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Impact of the size of silver nanoparticle integrated in an ARC based on silicon

Hichem Hachemi [a], *Abdellatif Zerga* [a], *Noureddine Sahouane* [b], *Batoul Benabadji* [a]

[a] *Third-generation Solar Cells, Research Unit of Materials and Renewable Energies (URMER), Abou BakrBelkaïd University, B.P. 119, Tlemcen, Algeria*

[b] *Research Unit in Renewable Energy in the Saharan environment URER.MS, Center for the development of renewable energies, CDER, Algeria*

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ABSTRACT

The controlled use of surface plasmon resonances in metallic nanostructures has attracted considerable interest in several areas, including photovoltaic devices. The diffusion of metal nanoparticles is a well-known phenomenon and has long been debated. The relative extent of absorption and dispersion depends on the size of the nanoparticles. The deposition of metal nanoparticles on the surface of the photovoltaic cell makes it possible to increase the absorption of the incident light and to reduce the optical losses. The most commonly used metal nanoparticle deposition techniques are: Bottom-up and Top-Down.

In this paper, we study the effect of the size of silver nanoparticles (np-Ag) integrated in an antireflection layer of silicon nitride. The plasmonic effect converts scattered ultraviolet rays (absorbed by np-Ag) into visible radiation. The use of the DDSCAT software has made it possible to calculate the dispersion and absorption of electromagnetic waves by nanoscale targets with arbitrary geometries using the dipole discrete approximation (DDA). DDA is used to model these particles arranged on a cubic lattice, as interactive sets of dipoles. The variance in np-Ag size allowed us to determine the optimal size that absorbs maximum UV radiation. The simulation results show that the optimal size of np-Ag is 110 nm.

1 Introduction

The intense research in the field of nanoparticles is motivated by the study of new materials to miniaturize more the electronic devices and increase efficiency [1]. The electromagnetic properties of metal particles have long been known, Wood [2] and Ritchie [3], and there's been great interest in last year's following the development of new techniques to deposit these nanoparticles. The plasmonic metal nanoparticles phenomenon has been studied in many applications such as optoelectronics imaging, photovoltaic and photo detection [4] because of its unique performance in a certain range of

* *Corresponding author. Tel.: +213 698210386 / 213 556548081.*
E-mail address: hichem.hachemi@yahoo.com

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plasmon resonance or wavelength. Indeed, the most significant losses in solar cells are due to the low absorption of photons of shorter wavelengths that are dissipated as heat. These losses represent 28% of the total loss figure (1).

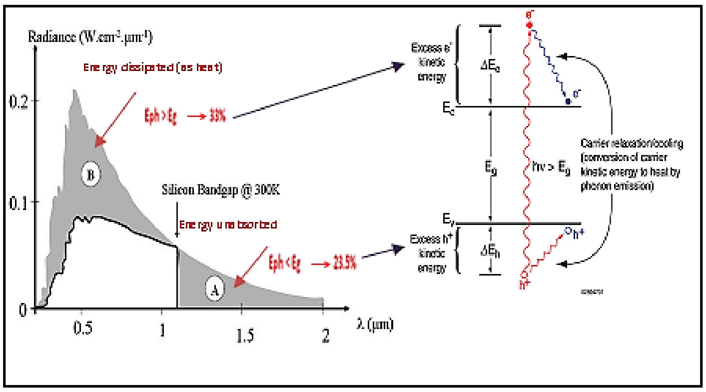

Fig .1 - Major intrinsic losses for a silicon photovoltaic cell. 1 - Loss of photons of longer wavelengths. 2 - Losses due to the excess energy of the photons [5]

In recent years, plasmonic particle layers (PPLs) made up of metal nanoparticles have been extensively studied as a way of improving solar cells [6–8]. Nanoparticles can efficiently scatter light [9–11] thereby increasing the optical path length of the incident light. Light coupling into the solar cell structure may be further enhanced if the PPL results in a high density of optical modes. This enables efficient thin film technology [12–15] and furthermore increases the absorption efficiencies of standard cells. Specific efforts have been made to incorporate Ag nanoparticles as Ag PPLs since Ag has relatively low absorbance in the wavelength range of interest [11]. In the most cost effective methods, Ag metallic nanoparticles (MNPs) are obtained chemically or by auto-aggregation of thin Ag layers [16, 17]. The first method can be applied to solar cells by spin coating [18, 19]. It has recently been proposed that the resulting material matrix incorporating Ag MNPs can be optimized as antireflective coating (ARC) [16, 20–22]. This type of structure usually shows an aging of Ag PPL [23] and therefore some authors have proposed the addition of a protective layer to avoid the degradation [24]. The optical response of the resulting system of PPL and enclosing matrix requires careful optimization in order to maximise light interaction with the underlying solar cell. Of particular importance is the separation between the PPL and the substrate, and consequences for Fano [25] interference losses between scattered and incident light. Localized surface plasmons are collective oscillations of the conduction electrons in the metal particles. The movement of the conduction electrons upon excitation with incident light leads to a bias charge accumulation on the surface of the particles. The metal support surface plasmons which are excited collective oscillation of free electrons, these oscillations are characterized by a resonance frequency. They can be located, for the metal nanoparticles or multiplication as in the case of planar metal surfaces. Precious metals resonances are mostly in the visible or infrared region of the electromagnetic spectrum.

MIE [26] was the first to explain this phenomenon. The properties of metallic nanoparticles have been studied within detail by Henglein and Kreibig [27–30]. The effect of variations in the size of nanoparticles about plasmon absorption in relation with the Mie theory is the major advantage [28, 30]. The silver nanoparticles (np-Ag) have much attracted attention of researchers during the last decade due to their small size and their new properties [31]. Their sizes nano scale makes suitable for many new applications with different properties of materials by volume [32]. A recent study shows that improving the efficiency of solar cells using TiO_2 and SiO_2 matrix encapsulate gold nanoparticles [33]. In this study, we chose silicon nitride (SiN) which is generally used as an antireflective layer in solar cells [34]. The following work presents a study often the absorption of silver nanoparticles (np-Ag) implanted in a silicon nitride matrix (SiN) from silicon with thickness of 78 nm.

2 Modeling of the effect of the size of silver nanoparticle

The controlled use of surface plasmonic resonances in metal nanostructures has raised a huge interest with regard to several application fields, of which one is photovoltaic devices. In this context, arrays of metal nanoparticles (NPs) (often referred as “plasmonic layers”) located at the front surface, in the active zone or at the rear surface of photovoltaic cells, are investigated as a means to enhance radiation absorption in the cell via mechanisms such as light scattering and nearfield effects [1]. For example, it has been demonstrated that silver or aluminum nanoparticles (NPs) at the front side of crystalline solar cells (c-Si) enhance the fraction of light Plasmonics reaching the active part of the device, boosting the total efficiency [2]. The scattering of metal nanoparticles is a well known phenomenon and has long been debated in many references. The relative extent of absorption and distribution depends on the size. For photovoltaic applications, the particle size must be optimized to ensure maximum propagation [35]. Bottom-up and Top-Down are two techniques for depositing nanoparticles in dielectric matrices. In order to deposit them, several objectives must be achieved, in particular the size and shape [36].

Our goal is to assess the role of the dielectric environment by comparing the effect of plasmonic layers either supported on- or embedded within the ARC. Among the different nanoparticles used, we find silver nanoparticles, that is a noble metal but more reactive than gold. Flexible enough, its mechanical strength is improved by the addition of copper and has excellent conductivity. Thus, it is well known for its ability to form small aggregates nuclearity in various matrices and supports. To assess the role of dielectric with plasmonic effect, we integrated np-Ag in an antireflection layer based SiN deposited on a surface of the silicon cell. We used the DDSCAT software to calculate the dispersion and absorption of electromagnetic waves by nano scale targets with arbitrary geometries using DDA (dipole discrete approximation). DDA is used to model these particles disposed on a cubic lattice, as interacting sets of dipoles [37]. This leads to solve a system of linear equations whose solution gives all values dipoles for giving outfield. The observable quantities such as absorption, distribution and extinction are then calculable from these dipoles. The main advantage of the DDA is that it is completely flexible with respect to the geometry of the target, however, it is necessary to use a low inter-dipole separation (d) compared to structural lengths in the target and the longest wavelength λ such that:

$$|m|k\ d < 1 \tag{1}$$

Where m is the complex refractive index of the target material, $k \equiv 2\pi/\lambda$, λ is the vacuum wavelength and d is the interlayer distance dipolar (distance between np-Ag).

In the DDA simulation, three important properties are considered for our structure. These include the size and density of the nanoparticles and the distance between them. These simulations revealed that there is an optimal design for these parameters where the effect of diffusion, absorption and plasmon causes progress in the overall performance of the solar cell. Figure 2 shows the structure of our device from solar cell based on silicon with silver nanoparticles incorporated in the silicon nitride (SiN).

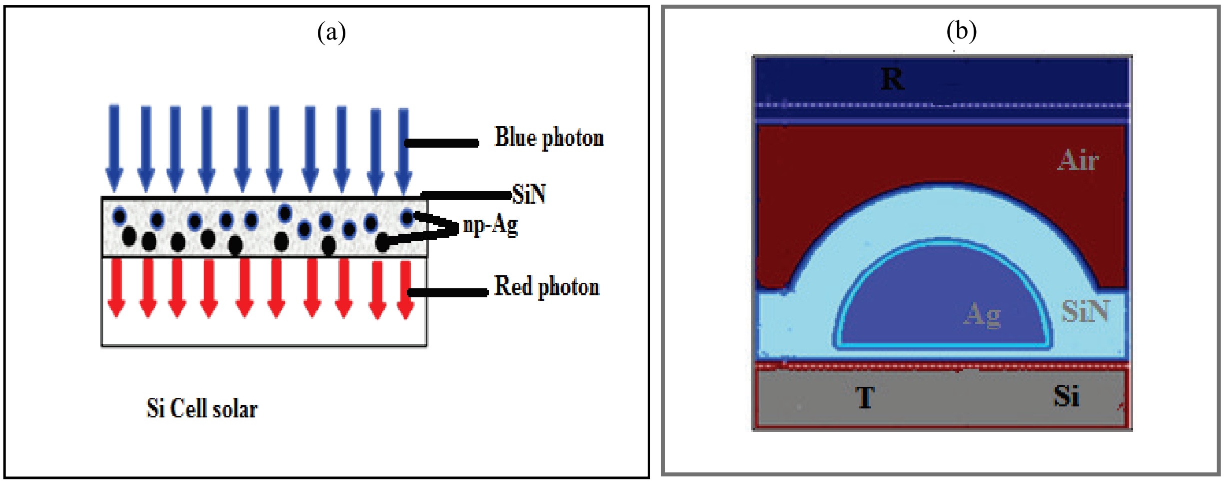

Fig . 2 – a) The geometry of the structure of the solar cell silicon-based simulated with np-Ag incorporated in the SiN matrix. b) Schematic view of the simulated geometry. The regions occupied by the different elements of the system are signaled by Air, Ag, SiN, T and R

Different size of np-Ag (1-150 nm), in DDA simulation, were used with a distance between nanoparticles (nps), which is (1- 15 nm) according to the following relationship:

$$\frac{a_{eff}}{d} = \left(\frac{3N}{4\pi}\right)^{1/3} \tag{2}$$

With a_{eff} is the effective radius of the target (the size of the nps). N is the number of lattice sites occupied in the target.

In scattering theory, there is an important relationship called optical theorem, which states that the effective extinction section is directly proportional to the imaginary part of the amplitude far field scattered in the direction of incidence. For small particles, the extinction is defined as the sum of the scattering and absorption. For these small particles in the quasi-static limit, the cross sections of scattering and absorption are given as follows: [38]

$$C_{abs} = \frac{2\pi}{\lambda}\,\mathrm{Im}(\alpha) \tag{3}$$

$$C_{sca} = \frac{1}{6\pi}\left(\frac{2\pi}{\lambda}\right)^4 |\alpha| \tag{4}$$

$$Q_{sca} = C_{sca} \,/\, \left(C_{sca} + C_{abs}\right) \tag{5}$$

α is the polarizability of the particle for a small spherical particle in vacuum, given by :

$$\alpha = 3V\left[\frac{\varepsilon / \varepsilon_m - 1}{\varepsilon / \varepsilon_m + 2}\right] \tag{6}$$

Q_{sca} is the scattering efficiency factor, Csca is the scattering cross section and Cabs is the absorption cross section.

In our simulation, we conducted several calculations for different sizes of np-Ag deposited in an antireflection layer based on refractive silicon nitride (SiN) having a refractive index n = 2.03 compared to the case of np-Ag deposited directly on the cell surface that considered the air in simulation (DDA). The np-Ag is spherical shape.

Our goal is to assess the role of the dielectric environment by comparing the effect of plasmonic layers either supported on- or embedded within the ARC. To this end, Ag NPs were embedded within the standard 78 nm SiN ARC. Much more work needs to be done to explain the effects of shape, dielectric of the surrounding medium around the particles, optimal spacing and distribution, on the photocurrent of solar cells. Simulation studies in the area of plasmonics particularly in the scattering by particles behaving like dipoles have helped in giving deep insight into the various processes described in this paper and acting as accurate optimisation tools. Cole and Halas [39] in their paper determine a model to suggest the ideal distribution of spherical metal nanoparticles, both nanoshells as well as nanospheres for optimum light harvesting of solar energy. According to their simulation 30% coverage of nanoparticles can scatter almost all the incident radiations and 105 nm silver colloidal particles can scatter 571.5W/m^2 out of the available 578 W/m^2 [35]. Using the SiO_2 matrix Antonio et al [40] showed that the absorption of the ultraviolet rays is quite large. He shows that growing an Ag plasmonic layer on top of c-Si solar cells terminated with the SiO_2 reference ARC leads to decrease the cell efficiency and the photo-generated current by about 30 %, an effect due to a drop in the transmitted light.

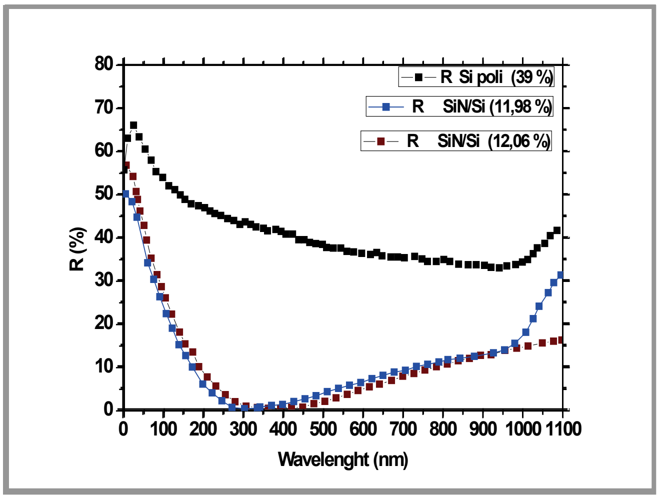

Fig. 3 - Losses reflectivity of a polished silicon surface and covered with a single layer antireflection

The figure 3 shows the losses reflectivity on polished silicon surface and covered with a single layer antireflection. This figure appeared a very good correlation between simulation results and the reflectivity measurements (integrating sphere) that we carried out. The differences recorded between the measured and simulated reflectivity for wavelengths greater than 1000 nm are caused by the reflection of the low energy photons on the back side of the silicon substrate (Si / air interface) which have not been captured [41]. Considered in simulations and for short wavelengths the difference is due to the instabilities of the light source. Nevertheless, the reflectivity simulations are quite satisfactory for conducting this study. It is noted that reflection losses are important for small and long wavelengths (UV and IR), this is caused by the nature of the antireflection layer used, in fact, the SiN (n = 2.03 and d = 78 nm) makes it possible to strongly reduce the reflectivity in the visible range, thus, a large part of the UV is reflected and another part will be absorbed at the surface of the cell. For infrared, a large part goes through the cell to be absorbed.

3 Results and Discussion

Third generation solar cells are often called the "future" of solar cells, as they promise low-cost and high-efficiency solar cells. This requires circumvention of the Shockley-Queisser limit of single band gap devices. In this case, the particle size, shape, dielectric environment and particle material were modified to study the impact of each on the trapping of light in solar cells. It has also been shown that the silver particles give an improvement in the length of the path much higher than the gold particles. For a more realistic simulation, the inter-particle interaction as well as the interaction with the substrate must be taken into account. The calculations were performed by DDSCAT simulator. It is a software package to calculate scattering and absorption of electromagnetic waves by targets with arbitrary geometries using the DDA. In this approximation the target is replaced by an array of point dipoles; the electromagnetic scattering problem for an incident periodic wave interacting with this array of point dipoles is then solved essentially exactly. The main advantage of the DDA is that it is totally flexible in terms of target geometry, being only limited by the need to use a weak interdipole separation in relation to the structural lengths in the target and the wavelength λ. The principle of this technique is to divide the particles to be studied into a number N of polarizable elements. For each element i, we obtain *Pi* and *ri*, which are respectively the polarizability and the position of center of the dipole. This makes it possible to obtain, for each element i, the *Pi* polarization resulting from its interaction with a local electric field E_{loc}. The local field created in each particle will be the sum of the contributions of the incident field and the fields of each of the other particles. From this, it is possible to calculate the total polarization P, and its absorption and diffusion.

In our studies, we considered two cases; the first is the nanoparticles incorporated into the SiN matrix and the second, when the nanoparticles will be deposited on the surface of the antireflection layer. The layer (SiN) considered has a thickness 78 nm and refractive index n = 2.03. The size of the nanoparticles was varied with average sizes of 1 to 10 nm with step 1 nm and 10 nm in sizes 10 to 150 nm to calculate absorption. Using DDSCAT, we have varied the sizes in the ddscat.par file. After simulation, the absorption of np-Ag is obtained as a function of the wavelength (figures 4). These results, presented in figure 4, allowed us to observe the impact of np-Ag in increasing the absorption of solar cells. Note that when the np-Ag varied between 10 and 100 nm, there is a relatively large increase in absorption between 200 to 350 nm compared to the variance from 1 to 10 nm. This absorption becomes greater when the np-Ag is sized from 110 to 150 nm. The figure 4c shows clearly the peaks between 200 and 400 nm. The transmission of np-Ag has appeared in figure 4c when the sizes are greater than 100 nm. The peaks are important when the size of np-Ag is at 110 to 150 nm (with λ = 250, 300 and 400 nm). These are due to the absorption of ultraviolet light by silver nanoparticles and convert into visible by plasmonic effect.

Comparing the effect of absorption of np-Ag embedded in SiN and np-Ag to surface (air) (Figures 5), the absorption of the cell becomes smaller then to np-Ag embedded in SiN. This lead us to conclude that the np-Ag embedded in a matrix SiN absorb more better the dissipated rays that allow increased efficiency of solar cells. This transmission of the rays towards to the visible gives a good absorption, which increases the absorption efficiency as shown in figures 6a, 6b for the sizes (150, 110 nm) respectively. The figure 6b shows us the scattering and absorption of np-Ag embedded in SiN with 110 nm. The silver particules have converter the short wavelength rays to visible rays and increase the absorption of solar cells.

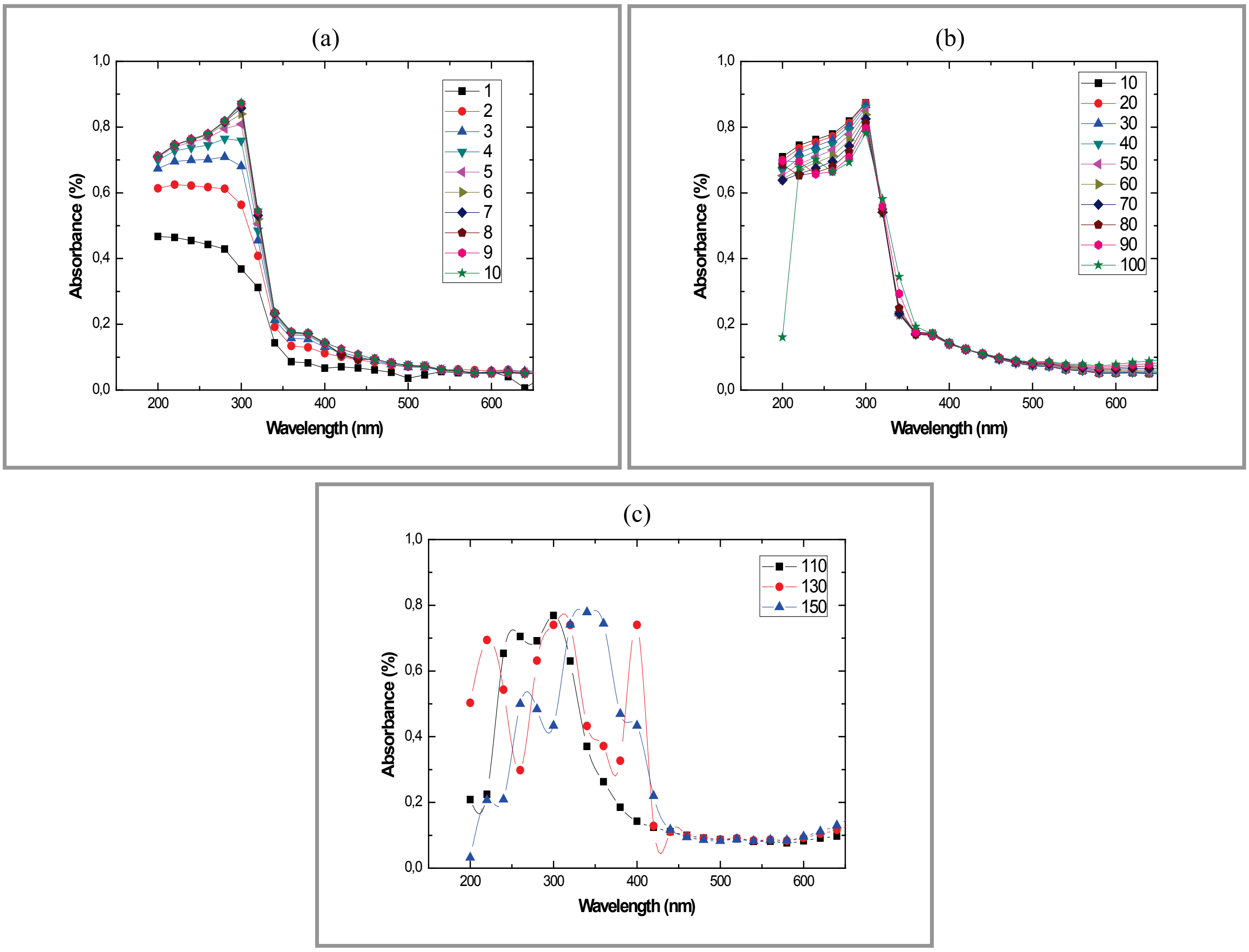

Fig . 4 - Absorbance of np-Ag incorporated into the SiN (np-Ag / SiN ; n=2.03).
a) in size from 1 to 10 nm b) in size from 10-100 nm c) in size from 110; 130; 150 nm.

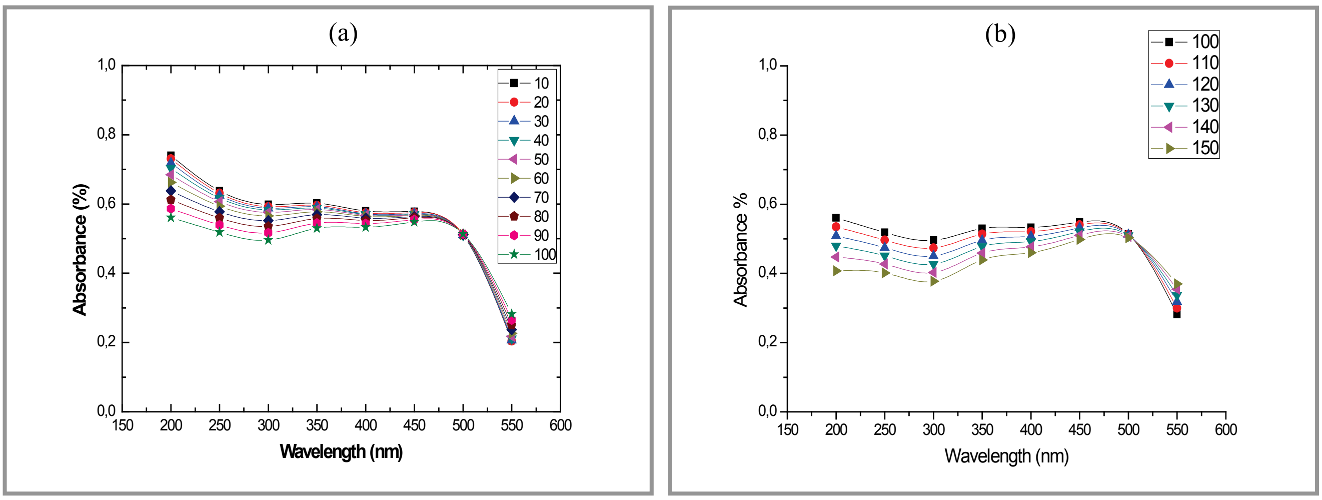

Fig . 5 - Absorption of cells solar based on silicon with np-Ag in surface (Ag / air "1").
Size from 10-100 nm, b) Size from 100-150 nm

In our case, the results clearly show that the size of nps-Ag between 10 and 150 nm increases the current generation of the cell covered by an anti-reflective layer based on SiN. The silver nanoparticules with 110 nm gives a maximum absorption.

The dissipated rays will be absorbed or converted to visible by this np-Ag. If the size of np-Ag is decreased, the absorption rate will be less. Moreover, if the incoming rays are very high (λ > 400 nm), the np-Ag absorption would be negligible.

Finally, the silver nanoparticles incorporated into a SiN matrix can increase the efficiency of our solar cell by absorbing the short wavelengths dissipated.

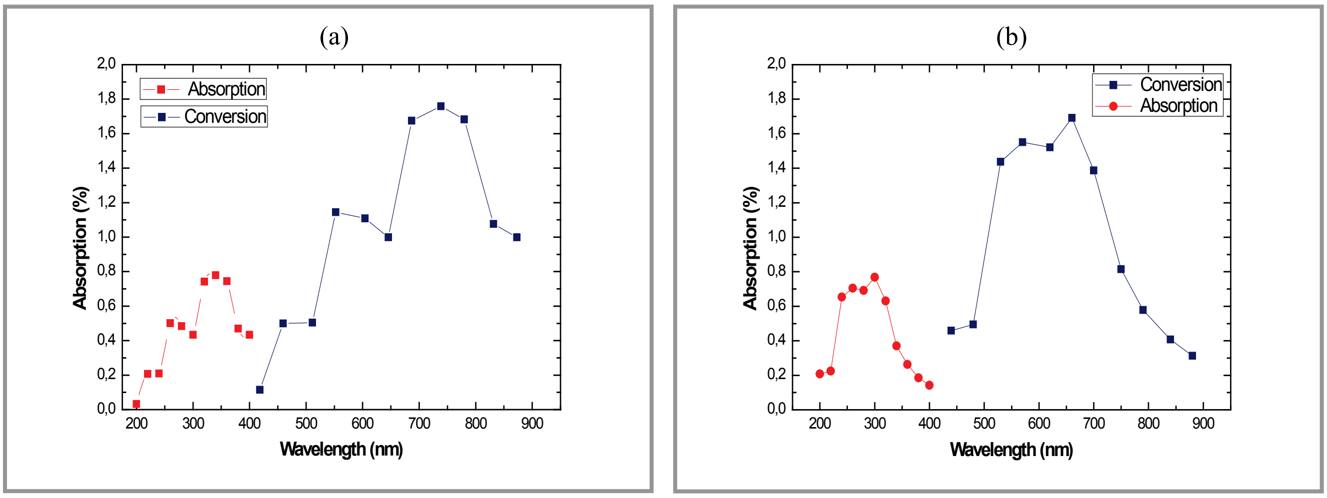

Fig . 6 - a) The conversion of np-Ag for a size 150 nm b) The conversion of np-Ag for a size 110 nm

4 Conclusion

In this paper, the absorption of silver nanoparticles (np-Ag) embeded in silicon nitride anti-reflective layers (SiN) on the performances of standard silicon solar cells are investigated. Using DDSCAT software, the maximum absorption rate of ultraviolet radiation loss in the case of the standard solar cells is determined. The variance of np-Ag size has allowed us to see the best size that can absorb a maximum of UV rays that are loss in a silicon solar cell. The contribution of np-Ag in the SiN anti-reflective layers has been able to improve the absorption of our solar cell.

The results show that the optimal size of np-Ag is 110 nm. The np-Ag can absorb the dispersed ultraviolet wavelengths and convert them into visible by plasmonic effect. It is interesting to note in all cases that in the high wavelength region (λ > 400 nm) the absorption of np-Ag is neglected. So, the elimination of these optical losses, can improve the spectral response of solar cells based on silicon in the ultraviolet region of solar spectrum.

Finally, it would be wise to complete our analysis by exploring the influence of the shape of silver nanoparticles (cylindrical, cubic, etc.), as well as the response of other metals, such as Al or Cu.

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