

Design of multi-layer anti-reflection coating for terrestrial solar panel glass

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Abstract. To date, there is no ideal anti-reflection (AR) coating available on solar glass which can effectively transmit the incident light within the visible wavelength range. However, there is a need to develop multifunctional coating with superior anti-reflection properties and self-cleaning ability meant to be used for solar glass panels. In spite of self-cleaning ability of materials like TiO_2 and ZnO , these coatings on glass substrate have tendency to reduce light transmission due to their high refractive indices than glass. Thus, to infuse the anti-reflective property, a low refractive index, SiO_2 layer needs to be used in conjunction with TiO_2 and ZnO layers. In such case, the optimization of individual layer thickness is crucial to achieve maximum transmittance of the visible light. In the present study, we propose an omni-directional anti-reflection coating design for the visible spectral wavelength range of 400–700 nm, where the maximum intensity of light is converted into electrical energy. Herein, we employ the quarter wavelength criteria using SiO_2 , TiO_2 and ZnO to design the coating composed of single, double and triple layers. The thickness of individual layers was optimized for maximum light transmittance using essential McLeod simulation software to produce destructive interference between reflected waves and constructive interference between transmitted waves.

Keywords. Anti-reflection; thin film; thickness; refractive index; interference; optical impedance.

1. Introduction

In photovoltaic (PV) module, the cover glass surface reflects more than 4% of incident light across the spectrum which needs to be effectively utilized for energy conversion. Additional loss due to soiling has become more common issue in substantial polluted areas due to dust coverage on solar panels [1]. Presence of microdepressions leads to the accumulation of dust and rain water droplets over the glass surface causing corrosion in solar panels. The optical parameters are affected by soiling effects which subsequently reduces the energy generation due to low light transfer thereby causing additional loss in the overall efficiency (1.17–1.42%) [2]. Therefore, under such circumstances, regular cleaning of solar modules becomes indispensable which might lead to abrasive cleaning of glass resulting into further development of scratches on the glass panels. To overcome these issues, glass surface properties can be modified by applying a suitable protective anti-reflective (AR) coating which possesses high transparency to visible light and self-cleaning ability.

However, the biggest challenge of this approach is the availability of material with refractive index lower than that of glass ($n < 1.5$). An ideal single-layer AR coating on a

glass surface in air ambient would require a material with a refractive index of 1.2 and there is no single material available with low refractive index. However, reflective losses can be minimized with single-layered coating of thickness equal to quarter of wavelength causing destructive interference of the reflected light waves from coated surface and substrate. Generally, magnesium fluoride (MgF_2) is used for single layer AR due to low refractive index of 1.14, however, less durable when used in outdoor environment [3]. Alternatively, porous SiO_2 is preferred due to reasonably low refractive index ($n < 1.45$) with good environmental stability and durability. Recently, authors have demonstrated improvement in light trapping in Si solar cells with maximum reduction in reflection by 85–100% in visible region of 400–550 nm when amorphous BaTiO_3 thin film with refractive index of 2.02–1.91 were deposited over Si solar cells [4]. However, single layer anti-reflection coating can minimize reflections at one specific wavelength typically at normal incidence which limits its use for broad visible range. In addition, they are inherently incapable to exhibit spectrally broadband reduction in reflectance over a wide range of angles of incidence.

Recently, multilayer AR design seems to be more promising due to the fact that material with refractive index lower than glass is no longer required [5,6]. Through proper selection of the material and layer thickness, the interference of light can be controlled by minimizing the reflection losses.

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Two or more materials of low (1.45–1.52) and high (2.0–2.3) refractive indices can be preferred depending on the level of anti-reflection required for specific application. In turn, these high refractive indices layers used as sequential layers along with low refractive index material is known to enhance the transparency of glass.

Particularly, TiO₂ and ZnO thin films have a wide optical band gap of 3.05 and 3.3 eV, respectively, shows excellent optical transmittance in visible and near infra-red region. In spite of self-cleaning ability of TiO₂ and ZnO, these coating on glass substrate has tendency to increase the reflectivity. To improve the anti-reflective property, a low refractive index material like SiO₂, which would enhance the light transmission has to be used in conjunction with TiO₂ and ZnO layers. Ye *et al* [7] developed abrasion resistant, hydrophobic SiO₂/TiO₂/SiO₂-TiO₂ triple coating with high transmittance of 98.4% at visible region using dip-coating method. Similarly, Li *et al* [8] designed and developed highly durable non-quarter wave double-layer TiO₂-SiO₂ and ZrO₂-SiO₂ using sol-gel technique which revealed transmittance up to 94.4 and 94.31%, respectively. Gao *et al* [9] developed highly transparent and UV-resistant superhydrophobic arrays of SiO₂-ZnO nanocomposite, coated on glass substrates showing minimal impact on solar cell device performance under AM1.5G illumination.

Self-cleaning phenomenon is based on the contact angle θ_c , if θ_c approaching to 0° it is known as super hydrophilic and if θ_c close to 150° then known as super hydrophobic. Hydrophilic coating carries away the dirt and other impurities with water droplets, whereas in case of hydrophobic coating the water droplets slide and roll over the surfaces thereby cleaning them. However, to achieve self-cleaning, a film not only must be super hydrophilic but also must be able to induce photocatalytic oxidative decomposition. The hydrophilic coatings doped with suitable metal oxides, chemically breaking down the complex dirt deposits by sunlight-assisted cleaning mechanism [10].

The self-cleaning ability of TiO₂ and ZnO coatings applied on solar glass are well studied. Zhang *et al* [11] reported the nanosized TiO₂ particles coated on the surface of the SiO₂ layer shows excellent surface wettability with ~1° contact angle and 99% transmission despite the high refractive index of TiO₂. Medina-Valtierra *et al* [12] demonstrated that the doping of metal in TiO₂ enhancing the self-cleaning property of nanostructured thin film. Ebert and Bhusan [13] studied self-cleaning and transparent properties of SiO₂, ZnO and ITO nanoparticle coatings on glass and plastic substrates (polycarbonate and PMMA) and found superhydrophobic nature θ_c (>150°) and high transmittance of visible light ($T \sim 90\%$) in terms of wettability and transmittance.

Development of multi-layer coating with optimized layer thickness using analytical approach is a difficult challenge and adds complexity to the fabrication process. Therefore, great emphasis should be laid on selection of materials and design of layers is crucial aspect prior to methodical evaluation of the coating for the desired requirement. With this aim, we intend to design multifunctional coatings with

superior anti-reflection properties in broadband range using cost-effective and high refractive index materials like TiO₂ and ZnO. In addition, these materials possess high self-cleaning ability to modify the surface wettability of the glass substrates which is another important parameter for reducing the reflection losses due to soiling. The design of the anti-reflection coating was made to achieve minimum reflectance and maximum transmittance in the wavelength range of 400–700 nm using Essential Macleod commercial software package [14]. This work is focussed on the development of a single, double and triple layers broadband designs. Therefore, the efficiency of PV module can be improved by depositing sequential layers of metal oxides possessing anti-reflection properties in broadband range.

2. Methodology

To achieve an optimal anti-reflection effect resulting in zero net reflection in the spectral range of interest 400–700 nm, the amplitude of the reflected beam at the air-coating interface and coating-substrate interface has to be equal and exactly one-half wave (180°) out of phase, resulting in destructive interference. This condition is met by the optical thickness of the anti-reflection coating layer (t) must be close to a quarter of the wavelength (λ) at which zero reflectance should occur (i.e., the reflected wave must be out of phase by π) expressed by equation (1):

$$t = \frac{\lambda}{4n_1} + m \frac{\lambda}{2n_1} \quad (m = 0, 1, 2, 3 \dots), \quad (1)$$

where n_1 is the refractive index of the thin-film layer [15]. Second condition is that the magnitude of front surface reflections must be equal to the magnitude of the sum of all other partial reflections in the thin-film layer. This condition can be met when refractive index of the single layered coating (n_1) equals to the square root of the product of the refractive indices of substrate (n_s) and medium (n_0) expressed by equation (1) [16]:

$$n_1 = \sqrt{n_s n_0}. \quad (2)$$

However, the region of low reflectivity for a single-layer optical coating occurs at thickness of $\lambda/4n_1$ which cannot cover the entire absorption width over a 400–700 nm visible range. It is often difficult to choose intermediate index materials with refractive indices that are close to ideal. To obtain the equivalent optical properties of an intermediate-index layer, a stack consisting of alternating layers of low- and high-index materials are used. The optimal performance is obtained when the refractive index of each layer is the geometric mean of the refractive indices of its two neighbouring layers. Each layer adds to a minimum to the reflectance spectrum, which can be used to increase the absorption width of the low reflectivity region. The multilayer stack design requires optimization of the individual layer thicknesses based on quarter-wavelength.

In this study, we use Essential Mcleod software which models the optical coating using the transfer matrix method to predict propagation of electromagnetic wave through the thin film stack. Optical interference matrix is an effective way to calculate reflectivity in multilayer thin film. Under the situation of zero degree incident angle of light, considering the optical thin film system of N layers, n_j is the refractive index and k_j the extinction coefficient, d_j the thickness of each layer, respectively, and n_0 the refractive index of air ($n_0 = 1$). The reflectance in multilayers is calculated by the optical admittance (Y) which is the ratio of the total tangential magnetic (C) and electric fields (B). According to the refractive index and thickness of each layer, interference matrix of each layer can be determined using characteristic matrix formulation with the following equation (3) [17]:

$$\begin{bmatrix} B \\ C \end{bmatrix} = \left\{ \prod_{r=1}^q \begin{bmatrix} \cos \delta_r & (i \sin \delta_r)/n_r \\ i n_r \sin \delta_r & \cos \delta_r \end{bmatrix} \right\} \begin{bmatrix} 1 \\ n_m \end{bmatrix}, \quad (3)$$

where $\delta_r = 2\pi N d \cos \vartheta / \lambda$ is the effective optical thickness of the layer at a given wavelength and n_m is the substrate refractive index. If q is the layer next to the substrate, then the order is

$$\begin{bmatrix} B \\ C \end{bmatrix} = [M_1][M_2] \dots [M_q] \begin{bmatrix} 1 \\ n_m \end{bmatrix}, \quad (4)$$

M_1 indicates the matrix associated with layer 1, and so on. As in the case of a single surface, n_o must be real for reflectance and transmittance should have a valid meaning. Using equations (3) and (4), reflectance (R) is derived to be:

$$R = \left(\frac{n_0 B - C}{n_0 B + C} \right) \left(\frac{n_0 B - C}{n_0 B + C} \right)^* . \quad (5)$$

In this study, anti-reflection coating design was optimized using SiO_2 , ZnO and TiO_2 layers to minimize the single surface reflection on glass for wavelength in the range of

Table 1. Coating design.

	Design	Materials	Refractive index	Extinction co-efficient	Coating thickness (nm)
1	Medium	Air	1.00000	0.00000	—
	Substrate	Glass	1.52083	0.00000	
2	Medium	Air	1.00000	0.00000	
	Layer 1	SiO_2	1.46180	0.00000	87.93
	Substrate	Glass	1.52083	0.00000	
3	Medium	Air	1.00000	0.00000	
	Layer 1	SiO_2	1.46180	0.00000	87.22
	Layer 2	TiO_2	2.34867	0.00000	54.29
	Substrate	Glass	1.52083	0.00000	
4	Medium	Air	1.00000	0.00000	
	Layer 1	SiO_2	1.46180	0.00000	87.22
	Layer 2	ZnO	2.03718	0.00000	62.59
	Substrate	Glass	1.52083	0.00000	
5	Medium	Air	1.00000	0.00000	
	Layer 1	SiO_2	1.46180	0.00000	87.22
	Layer 2	ZnO	2.03718	0.00000	125.17
	Layer 3	TiO_2	2.34867	0.00000	54.29
	Substrate	Glass	1.52083	0.00000	
6	Medium	Air	1.00000	0.00000	
	Layer 1	SiO_2	1.46180	0.00000	87.22
	Layer 2	ZnO	2.03718	0.00000	62.59
	Layer 3	TiO_2	2.34867	0.00000	54.29
	Substrate	Glass	1.52083	0.00000	
7	Medium	Air	1.00000	0.00000	
	Layer 1	SiO_2	1.46180	0.00000	87.22
	Layer 2	TiO_2	2.34867	0.00000	108.99
	Layer 3	ZnO	2.03718	0.00000	62.59
	Substrate	Glass	1.52083	0.00000	
8	Medium	Air	1.00000	0.00000	
	Layer 1	SiO_2	1.46180	0.00000	87.22
	Layer 2	TiO_2	2.34867	0.00000	54.29
	Layer 3	ZnO	2.03718	0.00000	62.59
	Substrate	Glass	1.52083	0.00000	

400–700 nm. The values of refractive indices for SiO_2 , ZnO and TiO_2 were selected using materials library database. To obtain the optical spectra, the reference wavelength was fixed at 510 nm with normal angle of incidence. To understand the effect of incident angle on anti-reflection properties, the angle of incidence was varied from 0 to 90° , while designing the layer thickness and subsequently reflection and transmittance spectras were simulated. Table 1 shows the details of the design parameters chosen for the present investigation.

3. Results and discussion

The reflectance and transmittance spectras were plotted as show in figure 1a and b, respectively. The uncoated glass covered with thin layer of air shows single-side reflectance of about 8% and transmittance of about 92% in wavelength range of 400–700 nm. For a single layered SiO_2 coating, the thickness obtained for reducing reflection to 3.0–3.2% is 87.93 nm with marginal increase in transmittance to 96.8–97.1%. For designs 3 and 4, the double layer

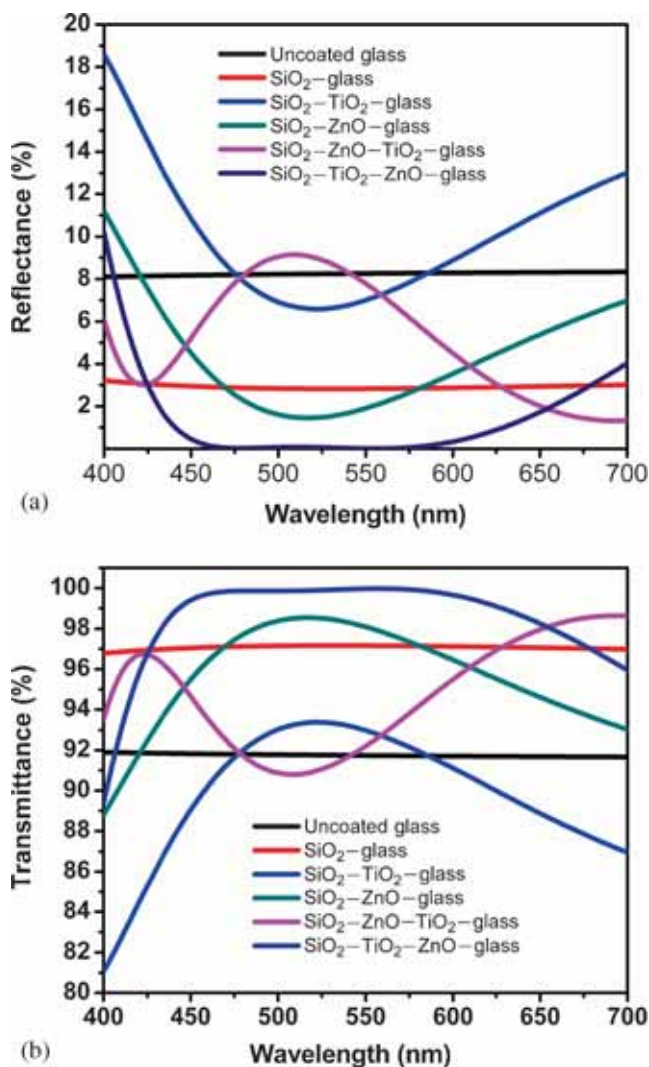


Figure 1. The modelled performance of single, double and triple layer coatings.

coating thickness obtained was based on quarter-quarter (Q-Q) wavelength theory. The surface reflection increased to 13.0–19.0%, while transmittance was decreased to 81–93.37% for simulated double layer coating comprising of 87.22 nm SiO_2 and 54.29 nm TiO_2 on glass substrate. Interestingly, double layer coating composed of 87.22 nm SiO_2 and 62.59 nm ZnO on glass, yielded a relatively lower reflectance of 7–11.5% and high transmittance of 89–98.45% in entire 400–700 nm wavelength range. This is explained by the Schuster diagram shown in figure 2 with SiO_2 as the low index of 1.46, while an upper bound range of refractive indices for TiO_2 and ZnO of 2.34 and 2.03, respectively. The area of shaded region shows the possible values of film indices for a quarter-quarter coating composed of SiO_2 – TiO_2 and SiO_2 – ZnO on glass. The narrow range of indices in case of SiO_2 – ZnO , contributes to the decrease in the reflectivity when compared to SiO_2 – TiO_2 coating.

However, in both these cases typical ‘V’ shaped characteristic curve was observed signifying that the anti-reflection property can be realized only at narrow wavelength range. The reflectance value was lowest for SiO_2 – ZnO double layer when compared to single SiO_2 layer and SiO_2 – TiO_2 double layer. In case of triple layer coating comprised of 87.22 nm SiO_2 –62.59 nm ZnO –54.29 nm TiO_2 (design 6) on glass, the reflectance increases to 9% at 510 nm which is more than the uncoated glass. However, triple coating composed of 87.22 nm SiO_2 –54.29 nm TiO_2 –62.59 nm ZnO coating (design 8) on glass showed a remarkable decrease in reflectance of <1% with high transmittance of 99% in the broad range of wavelength from 450 to 700 nm. This suggests that triple coating would be ideal for improving anti-reflection properties of PV glass modules in the broad range of visible portion of incident light.

Single layer SiO_2 coating on glass benefited with lowering the reflectance to 3% and transmittance to ~97% in

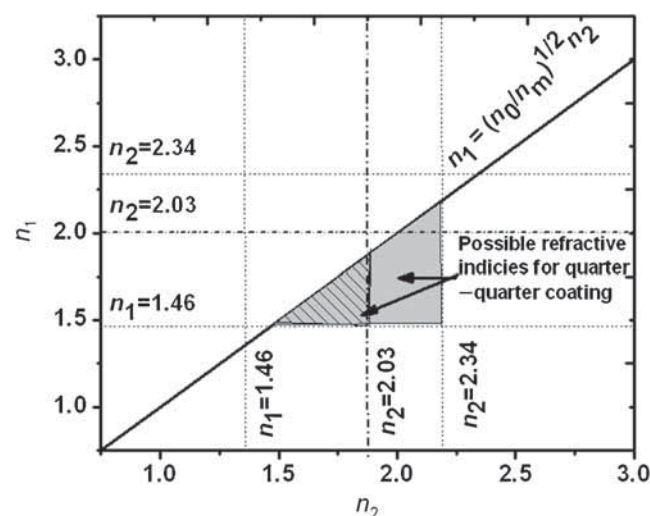


Figure 2. A Schuster diagram showing possible refractive indices for quarter-quarter wavelength coating composed of SiO_2 ($n_1 = 1.46$) – ZnO ($n_2 = 2.03$) and SiO_2 ($n_1 = 1.46$) – ZnO ($n_2 = 2.34$) on glass.

wavelength of 400–700 nm. Inclusion of TiO_2 with SiO_2 in double layer has resulted minima at wavelength 520 nm but resulted in higher reflectance towards lower and higher wavelengths, while inclusion of ZnO with SiO_2 resulted in decrease in reflectance and increment in transmittance in relatively broad wavelength range of 470–570 nm. Triple layer $\text{SiO}_2(1.46)/\text{ZnO}(2.03)/\text{TiO}_2(2.34)$ stack with $n_{\text{air}} < n_{\text{SiO}_2} < n_{\text{ZnO}} < n_{\text{TiO}_2} > n_{\text{glass}}$ showed two minima in reflection (first at 420 nm and second at 700 nm) and one maxima at 520 nm which does not meet the requirements of minimum reflection in 450–650 nm wavelength corresponding to maximum photon flux in the solar spectrum on earth. It was observed that when second and third layer was interchanged

in the stack i.e., $n_{\text{air}} < n_{\text{SiO}_2} < n_{\text{TiO}_2} > n_{\text{ZnO}} > n_{\text{glass}}$ minimum reflection with transmission $>90\%$ in 450–650 nm wavelength range. From these findings it is observed that by having a high refractive index layers sandwiched between two low index layer ($n_{\text{SiO}_2} < n_{\text{TiO}_2} > n_{\text{ZnO}}$) leads to minimum reflections and maximum transmittance when compared to the graded index layers ($n_{\text{SiO}_2} < n_{\text{ZnO}} < n_{\text{TiO}_2}$).

According to Thetford vector model [18], the two amplitude reflection coefficients are computed using multi-layer stack split into two parts on either side of the middle layer. At the reference wavelength of $\lambda_0 = 550$ nm, the quarter-wave length thickness of the multi-layer coating made the reflectance equal. A second value of wavelength (λ_1) shorter

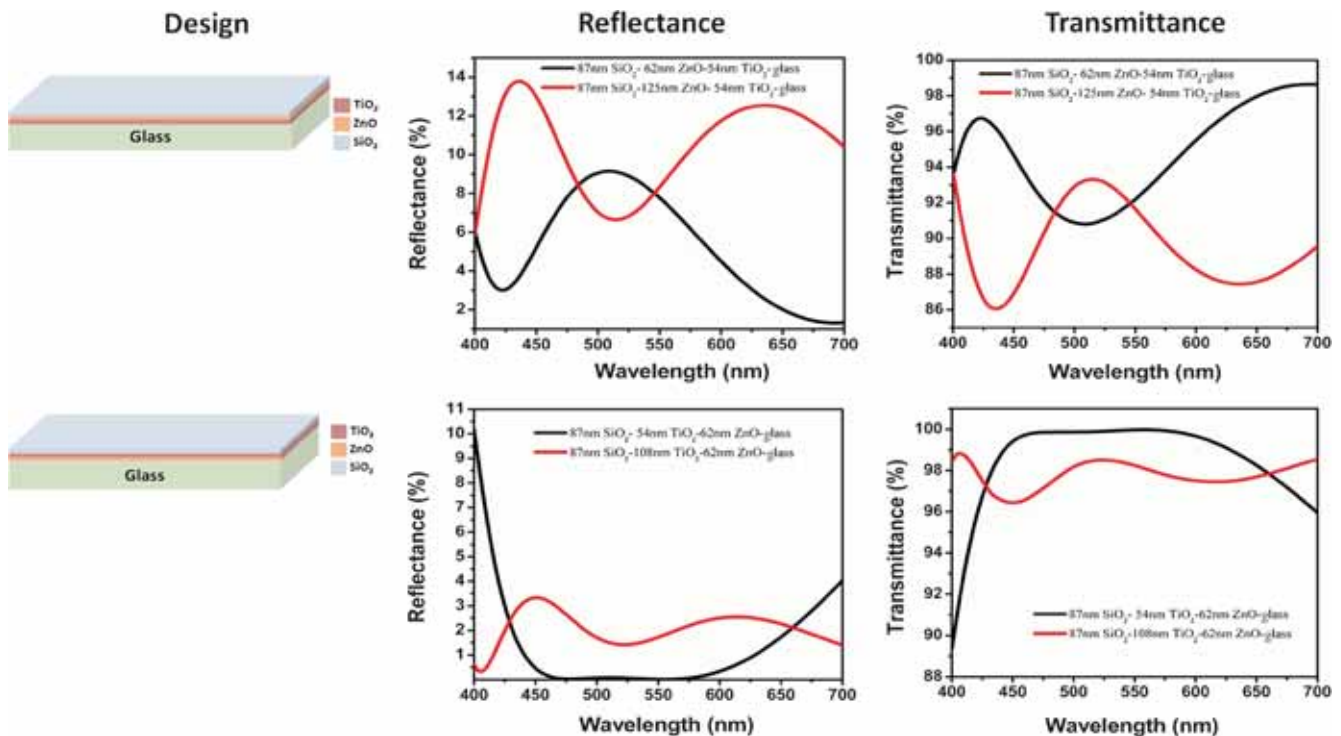


Figure 3. Computed spectra of reflectance and transmittance of a glass with SiO_2 – ZnO – TiO_2 and SiO_2 – TiO_2 – ZnO coatings on the top with thickness equivalent to Q–Q–Q and Q–H–Q wavelengths.

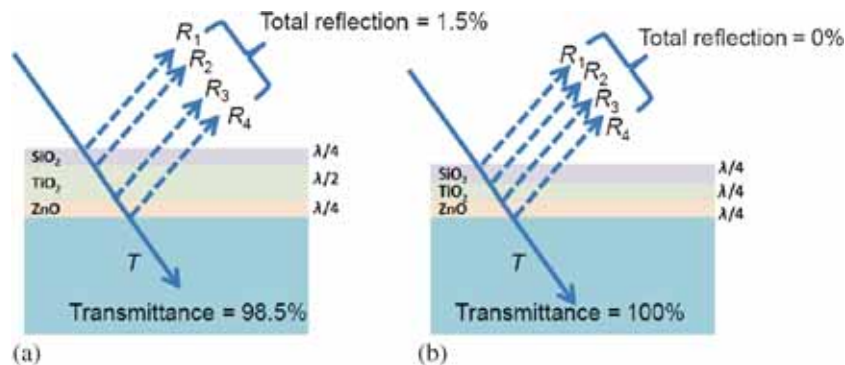


Figure 4. Schematic representation of reflection and transmittance occurring in (a) QHQ and (b) QQQ designs composed of SiO_2 (top layer) – TiO_2 (middle layer) – ZnO (bottom layer) triple coatings.

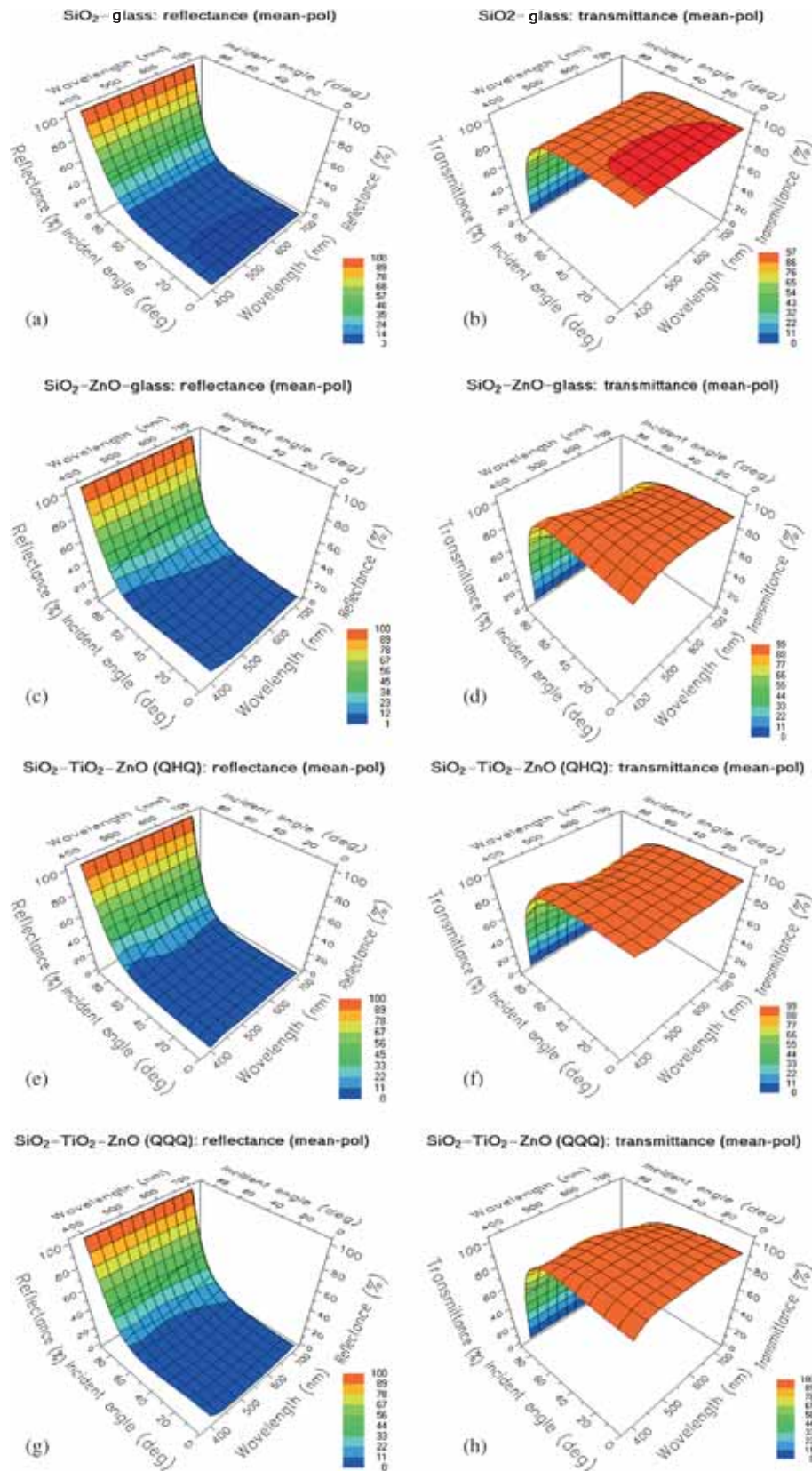


Figure 5. Computed 3D spectra of reflectance and transmittance (a–b) SiO_2 -coated glass, (c–d) SiO_2 -ZnO coated glass, (e–f) SiO_2 - TiO_2 -ZnO coating with thickness equivalent to Q–H–Q wavelength and (g–h) SiO_2 - TiO_2 -ZnO coating with thickness equivalent to Q–Q–Q wavelength.

than the first, at which the reflectance is equal, obtained with the expression related to change in reflectance with wavelength for each of the two structures. Therefore, phase condition was satisfied to give zero reflectance for the complete coating at the wavelength range of 450–650 nm. It is worth mentioning here that the thickness of the middle layer is a key factor to obtain the zero reflectance by satisfying the phase conditions at first and subsequent wavelengths.

Thickness of multilayer coating has a crucial role to play in minimizing the reflection from the glass surface. Herein, the thickness of the layers was chosen such that it obeys quarter-half-quarter (Q-H-Q) and quarter-quarter-quarter (Q-Q-Q) wavelength rule to obtain resultant zero reflectance. Instead, $\text{SiO}_2\text{-ZnO-TiO}_2$ coating on glass with thickness of each layer equivalent to Q-H-Q at 510 nm wavelength revealed high reflectance values of 6–14%, while transmittance value of 86–94% (figure 3). However, relative decrease in reflectance to 0.5–9% with transmittance of 91–98% was observed for Q-Q-Q coating ($\text{SiO}_2\text{-ZnO-TiO}_2$).

On the other hand, $\text{SiO}_2\text{-TiO}_2\text{-ZnO}$ coatings showed drastic improvement in the reflectance and transmittance values for both Q-H-Q and Q-Q-Q configuration in broad range of wavelength. This is due to the fact that second layer (TiO_2) has a high index of refraction than the first layer (SiO_2) and third layer (ZnO) and besides the optical thickness of TiO_2 is about quarter of the reference wavelength which lowers the reflectance down to 0.5–2.0%. In present case, the relative differences in refractive indices of middle and bottom layers have led to greater optical impedance mismatch at the layer interfaces with different angle of internal reflections and thereby broadening the reflectance band. Further, the thickness of intermediate layer of quarter-wavelength increases the optical impedance mismatch leading to higher minimum reflectance as in the case of Q-Q-Q coating design. A schematic shown in figure 4 represents the comparison of optical events taking place in $\text{SiO}_2\text{-TiO}_2\text{-ZnO}$ triple layer based on Q-Q-Q and Q-H-Q designs.

Given that the triple layer coating show very good suppression of reflectance, further we computed the mean polarized reflectance and transmittance spectra at incidence angle varying from 0 to 90° to understand its dependence on reflectance and transmittance spectra for simulated layers (figure 5). A comparison of different coatings shows that $\text{SiO}_2\text{-TiO}_2\text{-ZnO}$ coating clearly outperforms the single- and double-layered coatings in reducing the reflection of mean-polarized incident light, especially in the visible wavelength 400–700 nm regime. Similarly, in the case of transmittance spectra, computed for different layer thicknesses of $\text{SiO}_2\text{-TiO}_2\text{-ZnO}$ coating, Q-Q-Q wavelength coating outperforms when compared to Q-Q double and Q-H-Q triple coatings at wide range of incident angles. To validate these simulation results, experiments are being carried out by carefully controlling the individual layer thickness using magnetron co-sputtering technique. Optimization of process parameters for desired layer thickness will be carried out using response surface methodology.

4. Conclusions

The most interesting feature of the proposed design is the relatively small physical thickness of ~ 206 nm, making them feasible for practical deposition experiments. A particular feature of this design is that the refractive indices are confined between the high and low index components, which rules out the requirement for a material with refractive index close to that of the ambient (air). From this study, it is concluded that the thickness and refractive index of intermediate layer in Q-Q-Q or Q-H-Q play a dominant role in effectively cutting down the reflections through destructive interference of the reflected light. In addition, Q-Q-Q design is more appropriate for minimizing front surface reflections of glass. The layers proposed herein are standard materials for an optical coating and can be fabricated by simple deposition techniques. The designed coatings possess an ability to enhance the optical characteristics of solar photovoltaic module surface in broadband range. Therefore, such hybrid systems are highly interesting candidates and practically affordable for omni-directional anti-reflection coatings suitable for terrestrial solar panel.

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