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Development of $ZnO/SiO_2/Si$ Thin Films for Photothermoelectric Applications: Synthesis and Characterization

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Abstract: Photothermoelectric (PTE) conversion integrates photothermal and thermoelectric effects, where light irradiation generates a temperature gradient that induces voltage through the Seebeck effect. This study examines the influence of annealing treatment on the structural, morphological, and optical properties of $ZnO/SiO_2/Si$ thin films for PTE applications. ZnO films were synthesized on SiO_2/Si substrates modified via a hydrothermal process, with samples prepared under two conditions: without annealing and with annealing at 700, 1000, and 1100°C. X-ray diffraction (XRD) analysis showed that the unannealed sample had low crystallinity and small grain size, while annealed samples exhibited improved crystallinity and grain growth consistent with the wurtzite ZnO phase. Scanning electron microscopy (SEM-EDX) revealed that annealing transformed irregular, porous structures into more compact and uniform grains, with elemental composition approaching the stoichiometric Zn:O ratio. UV–Vis spectroscopy indicated that annealing broadened the absorption range (330–1100 nm) and reduced defect-related absorption. The enhanced crystallinity and optical absorption imply improved photothermal and thermoelectric potential. Overall, optimizing annealing temperature effectively enhances the light–heat–electricity conversion capability of $ZnO/SiO_2/Si$ thin films for photothermoelectric device applications.

Keywords: Annealing; Hydrothermal; Photothermoelectric; Solar cell efficiency; ZnO

Introduction

The sun is a renewable energy source that can be utilized both as heat and through direct conversion into electrical energy (Rifky et al., 2023). The direct conversion of solar energy into electricity is carried out by photovoltaics or solar cells (Hiendro & Suryadi, 2019; Busthomy & Widyartono, 2020), in which the movement of electrons between the positive and negative electrodes generates an electric current that can be used to power electronic devices. According to the Indonesia Solar Outlook 2023 report published by IESR, solar energy will play a crucial role in deep decarbonization in Indonesia by 2060 or earlier by 2050, with at least 88% of the installed capacity in 2050 expected to come from solar energy (Yanel & Saferi, 2023). In addition to light, solar radiation that reaches the earth's surface also carries thermal energy. Therefore, a fundamental question arises regarding how to utilize not only solar light but also its heat (Dewi et al., 2016). One possible approach is the application of photothermoelectric (PTE) technology.

Photothermoelectric (PTE) technology integrates multiple energy conversion mechanisms photothermal (PT) and thermoelectric (TE) to harvest solae energy more efficiently (Jin et al., 2022; Tee et al., 2022). In this

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hybrid concept, incident photons are converted into heat via the photothermal effect, which subsequently induces a temperature gradient across thermoelectric materials. This gradient drive charge carriers through the Seebeck effect, resulting in electrical power generation (Ananda et al., 2020). Thus, instead of being lost as waste heat, thermal energy from solar radiation is also converted into electricity, enhancing the overall energy conversion efficiency of solar systems (Zheng et al., 2024).

Previous studies Have Reported That Modifying ZnO-based materials can enhance their thermoelectric and optical properties. Rehman et al. (2020) and (Pradana & Widyartono (2020) compared the thermoelectric properties of ZnO and ZTO thin films grown on Si substrates, showing that ZTO exhibited higher Seebeck coefficients and power factors due to Sn incorporation in the ZnO lattice. Similarly, Zhang et al. (2024) demonstrated that $ZnO/SrTiO_3$ composites showed temperature-dependent increases in Seebeck coeffcients and infrared emissivity, confirming that thermal and optical behavior are strongly correlated. Moreover, dopant engineering such as Sb and Ni doping has been shown to improve both the photovoltaic and thermoelectric performanceof ZnO thin films, with Ni-doped ZnO exhibiting up to 13-fold improvement in thermoelectric output and 10-fold higher solar cell efficiency compared to undoped ZnO (Üzar et al., 2024).

Building on these findings, this research focuses on developing $ZnO/SiO_2/Si$ thin films as photothermoelectric (PTE) materials. The structure consists of a ZnO layer deposited on a SiO_2/Si substrate (Purbayanto et al., 2017; Jamaludin & Subramani, 2019 Arulraj, 2025). The novelty of this study lies in the integration of ZnO with modified SiO_2/Si substrates to explore how substrate modification influences light absorption, heat transfer, and charge carrier behavior in a PTE system. This combination is expected to simultaneously utilize solar light and heat, leading to improved energy conversion efficiency compared to conventional single-effect systems.

Therefore, this study utilizes $ZnO/SiO_2/Si$ thin film structures consisting of three layers, where a zinc oxide (ZnO) film is deposited on a SiO_2/Si substrate (Purbayanto et al., 2017). The development of $ZnO/SiO_2/Si$ thin films as photothermoelectric (PTE) materials is expected to exploit both solar light and thermal energy for conversion into electrical energy. The main focus of this research is to investigate the energy conversion mechanism in $ZnO/SiO_2/Si$ thin films for PTE applications. The methods employed include annealing and hydrothermal processes, as both techniques allow optimal control of thin film thickness and quality. In addition, this study aims to analyze the structure, optical properties, and morphology of the thin films to understand the influence of SiO_2/Si substrate modification on the photothermoelectric performance of $ZnO/SiO_2/Si$ thin films.

Method

This research was conducted using an experimental method to synthesis ZnO thin films on modified SiO_2/Si substrate. The synthesis procedure consisted of Si substrate preparation, substrate washing, substrate annealing at 700, 1000, and 1100, preparation of ZnO solution, and ZnO growth using the hydrothermal method. The synthesis results were then characterized using X-Ray Diffraction (XRD) to analyze crystal structure, Scanning Electron Microscope-Energy Dispersive X-Ray (SEM-EDX) to observe morphology and elemental composition, and Current-Voltage (I-V) measurements to determine electrical performance and photothermoelectric efficiency.

Materials

In this research, the materials used include silicon (Si), Zinc nitrate tetrahydrate ($Zn(NO_3)_2$ $4H_2O$), Hexamethylenetetramine ($C_6H_{12}N_4$), DI water, acetone, ethanol, and distilled water.

Research Procedure

The stages of the synthesis of SiO_2/Si subtrate procedure in this study are as follows:

Si Substrate Preparation

In silicon substrate preparation is done through substrate cutting. The Si substrate was cut to a size of 1 cm x 1 cm and 1.5 cm x 2.5 cm. using a cutting pen. Furthermore, the silicon pieces are stored in a Petri dish.

Substrate Washing

Substrate washing is done by putting the silicone into a glass beaker with ethanol solution for 3 minutes, then the silicone is transferred to a glass beaker in acetone solution for 3 minutes, and the silicone is transferred to a glass beaker in the solution using distilled water. Furthermore, the silicone is dried using tissue so that there is no liquid and dirt attached. The dried substrate was stored in a Petri dish.

Substrate Annealing Process

Substrate annealing is done by inserting silicon into a furnace tool with several temperature variations of 700, 1000, and 1100 with a hold of 30 minutes.

Preparation of ZnO Solution

Preparation of ZnO solution is done by weighing Zinc nitrate tetrahydrate ($Zn(NO_3)_2$ $4H_2O$) 2.61 grams and Hexamethylenetetramine ($C_6H_{12}N_4$) 1.4 grams, then mix into a glass beaker with 100 ml DI water closed using plastic warp. Then stirred using a magnetic stirrer at 600 rpm for 15 minutes.

ZnO Growth Using Hydrothermal Method

Prepare a substrate that has been anneled with several samples with a size of 1.5 cm x 2.5 cm given heat insulation. Then put the Si substrate into a glass beaker containing ZnO solution, cover using aluminum foil. Then heat the oven with a temperature of 100 °C for 10 minutes, after that put the glass jar containing the solution and the substrate into the oven with a temperature of 100 °C for 30 minutes. Then drying using a hotplate with a temperature of 250 °C for 15 minutes, put the substrate into a Petri dish.

Characterization

The synthesis results were then characterized using several techniques to analyze the $ZnO/SiO_2/Si$ thin films in this study.

X-Ray Diffraction (XRD)

X-ray diffraction is an effective method for analyzing the crystal structure of a material. This method is used to recognize the chemical composition in a sample (Rani, 2022). The basic principle of XRD used is diffracting light through the crystal gap. Light diffraction by crystals can occur when the wavelength is comparable to the interatomic distance, which is about 1 Angstrom (Febriliani et al., 2025). XRD uses x-ray radiation, electrons or neutrons. X-rays are photons with high energy having a wavelength of 0.5 to 2.5 Angstrom. When X-rays hit the material, some are absorbed, transmitted and partially scattered. This scattering will be detected by the XRD tool. There are scattered X-ray beams that eliminate each other because the phases are different and some reinforce each other because the phases are the same. X-ray beams that reinforce each other are referred to as diffraction beams (Hakim et al., 2019).

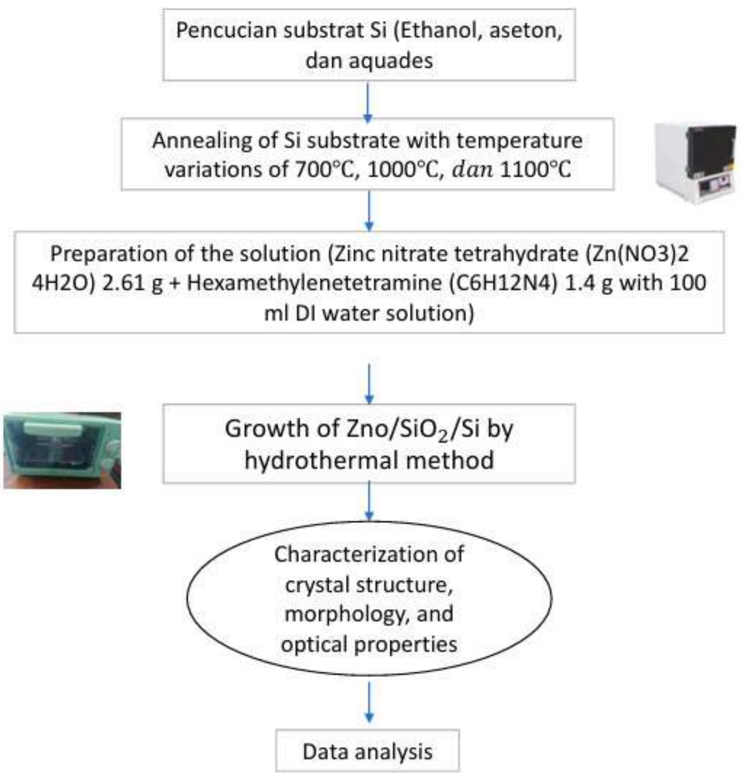

Figure 1. Flow diagram

Scanning Electron Microscope-Energy Dispersive X-Ray Spectroscopy (SEM-EDX)

SEM (Scanning Electron Microscope) - EDX (Energy Dispersive X-Ray Spectroscopy) is a method used to observe surface morphology, structure, and elemental distribution on various types of samples, such as natural, organic and inorganic materials, polymers, metals, and biological samples (Sahdiah & Kurniawan, 2023). This method uses an electron beam as a source with a wavelength tens of thousands of times shorter than the wavelength of visible light, resulting in images with much better resolution and image detail than optical microscopes (Masta, 2020). The working principle of the SEM tool is to utilize the scattering of electron beams on the surface of the sample and detect electrons that appear on the surface of the object (Septiano et al., 2021).

UV-Vis (Ultra Violet-Visible) Spectrophotometer

UV-Vis spectrophotometer is a device used to measure the wavelength and intensity of ultraviolet light and visible light absorbed by a sample. Ultra violet light and visible light contain enough energy to lift electrons from the outer shell of the atom to a higher energy level (Manurung et al., 2021). Ultra violet light is at a wavelength of 200-400 nm, while visible light has a wavelength of 400-800 nm (Suhartati, 2017). UV-Vis spectrophotometer is used to examine the nature of absorption (absorbance) and light transmission (transmittance) with samples at specific wavelengths (Cai & Guo, 2019).

Current Voltage (I-V)

I-V characterization is used to evaluate the performance of solar cells by measuring the relationship between electric current and voltage. I-V tests are carried out by giving light to the $ZnO/SiO_2/Si$ thin layer, then measuring the electric current and obtaining a current with voltage characterization curve (Andari, 2017).

Result and Discussion

The results of the synthesis and characterization process are as follows:

Crystal Structure of $ZnO/SiO_2/Si$

The results of XRD data of $ZnO/SiO_2/Si$ thin films with variations without annealing temperature and annealing temperature at 700 1000, and 1100℃ are plotted in Figure 2.

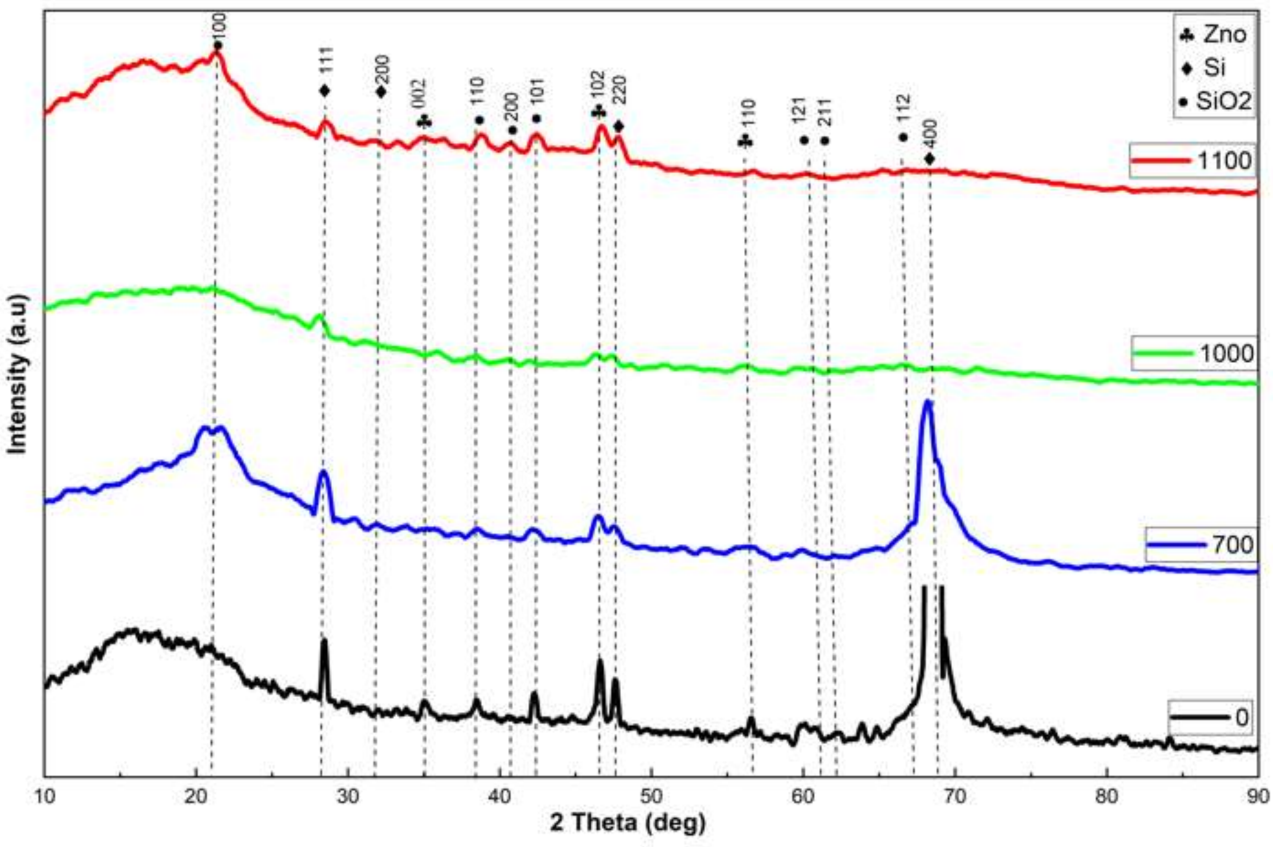

Figure 2. Diffraction peaks of $ZnO/SiO_2/Si$ without annealing temperature and annealing temperature (700, 1000, and 1100℃)

Table 1. XRD pattern results of $ZnO/SiO_2/Si$

Sample	2θ (°)	HKL	FWHM	Crystal Size (nm)
$ZnO/SiO_2/Si$-	35.32	002	0.39	
Without annealing	47.64	102	0.19	34.49
treatment	56.59	110	0.23	
	35.23	002	0.63	
$ZnO/SiO_2/Si$-700 ℃	47.54	102	0.63	12.82
	56.45	110	0.78	
	36.17	002	0.63	
$ZnO/SiO_2/Si$-1000 ℃	47.54	102	0.63	13.78
	56.34	110	0.63	
	36.14	002	0.47	
$ZnO/SiO_2/Si$-1100 ℃	47.87	102	0.41	18.34
	56.81	110	0.55	

Figure 2 All $ZnO/SiO_2/Si$ samples show ZnO peaks in the range of 2θ = 35.32 °, 47.64 °, 56.59 ° corresponding to diffraction 002, 102, and 110 having a hexagonal wurtzite crystal structure with a dominant peak in the 002 plane (Kareem et al., 2020). At 700 ℃ there is an increase in FWHM value and a decrease in crystal size, as well as a shift in the 002 peak to a smaller angle. Conversely, at higher temperatures (1000-1100 ℃), the FWHM value decreases and the crystal size increases and the peak shifts to a larger angle (Wagh et al., 2022). This phenomenon is in line with previous reports that annealing temperature can improve crystallinity, reduce lattice defects, and affect lattice parameters where the optimum temperature provides good crystal quality, while too high a temperature can cause morphological changes (Thi et al., 2020).

Morphology of $ZnO/SiO_2/Si$

SEM characterization was performed to identify the structure of the $ZnO/SiO_2/Si$ layer on the SiO_2/Si substrate. Figure 3 shows the morphological changes in the $ZnO/SiO_2/Si$ thin layer synthesized using the hydrothermal method without and with annealing process. The surface in Figure 3 (a) of the sample without annealing treatment shows small particles and agglomeration which indicates low crystallinity (Ade et al., 2021). After annealing at 700 ℃ (Figure 3 (b), the particle shape is clear, well dispersed, and high crystallinity (Khan et al., 2023). At 1000 ℃ Figure 3 (c), a hollow structure is formed due to the coalescence process and when the temperature gets higher at 1100 ℃ Figure 3 (d), the particle size merges into large grains. The addition of annealing temperature affects particle size, increases crystallinity, and particle shape, but at high temperatures it can reduce morphological quality for photothermoelectric applications.

Porosity measurement is carried out to determine the percentage of the volume of empty space contained in the sample. The results of porosity testing on $ZnO/SiO_2/Si$ thin films synthesized using the hydrothermal method with variations without annealing treatment and with variations in annealing temperatures of 700 ℃, 1000 ℃, and 1100 ℃, are shown in Table 2.

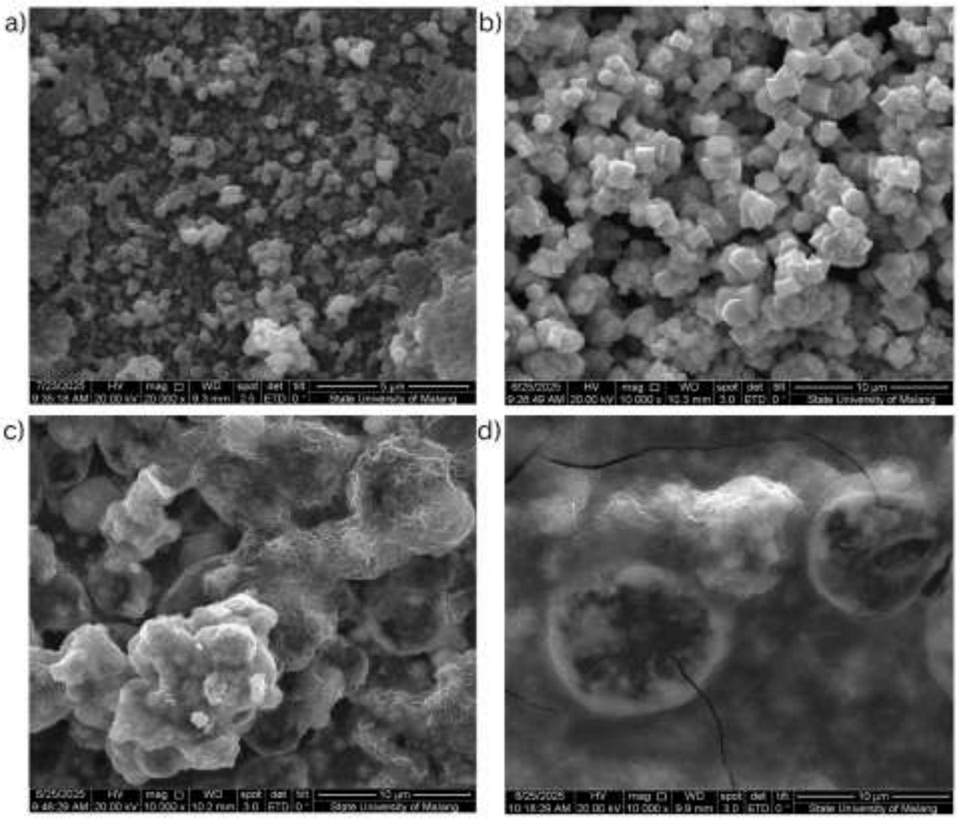

Figure 3. Scanning Electron Microscope (SEM) results on $ZnO/SiO_2/Si$ (a) without annealing treatment, (b) 700℃, (b) 1100 ℃, and (c) 1000℃

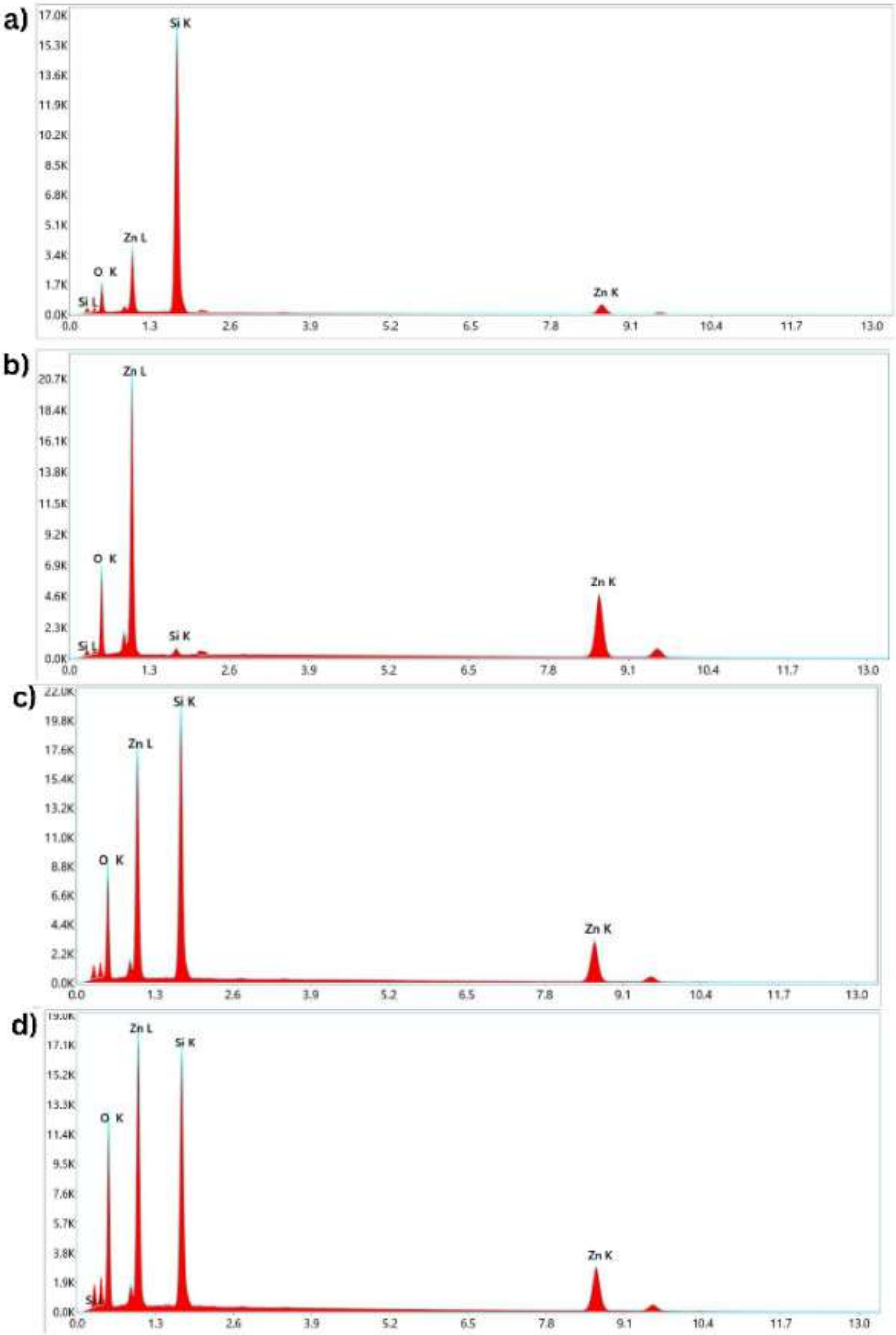

Figure 4. EDX results on $ZnO/SiO_2/Si$ (a) without annealing treatment, (b) 700°C, (c) 1000°C, and (d) 1100°C

Table 2. Porosity results on $ZnO/SiO_2/Si$

Sample	Porosity (%)
Without annealing treatment	71.26
700 °C	60.12
1000 °C	65.23
1100 °C	70.68

Table 2 shows that annealing treatment at 700°C leads to a decrease in porosity values caused by the densification and crystal grain growth process, where the ZnO particles form a denser and more homogeneous layer. This structure allows heat transfer and reduces thermal resistance between layers. In contrast, without annealing treatment, the porosity value is high, forming a layer structure that has not yet experienced density, while at temperatures of 1000 and 1100 °C the porosity value is still high, increasing due to the release of oxygen and structural degradation (microstructure damage). This is in accordance with the findings of Liang et al. (2020) who explained that high temperature annealing can trigger excessive crystal growth and increase the porosity value due to oxygen loss from ZnO particles. A similar study on ZnO synthesized using CVD method on Al substrate mentioned that ZnO coating synthesized at 400°C has the lowest thermal resistance with smooth surface, thus supporting more stable thermal performance (Liang et al., 2020).

Table 3. Atomic properties of $ZnO/SiO_2/Si$

Sample	Zn (%)	O (%)	Si (%)
Without annealing treatment	9.33	34.47	56.20
700 ℃	43.52	54.77	1.10
1000 ℃	18.78	50.19	31.03
1100 ℃	15.91	60.98	23.10

The results of EDX analysis in Figure 4 show the presence of Zn, Si, and O. The elements contained in $ZnO/SiO_2/Si$ with variations without annealing treatment and annealing variations of 70, 1000, and 1100 ℃ are seen in Table 3.

Optical Properties of ZnO/SiO₂/Si

Measurement of absorbance of $ZnO/SiO_2/Si$ thin films with variations without annealing temperature, 700, 1000, and 1100 ℃ is in the wavelength range of 330-1100 nm. The results of absorbance in $ZnO/SiO_2/Si$ thin films show that annealing treatment with temperature variations has an influence on the material in absorbing light. This phenomenon indicates a combination of optimal light trapping from the surface structure so that the possibility of the formation of impurity energy (defect states) in the band gap and allows the absorption of photons with lower energy than the material band gap. In the $ZnO/SiO_2/Si$ layer, nanostructures are formed by hydrothermal methods as light absorption, while the annealing process can optimize the surface structure, improve crystallinity, and generate defect states. So, this mechanism is in accordance with the behavior of black silicon (b-Si) which has absorption in the broad spectral range of surface nanostructures and defect states (Zhou et al., 2024).

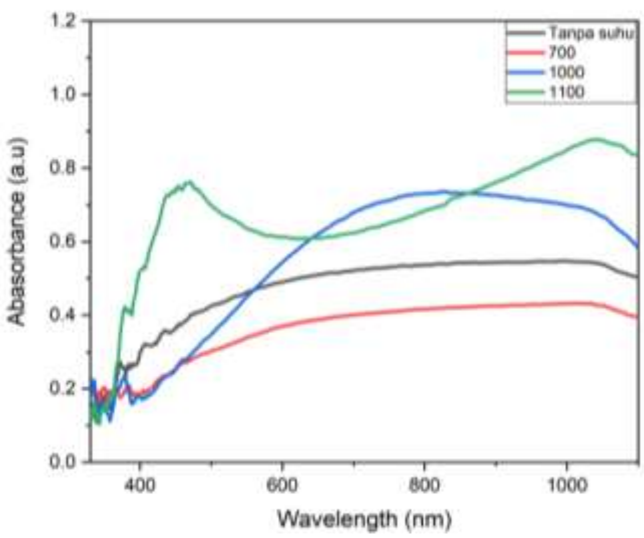

Figure 5. Absorbance results on $ZnO/SiO_2/Si$

EDX analysis in this study shows that at 700°C annealing, the O content is high (54.77%), and Zn is relatively low (43.52%), while Si is very low (1.71%). The Zn:O ratio = 0.80, so that at 700 ℃ annealing temperature close to the ideal Zn:O stoichiometric ratio variation of 1:1 indicates a lack of zinc vacancies that act as acceptors and contribute to the formation of defect levels that cause additional absorption in the visible region. In contrast, at 1100 ℃ annealing, the O content increased to 60.98% and Zn decreased to 15.91%, indicating reduced oxygen vacancies due to oxygenation and increased zinc vacancy formation. This condition is consistent with Das et al. who stated that annealing treatment in an oxygen-rich atmosphere can replenish oxygen vacuoles while increasing the amount of zinc vacuoles, thereby changing the optical properties through decreased n-type conductivity and increased defect-induced sub-bandgap absorption (Das et al., 2021).

This change in defect concentration affects the optical and electrical properties of the material. Oxygen vacancies can improve conductivity and optical response, but too much will increase the non-radiative recombination rate. Zinc vacancies tend to have fewer electrons, so the electricity generated is smaller and the performance drops. Zhou et al. (2024) emphasized that too low annealing temperature does not sufficiently improve crystallinity, while too high temperature can reduce the number of useful donor defects. Therefore, an optimum annealing temperature is needed that is able to balance crystallinity and the number of defects to maximize photothermoelectric performance (Zhou et al., 2024).

Efficiency of ZnO/SiO₂/Si

The efficiency of $ZnO/SiO_2/Si$ thin-film solar cells is calculated based on the J-V curve measurements under bright lighting conditions and temperature variations, as shown in Figures 6-7. The measurements were conducted using a solar simulator with a light intensity of 100 $mW.Cm^{-2}$. In Figure 5, the sample from I-V test to I-V+temperature test experienced an increase in efficiency value. The increase is due to the increase in annealing temperature (Iwantono et al., 2016) and the addition of temperature to the I-V characterization. The higher the annealing temperature, the ZnO crystal size increases and the morphology becomes denser, so that the resistivity decreases, the conductivity increases, and the number of ionized electrons increases which results in higher efficiency (Iwantono et al., 2016). Increasing the temperature before I-V testing by simulating hot conditions during the day shows that the electrical performance is improved through crystallinity improvement and defect reduction. Heating at the optimum temperature triggers desorption of molecules on the ZnO surface. Too high a temperature can trigger structural degradation and excessive grain growth, thereby increasing the electrical resistance and decreasing the electrical performance of $ZnO/SiO_2/Si$ thin films (Khalisha et al., 2025).

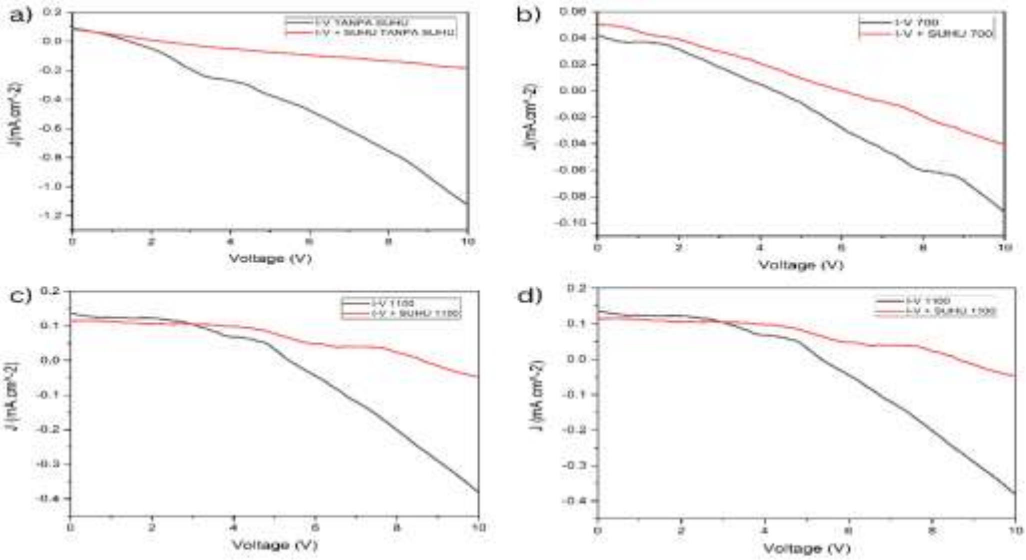

Figure 6. I-V and I-V+temperature results of $ZnO/SiO_2/Si$ (a) Without annealing temperature, (b) 700 °C, (c) 1000 °C, and (d) 1100 °C

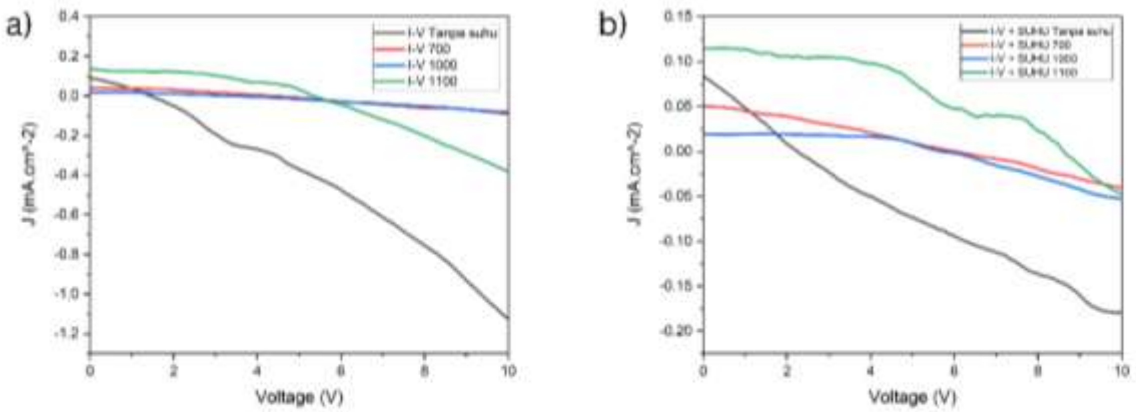

Figure 7. a) I-V results of all samples on $ZnO/SiO_2/Si$ and b) I-V+temperature results of all samples on $ZnO/SiO_2/Si$

Table 4. I-V and I-V + Temperature Parameter Data of Annealing Temperature Variation on $ZnO/SiO_2/Si$

Sample	J_{sc} T(mA. cm^{-2})	J_{sc} T′(mA. cm^{-2})	V_{oc} T(V)	V_{oc} T′(V)	FF T	FF T′	η T%	η T′%
without annealing treatment (0 °C)	0.08	0.07	1.45	2.14	0.36	0.40	0.04	0.06
700 °C	0.04	0.04	4.50	6.00	0.37	0.35	0.07	0.10
1000 °C	0.01	0.01	3.60	5.45	0.41	0.78	0.02	0.07
1100 °C	0.13	0.11	5.71	8.51	0.54	0.55	0.40	0.53

Description: T=initial measurement temperature; T'=measurement temperature after the sample is subjected to heat treatment.

Figure 6 (a) the highest efficiency occurred at 1100 °C annealing temperature of 0.402%. while the lowest efficiency at 1000 °C was 0.0291%. and in Figure 6 (b) the highest efficiency occurred at 1100 °C at 0.533% and the lowest efficiency occurred at 700 °C at 0.106%. As the annealing temperature increases, the crystal size increases and becomes denser, so the resistivity decreases. The resistivity value is affected by the ZnO layer which can minimize resistivity and increase conductivity. So that the number of ionized electrons increases which results in higher efficiency. This is in accordance with research conducted by Iwantono et al. (2016) and Liang et al. (2020).

Conclusion

This study demonstrates that surface modification of the SiO_2/Si substrate through annealing treatment significantly influences the crystal structure, optical characteristics, and morphology of $ZnO/SiO_2/Si$ thin films. Increasing the annealing temperature from 700, 1000, and 1100 °C enhances crystallinity and enlarges the crystal size in accordance with the wurtzite structure of ZnO. Furthermore, annealing affects the concentration of intrinsic defects, particularly oxygen and zinc vacancies, which play an important role in determining carrier concentration and light absorption behavior. The optimal annealing temperature improves light absorption over a broader wavelength range and results in more uniform, densely packed ZnO grains with reduced porosity. Although the J-V measurement reflects photovoltaic response, the observed improvements in crystallinity, optical absorption, and microstructure also indicate enhanced potential for photothermoelectric (PTE) performance, since better structural quality and controlled defect states can promote balanced photo-induced charge transport and thermal management. Therefore, controlling the annealing temperature of the SiO_2/Si substrate is a crucial step to optimize the overall photo-thermal-electric conversion performance of ZnO/SiO_2/Si thin films for future PTE device applications.

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Author Contributions
This article was written by four authors, namely L.F., R.K., C.I.Y., and E.L.

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Conflicts of Interest
The authors declare no conflict of interest.

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