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Development of Homogenous n-ZnO/TiO_2 Thin Film Dual-Layer Structure Utilizing the Spin-Coating Technique

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ABSTRACT

Titanium dioxide (TiO_2) and zinc oxide (ZnO) thin films have recently garnered attention as promising inorganic semiconductor materials in thin-film photovoltaic applications. Aside from a single layer, a combination of TiO_2 and ZnO layers has been studied since both materials have enormous potential in optical applications. This paper demonstrated deposition of n-ZnO/TiO_2 dual-layer thin films utilizing the sol-gel deposition via spin coating. One of the important steps for n-ZnO/TiO_2 dual-layer thin film to undergo the recrystallization process is the annealing treatment to produce a high-quality thin film. By varying different annealing temperatures, the optimal annealing setting for n-ZnO/TiO_2 dual-layer thin film was 500 °C for 2 hours. For structural properties, high intensity with preferred oriented n-(002)ZnO/(101)TiO_2 dual-layer thin film was achieved, showing anatase phase for TiO_2 and hexagonal wurtzite structure for ZnO. A compact, dense, and uniform surface morphology was observed for the n-ZnO/TiO_2 dual-layer thin film. The thickness was 266 nm for TiO_2 and 233 nm for the ZnO layer. The transmittance spectrum has also been achieved up to 70% at a wavelength of 400 nm. After the extrapolated plot, the band gap energy evaluated was 3.15 eV. In summary, the solar cell performance benefited from annealing treatment in refining the characteristics of the thin film.

Keywords: *n-ZnO/TiO_2 Dual-layer, Annealing temperature, Sol-gel deposition via spin coating, Homogeneous*

1. INTRODUCTION

Thin-film photovoltaic technologies are increasingly recognized as viable substitutes for traditional silicon-based solar cells. Their appeal lies in the reduced manufacturing costs and utilization of environmentally friendly materials. A range of absorptive layers, such as amorphous silicon (a-Si) [1], copper indium gallium selenide (CIGS) [2], cadmium telluride (CdTe) [3], and metal oxides [4], have been explored. Of these, metal oxide-based thin films stand out due to their non-toxic nature [5] and adaptable optical and chemical characteristics. In addition, these thin-film cells offer advantages such as ease of fabrication, efficient material deposition, and stable performance across varying temperature conditions [6]. As a result, this approach is gaining considerable interest as a viable option for photovoltaic solar cell applications.

Inorganic metal oxide semiconductors are widely utilized in thin film solar cells. Among materials that have been applied, titanium dioxide (TiO_2) has been widely researched due to its exceptional photocatalytic activity and optical behavior, making it suitable for applications in coatings, sensors, and catalysis [7]. Apart from its insensitivity to visible light while effectively absorbing near ultraviolet (UV) radiation, TiO_2 also possesses outstanding durability, a high refractive index, and a high dielectric constant. TiO_2 may be present in an amorphous structure or one of three crystalline configurations: tetragonal anatase, tetragonal rutile, or orthorhombic brookite. Among these phases, the anatase phase, despite being metastable, demonstrates exceptional optoelectronic properties [8].

Nevertheless, TiO_2 does have a disadvantage in its relatively high electron-hole recombination rate. Combining TiO_2 with other metal oxides to address this issue may help overcome the limitation [9]. The combination of n-type TiO_2 and n-type zinc oxide (ZnO) in bilayer structures has attracted increasing attention, primarily because both materials are inexpensive, readily available, and environmentally benign. Due to their comparable bandgap energies, around 3.3 eV, the integration of ZnO and TiO_2 in a dual-layer format results in favorable optical behavior and enhanced photoresponse under illumination [10]. ZnO is regarded as a promising transparent conductive oxide (TCO) for solar applications due to its thermal stability in hydrogen-rich environments and its capability to form at relatively low synthesis temperatures [11]. n-ZnO/TiO_2 bilayer thin films, which are widely applied as electron transport layers (ETLs) [12], aid in enhancing charge mobility while reducing recombination events [13], thereby increasing the overall efficiency of the device. Thus, the collaboration between TiO_2 and ZnO has been studied to optimize the crystallinity, electrical and optical properties of both materials.

Several fabrication methods have been explored for depositing n-ZnO/TiO_2 dual-layer thin films, including magnetron sputtering [14], chemical vapor deposition (CVD) [15], spray pyrolysis [16], atomic layer deposition (ALD) [17], and the sol-gel method [18]. When compared, the sol-gel spin-coating technique is found to have benefits such as low synthesis temperatures and cost-effectiveness [19]. It is also considered very practical in the formation of homogeneous and transparent thin films across different substrates [20]. In 2020, A. Timoumi et al. used the spin-coating technique to fabricate graphene oxide-titanium dioxide thin films on glass substrates. The films exhibited visible light transmittance of up to 80% with 3.71 eV for their optical band-gap energy, showing that spin coating can produce homogeneous thin films with high transmittance [21].

Various deposition parameters, such as the thickness of the acquired thin film, along with the spin speed [22] and annealing temperature [23] in the sol-gel spin coating method, may strongly affect the chemical, structural, and physical properties of the acquired thin film. Fundamentally, thin film materials are formed through random nucleation and growth processes, where condensing atomic or ionic species react on a substrate. Thermal annealing plays a crucial role in promoting recrystallization within the n-ZnO/TiO_2 dual-layer, leading to improved film quality through controlled grain growth and defect reduction [9]. In 2020, Lokesh K. S. et al. compared unannealed ZnO thin films with those that underwent annealing. Unannealed thin films often display amorphous structures with limited reproducibility, whereas those subjected to vacuum annealing tend to develop higher internal strain, mainly due to the formation of oxygen vacancies [24]. In addition, Y. Alaya et al., in 2023, demonstrated that TiO_2 thin films exhibited surfaces that were homogeneous and well-covered, with an increase in grain size as a result of annealing. Thus, it was proven that annealing enhances the overall thin film quality by reducing defects and increasing homogeneity [25].

This study employed sol-gel deposition by the spin coating method to deposit n-ZnO/TiO_2 dual-layer thin films onto fluorine-doped tin oxide (FTO) substrates. This research highlights how various annealing temperatures can affect the structural, morphological, and optical properties of the thin films. Different annealing temperatures were applied during the heating phase to determine the ideal parameters. The properties of n-ZnO/TiO_2 dual-layer thin films were characterised to achieve a high-quality crystalline thin film suitable for solar cell applications. By collaborating highly oriented n-(002)ZnO and (101)TiO_2 in a dual-layer film, significant improvements can be seen in its structural, morphological, and optical properties. By fine-tuning the heating process, the development of heterointerfaces within the thin film was enhanced, leading to an indirect improvement in photovoltaic performance through the efficient coupling of the TiO_2 and ZnO metal oxide semiconductors.

2. METHODOLOGY AND MATERIALS

Fabrication of the n-ZnO/TiO_2 dual-layer thin films was carried out using sol-gel spin-coating as the main deposition method. At first, TiO_2 and ZnO colloidal solutions were prepared using the sol-gel method. For the TiO_2 solution, the titanium (IV) butoxide precursor was combined with n-butanol and was agitated for 30 minutes. Then, a catalyst in the form of acetic acid was added dropwise into the precursor mixture. Deionized water was added next while stirring. Zinc acetate dehydrate, a precursor, was dissolved in isopropanol for the ZnO solution. Subsequently, diethanolamine (DEA) and deionized water were mixed in, and the milky suspension was stirred until it turned into a transparent solution. The prepared TiO_2 and ZnO sols were left to age at ambient temperature for 24 hours, which allowed them to become more viscous and fully homogenized. The FTO substrates underwent ultrasonic cleaning using acetone, ethanol, and distilled water to remove surface contaminants before the coating process.

The spin-coating process started with the deposition of TiO_2 film onto the pre-cleaned FTO substrates, followed by applying ZnO film. Each film was coated at 3000 revolutions per minute (rpm) for 30 seconds, and the cycle was repeated eight times to obtain the desired film thickness. After each layer was deposited, the substrate was dried on a hot plate at 100 °C for 2 minutes to facilitate solvent evaporation before the next coating cycle. Once the TiO_2 film was fully deposited, the films went through thermal annealing at 600 °C for one hour to improve crystallinity. Subsequently, the ZnO layer was coated onto the annealed TiO_2 film before a second annealing stage. The annealing step was conducted at varying temperatures using a closed-air furnace to determine the optimal annealing condition for the n-ZnO/TiO_2 dual-layer films.

A few characterization techniques were utilized to evaluate the characteristics of the n-ZnO/TiO_2 dual-layer thin films. X-ray Diffraction Spectroscopy was used to investigate the structural characterisation, with the 2θ range from 20 to 80° [Copper K-alpha (Cu Kα) radiation with a wavelength of 1.54 Å] for structural analysis. Field Emission Scanning Electron Microscopy (FE-SEM) was used to observe surface morphology at 50,000× magnification and an accelerating voltage of 15 kV. Meanwhile, optical properties evaluated from the transmittance spectra in the 300 to 800 nm wavelength range were obtained using the Ultraviolet-visible Spectroscopy. An overview of the fabrication steps for the n-ZnO/TiO_2 dual-layer thin film is shown in Figure

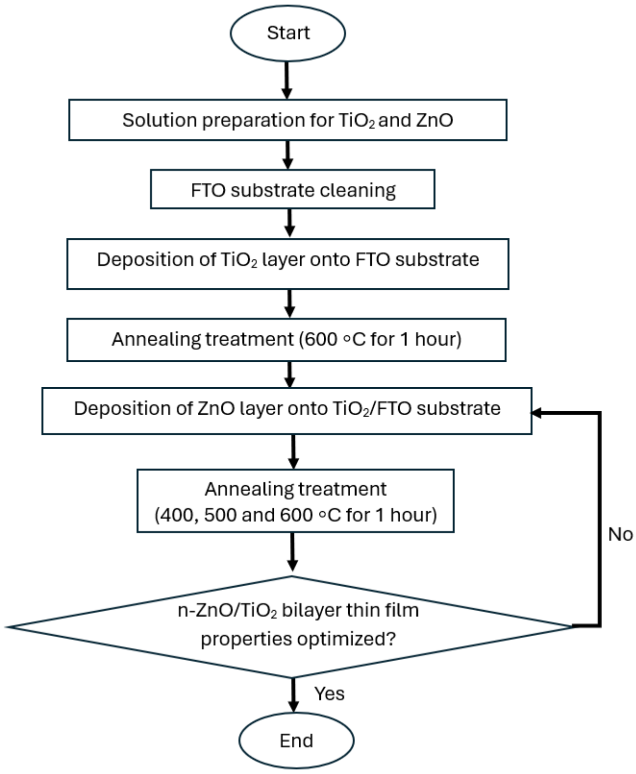

Figure 1. Flowchart of the fabrication of n-ZnO/TiO_2 dual-layer thin films.

3. RESULTS AND DISCUSSION

This section presents a detailed analysis of the n-ZnO/TiO_2 dual-layer thin film, focusing on its structural, morphological, and optical properties. The fabrication process includes depositing the n-TiO_2 film onto the FTO, followed by an annealing step to produce the n-TiO_2/FTO substrate. Consequently, a ZnO layer was coated onto the n-TiO_2/FTO to form a complete dual-layer structure. The resulting n-ZnO/TiO_2 films were then annealed at three different temperatures, 400, 500 and 600 °C, for a fixed duration of 2 hours, to evaluate the effect of annealing on the film's characteristics.

3.1. Structural Properties

X-ray diffraction (XRD) was used to study the structural characteristics of the fabricated n-ZnO/TiO_2 dual-layer thin films. The findings showed that after the deposition of TiO_2 onto the FTOsubstrate, the single layer of n-TiO_2/FTO consisted of 27% tin oxide (SnO_2) and 73% TiO_2. Two distinct TiO_2 peaks corresponded to the (101) and (200) orientation planes, which confirmed the anatase structure of the TiO_2 layer.

Upon annealing the ZnO layer at 400 °C, elemental analysis indicated that the n-ZnO/TiO_2 dual-layer thin film contained approximately 20% SnO_2, 42% TiO_2, and 38% ZnO. The XRD pattern revealed distinct peaks at 31.8°, 34.3°, 36.3°, 56.6°, 62.9°, and 68.0°, which correspond to the (100), (002), (101), (110), (103), and (112) planes of the ZnO crystal structure, respectively, confirming the presence of the hexagonal lattice which wurtzite phase. The results align with the hexagonal wurtzite phase, validated using Inorganic Crystal Structure Database (ICSD) file number 98-005-7450. Upon annealing at 500 °C, the thin film composition shifted to approximately 20% SnO_2, 33% TiO_2, and 47% ZnO. Additionally, the (002) ZnO diffraction peak at 36.3° became increasingly prominent as the annealing temperature rose from 400 °C to 500 °C, indicating enhanced crystallographic orientation. These results are consistent with findings reported by D. Muthee et al., who observed that elevated annealing temperatures promote improved particle crystallinity through the coalescence of smaller grains into larger ones [26].

After annealing at 600 °C, the thin film consisted of 18% SnO2, 14% TiO_2, 17% ZnO, and an additional composition of 51% zinc titanate (ZnTiO3). The intensity of the preferred (002) ZnO peak also decreased, and $ZnTiO_3$, an impurity, was detected. These findings are consistent with a previous study by Yang Li et al., which reported $ZnTiO_3$ formation at temperatures below 800 °C [27]. The decreased ZnO peak at (002) indicated that the ZnO crystal lattice was disturbed by the presence of $ZnTiO_3$. This phenomenon can weaken the material's structural stability and reduce its functional performance. This observation proves that the presence of impurities will disorganize the arrangement of atoms in the thin film, which will result in a reduction of crystallinity [28].

The Scherrer Equation (1) was used to find the crystallite size (D) of n-ZnO/TiO_2 dual-layer thin films, by using the data from the most intense peak corresponding to the (002) plane.

$$D = \frac{k\lambda}{\beta \cos \theta} \quad (1)$$

where K indicates shape factor (usually 0.9), λ represents the X-ray radiation with a wavelength of 1.54056 Å, θ represents the angle of diffraction (Bragg's law), and β represents the full width at half maximum (FWHM) for the particular diffraction peak. The n-ZnO/TiO_2 dual-layer thin film exhibited crystallite sizes of 21.03 nm, 26.77 nm, and 27.81 nm at annealing temperatures of 400, 500, and 600 °C, respectively. An increase in crystallite size was observed with rising annealing temperature, suggesting higher grain growth at higher annealing temperatures. The n-ZnO/TiO_2 dual-layer thin film annealed at 500 °C exhibited the largest crystallite size, suggesting a more uniform and well-aligned atomic arrangement of the film. The XRD analysis showed multiple narrow and well-defined diffraction peaks with low FWHM values, indicating a highly crystalline nanostructure formation. Additionally, the average lattice parameters of the n-ZnO/TiO_2 dual-layer thin film were calculated using Equation (2), which is based on the interplanar spacing formula for hexagonal crystal systems.

$$\frac{1}{{d_{hkl}}^2} = \frac{4}{3}\left[\frac{h^2+hk+k^2}{a^2}\right] + \frac{l^2}{c^2} \tag{2}$$

Where *hkl* represents the Miller indices of the crystal plane, $d_{hkl} = \lambda/(2 \sin \theta)$ is an offshoot from the angle of diffraction (Bragg's law) equation, λ representing the x-ray wavelength, and θ being the Bragg's diffraction angle in radians. It was found that the lattice parameters for the thin film were a = b = 3.2961 Å and c = 5.1234 Å. It is also important to note that the values of the lattice parameter were found to closely match the standard crystal lattice dimensions as recorded in the ICSD (98-016-9463) file, with the values of a = b = 3.2510 Å, and c = 5.2040 Å. Thus, the optimum annealing temperature was restricted at 500 °C since the thin film showed the highest preferred peak intensity, which indicated the highest crystallinity. The variation of crystallite size and lattice parameter is provided in Table 1. Meanwhile, Figure 2 presents the diffraction profile of n-ZnO/TiO_2 dual-layer thin films with various annealing temperatues.

Table 1 Size of the crystallites and crystal lattice parameters of n-ZnO/TiO_2 dual-layer thin films at various annealing temperatures

Annealing Temperature (ºC)	(002) plane orientation				Lattice parameter (Å)	
	Theta 2θ (°)	Intensity (%)	FWHM	Size of crystallite (nm)	a = b	c
400	34.4754	55.23	0.4133	21.03	3.1231	5.1217
500	34.4604	100.00	0.3247	26.77	3.2961	5.1234
600	34.4978	30.01	0.3125	27.81	3.1174	5.1183

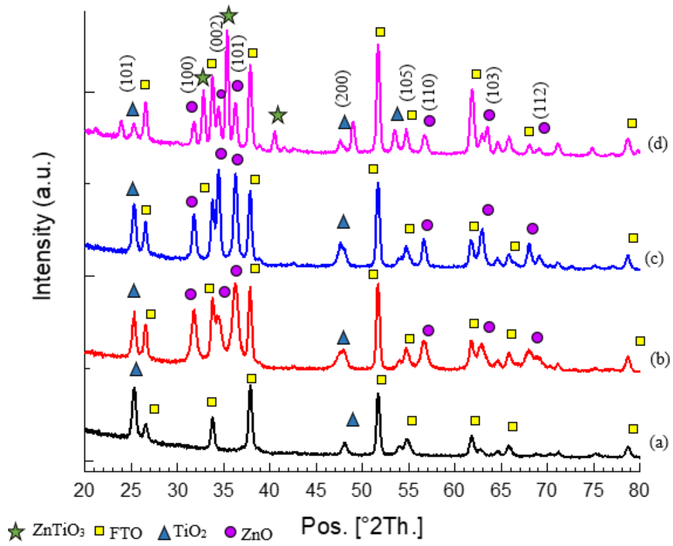

Figure 2. The diffraction profile of (a) n-TiO_2/FTO substrate annealing at a temperature of 600 °C and n-ZnO/TiO_2 dual-layer thin films annealing at temperatures of (b) 400 °C, (c) 500 °C, (d) 600 °C, respectively.

3.2. Morphological Properties

Morphological characterization was conducted using FE-SEM at a magnification of 50,000× to analyze the surface morphology of the n-ZnO/TiO_2 dual-layer thin films. The FE-SEM images revealed that the n-TiO_2/FTO substrate exhibited agglomerated fine-grained structures. Upon deposition of the ZnO layer, the resulting films grew homogeneously and thoroughly covered the underlying n-TiO_2/FTO surface. A noticeable increase in grain size was observed, which can be attributed to the presence of the additional ZnO layer. This observation aligns with previous studies, which reported that grain size increases with the number of deposited layers. This phenomenon is likely due to the interaction between the underlying and upper layers during the annealing process [29].

FE-SEM analysis of the n-ZnO/TiO_2 dual-layer thin film annealed at 400 °C revealed the presence of fine, closely packed grains on the surface. As the annealing temperature increased to 600 °C, a significant enlargement in grain size was observed. This trend aligns with the findings of F. Greuter et al., who reported that an increase in grain size corresponds to a more ordered atomic arrangement, resulting in fewer grain boundaries [30]. Subsequently, electrical properties will improve since atoms can easily move from grain to grain. However, some dark region among the grains was observed on the thin film annealed at 600 °C. The dark areas on the surface of the thin film indicated a less homogeneous and porous surface, as agreed by R. Hussin et. al. [19]. This film appeared more porous than others and exhibited lower surface density. When there are too many pores within the film, this will result in difficulty for carriers to travel between grains [31].

Since n-ZnO/TiO_2 dual-layer thin films that were annealed at 500 °C showed good agreement characteristics with previous structural properties, the thickness of the thin film has been evaluated. Cross-sectional FE-SEM images revealed distinct FTO, TiO_2, and ZnO layers, each exhibiting different thicknesses. The measured thicknesses of the TiO_2 and ZnO layers were approximately 266 nm and 233 nm, respectively. No visible defects, such as pores or irregularities in thickness, were observed at the interfaces, indicating that the films were uniformly deposited. This confirms the formation of a homogeneous dual-layer structure. The top-view images of the n-TiO_2/FTO substrate annealing at a temperature of 600 °C for 1 hour, along with the n-ZnO/TiO_2 dual-layer thin films annealing at various temperatures, are presented in Figure 3. Meanwhile, the cross-sectional image of the n-ZnO/TiO_2 dual-layer thin film annealed at 500 °C is shown in Figure 4.

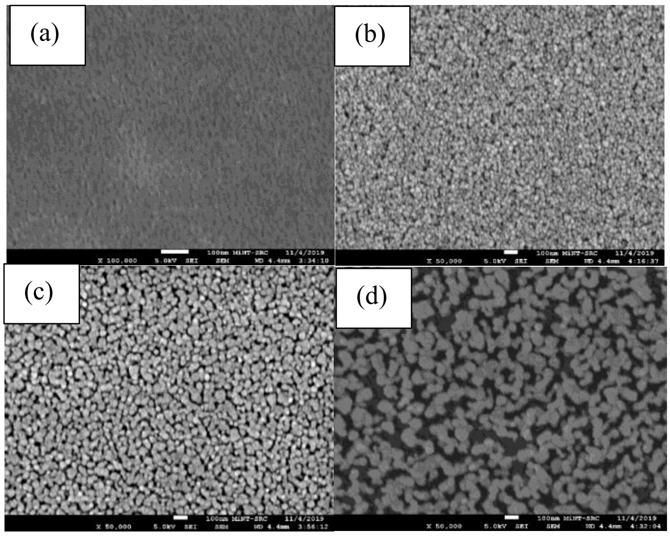

Figure 3. Morphological images of (a) n-TiO_2/FTO substrate followed by n-ZnO/TiO_2 dual-layer thin films processed by annealing at (b) 400 °C, (c) 500 °C, (d) 600 °C for 2 hours under magnification of 50,000x, respectively.

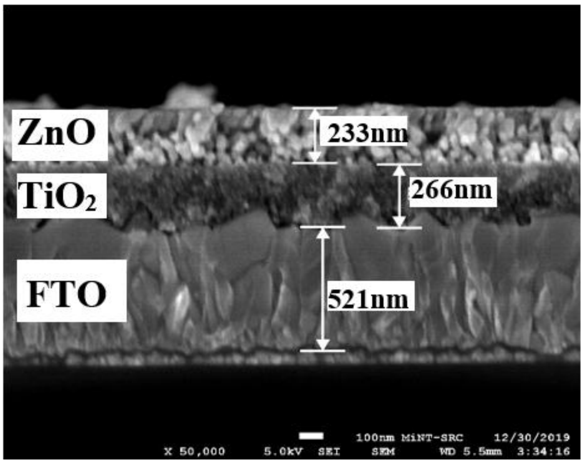

Figure 4. The cross-sectional diagram of n-ZnO/TiO_2 dual-layer thin films processed at an annealing temperature of 500 °C.

Homogeneous thin film refers to a thin material layer with a uniform composition and even thickness distribution across its entire surface. The thin film could also maintain consistent structural and optical properties without significant variations or defects [32]. From the results, it can be seen that the optimized n-ZnO/TiO_2 dual-layer thin films have consistent composition in each layer. The material composition remains the same throughout the film without any impurities. Impurities can cause non-uniform crystal growth, leading to inhomogeneous surface [33]. In addition, the optimized n-ZnO/TiO_2 dual-layer thin films have an even thickness distribution that can prevent the inhomogeneous surface of the thin film. Variation in thin film thickness in the entire layer can create localized recombination centres, which lead to a loss of charge carriers before contributing to current generation [34]. Thus, it will upset the performance of the photovoltaic mechanism in solar cell applications. This study's comparison with previous studies has been tabulated in Table 2.

Table 2 Previous studies on homogeneous TiO_2 and ZnO thin films for morphological properties

Thin Film Layer	Findings	Authors
TiO_2 and ZnO layer	The morphology of the TiO_2 porous layer was entirely covered by the perovskite film. The grain boundaries of the TiO_2 layer are clear and compact, indicating high crystallinity.	Yunfei Sun *et. al.* (2020) [35]
Carbon nanotube ZnO-TiO_2	Carbon nanotube ZnO-TiO_2 showed a thicker and smoother surface for the double oxide layers configuration. ZnO and TiO_2 nucleated as grains and coalesced to form a continuous layer.	László Péter Bakos *et al.* (2020) [17]
Nanocomposite ZnO-TiO_2-SnO_2/CeO_2	Nanocomposite ZnO-TiO_2-SnO_2/CeO_2 presented randomly distributed as bunch-like structures in spherical shape. The surface morphology appeared uniform in size.	H. Srinivasa Varaprasad *et. al.* (2021) [36]
Glass/TiO_2/ZnO (GTZ) bilayer films	The surface morphology of GTZ bilayer films significantly improved in the shape of spherical and ellipsoidal grains, proving that the crystal quality of the films is uniform.	Fariba Kheiri *et. al.* (2021) [37]
n-ZnO/TiO_2 dual-layer thin films	TiO_2 thin film grew homogeneously and was entirely covered by ZnO thin film. n-ZnO/TiO_2 dual-layer thin films have an even thickness distribution for each layer. The material's composition remains the same throughout the film without any impurities.	Nurliyana *et. al.*

3.3. Optical Properties

The potential transmittance of n-ZnO/TiO_2 dual-layer thin film is essential for the layer characterization. The transmittance spectra of the n-ZnO/TiO_2 dual-layer thin films were analyzed using ultraviolet-visible spectrophotometry. The results demonstrated that all samples, annealed at various temperatures, displayed transmittance within the visible wavelength range. Notably, the films annealed at 400 °C and 500 °C displayed fluctuating transmittance values, with peak transmittance reaching between 60% and 70%. In contrast, the film annealed at 600 °C displayed a significantly lower transmittance of approximately 45%. This reduction in optical transmittance is attributed to morphological changes, including increased porosity and a less compact surface structure [31]. Aside from the transmission of incident light between the layers becoming lower, it will affect the electrical properties due to higher series resistance based on previous studies by S. Hussein et. al. [38].

Furthermore, the optical band gap of the n-TiO_2/ZnO dual-layer thin film was determined by applying Tauc's Equation to determine the direct band gap of semiconductors, as presented below [39].

$$(\alpha h v)^{1/2} = A\,(h v - E_g)^{1/2} \quad (3)$$

In this context, α represents the absorption coefficient, h is Planck's constant, and ν denotes the photon frequency. Meanwhile, A and Eg indicate constant and optical band gap.By extrapolating the linear region of the Tauc plot of $(\alpha h\nu)^{1/2}$ vs (hν) for the n-ZnO/TiO_2 dual-layer thin film annealed at 500 °C, the optical band gap was estimated to be 3.15 eV. This value corresponds with the TiO_2 and ZnO layer band gap value as mentioned by P.L. Gareso et. al., which is 3.24 eV [40]. This value is similar and significant within the expected range for TiO_2 and ZnO thin film materials. This optimal band gap value facilitates efficient light absorption while enhancing the separation of photogenerated electron-hole pairs. As a result, the recombination rate is reduced, and the lifetimes of the charge carriers are prolonged—factors that are crucial for improving the performance of photovoltaic mechanisms [41]. Figure 5 illustrates the transmittance spectra of n-ZnO/TiO_2 dual-layer thin films annealed at different temperatures. The extrapolated Tauc plot for estimating the optical band gap of the dual-layer thin film processed with annealing at 500 °C is shown in Figure 6.

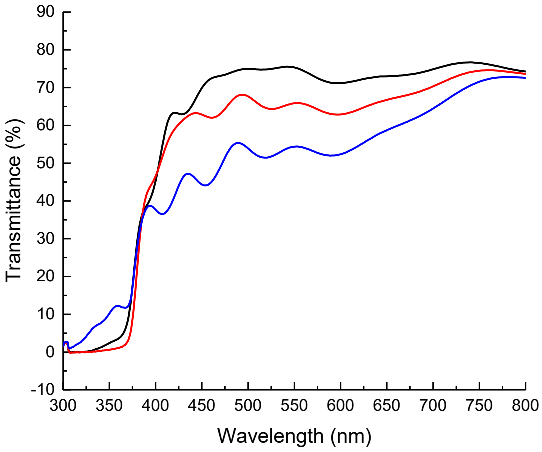

Figure 5. Spectra of transmittance for n-ZnO/TiO_2 dual-layer thin films with annealing temperatures of (a) 400 °C, (b) 500 °C, (c) 600 °C, respectively.

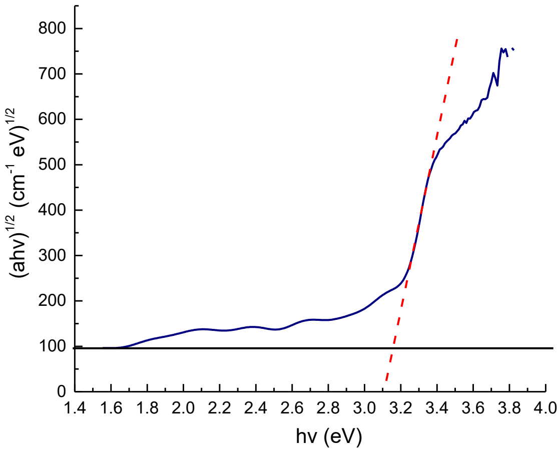

Figure 6. The band gap determined by extrapolation for n-ZnO/TiO_2 dual-layer thin film processed at an annealing temperature of 500 °C.

4. CONCLUSION

In conclusion, this study successfully demonstrated the fabrication of n-ZnO/TiO_2 dual-layer thin films utilizing the spin-on coating technique. The annealing temperature varied between 400 °C and 600 °C, with 500 °C as the optimal temperature. This value is chosen due to the properties of the dual-layer thin film. For structural properties, high intensity with preferred oriented n-(002)ZnO/(101)TiO_2 dual-layer thin film corresponded to the anatase and hexagonal wurtzite structure. Homogeneous n-ZnO/TiO_2 dual-layer thin film exhibited a dense and compact layer for morphological properties. The thickness was 266 nm for TiO_2 and 233 nm for the ZnO layer. A maximum transmittance of 70% was observed at a wavelength of 400 nm. After evaluating the extrapolated plot, the band gap energy obtained was 3.15 eV. These findings demonstrate that annealing substantially improved the characteristics of the n-ZnO/TiO_2 dual-layer thin film, making it a valuable advancement in the fabrication and development of thin-film solar cells.

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