



Pulsed laser deposition of ZnO and MoO₃ as reflection prohibitors on photovoltaic cell substrate to enhance the efficiency

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ABSTRACT

Purpose: With the ever-growing demand for conventional fuels, the improvement in the efficiency of the photovoltaic system is the need of the hour. Antireflection coatings enhance the availability of solar power by reducing the percentage of light reflected. A new coating has been developed to improve the solar cell's overall efficiency. This study focuses on enhancing the efficiency of the monocrystalline solar cell when a coating of ZnO-MoO₃ is applied at a certain thickness.

Design/methodology/approach: A layer of ZnO followed by MoO₃ is deposited on a Silicon solar cell substrate using a Pulsed Laser Deposition process. Due to the transmissivity d between the two materials, they act as excellent antireflection coating. The layer thickness has been engineered to lie in the maximum absorption spectrum of monocrystalline silicon solar cells, which is between 400 and 800 nanometers.

Findings: Based on the calculation of transmissivities for a given layer thickness of coating material, the coating has been done, and the efficiencies of the coated specimen were compared with the uncoated solar cell. The percentage improvement in the electrical efficiency of a single crystalline silicon solar cell with an anti-reflection coating at 1059 W/m² is about 35.7%.

Research limitations/implications: Among the available antireflection coating materials, the combination that provides better efficiency when coated on top of a solar cell is hard to find.

Practical implications: This anti-reflection coating could be a better solution to enhance the overall efficiency of the single crystalline silicon solar cell.

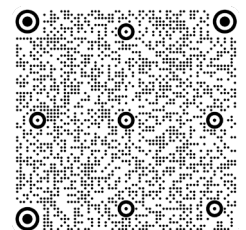
Originality/value: Although ZnO and MoO₃ coatings have been investigated separately for improvement in solar cell efficiency with varying levels of success, the hybrid coating of ZnO/MoO₃ with a performance enhancement of 35.7% is a great leap.

Keywords: Solar cell, Antireflection coating, Refractive index, Pulsed laser deposition

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MANUFACTURING AND PROCESSING

1. Introduction

Increased consumption of fossil fuels in the past few decades led to the depletion of fossil fuels and increased environmental degradation. This led to increased attention toward renewable sources of energy to meet the energy demand. Solar energy is one of the most pollution-free and environmentally friendly renewable energy sources available. Solar Energy can be converted into useful work by two major methods, namely, solar thermal energy conversion and the Photovoltaic module method. Among these two methods, the photovoltaic method is the better alternative, since it converts the light energy from the sun directly into electric power. This is due to the fact that solar photovoltaic cells do not have any moving parts in them. The compactness and modularity of the system are added advantages. These devices are constructed with an array of electrically connected solar cells. These modules have an energy conversion efficiency of around 22% [1]. The conversion efficiency is dependent on the type of PV module and the ambient conditions. This value of conversion efficiency shows that majority of the incident solar energy is dissipated as heat by the solar PV module. The monocrystalline silicon PV module is the widely used module. It consists of silicon wafers doped with impurities to form a p-n junction diode. The silicon substrate surface is prone to slinging bonds and similar crystal defects. Due to the presence of these defects, the electron-hole pairs in the module recombine, leading to the loss of efficiency. This phenomenon of recombination is known as surface recombination. This process can be minimized by surface passivation leading to an enhanced open circuit voltage and fill factor. Field effect passivation [2,3] is the mechanism by which semiconductor surface recombination is reduced. Also, an internal electric field can be provided, that alters the electron hole concentration at the surface. The surface field effect passivation is an efficient method for improving the efficiency of a solar module [4].

Conventionally, ZnO/SiO₂, TiO₂, and ZrO₂ doped with TiO₂ are the most frequently used coating materials to prevent reflection [5]. It has been found that when ZnO ceramic was coated on a monocrystalline photovoltaic module using atomic layer deposition, the conversion efficiency was found to increase by 6% [6]. On coating SiO₂ on the polycrystalline silicon solar cell substrate, the conversion efficiency improved by 0.3% due to surface passivation [2,7]. A combination of two materials as antireflection coatings were also been tried by the researchers to improve the conversion efficiency. Such combinations used by the researchers were Fluorine-doped ZnO, MoO₃, Al-doped ZnO, ITO, and TiO₂-SiO₂ [8]. From the listed materials, ZnO nanofilms are having less opaqueness to the light in the range of 400 to 700 nm. This

could enable a textured pattern on the substrate while coating. Chanta et al. proclaimed that the properties like diameter, shape, surface roughness, and thickness of the seed particles play a vital role in the refractive index of the ZnO nano-coating. It has been observed that ZnO increases the efficiency of the solar cell by 1.88% when it was used as an antireflective coating material [9]. Makableh et al. detailed the light scattering and lower reflection properties of ZnO nanofilm [10]. Suhandi et al. have found that the grown ZnO film displayed an optical band gap of 3.44 eV. With the help of UV-Vis spectrometry, they have demonstrated that the ZnO/TS nano films displayed reduced solar reflectivity [11].

Markov et al. have done a recent analysis on the suitable deposition technique that could be used to coat ZnO on solar cells. They analyzed ZnO films on the solar cells and found that the optimal temperature range during the deposition process would be between 20°C and 600°C [12]. Wang et al. have done pioneering work by identifying the near-infrared light capture property of ZnO nanoarrays. In addition, it also forms a hydrophobic surface which repels the water and provides a clear surface [13]. Other researchers have tried coating ZnO with different techniques like spin coating, sputtering, thermal evaporation, electrochemical deposition, hydrothermal method, chemical vapor deposition, and pulsed laser deposition with varying levels of success.

MoO₃ has also been studied extensively owing to its better refractive index and lower extinction coefficients. Pali et al. explained the above-said behavior of MoO₃ when coated on Au substrate. This enhanced the performance of organic solar cells by 6% [14]. Chen et al. discussed that the coating of MoO₃ nanolayers on the CZTSSe substrate lowered the series resistance and increased the fill factor. They have achieved the efficiency of solar cells by 26% while adding MoO₃ as a single layer on the substrate [15]. Lee et al. proved that the MoO₃ layer absorbs near-infrared and UV while coating on the silicon substrate. They claimed that MoO₃ covers a wide range of spectrums than the other absorbers [16].

It has been individually proved that ZnO and MoO₃ have the capability to enhance solar cell efficiency. In this purview, it can also be expected that if the layers of ZnO and MoO₃ are stacked on top of the substrate, an even more increase in transmissivity and thereby increase in solar cell efficiency can be expected. Thus, in the present work, the combination of ZnO-MoO₃ nanomaterials was selected as an antireflection coating.

2. Materials and methods

Highly transparent and conducting MoO₃ and ZnO pellets are formed from the ACS 99.99% (obtained from

Alfa aesar) purity powder with the help of a manual press. Then the pellets of MoO₃ and ZnO are sintered at the temperature of 530°C for 10 hours and 1300°C for 4 hours respectively in the furnace.

Background process of PLD method for ARC materials

Substrate temperature of PLD

The substrate temperature is controlled by adjusting the current and voltage through a platinum coil which is located behind the substrate. The current-voltage is adjusted by a regulator connected in a feedback loop with a temperature sensor. The temperature sensor is located right by one side of the substrate. The temperature is measured immediately before and after growth, and the average of these two measurements is taken as the deposition temperature [17]. Generally, it is in the range of 100°C to 250°C.

Gas working pressure

The oxygen pressure is the most critical process parameter reported. It is known to have a large impact on sheet resistance. A low oxygen pressure (10⁻⁶ m bar) will introduce oxygen vacancies in the ARC material. A high oxygen pressure (>10⁻² m bar) has been shown to induce island growth instead of the desired layer-by-layer growth mode.

Substrate to target distance

The distance between substrate and target will also affect the kinetic energy of the ablated atoms when they hit the substrate. As the ablated species move from the target to the substrate, they will frequently collide with the oxygen molecules present in the chamber. Each collision reduces the kinetic energy of the interacting atom. A longer distance means more collisions and thus lower kinetic energy of the species when they hit the substrate. The substrate target distance is altered by moving the substrate stage a way and towards the target. Generally, the target to substrate distance was in the range of 4.5 cm to 7 cm.

Laser fluence

Laser fluences are usually between 1 and 2 J/cm². However, lower values have also been reported. There are two ways to adjust the laser fluence. The intensity of the laser is controlled manually by adjusting the input voltage. The other option is to change the mask area. Increasing the mask area lets more of the beam through, thus more energy. However, this also increases spot area, which in turn lowers the laser fluence. The laser fluence is adjusted in both ways. The spot size is found by measuring the visible area left by the laser pulse on the target crystal. The energy of a laser pulse is measured by a detector placed in front of the growth

chamber where the laser pulse enters. A 10% loss of the chamber window is taken into account. The laser fluence has to be in the range of 2-4 J/cm². The optimum laser fluence to ablate the silicon source is to be 2.5 J/cm².

Off-axis angle

The off-axis angle is a measure of the substrate displacement relative to the target (and thus the material flux). Pulsed Laser Deposition (PLD) is generally thought of as a stoichiometric transfer. After growth, the film is assumed to have the same stoichiometry as the target. However, this is not always the case. The off-axis angle will be adjusted in the same plane as the incoming laser beam. A positive θ means away from the direction the laser beam enters. The angle is adjusted by moving the substrate stage. The deposition of the films is carried out by pulsed laser deposition method, inside a vacuum chamber using a Q-switched Nd: YAG laser (Quanta-Ray INDI series, Spectra-Physics) with 532 nm radiation at a 7 ns pulse having a cycle frequency of 10 Hz and maximum output energy of 200 m J.

3. Experimentation

PLD has become popular because of its relatively cheap and simple implementation and its ability to create high-quality oxide interfaces. A substrate and target (the material which is going to be deposited) is placed in a growth chamber facing each other. A pulsed laser is directed onto the target. When a laser pulse is absorbed by the target, species (atoms, ions, molecules, etc.) are ejected from the surface and moved towards the substrate. Right after being ejected, the species absorb some of the laser energy themselves, creating a plasma plume. When growing complex inorganics with PLD, the ejected species need to be mainly atomic, diatomic, or other low-mass particles. To achieve this, the energy of the laser pulse needs to be strongly absorbed in a small volume of the target. As a result, a small wavelength (UV) laser and a short pulse (nanoseconds) are required. In the growth chamber, a background gas is present, which affects the deposition in two ways. The gas often reacts chemically and is used to create the correct stoichiometry of the thin film. For example, an oxide grown under too low oxygen pressure will result in oxygen deficiencies in the film. The other use of background gas is that it reduces the kinetic energy of the ejected species. Since the often-high pulse energy is needed to achieve ablation, the ejected species themselves will have high energy. Atoms and ions with too high energy will be able to penetrate the substrate and cause defects in the material and damage the surface. When a background gas is present the species will collide with gas molecules and lose some of their kinetic

energy before reaching the substrate. When the species reach the substrate, they will be adsorbed. The deposited species will nucleate and grow epitaxially on the substrate. The substrate often has to be heated to a certain temperature for the nucleation to occur. One problem with PLD is that the material flux is both directional and has a small radius. If the substrate is too large, both nonuniform thickness and also variations in the stoichiometry will occur. The deposition chamber consists of three target holders. From the mentioned coating methods PLD technique gives an accurate uniformity of the thin film to be coated. This coating method gives maximum coating efficiency for thin film depositions. PLD is the most promising method for growing oxides such as high-quality ZnO because it has a high-speed deposition process under high partial pressure of oxygen. Figure 1 (a) and (b) show the PLD instrument and the coating operation.

The average size of the particle was found to be 175.5 nm calculated using the Debye Scherrer equation.

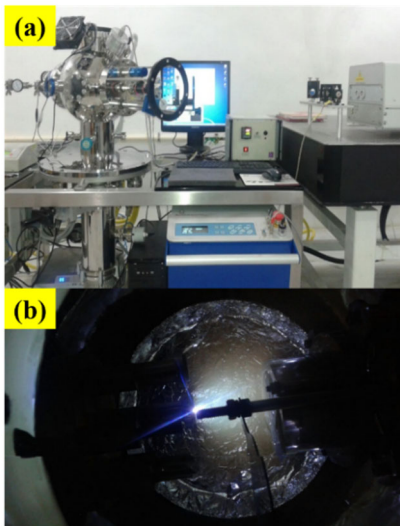


Fig. 1. a) Image of PLD equipment and b) coating operation

4. Theoretical modelling

Sukhatme S P, et al. have provided a relation to find the incident angle and incident total solar radiation falling on the surface. The declination angle (δ) is determined by using the following relation [18],

$$\delta \text{ (in degrees)} = 23.45 \sin \left(\frac{360}{365} (284 + n) \right) \quad (1)$$

where: n – particular day of the investigating month.

For a bare, uncoated solar cell, the maximum efficiency could be calculated based on the short circuit current and open circuit current as follows.

$$\frac{I_{sc}}{I_o} = \exp \left(\frac{eV_{oc}}{kT} \right) - 1 \quad (2)$$

where:

I_{sc} – short circuit current, A;

I_o – dark current, A;

e – charge of an electron, J/V;

V_{oc} – open circuit voltage, V;

k – Boltzmann constant, J/K;

T – absolute temperature, K.

Using the following equation, the maximum voltage that can be produced by the solar cell is obtained.

$$\left(1 + \frac{eV_m}{kT} \right) \exp \left(\frac{eV_m}{kT} \right) = 1 + \frac{I_{sc}}{I_o} \quad (3)$$

where: V_m – voltage at which the power is maximum, V.

The following expression can be used to evaluate the maximum current (I_m).

$$I_m = \frac{\left(\frac{eV_m}{kT} \right)}{\left(1 + \frac{eV_m}{kT} \right)} (I_{sc} + I_o) \quad (4)$$

The maximum power output (P_m) of the solar cell can be evaluated from the following equation.

$$P_m = \frac{\left(\frac{eV_m^2}{kT} \right)}{\left(1 + \frac{eV_m}{kT} \right)} (I_{sc} + I_o) = I_m \times V_m \quad (5)$$

For a coated photovoltaic module, the efficiency can be calculated using the following relation.

$$\eta_{max} = \frac{I_m \times V_m}{I_T \times A_c} \quad (6)$$

where: I_T – incident solar flux, W/m².

Refractive index calculation for ARC

Refractive index of an anti-reflective coating material

$$n_1 = \sqrt{n_0 \times n_2} \quad (7)$$

where:

n_1 – refractive index of ARC material,

n_0 – refractive index of the surrounding material (i.e. air medium),

n_2 – refractive index of semiconductor material (i.e. Si).

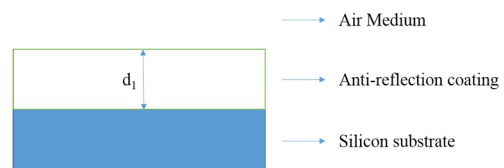


Fig. 2. 2-D view of solar PV panel with a coating

Figure 2 depicts the arrangement of coating on the substrate of the silicon solar cell. The optimum coating thickness can be calculated from the following expression,

$$2d_1 = \frac{\lambda}{4n_1} \quad (8)$$

$$d_1 = \frac{\lambda}{4n_1} \tag{9}$$

The following expression is Snell's law which helps to find the transmissivity of the solar cell without the anti-reflective coating.

$$\frac{\sin \theta_1}{\sin \theta_2} = \frac{n_2}{n_1}$$

$$\theta_2 = \sin^{-1} \left(\frac{\sin \theta_1}{\left(\frac{n_2}{n_1}\right)} \right) \tag{10}$$

Reflectivity when no anti-reflection coating,

$$\rho_i = \frac{\sin^2(\theta_2 - \theta_1)}{\sin^2(\theta_2 + \theta_1)} \tag{11}$$

$$\rho_{ii} = \frac{\tan^2(\theta_2 - \theta_1)}{\tan^2(\theta_2 + \theta_1)} \tag{12}$$

$$\rho = \frac{1}{2}(\rho_i + \rho_{ii}) \tag{13}$$

$$\tau_{r1} = \frac{1 - \rho_i}{1 + (2M - 1)\rho_i} \tag{14}$$

$$\tau_{r2} = \frac{1 - \rho_{ii}}{1 + (2M - 1)\rho_{ii}} \tag{15}$$

$$\tau_r = \frac{1}{2}(\tau_{r1} + \tau_{r2}) \tag{16}$$

Total transmissivity

$$\tau = (\tau_r \times \tau_a) \tag{17}$$

5. Results and discussions

The ARC material should have a refractive index of 1.85 to ensure proper light transmission [19]. The transmissivity and reflectivity of glass to silicon wafer are 0.4506 and 0.1943 respectively. In this case, 82% of the solar irradiance is transmitted across the air/glass medium as per the calculation. Before coating, it has been found that the overall efficiency of the solar panel at 40°C is 11.64%.

5.1. Influence of solar irradiation on power

Figure 3 shows the variation of power with the variation of solar irradiation. Without any antireflection coating, with the increase in intensity, power from the silicon solar cell is

found to increase. The power generated in the solar cell at 1022 W/m² of intensity was recorded as 0.03266 W. Similarly, at 1034, 1042, 1050 W/m² of solar flux, the power is recorded as 0.03340 W, 0.03380 W, and 0.03439 W respectively. At irradiation of 1059 W/m², the solar cell without coating generates the maximum power of 0.0365 W.

In the same way, at 1022 W/m², the solar cell with coating generates the power of 0.05175 W. At irradiation of 1034, 1042, 1050, and 1059 W/m², the power was recorded as 0.05400, 0.05520, 0.05590, and 0.05630 W. By comparing the results obtained, the single crystalline solar cell with antireflection coating gives an improved power in all the cases. The values are given in Table 1.

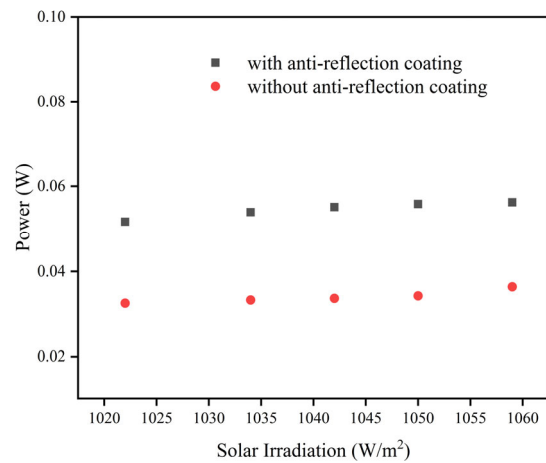


Fig. 3. Influence of coating on the solar cell on the power output at varying solar irradiation

5.2. Effect of solar irradiation on voltage

From Figure 4, it can be noted that the intensity of light radiation that falls on the solar cell influences the output voltage almost linearly. The solar cell without any antireflection coating provided a voltage of 1.15, 1.1, 1.16, 1.17, and 1.21 V at 1022, 1034, 1042, 1050, and 1059 W/m² of solar flux.

Table 1.

Observation of voltage, current, and power with and without antireflection coating at varying solar intensity

Intensity W/m ²	Without antireflection coating			With antireflection coating		
	Voltage V	Current A	Power W	Voltage V	Current A	Power W
1022	1.15	0.0284	0.03266	1.25	0.0414	0.05175
1034	1.16	0.0288	0.03340	1.28	0.0424	0.05400
1042	1.16	0.0292	0.03380	1.29	0.0428	0.05520
1050	1.17	0.0294	0.03439	1.30	0.0430	0.05590
1059	1.21	0.0302	0.03650	1.31	0.0430	0.05630

Similarly, the voltage of the solar cell with antireflection coating was noted at 1022, 1034, 1042, 1050, and 1059 W/m² which displayed the voltages as 1.25, 1.28, 1.29, 1.30, and 1.31 V, respectively. The silicon solar cell having no antireflection coating generated less voltage than the antireflection-coated silicon solar cell. The voltage variation data is shown graphically in Figure 4. From the figure, it is found that the voltage is found to increase while increasing the solar irradiation.

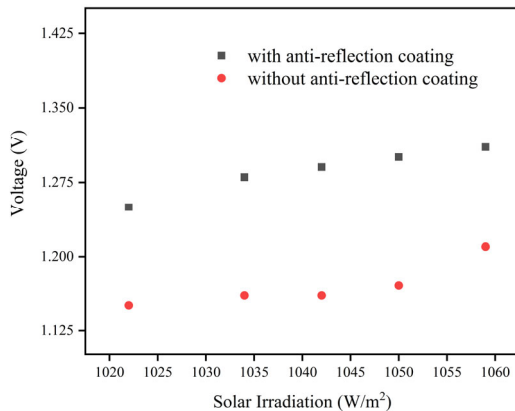


Fig. 4. Influence of coating on the open circuit voltage at varying solar irradiation

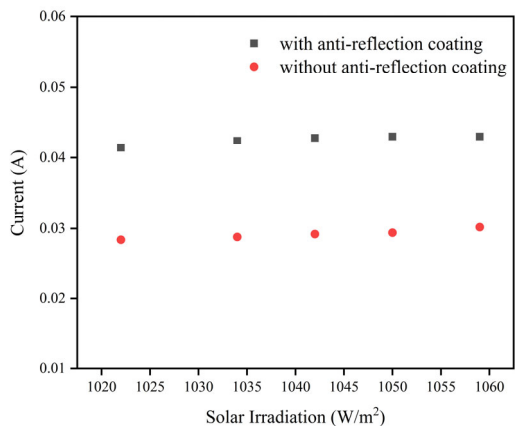


Fig. 5. Variation of current due to the coating at varying solar irradiation

5.3. Effect of solar irradiation on current

The current was recorded at various solar irradiation in the single crystalline solar cell without antireflection coating on the substrate. At 1022, 1034, 1042, 1050 and 1059 W/m² of solar flux, the current value was noticed as 0.0284, 0.0288, 0.0292, 0.0294 and 0.0302 A, respectively. Similarly,

silicon solar cell without antireflection coating was recorded at 1022, 1034, 1042, 1050, and 1059 W/m² of solar flux. At 1059 W/m² of solar flux, the current was noticed as 0.0430 A, which is higher than the current produced by the solar cell without any antireflection coating. From Figure 5, it has been noted that the current is linearly increasing with the increase in solar irradiation. This attributes to the increase in the overall conversion efficiency of the solar cell.

6. Conclusions

Based on theoretical calculations, the refractive index of ZnO-MoO₃ was found to be 1.85. The transmissivity and reflectivity of the solar cell substrate were calculated as 0.4506 and 0.1943, respectively.

- In the present work, ZnO and MoO₃ double layers were coated on the monocrystalline silicon solar cell substrate (2 cm x 2.5 cm) using a pulsed laser deposition technique with 355 nm and 2.5 J/cm² of laser fluence.
- The voltage, current, and power developed on the silicon solar cell substrate without the antireflection coating were found to be 1.21 V, 0.0302 A, and 0.03650 W only at 1059 W/m² of solar flux. Similarly, at 1059 W/m² of solar flux, the maximum current, voltage, and power were observed to be 1.31 V, 0.0430 A, and 0.05630 W.
- By coating the antireflection material on the silicon solar cell, the overall conversion efficiency is improved by 35.16%.

Additional information

The work presented in this paper was presented in “Two Days Virtual National Meet on Nano Interface Science (NIS-2021)”, Chettinad Academy of Research & Education, Chennai, India, 2021.

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