THE EFFECT OF SPUTTERING TEMPERATURE OF TiO_2/ITO-PEN PHOTOANODES IN DYE SENSITIZED SOLAR CELL

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ABSTRACT

Currently, the world is facing a major crisis related to the lack of sustainable, safe, and environmentally friendly energy resources. Dye sensitized solar cells (DSSC), another name for third-generation solar cells, have gained a lot of interest due to ease of production, cheapness, and environmental friendliness. The photoanode is among DSSC's most crucial components. In this research, the active layer on the TiO_2 photoanode was optimized to improve the efficiency of the DSSC. The active layer was deposited using Radio Frequency (RF) magnetron sputtering on an Indium tin oxide-polyethylene naphthalate (ITO-PEN) substrate. The sputtering temperature was varied to 25, 80, 120, and 160°C for one hour. The thin film TiO_2/ITO-PEN photoanode will be characterized by means of X-ray Diffraction (XRD), Ultraviolet-Visible (UV-Vis) spectroscopy, and solar simulator. The XRD analysis shows that the best crystal size is 14.55 nm for a sputtering temperature of 80°C. According to the UV-Vis data, optical absorption increases with increasing sputtering temperature. The wavelength range where the absorption peak occurs is 252–465 nm, and the smallest value of the energy gap is found at a sputtering temperature of 25°C with a value of 3.02 eV. For the TiO_2/ITO-PEN thin layer, the maximum efficiency was achieved at 0.12% at a sputtering temperature of 25°C.

Keywords: DSSC; ITO-PEN; Photoanode; Sputtering; TiO_2

Introduction

Currently, the world is facing a major crisis related to the lack of sustainable, safe, and environmentally friendly energy resources. Renewable energy sources are growing in importance due to the rapidly expanding population and the depletion of fossil resources.[1] In order to meet the world of expanding energy needs, solar energy is now the most suitable renewable alternative energy source.[2,3] Dye-sensitized solar cells (DSSCs) have drawn a lot of interest in the last several decades due to their straightforward, affordable, and eco-friendly production process.[4,5]

DSSC is a relatively new, promising photovoltaic technology with several applications because of its straightforward manufacturing method and affordable production costs.[6] Photovoltaics uses technology to directly convert sunlight into electrical energy.[1] In 1991, Grätzel invented DSSC as an alternative to Si crystal solar cells and is highly recognized worldwide compared to the previous solar cell because it is more effective than its previous solar cell in low light conditions and has a potential efficiency of 33%.[7,8] In an effort to improve power conversion efficiency (PCE), research has been conducted ever since DSSC were discovered in 1991.[9] The important parts of DSSCs are a coated Transparent Conductive Oxide (TCO) glass film substrate, photoanode, dye (dye attached to the

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semiconductor), electrolyte, and counter electrode.[5,10]

Photoanodes have a substantial effect on DSSC efficiency because they can affect light scattering, dye loading, and electron transport.[11] Among various semiconductor materials with a wide band gap, the most extensively used semiconductor material is titanium dioxide (TiO_2) due to superior performance, affordability, plenty accessibility, stability and large surface area for loading dyes.[12–15] The photoanode is composed of a TCO substrate, which serves as a working electrode in a DSSC, and a metal oxide semiconductor material adsorbed with dye molecules coated on it.[16] The best TCOs that have been created in the last thirty years are those that are fluoride-doped tin oxide (FTO) and indium-doped tin oxide (ITO), respectively.[17] In comparison to ITO, DSSCs made on FTO glass substrates are more electrically conductible and have less optical transparency. They are also more brittle and stiff. Plastic-based flexible DSSCs, namely ITO-PEN, have drawn a lot of attention because glass-based DSSCs are rigid. ITO-PEN has superior electrical conductivity, low cost, lightweight, and high flexibility.[18–20] This is supported by research conducted by Lee et al. based on TiO_2 with ITO-PEN substrates using the electrospray method, resulting in a DSSC efficiency of 2.09%.[21]

Additionally, one of the factors that is crucial to the creation, functionality, and marketing of DSSC is the synthesis method.[22] The sputtering technique is recognized as a common industrial production method. Large-area thin film production frequently uses the deposited films, which typically have good adherence to the substrate and are highly controllable with respect to the deposition parameters.[23] The sputtering technique is a good substitute since it is easy to use, reproducible, and can make homogenous films by adjusting the porosity and particle size of the film.[22,24] This method is attracting great interest because it can create dense or porous films at comparatively low temperatures.[25] The damage to the interface caused by ion bombardment during the sputtering process, however, is the main disadvantage of this technique. These impacts should be reduced since they have a negative effect on the final thin film performance.[25]

TiO_2 material was used as a photoanode in research on FTO substrates by Low et al. at various sputtering temperatures.[26] With the drawback of using the FTO substrate and the benefit of the ITO-PEN substrate above, The synthesis of a thin film of TiO_2 as a photoanode utilizing the ITO-PEN substrate and varying the sputtering temperature is a novel application of this research. It is anticipated that altering the sputtering temperature will regulate the deposition process of the TiO_2/ITO-PEN thin layer and minimize ion bombardment damage. The deposited TiO_2/ITO-PEN thin films were examined by XRD, UV-Vis spectroscopy, and solar simulator.

Methods

Materials

The materials used include Ti-Nanoxide BL/SP (Solaronix), ITO-PEN substrate, ready to use Ti-Nanoxide T/SP (Solaronix), ready to use Ti-Nanoxide R/SP (Solaronix), Target TiO_2 (with purity 99.9%), Titanium (IV) (triethanolaminato) isopropoxide sulotion (TTEAIP, Sigma Aldrich), aseton (Merck), etanol (Merck, 99.8%), 2-proponal (Merck), dye N719, elektrolit (Mesolyte TDE-250, Solaronix), surlyn (Meltonix 1170-25 sealing), and platisol T (Solaronix).

Synthesis TiO_2/ITO-PEN

The TiO_2 photoanode on ITO-PEN was grown using the sputtering synthesis method with various sputtering temperatures. Making the TiO_2/ITO-PEN photoanode begins with cleaning the ITO-PEN substrate using soaking for 15 minutes in purified water and soap in an ultrasonic bath. Subsequently, the substrate was exposed to acetone at 80°C and allowed to dry naturally. The blocking layer, which uses TiO_2 Blocking Layer (BL), is the initial layer of the photoanode. The solution was applied to the ITO-PEN substrate using the spin coating method, spinning at 3000 rpm for one minute. The films were then dried for 15, 15, and 30 minutes at temperatures of 100, 150, and 200°C, respectively. The TiO_2 layer is the second layer of the photoanode. Using the screen printing process, TiO_2 paste was applied on top of the BL having an active area of 0.5×0.5 cm^2 and dried for 15, 15, and 30 minutes at 100, 150, and 200°C, respectively. Using the RF magnetron sputtering coating process, the TiO_2 film was deposited with different sputtering temperature variations, namely 25, 80, 120, and 160°C for one hour at 80 Watts of power, 25% air pressure, and 5 rpm rotation. The film was submerged in a solution mixture of 20 mL of 2-propanol and 150 µL of Titanium (IV) (triethanolaminato) isopropoxide solution for 30 minutes at 80°C. Finally, ethanol was used to clear the TiO_2 coating, and it was then allowed to dry at room temperature, as in previous research.[27]

In a dark chamber, the TiO_2 film was soaked in sensitizer for 24 hours. The sensitizer was 0.07 mM N719 dissolved in ethanol and centrifuged for three hours at 400 rpm as shown in Figure 1. An iodide electrolyte is injected after the TiO_2 layer has been wet. The photoanode and counter electrode are then put together using binding clips to create a sandwich structure. The sample of photoanode TiO_2 with varying sputtering temperatures 25, 80, 120, and 160 W is indexed as TiO_2-25, TiO_2-80, TiO_2-120 and TiO_2-160, respectively.

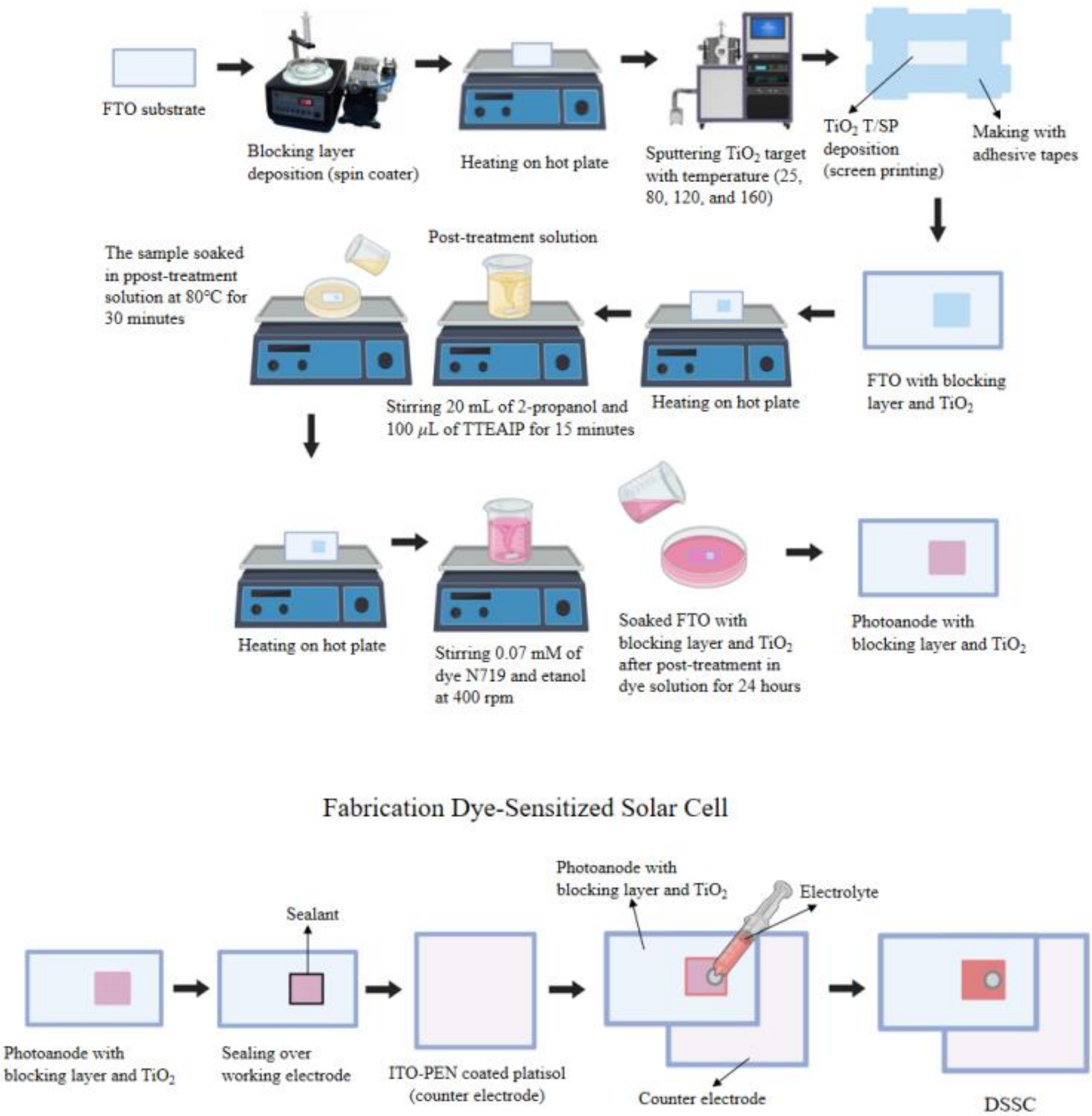

Figure 1. Synthesis of TiO_2 sputtering with variations in sputtering temperature

Characterization

The crystal structure of the TiO_2/ITO-PEN thin film was characterized using X-ray diffraction (XRD) type XRD E'Xpert Pro PANalytical at 2θ of 10−80° with $Cu_{k\alpha 1}$ = 0.15406 nm. Meanwhile, a Shimadzu UV-2600 spectrophotometer was used to examine the optical properties of the TiO_2/ITO-PEN thin film at a wavelength of 200−800 nm. The performance of the DSSC based on TiO_2 sputtering with variations in sputtering temperature was characterized using an I-V meter (Keithley 2400) under an illuminance of 100 mW/cm^2.

Result and Discussion

XRD

In this research, a thin film of TiO_2 has been effectively grown on an ITO-PEN substrate using the sputtering method conducted at various sputtering temperatures. XRD was used to evaluate the TiO_2/ITO-PEN thin film and ascertain its crystal structure. As can be seen in Figure 2, the XRD data produce a graph that illustrates the relationship between intensity and diffraction angle (2θ). The diffraction peaks indicate the crystal phase of TiO_2. The TiO_2 diffraction peaks are identified at 25.36°, 38.56°, 48.16°, 54.02°, 55.20°, and 63° with the hkl planes (101), (004), (200), (105), (211), and (204), and they exhibit the anatase phase.[28] The results of these TiO_2 peaks align with the research carried out by Bakr et al.[29]

Next, the Scherrer equation is used as a reference for calculating the crystal grain size of TiO_2, which is formulated with the equation (1).[30]

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

where D is the crystal size (nm), λ is the X-ray wavelength ($Cu_{k\alpha 1}$ = 0.15406 nm), k is the Scherrer constant (0.9), β is the FWHM (rad), and θ is the Bragg's angle (rad).[31] The crystal sizes obtained for each hkl plane (101), (004), and (200) are recorded in Table 1.

Table 1. Crystal size of TiO_2 with variations in sputtering temperature

Sample	2θ (°)	FWHM (rad)	*D* (nm)
TiO_2-25	25.83	0.55	14.82
TiO_2-80	25.71	0.51	15.98
TiO_2-120	25.79	0.56	14.55
TiO_2-160	25.78	0.42	19.40

In Table 1, it is shown that the crystal grain size increases with the rising sputtering temperature. At low temperatures, the nucleation site density is high, leading to the formation of small and dense crystallites. In contrast, at very high substrate temperatures, the nucleation density is lower and the surface diffusion of adatoms is greater, resulting in much larger crystallite sizes.[32]

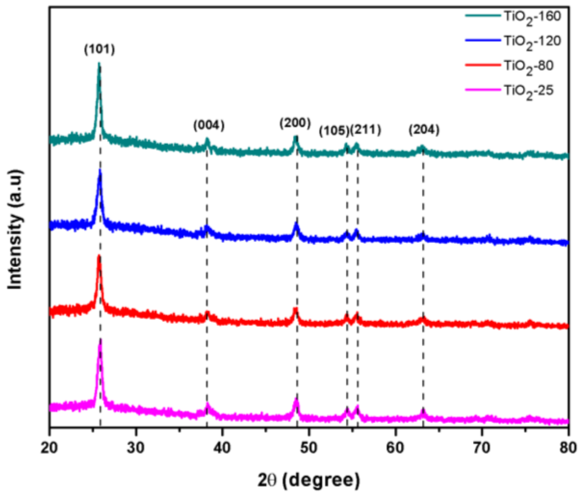

Figure 2. XRD pattern of TiO_2 with variations in sputtering temperature

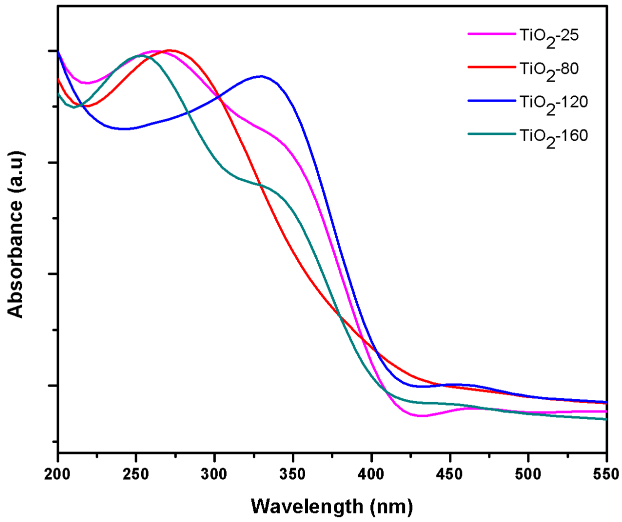

Figure 3. Spectrum absorbance of TiO_2 with variations in sputtering temperature

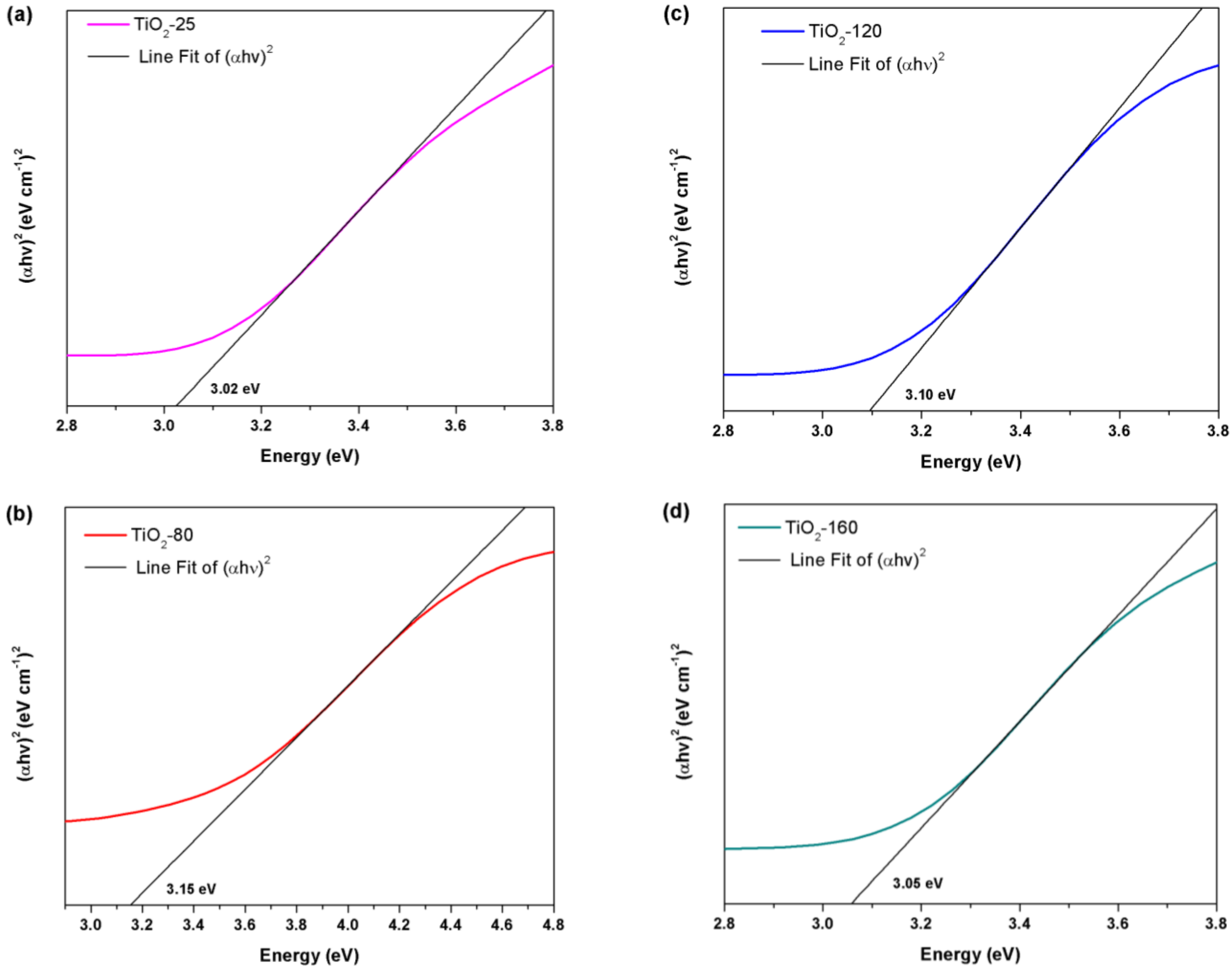

Figure 4. Band gap of TiO_2 with variations in sputtering temperature

Optical Properties Study

The optical properties and ultraviolet absorption of the TiO_2/ITO-PEN layer were studied in the wavelength range of 200−800 nm through UV-Vis characterization. The absorbance spectra of the TiO_2 layer at different sputtering temperatures is shown as Figure 3.

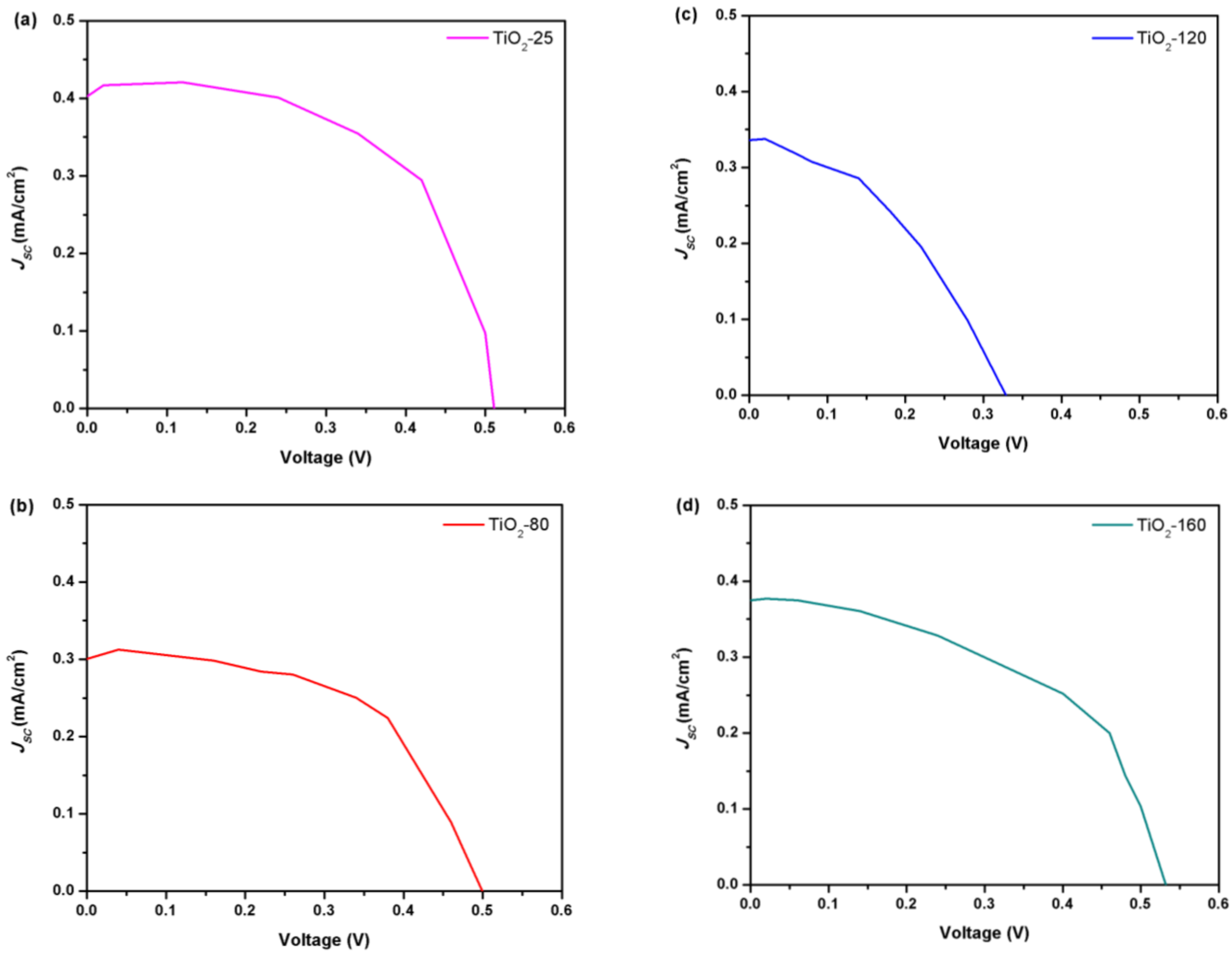

Figure 5. J-V characteristic of TiO_2 with variations in sputtering temperature

The maximum absorption area of the TiO_2/ITO-PEN layer shows a decrease with the increase in sputtering temperature. The wavelength ranges of 252–330 nm (UV region) and 455–465 nm (visible light region) contain the largest absorption peaks. It is evident that the temperature of growth affects the peak absorption.[33] Light scattering events cause the absorption window to increase and the absorption peak to shift to a longer wavelength, which in turn causes the optical band gap to diminish.[34,35] Higher light absorption will increase the number of electrons excited from the HOMO state to the LUMO state of the dye.[36] Consequently, the increase in the optical absorption coefficient will lead to an increase in the short-circuit current of the DSSC, and the efficiency of the DSSC will improve.[37]

The direct energy gap is obtained using the Tauc plot method through equation 2.[38]

$$(\alpha h v) = A(h v - Eg)^{1/2} \qquad (2)$$

Figure 4 shows the results for the direct band gap energy. The band gap of the TiO_2/ITO-PEN layer with varying sputtering temperatures shows energy gaps of 3.02, 3.15, 3.10, and 3.05 eV for TiO_2-25, TiO_2-80, TiO_2-120, and TiO_2-160, respectively. The band gap decreases when the sputtering temperature decreases. This indicates that the effect of optical confinement causes a shift in absorption to a narrower energy gap (blue shift). The particle size decreases as the width of this optical confinement narrows.[39] In this result, the smallest band gap is 3.02 eV with the sputtering temperature of 25℃. A small band gap allows for more efficient electron transfer, which can enhance photovoltaic performance.[40]

Performance DSSC Study

A common solar simulator system with an intensity of 100 mW/cm² is used to measure the current-voltage (J-V) characteristics of the

DSSC. With Pt serving as the counter electrode and a thin TiO_2 layer sensitized with N719 dye as the photoanode, DSSC is intended to create a sandwich structure. The J-V characteristics of the thin film TiO_2/ITO-PEN based DSSC with various sputtering temperature variations are shown in Figure 5.

Figure 5 shows that the lower the sputtering temperature, the higher the efficiency produced. From this research, the TiO_2-25 cell with a short-circuit current density (J_{SC}) of 0.40 mA/cm² has the highest efficiency of 0.12% among sputtering-based TiO_2 photoanode solar cells, as shown in Table 2. This power conversion efficiency was obtained through Equation 3.[41]

$$\eta = \frac{J_{SC} \times V_{OC} \times FF}{P_{in}} \quad (3)$$

With η being the overall efficiency, *Jsc* is the short-circuit current density, *FF* is the fill factor, *Pin* is the intensity of the incident light, and *Voc* is the open-circuit voltage.[41] The highest power and efficiency that the DSSC can generate will depend on the maximum current.

Table 2. The photovoltaic performance of TiO_2 with variations in sputtering temperature

Sample	J_{sc} (mA/cm²)	V_{oc} (V)	*FF*	η (%)
TiO_2-25	0,40	0,51	0,60	0,12
TiO_2-80	0,30	0,49	0,57	0,08
TiO_2-120	0,33	0,33	0,41	0,05
TiO_2-160	0,37	0,49	0,55	0,10

The increase in short-circuit current density (J_{SC}) is the primary reason for the improvement in efficiency. The current created is dictated by the number of photoelectrons from the dye molecules. Naturally, additional dye molecules will result in the production of more photoelectrons. The surface area and surface shape of the TiO_2 sample can easily affect the amount of dye molecules adsorbed on TiO_2.[42] Samples with the finest particle size provide the largest surface area at the TiO_2/N719 dye interface for the highest amount of dye that can be absorbed.[33] The increase in *Jsc* at higher sputtering temperatures should be regarded as an enhancement in electron transport characteristics; that is, TiO_2 becomes more conductive at higher sputtering temperatures and its rate of recombination for optically generated traveling electrons in the dye material decreases.[43]

Conclusion

In summary on ITO-PEN substrates, TiO_2 thin films have been effectively deposited by sputtering at different sputtering temperatures of 25, 80, 120, and 160 °C. The XRD results show that the obtained crystal grain size is 14.55 nm. The energy gap of the TiO_2/ITO-PEN thin film is in the region of 3.02–2.15 eV. The wavelength region of 252–465 nm is where the highest absorbance is found, and it tends to increase as the sputtering temperature increases. The best photovoltaic performance of DSSC is achieved at the lowest sputtering temperature of 25 °C, with a conversion efficiency of 0.12%. A low sputtering temperature enhances the efficiency because of decreasing crystal grain sizes and band gaps.

Acknowledgment

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