Journal of Current Science and Technology, January-April 2024 Vol. 14 No. 1, Article 11
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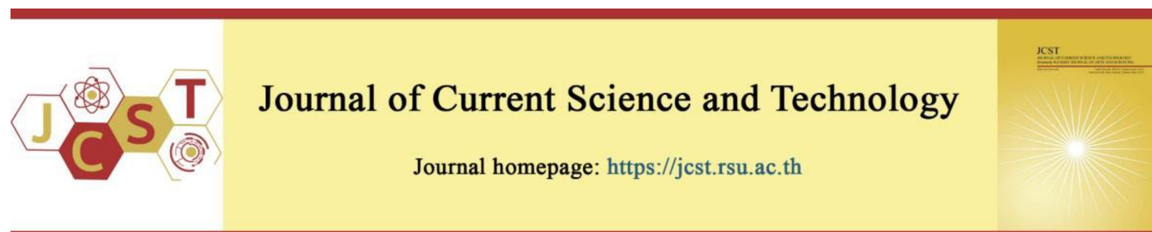
ISSN 2630-0656 (Online)

Cite this article: Mungchamnankit, A., & Limnonthakul, P. (2024). The optical properties characterization of hydrogenated silicon by spectroscopic ellipsometry for solar cell applications. *Journal of Current Science and Technology*, *14*(1), Article 11. https://doi.org/10.59796/jcst.V14N1.2024.11



The Optical Properties Characterization of Hydrogenated Silicon by Spectroscopic Ellipsometry for Solar Cell Applications

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Received 17 May, 2023; Revised 29 September, 2023; Accepted 6 October, 2023
Published online 6 December, 2023

Abstract

Amorphous and microcrystalline silicon play a critical role as layers in solar cell design, specifically in the absorption process layer which is crucial for the efficiency of sunlight conversion. This study explores the creation of hydrogenated amorphous silicon films (a-Si:H) via very high-frequency plasma-enhanced chemical vapor deposition (VHF-PECVD) and analyzes their optical properties and crystal structure. We determine the optical band gap of amorphous silicon using a Tauc plot and Spectroscopic Ellipsometry (SE) analysis of transmittance results. By comparing the data, we gain insights into the material's optical properties, aiding our understanding of the findings and enabling a swift evaluation of its optical band gap. Furthermore, we characterize the film's crystal structure using grazing X-ray diffraction. Our results reveal that a-Si:H on glass, produced by VHF-PECVD at a substrate temperature of 200°C with various hydrogen dilutions (R_H=H_2/SiH_4) ranging from 1.0 to 5.0, remains in the amorphous phase. The SE modeling provides the optical band gap of the a-Si:H film, with the lowest and highest optical band gap values occurring at RH 1.0 (1.79 eV) and R_H 3.0 (1.84 eV), respectively. We employed the Tauc-Lorentz model with five fitting parameters to extract optical properties and the band gap of amorphous silicon, including the real part of the dielectric function. This model incorporates three layers: the interface layer between the glass substrate and a-Si:H, the a-Si:H film, and surface roughness, yielding the most accurate model. This comprehensive analysis unveils the exact optical properties of the fabricated films, offering valuable insights for solar cell design and manufacturing. This research confirms that Spectroscopic Ellipsometry, a rapid and non-destructive method for measuring the optical band gap, can significantly benefit the solar cell fabrication industry.

Keywords: *spectroscopic ellipsometry; physical model; Tauc-Lorentz oscillator; very high-frequency plasma enhanced chemical vapor deposition.*

1. Introduction

Hydrogenated amorphous silicon (a-Si:H) is a popular material used for the production of solar cells due to its high optical absorption coefficient over a wide range of wavelengths, extending from visible to infrared. The band gap can be extended by adjusting the method and conditions of deposition. Additionally, a-Si:H is relatively easy to dope and can be easily prepared as either n-type or p-type by chemical vapor deposition (CVD) (Shin et al., 2014; Juneja et al., 2015; Lin et al., 2020; Dorostghol, & Kosarian, 2021; Zeng et al., 2022). As a result, numerous researchers in the field of a-Si: H solar cells continue to invest significant efforts in developing and optimizing a-Si: H-based alloys to enhance their efficiency. For instance, researchers have been experimenting with different compositions and structures, such as incorporating nanomaterials or altering deposition techniques, aiming to improve the overall performance of a-Si:

H solar cells. These efforts show the ongoing work to make solar energy better and more sustainable. Therefore, microcrystalline hydrogenated silicon has also been extensively researched for use in double junction solar cells to increase efficiency.

The structure and optical properties of hydrogenated silicon have been widely studied, and it is well-known that the microstructure of silicon films can range from amorphous to polycrystalline with varying grain sizes. Different microstructures of silicon films result in different efficiencies in solar cell structures. Spectroscopic ellipsometry (SE) has emerged as a powerful tool for studying the optical properties of silicon-based materials. Researchers have extensively utilized SE to investigate various aspects of silicon films, including refractive index, extinction coefficient, and thickness. The high precision and non-destructive nature of SE make it indispensable in characterizing thin films for solar cell applications. By employing SE, scientists have gained valuable insights into the optical behavior of silicon, enabling advancements in solar cell design and fabrication. The important step in SE modeling involves the physical model and parameters used in the fitting process. Normally, the dispersion models such as Forouhi and Bloomer (FB) (McGahan et al., 1994), Tauc-Lorentz (TL) (Jellison Jr, & Modine, 1996), and Cody-Lorentz (Cody, 1984) have been presented to extract the values of refractive index and extinction coefficient for amorphous silicon. The TL model and the Cody-Lorentz model, have been extensively utilized in the scientific community. These models provide a theoretical framework for understanding the optical properties of amorphous silicon, guiding researchers in optimizing the material's characteristics for solar cell applications. Their widespread usage underscores their significance in the field of photovoltaics. Additionally, employing the Bruggman Effective Medium Approximation (BEMA) to explain the crystallinity in semicrystalline silicon thin films (Tjaden et al., 2016) not only sheds light on the intricate application methodology of BEMA but also unravels the complexities inherent in utilizing this powerful technique. For this research, the BEMA was detailing the steps and rationale for choosing BEMA, the variations in deposition parameters led to diverse microstructures in a-Si:H films, consequently resulting in distinct optical properties. Moreover, Podraza et al. (2008) proposed a phase diagram to optimize the intrinsic layers of a-Si:H by radio frequency PECVD (RF PECVD) at low rates, achieving the maximum possible flow ratio of H_2 to SiH_4 while avoiding the amorphous to microcrystalline transition phase. Takatsuka et al. (2004) conducted a comparative study of direct current (DC), radio frequency (RF), and very high frequency (VHF) excitation for PECVD of intrinsic layers of a-Si:H, finding that the hydrogen dilution ratio is the strongest parameter affecting the microstructure of Si:H. Increasing the substrate temperature was found to decrease the energy gap.

Therefore, in this research, the study focused on optimizing the deposition parameters to control the microstructure of a-Si:H films, which significantly influences their optical properties and, consequently, their efficiency in solar cell structures. The spectroscopic ellipsometry (SE) emerged as a crucial tool, allowing precise determination of the dielectric constant and optical band gaps of the a-Si:H and μc-Si:H films. The optical band gap from SE can serve as an estimate of the electronic band gap. If the material behaves as a direct band gap semiconductor, the optical band gap will directly reflect the energy required to promote an electron from the valence band to the conduction band. While the indirect band gap, the relationship between optical band gap and energy band gap can be more complex. Optical transitions might involve phonons (quantized lattice vibrations) and can influence the measured optical band gap. However, even in these cases, the optical band gap obtained from SE can provide valuable information about the material's behavior and potential applications. In this study, the SE measurements were conducted across a photon energy range of 0.75 to 6.0 eV using a J. A. Woollam spectroscopic ellipsometer. The study also employed Grazing X-ray diffraction techniques to characterize the crystal structure of the films (Phae-ngam et al., 2023). The varying deposition parameters were utilized by SE and X-ray diffraction, the study provided valuable insights into the material properties, paving the way for enhanced solar cell design and fabrication. These findings contribute significantly to the ongoing efforts aimed at advancing solar energy technologies and making them more sustainable for the future.

2. Objectives

This study involves the preparation of hydrogenated amorphous silicon films using very high-frequency plasma-enhanced chemical vapor deposition (VHF-PECVD). The primary objectives of this research include investigating the optical properties and crystal structure of these films. To achieve this, the study employs advanced tools such as Spectroscopic Ellipsometer (SE) and Grazing X-ray Diffractometer (GIXRD) to extract precise data on the optical properties and optical band gap of amorphous silicon. Additionally, this research explores the often-overlooked factor of the hydrogen dilution ratio and its substantial impact on the properties of the a-Si:H film. Examining this factor closely has the potential to find better ways to improve material properties. Moreover, this research is poised to make significant contributions to the solar energy sector by enhancing the understanding of these fundamental properties. Furthermore, the study investigates the influence of the hydrogen dilution ratio on the properties of the a-Si:H film.

3. Materials and methods

The hydrogenated amorphous silicon films (a-Si:H) were deposited on 4 mm low ion glass substrates using very high frequency plasma enhanced chemical vapor deposition (VHF-PECVD) at the substrate temperature of 200°C. The film deposition was conducted at a very high frequency of 70 MHz. The various hydrogen dilutions (R_H=H_2/SiH_4) of 1.0, 2.0, 3.0, 4.0, and 5.0 were applied for deposition method. The selection of these particular ratios was based on the critical role played by hydrogen in the growth and properties of the amorphous silicon film. The use of hydrogen dilution can significantly impact the density, microstructure, and electronic properties of the amorphous silicon film, thus affecting the performance of the solar cell module.

The optical properties of the samples were measured using ex-situ spectroscopic ellipsometry with a variable angle of incidence. The ellipsometer, model VASE, was manufactured by J. A. Woollam Co., Inc., USA. The VASE is a traditional rotating analyzer ellipsometer (RAE) where the input polarizer is fixed, while the analyzer continuously rotates. The light source is a xenon arc lamp, and the detector is a solid-state type, with a monochromator located before the sample. The uncertainty associated with Spectroscopic Ellipsometry (SE) measurements can vary based on several factors. These factors include calibration procedures, environmental conditions, and the expertise of the operator. Therefore, in this SE of J. A. Woollam was calibrated. The Spectroscopic Ellipsometer (SE) calibration process initiates with meticulous instrument setup, aligning the sample and ensuring the stability of the light source. Angle of incidence calibration is achieved through null measurements on a standard sample (Si wafer with SiO_2 thickness 2 nm), where ellipsometry parameters (Ψ and Δ) are meticulously recorded at the null point. The critical aspect of wavelength calibration necessitates adjustments using known spectral lines and rigorous validation against standards such as silicon. This precision ensures the accuracy of SE measurements, laying the foundation for reliable scientific analyses. This instrument is capable of conducting dynamic scans at arbitrary wavelengths or spectroscopic scans from 190-1700 nm (or 0.73-6.5 eV), with incident angles of 65°, 70°, and 75°. Our thin film materials were deposited on 4 mm glass substrates, therefore, to avoid the effect of the amorphous phase of the glass substrate, we utilized grazing incident x-ray diffraction (GIXRD) to characterize the crystal structure of the thin films (Panyanon et al., 2021). The GIXRD, model TTRAX III, was manufactured by Rigaku. During measurements, the Cu-Kα (1.5426 Å) radiation was operated at 50 kV and 300 mA. The incident angle on the surface was 0.4 degrees, with a scanning speed of 2 degrees per minute at 2θ step of 0.02 degrees. The range of 2θ used was 20-75 degrees.

For the Tauc-Lorentz model (TL) was used in this SE modeling. It is a theoretical for analyzing the optical properties of materials, particularly amorphous and crystalline semiconductors. It describes the absorption coefficient as a function of photon energy. There are several parameters in the Tauc-Lorentz model, including the bandgap energy, the broadening parameter, and the oscillator strength. The bandgap energy is the minimum energy required for a material to absorb a photon of light, and the broadening parameter describes the width of the absorption peak. The oscillator strength measures the material's ability to absorb light. These parameters are referred to a numerical value used to describe a material’s behavior in the model. Specifically, the model describes the absorption of light by a material, and the parameters are used to specify the shape of the absorption curve. The

parameters in the Tauc-Lorentz model are used to fit the experimental data and determine the optical properties of the material, such as its refractive index and absorption coefficient. These parameters can be influenced by a variety of factors, such as the chemical composition, crystal structure, and temperature of the material. Therefore, we used Tauc-Lorentz model for these fittings.

4. Results and Discussion

The Spectroscopic Ellipsometry (SE) to investigate hydrogenated amorphous silicon films (a-Si:H) was prepared using very high-frequency plasma-enhanced chemical vapor deposition (VHF-PECVD). SE measurements were conducted over a wide photon energy range (0.75 to 6.0 eV) at incident angles of 65°, 70°, and 75°. The obtained ψ (°) data were analyzed to construct physical models describing the growth of a-Si:H on glass substrates. The measured data of ψ (°) were shown in Figure 1.

In the SE analysis, we first constructed a physical model to describe the physical structure of a-Si:H growth on glass substrates. The films were to be considered as physically possible film samples. We proposed 3 physical models as seen Figure 2.

In Figure 2 shows the three physical models were proposed: (a) a single-layer model (1L) representing an ideally homogeneous film, (b) a double-layer model (2L) incorporating inhomogeneity due to high growth rates, and (c) a triple-layer model (3L) considering a non-sharp interface layer during film growth. The dielectric function of the films was analyzed using the Tauc-Lorentz (TL) oscillator model, providing valuable insights into the optical properties of a-Si:H. The triple-layer model (3L) was found to best describe the microstructure of a-Si:H deposited on glass substrates, incorporating interface and top layers.

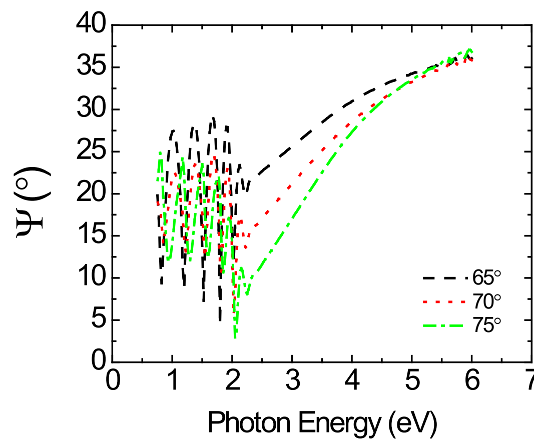
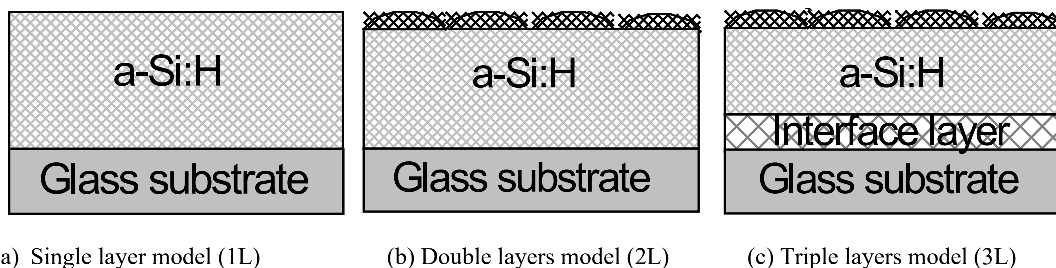


Figure 1 Schematic representation of a-Si:H film was prepared by VHF-PECVD at substrate temperature 200°C with hydrogen dilution 1.0 on glass substrate with various incident angle 65°, 70° and 75°



(a) Single layer model (1L) (b) Double layers model (2L) (c) Triple layers model (3L)

Figure 2 Schematic representation of physical model of a-Si:H film growth on glass substrate for SE analysis: (a) single layer model (1L), (b) double layers model (2L) and (c) triple layers model (3L).

We utilized the Tauc-Lorentz (TL) oscillator model extensively for our Spectroscopic Ellipsometry (SE) analysis. This model, developed by Jellison Jr, & Modine in 1996, incorporates the band edge of the material into its dielectric function. The TL oscillator is based on the Tauc joint density of state and the Lorentz oscillator, denoted by the abbreviation TL. According to the TL model, the absorption of the material follows a Tauc law formula:

$$\varepsilon_{imaginary(TL)}(E) \text{ or } \varepsilon_{2(TL)}(E)=\begin{cases} \frac{A\cdot E_0\cdot\Gamma\cdot(E-E_g)^2}{(E^2-E_0^2)^2+\Gamma^2\cdot E^2}\cdot\frac{1}{E}; & for \quad E>E_g \\ 0; & for \quad E\leq E_g \end{cases} \quad (1)$$

where A (eV) is the amplitude, E_0 (eV) is the peak transition energy, E_g (eV) is the energy gap and Γ (eV) is the broadening parameter. The four fitting parameters are A, E_0, E_g and Γ. The real part of the dielectric function ε_{real} is obtained by Kramers - Kronig integration, given by

$$\varepsilon_{real(TL)}= \varepsilon_{real}(\infty) + \frac{2}{\pi} P \int_{E_g}^{\infty} \frac{\xi\cdot\varepsilon_2(\xi)}{\xi^2-E^2} d\xi \quad (2)$$

where P stands for the Cauchy principle part of the integral and an additional fitting parameter $\varepsilon_1(\infty)$ has been included. The ξ is the angular optical frequency, which defines the oscillation frequency of the corresponding electromagnetic wave in the context of optics. The $\varepsilon_2(\xi)$ parameter refers to the imaginary part of the complex refractive index, which is proportional to the absorption coefficient of the material. From equations (1) and (2), the complex index of refraction, absorption coefficient and the energy gap of Tauc joint density of state and the Lorentz oscillator could also be found. The dielectric function of the Tauc-Lorentz model included five fitting parameters: are A, E_0, E_g, Γ and real part of the dielectric function ε_{real}. We employed these parameterizations to analyze the dielectric function of the a-Si:H film by applying them to the physical models we proposed earlier. The results of curve fitting for a-Si:H with R_H=1.0 are presented in Figure 3. The mean square error (MSE) results within the photon energy range of 0.75 to 6.0 eV were 45.53, 19.43, and 11.56 for models 1L, 2L, and 3L, respectively. It was evident that the 3L model yielded the most favorable results for this experiment. This model indicated the necessity of an interface layer and a top layer in the microstructure of this a-Si:H sample. Comparative analysis of the dielectric function and thickness layers for bulk a-Si:H (d_f) and the top layer (d_t) is presented in Table 1. Based on these outcomes, it was determined that the triple-layer model (3L) was the most suitable to describe the microstructure of a-Si:H deposited on a glass substrate. Additionally, the TL dispersion model was employed to explain the dielectric function of the a-Si:H film. Consequently, we utilized this method to determine the optical properties and parameterization for all samples. The summarized parameterization for a-Si:H prepared at various R_H values (1.0, 2.0, 3.0, 4.0, and 5.0) is presented in Table 2. The corresponding MSE values for these samples were 8.061, 9.897, 8.501, 7.361, and 6.434 for R_H 1.0, 2.0, 3.0, 4.0, and 5.0, respectively.

In Figure 4 compared the refractive index (n)-extinction coefficient(k) of all a-Si:H in this research and the reference material of Palik and Aspnes which measurement from bulk a-Si materials. The results show a decrease in both (*n, k*) values as the hydrogen dilution ratio (R_H) decreases. This observation indicates that the a-Si:H films formed an amorphous phase, confirming the amorphous nature of these films based on their optical constants.

Table 1 Parameter values obtained for fits for Tauc-Lorentz parameterizations with 3 physical models.

Model	Interface layer			Bulk a-Si:H					Top	
	d_i (nm)	Glass (%)	a-Si:H (%)	d_f (nm)	Amp (eV)	E_0 (eV)	Γ (eV)	E_g (eV)	d_t (nm)	Void (%)
Model 1L	-	-	-	496.8	110.51	3.4279	1.5160	1.53	-	-
Model 2L	-	-	-	454.6	162.25	3.8534	1.6545	1.57	7.4	50.00
Model 3L	32.8	50.86	49.14	430.8	228.93	3.5954	1.7344	1.79	9.4	39.60

Table 2 Parameter values obtained for fits for Tauc-Lorentz parameterizations with 5 sample of a-Si:H films which prepared by VHF-PECVD at various R_H.

R_H	Interface layer			Bulk a-Si:H						Top
	d_i (nm)	Glass (%)	a-Si:H (%)	d_f (nm)	Amp (eV)	E_0 (eV)	Γ (eV)	E_g (eV)	d_t (nm)	Void (%)
1.0	32.8	50.86	49.14	430.8	228.93	3.5954	1.7344	1.79	9.4	39.60
2.0	37.1	63.77	36.23	605.5	233.44	3.5879	1.8887	1.78	8.3	36.24
3.0	35.0	57.24	42.76	426.4	235.58	3.5404	1.8963	1.84	7.9	35.13
4.0	30.8	51.71	48.29	436.4	232.46	3.5498	1.9578	1.83	6.8	36.78
5.0	28.6	34.28	65.72	423.1	187.93	3.7030	2.0424	1.78	3.6	47.67

To thoroughly assess the optical band gap values of a-Si:H obtained from Spectroscopic Ellipsometry (SE) and the traditional Tauc-plot method mentioned earlier, we conducted a detailed comparison between the results obtained from both techniques. Using the Tauc-plot method, the optical band gap values were calculated according to Equation 3. This allowed us to meticulously analyze and compare the optical band gap results obtained from both methods, ensuring a comprehensive and efficient evaluation of the material's transparency characteristics in this research study. The absorption coefficient (α) was determined from Equation 3, when we know the extinction coefficient (k):

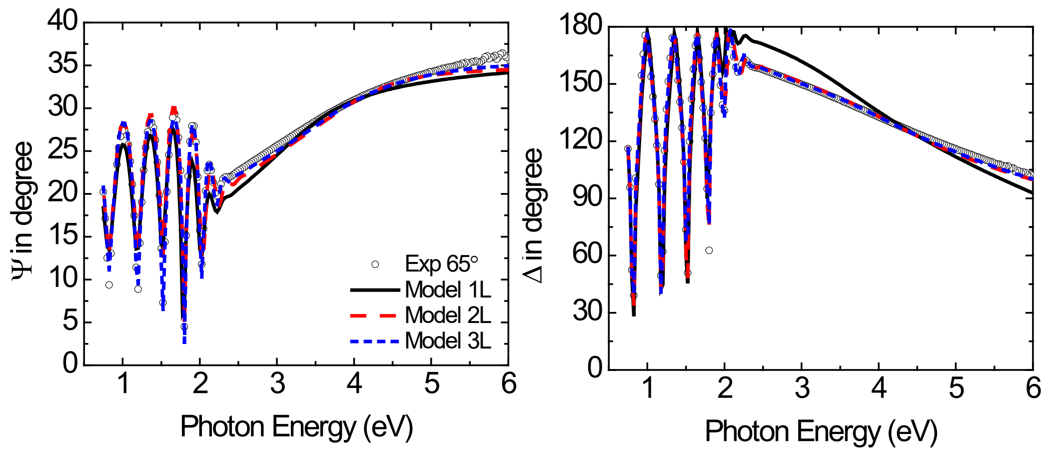
$$\alpha = \frac{4\pi k}{\lambda} \quad (3)$$

where α is absorption coefficient (cm^{-1}), λ is wavelength (cm) and k is extinction coefficient. The relation between energy gap and the absorption coefficient, α, and the energy $h\upsilon$ of incident photon is given by:

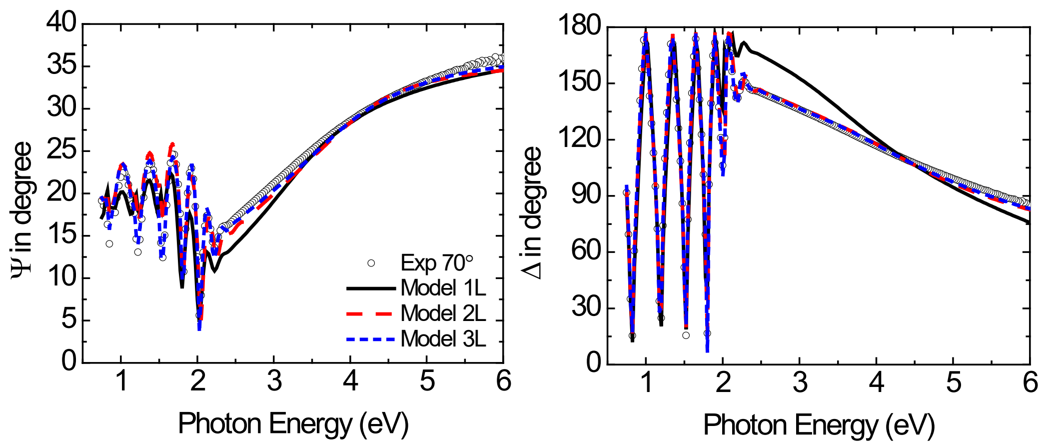
$$\alpha h\upsilon = C(h\upsilon - E_g)^m \quad (4)$$

where $h\upsilon$ is photon energy (eV), C is constant, E_g is energy gap and m is a constant depending on the optical transition mode of semiconductor.

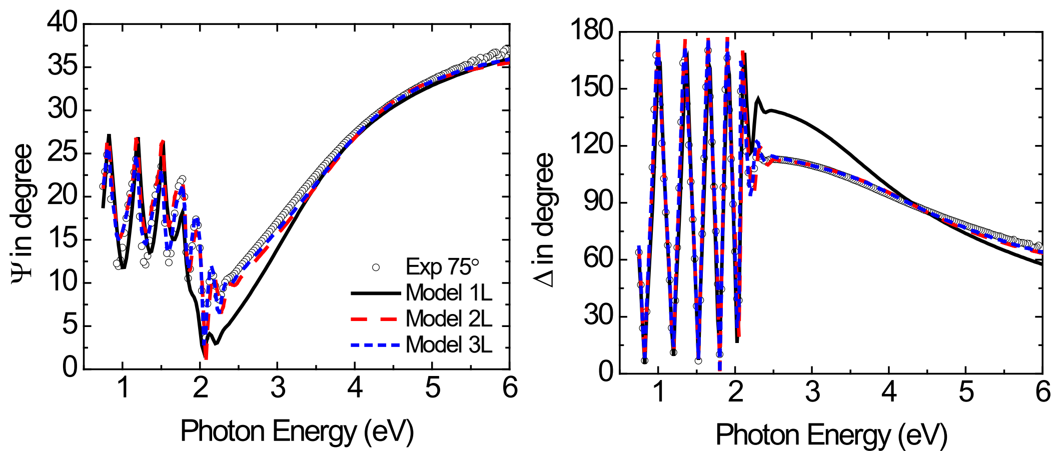
Using Equation 3 and the a-Si optical properties illustrated in Figure 4, which were obtained from the reference a-Si of Aspnes and Palik., we calculated the absorption coefficients (α) for a-Si:H samples at various hydrogen dilution ratios (R_H). The calculated coefficients were then plotted on Figure 5. For ease of comprehension, we plotted the absorption coefficients (α) on a logarithmic scale to visualize the significant changes. The results show the areas of strong light absorption in the materials. In Figure 4 shows the graph of the extinction coefficient exhibits a blue shift, it means that the a-Si:H films is absorbing light at higher energy levels. Foe comparisons the traditional method, the Tauc plot represents the energy of light (plotted on the y-axis) against $(\alpha h\nu)^2$ (plotted on the x-axis), where α is the absorption coefficient, h is Planck's constant, υ is the frequency of light, and m is a constant denoting the nature of the electronic transition (for a-Si:H, $m = 2$ for indirect transitions). The Tauc plot could determine the optical band gap by using linear regression. The cross-section point on the x-axis corresponds to the optical band gap of the material, which can be obtained through the equation provided in Equation 4. The optical band gap values obtained from the SE Tauc-Lorentz (TL) model (from Table 2) and the Tauc plot in Figure 6 are very close, indicating consistency between the methods. The optical band gap (E_g) at various hydrogen dilution ratios (R_H) (1.0-5.0) of TL model are: 1.79, 1.78, 1.84, 1.83 and 1.78, respectively. The optical band gap of Tacu plot is: 1.80, 1.80, 1.86, 1.85 and 1.81, respectively. The slight differences in the optical band gap values (not exceeding 0.04 eV) suggest that the SE technique effectively extracts the optical properties of the materials, allowing for precise determination of optical band gap using Tauc plots. This approach demonstrates the accuracy and reliability of the SE technique in determining the optical band gap of a-Si:H materials.



(a) fittings on ψ and Δ at incident angle 65°



(b) fittings on ψ and Δ at incident angle 70°



(c) fittings on ψ and Δ at incident angle 75°

Figure 3 Illustrations of generated and experimental data, curve-fitted with three physical models for the a-Si:H was prepared by VHF-PECVD at substrate temperature 200°C with hydrogen dilution 1.0 on glass substrate: (a) fittings on ψ and Δ at incident angle 65° b) fittings on ψ and Δ at incident angle 70° and (c) fittings on ψ and Δ at incident angle 75°

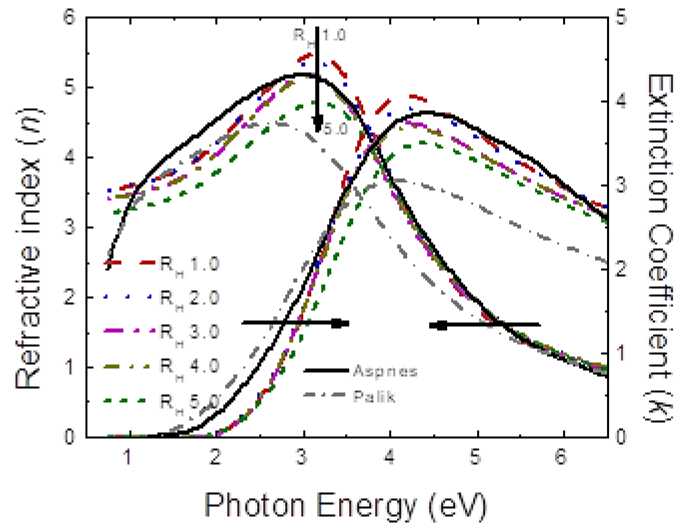


Figure 4 Optical constants of the a-Si:H film was deposited on glass substrate by VHF-PECVD with various hydrogen dilution (R_H), 1.0, 2.0,3.0,4.0 and 5.0, respectively. And compared with the reference a-Si of Aspnes and Palik.

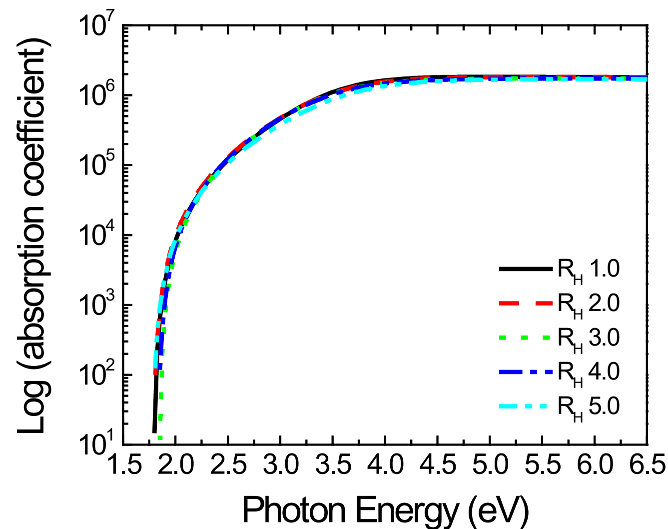


Figure 5 Absorption coefficient in log scale versus photon energy of a-Si:H on glass prepared by VHF-PECVD at substrate temperature 200°C with various hydrogen dilution (R_H=H_2/SiH_4) 1.0, 2.0, 3.0, 4.0 and 5.0, respectively.

For the crystalline phase of a-Si:H films were characterized by GIXRD. The XRD pattern was shown in Figure 7. In the result, we show only one result of XRD pattern due to all of sample which prepared a-Si:H at various R_H had the same result. Grazing X-ray Diffraction (GIXRD) analysis confirmed the amorphous nature of the a-Si:H films. The broadening peak of the glass substrate and the absence of distinct silicon peaks in the XRD pattern confirmed the amorphous phase of the deposited films. These comprehensive analyses provide a clear understanding of the optical and microstructural properties of the a-Si:H films, offering valuable insights for future applications in solar cell technology.

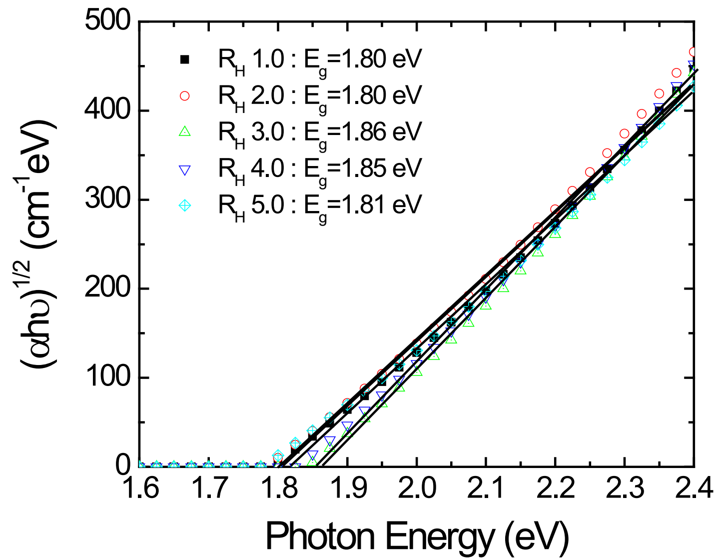


Figure 6 Tauc plot versus photon energy of a-Si:H on glass prepared by VHF-PECVD at substrate temperature 200°C with various hydrogen dilution ($R_H=H_2/SiH_4$) 1.0, 2.0, 3.0, 4.0 and 5.0, respectively.

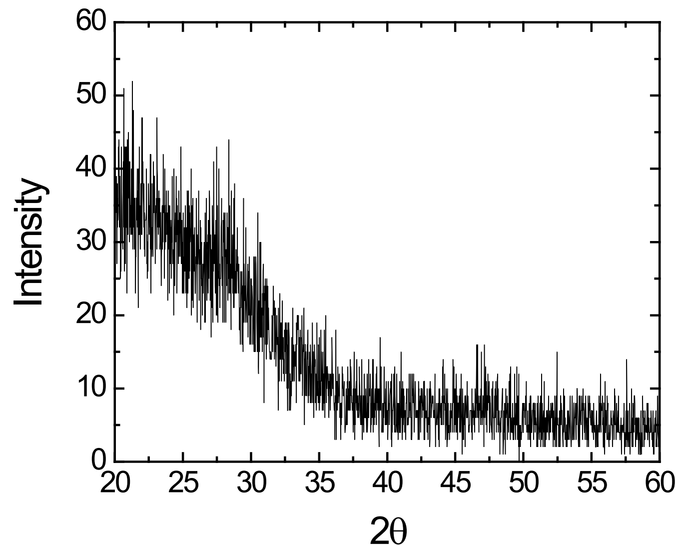


Figure 7 XRD pattern of a-Si:H on glass prepared by VHF-PECVD at substrate temperature 200°C with various hydrogen dilution ($R_H=H_2/SiH_4$) 1.0, 2.0, 3.0, 4.0 and 5.0, respectively.

5. Conclusion

The findings from Spectroscopic Ellipsometer modeling, our study delves into the intricacies of hydrogenated amorphous silicon (a-Si:H). We observed a noteworthy correlation between the hydrogen dilution ratio ($R_H = H_2/SiH_4$) and the material's optical properties. As R_H increases, indicating a higher concentration of hydrogen gas relative to silane gas in the reaction chamber, there's a corresponding rise in the energy gap. This phenomenon is rooted in the process of hydrogen dilution, which effectively reduces the hydrogen content in the a-Si film, thereby diminishing the density of states within the bandgap. Consequently, more energy is required to excite an electron from the valence band to the conduction band, leading to an increase in the optical band gap. However, when R_H in the preparation process was increased, there's also an SiH_3 radicals in the gas phase were increased. These radicals play a pivotal role in fostering the formation of Si-H bonds within the a-Si film. The

presence of these Si-H bonds introduces additional defect states within the energy gap. These states facilitate the excitation of electrons to the conduction band at lower energy levels, thereby potentially reducing the optical band gap. Consequently, at higher R_H values, the concentration of Si-H bonds and the associated defect states might outweigh the effect of reduced hydrogen content, causing a decrease in the optical band gap of the a-Si thin film. The complex relationship between hydrogen dilution and the optical properties of a-Si:H underscores the intricacy of the material's behavior. Significantly, our spectroscopic model not only unravels these complexities but also demonstrates its potential as a comprehensive tool. By employing this model, we can discern both the physical model of the material and its optical properties in a single analysis, providing a holistic understanding of a-Si:H behavior under varying R_H conditions.

6. Acknowledgements

We extend our sincere gratitude to the National Electronics and Computer Technology Center (NECTEC) for graciously supporting our research project through the provision of the spectroscopic ellipsometer and GIXRD characterization. In addition, we would like to express our deep appreciation to our esteemed colleagues and friends for their unwavering support and encouragement throughout the duration of this research.

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