Hydrophilic Antireflection and Antidust Silica Coatings

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ABSTRACT: We report on the optical and morphological properties of silica thin layers deposited by reactive RF magnetron sputtering of a SiO2 target under different oxygen to total flow ratios \([r(O_2) = O_2/Ar, \text{ranging from 0 to 25%}]\). The refractive index \((n)\), extinction coefficient, total transmission, and total reflectance were systematically investigated, while field-emission scanning electron microscopy, atomic force microscopy, and three-dimensional (3D) average roughness data construction measurements were carried out to probe the surface morphology. Contact angle measurements were performed to assess the hydrophilicity of our coatings as a function of the oxygen content. We performed a thorough numerical analysis using 1D-solar cell capacitance simulator (SCAPS-1D) based on the measured experimental optical properties to simulate the photovoltaic (PV) device performance, where a clear improvement in the photovoltage conversion efficiency from 25 to 26.5% was clearly observed with respect to \(r(O_2)\). Finally, a computational analysis using OptiLayer confirmed a minimum total reflectance of less than 0.4% by coupling a silica layer with \(n = 1.415\) with another high-refractive-index (i.e., \(>2\)) oxide layer. These promising results pave the way for optimization of silica thin films as efficient antireflection and self-cleaning coatings to display better PV performance in a variety of locations including a desert environment.

1. INTRODUCTION

The front cover glass required for photovoltaic (PV) module insulation is the first surface to receive irradiation toward the solar cell, and the first surface to limit the photon flux impinging due to optical losses, which can be counteracted by means of antireflective (AR) coatings. The soiling adherence inherently disrupts the intended function of the AR coatings, thus reducing the power output of the PV plants.\(^1\)–\(^4\) This issue is hence motivating the increased research attention toward AR and antisoiling coatings for photovoltaic solar panels, lasers, automobiles, optical instruments, and solar collectors.\(^5\)–\(^9\)

More specifically, the performance of PV modules has been found to be reduced by up to 22% due to dust encapsulation.\(^10\) A real-time measurement tool has recorded a performance drop of about 25%/\(\mu\text{m}\) for natural dust layer-encapsulated c-Si PV modules.\(^11\) Hence, it becomes critical to meet the conjunction requirements of enhanced antisoiling as well as antireflection properties. The improvement in optical and electrical efficiency is noticeable by integrating self-cleaning/antireflection coatings in PV modules.\(^10\)–\(^12\) To develop an optimized AR layer, the film has to satisfy the destructive interference conditions for light waves reflected from the glass/coating and coating/air surfaces.\(^8\)\(^9\) However, control of the coating thickness “\(t\)” plays a central role in developing such films as it has to be one-fourth of the incident spectrum wavelength \((\lambda/4 \times n_{ARC})\) for an optimized AR property. Additionally, the refractive index \(n_{ARC}\) of the AR layers should be \((n_s \times n_A)^{1/2}\), where \(n_s\) is the refractive index of the substrate and \(n_A\) is the refractive index of air.\(^10\) The calculation showed that values around 1.22 and 120 nm of refractive index and the layer thickness, respectively, are required to meet the need for \(n_s\) of around 1.5.

However, no material exists in nature with such a low refractive index value, whereas the lowest reported values are 1.39 for magnesium fluoride and 1.46 for silica.\(^13\) It has been demonstrated that introducing porosity in the coating can lead to the reduction of the refractive index. A three-dimensional (3D) crossed nanoporous layer has been found to be capable of Fresnel light reflection control with improved transmission.\(^14\) The physical vapor deposition process was used to grow such films on a low-alkali borosilicate glass with surface patterning to introduce superhydrophobicity.\(^14\) The contact angle (CA) was measured at 160° with a transmission range of

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95%. Later, a report by Zhang et al. confirmed that transmission can be improved further to achieve ~97% through a composition of silica–silica nanoparticle coating using the sol–gel technique. A remarkable refractive index of 1.21 has been successfully reported through controlled growth. However, the results showed superhydrophobicity with a CA of 5° with antifogging capacity. Such hydrophilic properties have been attributed to the impact of larger surface roughness. Hence, such results show a tradeoff between higher transmissivity and wettability. A lotus-leaf-like hierarchical structure results in measuring transmittance and water contact angle of 95% and 162°, respectively. Söüz et al. demonstrated that a thickness of 125–150 nm is a prerequisite in developing superhydrophobic AR layers through chemical composition modification, resulting in fine-tuning of the topography and mean roughness of the surface. In addition, a report by Kintaka and co-workers studied the usage of methyl tri-ethoxysilane (MTES) and tri-methylethoxysilane (TMES) as precursors to developing hydrophobic AR layers, where the CA increases from 22 to 108°. The stacking layers of SiO₂/TiO₂ have also been investigated and have resulted in a higher refractive index (n = 2.45) beyond the optimum value. However, although such layers demonstrated improved optical properties due to a minimal reflectance value, they were not qualified to be used as self-cleaning assemblies.

A silica thin film has the properties to show simultaneously high optical transparency and low refractive index values, suitable to be used as optical films from the near-ultraviolet to the near-infrared spectral range. This silicon oxide layer suppresses any parasitic light absorption in the visible range due to its suitable optical band gap and transmission range. Also, low refractive index values allow these layers to be coupled with high-refractive-index materials for antireflection coating applications by minimizing the total light reflectance. In addition, due to their high optical transparency, high hydrophobicity property, and the uniformity of their surface, silica layers are extensively used in various industrial and general-purpose applications including flexible displays, optics, bioengineering, ophthalmic, and energy generation.

Many techniques have been already employed to grow silica films, including electron beam evaporation, ion-assisted deposition, ion beam sputtering, magnetron sputtering (MS), sol–gel, and atomic layer deposition. Among all of these techniques, magnetron sputtering (MS) demonstrated the best deposition competency due to the fine control over the growth parameters, film-uniformity, homogeneity, the low density of pinholes to resist defects, and stable optical properties. Hence, nowadays, MS has become a state-of-the-art technique to grow pristine silica films even with large-scale capabilities. Moreover, having the ability to perform the reactive growth of these films under oxygen or any other reactive atmosphere help in controlling the film’s stoichiometry. The stable control over the growth process using the MS technique has eventually enabled many photovoltaic (PV) industries to grow thin layers of silica layers as antireflection coatings for PV modules. In general, growth parameters play a tremendous role in understanding film quality, which becomes critical even with the stoichiometry ratio. Reactive MS deposition adds an advantage to grow oxygen-rich or oxygen-poor silica films with nanoporous morphology, where process gas has a direct effect on altering the layer morphology.

In this work, we have investigated the optical and microstructural properties of 100 nm silica thin films grown on a glass substrate by the radio-frequency (RF) sputtering technique. The main goal was to develop an optimized deposition recipe of silicon oxide thin films (SiOₓ) with a controllable roughness using the sputtering system for antireflection and antistain coating applications. More specifically, we aim at pointing out the multiple correlations existing between the deposition parameters (i.e., mainly oxygen content), the morphological and structural properties of the grown films, their optical characteristics, their surface wettability properties, and the associated output PV performance. In addition, the effect of oxygen plasma treatment of glass is studied and results are compared with deposited silica film properties. Furthermore, we simulated the effect of multistack layers with various refractive indexes against PV performance. The thin films were grown at a constant process temperature of 200° C under different oxygen to total flow ratios [r(O₂) = O₂/Ar, ranging from 0 to 25%]. A maximum oxygen flow of r(O₂) of 25% has been set based on the chamber’s set pressure tolerance level to prevent any arcing issue. The associated oxygen flows bring the deposition pressure to 1.1 × 10⁻³ Torr, considering the sputtering deposition chamber size. Hence, it has been advised to not reach high-pressure (>1 × 10⁻³ Torr) reactive sputtering, which provides a high degree of ionization of the sputtered species to control the arc of the target. Ellipsometry and UV–vis spectroscopy were used to extract the optical properties, while contact angle measurements were performed for hydrophilicity properties. Furthermore, 3D roughness measurements by Dektak Stylus and contact-angle atomic force microscopy (AFM) measurements were applied to probe the surface topology and field-emission scanning electron microscopy (FESEM) was used for determining the film’s microstructure. Finally, numerical simulations were carried out using SCAPS-1D and OptiLayer simulators to implement the experimentally measured optical properties (especially the coupling with various obtained refractive indexes) to determine their effects on the solar cell device performance. Our simulated results demonstrated a clear improvement in the photoconversion efficiency from 25 to 26.5% with respect to r(O₂) in the silica films and enabled us to computationally screen oxide materials and assess their potentials before testing them experimentally with energy conversion devices.

2. RESULTS AND DISCUSSION

2.1. Structural Measurements. Figure 1a shows the grazing-incidence X-ray diffraction (GIXRD) pattern from 10 to 90° for silica thin films. All films show amorphous phases and confirm that a higher annealing temperature is required for the crystalline phase state. Figure 1b shows the X-ray photoelectron spectroscopy (XPS) survey of the samples grown at r(O₂) = 25% and 200 °C process temperature and its silicon (Si 2p) and oxygen (O 1s) spectra, after monatomic etching, along with oxygen profiling from the XPS study of the thin films grown at oxygen concentration r(O₂) = 0–25%. Regarding the oxygen chemical state analysis, carbon spectra fitting has been used to identify the amount of oxygen related to carbon species (i.e., C–O and C=O), which can be deduced from the total oxygen to conclude the oxide-related oxygen (C-metal bond). All of the samples show mainly the Si and O signals with clear SiOₓ chemical states, with Si 2p locating at 103 eV. The stoichiometry of the SiOₓ samples was...
found to be SiO$_{1.85}$, SiO$_{1.85}$, SiO$_{1.91}$, SiO$_{1.86}$, SiO$_{1.89}$, and SnO$_{1.91}$ based on the partial oxygen pressure $r$(O$_2$) = 0–25%.

As studied, silica stoichiometry has been identified for both cluster etching and monatomic etching.

2.2. Optical Measurements. The optical properties of the films were investigated through UV−vis−NIR spectroscopy. Absorptance spectra were determined from

$$A(\%) = 100 - (T + R)$$

where $A$ is the absorptance, $T$ is the transmittance, and $R$ is the reflectance.

Figure 2a displays the optical transmittance and absorptance spectra of the sputtered silica deposited on glass for various $r$(O$_2$) oxygen contents. Figure 2b shows the transmittance and absorptance spectra for the sputtered silica deposited on glass for various $r$(O$_2$) oxygen contents. The optical transmission was measured to be >90%, while reflectance was below <9% in the visible range, confirming thereby that the deposited films are highly transparent within the range of $r$(O$_2$). All films with $r$(O$_2$) > 5% present low absorptance (<2%) in the visible and NIR regions of the spectra. In the UV−Vis range, a Burstein−Moss shift, (i.e., a blue shift of the absorption edge with increasing charge density (Ne) when decreasing $r$(O$_2$)) may also be observed. The optical absorptance of the films in the NIR was found to strongly decrease with increasing $r$(O$_2$), which could be associated with an increase of the carrier concentration (Ne). Note that the silica films associated with $r$(O$_2$) = 0% show higher absorptance in the visible range compared with the rest of silica layers for $r$(O$_2$) = 5–25%. The deduced absorptance demonstrates interesting values that are very critical to develop a high-quality antireflection coating as parasitic absorption reduces the overall performance of a solar cell. Overall, only a minimal difference was noticed in T\%, R\%, and A\% with respect to $r$(O$_2$), which might be associated with the saturated oxidization effect occurring throughout the bulk of the silica films. Hence, as the oxygen content has just a slight effect on the optical properties of the films, other factors, like the porosity of the films and their morphological properties, become critical to our study.

Figure 3 shows the extinction coefficient and the refractive index ($n$) spectra with respect to the oxygen contents in silica films. The measured refractive index ($n$) value taken at 633 nm (deduced from ellipsometry measurements) varies between 1.4 and 1.5, which corroborates well with the data for coatings employed as AR. As expected, the extinction coefficient increases with higher energy for all oxygen contents $r$(O$_2$). Since the extinction coefficient is a direct function of the absorption coefficient, higher photon energy results in increasing interaction of photons and electrons to observe such a trend.

Figure 4a displays the refractive index values taken at 633 nm of the various sputtered silica films on glass for $r$(O$_2$) in the 0–25% range. For the naked uncoated glass substrate, RI was measured at 1.521 ± 0.5%. For the coatings, without any oxygen flow (i.e., $r$(O$_2$) = 0%), the value starts at 1.510 ± 0.5%, then decreases to 1.492 ± 0.5%, and follows decreasing to 1.412 ± 0.5%, for $r$(O$_2$) = 5 and 25%.

Figure 3a shows the measured band gap ($E_g$) of the silica layers deduced from the absorptance data. The absorbance of oxide films for $r$(O$_2$) in the 0–25% range is calculated using the following equation

$$A = 2 - \log_{10} T(\%)$$

Figure 1. (a) GIXRD patterns of the thin films grown at $r$(O$_2$) = 25% and 200 °C process temperature. (b) XPS survey, along with Si 2p and O 1s spectra of films grown at $r$(O$_2$) = 25% shown in the insets. The inset on the left shows oxygen profiling from the XPS study of silica thin films grown at oxygen concentration $r$(O$_2$) = 0–25%.
where \( A \) is the absorbance and \( T \) is the transmission. Tauc’s curve using the optical absorption data gives the band gap data. The equation to calculate the absorption coefficient is \[ \alpha(h\nu) = A(h\nu - E_g)^{0.5} \] (3) where \( \alpha \) is the absorption coefficient, \( h\nu \) is the photon energy (eV), and \( E_g \) is the energy band gap. 

\( E_g \) was found to increase proportionally with the oxygen content to the silica films, which corroborates perfectly the increase of film transparency with respect to oxygen, as shown in Figure 5b. However, since the band gap is assumed to be a bulk property, it is possible that a decrease in the fluctuation of the Si–O–Si dihedral angle and the Si–O interatomic distance with respect to \( r(O_2) \) leads to an increase in the energy gap. However, an accurate measurement of the band gap variation may involve advanced characterization methods, like synchrotron radiation photoelectron spectroscopy of the valence band or reflection electron energy loss spectroscopy, where the band

Figure 3. Ellipsometry measurement of (a) refractive index (RI) and (b) extinction coefficient spectra of the sputtered silica on glass for various \( r(O_2) \) oxygen contents.

Figure 4. Histogram representation of (a) refractive index measured at 633 nm of the sputtered silica on glass for \( r(O_2) \) in the 0−25% range and (b) calculated silica band gaps for different oxygen partial pressures.

Figure 5. Contact angle measurements to assess the hydrophilicity of sputtered silica on glass substrates for various \( r(O_2) \) values: (a) uncoated glass substrate and hydrophilic (CA ~ 50°), (b) 0% and hydrophilic (CA ~ 17.5°), (c) 5% and hydrophilic (CA ~ 6.4°), (d) 10% and hydrophilic (CA ~ 5.8°), (e) 15% and superhydrophobic (CA ~ 4.2°), (f) 20% and superhydrophobic (CA ~ 3.8°), and (g) 25% and superhydrophilic (CA ~ 1.5°).
gap is determined from the differential inverse inelastic mean free path,\textsuperscript{39} which permits to determine at the same time the optical constants and to distinguish the loss features associated with the presence of defects on the silica surface.

2.3. Hydrophilicity Properties of SiO\textsubscript{2} Thin Films. A hydrophobic surface has been previously proposed as an antidust coating.\textsuperscript{48} As shown in the image sequence of Figure 5, compared to the reference naked glass that showed the lowest hydrophilicity with a CA of about 50\(^\circ\), a changing trend is observed with respect to the oxygen content within the silica coating. As studied previously, wettability depends significantly on surface roughness.\textsuperscript{49}

The contact angle of the films decreases as a function of \(r(O_2)\), which also proportionally depends on the surface roughness as demonstrated further. Previous studies have confirmed that lower surface roughness (i.e., smoother) results in superhydrophobicity,\textsuperscript{30,51} thus, one may expect that increasing roughness will increase the film’s hydrophilicity. The highest angle of 17.5\(^\circ\) has been observed for films grown in the absence of oxygen flow during the deposition (i.e., \(r(O_2) = 0\%\)); then, the more the oxygen we incorporated, the more hydrophilic the films became.

Figure 6 shows also a clear correlation between the refractive index and hydrophobicity properties, suggesting that adding oxygen to the film plays a role not only in terms of optical properties (i.e., films become more transparent) but also in their morphological properties. In general, hydrophobicity does not spread any water droplet on the surface to roll off. Hence, a significant gain in terms of optical transmission and spectroscopic properties (i.e., films tend to diffuse the light) can be confirmed using roughness measurements as described further.

![Figure 6. Summary of the measured contact angle measurements of sputtered silica for various \(r(O_2)\) values along with the reference sample, and its relationship to the refractive index. The dashed lines are a guide for the eye.](https://dx.doi.org/10.1021/acsomega.0c05405)

\[ \text{Figure 7. Typical SEM micrographs showing the surface morphology of the extreme oxygen content values: (a) } r(O_2) = 0\% \text{ and (b) } r(O_2) = 25\%. \]

Without the additional oxygen content (i.e., \(r(O_2) = 0\%\)) and silica with the richest oxygen content (i.e., \(r(O_2) = 25\%\)). In both cases, films seem dense, homogenous, and pinholes-free with no voids observed and cover the entire substrate surface uniformly, which is a crucial factor for the optoelectronic properties of the device in which these films are to be implemented and is one of the main characteristics of films deposited by the magnetron sputtering technique. In addition, the surface roughness in Figure 7b seems higher compared to that in Figure 7a due to the smaller grainlike morphology.

Corrugation has been suggested to reduce the hydrophilicity by means of patterning the surface texture.\textsuperscript{55}

As shown below, for a flat surface

\[ \cos \theta = \frac{\gamma_{lv} - \gamma_{sl}}{\gamma_{lv}} \]

\[ \gamma_{lv} \times \cos \theta = \gamma_{sv} \]

with corrugation,

\[ \gamma_{lv} \times \cos \theta = \varphi (\gamma_{lv} - \gamma_{sl}) - (1 - \varphi) \gamma_{lv} \]

where \(\varphi\) is the fractional area covered by the pattering on the surface, \(\theta\) is the angle of the droplet, \(\gamma_{lv}\) is the liquid to vapor tension, \(\gamma_{sv}\) is the tension from the surface to vapor, and \(\gamma_{sl}\) is the surface to liquid tension.

This mechanism is well established in developing potentiometric nanobiosensors to improve the sensitivity by capturing the essential volume of molecules.\textsuperscript{55} In our case, this technique could be adapted through surface engineering, which will eventually improve light management within the AR layer by corrugation.

As a matter of fact, interestingly, glass samples treated with oxygen plasma for just 5 min have demonstrated superhydrophilicity similar to silica deposited at \(r(O_2)\) of 20\% (CA \(\sim 4^\circ\), see Figure 8). The bombardment of the glass surface by oxygen atoms during plasma treatment may increase the surface roughness and surface tension accordingly. Moreover, AFM analysis results (Figure 9) corroborate well with this assumption and confirm that the surface roughness increases by almost 300\%, going from 1 nm for untreated glass to about 3 nm after 5 min of plasma treatment. Additionally, this is also corroborated by SEM images shown in Figure 10.
Indeed, to increase light management, silica films need to be free from voids and discontinuities and have large particles/ grains. Figure 10 shows the FESEM micrographs of the thin films of typically 100 nm thickness, grown under $r(O_2)$ of 0 and 25%, respectively. Films grown at 25% $r(O_2)$ show smaller grains and can be correlated with the higher surface roughness value (as measured in Figure 8).

2.4. Surface Roughness Properties of Silica Thin Films. Figure 11 shows the 3D mode average surface roughness ($R_a$) of the silica films as a function of $r(O_2)$. A changing morphology is clearly observed as soon as silica is deposited, and the $R_a$ increases already from 6.35 nm for the uncoated reference glass substrate (Figure 11a) to 14 nm for silica grown at $r(O_2) = 0$% (Figure 11b). Following this trend, the surface roughness keeps increasing with respect to the oxygen content and reaches its highest measured value of 43.61 nm for the films grown at $r(O_2) = 25$%. As studied previously, the same trend has been reported for metal-oxide thin films, where increasing partial pressure of oxygen results in decreasing refractive index, while the roughness increases.56 Especially, looking at Figures 4a and 11, we can also notice that the silica layer has a trend of lowering the refractive index with respect to $r(O_2)$, which suggests that incorporating oxygen into the film during their growth yielded an increasing rough surface. Typically, the self-cleaning surface requires to be modified through varying deposition parameters such as background oxygen pressure, deposition rate, deposition power, etc.57

In addition, self-assembling organosilane monolayers have been suggested as a competitive alternative to make a surface hydrophobic by reducing the surface energy.44 In sum, the film roughness increases with increasing oxygen content and the mean free path of the vapor particles decreases when the oxygen partial pressure increases. Such a phenomenon results in reducing the kinetic energy with a decrease of surface mobility,59 which eventually leads to a porous microstructure.

Indeed, it has been demonstrated that the packing density decreases for the films grown at higher oxygen contents.59 One can note that the surface roughness values given by the two techniques (AFM and 3D roughness measurements by the Dektak Stylus profilometer) are different, which is due to the well-known difference in these techniques in terms of resolution and tip-surface convolution.

2.5. Numerical Analysis. During the calculation using SCAPS and OptiLayer software, both the absorption coefficient and band gap of the silica layers were deduced from the experimental data. Results are displayed in Figure 12. The initial value of the silica layer was set at 20 nm. This value was reported as an initial starting value for antireflection coating.44 Furthermore, a set of thicknesses between 20 and 200 nm was launched to optimize the ARC layer thickness. It is worth pointing out that an optimum thickness of the ARC layer is essential to maximize the absorbed light. Hashmi et al.44 has reported 100 nm thick silica as the optimum ARC layer, which is in close agreement with our reported value.

As studied previously, the optimum thicknesses and efficiencies with TiO$_2$, ZnO, ZnS, SiO$_2$, and Si$_3$N$_4$ ARC for the solar cell are 62.4, 78.4, 63.5, 101.3, and 74.3 nm and 19.73, 20.34, 19.83, 18.99, and 20.35%, respectively. The reason for such an efficiency increase is the reduction in light reflection.44 Such results keep the option of further performance improvement through fine-tuning of the ARC layer. As calculated, adding an oxygen-rich $r(O_2) = 25$% AR layer shows an improved photoconversion efficiency (PCE) of up to 27%, as shown in Figure 12d. This increase is due to the reduction in light reflection (i.e., increase of photon absorption).44 In general, the front ARC layer in solar cells plays simultaneously the role of an optical window layer and an antireflection coating. Hence, it is highly required to develop such an oxide layer with low (ideally zero) light absorption and displaying a

Figure 8. Oxygen plasma treatment of the reference glass substrate. Contact angle measurement of the substrate: (a) preplasma treatment and (b) postplasma treatment.

Figure 10. SEM analysis of the (a) untreated glass substrate and (b) 5 min plasma-treated glass substrate.

Figure 9. AFM analysis of the (a) untreated glass substrate and (b) 5 min plasma-treated glass substrate.
refractive index close to the symmetrical mean of those of silicon and air. The calculated values confirm that silica layers developed with a high oxygen concentration \( r(O_2) = 25\% \) results in a higher \( J_{sc} \) (47 ma/cm\(^2\)) and \( V_{oc} \) (0.750 V). Such performance is mainly due to the improvement of optical properties and related to improving \( J_{sc} \) as expected, due to less reflection, resulting in higher solar spectrum absorption in the Si absorber layer. Again, \( V_{oc} \) is a function \( J_{sc} \); hence, a gain is expected. However, a clear decrease in both \( V_{oc} \) and \( J_{sc} \) has been calculated for silica layer thickness above 120 nm for oxygen concentration \( r(O_2) = 25\% \), resulting, in turn, in a clear decrease in photoconversion efficiency, from 27.1 to 24.0\% when the thickness increases from 120 to 200 nm. Lower \( r(O_2) \) was found to yield a lower PV performance and could be attributed to a parasitic absorption as evidenced by the lower band gap energy calculation.

In addition, OptiLayer software has been used to find out the minimum achievable reflectance for stacking of oxide layers. A two-layer antireflection design is simulated using this software. The desired wavelength range has been set from 420 to 1200 nm during this analysis. Our simulations pointed out that silica grown at \( r(O_2) = 10\% \) with a refractive index of 1.415 can be used with another optimized oxide layer of 2.35 refractive index to have a minimum reflectance of 0.4\%. Such high-low refractive index stacking layers interfere with the incident light destructively since they are exactly out of phase. Therefore, a minimal reflectance is achievable and opens the way to develop an optimum ARC layer while maximizing its photon absorbance. However, development of such stacked ARC layers may add-up extra cost for large-scale PV modules.

### 3. SUMMARY

In solar cells, it is vital to develop transparent oxide layers to serve as antireflection as well as antidust coatings specifically under a desert environment to boost the performance. In this work, a detailed study has been conducted on the optical and morphological properties of silica thin layers deposited by reactive RF magnetron sputtering of a SiO\(_2\) target under different oxygen to total flow ratios \([r(O_2) = O_2/Ar, \text{ranging from } 0 \text{ to } 25\%]\). The optical transmission was measured to be > 90\%, while reflectance was below < 9\% in the visible range, confirming that the deposited silica films were highly transparent. In addition, all films with \( r(O_2) > 5\% \) presented a very low absorptance below 2\% in the visible and NIR regions of the spectra. The optical absorptance of the films in NIR was found to decrease with respect to the oxygen content, which could be associated with an increase in carrier concentration. The refractive index values were deduced from ellipsometry measurements and varied between 1.4 and 1.5, which corroborates well with the data for coatings employed as AR from the relevant literature. The associated band gaps were deduced from the absorptance data and found to increase proportionally with respect to oxygen, which translated into more transparent coatings. The wettability of these silica layers was assessed by contact angle measurements, which showed that hydrophilicity increased with respect to \( r(O_2) \), corroborating our assumption about the surface roughness analysis. Indeed, this finding was also demonstrated by FESEM, AFM, and 3D mode average surface roughness analyses. On the other hand, reference glass samples treated with oxygen plasma for just 5 min have demonstrated superhydrophilicity similar to silica deposited at high \( r(O_2) \) doses. Moreover, our numerical analysis using SCAPS-1D showed an improvement of the PV performance globally, where a PCE of up to 27\% was calculated for an optimized silica thickness of 100 nm (and a \( r(O_2) \) of 25\%), whereas without any reactive oxygen, the PCE was about 25\%, which is attributed to the reduction of light reflection and thereby an increase of photon absorption. Finally, using OptiLayer, we obtained a calculated total reflectance of less than 0.4\% by...
coupling a SiO₂ layer of \( n = 1.415 \) with another high-refractive-index (>2) oxide layer. This latter result supports strongly the ongoing quest for efficient antidust and antireflection coatings.

4. EXPERIMENTAL SECTION

Soda lime glass (SLG) with a dimension of 1” (width) \( \times \) 3” (length) was used as substrates for the growth of silica thin films. SLG substrates were first cleaned in an ultrasonic bath with acetone, isopropanol, and deionized water for 10 min each. Later, the substrates were blow-dried with nitrogen. MS deposition was performed using a Torr magnetron sputtering tool with a base pressure of \( 5 \times 10^{-5} \) Torr, respectively, under oxygen and argon flow as process gases, with a rotating substrate mode. The purity of oxygen and argon gases was 99.995%. The SiO₂ sputtering target (Kurt J Lesker) with a purity of 99.995% was used. All of the films were grown at a stable process temperature of 200°C with the same deposition rate of 0.5 Å/s.

The argon flow rate was kept constant at 200 sccm during the deposition process to ignite the RF plasma, whereas the oxygen flow rate was varied between 0 and 50 sccm. This leads to oxygen to total flow ratios \( r(O_2) = O_2/Ar \) ranging from 0 to 25%. Structural properties were investigated by an X-ray diffractometer (Rigaku) at diffraction angles 2θ and an XPS Escalab 250Xi (Thermo Fisher Scientific). XPS spectra analysis and fitting were conducted using Avantage software. The source is monochromatic Al Kα, and its energy is 1486.68 eV. The pass energy was 20 eV for all narrow scans and 100 eV for survey scans. The number of scans for each spectrum was 10 for high-resolution spectra and 1 for survey spectra.

The optical properties of the films were investigated by UV–vis spectroscopy (PerkinElmer Lambda 1050 UV/VIS/NIR) as well as ellipsometry (Horiba UVISEL 2). Field-emission scanning electron microscopy (FESEM) was employed to investigate the morphology of the oxides using a JEOL 7610. Film thickness and 3D average roughness measurements were performed with a Dektak stylus profilometer that uses a probe in a contact mode with the surface. This measurement is performed mechanically with a feedback loop that monitors the force from the surface with a map resolution of 0.833 μm/point. Because the stylus profilometer involves physical movements in X, Y, and Z directions while maintaining contact with the surface, it is slower than the noncontact mode technique. The surface topology was characterized using a Bruker atomic force microscope operating in contact mode.

The contact angle measurements of the films were performed by a Kruss tool to assess the wettability (hydrophilicity/hydrophobicity) of the silica films. For the optimized silica layer, CA was performed 10 times for each sample, and the droplet volume was 2 μL.

Plasma treatment of the glass samples was performed under ambient oxygen using an MTI compact plasma reactor at 11 W for 5 min. This study enables us to understand the changes in surface morphology through plasma treatment. In general, an energetic plasma is created through high-frequency voltages (typically kHz to MHz) to ionize the oxygen gas. To test the reproducibility of the results, 25 samples were deposited during each run for a fixed oxygen pressure starting from \( r(O_2) = 0\% \). This process has been followed three times for each oxygen pressure to point out the optimized growth.
The fluctuation was within 5% for the total flow ratios \( [r(O_2) = O_2/Ar] \) ranging from 0 to 25%. Our deposition technique has adapted the pristine growth of silica layers with scalability through varying the background oxygen pressure. In addition, from the characterization results, it has been identified that stoichiometric growth of silica layers has been achieved for all of the samples, which is very significant for improved device performance.

The numerical analyses have been performed using 1D-solar cell capacitance simulator (SCAPS-1D) software. The analyses were performed using the experimentally measured optical properties (band gap, absorption coefficient) of the samples prepared in this work, while other parameters including electron affinity, electron mobility, hole mobility, density of states, dielectric permittivity, carrier density, and thermal velocity were extracted from the relevant literature.44 The second set of simulations was carried out using OptiLayer software to point out the minimum achievable residual reflectance.45 To calculate the optical reflectance, the software follows the theory of normal angle of incidence and oblique incidence case (s-polarization) and two-component AR designs with the highest and lowest available refractive indices to form optimal AR designs.

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**Notes**

The authors declare no competing financial interest.
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