabricate and characterize multijunction $Cu/Cu_2O/ZnO/FTO$ solar cell due to its simplicity, reproducibility, and economic factors. Current-voltage characteristics indicated an increase in current and efficiency when a current collection grid was used. The fabricated tandem solar cell showed an open voltage V_{OC} of 720 mV, a short circuit I_{SC} of 3.25 mA, a fill factor FF of 0.68, a maximum power P_{max} of 1.61 $mW\,cm^{-2}$, and a conversion efficiency (η) of 1.57%. Based on the findings, it can be concluded that the fabrication process and characterization techniques employed in this study resulted in the successful development of a $Cu/Cu_2/ZnO/FTO$ tandem solar cell with enhanced performance. The use of ZnO-Mg films contributed to improved absorption coefficient and energy band gap, leading to higher efficiency.

INTRODUCTION

Solar cells are electronic devices that use the photovoltaic (PV) effect to directly convert solar energy into electrical power. Along with other additional benefits, the process of converting solar energy into electricity is pollution-free and seems a good workable answer to the world's energy issues, provided that viable, affordable direct conversion methods can be developed. (Abdu *et al.,* 2009). The two main processes of a solar cell are the photo-generation of charge carriers in a light-absorbing material and the separation of the charge carriers to a conductive contact that will transmit the electricity. A solar cell consists of a junction formed between a p-type and an n-type semiconductor, either of the same material (homojunction) or different materials (heterojunction). Due to their non-renewable nature and constantly rising demand, the world's energy supplies are not predicted to last for very long.

In addition to not being renewable, they mostly come from fossil fuels and play a significant role in the issue of global warming. The global community is concentrating its attention on alternative energy sources due to the significant pollution and depletion issues with the energy sources mentioned above, and solar energy is a very promising option. The sun emits this energy as electromagnetic radiation in the (0.2 – 0.3 μm) spectral range, which spans from ultraviolet to infrared. With a predicted continuous radiative energy output of more than 10 billion (10^{10}) years, the sun has a stable lifespan. (Sze, 1981). Solar cells respond linearly to solar flux, which gives them an advantage over thermal conversion modes in practice.

In contrast to solar thermal systems, which require time to reach operating temperatures, solar cell systems do not have inertia and generate output at a level proportional to solar intensity.

The population of the world will continue to influence its energy demand as well as countries development, multi-junction solar cells will continue playing a vital role due to their ability in efficient conversion of solar energy without being bound up to **33%.** (Adil, B, and Maksym, Y., 2022). Photovoltaic cells made of silicon and germanium, which are currently in use, eliminate fuel transport and storage issues but are hindered by the current cost of the materials and fabrication (Musa *et al.*, 1998).

Copper (I) oxide (Cu_2O) is a non-stoichiometric defect p-type semiconductor (Walter, 1951), and its potential for

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the design of solar cells has been recognised since 1920. Cuprous oxide semiconductors are promising elements of a whole functional oxide solar cell material because of their photo electronic properties, band gap energy of 2.1eV, environmentally friendly properties such as non-toxicity and low material cost (Fujimoto *et al.*, 2013; Noda *et al.*, 2013). ZnO is a transparent oxide that is widely used in many different applications, including thin-film solar cells. ZnO is considered a potential material for transparent electrodes and electronics because of its wide band gap

Since 1920, the potential of copper (I) oxide (Cu_2O), a non-stoichiometric defect p-type semiconductor (Walter, 1951) for solar cell design has been acknowledged. Because of their photoelectric properties, 2.1 eV optical band gap energy, non-toxicity, and low material cost, cuprous oxide semiconductors hold great promise as components of a complete functional oxide solar cell material (Fujimoto *et al.*, 2013; Noda *et al.*, 2013). ZnO is a transparent oxide that finds extensive use in thin-film solar cells and other applications. ZnO direct wide band gap makes it a promising material for transparent electrodes and electronics. (Gordon *et al.*, 2002).

The efficiency of a solar cell can be increased by stacking multiple solar cells with a range of band gap energies, resulting in a multi-junction solar cell (NREL, 2021). Since Group III-V compound semiconductors can be grown with excellent material quality and have direct band gaps, which produce a high absorption coefficient, they are good candidates for the fabrication of multijunction solar cells (Dimroth and Kurtz, 2007; Sagol *et al.*, 2007).

Because transparent conducting oxide thin films are widely used in optoelectronic devices like touch screens, liquid crystal displays, solar cells, and light-emitting diodes, research on them has changed over the past few decades (Chen *et al.*, 2021).

Transparent Conducting Oxides (TCOs) should have both high electrical conductivity and high optical transparency in the visible region (Kim *et al.*, 2015). Because of its high electrical conductivity and transparency, indium tin oxide (ITO) is used extensively; however, finding a non-toxic, affordable substitute has proven difficult. (Kim *et al.*, 2015). Because of its abundance, non-toxic, and favourable optical and electrical qualities, zinc oxide (ZnO) is a preferable substitute for ITO. (Kim *et al.*, 2015). ZnO is an n-type semiconductor material with a wide band gap of 3.3eV and is used in a variety of applications, including electronic devices and sensors (Chen *et al.*, 2021). Intrinsic ZnO is transparent, but it must be doped to enhance its n-type conductivity. ZnO layers can be doped with dopants such as Al, Ga, In, Mg, and F to improve their n-type conductivity (Chin *et al.*, 2016). The electrical and optical properties of ZnO can be optimized by controlling the deposition conditions and the doping process (Chin *et al.*, 2016). In this work, Cu / Cu_2O Schottky barrier solar cells were fabricated and characterized, and ZnO layers were grown on the Cu_2O surfaces to form a multi-junction solar cell structure of Cu / Cu_2O/ZnO / FTO. The ZnO was doped with Mg to improve its conductivity.

MATERIALS AND METHOD FOR THE CELL FABRICATION

This section discussed in detail the materials used for the fabrication of Cu / Cu_2O/ZnO / FTO tandem solar cell.

Spray pyrolysis deposition of Cu / Cu_2O/ZnO / FTO tandem solar cell

High-efficiency solar cells can be achieved through the development of tandem or multijunction solar cells. Tandem junction involves stacking two or more p-n junctions on top of each other, while multijunction cells split the solar energy spectrum into several sub-bands and treat each sub-band separately with an appropriate solar cell material. The most common tandem solar cells consist of three junctions; however, research is underway to explore designs with more junctions (Dimorth and Kurtz., 2007).

Tsakalakos *et al.* (2010) reported that a theoretical efficiency of 44%, 54%, and 66% could be achieved by two-junction, three-junction, and an infinite stack of multijunction solar cells, respectively.

Steps for the spray pyrolysis deposition.

a) Substrate Pre-cleaning

The FTO substrate of thickness 1 mm and dimension 75 mm by 25 mm was washed with propanol and dried in a spin coater (Lab science model 800) at 3000 rpm.

b) Substrate Etching

A laser cut resist in the form of adhesive film was applied on the substrate, which was subsequently covered with zinc powder and etched with 2M HCL Solution to remove FTO from the "etch area"). Final etching was carried out after the adhesive film was removed and the substrate was washed thoroughly with propanol and dried. Final cleaning was carried out by heating the substrate to 400^{o}C/30min on a hot plate to oxidize organic surface contaminants.

c) Deposition of ZnO and ZnO-Mg

The spray pyrolysis was used to deposit the undoped ZnO and the Mg-doped ZnO from 0.3M of zinc acetate dihydrate. $[Zn(CH_3COO)\ _2 2H_2O]$ (Baker Chemical Co.) and 0.03M Magnesium acetate $[Mg(CH_3COO)_2.4H_2O]$(MERCK) which were dissolved in a mixture of methanol and deionized water. This was done to increase the electrical conductivity of

ZnO as magnesium is a group II element with fewer electrons in its outer shell than zinc, leading to the introduction of free charge carriers (holes) in the material. To stop zinc hydroxide from precipitating, 0.5 ml/40 ml of acetic acid (MERCK) was added to the prepared solution while the methanol to deionised water ratio was kept at 2:1. (Muchuweni *et al.*, 2017).

The prepared solution was sprayed directly onto the substrate, which was set on a hot plate that was at 400 ℃, using an infusion syringe that was pumped at a steady flow rate of 0.1 millilitres per minute. The sprayed solution was atomised at 6 kV using a stream of compressed air through a nozzle positioned 12 mm above the substrate for a 5-minute deposition period.

d) Metal Back Contact Formation

Carbon conductive paste (Elcocarb solaronix) was applied as the back metal contact by the doctor blade method, followed by annealing at 200℃ for 30 minutes.

Plate 1: The fabricated $Cu / Cu_2O/ZnO / FTO$ tandem Cell Structure

CHARACTERIZATION OF THE CELL

Optical Measurement

The UV/VIS/NIR spectrophotometer (Perkin-Elmer, America) was used to measure the materials' optical characteristics in the 250–700 nm wavelength range. This range includes the interaction of light with the slide film in the visible and ultraviolet light spectrums. The transmittance, reflectance, and absorption were measured.(Huiling *et al.*, 2011).

The Tauc model used in the absorption region provided by Muchuweni *et al.* was employed to estimate the optical band gap (E_g).(Muchuweni *et al.*, 2016).

$$(\alpha h v)^2 = B(hv - E_g) \quad (1.0)$$

where B is an energy-independent constant, and hv is the incident photon's energy. The E_g can be determined by extrapolating the linear part of the absorption edge from the Tauc plot of $(\alpha h v)^2$ versus hv. Additionally, the Beer-Lambert law can be used to determine the optical absorption coefficient (α) in the strong absorption region.(Sandeep *et al*,, 2012; Mohammed *et al*, 2017).

$$\alpha = 2.303\frac{A}{d} \quad (1.1)$$

with d and A as the thickness of the film and absorbance, respectively.

I-V Characterization

The I-V characterization measurement is required to assess a solar cell's performance. The open-circuit voltage(V_{oc}), the short-circuit current density(I_{sc}), the fill factor (FF), and the electrical power conversion efficiency(η) are the most crucial solar cell parameters. These parameters determine a solar cell's output performance. Solar cells have typical diode characteristics in the dark because of the barrier layer, which is necessary for the photovoltaic effect. When illuminated, a current component is added that is almost independent of voltage and shifts the characteristic along the current axis by J_{sc} , known as the short circuit current density.

The short-circuit current density (J_{sc}) as given in equation (3.2) is the current that flows through the junction under illumination at zero applied voltage (i.e., V = 0). In the ideal case (when R_s and R_{sh} effects are negligible) it is equal to the light-generated current density (J_L).

According to equation (3.2), the current that passes through the junction when there is no applied voltage (i.e., V = 0) is known as the short-circuit current density (J_{sc}). In the ideal case, when the effects are minimal, it is equivalent to the light-generated current density (J_{sc}).

$$J_{sc} = J_L - J_0\left[exp\left(\frac{qV}{AKT}\right) - 1\right] \quad (1.2)$$

An electrical device's voltage on open circuit is known as its open circuit voltage. When the device is operating at open circuit voltage, the terminal current density is zero (i.e $J = 0$).

$$V_{oc} = \frac{AkT}{q}\ln\left(\frac{J_{sc}}{J_o}\right) \quad (1.3)$$

The fill factor is a measure of the deviation of the real J-V characteristic from the ideal one, given as the ratio of the maximum output power that can be extracted from the cell to the product of J_{sc} and V_{oc} and is a measure of the "squareness" of the J-V curve

The fill factor, which is a measure of the "squareness" of the J-V curve, is the ratio of the maximum output power that can be extracted from the cell to the product of J_{sc} and V_{oc}. It quantifies the difference between the real and ideal J-V characteristics (Musa, 2010).

$$FF = \frac{P_{max}}{J_{sc}V_{oc}} \quad (1.4)$$

The electrical power conversion efficiency of a solar cell is expressed as the ratio of the power output to the power input.

$$\eta = \left(\frac{FF \times J_{sc}V_{oc}}{P_{in}}\right) \times 100\% \quad (1.5)$$

RESULTS AND DISCUSSION

Optical measurement analysis for Cu_2O

When the linear part of the absorption edge for Cu_2O is extrapolated to $(\alpha h\nu) = 0$, the energy band gap of 2.11 eV was obtained, as shown in Figure 1's plot of $(\alpha h\nu)^2$ versus $(h\nu)$. This value agrees with previous reports by Katayama *et al.* (2004), Huiling *et al.* (2011), Bessek *et al.* (2005), and Huang *et al.* (2009).

The optical band gap measurement for ZnO

The UV-Vis spectra graph of $(\alpha h\nu)^2$ versus $(h\nu)$, shown in Figure 2, was plotted for ZnO thin film, and the energy band gap was found to be 3.3eV. The result is in agreement with the result of Katayama, (2004).

I-V Characterization Result for Cu / Cu_2O/ZnO / FTO Solar Cell.

The I-V and P-V characteristic curves for the fabricated tandem solar cell were plotted using the values obtained with the solar cell simulator. The short circuit currentI_{sc} and the open circuit voltage V_{oc}values for the cell were obtained from the curve in Figure 3.

The open circuit voltage, V_{oc} and the short-circuit current I_{sc} values for the cell doped with magnesium were found to be 720 mV and 3.25 mA, respectively. The maximum power point of the cell was obtained from the P-V curve (Figure 4) as 1.61mWcm^{-2}. The fill factor, *FF, was* found to be 0.68, while the corresponding electrical power conversion efficiency *is* η = 1.57%. Thus, the fabrication and characterization of the Cu/Cu_2O /ZnO/FTO tandem solar cell yielded promising results using the electrostatic spray pyrolysis technique.

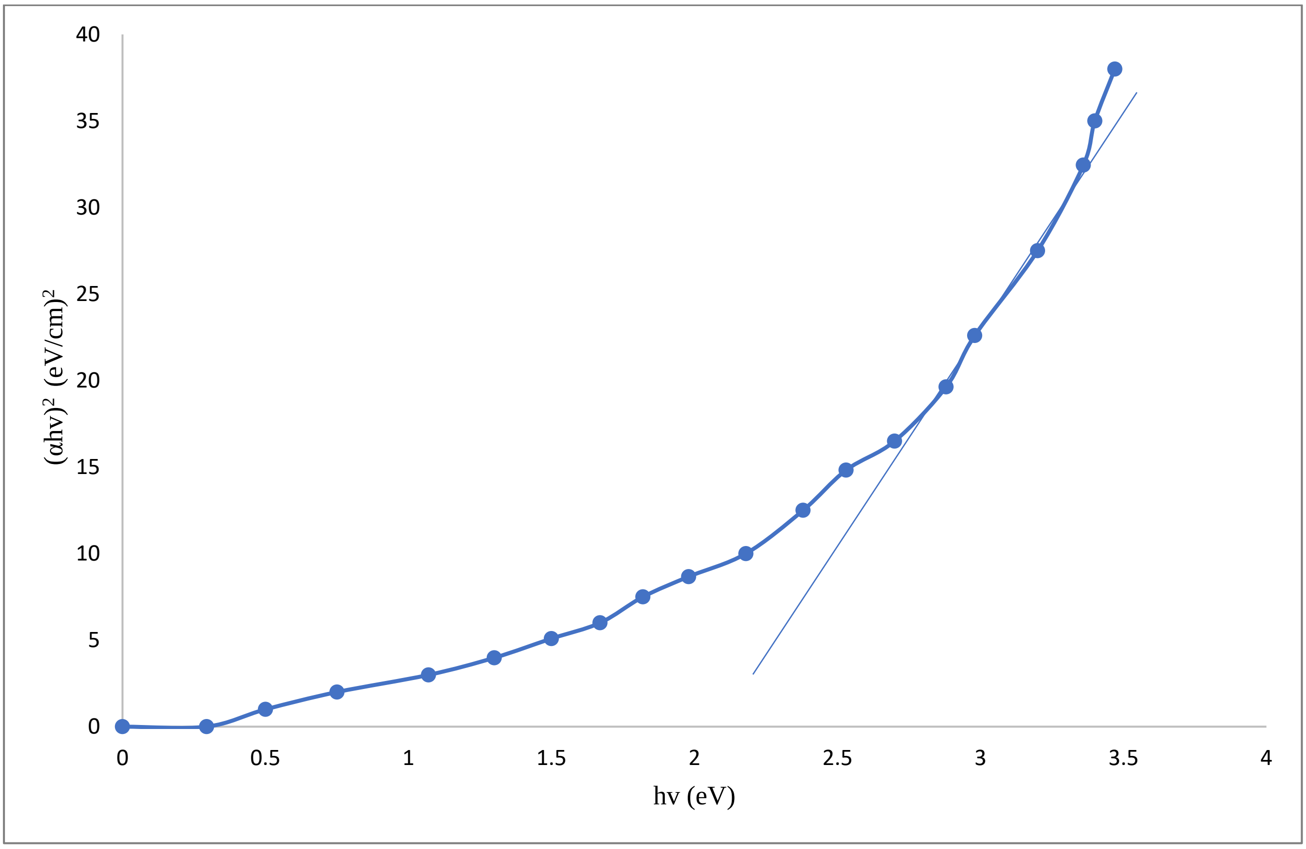

Figure 1: A graph of $(\alpha h\nu)^2$ versus $(h\nu)$ for the optical band gap energy of Cu_2O

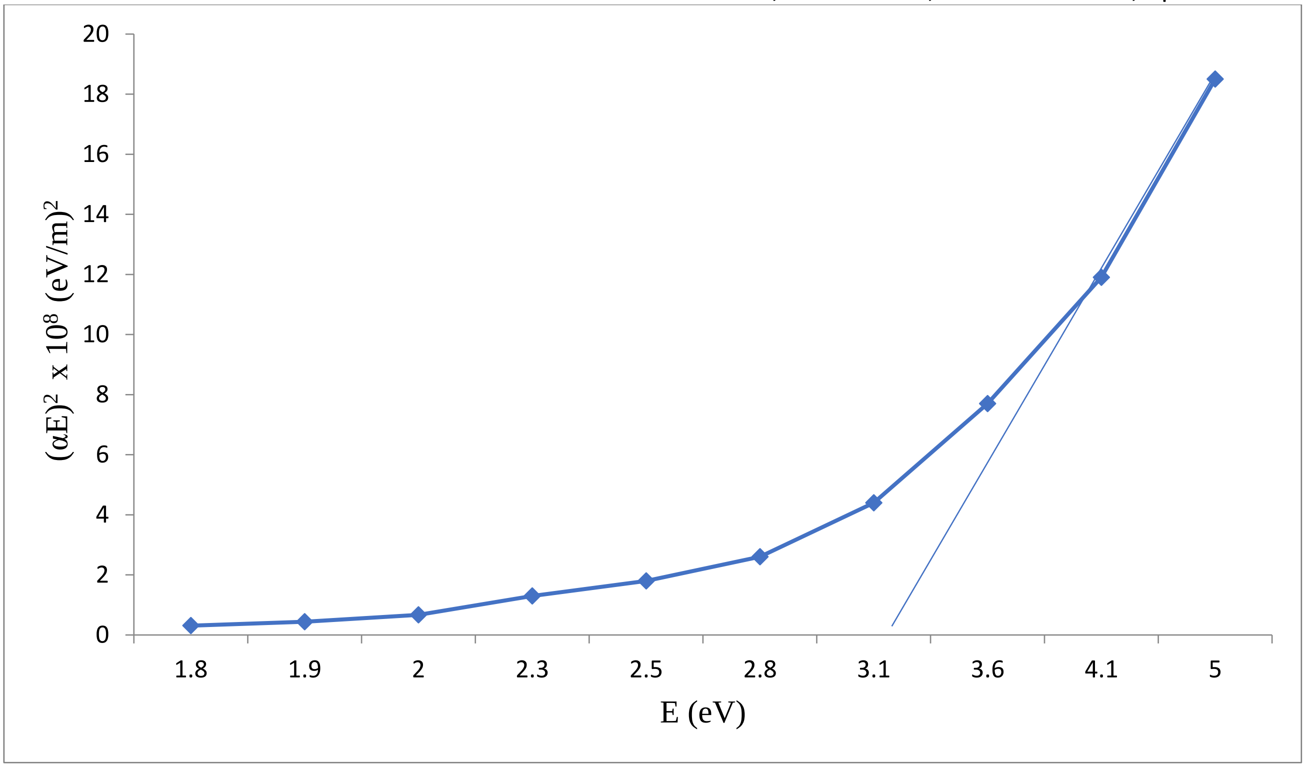

Figure 2: A graph of $(\alpha h\nu)^2$ versus $(h\nu)$ for the optical band gap energy of ZnO

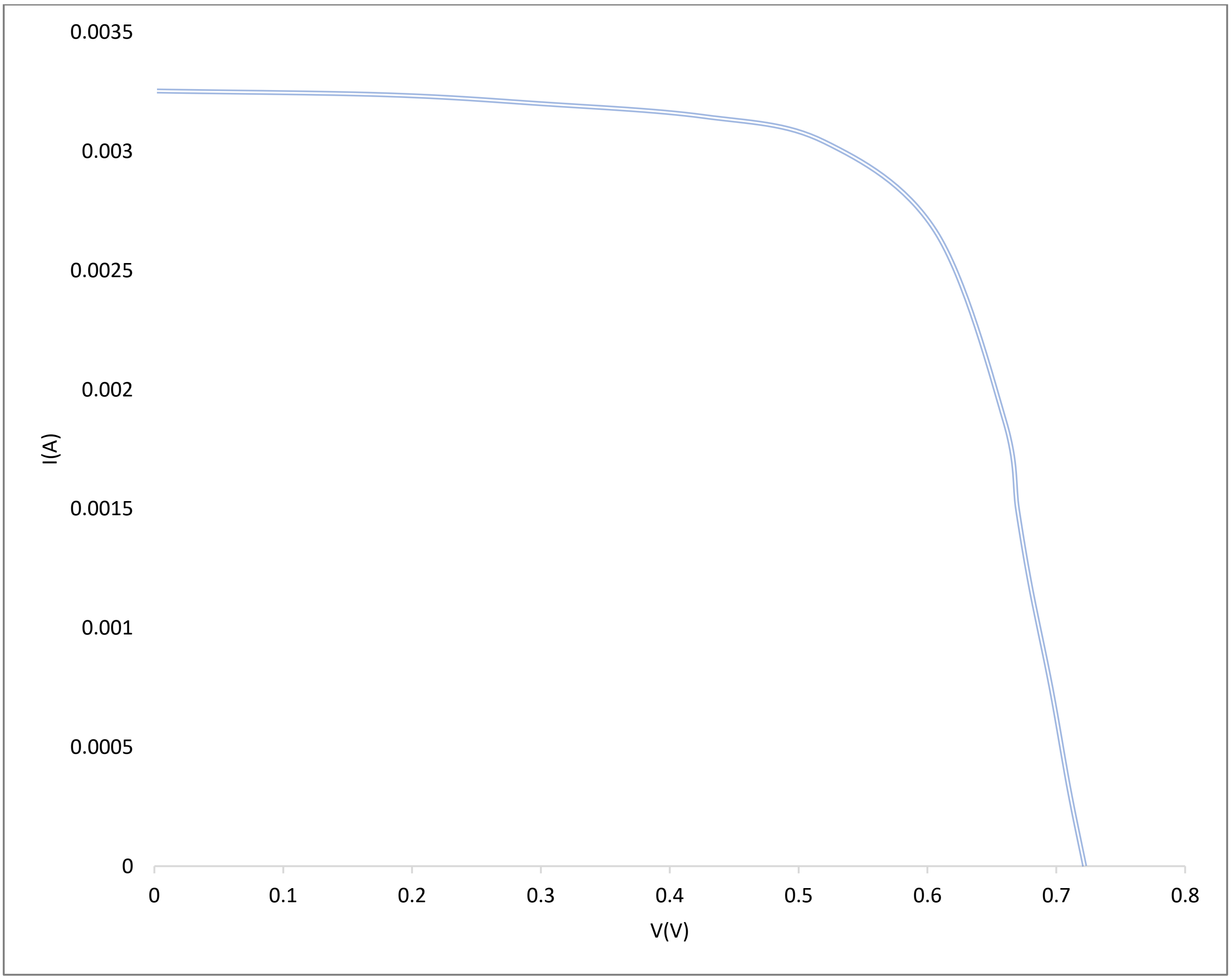

Figure 3: The I-V characteristic curve for the doped fabricated solar cell

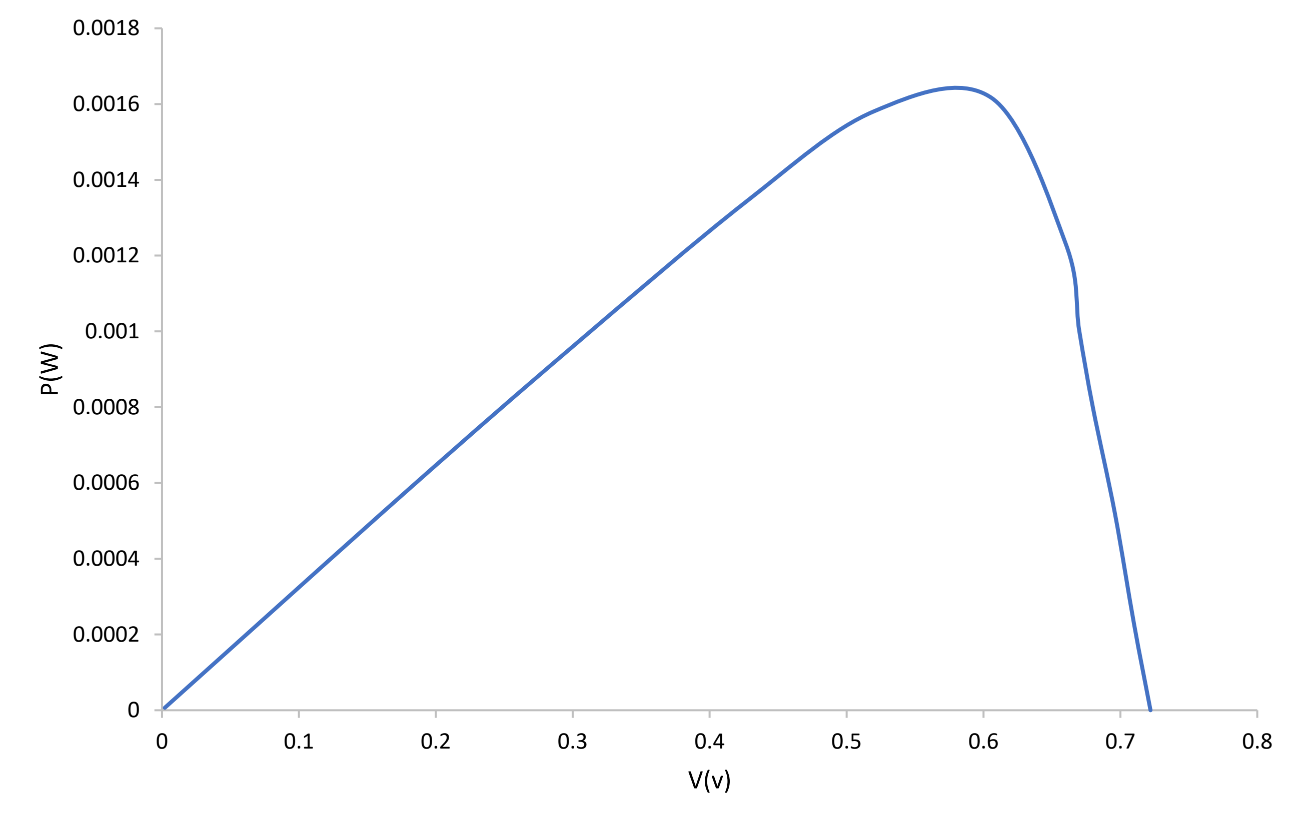

Figure 4: The P-V characteristic curve for the doped fabricated solar cell

CONCLUSION

The electrostatic spray pyrolysis method was successfully and effectively utilised to fabricate the Cu/Cu_2O /ZnO/FTO tandem solar cell, which yielded promising results, making the method economical and highly reproducible, with the optical band gap measurement of 2.11 eV and 3.3 eV.

While the I-V characteristic measurements of the most important parameters of the fabricated magnesium-doped tandem solar cell, which provided the output performance of a solar cell, the open-circuit voltage (V_{oc}), the short-circuit current density (I_{sc}), the maximum power point of the cell (P_{max}), the fill factor (FF) and the electrical power conversion efficiency (η) were found to be 720 mV, 3.25 mA, 1.61 mWcm^{-2}, 0.68 and 1.57%, respectively.

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