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THE IMPLICATION OF ANNEALING TEMPERATURE ON ZINC OXIDE (ZnO) BASED PEROVSKITE METHYLAMMONIUM LEAD BROMIDE ($CH_3NH_3PbBr_3$) USING HYDROTHERMAL BATHING AND SPIN COATING DEPOSITION METHODS

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Abstract: The implication of annealing temperature on zinc oxide (ZnO) based perovskite, Methylammonium lead Bromide ($CH_3NH_3PbBr_3$) using hydrothermal bathing and spin coating deposition methods was investigated; which is an advantageous thin film technique for deposition of large films at ambient or low temperature. The zinc oxide (ZnO) based perovskite, Methylammonium lead Bromide ($CH_3NH_3PbBr_3$) were successfully deposited on the substrate (Glass slide) at a deposition time of 3 hours and were annealed at different temperatures of 100˚, 130˚, 160˚,190˚ and 200˚ degree Celsius respectively. The optical and morphological characterization of the zinc oxide nanoparticles and the perovskite material sequentially deposited on the substrate (Glass slides) annealed at different temperatures of 100˚, 130˚, 160˚, 190˚, 200˚ degree Celsius at deposition time of 3 hours were carried out using the UV – Visible Spectrophotometer and x- ray diffractometer respectively. It shows a transmittance between 1.42 a.u to 1.48 a.u of the thin film within the wavelength band of 350nm – 1500nm and a direct allowed band gap of 3.33 eV – 3.35 eV. UV- VIS spectroscopy revealed that the zinc oxide nanoparticles and the perovskite material sequentially deposited on the substrate (Glass slides) and annealed at temperatures of 130˚degree Celsius at deposition time of 3 hours has the highest efficiency because it absorbed readily at the least absorbance of 3.38 a.u and transmitted highly at a transmittance of 1.48 a.u within the visible region when compared to other annealing temperatures.

Keywords: ZnO nanoparticles, Methylammonium lead Bromide ($CH_3NH_3PbBr_3$), Optical properties: Band gap, Hydrothermal and spin coating deposition techniques.

INTRODUCTION

A solar cell is a modular device that changes the incident solar beams into electrical energy through an electrochemical reaction (the photovoltaic effect). Thin film is prevalent and real photocell for collection of solar energy. Perovskites, in contrast to crystalline silicon, consist of a family of materials, named after the minerals

that make them up, and also named after Lev Perovsky. Perovskite was first studied as an absorbing material in 2006, with results published in 2009. In the solar industry, perovskites remain the utmost optimistic of the third-generation photovoltaics cell. In just five years of existence, the proficiency of perovskite solar cells increased from less than 4% to more than 20%, and after a little over 15 years the efficiency increased even further, reaching 30% perovskite solar cell efficiency [16]. Several limitations can affect the productivity of perovskite solar cells; Solvent selection [1], deposition approach [7,16,11], precursor concentration [15,9,6], the use of different electron- and hole-transporting materials, and different architectures. obtaining the suitable hardening temperature is likewise a significant parameter for improving solar cell efficiency. Furthermore, thermal annealing positively affects cell efficiency to a positive limit. Overall, thermal annealing is a key step for initiating or accelerating intermolecular reactions throughout perovskite film formation [20]. In this study, the implication of annealing temperature on zinc oxide (ZnO) based perovskite, Methylammonium lead Bromide ($CH_3NH_3PbBr_3$) was investigated. Thin film is common and current photocell for collection of solar energy. In some cases, the total energy collected by these photocells remain directly proportionate to the surface area [18]. When a solution of a material or substance is deposited on the substrate, is known as thin film [19]. When these thin films are exposed to electromagnetic radiation such as solar radiation, their optical properties are found to be better in rating. Throughout the exposure, their optical properties take up dissimilar wavelength of the radiation [19], such process of optical properties changing by wavelength is identified as Spectral selectivity as seen in work of [2]. The optical as well their solid-state properties were studied. Hence, it is very important to give little theoretical consideration and calculation of some of these parameters. These consist of transmittance (T), absorbance (A) and as well reflectance (R) which is used in the calculation of absorption coefficient (α) and band gap measurement.

THE TRANSMITTANCE

It is defined that transmittance of a material is the portion of light that passes through to the supplementary side of a surface. It can be observed that these light rays are either transmitted, reflected, or absorbed when light passes through any material [5]. Transmittance and reflectance are closely related.

Transmittance is well-defined as a fraction of the intensity of incident light (I) to the quantity or amount of intensity going over the object (I_0). The transmittance is denoted as T.

In an equation form, the transmittance T_λ is given as:

$$T_\lambda = \frac{I}{I_0} \qquad 1$$

where I is the amount of the radiation emanating from the sample and the incident radiation intensity is denoted as I_0 [5]. In these calculations, scattering and reflection are measured to be close to zero.

Relative to absorbance

Transmittance is associated to absorbance A as given in giving in equation 2 :

$$T = 10^{-A} \qquad 2$$

Relation to optical depth

Transmittance is associated to optical depth τ_λ as

$$T = e^{-\tau} = 10^{-\tau_{dB}}$$

From the above equation for the inverse problem, the transmittance is hence given as:

$$\tau = -\ln T = -\ln\left(\frac{I}{I_0}\right) \quad 3$$

Hence optical depth in decibels can be expressed as:

$$\tau_{dB} = -10\log_{10} T = 10\log_{10}\left(\frac{I_0}{I}\right) \quad 4$$

ABSORBANCE (A)

Absorbance (A) is simply the amount of light captured or absorbed by a material. Whereas Transmittance simply stands as the number of light rays that passes through a material. Absorbance as well transmittance remain frequently applied in spectrophotometry and is stated as [13]:

Absorbance is given as follows:

$A = \text{Log}_{10} (I_0/I)$ 5

The intensity of the incident rays is denoted as I_0 , I denotes intensity of that light rays subsequently as it passes through the sample

$T = I/I_0$ and $\%T = 100\ (T)$ 6

The expression can be used to compute absorbance from percent (%) of transmittance

$A = 2 - \log_{10} (\%T)$ 7

REFLECTANCE

Reflectance is the portion of all radiant change falling upon a surface that is reflected and that changes giving to the wavelength dispersal of the falling radiation. The foundation of this method is that dissimilar substances display exceptional reflectance and emission characteristics as a function of wavelength across the electromagnetic spectrum [12].

Hence,

$R + T + A = 1$ 8

$R = 1- A -T$ 9

Where L = absorbance of light without any optical loss. Therefore, L = A + S where, A = absorbance, S = optical loss

It follows that if the optical loss is negligible, then L = A, that is equation (8) holds under that condition. therefore, R = 1 – T – L

$R = 1 – T – (A + S)$ 10

If, L = A

$R = 1 – (T + A)$ 11

EXPERIMENTATION:

Material preparation involves the Synthesis of Methylammonium bromide (CH_3NH_3Br) by reacting 38ml of methylamine with 40mls of Hydrobromic acid (HBr) in a 250 ml rounded bottomed flask at O˚C (maintained at this temperature with ice block), and stirring the yellow solution without heating for 2hrs to 3hrs. Subsequently, the methylammonium bromide was dried up at 60˚C in an oven. In conclusion, the Methylammonium bromide powder produced was stored at 25˚C in an oven. Also, Lead bromide **(**$PbBr_2$**)** was synthesized by reacting 5g of

lead nitrate Pb $(NO_3)_2$ with 5g of potassium bromide (KBr) in a 250ml round bottomed flask to yield lead Bromide ($PbBr_2$). Also, the solution was stirred continuously and heated to 80˚C at the same time for 20 Minutes. To obtain the residue lead bromide **(**$PbBr_2$**)** which is white in colour, the solution was filtered using filter paper and left to cool. The seed layer is a material that provides a nucleation site or a surface for an easy adhesion of the zinc oxide nanoparticles on the substrate. The seed layer was prepared by melting 1.32g of zinc acetate in 60ml of ethanol. The mixture was incited in the magnetic stirrer machine for two hours, during which 1.5ml of distilled water was added drop wise. Solution of the Precursor used for deposition was achieved by dissolving 1.49g of zinc nitrate; Zn $(NO_3)_2.6H_2O$ in 0.70g of Hexamethylenetetramine ($C_6H_{12}N_4$). The mixture was uninterruptedly stirred in the magnetic stirring machine for three hours.

In the treatment of substrate**,** the Glass slide was ultrasonically cleaned for 10 minutes in acetone and deionized water sequentially, using the ultrasonic machine in other to remove any contaminations. In the growth of zinc oxide nanorod's, a thin seed layer of zinc oxide is deposited on the substrate by spin coating machine. The seedling of zinc oxide reduces the thermodynamic barrier by giving nucleation sites and also aids to progress the aspect proportion of the gained rods. The spin coating of the thin seed layer of zinc were repeated for five times by spin coating the seed layer solution on the Glass slides using the spin coating machine at the rate of 3000 rpm for 30 seconds and evaporated in the oven for 10 minutes. The zinc oxide (ZnO) nanoparticles, was put on the substrates by hydrothermal bathing using the acid digester and heating in the oven for deposition time of 3 hours at 90˚C. There are various deposition methods of methylammonium lead Bromide on substrates, which consist of one-step as well two-step sequential deposition methods. The one-step spin coating technique was used to directly deposit the perovskite material from a precursor solution mixture of $PbBr_2$ and CH_3NH_3Br in a polar solvent such as r-butyrolactone (GBL) or N-N- Dimethylformide (DMF) using a spin coating machine revolving at 3000 rmp for 30 seconds, followed by annealing at different temperatures of 100˚, 130˚, 160˚, 190˚, 200˚ degree Celsius to eliminate the additives and form the perovskite thin films. The two-step sequential method involves spin coating the precursor homogenous mixture of lead iodide ($PbBr_2$) in a polar solvent of N-N- dimethylformide (DMF) solution firstly, followed by deposition of the precursor solution of methylammonium iodide in a polar solvent of isopropanol also followed by annealing at different temperatures.

RESULTS AND DISCUSSION

The zinc oxide nanoparticles and the perovskite material were successfully deposited by hydrothermal and spin coating deposition methods respectively on the Glass slides and annealed at different temperatures of 100˚, 130˚, 160˚, 190˚, 200˚ degree Celsius at deposition time of 3 hours and was duly characterized to obtain the optical properties; absorbance, transmittance and as well as reflectance using the UV – Visible Spectrophotometer.

The graph of spectral absorbance, transmittance and as well reflectance versus wavelength are given in figure 1, 2 and 3 respectively. The optical properties revealed in figure 1, 2 and 3 showed that zinc oxide (ZnO) based perovskite, Methylammonium lead Bromide ($CH_3NH_3PbBr_3$) annealed at 130˚ C has the least absorbance of 3.38 a.u, reflectance of - 2.4 a.u and high transmittance of 1.48 a.u when compared to other samples in the UV – VIS region. The calculated annealing temperature in the current study was close to that stated by [14].

Figure 4 to 8 comprises the graph of absorption coefficient in contrast to photon energy (eV) of the five films annealed at dissimilar temperatures of 100˚, 130˚, 160˚, 190˚and 200˚ degree Celsius respectively. Determination of band energy (Eg) was frequently essential to advance the electronic band structure of thin film. Its Absorption coefficient (α) was associated to energy (hv) of incident photon by a process called Tauc's plot technique [4].

$$\alpha h\nu = \beta (h\nu - Eg)^p \qquad 12$$

where it follows that β remained a constant and p remained an index that categorized the optical absorption procedure and was equal to ½, 2, 3/2 or 3 used for direct allowed, indirect allowed, direct forbidden and indirect forbidden transition correspondingly. In the current study of zinc oxide nanoparticles and the perovskite material, the value of p of equation (12) was assigned as ½ [8].

The band gap variation of zinc oxide nanoparticles and the perovskite material with different annealing temperature was shown in figure 4 to 8. The optical band gap energy (Eg) was gotten by deducing the linear portion of the Tauc's plot to cut the energy axis at $(\alpha h\nu)^2 = 0$ [3].

The optical band gap energy for the zinc oxide nanoparticles and the perovskite material grown at 100˚C stayed 3.35 eV. As the temperature was raised to 130˚, 160˚, 190˚and 200˚ degree Celsius respectively, the optical band gap shifted to 3.34 eV, 3.34 eV, 3.34 eV and 3.33 eV correspondingly. The band gap energy of ZnO nanoparticles and perovskite materials decreased with temperature.

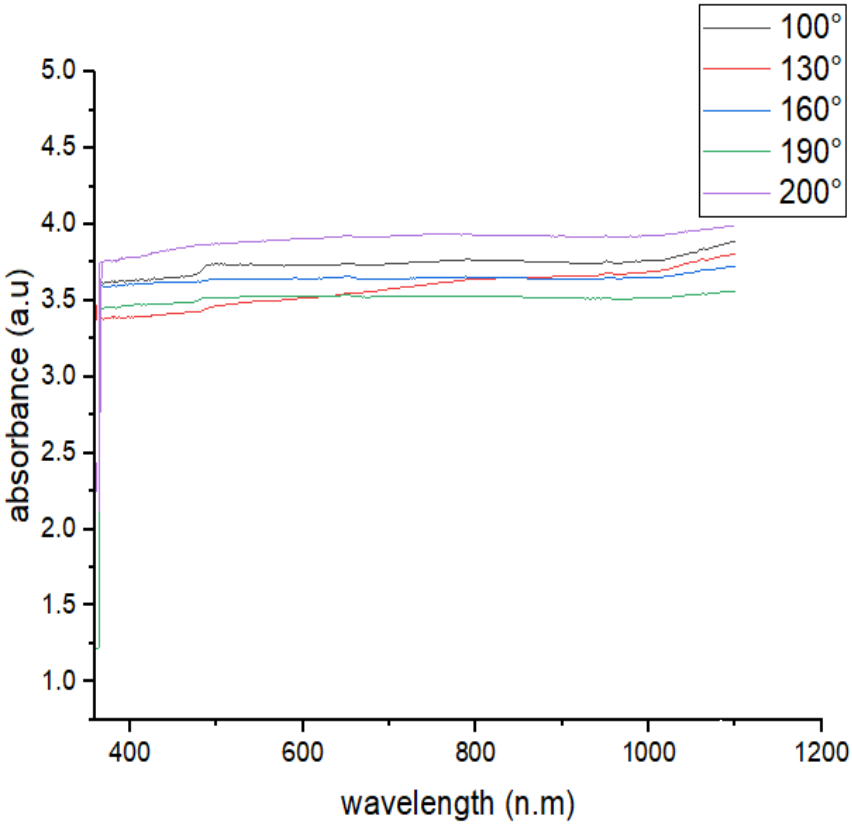

FIGURE1: Absorbance spectra of ZnO nanoparticles and perovskite materials

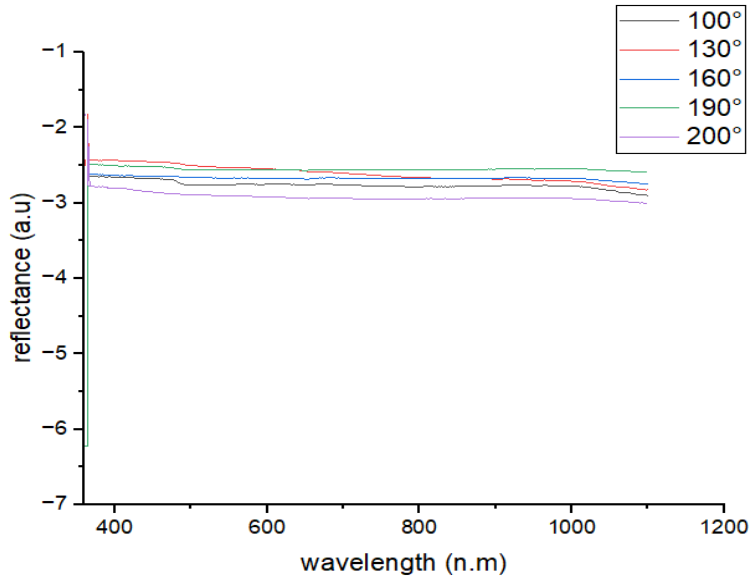

FIGURE2: Reflectance spectra of ZnO nanoparticles and perovskite materials

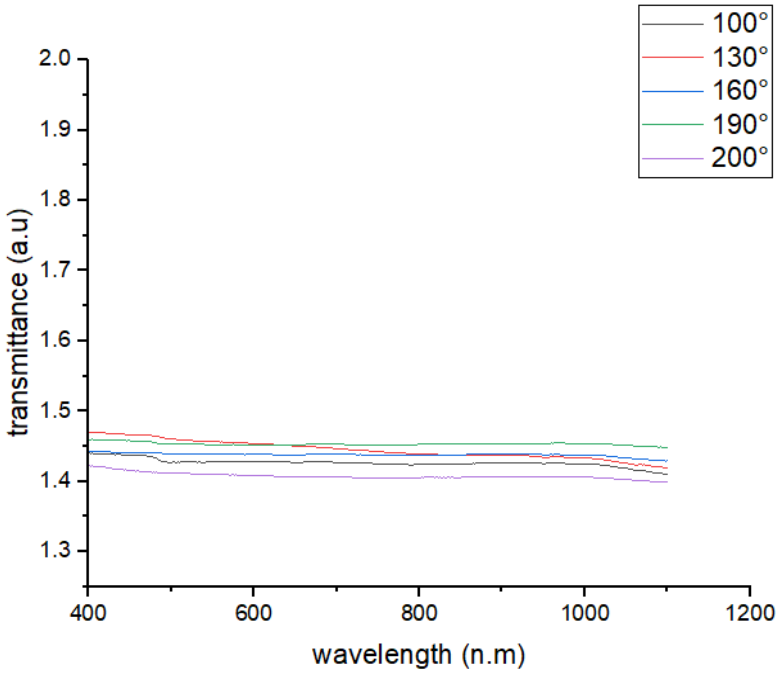

FIGURE 3: Transmittance spectra of ZnO nanoparticles and perovskite materials

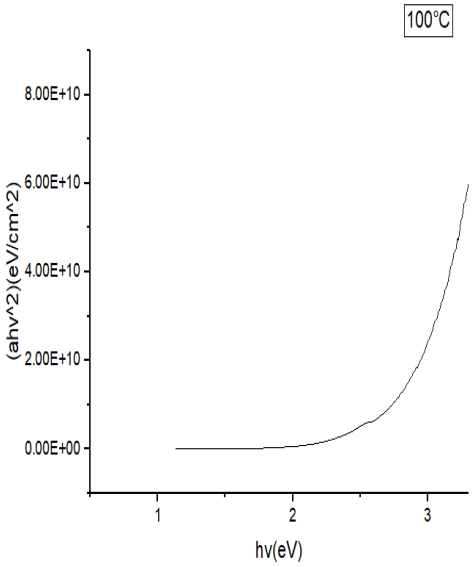

FIGURE 4: Energy band gap of the ZnO nanoparticles and perovskite materials annealed at 100˚C

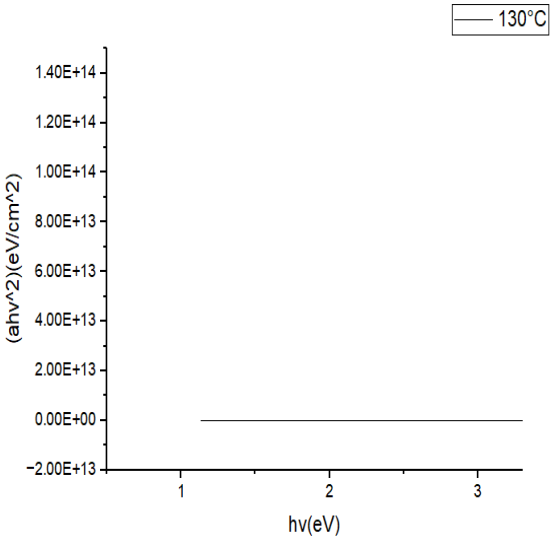

FIGURE 5: Energy band gap of the ZnO nanoparticles and perovskite materials annealed at 130˚C

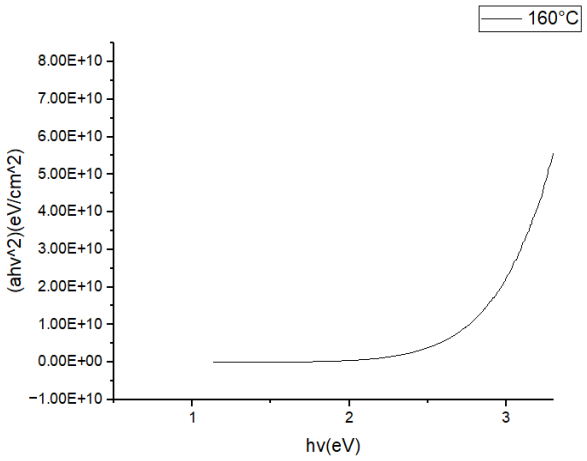

FIGURE 6: Energy band gap of the ZnO nanoparticles and perovskite materials annealed at 160˚C

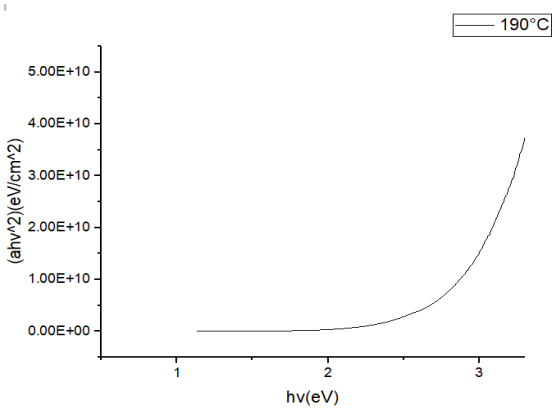

FIGURE 7: Energy band gap of the ZnO nanoparticles and perovskite materials annealed at 190˚C

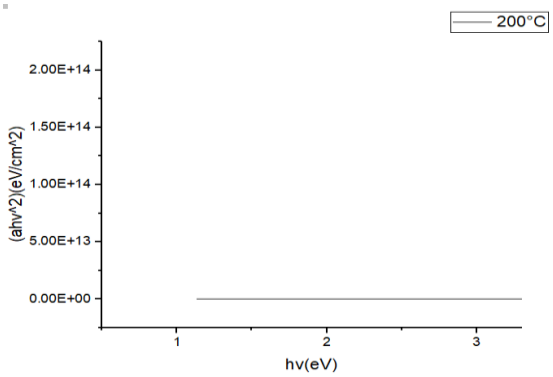

FIGURE 8: Energy band gap of the ZnO nanoparticles and perovskite materials annealed at 200˚C

X-RAY DIFFRACTION RESULTS (XRD): The crystal structure of the deposited Zinc oxide (ZnO) and Perovskites materials were obtained by X- ray Diffractometer (XRD), MIDI 10 mini range of 10˚ - 80˚ using Cukα radiation (λ= 1.5406Å̃). The slides containing the deposited Zinc oxide (ZnO) thin films and perovskite materials were analyzed by X- ray Diffractometer in other to determine crystalline structure of the deposited materials produced, crystalline size of deposited materials and the inter – planar spacing of the given atoms. The x – ray diffraction results of ZnO nanoparticles and perovskite materials increased with temperature. The grown zinc oxide-based perovskite was of untainted sample and no diffraction peak as of slightly additional impurities were noticed. Completely, the diffraction peaks are somewhat sharp as shown in fig. 11 to 13; and least as shown in fig. 9 and 10. The pattern displayed for diffraction peak at 2θ values between 10˚ and 80˚ revealed that, ZnO nanoparticles and perovskite materials annealed at 200˚C has the highest peak values of 28.5˚, with a corresponding intensity value of 53 a.u.

The particle size values of the crystals were calculated from Debye Scherrer formula equation.

$$D = \frac{0.9\lambda}{\beta\cos\theta} \qquad 13$$

Where D is taken as the grain size, λ stands as the X – ray wavelength (0.15406nm), β stands as the full width of Half maximum intensity (FWHM) in radian of the diffraction peak and θ remains the diffraction angle [6]. The value of D is about 2.9957 x 10^{-4}nm.

The inter – planar spacing deducted from Braggs law

$$d_{hkl} = \frac{n\lambda}{2sin\theta} \qquad 14$$

λ = 1.5406Å̃, $\theta = \frac{2\theta}{2}, n = 1$

Gives a mean value of the inter – planar spacing ‘a’ after putting in all the data above for various peak values (2θ) as d_{hkl} = 2.03607Å̃.

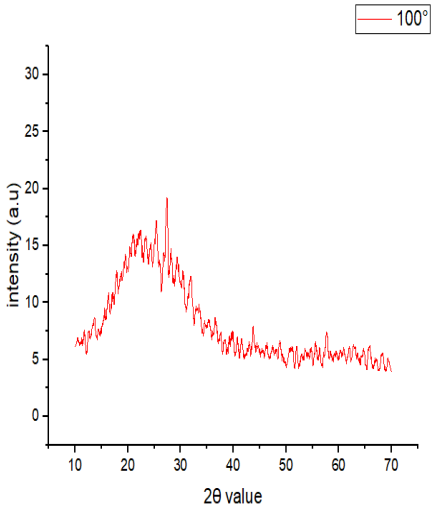

FIGURE 9: XRD result of the ZnO nanoparticles and perovskite materials annealed at 100˚C

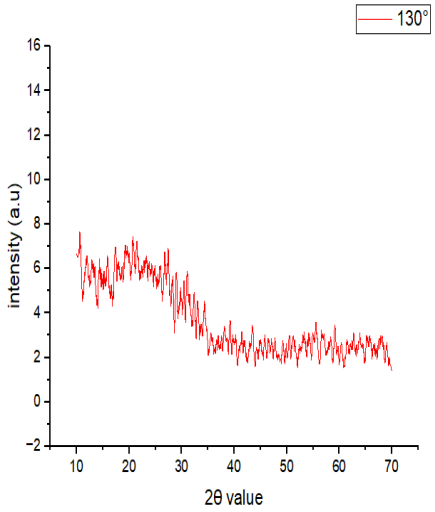

FIGURE 10: XRD result of the ZnO nanoparticles and perovskite materials annealed at 130˚C

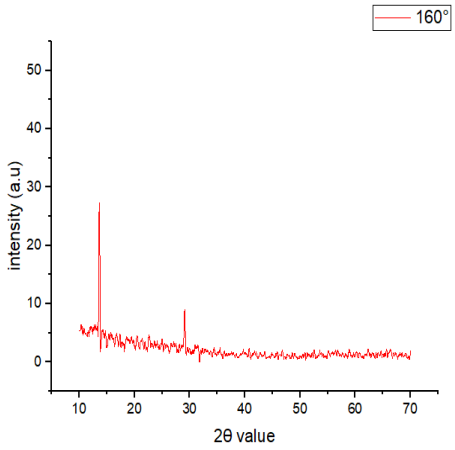

FIGURE 11: XRD result of the ZnO nanoparticles and perovskite materials annealed at 160˚C

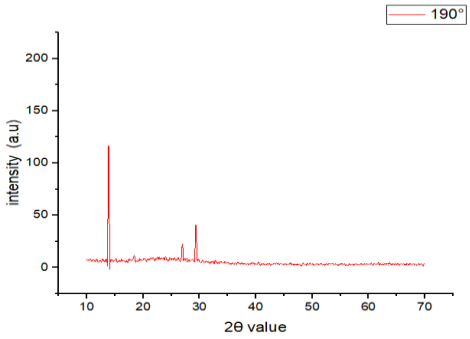

FIGURE 12: XRD result of the ZnO nanoparticles and perovskite materials annealed at 190˚C

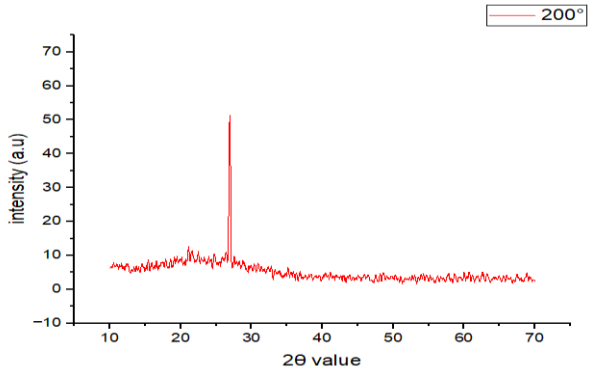

FIGURE 13: XRD result of the ZnO nanoparticles and perovskite materials annealed at 200˚C

CONCLUSION

The zinc oxide (ZnO) and perovskite material have been successfully deposited by hydrothermal bathing and spin coating methods at deposition time of 3 hours and annealed at dissimilar temperature of 100˚C, 130˚C, 160˚C, 180˚C and 200˚C. UV- Vis spectroscopy revealed that Zinc oxide (ZnO) and perovskite film annealed at 130˚C has the highest efficiency because it absorbs least at 3.38 a.u and transmits highly at 1.48 a.u within the visible region when compared to other annealing temperatures and these properties or characteristics approves the films good materials for fabrication of solar cells. Finally, it has been shown that hydrothermal bathing and spin coating methods offers good deposition technique that results in adequate film properties for use in solar energy device fabrication process and are inexpensive for thin film deposition.

REFERENCES

Akihiro Kojima (2009), Organometal halide perovskites as visible-light sensitizers for photovoltaic cells J. Am. Chem. Soc.

Babatunde, E.B and Uhuegbu, C.C. (2009): spectral selectivity characteristics of FeCuS thin film and its possible solar energy applications. Global jour. of Engg. And Tech. Vol. 2, 193 – 100.

Deependra D.M., Agni D., and Buddha R.S. (2014): effect of annealing on optical properties of Zinc Oxide thin films prepared by homemade spin coater. Nepal journal of Sci. and Tech. Vol. 15, No. 2, 111-116.

Ding, R., C.Xu, B. Gu, Shi Z., Wang H., L. Ba and Xiao Z. (2010): effect of Mg incorporation on microstructure and optical properties of ZnO thin films prepared by sol-gel method. J. mater.Sci.Technol. 26(7):601-604.

Electrical 4 u (2020); transmittance (formula and transmittance to absorbance calculation). https://www.electrical4u.com/what-is-transmittance

Feng Xian Xie, (2015) Vacuum-assisted thermal annealing of $CH_3NH_3PbI_3$ for highly stable and efficient perovskite solar cells ACS Nano.

Guichuan XingLong (2013), range balanced electron-and hole-transport lengths in organic-inorganic $CH_3NH_3PbI_3$Science.

Gumu, C., Ozkendir O.M., Kavak H., and Ufuktepe Y. (2006): structural and optical properties of Zinc Oxide thin films prepared by spray pyrolysis method.J. Optoelectronics and Advanced materials 8:299-303.

Henry J. Snaith (2013): Perovskites: the emergence of a new era for low-cost, high-efficiency solar cells J. Phys. Chem. Lett.

Khan, Z.R., Khan M.S., and Zulfequar M. (2011): Optical and structural properties of ZnO thin films fabricated by sol-gel method. Mat.Sc. and appl. 2: 340-345

Martin A. Green*; Anita Ho – Baillie; Henry j. Snaith,* (2014) The emergence of perovskite solar cells Nat. Photon.

Merriam – webster, incorporated (2023): Britannica.com: encyclopedia article about reflectance.

Molecular Devices (2022): what is absorbance? https://www.moleculardevices.com/technology /absorbance - detection -work.

Mousoi A.k.; Mehde M.S., Gebori, A.M. (2020): Annealing temperature effects on the performance of the perovskite solar cells. IOP Conf.Ser.: Mater.Sci.Eng.757 012039.

Peng Gao*; Michael Gratzel; Mohammed K. Naseeruddin, (2014)* Organohalide lead perovskites for photovoltaic applications Energy Environ. Sci.

Samuel D. Stranks (2013), Electron-hole diffusion lengths exceeding 1 micrometer in an organometal trihalide perovskite absorber Science. dio: 10.1126/science.124382.PMID:2413696

Solar Magazine (2022), Perovskite Solar Cells: An In-Depth Guide + Comparisons with Other Techs. https://solarmagazine.com/solar-panels/perovskite -solar-cells.

Uhuegbu, Chidi.C. (2010): solution growth technique for iron Copper Sulphide ternary thin film and its optical characteristics. American journal of scientific and industrial research.1(3):392 – 396.

Uhuegbu, C.C. and Babatunde E.B. (2012): some solid properties of FeCus thin film applicable in spectral selectivity application in solar energy. Canadian journal on scientific and industrial research Vol. 3 No. 2, 69 – 82.

Hanadi M., Asya M., Abdelaziz B., (2020): effect of annealing treatment on the properties of inverted solar cells based on mixed halide perovskite, Vol.119, http://www.science direct.com/science /article /pii/s1386947719317308.