Investigation of an Antireflective Coating System for Solar Cells based on Thin Film Multilayers

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ABSTRACT

The optical loss due to reflection is a significant barrier to the quantum efficiency of solar cells. In this work, an antireflective coating based on multilayers of metal oxides (TiO_2, SiO_2, ZnO) was prepared with the spin coating method. The coatings' antireflective, hydrophobic, and photocatalytic properties were examined. Based on the requirements met by the refractive index, a methodical selection of material and thickness for each layer was made in order to achieve near-zero reflection. The performance of different coating systems was examined by evaluating the percentage transmittance in the visible light range (400 nm - 800 nm). The optical properties of the obtained samples were studied with regard to transmittance and reflectance. The surface wettability of antireflective coating films was assessed by measuring the Water Contact Angle (WCA). The photocatalytic characteristics were evaluated by analyzing of the degradation of 0.02 mM Methylene Blue (MB) solutions after sunlight exposure for varying durations at midday.

Keywords-antireflective coating; multilayer; photocatalytic property; solar cell

I. INTRODUCTION

Energy is essential for human progress, and throughout history, human civilization has continuously sought sustainable energy sources [1]. Solar energy is one of the most important renewable energy sources. It enables abundant photons to be efficiently transformed into electricity while minimizing carbon emissions [1–3]. A significant percentage of solar radiation is reflected from the silicon solar cell surface and does not contribute to the generation of the electron-hole pairs, which reduce the energy conversion efficiency [4-5]. The optical loss from reflection is a significant barrier to solar cells' quantum efficiency. Therefore, various solutions have been suggested to mitigate the optical loss induced by high reflectivity and boost the energy capture potential of solar cells. These solutions encompass the application of Antireflective Coatings (ARCs), surface texturization, or a synergistic combination of both approaches, aiming to extend the optical path of light within solar cells [6-7]. These layers work as anti-reflection and self-cleaning coatings on photovoltaic panels. The goal of creating antireflection films is to get the substrate that has one coated on it as close to the refractive index of the incident medium as possible without losing transmittance. Several materials have been used as ARCs such as MgF_2, SiO_2, TiO_2, ZnO, Al_2O_3, ZnS and $BaTiO_3$ [8- 15]. The anti-reflection and self-cleaning coatings films can be made by single layer [6, 16-17] or multilayers [7-9]. A single-layer anti-reflection coating minimizes reflections at a specific wavelength, usually under normal incidence. However, this limitation restricts its effectiveness across the broad visible spectrum. Additionally, these coatings are unable to achieve broad-spectrum reduction in reflectance across a wide range of incident angles. To overcome these limitations, the multilayer anti-reflection design holds greater promise because it eliminates the need for materials with a refractive index lower than glass [8, 18]. Titanium dioxide (TiO_2) and Silicon dioxide (SiO_2) are the most commonly used materials for multilayer anti-reflection coatings because their higher transparent properties [9, 19]. TiO_2 and Zinc oxide (ZnO) show excellent optical transmittance in visible and near infra-red wavelength regions [20-21]. Creating multi-layer coatings with different materials and optimized layer thickness presents a challenging task. The complexity involved in the fabrication process necessitates careful consideration of material selection and layer design [22]. This work follows a similar approach, focusing on enhancing antireflective coatings based on $SiO_2/TiO_2/ZnO$ triple layer ARCs. Multilayers were prepared using sol-gel spin coating technique. Optical and photocatalytic properties of triple layer ARCs were investigated.

II. EXPERIMENTAL DETAILS

A. Materials

Hydrochloric acid (HCl, 37%), absolute ethanol (EtOH, 99.9%), zinc acetate dihydrate [Zn $(CH_3COO)_{22}H_2O$], titanium butoxide (TBOT, $Ti(OBu)_4$), tetraethyl orthosilicate (TEOS, $Si(OC_2H_5)_4$), acetic acid (CH_3COOH), methanol (CH_3OH), $Si(OC_2H_5)_4$: CH_3OH: CH_3COOH, and $Ti(OBu)_4$: CH_3OH: CH_3COOH: DI and deionized water were used.

B. Cleaning Substrate

Prior deposition, the substrates were ultrasonically cleaned with deionized water, acetone, and hydrochloric acid (pH = 1) sequentially for 15 min to enable the film to stick to the substrate steadily.

C. Preparation of the ZnO Layer

The ZnO layer was prepared by the sol-gel spin coating method onto a glass substrate using zinc acetate dihydrate [Zn $(CH_3COO)_{22}H_2O$] as a source of Zn. A necessary quantity of zinc acetate was dissolved in absolute ethanol and m-cresol to create the precursor solution (0.2 M), which was then rapidly agitated at 65 °C for 2 h to produce a precise and uniform solution. The produced solution was allowed to stand at room temperature for 24 h in an airtight beaker to increase its homogeneity before being ready for deposition. The solution was deposited onto a pre-cleaned glass substrate by a single wafer spin coater. Following the substrate's placement on the spin coater's substrate holder, 0.7 ml of coating solution was dropped and spin-coated at 2000 rpm for 30 s in an air and dried on a hot plate at 180 °C for 5 min. The above-mentioned process was performed five times to get the required ZnO film thickness.

D. Preparation of TiO_2 and SiO_2 layers

Firstly, 1.5 ml of acetic acid were added to methanol (25 ml) and stirred for 10 min with a magnetic stirrer at ambient temperature. The titanium butoxide solution was added drop by drop, and stirred for 2 h at room temperature. After being aged for 24 h, the TiO_2 solution became transparent. Then, 25 m of methanol ml and 1.5 ml of acetic acid were combined, and the mixture was swirled for 10 min while 2.5 ml of tetraethyl orthosilicate was added dropwise every minute. The prepared SiO_2 solution was mixed continuously for 2 h and aged 24 h. TiO_2 and SiO_2 layers were deposited using spin-coater at 3000 rpm for 30 s. After the coating, thin film samples were annealed at 600 C (TiO_2) and 500 C (SiO_2) for 1 h.

E. Procedure

To attain the desired anti-reflection outcome within the spectral range of 400-800 nm, it is necessary for the reflected beam's amplitude at both the air-coating interface and the coating-substrate interface to be identical and precisely out of phase by one-half wave (180°), leading to destructive interference. This will effectively eliminate any net reflection. To satisfy this requirement, the anti-reflection coating layer's optical thickness (t) needs to be approximately one-fourth of the wavelength (λ) at which zero reflectance is desired [23]. The sketch of the multilayer stack necessitates the optimization of the thicknesses of each individual layer, taking into consideration the quarter-wavelength principle [24]. Therefore, we consider two samples, S1 and S2, in which the layer thickness was quarter-quarter-quarter (λ/4 - λ/4 - λ/4) wavelengths for sample S1 and quarter-half-quarter (λ/4 - λ/2 - λ/4) wavelengths for sample S2. The wavelength λ was selected to be λ = 550 nm which corresponds to the highest intensity of the solar spectrum. The thickness of each layer was accurately controlled by the spin coating speed. Schematic illustration and refractive index profiles of $SiO_2/TiO_2/ZnO$ ARCs are shown in Figure1.

F. Characterizations

The T(λ) spectrum, which represents direct transmittance at normal incidence, was predominantly obtained using a PerkinElmer Lambda 1050 spectrophotometer across the wavelength range of 400–800 nm. The measurement utilized a step length and slit width of 5 nm. To study the chemical structure of the layers, FTIR measurements were carried out on a Fourier transform infrared spectrometer. (FT-IR, IS10, Themo Scientific). Thin film thickness and refractive index and were measured by a spectroscopic ellipsometry (SENTECH SE8000PV). The surface wettability of ARC films was assessed by measuring the water contact angle (wca) using a CAM 200 Optical Contact Angle Meter from KSV Instruments. The sessile drop method was utilized, involving the careful placement of a 6 µl droplet of distilled, deionized water on the surface using a micro syringe. Subsequently, images were captured to determine the angle formed at the liquid/solid interface. The photocatalytic properties were studied by evaluating the decomposition of 0.02 mM MB solution samples after they were retained in a sunbath for different periods (2 h, 4 h and 6 h) at noon.

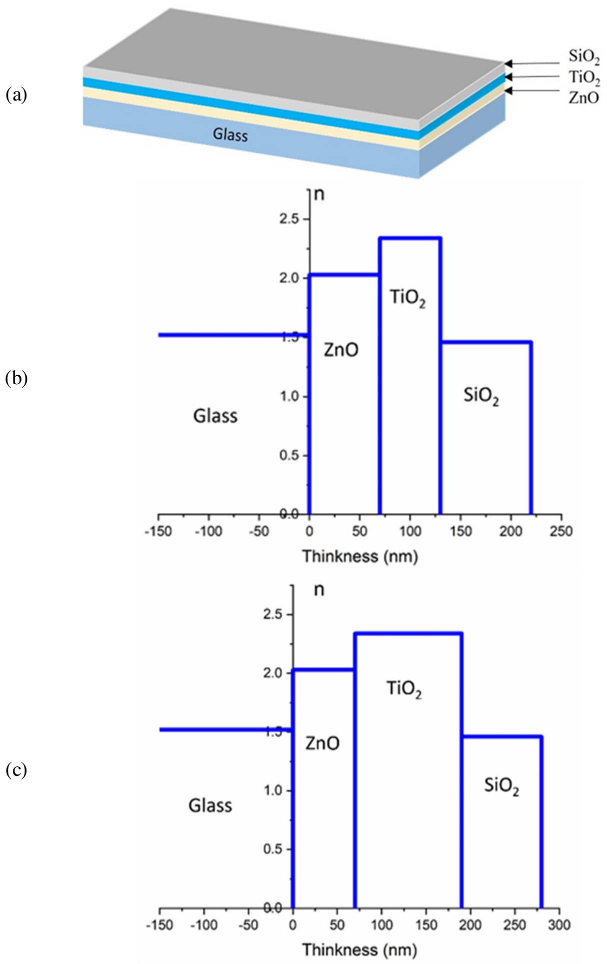

Fig. 1. (a) Schematic illustration of $SiO_2/TiO_2/ZnO$ ARCs. (b) and (c) Refractive index profile of samples (a) S1 and (b) S2.

TABLE I. REFRACTIVE INDEX AND LAYER THICKNESS OF THE STUDIED SAMPLES

Sample		S1	S2
Layer	**Refractive index**	**Layer thickness**	**Layer thickness**
SiO_2	1.46	90nm	90nm
TiO_2	2.34	60nm	120nm
ZnO	2.03	70nm	70nm

III. RESULTS AND DESCUSSION

Fourier Transform Infrared (FTIR) measurement, a non-destructive technique, is employed to investigate the vibrational behavior of bonds in organic molecules as well as the metal-oxygen bonds in metal oxides. Figure 2 depicts the FTIR spectrum of $SiO_2/TiO_2/ZnO$ triple-layer coating in the wavenumber range 400–4000 cm^{-1}. As we can see, the two samples exhibit almost the same absorption peaks. The band located between 457 cm^{-1} and 487 cm^{-1} corresponds to Zn–O vibration mode which approves the formation of the ZnO layer. The spectra exhibit other peaks around 975 cm^{-1} corresponding to the vibration of Si-O-Ti, a peak at 1000 cm^{-1} which can be attributed to the stretching vibration of Ti-O. Additionally, there is a peak at 1126 cm^{-1} attributed to Si-O-Si. Furthermore, there are broad peaks at 3430 cm^{-1}, which are associated with the stretching mode of water and hydroxyl groups [25-26].

Figure 3 presents the transmittance spectra of $SiO_2/TiO_2/ZnO$ triple-layer ARCs at normal incidence as well as the recorded transmittance and reflectance spectrum of glass substrate. The prepared triple-layer broadband anti-reflective coating demonstrated a marked improvement in transmittance levels compared to uncoated glass. The uncoated glass reveals a transmittance about 93%. It can be obvious that S1 and S2 exhibits excellent broadband antireflection ability in the wavelength range of 400-800 nm. Sample S1 (90 nm SiO_2 / 60 nm TiO_2 / 70 nm ZnO) shows a high transmittance at 450-600 nm with an average transmittance at that region around 99.11% which can be considered as a good result. However, sample S2 (90 nm SiO_2 / 120 nm TiO_2 / 70 nm ZnO) reveals an average transmittance about 95.58% in this region. At the entire visible region, the average transmittance was 96.04% for sample S1 and 97.35% for sample S2. Similar result has been obtained on simulation investigation using these materials ($SiO_2/TiO_2/ZnO$) [27]. Table II presents a comparison of our results with those obtained using both similar and different materials. As we can see, the proposed ARC multilayer exhibits an excellent optical property compared to other samples. This result indicates that applying three layers of coating would be optimal for enhancing the antireflection characteristics of photovoltaic glass modules across a wide spectrum of visible light.

TABLE II. COMPARISON OF OUR RESULTS WITH THOSE OBTAINED USING SIMILAR AND DIFFERENT MATERIALS

Materials	Transmittance	Range	Reference
$SiO_2/TiO_2/ZnO$	99.11% (S1) 95.58% (S2)	450-600 nm	Our work
ZnO /SiO_2	96.10%	300–1200 nm	[7]
$SiO_2/TiO_2/ZnO$ (simulation)	99.5%	450-700 nm	[9]
MgF_2/SiO_2	98.89%	400-800 nm	[12]
TiO_2/ZnO	94.71%	450-600 nm	[20]
Porous ZnO/SiO_2/TiO_2	98.56%	350-700 nm	[32]

The presence of dust and dirt particles on the surface of the Anti-Reflective (AR) coated glass leads to a reduction in the transparency of the covers, sometimes even more so than with uncoated glass. This decrease in cover transparency results in a reduction in the amount of solar radiation that reaches the absorber, ultimately leading to a decline in the efficiency of the solar plant. Therefore, development of films that possess both high transparency and self-cleaning capabilities appears to be a promising solution. The effectiveness of hydrophobic ARC in solar cells extends beyond their ability to repel water. A highly hydrophobic surface can provide self-cleaning properties to the ARC by enabling water to roll off and remove contaminants from the surface [28]. Hydrophobicity is typically evaluated through water contact angle measurements. When the value of the water contact angle increases, it signifies a greater hydrophobicity of the surface.

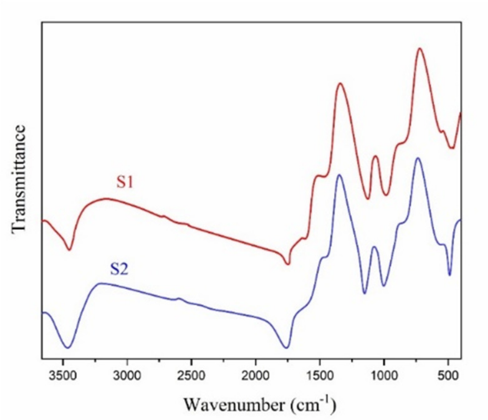

Fig. 2. FTIR spectra of samples S1 and S2.

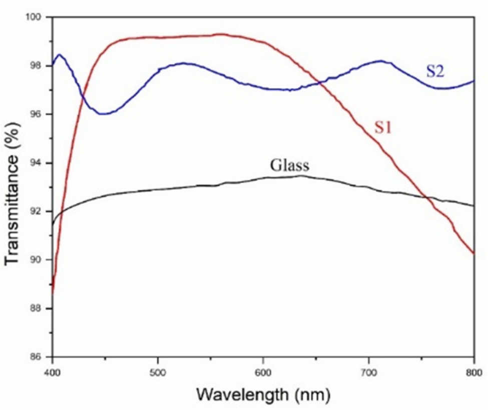

Fig. 3. Transmittance spectra of $SiO_2/TiO_2/ZnO$ triple-layer ARC.

To enhance the hydrophobicity of the ARC, the surface was treated with hexamethyldisilane (HMDS), resulting in significantly improved self-cleaning properties. The Water Contact Angles (WCAs) were assessed at five distinct locations on each sample surface to estimate any potential measurement errors. Figure 4 shows the evolution of WCA of the $SiO_2/TiO_2/ZnO$ triple-layer broadband coating as a function of HMDS concentration. As we can see, the HMDS treatment significantly enhances the WCA. The observed phenomenon can be explained by the fact that as the concentration of HMDS increases in hexane, the number of $–OSi(CH_3)_3$ groups attached to the silica surface also increases [29]. Consequently, this leads to a significant rise in the WCA, from an initial value of 38.61° to a final value of 92.77°.

The photocatalytic performance of ARCs films is considered a crucial point in the self-cleaning of solar cell panels which enhance and maintain its efficiency [30]. To investigate the photocatalytic properties, the uncoated glass and the triple-layer broadband coating slides 2 cm^2 in size were flooded in the 50 mL MB. The initial concentration of MB in the solution was 0.02 mM. To study the catalytic activity, the degradation of MB was carried out under direct sunlight for different periods (2, 4, and 6 h). Figure 5 depicts the change in the absorbance spectra of MB versus the wavelength in different period time. The absorption peaks were at 655 nm, and the peak intensity decreased regularly with the increase period of exposure. In simpler terms, as the period time increased, the degradation of MB also increased [31]. This implies that the coatings might have self-cleaning properties, potentially removing other organic contaminants from the surface of PV panels. Ultimately, this could lead to improved efficiency in utilizing solar light.

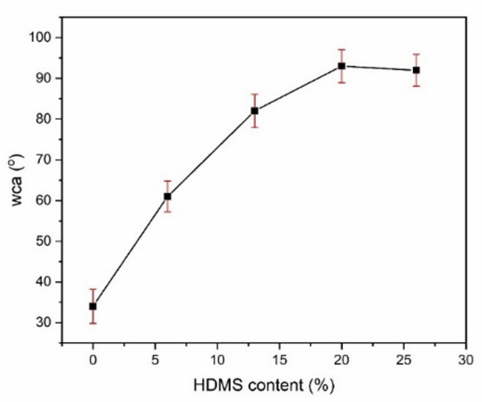

Fig. 4. Water contact angle versus concentration of HMDS.

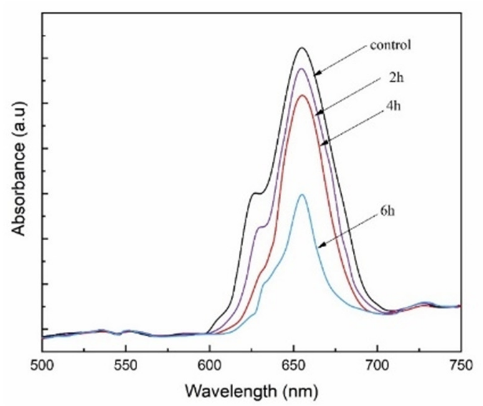

Fig. 5. Variation of absorbance spectrum of methylene blue with wavelength.

IV. CONCLUSION

A multilayer antireflective coating (ARC) has been designed for the 400 nm-800 nm wavelength range. The coating consists of three layers and has been prepared by sol-gel spin coating on glass substrates. The coated structure reveals an excellent antireflective, hydrophobic, and photocatalytic properties. The triple layer ARC reveals a high transmittance at 450-600 nm wavelength range. The hydrophobicity of the ARCs has been enhanced significantly by treating the surface with HDMS resulting in significantly improved self-cleaning properties. Therefore, the proposed ARC films combine both properties, i.e. high transmittance and self-cleaning, which can significantly improve photovoltaic solar cells' efficiency.

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