



Research Article

Silicon dioxide thin films prepared by spin coating for the application of solar cells

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ABSTRACT

In this study, Silicon Dioxide (SiO₂) thin films processed by the spin coating method was studied with prepared solutions. Antireflection coating effect of deposited SiO₂ thin films on crystalline silicon substrates was analyzed after optimizing the solution, deposition, and thermal treatment processes. The effect of ethanol dilution of the solution was investigated as well. Spectrophotometer reflectance measurements, Scanning Electron Microscopy (SEM) measurements and Afors-het based simulations were carried out. For the prepared solution based SiO₂ thin films, the annealing temperature of 950 °C for 7 min in the air was determined as optimum. The minimum surface reflectance of SiO₂ coated silicon surface could be reduced below 10% depending on the applied process. Based on the silicon solar cell device simulations, it was revealed that efficiency of a solar cell could be improved 4.23% more thanks to the antireflection coating effect.

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1. Introduction

The reflection of the light coming on the surface of the solar cells is an important factor affecting the efficiency [1]. Anti-reflective layers (ARLs) are playing an important role in minimizing reflection losses on the surface of solar cells. By applying one or more ARL layers to photovoltaic cells [2] or to other optoelectronic devices, the reflection of incoming light can be reduced, thus the performance of the device can be improved [3, 4]. However, high production and raw material expenses increase the cost per unit watt. Energy costs can be reduced by reducing costs in production methods. As an effective thin film coating approach, the ARLs can be easily prepared by sol-gel based coating processes such as dipping, spinning, meniscus, and spray pyrolysis to obtain quality optical thin films [5-8]. Compared to physical and chemical steam methods, the sol-gel method has many advantages such as being cheaper, easily adaptable to industry scale and mass production, working under normal atmospheric conditions, high purity, ultra-homogeneity, and working at the desired

molarity and temperature [9-11]. For anti-reflection coatings, highly transparent materials such as SiO₂, Si₃N₄, TiO₂, Al₂O₃, Ta₂O₅, and SiO₂-TiO₂ are used for c-Si solar cells [12].

High-quality anti-reflection film coatings are generally produced by methods that require vacuum, such as thermal evaporation, atomic layer deposition, chemical vapor deposition, or plasma-enhanced chemical vapor deposition method, etc. [13]. However, these methods are quite costly compared to the sol-gel method. In addition, most of these methods are performed with atmosphere control. On the other hand, the sol-gel method including spin coating is generally carried out in ambient air. The solution needs to be prepared properly to meet the aim of application which will enhance antireflection properties in current study.

In this study, SiO₂ thin films created by the spin coating method and their antireflection properties were investigated. The used solution shows differences in terms of content ratios and experimental stages compared to the literature. In addition to the explained preparation and optimization process of the SiO₂ solution, the effect of

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ethanol dilution of the solution was investigated as well. Moreover, the effect of ARL on c-Si solar cells obtained by SiO₂ thin films was observed using Afors-het simulations.

2. Experimental

The initial SiO₂ solution was prepared with volume ratio of 3: 2: 4; TEOS (C₈H₂₀O₄Si - %99), Ethyl alcohol and water were mixed with a magnetic stirrer for half an hour at room temperature. Then, 150 µL of HCl was added to clear the solution. Subsequently, 8 mL of ethyl alcohol was added to the prepared solution to dilute it and it was kept at room temperature for 24 hours (would be called “SiO₂-Sol” hereafter). Polished p-type CZ-Si with 725 µm thickness and 1.5 cm × 1.7 cm dimensions was used as a substrate. Substrates were cleaned with ethanol in an ultrasonic bath for 15 min and then air-dried. Then, they were further cleaned with purified water in ultrasonic bath for 15 min and finally the drying step in air was followed.

Coating was carried out using the spin coating process. SiO₂ thin films were coated on the Si substrate at 4000 rpm for 30 s. Annealing temperature, annealing time, and thin film thickness optimizations were carried out in order to achieve the minimum reflectance value for the thin film coated on the c-Si surface. To adjust the desired film thickness of the optimized solution, different amounts of solution (100 µL, 50 µL, 35 µL and 25 µL) were dropped onto the substrate surface and the coating was completed. Subsequently, samples were prepared to adjust the annealing temperature in atmosphere for 10 min from 550 °C to 1000 °C. To adjust the annealing time, annealing was performed at different temperatures for periods from 1 min to 30 min. Moreover, in order to observe the effect of further dilution of the initial SiO₂-Sol, four different diluted solutions of Sol:Ethanol (1:1, 1:2, 1:5, 1:10 in volume) were prepared. Reflectance behaviors of the films coated by further diluted SiO₂-Sols were compared to that of the films coated by undiluted SiO₂-Sol. Optical and morphological characterizations of thin films were performed with the Spectrophotometer reflection measurements and Scanning Electron Microscopy (SEM) measurements. Finally, simulations of c-Si solar cells were performed using Afors-het software.

3. Result and Discussion

To examine the characteristic properties of the coated SiO₂ thin films with varying thicknesses, thin films were formed by the spin coating method by dropping the same molarity solution in different amounts on the substrate. Depending on the amount of solution used on the substrate, the corresponding reflectance spectra of coated Si surfaces are shown in figure 1. In addition, the minimum reflectance and average reflectance values of this graph are shown in Table 1 in the range of 350 nm to 1000 nm. It

was observed that there was a shift in the minimum point of the reflectance spectra with the amount of solution dripped onto the substrate. The shift can be attributed to the increase of the formed film thickness [14, 15]. The minimum reflectance value increased from 9.60% to 11.50% as the amount of solution increased Table 1.

In addition, it was observed that the average reflectance value varied from 19.02% to 19.62% depending on the increasing amount of solution. 35 µL was set as the optimum amount of drop for the rest of the experiments because it was in the appropriate wavelength range and provided lower average reflectance (19.02%) than that of the other samples.

By using thin films created under the same conditions with the same experimental parameters, annealing temperature optimization was performed between 550 °C and 1000 °C for 10 min. The reflectance spectra of the samples prepared depending on the temperature were shown in figure 2. In addition, Table 2 presents the minimum and the average reflectance values for these samples in the wavelength ranging from 350 nm to 1000 nm.

The minimum reflectance value increased from 10.30% to 11.50% when the temperature increased from 550 °C to 750 °C; however, after 750 °C, it decreased again to 9.50% when the annealing temperature was 1000 °C (Table 2). Due to the minimum and average reflectance values as well as the appropriate wavelength range, 950 °C was considered as the optimum temperature.

Table 1. Minimum and average reflection values depending on the amount of SiO₂-Sol coated on the Si surface

Amount (µL)	Average Ref. (350-1000 nm)	Min. Ref (%)
100	19.26	11.50 (555nm)
50	19.62	11.00 (585nm)
35	19.02	10.10 (615nm)
25	19.20	9.60 (675nm)

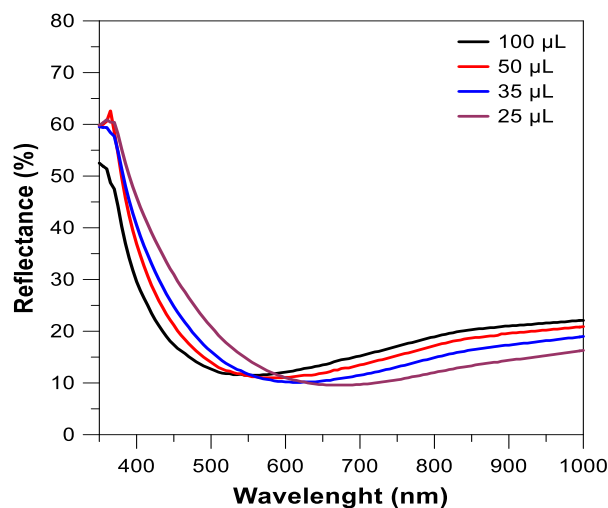


Figure 1. Reflectance spectra of the Si surface depending on the amount of coated SiO₂-solution

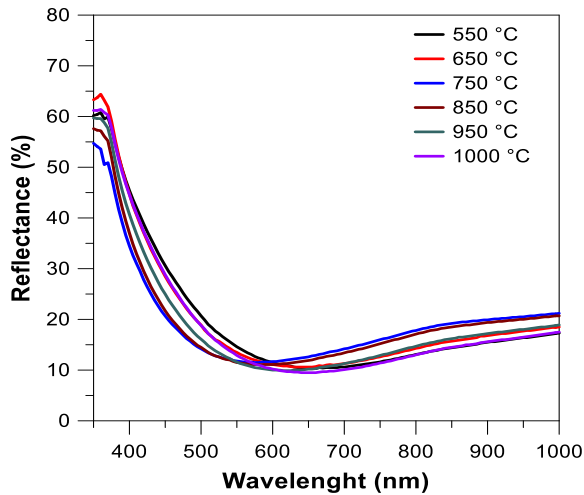


Figure 2. Reflectance of SiO₂-coated-Si surface depending on different annealing temperatures

Table 2. Minimum and average reflection values depending on annealing temperature

Temperature (°C)	Average Ref. (350-1000nm)	Min. Ref (%)
550	19.74	10.30 (660nm)
650	19.98	10.60 (645nm)
750	19.47	11.50 (580nm)
850	19.48	11.10 (590nm)
950	18.93	9.90 (620nm)
1000	19.09	9.50 (650nm)

The prepared samples were annealed in the air atmosphere at 950 °C from 1 min to 30 min in order to observe the effect of the annealing time. Reflectance spectra by annealing time are shown in figure 3.

As the annealing time increased from 1 min to 30 min, minimum reflectance values decreased continuously from 13.50% to 9.40%. In contrast to the minimum reflectance value, although the average reflectance values decreased from 20.61% to 19.26% when the annealing time was increased from 1 min to 10 min, it was increased up to 20.06% at 30 min.

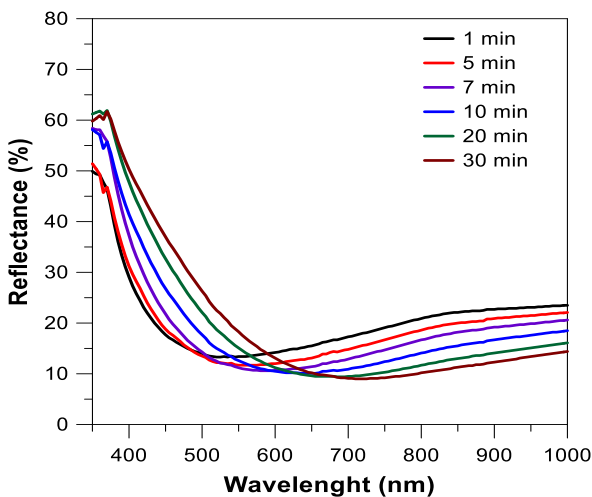


Figure 3. Reflectance of SiO₂-coated-Si surface depending on different annealing times

Table 3. Minimum and average reflection values depending on annealing time

Time (min)	Average Ref. (350-1000 nm)	Min. Ref (%)
1	20.61	13.30 (535nm)
5	19.34	11.70 (565nm)
7	19.26	10.60 (590nm)
10	18.99	10.10 (645nm)
20	19.54	9.40 (690nm)
30	20.06	9.00 (720nm)

Since the dilution of the sol-gel prepared solutions with ethanol was reported in the literature [16, 17], attention was given to the effect of ethanol dilution on the reflection properties of the resulted thin films. The SiO₂-Sol is the optimized solution in terms of smooth and uniform coatability. The effect of further dilution of the SiO₂-Sol with ethanol (Sol:Ethanol in volume, 1:1, 1:2, 1:5, 1:10) on the antireflection behavior can be seen in figure 4. Reflectance of the c-Si surface was increased by adding additional ethanol into the solution comparing with that of the SiO₂-Sol. Moreover, the minimum reflection point was apart from the desired visible region. Therefore, no improvements could be observed at the end of further dilution of the SiO₂-Sol.

Based on the results, annealing temperature of 950 °C for 7 min in air was determined to be the optimum conditions for the prepared SiO₂ thin films. As seen in figure 4, the average reflectance value decreased from approximately 40% (bare c-Si surface) to 19.26% with the coating of SiO₂ thin film on the c-Si surface.

Figure 5 shows morphology of the optimum SiO₂ thin film formed on c-Si substrate with coating speed of 4000 rpm and annealed at 950 °C for 7 minutes. It is observed that the nanostructured SiO₂ film has been formed and possesses a porous structure.

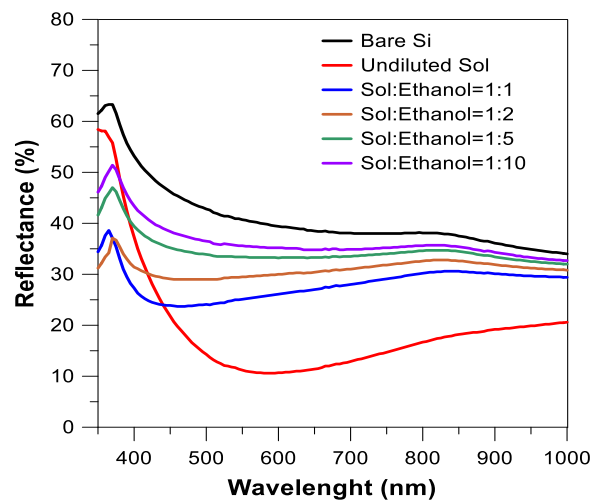


Figure 4. Reflectance spectra of the c-Si surface depending on the further dilution of SiO₂-solution

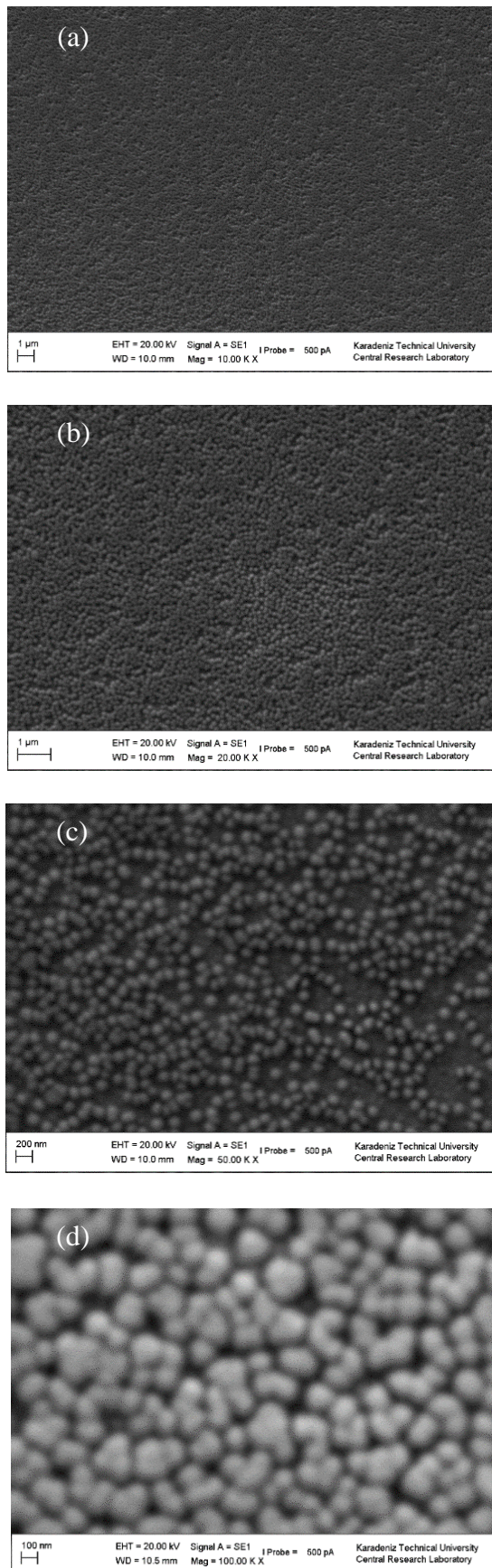


Figure 5. SEM micrographs of SiO₂ thin films a) ×10k, b) ×20k, c) ×50k, d) ×100k

The average reflectance values obtained from the optimum SiO₂/c-Si structures were applied to the c-Si solar cell as ARL by using Afors-het simulation [18]. For c-Si

solar cell, some initial parameters including emitter thickness, emitter doping concentration, base thickness, base doping concentration, back surface field (BSF) thickness, BSF doping concentration, series resistance, and parallel resistance were determined in Afors-het software (Table 4). I-V graph for c-Si solar cell with or without SiO₂ coating as an anti-reflection coating is shown in figure 6 with the characteristic parameters of solar cells given in Table 5.

While the power conversion efficiency (PCE) of the solar cell without ARL coating was determined as 11.68%, the efficiency of the solar cell with reduced reflection was obtained as 15.91%. A 4% increase in efficiency was achieved as a result of an increase in the short-circuit current density (J_{sc}) from 20.87 to 28.33 mA/cm² and an increase in the open-circuit voltage (V_{oc}) from 685.1 to 693.40 mV due to a decrease in reflection on the silicon surface. Similar impacts of the increase of J_{sc} and V_{oc} on PCE has also been reported elsewhere practically [19, 20], which confirms the importance and great impact of ARLs on the performance of solar cells.

Table 4. Some initial parameters set in Afors-het for c-Si solar cell

Emitter thickness (μm)	0.3
Emitter doping concentration (cm ⁻³)	1.0×10 ²⁰
Base Thickness (μm)	200
Base doping concentration (cm ⁻³)	1.5×10 ¹⁶
BSF doping concentration (cm ⁻³)	1.0×10 ¹⁹
BSF thickness (μm)	7
Series resistance (Ω cm ²)	0.8
Parallel resistance (Ω cm ²)	10000

Table 5. Characteristic parameters of SiO₂ coated and uncoated solar cell as ARL

	J_{sc} (mA/cm ²)	V_{oc} (mV)	FF (%)	Eff (%)
non-ARL	20.87	685.10	81.64	11.68
with-ARL	28.33	693.40	81.01	15.91

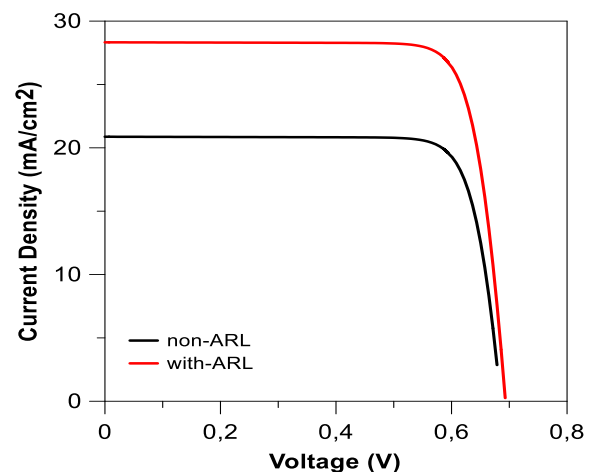


Figure 6. I-V graph of an uncoated and SiO₂ coated solar cell

4. Conclusion

In this study, SiO₂ based spin coating processed ARL thin films were introduced. ARL effect of the prepared SiO₂ films were provided with the recipe of the optimum solution and the optimum processing conditions. Average reflection (350 – 1000 nm) of c-Si surface could be reduced from 40.5% to 19.26%; the minimum of it could be <10% depending on the process. Simulations related to the effect of ARL on c-Si solar cell device were also shown by increasing the PCE of a solar cell from 11.68% to 15.91%. These results can contribute to the development of solar cells through solution-based, non-vacuum, and simple spin-coating processing.

Declaration

The authors declared no potential conflicts of interest with respect to the research, authorship, and/or publication of this article. The authors also declared that this article is original, was prepared in accordance with international publication and research ethics, and ethical committee permission or any special permission is not required.

Author Contributions

I. Kanmaz and A. Üzüm conceived the study together. I. Kanmaz conducted the experiments, analyzed the data and drafted the paper. A. Üzüm supervised the research, contributed the data analyze and revised / finalized the paper.

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