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The novel usage of spectroscopic ellipsometry for the development of amorphous Si solar cells

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ABSTRACT

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Keywords: a-Si:H Thin film Si solar cell Spectroscopic ellipsometry (SE) We present the novel use of spectroscopic ellipsometry (SE) for the development of a-Si:H solar cell. SE is a very fast and useful tool to measure various optical properties of thin film. In the case of a-Si:H thin film analysis, generally, SE is used to estimate the film thickness, roughness, void fraction, optical constants such as (n,k), and so forth. In this study, optical parameters from SE measurements were analyzed with relation to structural and electrical properties of a-Si:H thin film for solar cell. By analyzing IR absorption spectra and conductivity measurements, it was affirmed that $\langle \varepsilon_2 \rangle$ and parameter *A* by Tauc–Lorentz model fitting of SE data are representative parameters qualifying a-Si:H thin film, and that they have close relationships with FF and light induced degradation property of solar cells. Based on the analysis, solar cells that have i-layers with various E_g were optimized. By this research, easier and faster methodology to develop a-Si:H thin film for thin film Si solar cells using SE measurements was established.

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1. Introduction

Thin film Si solar cell is one of the most promising technologies to reduce cost of the electricity generation. However, its low conversion efficiency is considered as a neck point to overcome. To improve the efficiency, multi-junction approaches such as a-Si:H/µc-Si:H double junction or a-Si:H/a-SiGe:H/µc-Si:H triple junction are being researched. In the multi-junction, optimization of the thin film deposition process is necessary to control the band-gap (E_g) and the quality of amorphous silicon (a-Si:H), which are appropriate to each junction cell.

In order to qualify properties of a-Si:H thin film, various methods are being used, such as conductivity measurement [1], Reflectance-transmittance (R–T) measurement [1], Fourier-transformed infrared (FTIR) measurement [2,3], constant photocurrent measurement (CPM) [4], spectroscopic ellipsometry (SE) measurement [5,6], and so forth. Among them, SE is a very sensitive tool to analyze optical properties of thin film, and these properties must be related very closely to film's structural and optoelectronic properties. Therefore, if the SE results of a unit layer are correctly analyzed, the performance of the solar cell composed of the layer could be predicted, and optimization of the solar cell might be easier and faster.

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In this study, at first, dielectric function $\langle \varepsilon_2 \rangle$ was measured by SE and optical parameters were evaluated by Tauc–Lorentz model fitting [5,6]. Secondly, the relationship between these parameters and the structural and optoelectronic properties that are represented by FTIR spectra and conductivity was analyzed. Then these optical parameters and solar cell performances such as $V_{\rm oc}$, $J_{\rm sc}$, FF, efficiency, and light induced degradation property were investigated. Finally, with these results, solar cells that have ilayer with various E_g were fabricated.

2. Experimental details

300–500 nm thick intrinsic a-Si:H thin films were deposited on corning 1747 glass by means of 13.56 or 40 MHz plasma enhanced chemical vapor deposition system. This system is a cluster type equipment that has three PECVD chambers for p, i, n type Si separately, and one sputter chamber for ZnO:Al layer [7]. Intrinsic a-Si:H thin films for analysis of SE and other film properties were prepared in the so-called high pressure high power regime, which is generally used for microcrystalline layer deposition. Samples were classified into two sets by CVD working pressure and RF power. Set-1 samples were deposited at working pressure of 4 Torr and RF power of 200 W condition, and set-2 samples were deposited at 8 Torr/300 W. Substrate temperature was lower than 200 °C in all the samples.

SE measurements were performed using the product of J.A. Woollam company which has photon energy range of 1.2–5 eV.

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This equipment includes well-made software to fit experimental data and modeled parameters. Ellipsometric spectra (ψ , Δ) were measured to deduce dielectric function $\langle \varepsilon_2 \rangle$, and the Tauc–Lorentz (T–L) model and multi-layer model of roughness layer/ a-Si:H/glass stack were also used to derive optical parameters [6,8,9]. T–L empirical model is expressed as following equation:

$$\varepsilon_{2}(E) = \frac{AE_{0}C(E-E_{g})^{2}}{(E^{2}-E_{0}^{2})^{2}+C^{2}E^{2}}\frac{1}{E}, \quad E > E_{g}$$

$$= 0 \quad E \le E_{g}$$
(1)

where parameters *A*, *E*₀, *C*, and *E*_g (in eV) represent, respectively, an amplitude factor, the oscillator resonance energy, the oscillator broadening parameter, and the Tauc optical gap. The symbols < > are used to remark they are psuedo-dielectric function. Al-coplanar electrodes deposited by an e-beam evaporation system were used to measure dark and photo conductivities for silicon layers on bare glasses. FTIR spectra were measured in attenuated total reflection (ATR) mode, and Si–H bonding fraction (Si–H(%)) and Si–H₂ bonding fraction (Si–H₂(%)) were estimated by fitting two Gaussians of which middle positions are around 2000 and 2090 [2,3], which are IR spectrum of stretching modes, respectively.

With the cluster deposition system mentioned above, solar cells were grown in the p-i-n sequence on Asahi U-type TCO glass and had the following structure: TCO glass/p-a-Si:H(~10 nm)/ intrinsic(i) a-Si:H(~300 nm)/n- μ c-Si:H(~40 nm)/ZnO:Al. 1 cm² size metal contacts were defined using shadow mask, and Ag and Al back contact were deposited with e-beam evaporation system. Solar cells were characterized by light *I*-*V* measurement, and *V*_{oc}, *J*_{sc}, FF and efficiencies were deduced from these *I*-*V* curve. Light soaking (LS) treatment of solar cells was performed for 1000 h keeping at 50 °C, and all photocurrent versus voltage measurements and LS treatments were performed under air mass (AM)1.5, 100 mW/cm².

3. Results and discussion

3.1. a-Si:H thin films analysis

Set-1 samples were deposited in the H₂/SiH₄ gas flow ratio (*R*-ratio) range of 4–15. Fig. 1 shows dielectric function $< \varepsilon_2 >$ measured by SE (solid line) and fitting results (dashed line). In this



Fig. 1. Dielectric function $< \varepsilon_2 >$ spectra of amorphous Si thin film samples (set-1). As *R*-ratio increased, peak value increased, and corresponding photon energy decreased slightly.

set, as *R*-ratio increased, $< \varepsilon_2 >$ curve moved upwards and photon energy corresponding to peak $< \varepsilon_2 >$ value shifted to slightly lower energy. Among optical parameters from T–L model fitting as shown in Table 1, parameter A value increased with Rratio; however, Eg decreased to around 1.75 eV and increased again. These trends were also shown in other pressure and power conditions, although they are not shown in this report. On the other hand, other parameters E_0 and C did not have clear tendency. With this set-1, FTIR spectra were measured and Si-H(%) and Si-H₂(%) were calculated as also shown in Table 1. Si-H(%) increased and Si-H₂(%) decreased as *R*-ratio increased. As many researchers have investigated, less $Si-H_2(\%)$ means lower void fraction [10], denser structure [9], or lower defect density [2]. Therefore, it could be concluded that increase in $< \varepsilon_2 >$ and A with R-ratio means better film quality. However, parameter A corresponds to amplitude of Tauc-Lorentz model, which is empirical formula for ε_2 [8], and it is natural that increase in $< \varepsilon_2 >$ peak should lead to increase in A parameter if fitting is properly performed. Therefore hereafter, only $\langle \varepsilon_2 \rangle$ will be considered to gualify a-Si:H.

With set-2 samples, optical parameters estimated from SE analysis and conductivity (both dark-, photo-) were measured as shown in Table 2. Similarly with set-1, $\langle \varepsilon_2 \rangle$ increased with *R*-ratio, and photo-conductivity and photo-/dark-sensitivity also increased, which means that optoelectronic quality of a-Si:H film improved with *R*-ratio. From these set-1 and 2 results, we could infer that $\langle \varepsilon_2 \rangle$ represents quality of a-Si:H thin film, and intrinsic layer with higher $\langle \varepsilon_2 \rangle$ value might lead to higher conversion efficiency. Then, we can get very powerful methodology to optimize a-Si:H solar cells, because SE is very simple and fast measurement tool compared with other analysis methods such as conductivity measurement or FTIR measurement.

3.2. Solar cell performances

Sonobe et al. [3] reported that a-Si:H film with lower Si–H₂(%) showed lower light induced degradation property and also lower Si–H₂(%) indicated higher film quality. They also reported that among solar cell performance variables, FF is strongly affected by intrinsic layer quality. With these results, we could predict that intrinsic layers with higher $<\varepsilon_2 >$ value would lead to higher conversion efficiency, especially with higher FF.

Fig. 2 is the plot of FF and efficiency versus $\langle \varepsilon_2 \rangle$ of solar cell's intrinsic layer. Additionally, Fig. 2 includes stabilized cell performances as well as initial FF and efficiencies. Initial FF increased as $\langle \varepsilon_2 \rangle$ of intrinsic layer increased, and saturated when $\langle \varepsilon_2 \rangle$ is higher than about 22. The tendency of initial efficiency was almost same as FF, that is to say, it increased with $\langle \varepsilon_2 \rangle$ However, efficiency showed slight decrease when $\langle \varepsilon_2 \rangle$ was higher than 23. This decrease in efficiency was due to the

Table 1

Parameters from SE analysis and FTIR results with respect to H_2/SiH_4 gas flow ratio (*R*-ratio) (set-1 samples). $< \varepsilon_2 >$ peak value and *A* increased as *R*-ratio increased, and Si-H(%) had same tendency.

Samples			SE analysis				FTIR analysis	
R-ratio	$< \varepsilon_2 >$ peak	D.R. (nm/s)	Eg (eV)	Α	En	С	Si–H (%)	Si-H ₂ (%)
4 7 10 15	17.2 19.7 20.3 21.9	3.3 2.91 2.23 1.65	1.797 1.754 1.750 1.762	139 187 196 209	3.764 3.654 3.71 3.623	2.138 2.291 2.46 2.404	29 49.4 57.7 71.7	71 50.6 42.3 28.9

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