**Minimizing the ohmic resistance of wide-bandgap perovskite for semi-transparent and tandem solar cells**

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**Abstract**

To overcome the efficiency limit of perovskite single-junction solar cells, it is vital to develop various types of tandem solar cells. Especially, wide-bandgap (WBG) perovskite solar cells (PSCs) have played an important role in high-efficiency tandem solar cells. Herein, an indium zinc oxide (IZO)-based interfacial structure was developed to improve the performance of a WBG PSC and used as the transparent electrode for semi-transparent (ST) PSCs. This approach minimized ohmic contact between the electron-transport layer and metallic electrode, which also accelerated electron transfer and suppressed trap-assisted carrier recombination. As a result, the WBG PSC (1.71 eV) showed the best power conversion efficiency (PCE) of 19.26% and improved operational stability. When the optimized ST-PSC was used as the ST-top cell, perovskite/CdTe four-terminal (4T) and perovskite/silicon (double-side polished) two-terminal (2T) tandem solar cells achieved a maximum efficiency of 22.59% and 26.34%, respectively.

**Keywords**

Wide-bandgap solar cell; Indium zinc oxide; Ohmic contact; Perovskite/CdTe tandem solar cell; Perovskite/silicon tandem solar cell

1. **Introduction**

Silicon and cadmium telluride (CdTe) solar cells are some of the most widely commercialized photovoltaic devices.1-5 The highest power conversion efficiency (PCE) of silicon and CdTe solar cells in the laboratory have reached 26.7% and 22.1%, respectively.6 Tandem solar cells can be developed to further enhance the PCE. The PCEs of metal halide perovskite solar cells (PSCs) have increased dramatically in the past decade, and the certified PCE of single-junction devices has reached 25.7%.6 The ability of perovskite materials to tune the bandgap within the range of 1.2–3.0 eV makes them suitable for tandem solar cells with other types of solar cells. 7

Highly-efficient perovskite-based tandem solar cells, such as perovskite/perovskite, perovskite/CdTe, and perovskite/silicon tandem solar cells, require the use of efficient wide-bandgap (WBG) (~1.7–1.8 eV) PSCs as the top cell.8-10 However, WBG perovskite solar cells have lower PCEs because of their severe open-circuit voltage deficit, phase segregation, and high trap density.11-13 Improving the quality of WBG perovskite films can solve these problems, and the specific methods include crystallization engineering, additive engineering, and surface passivation.14-18 For example, Yu et al. reported that using Pb(SCN)2 and phenyl-ethylammonium iodide as additives synergy improved the film crystallinity.19 Yu et al. introduced alkali pseudo-halide KBF4 into the perovskite precursor solution to reduce the number of defects in the WBG perovskite film. The champion WBG PSC (bandgap = 1.74 eV) delivered a high open-circuit voltage (*V*OC) of 1.21 V and a power conversion efficiency (PCE) of 17.49%.20 Chen et al. proposed an interfacial engineering strategy to form a 2D/3D perovskite heterojunction using 2-thiopheneethylammonium chloride (TEACl). The device treated with TEACl delivered a PCE of 20.31% with a 1.68 eV bandgap.22 It can be seen that the quality of perovskite films has been studied extensively, but interfacial contact is a non-negligible factor affecting device’s performance. The semiconductor-metal interfacial contact modes include Schottky contact and ohmic contact. Schottky contact means that at the metal-semiconductor interface, the energy band of the semiconductor bends to form a Schottky barrier, resulting in large interfacial resistance. Ohmic contact is a low-resistance junction (non-rectifying) that conducts current from the metal to the semiconductor. For solar cells, reducing contact resistance can be used to improve a device's performance.22-24

For PSCs with a p-i-n structure, PCBM is commonly used as the electron transport layer (ETL).25, 26 However, when the metallic electrode (Ag, Al, or Au) was directly evaporated on the PCBM surface, Schottky contact formed between PCBM and the metallic electrode, resulting in charge accumulation and relatively large series resistance, which decreased the device’s performance. In order to solve the problem’s mentioned above, a copolymer was used as a buffer layer between PCBM and the metallic electrode to improve ohmic contact.27, 28 To decrease the energy offset, Yang et al. used a polyethyleniemine ethoxylated (PEIE) layer between PCBM and the metallic electrode.29 To improve the electron extraction ability, Hang et al. used an Al-doped ZnO (AZO) layer between the PCBM and electrode.30 Brinkamn et al. introduced a bilayered aluminum-doped ZnO (AZO)/tin oxide (SnO*x*) electron-extraction layer (EEL) between the PCBM and electrode to improve the stability of the device.31 For p-i-n type perovskite solar cells, a compact layer of SnO*x* was deposited by atomic layer deposition (ALD) to suppress the ingress of moisture and enhance electron transport. This improved the performance and stability of the device.31,32 For semi-transparent (ST) solar cells and tandem solar cells, ALD-SnO*x* is an important buffer layer for preventing sputtering damage.33 However, although the main interfacial contact mode between the SnO*x* layer and metallic electrode is ohmic contact, the interfacial contact resistance is non-negligible. Interfacial contact can be improved by reducing the interfacial contact resistance, thus improving the device's performance.

In this work, ALD-SnO*x* was used as both the ETL and sputtering buffer layer. We sputtered a layer of indium zinc oxide (IZO) as the interfacial layer between the SnO*x* and Ag electrode to improve the device’s performance by reducing contact resistance. After introducing the IZO layer, the electron extraction ability was improved. Specifically, charge transfer from the perovskite to the ETL was more efficient, and non-radiative recombination was suppressed. IZO could also be used as the transparent electrode in ST solar cells.34 Thus, the champion p-i-n WBG perovskite solar cell (1.71 eV) delivered a maximum PCE of 19.26%. More importantly, by combining the ST-PSC with a CdTe bottom cell, a four-terminal (4T) perovskite/CdTe tandem solar cell with a PCE of 22.59% was obtained. This is one of the highest PCEs of perovskite/CdTe tandem solar cells. Finally, to verify the universality of the WBG ST-perovskite solar cell, a two-terminal (2T) perovskite/silicon (double-side polished) tandem solar cell with a PCE of 26.34% was obtained.

**2. Results and discussion**

A schematic of the structure of the device in this work is shown in Fig. 1a, where PTAA and PCBM acted as the hole transport layer (HTL) and ETL, respectively. We sputtered an IZO layer with a thickness of 100 nm between the SnO*x* and Ag electrode. An impermeable layer of SnO*x* was deposited by ALD. ALD-SnO*x* can be used as an ETL to enhance charge transfer and as a buffer layer to protect underlying layers from sputtering damage.31 The photovoltaic parameters of the devices with/without an IZO layer are presented in Fig. 1b, and the corresponding device parameters are summarized in Table 1. From the statistical data of the devices, it can be concluded that the performance of the WBG perovskite solar cells (1.71 eV, Fig. S1, Supporting Information) with an IZO layer was significantly improved. Fig. 1c displays the current density-voltage (*J*-*V*) curves of the champion WBG PSCs. Devices without an IZO layer (denoted as REF devices) showed a maximum PCE of 18.33% with a *V*OC of 1.173 V, a short-circuit current density (*J*SC) of 19.63 mA/cm2, and a fill factor (FF) of 79.54%. The device with an IZO layer (denoted as the IZO device) achieved a PCE of 19.26%, a *V*OC of 1.187 V, a *J*SC of 19.77 mA/cm2, and a FF of 82.11%. The IZO device showed enhanced performance because of the high *V*OC and FF, which were mainly ascribed to the efficient charge extraction/transport and a reduction in contact resistance, trap state density, and non-radiative recombination. These will be discussed in detail below.

Steady-state power output at maximum power point (MPP) tracking was carried out. The current density of the IZO device remained at 18.61 mA/cm2, and the PCE remained at 18.62% under continuous tracing under 1 sun illumination for 2000 s (Fig. 1d). Fig. S2 shows that the current density remained at 17.99 mA/cm2, and the PCE remained at 17.37% under continuous tracing under 1 sun illumination for 2000 s for the REF device. Fig. 1e shows the external quantum efficiency (EQE) spectrum and integrated *J*SC of devices with/without an IZO layer, which were measured by using a quantum efficiency measurement system (Enlitech QE-R). The integrated current densities were 19.55 mA/cm2 and 20.10 mA/cm2 for the REF and IZO devices, respectively, which are consistent with the *J*SC obtained from *J*−*V* measurements. The hysteresis index (H-index) was calculated with the equation of *H-index* = (*PCE*RS – *PCE*FS)/*PCE*RS, where *PCE*RS and *PCE*FS represent the PCE from the reverse scan (RS) and forward scan (FS), respectively.35 The hysteresis index of the IZO device was only 0.2% (Fig. 1f), so it can be ignored. We compared the stability of the REF and IZO devices under different environmental conditions (Fig. S3). The IZO devices maintained 98% of their initial efficiency after being stored in an inert environment for 4164 h, while the REF device decreased to 90% of its initial efficiency after 2544 h under the same conditions. The IZO device maintained 87% of its initial efficiency after being stored in ambient air at ~30% RH for 3669 h, whereas the REF device only retained 83% of its initial efficiency after 2049 h under the same conditions. As for the thermal stability at 85 oC, the IZO devices only showed an 18% loss in PCE after 396 h, while the REF device decreased to 77% of its initial efficiency after 204 h. Such results indicate that an effective interfacial contact by IZO layer can significantly improve the device stability.



Fig. 1. (a) Schematic structure of WBG PSCs (1.71 eV) with/without an IZO layer. (b) Statistics of the photovoltaic parameters for the REF device and the IZO device. (c) Best *J*-*V* characteristics of WBG PSCs (1.71 eV) with/without an IZO layer. (d) MPP tracking of the champion IZO device. (e) EQE spectra. (f) *J*-*V* curves of the champion IZO device based on the forward and reverse scans.

Table 1. Summary of photovoltaic parameters for the REF and IZO devices (1.71 eV)

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| **Device configurations** | ***J*sc**  **(mA/cm2)** | ***V*oc**  **(V)** | **FF**  **(%)** | **PCE**  **(%)** |
| **REF** | 19.38±0.23  （19.63） | 1.164±0.007  （1.173） | 77.51±1.55  （79.54） | 17.49±0.38  （18.33） |
| **IZO** | 19.86±0.08  （19.77） | 1.170±0.009  （1.187） | 81.21±0.64  （82.11） | 18.88±0.22  （19.26） |

Parameters of champion devices are in brackets.

To determine the chemical composition of IZO deposited by sputtering, X-ray photoelectron spectroscopy (XPS) was conducted (Fig. S4). The binding energies were analyzed by examining the core level spectra of In 3d (444.2 and 451.8 eV), Zn 2p (1021.6 and 1044.7 eV), and O 1s (529.7 and 531.7 eV), as shown in Fig. S4. The existence of strong In, Zn, and O peaks indicated that the sample included a mixture of In2O3 and ZnO. We conducted ultraviolet photoelectron spectroscopy (UPS) to measure the energy spectra of the IZO, as shown in Fig. 2a. The work function and valence band levels were calculated by subtracting the Fermi level from the vacuum energy level.36 In combination with the band gap (Fig. S5, the Tauc-plots of IZO), we calculated the energy positions of the valence band (VB) and conduction band (CB). The value of the CB of the IZO layer was calculated to be -4.3 eV. Previous literature has reported that the work function of Ag is -4.3 eV, and the conduction band of PCBM and ALD-SnO*x* were both-4.2 eV.37 The energy diagram of the ETL and electrode in PSC is depicted in Fig. 2b, which indicates favorable band alignment for electron extraction.37



Fig. 2. (a) Cutoff spectra of IZO (100 nm). (b) The energy level diagram of the relevant layers in PSCs. (c) Schematic of the TLM measurement. (d) Resistance-distance curves obtained by linear fitting to obtain *R*C values.

TLM measurements were used to determine changes in the contact resistance (*R*c) with/without an IZO layer. A schematic of the TLM measurement is shown in Fig. 2c. *R*C was calculated by the following equation:38,39

where *R*T is the total resistance measured from the current-voltage response curves between the two electrodes, *l* is the distance between the two electrodes, *w* is the width of the electrode, and *R*SH is the sheet resistance of the layer outside the contact region. As shown in Fig. 2d, 2*R*C was obtained from the intercept by the linear fitting of the resistance vs distance curves of the REF and IZO samples. The resistance of Ag electrodes at different distances was calculated from the *I*–*V* curves, as shown in Fig. S6. As a result, *R*C of the REF sample was 5.2 Ω, while that for the IZO sample was 4.7 Ω. The contact resistance between the IZO layer and Ag electrode of the IZO sample was lower than that between the SnO*x* layer and Ag electrode of the REF sample. This means that the IZO interfacial layer was beneficial for charge collection in perovskite solar cells.

The improvement of the interfacial contact between ETL and metal electrode can suppress the non-radiative recombination effectively.28 To further explore the non-radiative recombination behavior, the relationship between light intensity and *V*OC was investigated, as shown in Fig. 3a. This was used to evaluate the trap-assisted recombination mechanism in PSCs. Based on previous reports, when the ideality factor (*n*) is close to 1, Shockley-Read-Hall recombination is suppressed.40 The ideality factor is calculated by the following equation:41

where *n*, *k*B, *V*, *T*, and *q*e are the ideality factor, Boltzmann constant, voltage, absolute temperature, and elementary charge, respectively. As shown in Fig. 3a, the ideality factor was 1.53 for the REF device, which decreased to 1.44 for the IZO device. This indicates further suppression of trap-assisted carrier recombination for the IZO device. To further confirm the efficient charge extraction/transport with a suppression in the trap-state density, we prepared electron-only devices (ITO/SnO2/PVK/PCBM/ALD-SnO*x*/Ag and ITO/SnO2/PVK/PCBM/ALD-SnO*x*/IZO/Ag) to analyze changes in the electron trap states by using the space-charge limited current (SCLC), as shown in Fig. 3b. The defect density (*n*t) was calculated using the following equation:43

where *V*TFL, *e*, *L*, *ε*, and *ε*0 are the trap-filled limit voltage, elementary charge of the electron, thickness of perovskite (~450 nm, Fig. S7), relative dielectric constant of the perovskite (32), and vacuum permittivity, respectively. This shows that the IZO electron-only devices exhibited a lower voltage value (0.33 V) than the REF electron-only device (0.43 V). The calculated electron trap densities were 7.52×1015 cm-3 and 5.77×1015 cm-3 for the REF devices and IZO devices, respectively, which indicates the IZO interlayer can reduce the density of interfacial traps between the layers of SnO*x* and Ag contact. This result is in agreement with the measured ideality factor.42 The dark current density of the IZO device was almost one order of magnitude lower than that of the REF device, as shown in Fig. 3c. This shows that the IZO layer reduced current leakage and non-radiative recombination due to the high recombination resistance.43 Open-circuit voltage decay (OCVD) measurements can be used to reflect the recombination of charge carriers effectively. As shown in Fig. 3d, a longer *V*OC decay time showed that the IZO devices had a lower interfacial recombination rate.44



Fig. 3. (a) Measured *V*oc plotted against light intensity (dots), together with linear fits (solid lines) to the data of PSCs. (b) Dark *J*-*V* curves for the electron-only devices with/without the IZO layer. (c) The dark *J*-*V* characteristic curves of the REF and IZO devices. (d) Open-circuit voltage decay.

After using IZO as the interfacial layer to prepare high-performance WBG perovskite solar cells, we used IZO as the transparent electrode to prepare high-performance WBG ST-PSCs. For comparison, we prepared ST-PSCs based on IZO and ITO as the transparent electrode, respectively. As a result, IZO-based ST-PSCs show improved performance with negligible *J*-*V* hysteresis, as shown in Fig. 4a-b, Fig. S8a and Table S2. Compared with opaque PSCs (see Fig. 1c and Table 1), ST-PSCs had a lower *J*SC and *V*OC, but a higher FF was obtained due to excellent interfacial contact, thus PCE decay was not obvious. For the IZO-based ST-PSC, current density was maintained at 18.02 mA/cm2, and the PCE was maintained at 17.88% under continuous tracing under 1 sun illumination for 1000 s (Fig. S8 b). Compared to the Glass/Perovskite/PCBM/SnOx/ITO film, it can be observed that the Glass/Perovskite/PCBM/SnOx/IZO film shows more efficient PL quenching and lower charge transport lifetime (Fig. 4c-d and Table S1), indicating the presence of better contact interfaces, more efficient charge separation and lower interfacial trap density.45,46



Fig. 4. (a) Best performed *J*–*V* characteristics of the ITO ST devices and the IZO ST devices. (b) Statistics of photovoltaic parameters for the ITO ST devices and the IZO ST devices. (c) Steady-state PL spectra and (d) time-resolved PL spectra of Glass/Perovskite/PCBM/SnO*x*/ITO and Glass/Perovskite/PCBM/SnO*x*/IZO.

We further constructed a 4T perovskite/CdTe tandem solar cell whose structure is shown in Fig. 5a. The incident light came from the ITO/PTAA side of the ST-PSC. Here, a 6.25 cm2 perovskite filter prepared under the same processing conditions as the ST-PSC was used as the mask to measure the performance of filtered-CdTe, and the size of each device is shown in Table 2. Higher-energy photons were absorbed by the perovskite top cell, while lower-energy photons passed through the top cell and were absorbed by the CdTe bottom cell. As shown in Fig. 5b, the CdTe solar cell exhibited a PCE of 18.43%, with a *J*SC of 29.82 mA/cm2, a *V*OC of 0.799 V, and a FF of 77.35%. After the light was filtered by the ST perovskite top cell, the PCE of the CdTe bottom cell decreased to 4.39%, with a *J*SC of 7.35 mA/cm2, a *V*OC of 0.759 V, and a FF of 78.79%. The lower *J*sc of the filtered CdTe bottom cell was because that most of the short-wavelength photons were absorbed by the perovskite top cell. Only a portion of the photons in the near-infrared region could be absorbed by the CdTe bottom cell. The intensity of sunlight reaching the CdTe bottom cell was lower than that of the standalone CdTe cell, and the CdTe bottom cell had a lower *V*OC. To demonstrate the benefits of using IZO as TCO, we have prepared 4T tandem solar cell with ST-PSC using ITO layer as TCO. The *J*-*V* curves and corresponding photovoltaic parameters, and EQE spectra are shown in Fig. S9 and Table. S2. By comparison, the 4T PVK/CdTe tandem device using IZO as TCO showed better device’s performance (Table. S3).Overall, the ST perovskite top cell and the filtered CdTe bottom cell achieved a total PCE of 22.59% in the 4T perovskite/CdTe tandem solar cells. Table 2 shows the corresponding device parameters. Fig. 5c shows the EQE of the perovskite top cell and the filtered CdTe bottom cell. The integrated photocurrent densities were 19.08 mA/cm2 and 7.01 mA/cm2 for the perovskite top cell and filtered CdTe bottom cell, respectively, which are very close to the *J*-*V* measurements.



Fig. 5. (a) Schematic of device configuration, (b) *J*-*V* curves, and (c) EQE spectra of the 4T perovskite/CdTe tandem cell.

Table 2. The photovoltaic parameters of solar cells

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| **Device** | ***J*sc**  **(mA/cm2)** | ***V*oc**  **(V)** | **FF**  **(%)** | **PCE**  **(%)** | **Size**  **(cm2)** |
| **Perovskite (semi-transparent)** | 18.91 | 1.161 | 82.89 | 18.20 | 0.09 |
| **CdTe Cell** | 29.82 | 0.799 | 77.35 | 18.43 | 0.17 |
| **Filtered CdTe Cell** | 7.35 | 0.759 | 78.79 | 4.39 | 0.17 |
| **4T tandem** |  |  |  | 22.59 |  |

To verify the universality of the WBG ST-perovskite solar cell, we constructed a 2T perovskite/silicon tandem solar cell, as shown in Fig. 6a. It is important to deposit uniform NiO*x* film with full coverage on the silicon bottom cells by sputtering method for high-performance tandem solar cells.47 We also found that the PCE of Si/PVK tandem device without NiO*x* is lower due to the poor FF (Fig. S10 and Table S4), which may be attributed to the imperfect contact between Si and PSC. Besides, we also fabricated tandem solar cells based on both PCBM and C60, and found that C60 based tandem devices showed better PCE, as shown in Fig. S10. The improved performance may be ascribed to the homogeneity of C60 layer by evaporation method. Thus, the two-terminal perovskite/silicon tandem solar cell delivered a PCE of 26.34% (steady-state PCE of 25.63%, Fig. 6d), with a *J*SC of 17.43 mA/cm2, a *V*OC of 1.876 V, and a FF of 80.53% (Fig. 6b). The corresponding EQE spectra are shown in Fig. 6c. The integrated *J*SC values were 17.97 mA/cm2 and 17.58 mA/cm2 for the perovskite and silicon subcells, respectively, which matched the *J*SC value obtained from *J*-*V* measurements. Meanwhile, we have prepared 2T tandem solar cells with ITO layer as TCO, and corresponding *J*-*V* curves, EQE spectra, MPP tracking and photovoltaic parameters are shown in Fig. S11 and Table S2. It can be seen that tandem solar cells based on IZO show better performance (Table S3).



Fig. 6. (a) Schematic of the device configuration, (b) *J*-*V* curves, (c) EQE spectra of the 2T PVK/silicon tandem cell, and (d) MPP tracking of the champion tandem device.

**3. Conclusion**

In this study, we developed a method to fabricate efficient WBG PSCs by introducing an interfacial layer IZO. The improved performance of the PSCs was mainly ascribed to the reduction in contact resistance, efficient charge extraction/transport, and reductions in the trap state density and trap-assisted carrier recombination. We also used IZO as the transparent electrode to prepare high-performance WBG ST-PSCs, a 4T perovskite/CdTe tandem solar cell with an efficiency of 22.59%, and a 2T perovskite/silicon tandem solar cell with an efficiency of 26.34%.

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## Conflicts of interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## Author Contributions

**Haoran Ye**: Conceptualization, Methodology, Data curation, Writing – original draft. **Weiquan Xu**: XPS, UPS, PL, and TRPL measurements, Material preparation. **Fei Tang**: Improvement of PVK/silicon tandem solar cells. **Bohao Yu**: Improvement of ALD method. **Cuiling Zhang:** Modification ofperovskite formula. **Nanxi Ma**: Improvement of IZO sputtering method. **Feiping Lu**: Review of the manuscript. **Yuzhao Yang**: Formulation of overarching research goals and aims, Provision of resources, Acquisition of the financial support for the project leading to this publication, and Review of the manuscript. **Kai Shen**: Provision of CdTe solar cells, Review of the manuscript. **Weiyuan Duan**: Provision of silicon solar cells, Review of the manuscript. **Andreas Lambertz:** Provision of silicon solar cells, Review of the manuscript. **Kaining Ding:** Provision of silicon solar cells, Review of the manuscript. **Yaohua Mai**: Provision of resources, Oversight and leadership responsibility for the research activity planning and execution.

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