

# Sputtered WOx thin film as the electron transport layer for efficient perovskite solar cells

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Received: 27 December 2021 / Accepted: 17 March 2022 / Published online: 31 March 2022 © The Author(s), under exclusive licence to Springer-Verlag GmbH, DE part of Springer Nature 2022

#### Abstract

The electron transport layer (ETL) is critical in perovskite solar cells (PSCs) as it controls the optics of the complete solar cell. This study uses an industrially viable RF magnetron sputtering technique to prepare the tungsten oxide (WO<sub>x</sub>) ETL for PSCs. Necessary morphological and optoelectronic investigations were carried out to ensure the high-quality WO<sub>x</sub> thin-film. The influence of the deposition power on the ETL thickness and PSC optics were systematically investigated. A three-dimensional (3D) finite-difference time-domain (FDTD) approach analyses the optics and optimization of the complete solar cell. The investigations allow the optimized planar PSC to determine the  $J_{SC}$  of > 21 mA/cm<sup>2</sup>. The optical performance of the planar device is limited due to higher optical losses; hence, the current study proposes a PSC design embedded with Ag nanoparticles. The proposed PSC can improve the  $J_{SC}$  by ~ 17% (up to 24.5 mA/cm<sup>2</sup>) than the planar device owing to improved light trapping, further boosting the PSC's energy conversion efficiency (ECE). A detailed discussion on film realization and solar cell optics is provided.

Keywords  $WO_x$  thin film  $\cdot$  Electron transport layer  $\cdot$  Magnetron sputtering  $\cdot$  Perovskite solar cell optics  $\cdot$  FDTD

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#### 1 Introduction

Recently, organic-inorganic hybrid perovskite solar cells (PSCs) have become a game-changer by showing their radical rising in energy conversion efficiency (ECE) over 25% from 3.8% within a short time [1, 2]. The ECE of the record PSC is comparable to the market-leading silicon (Si) solar cell's efficiency [3-5]. Importantly, the production cost of PSCs is much lower than the traditional Si solar cells. Perovskites are considered to be the most exciting material system due to their excellent optoelectronic properties, such as tunable bandgap (1.2-3.17 eV), low exciton binding energy (~20 meV), high absorption coefficient (~ $10^5$  cm<sup>-1</sup>), high carrier mobility ( $\sim 20 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ ), and simple deposition technique, allowing for various optoelectronic device applications [2, 6-12]. The bandgap tunability of the perovskite material lets achieving high-efficiency tandem solar cells (TSCs) and vertically stacked image sensors [13-17]. According to the detailed balance theorem, the ECE of the perovskite-based TSCs can go beyond 45% if the accurate material bandgaps are selected [18–20]. The perovskite absorber is placed between two charge transport materials in a simple PSC structure. The electron transport layer (ETL) plays a vital role in extracting the photogenerated electrons and transporting them to the external circuit. Besides, ETL blocks hole not to be transferred through it and suppress charge recombination [19]. Hence, ETL should have physical properties, such as high charge mobility, energy level alignment with perovskite, high carrier concentration, etc.

So far, titanium oxide  $(TiO_2)$  is the most widely used ETL for PSCs owing to its superior qualities in terms of transparency and suitable energy levels [2, 19]. However, the TiO<sub>2</sub> ETL shows high charge accumulation within the layer junction, impeding the electron extraction as the conduction band minimum (CBM) is slightly higher than the CBM of the perovskite absorber [21]. Decomposition of TiO<sub>2</sub> under ultraviolet (UV) exposure, poor carrier mobility  $(\sim 1 \text{ cm}^2/\text{Vs})$ , high-temperature annealing, and non-radiative loss due to oxygen vacancy formation are the most common challenges on the way of successful use of TiO<sub>2</sub> as ETL in PSCs [15, 22, 23]. In search of alternative electron transport materials (ETMs), recently, a few metal oxides, such as SnO<sub>2</sub>, ZnO, and WO<sub>x</sub>, have been studied; among them, tungsten oxide  $(WO_x)$ , a comparatively novel material, can be well-thought-out of TiO<sub>2</sub> due to its higher carrier mobility (10–20 cm<sup>2</sup>/Vs), higher chemical stability, low material consumption, low-temperature processing cost [15, 24–26]. In addition to that, a comparatively higher energy bandgap of WO<sub>x</sub> than TiO<sub>2</sub> confirms the passing of most visible lights through it and less absorption to the UV exposure [26]. The complete solar cell's optics greatly depend on the ETL or the front contact; thus, a rigorous optimization is required to use WO<sub>x</sub> as an ETL for PSC fabrication. Until now, a few works have been demonstrated on WO<sub>x</sub> preparation using several synthesizing techniques, including sol-gel, chemical vapor deposition, sputtering, and electron-beam evaporation [22, 23]. However, up to our knowledge, no work has yet studied the preparation of such ETLs along with the underlying device optics while using WO<sub>x</sub> in PSCs. Hence, most literature focused on the electronic properties (e.g., electron mobility, carrier lifetime, and resistance) of WO<sub>x</sub>, which impacts the photovoltaic parameters of PSCs [15, 22, 23]. However, the current study aims only at the optics of PSCs, which do not require detailed electrical parameters of the deposited films for the solar cell characterization [15, 19, 26]. In this work, we considered the RF magnetron sputtering method to realize the WO<sub>x</sub> thin film, which can allow controlling precise film thickness, film uniformity, and elemental composition. Necessary material characterizations confirmed the high quality of the deposited WO<sub>x</sub> film. Then, extracted optical properties of WO<sub>x</sub> film are used to investigate the optics of the WO<sub>x</sub>-based PSCs. Moreover, we proposed a PSC structure integrated with a silver nanoparticle to achieve improved light trapping, increasing the quantum efficiency (QE) and short-circuit current density  $(J_{SC})$ . A three-dimensional (3D) finite-difference time-domain (FDTD) integrated with the particle swarm optimization (PSO) algorithm is used to study optics and optimize the solar cell. Findings from such a multiphysics approach can provide a great agreement with the experiment, which is confirmed by our previously published works [13, 26]. A detailed investigation of the film preparation, characterization, optimization, and device modeling and simulations is provided in the following discussions.

#### 2 Methods

#### 2.1 Experimental details

WO<sub>x</sub> thin films were deposited by a magnetron sputtering machine (PRO LINE PVD75, Kurt J. Lesker Company). A sputter target of WO<sub>x</sub> (Kurt J. Lesker company) with purity 99.99% was used. The target was connected to an RF power source. A cleaned glass substrate was placed above the target. The substrate holder was rotated at a speed of 20 rpm. The working pressure of the deposition chamber was maintained at 15 mTorr during the deposition. The RF power was varied from 50 to 90 W to develop different thin films at room temperature while keeping the deposition time for 30 min. A surface profilometer named DektakXT-A from Bruker Corporation was used to measure the thickness of the films. Elemental compositions of deposited films were revealed using EDX spectroscopy (Zeiss EVO 18 Research). NANOSURF C3000 was used to measure the surface roughness of WO<sub>x</sub> films. The UV-Vis-NIR spectrometer (UH-4150) was used to determine the deposited thin film's optical properties (transmittance, reflectance, and absorbance).

#### 2.2 Device modeling and optical simulation method

Finite-difference time-domain (FDTD) optical simulations embedded with the particle swarm optimization (PSO) algorithm were carried out to investigate the optics and optimization of the solar cells in three dimensions [27-30]. Complex refractive indices of materials were used as input parameters for the FDTD environment. The optical constants of the used materials were determined from the optical investigation of this study and previously published works [2, 19, 31]. The optical wave propagation for the device was modeled from 300 to 850 nm wavelength that is close to the perovskite absorber bandgap. The incident wavelength is circularly polarized, with an amplitude of 1 V/m. Hence, electric field and field intensity can be measured as V/m and  $(V/m)^2$ , respectively, leading further to calculate the absorbed timeaverage power density within a unit cell. The Supporting Information (Section S1) provides a more detailed description of the optical simulation method.

#### 3 Results and discussion

#### 3.1 Realization of high-quality WO<sub>x</sub> thin film

The film quality primarily depends on its physical, optical, and electrical properties. In the case of the RF sputtering technique, such material properties greatly depend on the sputtering power, which influences the film thickness and atomic percentage of W and impacts the film's optoelectronic properties.

In this work, the deposited WOx thin film exhibited a thickness range between 75 and 118 nm while varying the sputtering power from 50 to 90 W. At the same time, the atomic percentage of W is changed from 21.37 to 31.03. The increment of sputtering power ensures the flow of higher energy to the target, resulting in a higher number of ions from the target are sputtered that confirms a higher rate of deposition [32]. Table 1 illustrates the effect of sputtering power on the film thickness and atomic percentage of W. The elemental composition of WO<sub>x</sub> thin films developed with different sputtering powers can be understood by the EDX results presented in Fig. 1.

AFM analysis was carried out to determine the surface roughness of the developed  $WO_x$  films. Figure 2 illustrates the 2D and 3D AFM images of developed films, showing that the average and the root mean square (RMS) surface roughness vary from 1.875 to 3.86 nm and 2.314 nm to 4.717 nm, respectively, with the increment of sputtering power from 50 to 90 W. Resulting an enhanced film roughness with an increase of the sputter power. The roughness data are presented in Table 2.

The change of optical properties against the WO<sub>x</sub> film thickness can be obtained from the wavelength-dependent transmittance (*T*), reflectance (*R*), and absorbance (*A*) spectra presented in Fig. 3a–c. Overall, an increase in the film thickness reduces the transmittance and improves the absorbance, which clearly supports the rising behavior of the film absorption. In addition to that, the absorption towards the higher wavelengths while increasing the film thickness.

The deposited film exhibits almost 80% transmittance and substantial absorption in the UV region, ensuring the development of a high-quality ETL in PSCs. However, the

 
 Table 1
 Thickness and atomic % of W of the films developed at different sputtering powers

| RF sputtering power<br>(W) | Thickness (nm) | Atomic % of W |
|----------------------------|----------------|---------------|
| 50                         | 75             | 21.37         |
| 70                         | 93             | 26.45         |
| 90                         | 118            | 31.03         |

reflection does not follow a common trend, which substantially depends on the film-air and film-substrate interfaces. As a result, the WO<sub>x</sub> can control the optics of the solar cells once it is used as a front contact or potential ETL. The absorption coefficients ( $\alpha = 2.303 \text{ A/d}$ ; A = absorbance and d = film thickness) are also calculated for the films with different thicknesses shown in Fig. 3d, which are used to determine the values of important parameters such as refractive index, extinction coefficient, and optical conductivity.

Developed films exhibit high values of  $\alpha$  in the range of  $10^4 \,\mathrm{cm}^{-1}$ , comparable to reported values of WO<sub>x</sub> developed by reactive sputtering [15]. The energy bandgap  $(E_g)$  of the film was determined by Tauc's relation  $[(\alpha hv)^2 = (hv - E_g)]$ [33], plotting  $(\alpha hv)^2$  versus  $E_g$  as illustrated in Fig. 4a. As the WO<sub>x</sub> shows a direct energy bandgap, the  $E_{g}$  is derived by extrapolating the linear part of the curve to the horizontal axes where  $(\alpha hv)^2 = 0$  for each film. The estimation provides a value of 3.6, 3.4, and 3.19 eV for 75, 93, and 118 nm film thickness. The increase in structural defects (formation of the localized states in the gap) can influence the incremental behavior of the  $E_{g}$  concerning the film thickness. Subsequent, optical constants of the deposited WO<sub>x</sub> films were obtained by calculating refractive index (n) and extinction coefficient (k), which are the main parameters used in the numerical simulation while investigating the optics of the solar cells.

The following expressions can define the refractive index and extinction coefficient of a material, which directly relates to the reflectance and absorbance of the film [15, 34]:

$$n = \frac{1+R}{1-R} + \sqrt{\frac{4R}{(1-R)^2} - k^2},$$
 (1a)

$$k = \frac{\lambda \alpha}{4\pi}.$$
 (1b)

It is noted that the absorption index is closely related to the refraction of the film. The spectral variations of n and k against incident wavelengths for different WOx films thicknesses are presented in Fig. 4b, c, respectively. The values of optical constants of WO<sub>x</sub> films are analogous to the previously published results in the literature. Moreover, the film's optical conductivity ( $\sigma_{opt} = \sigma nc/4\pi$ ) was determined to observe the optical response in the deposited thin film. Figure 4d shows the dependency of  $\sigma_{\rm opt}$  for different WO<sub>x</sub> films as a function of light wavelength. It increases with the film thickness, which may also be related to the increment of charge carrier as thicker films contain more sputtered atoms confirming the higher number of charge carriers [35]. In the following, we use the derived optical constants of the deposited WO<sub>x</sub> thin films as a potential ETL in simulating the WO<sub>x</sub>-based PSC devices.

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**Fig. 1** EDX analysis of the films developed at: **a** 50 W, **b** 70 W, and **c** 90 W sputtering power



## 3.2 Optics in WO<sub>x</sub> thin film-based perovskite solar cells

The investigation started with the utilization of  $WO_x$  as an ETL to develop a planar PSC, where  $WO_x$  was considered to be deposited on a ~ 80-nm-thick ITO substrate, followed by a 400 nm CsFAMA triple cation perovskite absorber and 30 nm NiO HTL. The thickness of the  $WO_x$  ETL was varied from 30 to 400 nm to optimize the device performance. A 100 nm thick Ag was finally considered as a back reflector to complete the device. Our investigation shows that the optical performance of planar PSCs is only maximized while the ETL is 75 to 118 nm, which is also consistent with the thicknesses of deposited films. A schematic of the planar devices (Flat-A, Flat-B, and Flat-C) is depicted in Fig. 5a, e, i for the  $WO_x$  thickness of 75 nm, 90 nm, and 118 nm,

istics of the solar cells were studied by 3D FDTD optical simulations, which gives an excellent agreement between simulation outcome and experimental results. To find the effect of deposition power (film thickness) on the optics of PSCs, time-averaged power densities of planar devices from different ETL thicknesses were calculated, allowing us to understand the optics within a device. Figure 5b–d illustrates the investigated PSC's cross-sectional time-averaged power density plots for an incident wavelength of 450 nm, 600 nm, and 760 nm with the deposition power of 50 W, 70 W, and 90 W, respectively, along with the device schematic (Flat-A) shown in Fig. 5a. Similarly, simulated power densities of the other planar devices (Flat-B and Flat-C) are demonstrated in Fig. 5f–h, j–l. In the case of flat solar cells, power densities were derived from the constructive and destructive interfaces

respectively. The device modeling and optical character-

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**Fig. 2** 3D AFM images along with 2D images (inset) of the WO<sub>x</sub> thin films, synthesized at different sputtering powers: **a** 50 W, **b** 70 W, **c** 90 W; **d** average roughness and RMS roughness of prepared WO<sub>x</sub> films against RF sputtering powers

 Table 2
 Average and RMS surface roughness of the films sputtered at different RF powers

| RF sputtering power (W) | Average roughness $R_a$ (nm) | RMS<br>roughness<br>$R_q$ (nm) |
|-------------------------|------------------------------|--------------------------------|
| 50                      | 1.875                        | 2.314                          |
| 70                      | 2.88                         | 3.554                          |
| 90                      | 3.86                         | 4.717                          |

where no scattering has been observed. For shorter wavelengths (< 450 nm), most photons are absorbed by the WO<sub>x</sub> ETL, considered as a parasitic loss, which does not contribute to the quantum efficiency (QE) or short-circuit current density ( $J_{SC}$ ).

At 450 nm (Fig. 5b, f, j), photons are absorbed by the vicinity of the  $WO_x$ /perovskite interface and penetrate to

the few hundred nanometers of the perovskite bulk. The incremental wavelength improves photon absorption in the perovskite absorber (Fig. 5c, g, k). A light goes to the back contact for a longer wavelength (700 nm) and gets reflected; however, after 800 nm wavelength, close to the perovskite  $E_{\rm s}$ , no absorption (almost zero) occurs in the device. A slight change in power densities can be observed between different planar devices only in the case of shorter wavelengths; in contrast, all flat devices follow almost the same characteristics for longer wavelengths. The device optics can be further understood by the simulated QE and optical losses. Figure 6 compares the QEs, absorption loss, and reflection loss of PSCs, while the ETL layer varies from 75 to 118 nm, which further facilitates understanding of the optimum deposition power for WO<sub>x</sub>-based PSCs. QE, reflection, and parasitic losses of PSCs for the ETL thickness of 30, 50, and 400 nm are provided in the Supporting Information (Fig. S1a-c).



Fig. 3 Wavelength-dependent  $\mathbf{a}$  transmission,  $\mathbf{b}$  absorbance,  $\mathbf{c}$  reflectance, and  $\mathbf{d}$  absorption coefficient of the deposited films with different thicknesses

It has been found that the overall device absorption and reflections are not considerably affected by the thickness of the WO<sub>x</sub> layer; however, the ETL thickness impacts the absorption and  $J_{SC}$  of the solar cell. An increased ETL thickness enhances the parasitic absorption loss in the shorter wavelength regions, which do not contribute to the QE or  $J_{\rm SC}$  of the PSC (Fig. S1a, c). In this study, the  $J_{\rm SC}$  is maximized  $(21.1 \text{ mA/cm}^2)$  when the ETL thickness is 118 nm. On the other hand, with increased ETL layer (400 nm) leads to a drop in the  $J_{SC}$  by ~13% (18.6 mA/cm<sup>2</sup>). An increase in the deposition power leads to a rise in the WO<sub>x</sub> absorption and some changes within the spectral range (<700 nm) in the QE and reflection; however, it does not drastically affect the photon absorption of the solar cell. Flat-C has slightly higher absorption than Flat-A and Flat-B, which can be confirmed by the calculated  $J_{SC}$ s (Flat-A = 21 mA/  $cm^2$ , Flat-B = 21 mA/cm<sup>2</sup>, and Flat-C = 21.1 mA/cm<sup>2</sup>). A comparison of  $J_{SC}$ s for different thicknesses of ETL is shown in Fig. S1(d). This optical phenomenon is clearly validating the explanation described by the QE and power density plot. It is also seen the NiO/Ag has no major absorptions that can increase the parasitic absorption or decrease the QE. As the optical performance of the planar device is restricted due to high reflection and absorption losses, as a next step, we propose a PSC structure that is embedded with a silver (Ag) nanoparticle (NP) in the bulk of the perovskite absorber. The NP-PSC considers the optimized planar device structure (Flat-C), where only an Ag NP with a radius of 50 nm is embedded in the unit cell, as depicted in Fig. 7a. The Ag NP significantly influences the optics of the complete solar cell through a scattering of incident light. The photon scattering improves the light trapping, leading to enhanced QE over 850 nm, leading to an increased  $J_{SC}$  of the solar cell. Compared to the planar solar cell, the NP-PSC improves the QE and reduces the reflections substantially shown in Fig. 6, facilitating increasing the  $J_{SC}$  by ~ 17%. The



Fig. 4 a Determining the deposited films' energy bandgap ( $E_g$ ) using Tauc's plot. A comparison of **b** refractive index, **c** extinction coefficient, and **d** optical conductivity between different thickness WO<sub>x</sub> films prepared by the sputtering method

NP-PSC exhibits a  $J_{SC}$  of 24.5 mA/cm<sup>2</sup>. The power density and electric distribution plots of the proposed NP-PSC can help to further understand the optical wave phenomena on the device optics.

Figure 7b–d, e–g shows the cross-sectional power density and electric field distribution plots of the simulated NP-PSC for an incident wavelength of 450 nm, 600 nm, and 760 nm. Figure 7 clearly demonstrates how the NP contributes to photon scattering and enhanced light trapping, confirming the description presented in QE and reflection plots shown in Fig. 6. The standing wave pattern formed in the case of NP-PSC is due to the scattering effect. The power densities are substantially improved in the case of longer wavelength in the NP-PSC shown in Fig. 7d compared to the planar device (Fig. 5d, h, l). It has to be noted that the photon absorption can be further enhanced via improved light incoupling in a solar cell. Light incoupling can be increased by nanotexturing (e.g., pyramid, moth-eye, and nano-dome) the solar cell's front contact [27, 31, 36, 37]. Nevertheless, the dimension of the nanotexture plays a major role in achieving optimum light incoupling [13, 36]. Such nanotextures can be prepared by nanoimprinting and lithography processes. More details on light incoupling and light trapping in PSCs can be found in the literature [29, 31, 36]. Hence, it can be concluded that the deposited WO<sub>x</sub> as an ETL has great potential to improve the optical performance of the PSCs, and such ETL can also be applied to the realization of TSCs [19, 38, 39]. By considering the electrical parameters ( $V_{OC}$ =1.18–1.2 V and FF=82–84%) [2, 40, 41] of the best PSC along with the improved optics in the current study, the ECE of the NP-PSC can be estimated to be over 24%.



Fig. 5 A schematic cross-section of a PSC with the  $WO_x$  thickness of a 75 nm, e 93 nm, and i 118 nm. Simulated cross-sectional power density maps, at the center of the unit cell, for the PSC under mono-

chromatic wavelength of **b**, **f**, **j** 450 nm, **c**, **g**, **k** 600 nm, and **d**, **h**, **l** 760 nm for the deposition power of 50 W (**b**–**d**), 70 W (**f**–**h**), and 90 W (**j**–**l**), respectively

#### 4 Summary

In the first part of this study, tungsten oxide  $(WO_x)$  thin films were successfully synthesized using the RF magnetron sputtering method at room temperatures, which are used as the potential electron transport layer (ETL) for perovskite solar cells (PSCs). The influence of sputtering power on film thickness has been systematically studied. EDX analysis confirmed the incremental behavior of W atomic % with the increment of thickness due to the increasing sputtering power. Transmittance, absorbance, and reflectance measurement allowed determining the prepared thin films' bandgap (varied from 3.19 to 3.6 eV) and optical constants (refractive index and extinction coefficient). The second part focuses on

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Fig. 6 Comparing the **a** quantum efficiencies, **b** reflections, and  $WO_x$  absorption between different planar solar cells and the solar cell embedded with the Ag nanoparticle. The deposition power of 90 W was considered for the realization of the nanoparticle solar cell



Fig. 7 a A schematic cross-section of a PSC embedded with an Ag nanoparticle (NP-PSC) with a radius of 50 nm. Corresponding simulated cross-sectional **b**–**d** power density maps and **e**–**g** electric field distribution maps for incident wavelengths of 450 nm, 600 nm, and 760 nm

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investigating the optical characteristics of PSCs with the use of optimized WO<sub>x</sub> as an ETL. The optics and optimization of the solar cells were studied by three-dimensional finite-difference time-domain (FDTD) optical simulations integrated with the particle swarm optimization (PSO) algorithm. In the current study, the effect of sputtering power on the optical performance of the planar device has been thoroughly investigated. Finally, to overcome the limitation of a planar device, a PSC embedded with an Ag nanoparticle (NP) has been proposed to mitigate optical losses and enhance optical performance through improved light trapping. Incorporating NPs can significantly enhance the quantum efficiency in the longer wavelengths. The NP-PSC improved the  $J_{SC}$  by ~ 17% compared to the planar architecture. The  $J_{\rm SC}$  of the NP-PSC was found to be 24.5 mA/cm<sup>2</sup>. By considering the optimum electrical properties of a solar cell, the energy conversion efficiency of the proposed PSC is estimated to be beyond 24%. It is believed that the prepared  $WO_x$  can also be applied to the realization of high-efficiency tandem solar cells.

Supplementary Information The online version contains supplementary material available at https://doi.org/10.1007/s00339-022-05500-5.

Acknowledgements We acknowledge the Universiti Kebangsaan Malaysia, Malaysia for financial support through the Long-term Research Grant Scheme (LRGS/1/2019/ UKM-UKM/6/1). The work is partly supported by the Bangabandhu Fellowship Trust (BBFT), Bangladesh. The authors extend their appreciation for funding to Researchers Supporting Project number (RSP-2021/34), King Saud University, Riyadh, Saudi Arabia. We also acknowledge the Bangladesh Council of Scientific and Industrial Research for supporting using their experimental laboratories.

#### Declarations

Conflict of interest There are no conflicts of interest to declare.

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