A Theoretical Study for Designing Optical Multilayer Films Using NdF₃/ThF₄

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Abstract

An idea of a colored glaze is presented in this study to hide and dispose all the obstacles of using solar systems as facades integrated with buildings. This aim is achieved by designing multilayer optical interference filters by using Mat lab program . Appropriate dielectric materials, namely NdF_3 of high refractive index (n_H =1.6) and ThF_4 of low refractive index (n_L =1.5143) were employed. Quarter wave thicknesses of high (H) and low (L) refractive index were deposited on a microscopic slide substrate with n=1.513 and 550 nm design wavelength (λ_o). Two optical models were designed, which are Air//HL//glass and Air//LH//glass, for even numbers of layers (2-32 layers). The challenge in this study is to find the most efficient design which has lower solar reflectance ($R_{sol.}$) and higher solar transmittance ($T_{sol.}$) to raise the efficiency of the solar systems and, in parallel, obtain the colored reflection to achieve the esthetic appearance of the buildings integrated with the solar system facades. The $T_{sol.}$ value was high (94-95 %), whereas the $R_{sol.}$ was very low (4-5 %). Hence, the efficiency of the solar system was increased. The two optical models exhibited green color reflectance in the visible region. The first design, i.e. Air/HL/glass, showed higher values of $R_{vis.}$ and the merit factor (M) than the second model, resulting in a higher potential of coloration. The first design requires fewer materials and layers, thus, it is more cost-effective as compared to the second one.

Keywords: Colored glazing , Multi layers films , Facades collectors , Thin films .

دراسـة نظريـة لتصميم اغشيـة بصريـة متعددة الطبقات باستخدام ThF₄/NdF₃

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الخلاصة

تقدم هذه الدراسة فكرة بسيطة للحصول على طلاء زجاجي ملون لإخفاء و التخلص من جميع معوقات استخدام المنظومات الشمسية كواجهات متكاملة مع المباني من خلال تصميم مرشحات تداخل بصرية متعددة الطبقات باستخدام برنامج الماتلاب . تم استخدام مواد عازلة مناسبة (NdF₃) ذو معامل الانكسار العالي 1.6 = nₕ و (ThF₄) ذو معامل الانكسار الواطيء 1.5143 = nₗ و بسمك ربع طول موجة (H) و (L) لمعامل الانكسار العالي والواطيء على التوالي ، رسبت على ركيزة لشريحة مجهرية لها (1.513) = n و للطول الموجي للتصميم (λₒ) مقداره (550) نانومتر . تم تصميم نموذجين بصريين Air // HL // glass و glass // Air // LH لطبقات زوجية من (32−2) طبقة . يتمثل التحدي في هذه الدراسة في

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العثور على أفضل تصميم يمتلك اقل انعكاسية شمسية (.R_{sol}) و اكبر نفاذية شمسية (.T_{sol}) لرفع كفاءة
أنظمة الطاقة الشمسية وفي نفس الوقت الحصول على الانعكاس الملون للحصول على مظهر جمالي للمباني
المتكاملة بواجهات أنظمة الطاقة الشمسية . تمتلك النفاذية الشمسية (.T_{sol}) قيمة عالية تتراوح ما بين (94–
95)% ، وان الانعكاسية الشمسية (.R_{sol}) تمتلك قيمة قليلة جدًا وتتراوح ما بين (5–4)%، لذا فإن كفاءة
الأنظمة الشمسية سوف تزداد. يظهر النموذجان البصريان انعكاس اللون الأخضر في المنطقة المرئية. أظهر
التصميم الأول Air/HL/glass بأن قيم .R_{vis} وعامل الجدارة (M) له اعلى من قيم النموذج الثاني ، يشير
هذا إلى إمكانية كبيرة للتلوين . ان النموذج الاول يتطلب مواد أقل وعددًا أقل من الطبقات ، لذلك سوف يكون
اكثر فعالية من حيث التكلفة من التصميم الثاني .

1.   Introduction

  The fast industrial and economic development continuously increases energy demand [1]. The resources of renewable energy can be defined as clean resources that minimize environmental effects, produce zero or minimum secondary wastes, and are considered as sustainable based on the economic, energy, and social needs [2]. The renewable energy resources are characterized by the varied energy supply choices [3], reduced dependence on fossil fuels [4], increased net employment, progressive creation of export markets [5] , and reduced climate change and greenhouse gas emissions [6, 7]. The resources of renewable energy include solar , wind , biomass, geothermal, hydropower, and marine energies [8-10]. One of the most obvious resources is solar energy which is used for new buildings that have to meet nearly zero energy building standards, where the harvesting of energy from the surrounding environment is of utmost importance. Building-integrated photovoltaic and solar collectors are considered as a way to efficiently implement multi-functional building skins with respect to aesthetic, economic, and technical solutions . Parts of conventional building components and materials, such as facades and roofs, can be replaced by building-integrated photovoltaic (BIPV) and building-integrated solar thermal (BIST) units [11]. Technological advances in BIPV and BIST that can provide electricity and heating to buildings and their local environment were efficiently used in transforming building envelopes into energy generators based on renewable energy   The conventional BIPV and BIST facade characteristics have been modified in appearance and color to address such conflicts, leading to an on-going customization trend in BIPV and BIST façade design [12]. Coloring plays a significant role in the acceptance of BIPV and BIST applications. The aesthetics aspect of the whole building is determined by BIPV and BIST modules. In many recent projects, the PV and ST collectors, as active parts of the building envelope, are not recognizable. The fundamental requirement for the acceptance of PV and ST facades in buildings is the coloring of PV and ST collectors, which implies their ability to be camouflaged or designed. In BIPV and BIST glass facades, the conventional PV and ST material language can be hidden behind colored patterns that completely disguise the original materiality of the PV cells and ST collectors [13]. The technologies of the thin films, such as physical vapor deposition (PVD) and chemical vapor deposition (CVD), permit a new approach for generating colors with interference coatings for PV and ST modules, characterized by homogeneity, high color saturation, and low power impact [14, 15]. Antireflection coatings (ARCs) play a vital role in increasing transmittance and reducing reflectance [16]. The amorphous silicon, or CIS thin film solar cells, have a black color, while CdTe cells have a greenish look [17]. The absorbers in the solar thermal collectors assist the heat collection function, being usually colored with black or dark blue. The selective coatings are often the cause of the absorber sheet color, which is used to optimize absorption and reduce emission losses. PV products confer more freedom compared to the collectors [18]. In 2005, Schular *et al*. studied the optical behavior, $R_{vis.}$, $T_{sol.}$, CIE colored coordinates, and merit factor (M) for a multilayer thin film by making a simulation. They also investigated the potential of quarter wave thickness stacks [19]. In the same year, Schular *et al*. conducted a reflectance spectra

simulation for (W) design by the deposition of the dielectric materials SiO$_2$ and TiO$_2$ on both sides of a glass substrate with 10 to 800 nm design wavelength. They also studied a reflectance spectra (V) simulation design for 3 layers, where the glass substrate was coated only in one side. The researchers computed the angle dependence of reflectance spectra for quarter wave stacks [20]. Stacks of 3, 4, and 5 layers were fabricated using sol-gel dip technique by Schular *et al.*, who utilized SiO and mixed oxides of silicon titanium deposited on a glass substrate. The parameters of color CIE color coordinates, R$_{vis.}$, T$_{sol.}$ and merit factor were studied. The resulting coating had an M value of up to 2.4 and a reflection of bright color [21]. In the present study, appropriate dielectric materials, namely, NdF$_3$ of high refractive index (n$_H$ =1.6) and ThF$_4$ of low refractive index (n$_L$ =1.5143), were employed with quarter wave thicknesses for high and low refractive index layers. They were deposited on a microscopic slide substrate with n=1.513 and 550 nm designed wavelength ($\lambda_\circ$). Two optical models were designed; the first model is Air//HL//glass for even layers and the second is Air//LH//glass for even layers (2-32 layers). The challenge in this study is to find the best design which has lower R$_{sol.}$ and higher T$_{sol.}$ values to raise the efficiency of the solar systems and, simultaneously, achieve the colored reflection and gain the esthetic aspect of the solar systems used as building facades .

2. Theory

In a design of optical multilayer coatings, refractive index and thickness of each layer must be determined in accordance with specifications of spectral reflectance [22]. The characteristic matrix of a dielectric thin film takes a very simple form if the optical thickness is an integral number of quarter or half waves. That is, if

$$\delta = m(\pi/4) \qquad\qquad m = 0, 1, 2, 3...$$

for *m* even, cos δ = ±1, and sin δ = 0, so that the layer is an integral number of half wavelengths thick, and the matrix becomes

$$\pm \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}$$

This is the unity matrix which can have no effect on the reflectance or transmittance of an assembly. For *m* odd, sin δ = ±1, and cos δ = 0, so that the layer is an odd number of quarter wavelengths thick, and the matrix becomes [23]

$$\pm \begin{bmatrix} 0 & i/\eta \\ i\eta & 0 \end{bmatrix}$$

In general, the optical properties of thin filters can be computed by numerical simulations using the method of the complex matrix multiplication, where a characteristic matrix represents each layer. The assembly of a multilayer stack on a substrate can be described as

$$\begin{bmatrix} B \\ C \end{bmatrix} = \prod_{r=1}^{q} M_{r.} \begin{bmatrix} 1 \\ \eta_{sub}(\lambda) \end{bmatrix} \qquad\qquad (1)$$

where Equation (1) is called matrix of the assembly. The optical admittance of the parallel components of the incident electromagnetic wave at the outermost surface is given by Y (λ) = H(λ)/E(λ) = C/B . M$_r$ is the characteristic matrix of each layer, η_{sub} (λ) is the optical admittance of the substrate, and q is the number of layers in the stack. Negligible absorptance is assumed, which is consistent with the quasi-nil-absorptance requirement and with the used dielectric coating materials. The reflectance is then given by

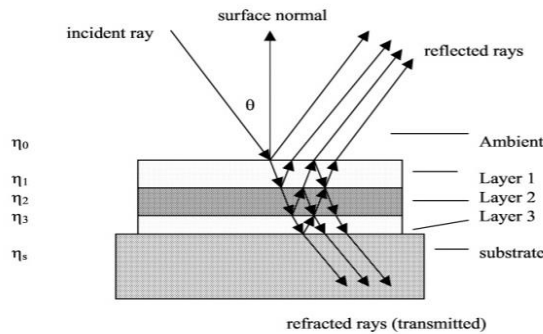$$R(\lambda) = \left(\frac{\eta_0(\lambda) - Y(\lambda)}{\eta_0(\lambda) + Y(\lambda)} \right)^2 \qquad\qquad (2)$$

The optical reflection can be changed due to the morphological difference in films [24]. The transmittance is given by

$$T(\lambda) = \frac{4. \eta_0(\lambda)Y(\lambda)}{\left(\eta_0(\lambda) + Y(\lambda)\right)^2} \tag{3}$$

where $\eta_0(\lambda) = 1$ for the incident medium air. For non-absorbing media, the energy conservation equation is simply

$$R + T = 1 \tag{4}$$

**Figure 1-**cross section diagram for multilayer thin film- substrate system [25].

Equation (4) is very general and valid for spectral values as well as for integrated quantities, such as T_{sol} or R_{vis}. T_{sol} is defined as the ratio between incident and transmitted solar radiation, whereas R_{vis} is defined as the ratio between incident and reflected daylight (CIE D65) weighted by the photopic luminous efficiency function $V(\lambda)$ of the human eye [25]. The angular dependence can be compensated, which is inherent to planar multilayer stacks due to the complex interactions in these functional multilayer thin films. High color saturation and, in the same time, high transmittance can be achieved, which are the requirements of the filter function by narrow reflectance peaks. To protect the functional layer against weathering effects, a glass cover is applied to the rear surface of the module, which can be manufactured by a sputter coating technology [26]. At the wavelength for which the layers are exact quarter waves, the reflected beam can form when all the multiple beams emerging from the top surface and the interference are at maximum. The width of the high reflectance zone is restricted and the higher the ratio of the high to low index the broader is the region, but it is always rather restricted obviously at one half of restricted. The design wavelength the layers become half waves and their interference effects vanish. If a dielectric substrate is coated with a dielectric film that is also made of a dielectric material in an incident medium, see Figure 1, then the film will support multiple beam interference [23].

2. Results and Discussion

A Matlab program was used in this study to design two optical models for quarter wave thicknesses. Two, dielectric materials, which are NdF_3 of high refractive index ($n_H = 1.6$) and ThF_4 of low refractive index ($n_L = 1.5143$) were deposited for a thickness of quarter wave on a microscopic slide substrate for a designed wavelength ($\lambda_\circ$) of 550 nm. The first optical model is Air/HL/Glass, while the second is Air/LH/Glass for even layers (2-32 layers). These two optical models work as multilayer optical interference filters that exhibit green colored reflectance in the visible region, which provide building integration solar collectors, or PV facades in an esthetic aspect. They also have high transmission in the near IR region, which is considered as a solar gain to supply the buildings with heating or electricity. The results reveal that the increase in the number of layers of the two optical models, and the even numbers of these layers, l produce an increase in peak high. This increase exhibit regular in

the Air/HL/Glass design. However, in the Air/LH/Glass design, it was noticed that the peak is inverted in figure, which has two layers of NdF_3 and ThF_4. It was also observed that, with increasing the number of layers, two peaks will appear. With the gradual increase in the layers number, these peaks will merge and appear as one regular peak in the case of 30 layers . All these behaviors are illustrated in Figure 2.
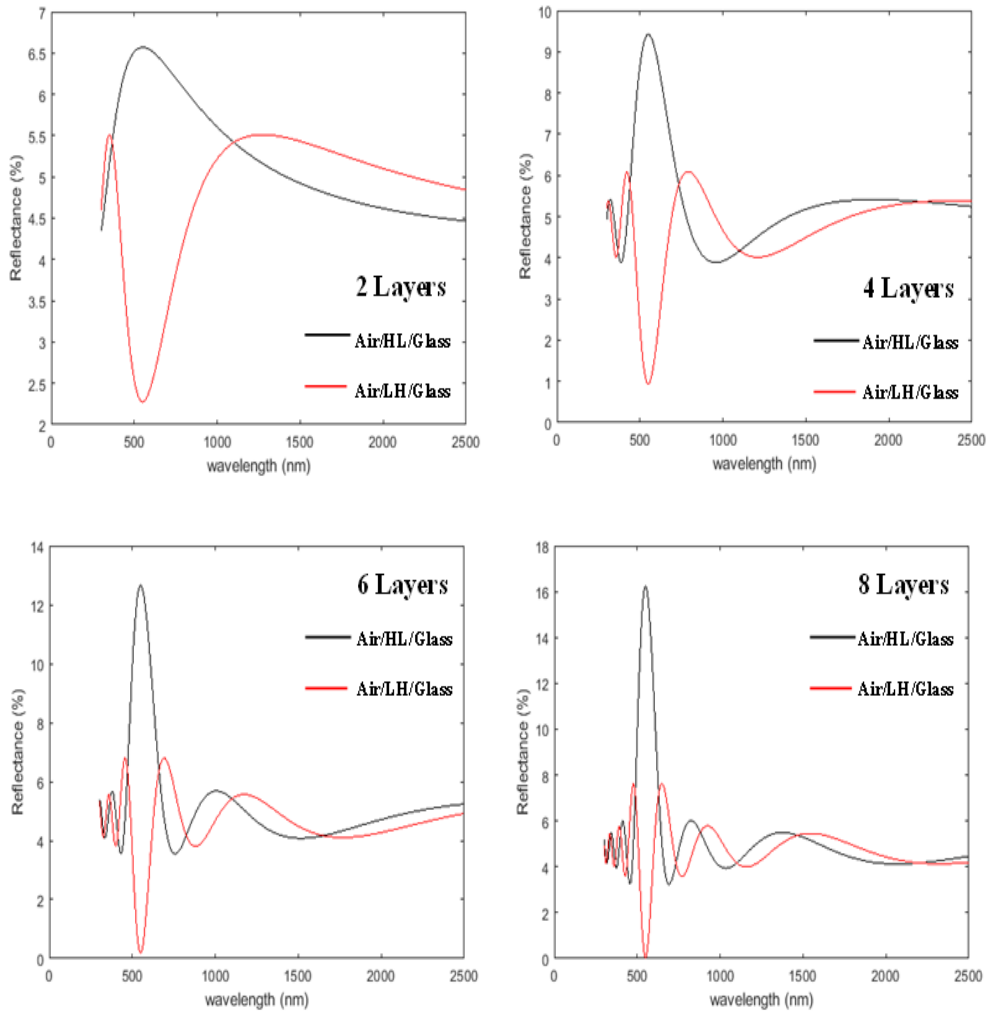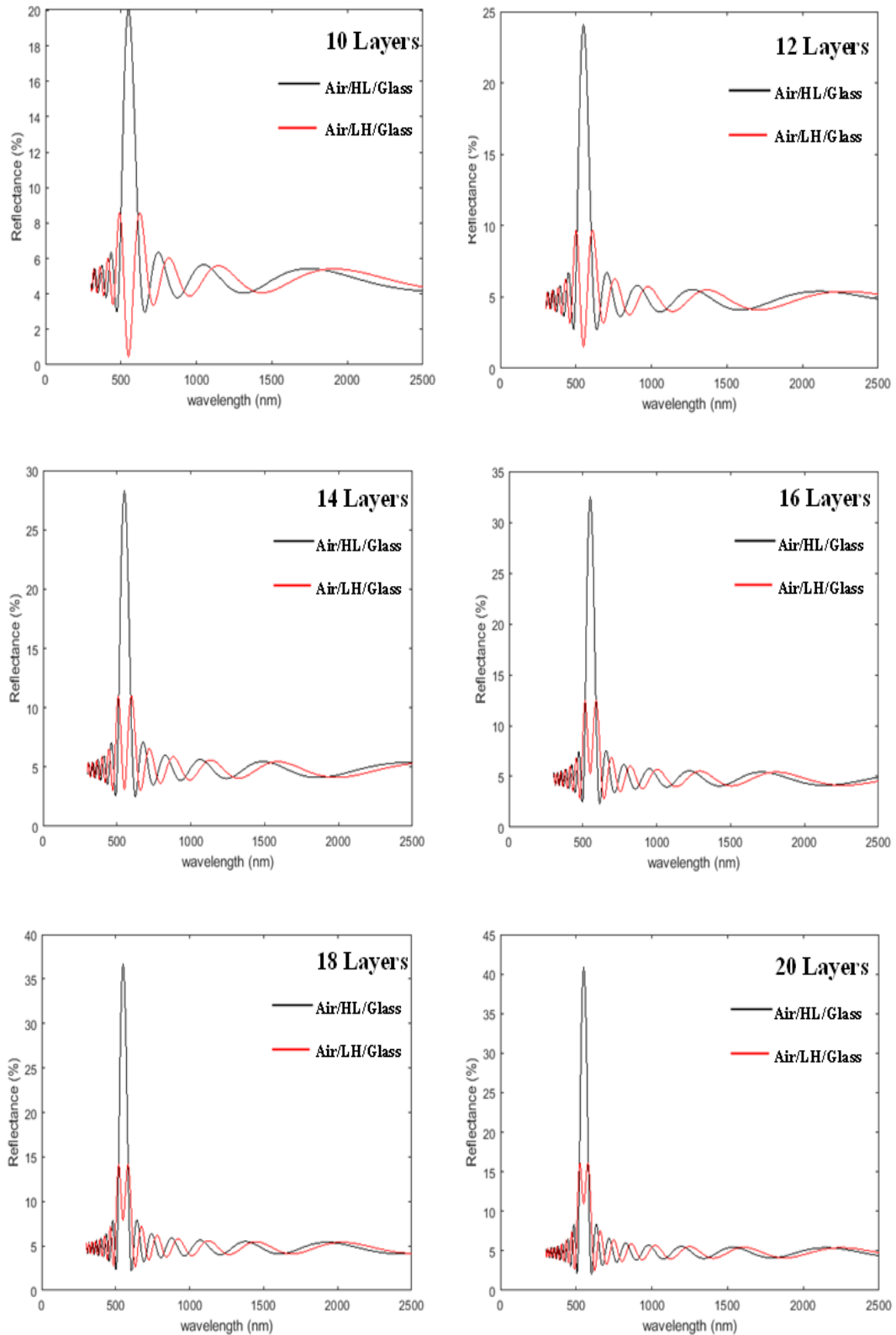
Figure 2 -Reflectance spectra of quarter wave thicknesses for even layers (2-32 layers) and for the two designs (Air/H/L/Glass and Air/L/H/Glass) at λ₀=550 nm.
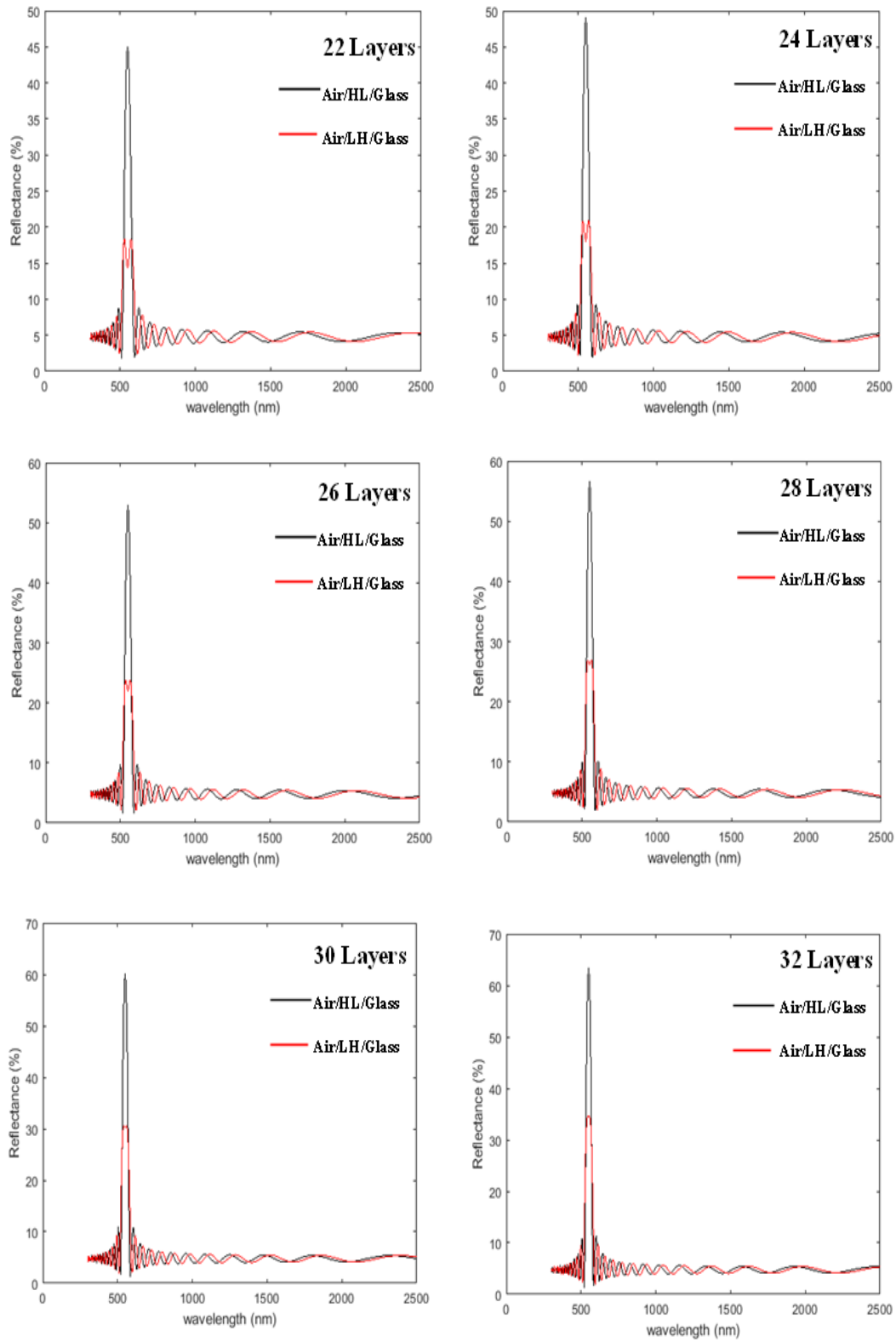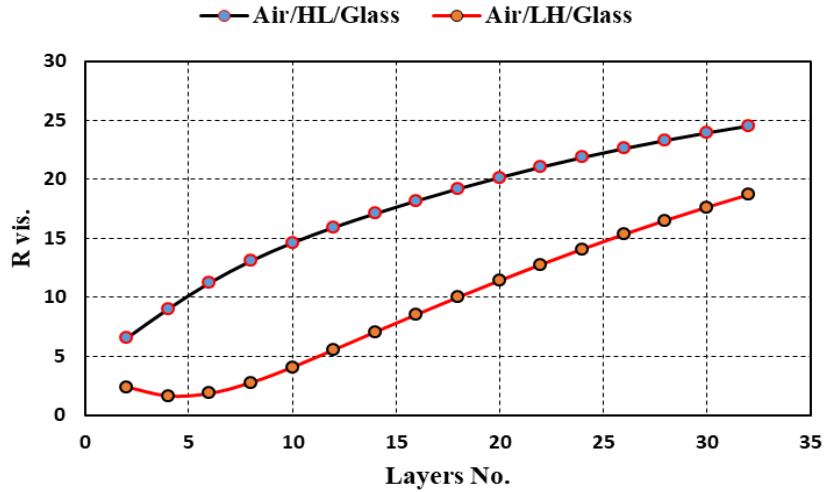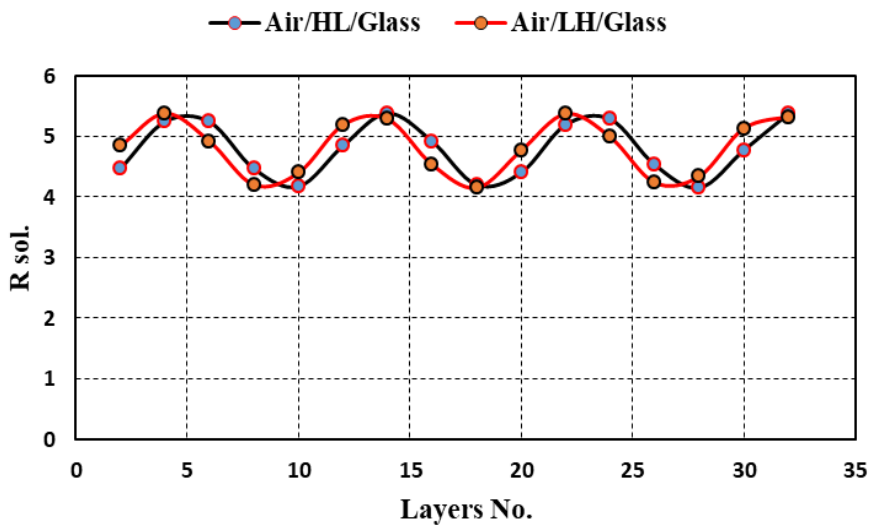
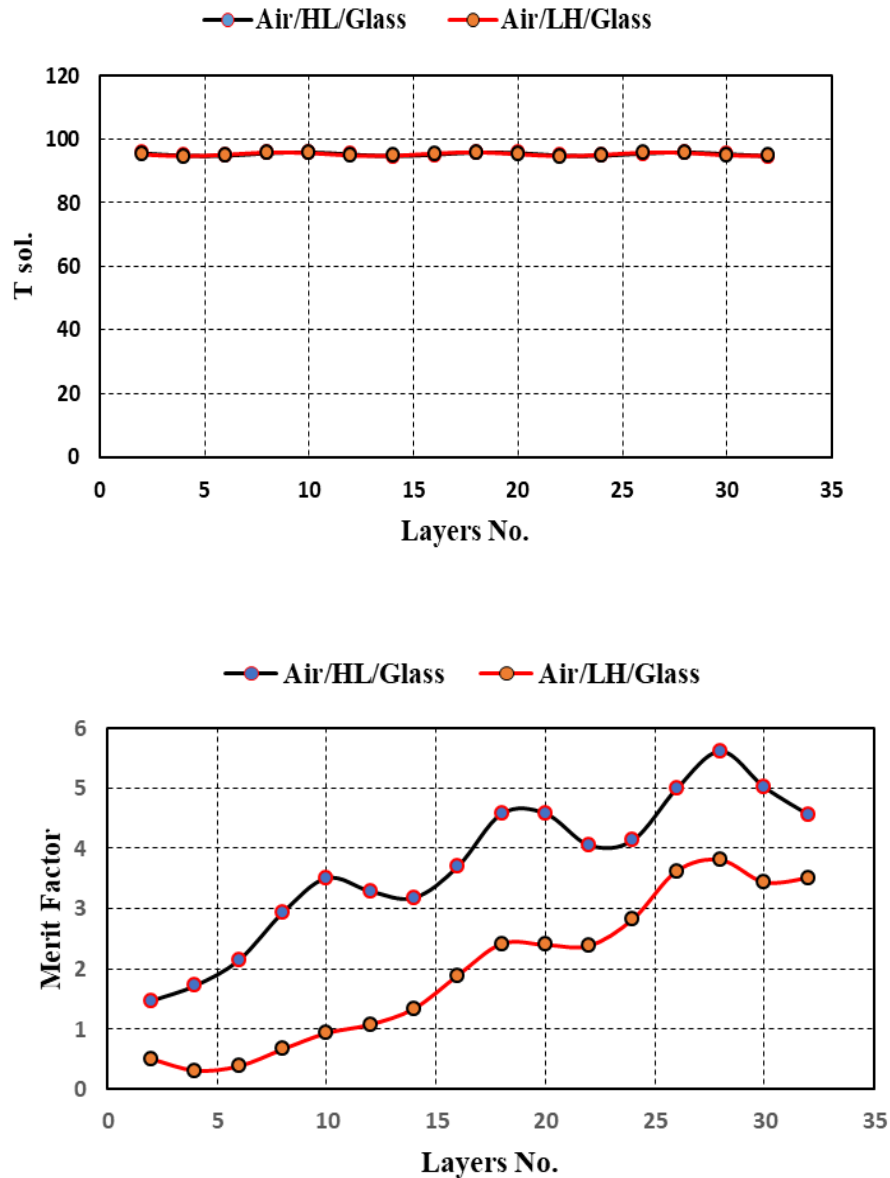**Figure 3 -** Rvis. value versus number of layers for air/HL/glass and air/LH/glass models.

Figure 4 - Tsol. value versus number of layers for air/HL/glass and air/LH/glass models.

The figure also shows that the peak of reflectance reaches to ≈ 65% in Air/H/L/Glass design and to ≈ 35% in Air/L/H/Glass design for 32 layers. The optical properties of the multilayer optical interference filters were computed theoretically for the two optical models. Figure 3 explains the behavior of $R_{vis.}$ for the two models with increasing layers number. The two cases exhibit an increase in visible reflectance values, which reaches 24.5258 in Air/H/L/Glass model of 32 layers and 18.6959 in Air/L/H/Glass, where 12% of $R_{vis.}$, which is already considerable for a color [19]. The behavior of Air/H/L/Glass model is better than that of Air/L/H/Glass model. This implies that Air/H/L/Glass model is better for colored visible reflectance used for solar collectors or photo-voltaic cells in integrated building facades. In the same time, the values of solar transmittance ($T_{sol.}$) become high and its values vary (94-95 %). Hence, the near infrared region remains as an antireflection region. Figure 4 explains the matching in $T_{sol.}$ values for the two models. While the values of the solar reflectance ($R_{sol.}$) for the two models vary from 4 to 5 %, which are considered as very low values compared with $T_{sol.}$ values (Figure 5) . Thus, the efficiency of the thermal solar collector or the photo-voltaic cells increases.

Figure 5 - Rsol. value versus number of layers for air/HL/glass and air/LH/glass models.

Figure 6- Merit factor (M) versus number of layers for air/HL/glass and air/LH/glass models.

The overall potential of the colored thermal solar collectors or PV cells is promising. This can be expressed by the value of M which describes the energy efficiency of the visual perception. A value of M= 6 indicates a high potential of coloration [19]. In a previous study presented by Schular *et al*. [21], it was exhibited that the resulting coating has an M value of up to 2.4, while our results exhibit a progressive increase with oscillation for the merit factor M (2-32 layers) for the two models. The value of M reaches to 5.6 to ≈6 for the air/HL/glass model with 28 layers. This indicates a high potential of coloration. While the air/LH/glass model has a value of M between 0.3 and 3.6, which implies a lower coloration efficiency. Figure 6 shows the relation between merit factor and the layers number.

3. Conclusions

1-   The results of this study show that the Rvis. value for air/HL/glass model is higher than that for the air/LH/glass model. The coatings exhibited green color reflectance which

2-   will add an esthetic aspect to the solar systems.

3- The coating of the two models has a high $T_{sol.}$ value (94-95 %). Hence, the near infrared region remains as an antireflection region. The R_{sol} value for the two models varies from 4 to 5 %, which is considered as very low compared with $T_{sol.}$ value. Thus, the efficiency of the solar systems will increase.

4- The results exhibit a progressive increase in the merit factor with the increase in oscillation for the two models. The values of M reach to ≈6 for air/HL/glass model, while air/LH/glass model has a range of 0.3-3.6. This indicates that air/LH/glass model is less efficient for coloration than the air/HL/glass model.

5- The Air/HL/glass model show higher values of $R_{vis.}$ and M than the second model, resulting in a high coloration potential. This model requires fewer materials and layers. Hence, it will be more cost-effective.

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