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PAPER

Optimization of multilayer anti-reflection coatings for efficient light management of PEDOT:PSS/c-Si heterojunction solar cells

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Keywords: reflection, AR coating, PEDOT:PSS, refractive index, transfer matrix, PEDOT:PSS/c-Si heterojunction solar cell

Abstract

In this paper, we present a theoretical model for the optimization of multilayer anti-reflection coatings for PEDOT:PSS/c-Si heterojunction solar cell based on optical interference transfer-matrix theory. A comprehensive idea of designing multilayer anti-reflection coatings (ARCs) on the solar cell and minimization of the overall reflectance is provided in this work. Optical reflectance values for various single, double and three layer anti-reflection coatings on PEDOT:PSS deposited c-Si substrate have been deduced using a MATLAB program and compared with that of the measured value. The reflectance value is calculated to be lower than 4% in the visible wavelength spectra for ARC by alternately using high and low refractive index materials. This low value of reflectance suggests that the anti-reflection coating layers proposed in this study can be employed as the standard ARC materials for optical coatings of the PEDOT:PSS/c-Si heterojunction solar cells.

1. Introduction

In recent years, photovoltaics has been extensively explored as a promising renewable clean energy technology to meet up with the issues of global energy crisis and green-house effects. In the last few decades, solar cells based on crystalline silicon (c-Si) have dominated the photovoltaic technology with the market share of over 90% of the global market although it requires a very high temperature of ~1400 °C for the fabrication [1]. Recently, an efficiency of 26.33% for the c-Si solar cell using back contact device structure with a practical module of 180 cm^2 has been reported [2]. Nowadays, polymer/silicon heterojunction solar cell with poly(3,4-ethylenedioxythiphene):poly (styrene sulfonate) (PEDOT:PSS) i.e. PEDOT:PSS/n-Si structure is being comprehensively studied due to its high efficiency and low-cost as it combines the advantages of organic materials being inexpensive and simple to process on Si substrate reducing high technological demands [3–7]. The efficiency of this type of c-Si/organic heterojunction solar cells has already reached to 13%–20% [7–14]. Most recently, all solution-processed PEDOT:PSS/n-Si solar cell modules also have been demonstrated consisting of ten-units of series-connected 2×2 cm^2 (4 inch) sized cells exhibiting a output power of 0.37 W (7.3 W) with a power conversion efficiency (PCE) of 13%–14% (11%–12%) [15].

Among the different constrains to fabricate high efficiency PEDO:PSS/n-Si heterojunction solar cells, the reflection of the incident light by the surface of solar cell is a key source of losses for photovoltaic conversion as Si surface reflects more than 30% as well as PEDOT:PSS on Si substrate reflects ~20% of the incoming light [10, 16]. These optical losses can be minimized by designing anti-reflection structure for crystalline silicon solar cells as it ensures a high photocurrent by reducing the reflectance.

So far, various anti-reflection coatings (ARCs) deposited on silicon solar cells have been optimized theoretically and experimentally to lessen the optical losses in the solar cells [17]. In addition, the double antireflection coatings (DARCs) and multilayer antireflection coating (MARCs) using different types of materials such as SiO_2/Si_3N_4, SiO_2/TiO_2, ZnS/MgF_2, and SiN_x:H/SiO_xN_y/SiO_x have been reported by many

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authors [16, 18–20]. However, these ARC layers reported in the previous studies require high temperature and high vacuum deposition technologies. On the other hand, ARCs for the PEDOT:PSS/c-Si heterojunction solar cells require low temperature deposition process because the processing temperature higher than 200 °C severely deteriorates the properties of PEDOT:PSS polymer and hence the performance of the solar cells [12]. Although, PEDOT:PSS having refractive index (RI) of ~1.6 reduces the reflectance of the silicon to about 20%, it is still high for reducing the optical loss in the PEDOT:PSS/c-Si heterojunction solar cell [10, 21]. This is happened due to the reason that PEDOT:PSS as a single ARC layer can reduce reflection at a specific wavelength at normal incidence and unable to provide broadband reduction in reflectance over a wide range of incidence angles [22, 23].

However, there are few reports on the use of low temperature solution-processed DARC structures for the PEDOT:PSS/c-Si heterojunction solar cells such as TiO_2, MoO_x and Nafion which include PEDOT:PSS as one of the DARC layers [10, 12, 21, 24]. Although, texturing of the silicon can reduce reflectance which enhances the short circuit current, but it additionally introduces huge defects at the PEDOT:PSS/c-Si interface resulting lower open circuit voltage and hence photovoltaic performance of the solar cell [24–26]. Therefore, the deposition of multilayer antireflection coatings (MARCs) on solar cell with flat silicon substrate seems to be a good solution to further reduction of optical losses as well as avoiding defects for achieving higher photovoltaic performance of the solar cells.

In the present work, we report a theoretical design guideline for the optimization of MARCs for PEDOT:PSS/c-Si heterojunction solar cells using optical interference transfer-matrix theory with introducing few new materials such as $SrTiO_3$ reported to be produced by solution-process. We also compare the simulated reflectance value with the measured value of reflectance of PEDOT:PSS on crystalline silicon.

2. Experimental details

The samples for the measurement of thickness and reflectance of PEDOT:PSS polymer were prepared on one side polished CZ crystalline n-type silicon substrates with thickness and resistivity of 300 μm and 0.1–0.3 Ω·cm, respectively. The substrates were cleaned first in acetone for 10 min followed by isopropyl alcohol (IPA) and de-ionized (DI) water cleaning for 10 min each in an ultrasonic bath. Then these substrates were immersed in HF (5%) solution for 2.5 min and then rinsed in DI water and then dried by N_2 gas blower. After that, conductive PEDOT:PSS (Clevios PH1000) with 7 wt% Ethylene glycol (EG) and 0.2 wt% Zonyl surfactant was spin coated on the cleaned c-Si substrate at 2500 rpm for 1 min. The samples were then annealed at 140 °C for 30 min to remove the residual solvent.

The thickness of PEDOT:PSS thin films deposited on c-Si was measured by Dektak-150 thickness profilometer and the reflectance of flat silicon and PEDOT:PSS on flat silicon was measured by Shimadzu UV–vis spectrophotometer (UV-2600 Shimadzu).

3. Design methodology for MARCs

The optical interference matrix, commonly known as transfer matrix, method has been employed to design multilayer anti-reflection coatings on the substrate material having refractive index, n_s [22, 27–29]. The idea behind this method is to adjust the electric and magnetic field of the incident light on the surface of multilayer optical coatings [29]. The optical admittance, Y is used to calculate the reflectance in multilayers, which is the ratio of the normalized total tangential electric field (B) and magnetic field (C). Under the situation of zero degree incident angle of light, the general form of the transfer matrix for N-layer anti-reflection coatings is given by [27]

$$\begin{pmatrix} B \\ C \end{pmatrix} = \left\{ \prod_{j=1}^{N} \begin{bmatrix} \cos\delta_j & \dfrac{i\sin\delta_j}{n_j} \\ in_j\sin\delta_j & \cos\delta_j \end{bmatrix} \right\} \begin{pmatrix} 1 \\ n_s \end{pmatrix} \tag{1}$$

where, n_j represents the refractive index of the jth layer, δ_j is the effective optical thickness of the jth layer, and n_0 is the refractive index of air ($n_0 = 1$), respectively. The effective optical thickness (phase thickness), δ_j is given by

$$\delta_j = \frac{2\pi}{\lambda} n_j d_j \cos\theta_j \tag{2}$$

where d_j is the physical thickness of the jth layer, θ_j is the angle of incidence in the layer jth layer. The optical admittance, Y is given by the ratio

$$Y = \frac{C}{B} \tag{3}$$

This transfer matrix is usually used to calculate the reflectance of a stack of thin film of N layers where each layer is represented by a 2×2 matrix. The order of the characteristic matrix at a wavelength λ for the assembly of N layers is given by

$$\begin{pmatrix} B \\ C \end{pmatrix} = [M_1][M_2][M_3]\ldots\ \ldots[M_N]\begin{pmatrix} 1 \\ n_s \end{pmatrix} \tag{4}$$

where, M_1 is associated with layer 1, M_2 is associated with layer 2 and so on. Thus, the N-th layer is represented by a 2×2 matrix, M_N of the form

$$M_N = \begin{bmatrix} \cos\delta_N & \dfrac{i\sin\delta_N}{n_N} \\ in_N\sin\delta_N & \cos\delta_N \end{bmatrix} \tag{5}$$

Then from the above equations, it is possible to determine the reflectance, R of the stack of layers by calculating the reflection coefficient, r. The r is given by

$$r = \left(\frac{n_0 - Y}{n_0 + Y}\right) \tag{6}$$

The reflectance, R is given by [27]

$$R = |r|^2 = \left|\frac{n_0 - Y}{n_0 + Y}\right|^2 \tag{7}$$

3.1. Double-layer antireflection coating

In the case normal incident of light, the transfer matrix for double layer anti-reflection coatings becomes [27]

$$\begin{pmatrix} B \\ C \end{pmatrix} = \begin{bmatrix} \cos\delta_1 & \dfrac{i\sin\delta_1}{n_1} \\ in_1\sin\delta_1 & \cos\delta_1 \end{bmatrix}\begin{bmatrix} \cos\delta_2 & \dfrac{i\sin\delta_2}{n_2} \\ in_2\sin\delta_2 & \cos\delta_2 \end{bmatrix}\begin{pmatrix} 1 \\ n_s \end{pmatrix} \tag{8}$$

3.2. Three-layer antireflection coating

Accordingly, the transfer matrix for three layer ARCs equation for normal incidence of light can be written as follows [27]

$$\begin{pmatrix} B \\ C \end{pmatrix} = \begin{bmatrix} \cos\delta_1 & \dfrac{i\sin\delta_1}{n_1} \\ in_1\sin\delta_1 & \cos\delta_1 \end{bmatrix}\begin{bmatrix} \cos\delta_2 & \dfrac{i\sin\delta_2}{n_2} \\ in_2\sin\delta_2 & \cos\delta_2 \end{bmatrix}\begin{bmatrix} \cos\delta_3 & \dfrac{i\sin\delta_3}{n_3} \\ in_3\sin\delta_3 & \cos\delta_3 \end{bmatrix}\begin{pmatrix} 1 \\ n_s \end{pmatrix} \tag{9}$$

3.3. Simulation in MATLAB

The reflectance spectra have been simulated in MATLAB software using the transfer-matrix by varying the refractive indices and thicknesses of different layers. The thickness of the PEDOT:PSS layer on the silicon substrate has been kept fixed in the simulation. The reference wavelength for the calculation of the thickness of the layer has been considered as $\lambda_o = 650$ nm. In this work, the wavelength has been taken per nanometer to calculate the broadband reflectance of the anti-reflection coatings in the range of 300–1200 nm. All the calculations have been done by taking the normal angle (0°) of the incident light [27, 28].

The parameters of the program are as follow:

(a) Refractive index (RI) for each coating layer.

(b) Optical thickness for each layer.

(c) Refractive index of the substrate.

4. Results and discussion

4.1. PEDOT:PSS/c-Si heterojunction solar cells: PEDOT:PSS as a single ARC layer

The silicon/organic heterojunction can be fabricated by simple deposition techniques such as spin coating, chemical mist deposition, electrospray deposition and ink jet, which have a large degree of freedom in selection

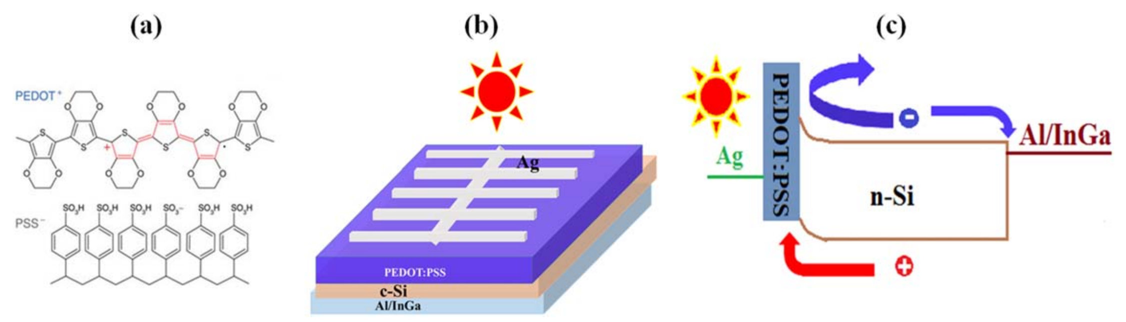

Figure 1. (a) The chemical structure of PEDOT:PSS, (b) The schematic diagram of the PEDOT:PSS/c-Si heterojunction solar cell and (c) The schematic energy diagram of the PEDOT:PSS/c-Si solar cell.

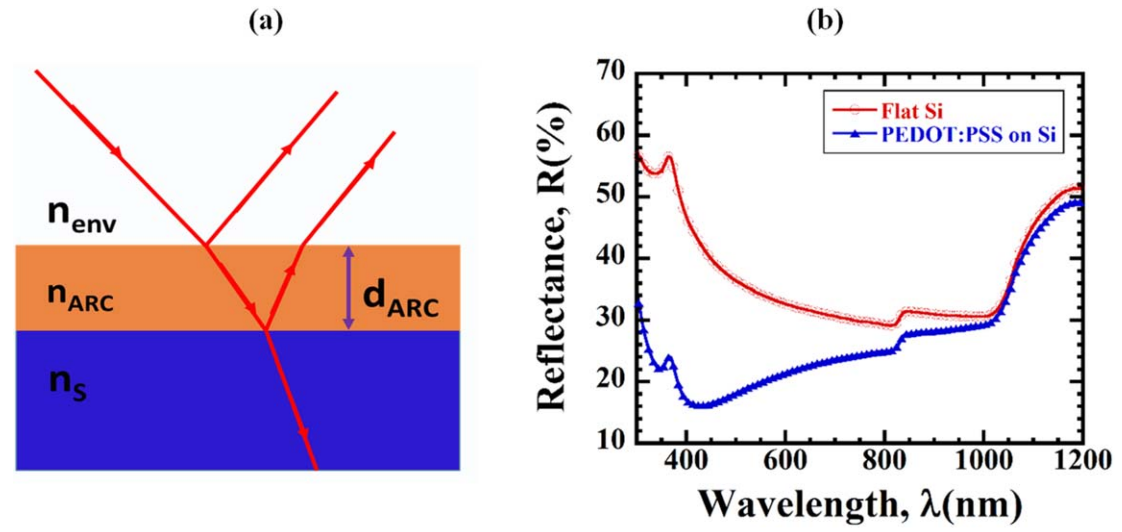

Figure 2. (a) The schematic diagram of the single layer ARC design and (b) The measured reflectance spectra of flat Si and 80 nm thick PEDOT:PSS thin film on flat Si.

of materials to deposit the organic polymer on crystalline silicon [3–7]. PEDOT:PSS is the most preferred polymer due to its high transparency, hole-conducting ability, suitable work function [30–32]. Figures 1(a)–(c) show the chemical structure of PEDOT:PSS, the schematic diagram and schematic energy diagram of PEDOT:PSS/c-Si heterojunction solar cell, respectively. The conductive PEDOT:PSS polymer comprises two ionomers. One is conjugated hydrophobic and conductive polymer, PEDOT which conducted positive charge. The other ionomer is hydrophilic and insulating PSS chain which conducted negative charge. In this photovoltaic structure, crystalline silicon is used as an effective light absorber, whereas the organic film mainly acts as hole transporting layer in addition to the antireflection coating [33–35].

Figure 2(a) shows schematic diagram of the single ARC layer. In order to act a single layer as an ARC layer to obtain net zero reflection in the spectral range of 300–1200 nm, the amplitudes of the reflected waves at the air-ARC interface and ARC-substrate interface have to be identical and precisely one-half wave (180°) out of phase, resulting in destructive interference. This condition is fulfilled when the optical thickness of the ARC layer (d_{ARC}) is equal to a quarter of the wavelength (λ) at which reflectance is zero. This condition can be expressed mathematically by equation [22]

$$d_{ARC} = \frac{\lambda}{4n_{ARC}} + m\frac{\lambda}{2n_{ARC}}(m = 0, 1, 2, 3\ldots) \tag{10}$$

where, n_{ARC} is the refractive index of the thin-film ARC layer [36]. Another condition that must be meet is that the magnitude of front surface reflections must be identical to the magnitude of the sum of all other partial reflections in the thin-film ARC layer. The ARC should fulfill the following condition in order to reduce the reflection down to zero at that wavelength [37]

$$n_{ARC} = \sqrt{n_S n_{env}} \tag{11}$$

where, n_S is the refractive index of substrate and n_{env} is the refractive index of environment, respectively.

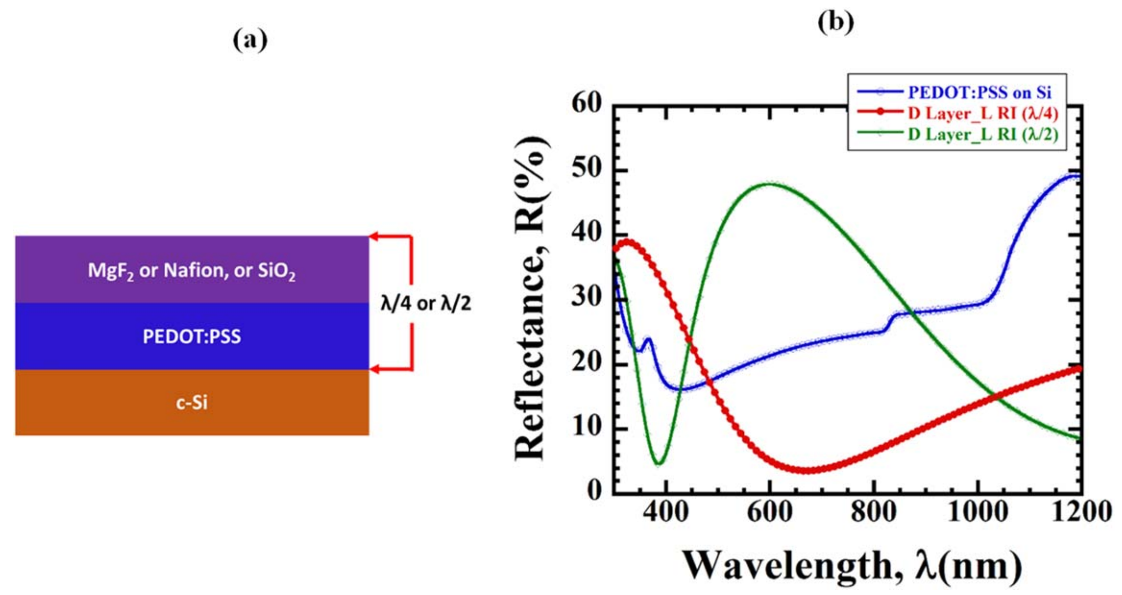

Figure 3. (a) The schematic diagram of double layer AR coatings design with low-index materials and (b) Reflectance of the double layer ARCs using low-index material and PEDOT:PSS on Si Substrate, respectively.

However, PEDOT:PSS as a single ARC layer cannot fulfill the above two criteria well because the single layer $\lambda/4$ ARCs reduce the reflectance for limited (almost single) wavelengths and incidence angle, the refractive index of the PEDOT:PSS is ~1.6 and the thickness of the PEDOT:PSS film should be in the range of 80–100 nm for efficient photovoltaic performance of PEDOT:PSS/c-Si heterojunction solar cell [3–14, 21, 38]. These two factors violate the condition for a single ARC layer as it is necessary to deposit a 68 nm ARC layer with an RI of 2.02 on silicon substrate having refractive index of ~4.0 at 550 nm [38, 39]. Figure 2(b) shows the measured reflectance spectra of flat Si and 80 nm thick PEDOT:PSS film on flat Si substrate as a function of wavelength. It is seen from this figure that PEDOT:PSS reduces the reflectance of the silicon substrate to ~22% at 650 nm wavelength. Therefore, it can be concluded that PEDOT:PSS can act as a single ARC layer in addition to hole transport for PEDOT:PSS/c-Si heterojunction solar cells [10, 12, 21, 24, 33–35]. For further reduction of the reflectance of the solar cell structure, DARC and MARC layers have been designed using transfer matrix and discussed in thefollowing sections. The materials for the construction of upper layers of ARCs on the PEDOT:PSS have been chosen such that they have low cost and easy availability.

4.2. Double layer ARCs design

Double-layer ARCs are generally used in solar cells for minimizing the reflectance for a specific wavelength spectra. In the design of double-layer ARCs, the coating film having the lowest refractive index is usually placed at the top to face the air, and the other layer is placed on the ascending order of its refractive index [38]. However, in this work, as PEDOT:PSS does not follow the design condition of AR coating, materials with low and high refractive indices have been placed at different positions with an overall optical thickness of $\lambda/4$ or $\lambda/2$ to optimize the reflectance spectra for PEDOT:PSS/c-Si heterojunction solar cells.

4.2.1. Design with low-index materials

Solution-processed materials with low (L) refractive indices (1.2–1.4) such as MgF_2, Nafion and SiO_2 can be used to minimize the reflectance loss over a broadband spectral wavelength [21, 40, 41]. Figure 3(a) shows the block diagram of the DARCs layer design for PEDOT:PSS/c-Si heterojunction solar cells. The measured reflectance spectra of PEDOT:PSS and simulated reflectance spectra as a function of wavelength of the double ARC layers for PEDOT:PSS/c-Si solar cell are shown in figure 3(b). It is seen from the figure that double ARC layer designed with a quarter-wavelength ($\lambda/4$) thickness at the reference wavelength of 650 nm for normal incidence shows the lowest reflectance minimizing the reflectance lower than 4% in the visible wavelength. The thicknesses of PEDOT:PSS and materials with RI of 1.35 for the corresponding DARCs are 80 and 20 nm, respectively. The simulated reflectance spectra of the DARCs with $\lambda/2$-wavelength thickness are highly increases in the visible range as compared to the $\lambda/4$-wavelength DARCs as also shown in the figure. The thicknesses of PEDOT:PSS and other material with RI of 1.35 for the DARCs design are 80 nm of 120 nm, respectively in this case. These results indicate that reflectance of DARCs with low-index materials is highly affected by the thicknesses of the

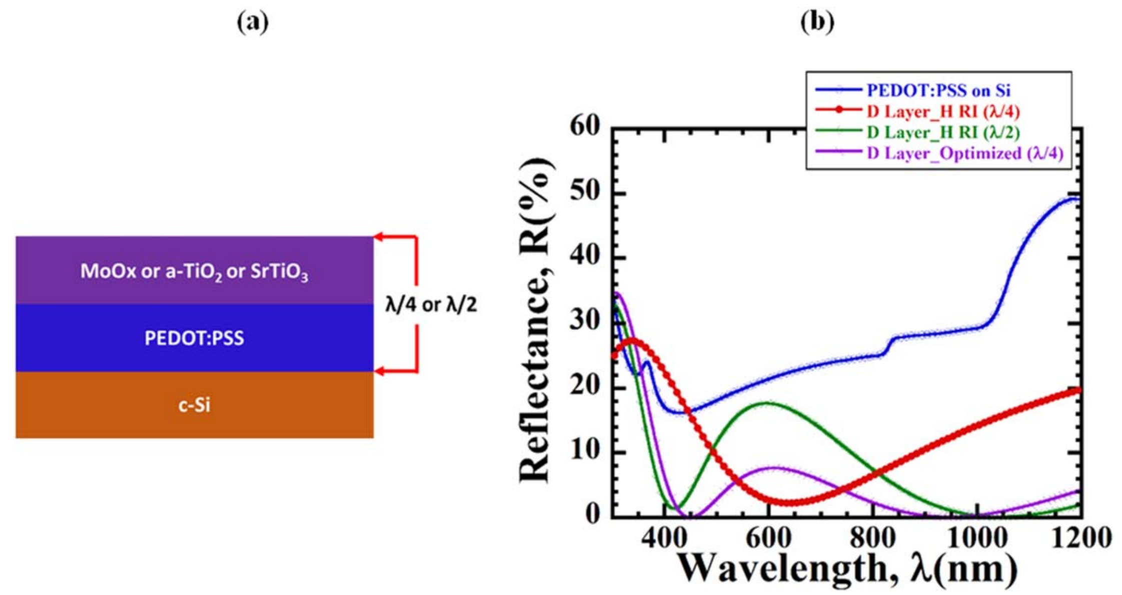

Figure 4. (a) The schematic diagram of double layer AR coatings design with high-index materials and (b) Reflectance spectra of the double layer ARCs using high-index material and PEDOT:PSS on Si Substrate, respectively (Reflectance of PEDOT:PSS on Si is shown as a guide to the eye).

coating layers. Therefore, it can be concluded that materials having RI of 1.35 can be used for double layer ARCs with $\lambda/4$- wavelength thickness for achieving the lowest reflectance.

4.2.2. Design with high-index materials

Solution-processed materials with high (H) refractive indices (2.0–2.5) such as MoO_x, anatase TiO_2, and $SrTiO_3$ etc can be used to minimize the reflectance loss over a broadband spectral wavelength [12, 24, 42]. The block diagram of DARCs layer design with high-index materials for PEDOT:PSS/c-Si heterojunction solar cells is shown in figure 4(a). The variation of reflectance with wavelength of the corresponding double ARC layers and that of PEDOT:PSS is shown in figure 4(b). It is seen from the figure that the reflectance spectra for DARCs with $\lambda/4$-wavelength thickness shows the lowest reflectance minimizing the reflectance close to 2% at 650 nm. The thicknesses in this DARCs design with high-index materials are 80 nm for PEDOT:PSS and 20 nm for materials with RI of 2.10. It is also seen from the figure that the reflectance spectra for DARCs with $\lambda/2$ thickness are W-shaped which indicates that reflectance reaches to a minimum value for two wavelengths, which contribute to minimize reflectance over a wide range of wavelength (300–1200 nm) [43]. The thicknesses of PEDOT:PSS and materials with RI of 2.10 for $\lambda/2$-wavelength DARCs design are same 80 and 80 nm, respectively. Therefore, it can be concluded that materials with RI of 2.10 can be used for double layer ARCs design to achieve the lowest reflectance compared to the measured value of PEDOT:PSS on Si substrate.

The optimized condition for reducing reflectance loss to the lowest value for DARCs layer design is also shown in figure 4(b). The optimized condition for DARCs layer design is using a materials having RI and thickness of 2.45 and 63.8 nm, respectively on top of 80 nm thick PEDOT:PSS thin film.

4.3. Three layer ARC design

In the present study, three layer anti-reflection coatings (ARCs) have also been design on the Si substrate for minimizing the reflectance loss of PEDOT:PSS/c-Si heterojunction solar cells. Figure 5(a) shows the cross section of the MARCs layer design for the PEDOT:PSS/c-Si heterojunction solar cell. Different types of materials having low (L) or high (H) refractive indices are placed as Layer 1 or Layer 2 on the top of PEDOT:PSS for achieving lowest reflectance. The reflectance spectra as a function of wavelength for triple-layer MARCs are depicted in figure 5(b). It is seen from the figure that w-shaped spectra shifted to lower wavelength for triple-layer MARCs except for the design with Layer 1 having high-index and Layer 2 having low-index, respectively for a thickness of $\lambda/2$-wavelength. This means that wavelength for minimum reflectance resides in the longer wavelength region in the spectra and triple-layer MARCs design does not lead to a remarkable reduction in the reflectance for the PEDOT:PSS/c-Si solar cells. As observed in the figure, a sequential layer of PEDOT:PSS, Layer 1 having RI of 2.45 and Layer 2 having RI of 2.1 with the optimized layer thicknesses of 80, 20 and 20 nm, respectively can be organized on the top of silicon substrate for achieving the lowest reflectance than PEDOT:PSS.

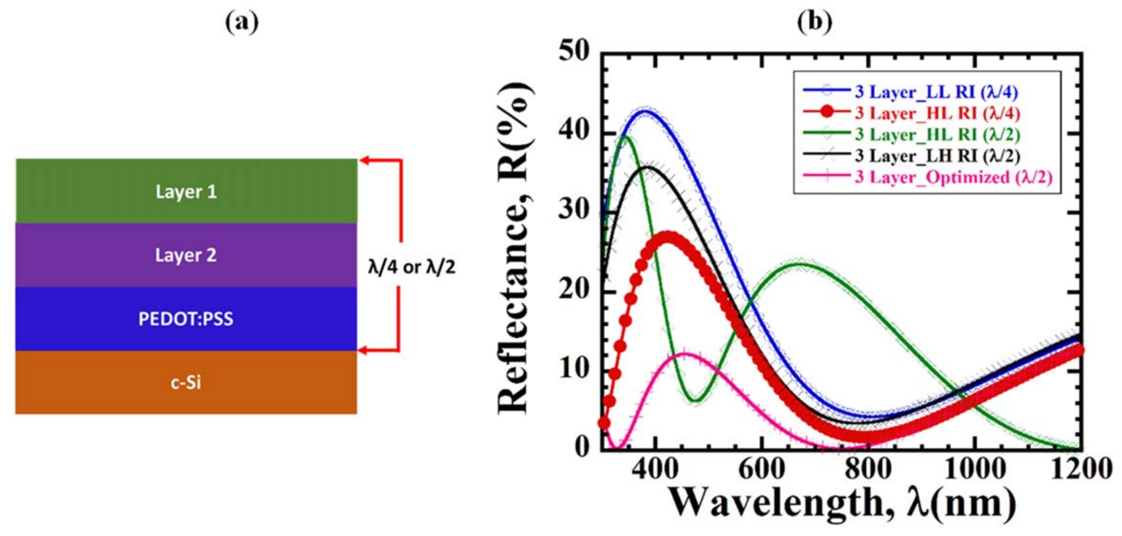

Figure 5. (a) The cross-section of three layer ARCs design and (b) Reflectance spectra of the three layer ARCs using high (H) and low (L) RI materials on 80 nm PEDOT:PSS coated Si Substrate, respectively.

Table 1. Tabular representation of the overall AR coatings design for PEDOT:PSS/c-Si solar cells.

Design	Materials	Refractive index	Optimized layer Thickness (nm)	Total coating Thickness (nm)
1. Medium	Air	1.0		
Layer 1	PEDOT:PSS	1.60	80	80
Substrate	Si	4.00		
2. Medium	Air	1.00		
Layer 1	MgF_2/Nafion/SiO_2	1.35	20	
Layer 2	PEDOT:PSS	1.60	80	100
Substrate	Si	4.00		
3. Medium	Air	1.00		
Layer 1	MoO_x/a-TiO_2/$SrTiO_3$	2.0–2.5	20	100
Layer 2	PEDOT:PSS	1.60	80	
Substrate	Si	4.00		
4. Medium	Air	1.00		
Layer 1	TiO_2	2.1	20	120
Layer 2	$SrTiO_3$	2.45	20	
Layer 3	PEDOT:PSS	1.60	80	
Substrate	Si	4.00		

4.4. Overall anti-reflection coatings design

The details of the design parameters for anti-reflection coatings used in this study are shown in table 1 optimized for $\lambda/4$-wavelength. The ARCs layer design has been optimized using SiO_2, MgF_2, Nafion, MoO_x, TiO_2 and $SrTiO_3$ layers to minimize the reflection for wavelength in the range of 300–1200 nm. In addition, materials library database has been used to select the values of refractive indices for SiO_2, MgF_2 Nafion, TiO_2 and $SrTiO_3$.

5. Conclusion

We demonstrate a model based on transfer matrix approach to design and simulate multilayer anti-reflection coatings in the visible and near IR range starting from double-layers to three-layers for the solution-processed PEDOT:PSS/c-Si heterojunction solar cells using MATLAB program. The most interesting feature of this ARCs layer design is that all of the ARC layers have small physical thicknesses. The reflectance spectra have been calculated in the wavelength range of 300–1200 nm and found to be lower than 4% in the visible wavelength when materials with high and low refractive indices are used for the DARC. The ARC layers proposed herein are standard materials for an optical coating and can be fabricated by simple low temperature solution process.

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Disclosure statement

The authors declare no competing financial interest.

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