The study of the graded index anti-reflective coating based on silicon oxynitride

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The main purpose of the present paper is the study of the effect of different parameters of multi-layer anti-reflective coatings ARCs on the reflectivity of solar cells Silicon based. The distribution of the refractive index along different layers is performed following a linear gradient variation from the substrate to the ambient within predefined limits. Firstly the model of Oxynitride multi-layer anti-reflecting coatings was presented which is intended to be used in the evaluation of the performance and the process design of the ARCs in practical applications of solar cells. Secondly the presented model was tested under simulation face to different parameters constraints such as the thickness of the ARCs, the index gradient range variation, the gradient distribution and finally the incidence angle, where the main aim is to improve the quality of the solar cells based on the reflectivity evaluation. The obtained results were also compared to other previous works, whereas focusing on the advantages that have been brought by the proposed model. This paper is concluded by the representation of the main advantages that are needed to be validated by experimental measurements.

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1. Introduction

The materials used for manufacturing solar cells, such as (Si, $GaAs$, $CdTe$, SiO_2, SiN_4, ZnS, MGF_2, Al_2O_3, TiO_2...) have high refractive indices, consequently, the reflection losses are considered to be high. It is well known that the reflection of a silicon surface is over 30% due to its high refractive index. Indeed, it is found that if more than 35% of incident light flux is reflected, this will affect significantly the performance and the efficiency of photovoltaic devices. Therefore, to improve the quality of solar cells, especially in term of efficiency, there are several ways to reduce the optical losses in solar cells, such as:

- Minimization of the top contact coverage of the cell surface, unfortunately this methods may contribute for increasing the series resistance;
- Anti-reflection coatings can be used on the top surface of the cell.
- Surface texturing, this may decrease the reflectivity;
- Increasing the thickness of the solar cell, this can increase the absorption, although the light which absorbs more than a diffusion length from the junction has a low collection probability and will not contribute to the short circuit current;
- The optical path length in the solar cell may be increased by a combination of surface texturing and light trapping.

The present paper is focusing mainly on the method of the antireflection coatings (ARC) based on multi sub-layers where a special property is taken into account for the distribution of the refractive index along used sub-layers. The antireflective coatings (ARC) are generally used on the top surface of the solar cell to reduce the reflection losses. The ARC can be realized by different methods according to various conceptions. It can be simply realized by depositing a dielectric layer on the solar cell. Where, different materials such as (SiO_2, SiN_4, Al_2O_3, TiO_2, ZnS and MGF_2) can be deposited by various methods such as, evaporation, Chemical vapor deposition (CVD), plasma chemical vapor deposition (PECVD), spray pyrolysis, sputtering and other evaporation techniques. These simple antireflection coating can ensure a reduced reflection losses with an average value of 12%, their efficiency does not extend to the entire range of the solar cells sensitivity [1],[3]. More complex structures based on multilayer antireflective coatings: V_{coat}, W_{coat} ... can achieve better results, but their realization is more difficult [2]. Another way of realizing antireflective coating is consisting of depositing a dielectric layer with a refractive index which decreases gradually from the substrate to the environment [4]. Theoretically, if the index decreases from n_s to n_0 the reflection will be nil [1-4]. Unfortunately, this cannot be practically feasible as the materials with high optical index are generally absorbent.

In the present paper the last abovementioned category of antireflective coating is used. Where, the Silicon Oxynitride layers with graded index are investigated and implemented for solar cells application. Indeed, SiOxNy was proved to be effectively ay very interesting antireflection material, which could also efficiently ensure the protection of the surface and the interface of semiconductor devices, this can lead to significant enhancement of the devices lifespan. On the other side, it was also proved that the Oxynitrides based antireflection coatings might be more cost effective compared with other antireflection coatings. In the present paper SiOxNy will be investigated as a multi-layer antireflection under special refractive index distribution represented by antireflection coatings with linear graded index that were made by PECVD deposition of silicon Oxynitride (SiN_xO_y) [5-6]. Measurements of reflectance were recorded. A theoretical model was developed to study the behavior of these AR coating at normal and oblique incidence. Comparison of theoretical curves with experimental measurements of reflectance factor was used to validate the model. A comparative study between the performance of a graded index ARC and that of a traditional Single-Coating AR (silica or silicon nitride) shows clearly the improvement made by the presented ARC from point of view of the sensitivity towards the thickness of layers and the angle of incidence. This study is based on the determination of the optimal values of sensitive parameters (Thickness, index etc.) for the implementation of graded index ARC. It is obvious that the multi-layers ARCs allow improving the efficiency of solar cells with oblique incidence, which can be obtained with simple implementation methods. In Fig. 1 the reflectivity $R(\lambda)$ is presented using one ARCs layer for different materials (MgF_2, SiO_2, Al_2O_3, TiO_2) with their optimum thickness, similar results are obtained in [7]. Indeed these antireflective coatings materials are commonly used with crystalline silicon solar cells, their refraction index are given for precise value of the wavelength in Table. 1. As it is shown, the best ARC can be obtained with TiO_2 ($d = 60nm$). In the same time the minimum value for the different materials is also obtained nearly around the same value of the wavelength $\lambda_{opt} = 550nm$.

Table 1. The optimum reflectivity for different materials combination of double layer ARCS.

Materials	TiO_2	Al_2O_3	SiO_2	MgF_2
$d_{opt}(nm)$	60	72	94	100
$R_{min} = R(\lambda_{opt})$	0.06903	3.669	6.616	9.501

The reflectivity $R(\lambda)$ can be calculated as follows [4],[6-9]:

$$R(\lambda) = \frac{r_1^2 + r_2^2 + 2r_1r_2\cos(2\theta_1)}{1 + r_1^2r_2^2 + 2r_1r_2\cos(2\theta_1)} \quad (1)$$

Where:

$$r_1 = \frac{n_0 - n_1}{n_0 + n_1} \quad r_2 = \frac{n_1 - n_2}{n_1 + n_2} \quad \theta_1 = \frac{2\cdot\pi\cdot d_1\cdot n_1}{\lambda}$$

It is well known that among the terrestrial solar spectrum around the wavelength $\lambda = 500nm$ has the peak energy, on the other side the relative spectrum response of silicon cells has the peak value within the wavelength range of $\lambda = 300 - 900\ nm$, Hence the best anti-reflection is located within the wavelength range of $\lambda = 500 - 700\ nm$. In the present work the reflectivity study take into account the wavelength range of $\lambda = 400 - 800\ nm$ to cover all possible regions of the best energy spectrum quality. The dependence of the reflectivity on the three parameters the wavelength, the thickness and the refractive index for single layer ARCs is shown in Fig. 2(a), Fig. 2(b), Fig. 2(c).

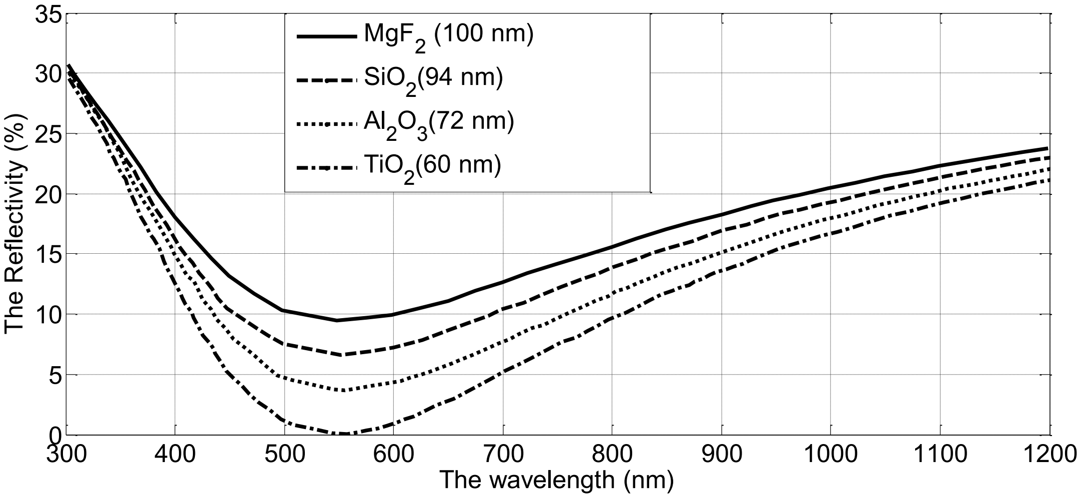

Fig. 1. The reflectivity of single layer antireflection coating.

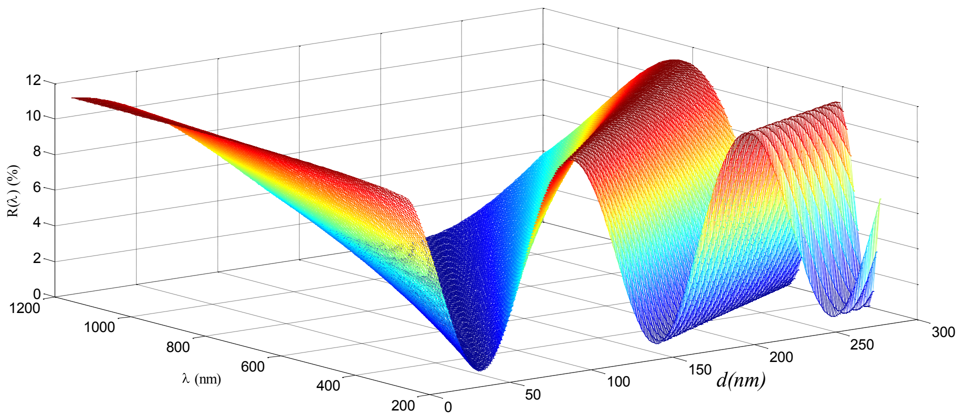

Fig. 2(a). The reflectivity versus the thickness and the wavelength ($n_1 = .38$).

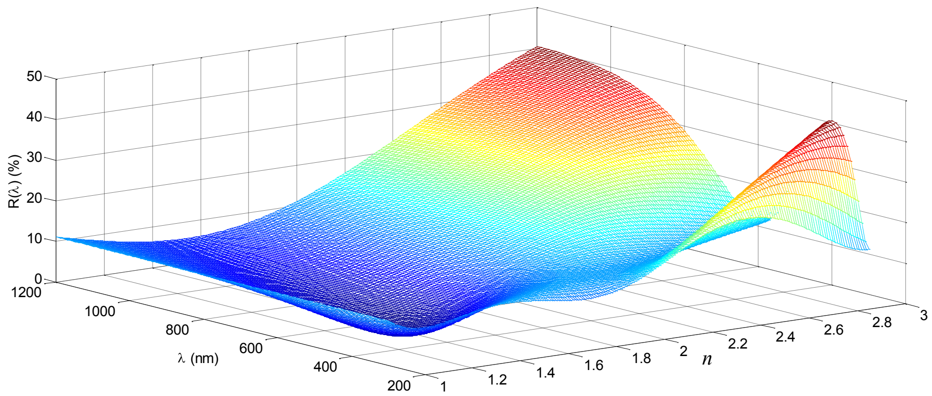

Fig. 2(b). The reflectivity versus the refractive index and the wavelength ($d = 00\,nm$).

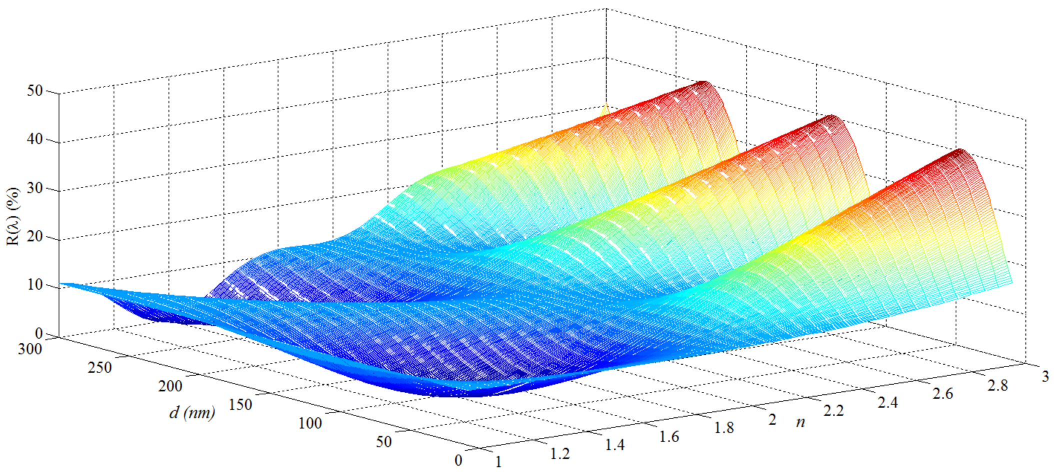

Fig. 2(c). The reflectivity versus the refractive index and the wavelength ($\lambda = 550\,nm$).

Among all antireflection coatings Materials, TiO_x is one of commonly used ARCs for the preparation of crystalline silicon solar cells. Where it is usually used as an ideal ARCs due to its high refractive index, and its transparent band center which coincides perfectly with the visible solar spectrum. On the other side; the silicon nitride (SiO_x) is another commonly used ARC. It is well known that the SiO_x film has been widely used in semiconductor production as an efficient surface passivation layer which can contribute strongly for the improvement of the conversion efficiency of the solar cells [8]. Hence; recently the use of SiO_x thin film as antireflection coatings has attracted researchers to be included in several applications, especially in solar cells [7-11]. In Fig. 3 the reflectivity $R(\lambda)$ is presented using double layer ARCs with the combination of the aforementioned materials such as ($MgF_2 + Al_2O_3$, $MgF_2 + TiO_2$, $SiO_2 + Al_2O_3$, $SiO_2 + TiO_2$) where the thickness of each material layer is kept the same as in Fig. 1, Table. 2 is representing this materials, the thickness, and the minimum reflectivity obtained versus the wavelength. The reflectivity $R(\lambda)$ can be calculated as follows [7-10]:

$$R(\lambda) = \frac{r_1^2 + r_2^2 + r_3^2 + r_1^2 r_2^2 r_3^2 + R_1(\lambda) + R_2(\lambda)}{1 + r_1^2 r_2^2 + r_1^2 r_3^2 + r_2^2 r_3^2 + R_1(\lambda) + R_2(\lambda)} \quad (2)$$

Where:

$$R_1(\lambda) = 2r_1 r_2 (1 - r_3^2)\cos(2\theta_1) + 2r_2 r_3 (1 - r_1^2)\cos(2\theta_2)$$

$$R_2(\lambda) = 2r_1 r_3 \cos 2(\theta_1 + \theta_2) + 2r_1 r_2^2 r_3 \cos 2(\theta_1 - \theta_2)$$

$$r_1 = \frac{n_0 - n_1}{n_0 + n_1} \quad r_2 = \frac{n_1 - n_2}{n_1 + n_2} \quad r_3 = \frac{n_2 - n_3}{n_2 + n_3}$$

$$\theta_1 = \frac{2 \cdot \pi \cdot d_1 \cdot n_1}{\lambda} \quad \theta_2 = \frac{2 \cdot \pi \cdot d_2 \cdot n_2}{\lambda}$$

The reflectivity $R(\lambda)$ of the double layer ARCS for the four materials are given in Fig. 3. It is obviously shown that each combination represents two minimum, where these minima are obtained nearly around the wavelength of $\lambda = 430\ nm$, contrary the other minima that are presenting the global minima of all the combination within a large range of the wavelength $\lambda = 730 - 930\ nm$, Hence the double layer ARCS of $SiO_2 + TiO_2$ can be considered to be the best with $(\lambda = 440\ nm, R = 0.9951)$ and $(\lambda = 820\ nm, R = 0.0890)$. Similar results were obtained in [7].

Table 2. The optimum reflectivity for different materials combination of double layer ARCS.

Materials $d_{1opt} + d_{2opt}(nm)$	$MgF_2 + Al_2O_3$ $100 + 72$	$MgF_2 + TiO_2$ $100 + 60$	$SiO_2 + Al_2O_3$ $94 + 72$	$SiO_2 + TiO_2$ $94 + 60$
$R_{min1} = R(\lambda_{opt1})$ $\lambda_{opt1}(nm)$	3.5974 420	0.6375 460	3.8634 410	0.9951 440
$R_{min2} = R(\lambda_{opt2})$ $\lambda_{opt2}(nm)$	2.195 880	0.2709 730	1.446 930	0.0890 820

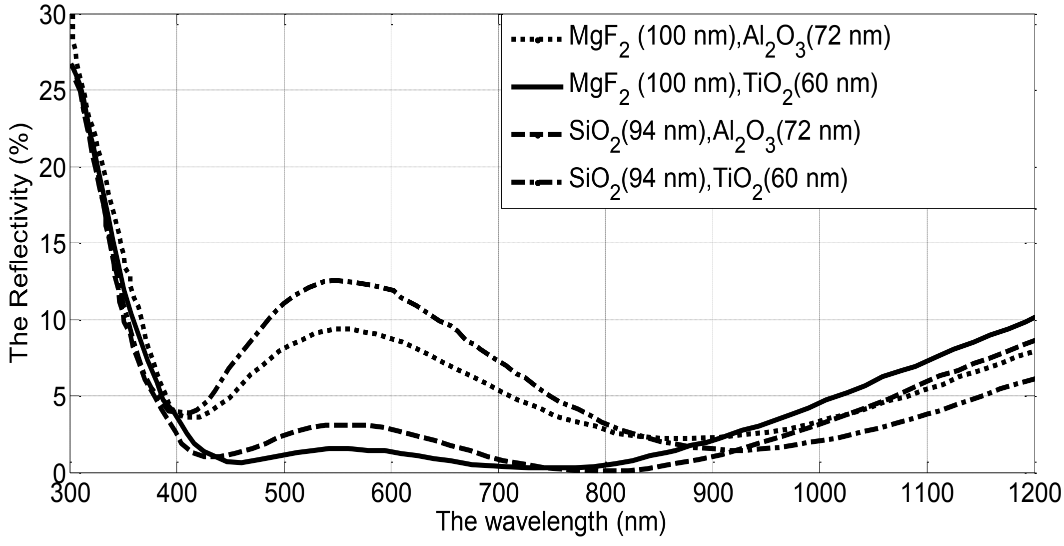

Fig. 3. The reflectivity of double layer antireflection coating.

Table 3. The refractive index of common anti-refractive materials [8], [11]

Materials	**Refraction index (** n_i **)**
MgF_2	1.38
SiO_2	1.46
Al_2O_3	1.76
Si_2N_4	2.05
Ta_2O_3	2.2
ZnS	2.36
SiO_x	1.8 – .9
TiO_2	2.62

The wavelength 590nm (the corresponding energy is 2.1eV) was used in calibration.

2. Theoretical Model

The model used allows the calculation of the reflection factor R of multi-layer structure dielectric versus the wave length of the incident light. For these calculations, the method of the characteristics matrixes is used [3],[12-18]. This method takes into account firstly the electromagnetic field evolution E and H versus their position in the structure. The propagation is generally understood in each layer and is represented by a single matrix M_j called characteristic matrix. The reflected field is then calculated from the global field. This method has the advantage of a very simple formulation. The Oxynitride layer gradient is represented by a stack of n layers of equal thickness d_j, isotropic, transparent, with indices n_j gradually decreasing from the substrate to the environment. The substrate and the environment are considered semi-infinite.

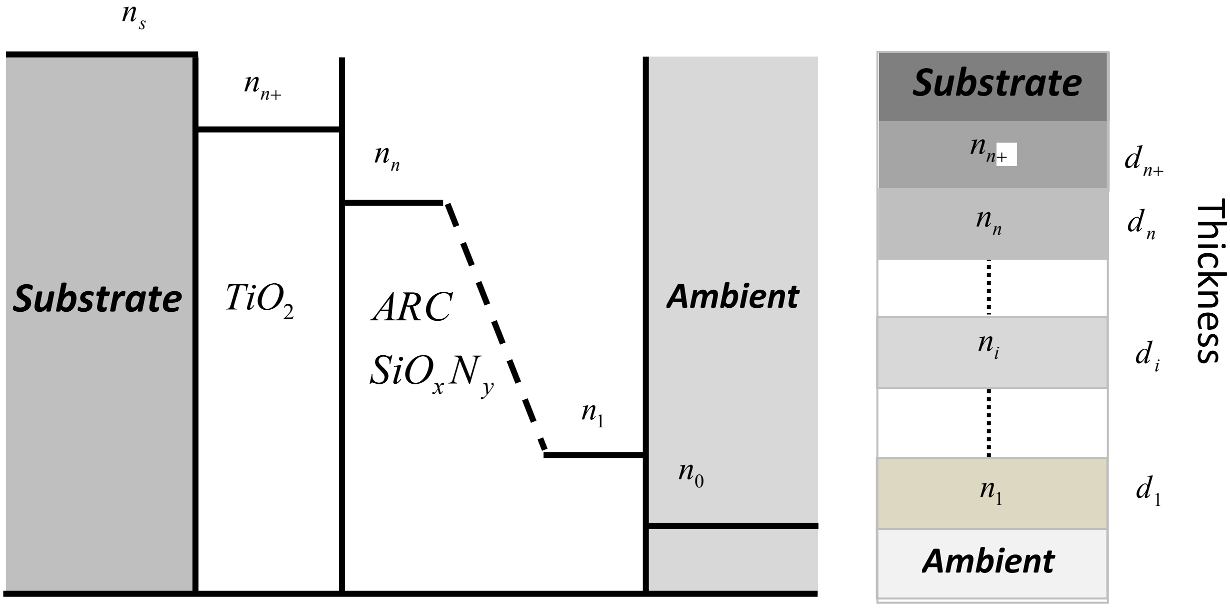

Fig. 4. A multi-layer dielectric system

2.1Reflectivity calculation

$n_s = \imath_s + k_s$ it is complex and varies with the wavelength. $n_0 =$ for the air. The characteristic matrix of the J^{th} layer is expressed as:

$$M_j = \begin{bmatrix} \cos\beta_j & -i\cdot\dfrac{\sin\beta_j}{p_i} \\ -i\cdot p_j\cdot\sin\beta_j & \cos\beta_j \end{bmatrix} \qquad (3)$$

Where:

$$\beta_j = \frac{2 \cdot \pi \cdot n_j \cdot d_j}{\lambda} \cdot \cos\Phi_j \qquad (4)$$

$$p_j = \begin{cases} n_j \cdot \cos\Phi_j & polarized\ wave\ type\ (S) \\ \dfrac{n_j}{\cos\Phi_j} & polarized\ wave\ type\ (P) \end{cases} \qquad (5)$$

The Φ_j is determined from the Snell-Descart law:

$$n_0 \cdot \sin\theta = n_j \cdot \sin\Phi_j \qquad (6)$$

For the two types of polarization the following expression can be written as:

$$\begin{bmatrix} E_r \\ E_i \end{bmatrix} = \frac{1}{2} \cdot \begin{bmatrix} 1 & -\dfrac{1}{n_0} \\ 1 & \dfrac{1}{n_0} \end{bmatrix} \prod_{j=1}^{n} M_j \cdot \begin{bmatrix} 1 \\ \dfrac{1}{n_s} \end{bmatrix} \qquad (7)$$

E_r : Reflected field, E_i : Incident field;

p_j is written differently in M_j according to the type of polarization. For polarized wave type (S) the reflection factor is give as follows:

$$r_s = \left(\frac{E_r}{E_i} \right) \qquad (8)$$

Hence; the reflectivity can be deduced from the following expression:

$$R_s = r_s \cdot r_s^* \qquad (9)$$

For polarized wave type (P) the reflection factor and the reflectivity are given respectively as follows:

$$r_p = \left(\frac{E_r}{E_i} \right)_p, \qquad (10)$$

$$R_p = r_p \cdot r_p^*$$

Where: r^* and r_p^* are the conjugates of r and r_p respectively.

For non-polarized light, like the sunlight the reflectivity can be calculated by the following expression:

$$R = \frac{R_s + R_p}{2} \qquad (11)$$

It is important to note here that this model can be used for finding the theoretical curve $R(\lambda)$ for different incidence angles with the whole multilayer systems. In the present paper, the first step is definite the quality of the refractive index variation along the different layer materials used in the ARCS. Indeed the variation of the reflective index with the number of layers of the ARC is based on a linear variation, this kind of ARC is called multi-layer graded index, where the main aim is to minimize the reflection losses. In this case, the reflection index n_j is determined by the proposed following expression:

$$n_j = n_1 - \frac{j-1}{n-1} \cdot (n_1 - n_n) \qquad (12)$$

The n_j index is considered real and constant for each layer. In this study, the method of Bruggeman is used, it allows the determination of the optic index of each layer versus the wavelength.[19-20] . On the other side; with this method the gradient profile can be modified easily and similar results can be obtained.

2.2 Determination of layers number n for the used ARC

The $R(\lambda)$ curves have been carried out by varying the number of sub-layers constituting the ARC with linear gradient for varying thicknesses. It is noticed that starting from a number of sub-layers of $n = 20$ the best minimum value of the reflectivity is obtained and a very tiny variation towards from best minimum can be obtained by adding high number of sub-layers, it can be said that a convergence toward the desired value is reached or there is a saturation for getting best minimum value. This number of layers is chosen for the ARCs used in the present work.

3. The Model Validation

To validate the model presented in this paper, the obtained simulation results has been compared with experimental measurements. Therefore, a sample was implemented by PECVD from a mono-crystalline Si substrate on which was deposited a silicon oxynitride ($SiNO$) with a thickness of 260 nm, its index range varies from 1.95 on the surface substrate to 1.47 of the ambience surface based on a linear variation. Measurements of the reflectance factor $R(\lambda)$ with normal incidence have been reported. Figure .5 shows the reflectivity of the theoretical and the experimental results of graded index ARC and the bared mono-crystalline silicon Si . As it can be seen clearly, the reflectivity is decreased remarkably with the use of the ARC, on the other side; the experimental and theoretical results of the reflectivity prove clearly the validity of the theoretical model sued in the simulations. The minimum value is obtained for both models at $\lambda = 590\ nm$ with

$R(\lambda_{ry}) = 2.7953$ and $R(\lambda_{ry}) = 2.0866$ for the experimental and the proposed theoretical model respectively, a deviation of $\Delta R(\lambda_{ry}) = 0.7087\,\%$ is occurred which explain the accuracy and the matching of the theoretical model to the real model. This wavelength $\lambda = 590\ nm$ is very close to the peak power of the solar spectrum. It is important to note that the using of the ARC has improved greatly the reflectivity, it can be remarked that a deviation of $\Delta R(\lambda_{ry}) = 35.84\,\%$ between the results of the experimental results. Hence, the use of the graded index ARC is solar cells can decreases drastically the losses that can be chiefly effect the power by lowering the short-circuit current.

In the study of the behavior of the graded index multi-layers ARCS, the main sensitive parameters of the coating structure are:

- The thickness of the ARCs, concerning the coating thickness $d\,(nm)$ and the thickness of each layer $dj(nm)$, where: $d\,(nm) = \sum_{j=1}^{n} d_j\,(nm)$;
- The wavelength of the incident light $\lambda\ nm)$;
- The range of the refractive index n_j variation in different layers between the substrate and the ambient;
- The angle of incidence θ on the contact surface with the ambient;
- The distribution of the index gradient along the different layers of the coating.

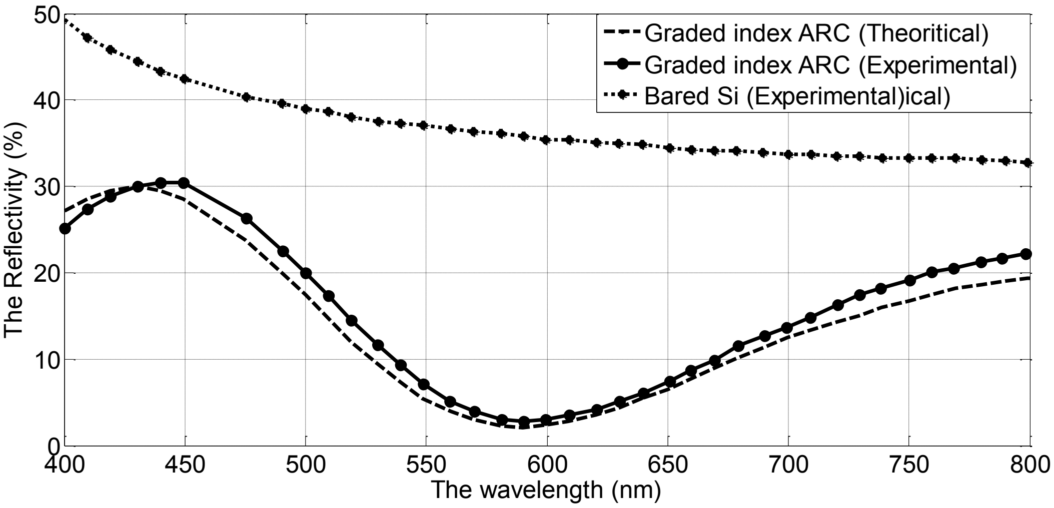

Fig. 5. The reflectivity for the bared Si and graded index ARCs model.

The obtained reflectivity of such coating is depending on these structural parameters. However for solar cell , the spectrum distribution of incident light can be known previously depending on the location and the position of the solar cell surfaces, consequently the desired reflectivity $R(\lambda$ of the multi-layer ARCS can be achieved based on the values of the basic parameters n_j, $dj(nm)$, $d\,(nm)$. On the other side, as it was mentioned in equation (12) the index gradient is taken to be linearly graded based on the two limit values of the first layer refractive index n_1 and the last layer refractive index n_n Fig. 6. The all n layers of the ARCs are considered to have equally thickness distribution among all the layers, therefore each layer has a thickness $dj(nm)$ as follows:

$$d_j\,(nm) = \frac{d\,(nm)}{n} \tag{13}$$

In the present work it is demonstrated that $n = 20\ layers$. Which means that:

$$d_j\,(nm) = \frac{d\,(nm)}{20}$$

The present work is focusing mainly of the effect of the basic parameters such as: the thickness of the ARCs $d\,(nm)$, the limits of the refractive index n_1 and n_n that effect the refractive index gradient variation along the whole layers and the angle of incidence on the reflectivity of the proposed graded index principle of the multi-layer ARCs.

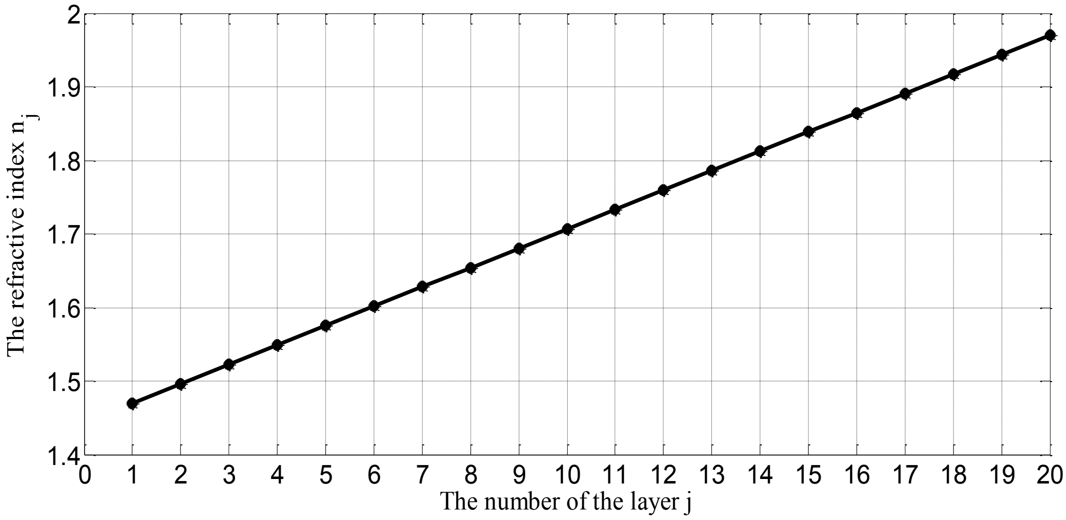

Fig. 6. The refractive index of each layer.

4. The Study of the ARC with linear gradient

To understand the influence of the aforementioned parameters on the reflectivity of multi-layer ARCS under graded index or linear gradient refractive index vacation, the effect of each of the parameters has been studied separately. The range of the wavelength variation is taken $\lambda\ nm) = 400 - 800$ which is including practically the solar spectrum distribution.

4. 1 The Effect of the thickness variation

Figs .7 and .8 present the reflectivity versus the wavelength $R(\lambda)$ for different thickness ($d\,(nm)$). Where the limit values of the refractive index has been taken as $n_1 = 1.5$ and $n_n = 2.0$, while the conditions presented in equations (12) and (13) are taken into account. It can be clearly remarked from Fig. 6 that the increase of the ARCs thickness $d\,(nm)$ implies the increase of the wavelength which achieve the minimal reflectivity $R_{\min}(\lambda_{ry})$, the same remark can be deduced from Fig. 7. Whereas the values of $R_{\min}(\lambda_{ry})$ with greater value of the thickness $d\,(nm)$ are better, this can be seen clearly in Fig. 9. Therefore the values of the thickness with the range $d\,(nm) = 240 - 280$ allow to ensure less light losses under less reflectivity value within the wavelength range $\lambda\ nm) = 400 - 800$ which can fulfill the requirement imposed by the solar spectrum distribution. Consequently the Oxynitride multi-layer ARCs is less sensible to the variation of the thickness within an acceptable range of variation, this can be considered as a good advantage of using such ARCs.

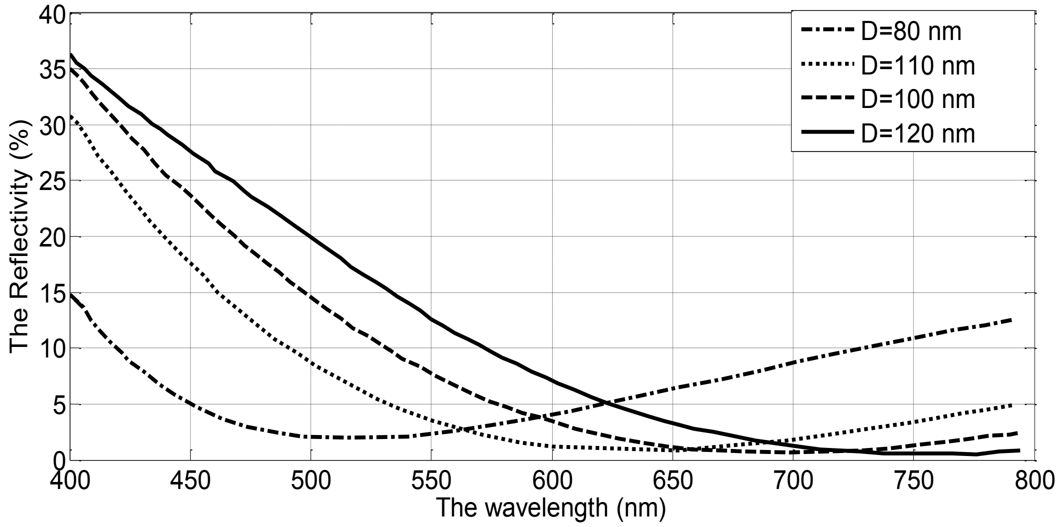

Fig. 7. The effect of the thickness on the ARCs graded index reflectivity.

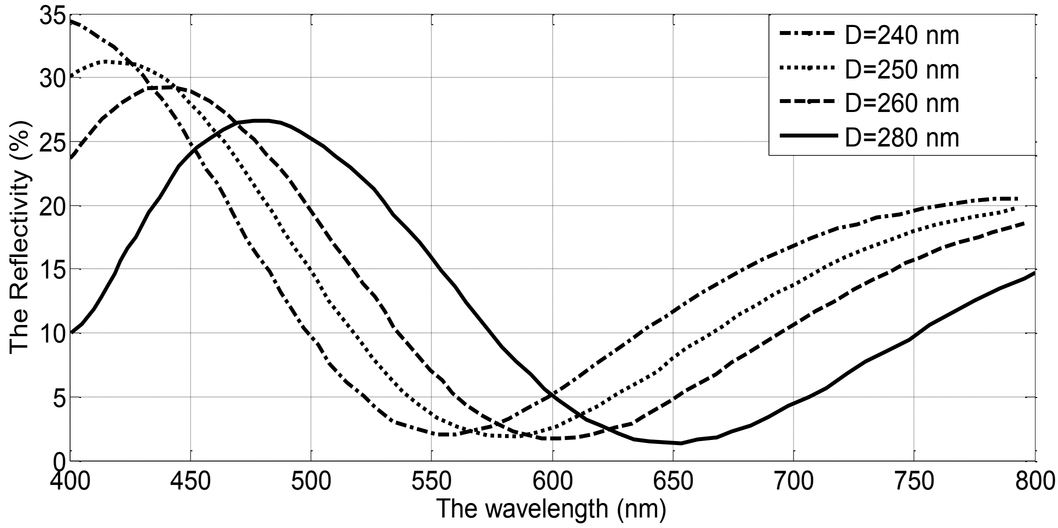

Fig. 8. The effect of the thickness on the ARCs graded index reflectivity.

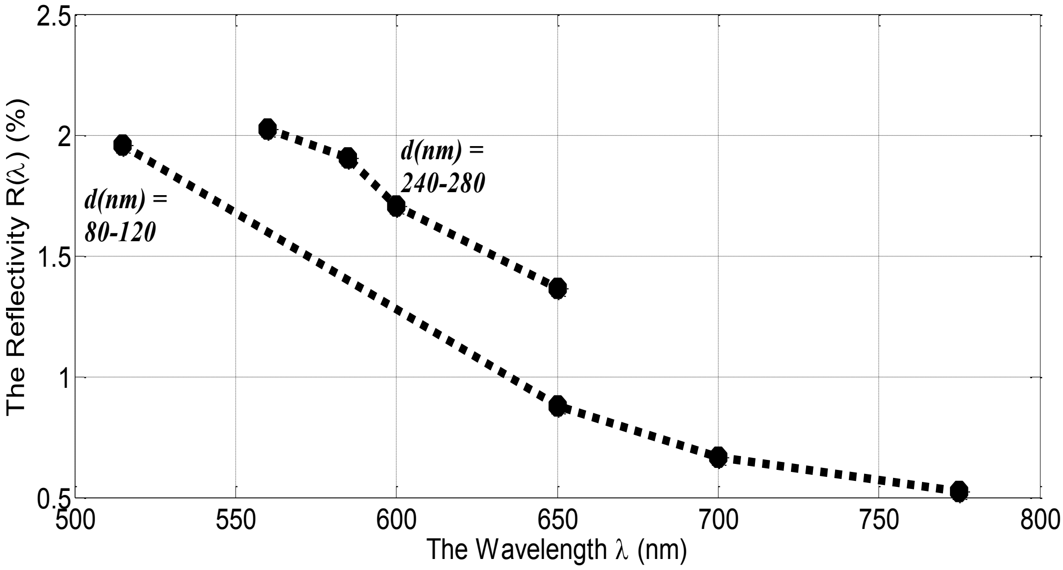

Fig. 9. The effect of the thickness on the reflectivity $R_{\min}(\lambda_{opt})$.

4.2 Effect of the gradient variation

The figs. 10 and 11 present the curves of the reflectivity versus the wavelength ($\lambda\ nm) = 400 - 800$) for different values of n_1 and n_n. It is obvious that the refractive index of the intermediary layers is varying linearly following the expression presented in equation (12). The analysis of the mentioned effect is performed under three rang of reflective index variation $\Delta = n_n - n_1$ under two different values of the ARCs thickness which is taken equal to 260 nm in Fig. 10 and equal to 100nm in Fig. 11.

These curves show that an increase of the gradient limits results in a decrease of the reflectivity minimum for all wavelengths with a shift toward the largest wavelengths. It can be seen clearly that with the greater value of Δ (1.45 -2.50) a nearly ideal case of reflectivity can be reached at $\lambda_{opt} = 560\ nm$ with $R_{\min}(\lambda_{opt}) = 0.1685$. Contrary for the ARCs thickness value of 100 nm, the reflectively of different graded index have approximately the same behaviors Fig. 11. The proposed linear variation of the refractive index within the different layers of the presented ARCs allows to decrease the reflectivity within acceptable practical value of wavelength variation which presents another advantage of using the Oxynitride multi-layer ARCs.

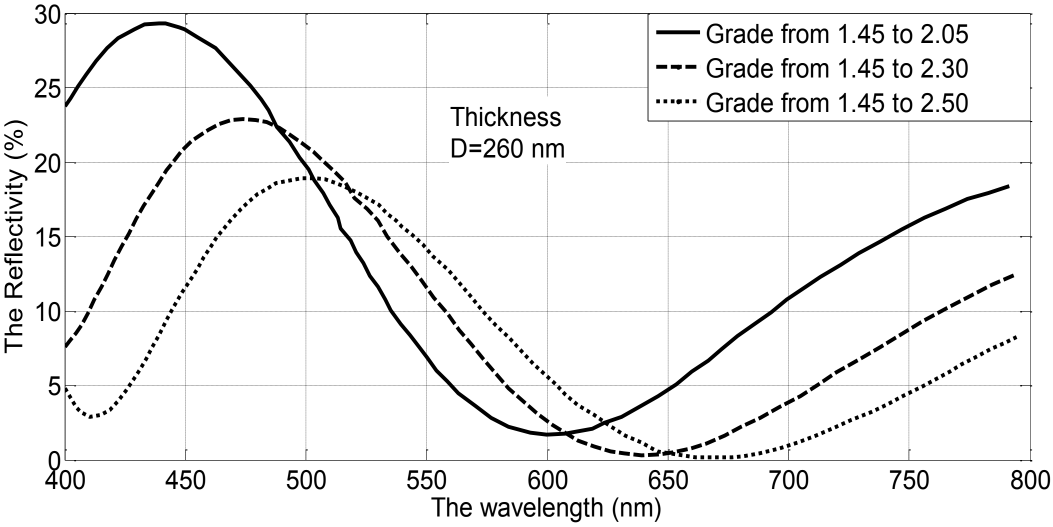

Fig. 10. Effect of the gradient variation on the reflectivity of a linear graded ARC with thickness of 260nm

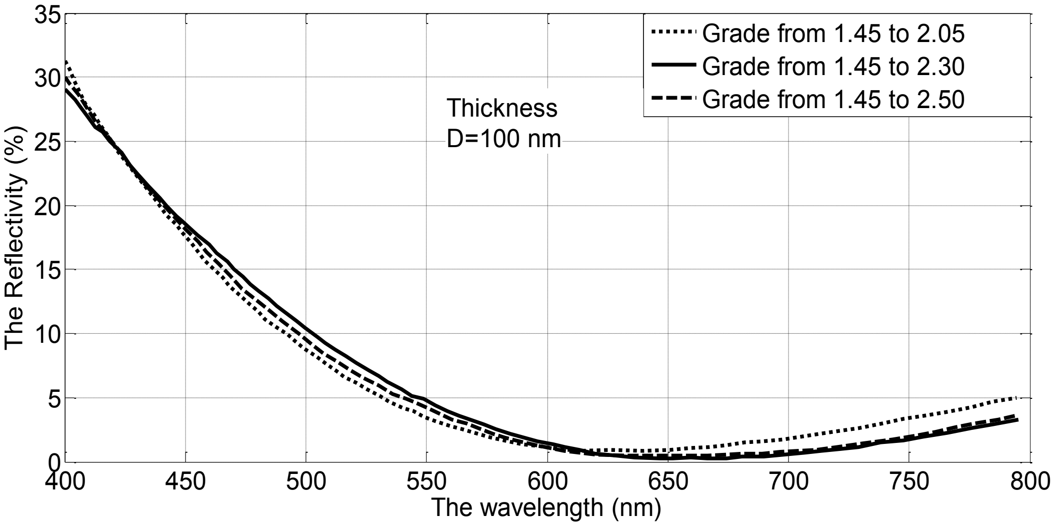

Fig. 11. Effect of the gradient variation on the reflectivity of a linear graded ARC with thickness of 100nm

4.3 The Effect of the incidence angle

The figs. 12 and 13 present the curves of the reflectivity versus the wavelength ($\lambda\ nm) = 400 - 800$) for different values of the incidence angle. Where the limit values of the refractive index has been taken as $n_1 = 1.5$ and $n_n = 2.0$, while the conditions presented in equations (12) and (13) have been taken in consideration. It is clear that with the increase of the incidence angle the minimum reflectivity decrease towards the best minimum, while the corresponding wavelength increases Fig. 12. The same remark can be deduced from Fig. 13. In Fig. 14 the effect of the incidence angle under both values of the ARCs thickness are presented. The reflectivity under the thickness $d\ (nm) = 260$ is considered to be better based on the wavelength range variation which can fulfill the requirement of the solar spectrum distribution. It can be concluded that the use of the Oxynitride ARCs can be used face to the incidence angle variation to keep better efficiency of the solar cells.

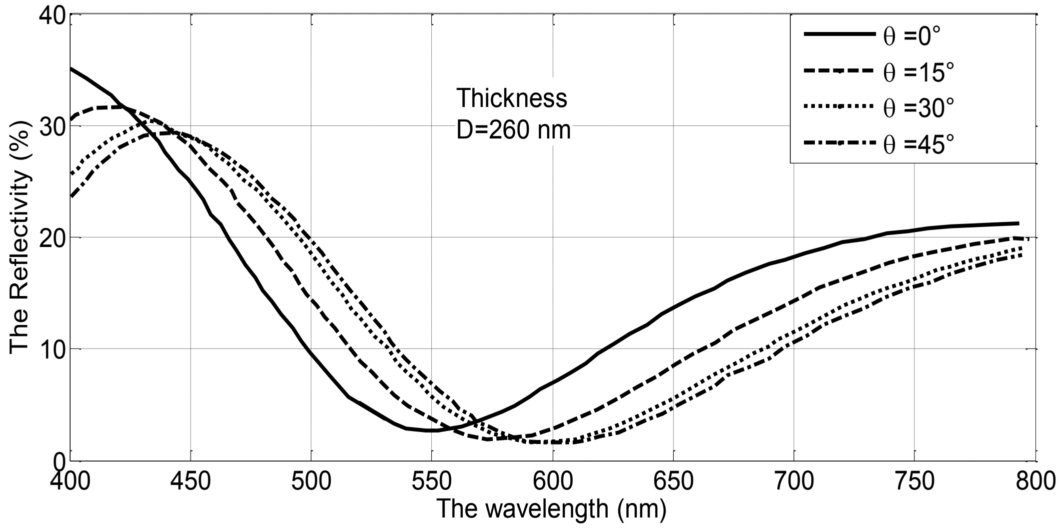

Fig. 12. Effect of the incidence angle variation on the reflectivity of a linear graded index ARCs with thickness of 260nm

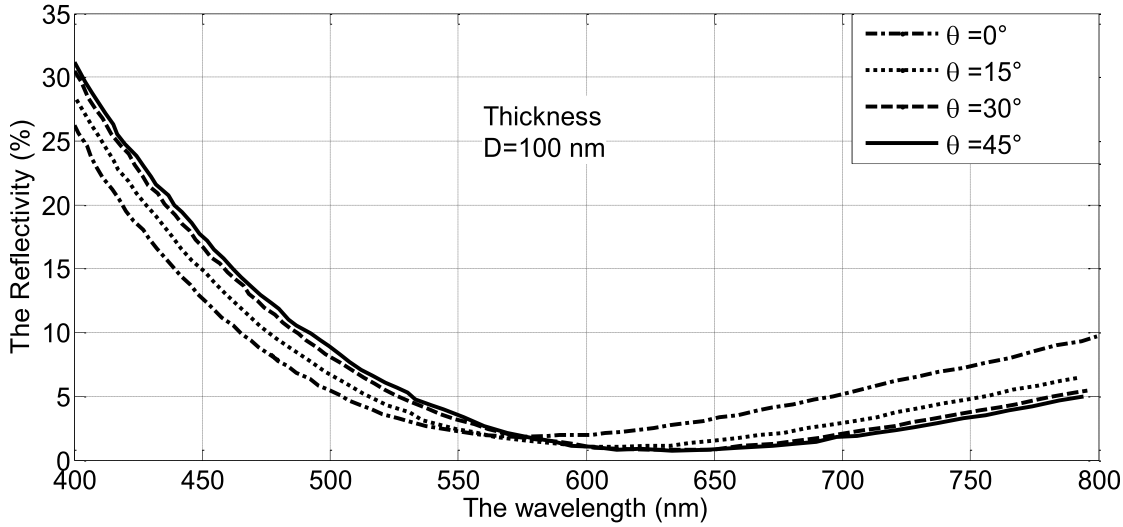

Fig. 13. Effect of the incidence angle variation on the reflectivity of a linear graded ARC with thickness of 100nm

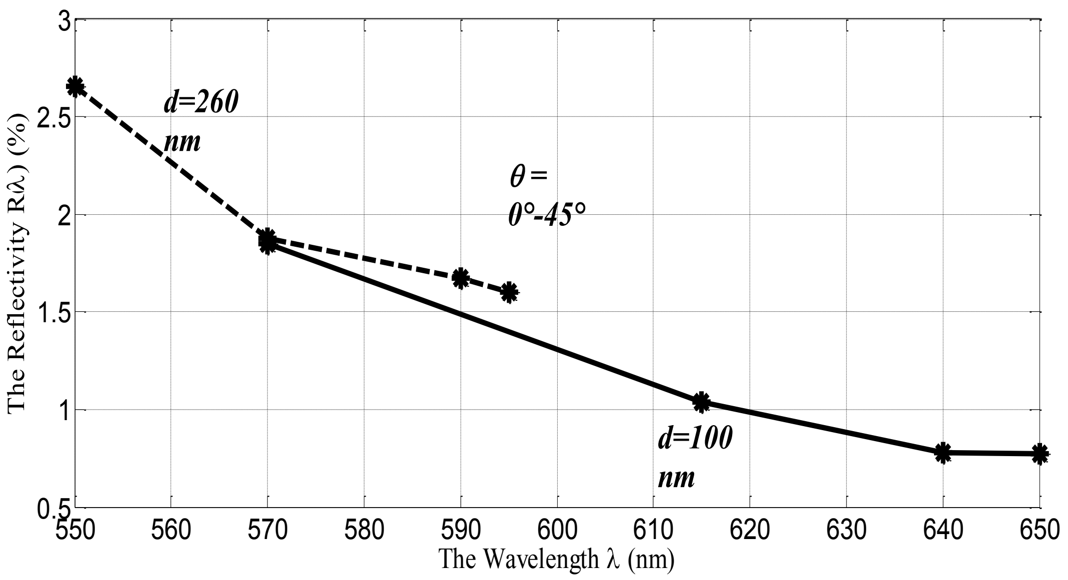

Fig. 14. Effect of the incidence angle variation on the reflectivity of a linear graded ARC with thickness of 100nm

5. Conclusion

In the present paper the multi-layer anti-reflective coatings (ARCs) with linear graded index of the Oxynitride materials have been studied under different constraints of thickness variation, range variation of the graded index and the incidence angle variation, it is found that:

- The Oxynitride ARCs graded index is less sensitive to the thickness variation.
- Their behavior face to the oblique incidence angle is better.
- The linear graded index behaviors can greatly increase the efficiency of the solar cells, decrease the reflectivity, hence; minimize the light losses

These results have been confirmed by using the simulator of solar cells PC1D version 5. However measurements on real solar cells are needed for more credibility. The performance of the ARC graded index can be improved by the interposition of the silicon Oxynitride and a quarter wave layer TiO2. The first results are very encouraging compared to the previous published results, while the theoretical obtained results need to be confirmed by experimental measurements.

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