Room-Temperature-Processed ZrO$_2$ Interlayer toward Efficient Planar Perovskite Solar Cells

Jiawen Sun,$^\dagger$ Yuzhu Li,$^\dagger$ Naiwei Tang, Yang Zhou, Xiang Zhang, Xubing Lu,* Xingsen Gao, Jinwei Gao, Lingling Shui, Sujuan Wu,* and Jun-Ming Liu

**ABSTRACT:** The Sn-doped In$_2$O$_3$ transparent conductive (indium tin oxide (ITO)) electrode in planar perovskite solar cells (PSCs) is modified by a zirconia (ZrO$_2$) interlayer with a low-temperature process. Here, the ZrO$_2$ film is prepared by ultraviolet (UV) treatment at room temperature. The effects of the inserted ZrO$_2$ interlayer on the performance of CH$_3$NH$_3$PbI$_3$ is systematically studied. After optimizing the process, the champion efficiency of PSCs with a UV-treated ZrO$_2$ interlayer is 19.48%, which is larger than that of the reference PSC (15.56%). The improved performance in the modified devices is primarily ascribed to the reduced trap states and the suppressed charge recombination at the ITO/SnO$_2$ interface. Our work provides a facile route to boost the photovoltaic performance of PSCs by modifying the interface of the transparent conductive electrode at room temperature.

**KEYWORDS:** ITO/SnO$_2$ interface modification, room-temperature processed ZrO$_2$ interlayer, ultraviolet (UV) treatment, planar perovskite solar cell, photoelectric properties

1. **INTRODUCTION**

Recently, organic–inorganic halide perovskite solar cells (PSCs) have attracted considerable attention because of their excellent photoelectric properties.1–3 The certified power conversion efficiency (PCE) is over 25.0%.4–9 Usually a compact electron transfer layer (ETL) is used to block holes and transport electrons in PSCs. The pinholes in the ETL can cause shunt pathways and direct contacts of the conductive substrate and the perovskite film, which will result in severe carrier recombination at the interface.9 The thicknesses of ETLs in PSCs should be appropriate to efficiently transfer electrons and block holes. A thicker ETL will reduce transmission and increase series resistance ($R_s$), which will harm performance of devices. Moreover, the current leakage will be inevitable in PSCs with thin ETLs.

A lot of efforts have been made to modify the ETL layer to achieve better photovoltaic performance.10 Although most of the researchers have tried to modify the interface between the ETL and the perovskite film of PSCs,7 a little work has been made to modify the transparent conductive electrode/ETL interface. It has been found that the charge recombination at the electrode/ETL interface will also severely affect the open-circuit voltage ($V_{oc}$) and PCE of devices.11 Currently, tin oxide (SnO$_2$) films have become one of the most popular ETLs. However, there are still many problems needing to be solved. For example, it is difficult to obtain a SnO$_2$ ETL with a suitable thickness because of the low concentration of the SnO$_2$ precursor solution,12 resulting in defects and recombination at the electrode/SnO$_2$ interface. Thus, a simple SnO$_2$ ETL cannot effectively block holes and suppress charge recombination.13 To solve these problems, polyethylene glycol has been introduced into the SnO$_2$ precursor solution to fabricate the SnO$_2$ ETL with high coverage, as reported by Wei et al.12 Fang et al. have improved the photovoltaic performance of PSCs by modifying the indium tin oxide (ITO)/SnO$_2$ interface with a MgO layer annealed at 400 °C.15 It is one of the significant methods to modify the transparent conductive electrode/SnO$_2$ interface to suppress charge recombination. The related mechanism needs to be further investigated. Wide band gap materials such as SrO$_2$, CaCO$_3$, MgO, and ZrO$_2$ can be used to prevent the electron back transfer for high-performance PSCs.14–20 However, it is necessary to prepare them with high-temperature process. The PSCs can be fabricated by a low-temperature process. ETLs modified by insulating the oxide usually need to be annealed at high temperatures, which will increase costs and hinder their application on flexible plastic substrates. The ultraviolet (UV)

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annealing-ZrO₂ is used to modify the ITO/SnO₂ interface, lower valence band level can help to block holes, and improve the quality of the SnO₂ recombination during the charge transfer process. 13, 25, 26 A charge transportation at the ITO/SnO₂ interface. attributed to the reduced trap state density and the promoted

Based on these considerations, the UV-ZrO₂, or ZrO₂ film annealed at 160 °C (annealing-ZrO₂) has been deposited on an ITO substrate to modify the ITO/SnO₂ interface. The effects of ZrO₂ modification on the photovoltaic characteristics of PSCs are systematically investigated. The structure of our devices is ITO/ZrO₂/SnO₂/CH₃NH₃PbI₃-Cl/spiro-OMeTAD/Ag. For the convenience of presentation, the “reference-PSC/film” corresponds to the structure in which there is no ZrO₂ layer between ITO and SnO₂, “UV-PSC/film” and “annealing-PSC/film,” in which the UV-ZrO₂ or annealing-ZrO₂ is used to modify the ITO/SnO₂ interface, respectively. Under the optimum condition, the UV-PSC and annealing-PSC achieve the champion efficiency of 19.48 and 18.51%, respectively, which is greatly better than the value of reference-PSC (15.6%). The improved performance can be attributed to the reduced trap state density and the promoted charge transportation at the ITO/SnO₂ interface.

2. RESULTS AND DISCUSSION

Figure 1a,b shows the schematic illustration of the device structure and the corresponding cross-sectional scanning electron microscopy (SEM) image, respectively. Figure 1c presents the energy level diagram of materials used in PSCs. (d) XRD patterns for UV-ZrO₂ and annealing-ZrO₂ films.

![Figure 1](https://example.com/figure1.png)

Figure 1. (a) Schematic illustration of the device structure. (b) Cross-section SEM image of PSC. (c) Energy level diagram of materials used in PSCs. (d) XRD patterns for UV-ZrO₂ and annealing-ZrO₂ films.

Ozone process can induce photocatalysis to remove organic ligands and spontaneously crystallize. A high quality film with low pinhole density can be obtained by the UV process. Moreover, the UV-treated ZrO₂ (UV-ZrO₂) layer is compatible with the low-temperature process in PSCs. Until now, the UV-ZrO₂ film has not been used to modify the ITO/SnO₂ interface. In addition, the wettability of ITO is significantly improved after the deposition of UV-ZrO₂, which will help to improve the quality of the SnO₂ film. ZrO₂, and ITO/annealing-ZrO₂ thin films. The microstructure of the three films has also been characterized by atomic force microscopy (AFM), as shown in Figure 2d–f.

The root-mean-square (rms) roughnesses of ITO, ITO/UV-ZrO₂, and ITO/annealing-ZrO₂ thin films are 1.14, 4.37, and 4.48 nm, respectively. The slightly increased roughness in ITO/UV-ZrO₂ and ITO/annealing-ZrO₂ may be related to the presence of ZrO₂ nanocrystals. The top-view SEM images and the water contact angles of ITO/SnO₂, ITO/UV-ZrO₂/SnO₂, and ITO/Annealing-ZrO₂/SnO₂ are also investigated, as presented in Figure S2a–c. Figure S2d–f shows the AFM images of ITO/SnO₂, ITO/UV-ZrO₂/SnO₂, and ITO/annealing-ZrO₂/SnO₂, respectively. All films exhibit similar surface topography and hydrophilicity. The rms for ITO/annealing-ZrO₂/SnO₂ and ITO/UV-ZrO₂/SnO₂ are slightly larger than that of ITO/SnO₂, while they are similar in the ITO/annealing-ZrO₂/SnO₂ and ITO/UV-ZrO₂/SnO₂ films. Therefore, the inserted ZrO₂ interlayer has little effect on the microstructure of the SnO₂ film.

In order to obtain the best device, the preparation process of the ZrO₂ layer is optimized. Figures S3a and S5a show J–V curves of UV-PSCs and annealing-PSCs prepared by the Zr(AC)₄ precursor solutions with different concentrations, respectively. The Vₘₒₖ short-circuit current density (Jₘₒ), fill factor (FF), and PCE of PSCs at various concentrations are demonstrated in Figures S3b,c and S5b,c, respectively. Tables S1 and S3 demonstrate the detailed parameters, respectively. The PCE increases with the concentration of Zr(AC)₄ precursor solutions at the beginning. Then, the concentrations continue to increase and the performance of the device becomes worse. The UV-PSCs and annealing-PSCs demonstrate the highest PCE at 0.5 mg/mL. Thus, the optimum concentration (0.5 mg/mL) is the condition in our previous experiments (Figures 3–6, S2, S4, S6–S9). In addition, the processing times for UV-PSCs and annealing-PSCs were optimized. Figures S4a and S6a show J–V curves of UV-PSCs and annealing-PSCs with different treatment times, respectively. Figures S4b,c and S6b,c demonstrate the changes of corresponding photovoltaic parameters with different times. The corresponding photovoltaic parameters are presented in Tables S2 and S4. It is found that the optimal time for UV-PSCs and annealing-PSCs is 45 and 60 min, respectively. All samples in Figures 3–6, S2, S3, S5, and S7–S9 were prepared under this condition.

To study the effects of UV-ZrO₂ and annealing-ZrO₂ films, Figure 3a demonstrates the J–V curves of reference-, UV-, and annealing-PSCs. Table 1 presents the corresponding photovoltaic parameters. The UV-, annealing-, and reference-PSCs achieve their champion PCE of 19.48% (Vₘₒₖ: 1.068 V, Jₘₒ: 23.32 mA/cm², FF: 0.782), 18.51% (Vₘₒₖ: 1.042 V, Jₘₒ: 22.86 mA/cm², FF: 0.761), and 15.56% (Vₘₒₖ: 0.985 V, Jₘₒ: 20.35 mA/
cm², FF: 0.759), respectively. Obviously, the UV- and annealing-PSC achieve higher PCE, suggesting that the inserted ZrO₂ interface layer can enhance the performance of PSCs. To verify the reproducibility, 40 devices for each type of PSCs were prepared. Figure 3b–f demonstrates the distributions of detailed parameters. Compared to the reference-PSCs,
The UV- and annealing-PSCs demonstrate the enhanced photovoltaic parameters. Moreover, the UV-PSCs show the best performance. These results confirm the positive effect of the inserted ZrO2 layer. Related mechanisms will be discussed in the following. Tables S5−S7 in the Supporting Information have listed the values of $R_{sh}$, $R_s$ and $R_{sh}/R_s$ extracted from the J−V measurement from 40 devices for each type of PSC. The FF is related to the $R_{sh}/R_s$.27 The larger average $R_{sh}/R_s$ contributes to higher FF values in UV- and annealing-PSCs.28 To better characterize the performance of devices, the hysteresis behavior has been investigated. Figure S7 demonstrates the J−V curves of the three PSCs measured under different scan directions. According to the report, the hysteresis index (HI) factor can be used to evaluate the J−V hysteresis behavior.29 The calculated HI for the three PSCs are 0.032, 0.064, and 0.265, respectively. Obviously, hysteresis has

**Table 1. Photovoltaic Parameters of UV-, Annealing-, and Reference-PSCs**

<table>
<thead>
<tr>
<th>Type</th>
<th>$V_{oc}$ (V)</th>
<th>$J_{sc}$ (mA/cm²)</th>
<th>FF</th>
<th>efficiency</th>
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<tr>
<td>reference-PSC</td>
<td>0.985</td>
<td>20.35</td>
<td>0.759</td>
<td>15.56% (14.77)</td>
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<tr>
<td>UV-PSC</td>
<td>1.068</td>
<td>23.32</td>
<td>0.782</td>
<td>19.48% (19.01)</td>
</tr>
<tr>
<td>annealing-PSC</td>
<td>1.042</td>
<td>22.86</td>
<td>0.761</td>
<td>18.51% (17.63)</td>
</tr>
</tbody>
</table>

*a Champion efficiency. b Average PCE for 40 PSCs. c Measured $J_{sc}$. d Integrated current density from EQE.

**Figure 5.** Dark I−V curves for (a) SnO₂, (b) UV-ZrO₂/SnO₂, and (c) annealing-ZrO₂/SnO₂ devices. Green lines are the fitting results of the measured data (square); (d) steady-state PL and (e) time-resolved photoluminescence (TRPL) spectra for reference-, UV-, and annealing-films, respectively. Solid curves correspond to the fitting results. (f) Photovoltage decay curves for reference-PSC, UV-PSC, and annealing-PSCs.

**Figure 6.** (a) Nyquist plots of reference-, UV-, and annealing-PSCs. The experimental data and the corresponding fitting result correspond to open symbol and solid line, respectively. Inset: the equivalent circuit model used to fit the Nyquist plots. (b) Histograms of $R_{rec}$ and $R_{tra}$ acquired from the fitting results. (c) Dark $J$−$V$ curves of reference-, UV-, and annealing-PSCs. Light intensity dependence of (d) $V_{oc}$; (e) $J_{sc}$; and (f) FF for reference-, UV-, and annealing-PSCs.
been reduced after the insertion of the ZrO2 layer, and the UV-PSC shows the lowest HI, suggesting that the passivation effect of the UV-ZrO2 layer is better than that of the annealing-ZrO2 layer. To confirm the reliability of J−V results, the stable power output was measured at their optimal bias voltage (V_{opt}) under AM1.5 illumination, as shown in Figure 3g. Reference-, UV-, and annealing-PSCs, respectively, achieve their stabilized PCE of 13.29, 18.63, and 17.54%, which are similar to their corresponding average PCE of J−V measurement. In addition, air stability of PSCs has been investigated. Figure S8 presents the evolution of the normalized PCE for the three types of devices stored in air, indicating that all devices exhibit similar air stability. The possible reason may be attributed to the fact that the intrinsic properties of the perovskite material determine the stability of PSCs. The effect of the ZrO2 interlayer on the absorption and transmission of ETL was also investigated. Figure S9a presents the transmittance spectra of ITO/SnO2, ITO/UV-ZrO2/SnO2, and ITO/annealing-ZrO2/SnO2. All films show a similar transmittance to the ITO substrate, suggesting that the ZrO2/SnO2 layer has a negligible effect on the light transmittance. Figure S9b shows the absorption spectra of the three PSCs. Just similar to the transmittance, their absorbance spectra are similar. Figure 3h demonstrates the EQE and their corresponding integrated current density curves of the three devices. The integrated current density for the three PSCs are 19.78, 23.14, and 22.53 mA/cm². They are consistent with the J−V results. Compared to the similar transmittance and absorbance spectra of the three perovskite films, the EQE and integrated current density are significantly increased after ZrO2 insertion, which may be due to the reduction of defects and the suppressed charge recombination at the ITO/SnO2 interface. They will be discussed in the following.

To further study the microscopic effects of the inserted ZrO2 layer, the average photocurrent and contact potential difference (CPD) of the three ETLs are studied by C-AFM and KPFM, respectively. Figure 4a−c shows the KPFM images of the corresponding ETLs, and Figure 4d demonstrates the histograms of average CPD for the three films. Obviously, the UV-ZrO2/SnO2 and annealing-ZrO2/SnO2 films demonstrate the lower average CPD values compared to the reference SnO2 film, suggesting that the insertion of UV-ZrO2 or annealing-ZrO2 film increases the work function of the SnO2 film. This higher energy level enables more efficient electron extraction. This can explain the enhanced V_{oc} in the modified devices. The local current distributions can be investigated by C-AFM, which is associated with local photocurrent properties of current density. The C-AFM images of the three ETLs are shown in Figure 4e−g. Figure 4h presents the histograms of the average photocurrent for the three ETLs. The values of average photocurrent rise from 22 pA for the SnO2 film to 181 pA for the UV-ZrO2/SnO2 film and 137 pA for the annealing-ZrO2/SnO2 film. The average photocurrent value of C-AFM is in accordance with the current density of devices, which also explains the increased J_{sc} in the UV-PSCs and annealing-PSCs.

Table 2. Fitting Results of TRPL Spectra

<table>
<thead>
<tr>
<th>ETL Type</th>
<th>τ1 (ns)</th>
<th>τ2 (ns)</th>
<th>B1</th>
<th>B2</th>
<th>A1 (%)</th>
<th>A2 (%)</th>
<th>τ_{mean} (ns)</th>
</tr>
</thead>
<tbody>
<tr>
<td>reference-film</td>
<td>72.67</td>
<td>78.41</td>
<td>0.967</td>
<td>0.329</td>
<td>76.55</td>
<td>26.85</td>
<td>76.68</td>
</tr>
<tr>
<td>UV-film</td>
<td>13.69</td>
<td>19.78</td>
<td>0.972</td>
<td>0.279</td>
<td>83.85</td>
<td>29.33</td>
<td>17.28</td>
</tr>
<tr>
<td>annealing-film</td>
<td>83.31</td>
<td>46.21</td>
<td>0.914</td>
<td>0.857</td>
<td>26.32</td>
<td>34.21</td>
<td>37.74</td>
</tr>
</tbody>
</table>

The trap-state density (N_{trap}) of the ETL has been characterized with space-charge-limited current method. The device structures are ITO/SnO2/PCBM/Au and ITO/annealing (or UV)-ZrO2/SnO2/PCBM/Au. Figure 5a−c shows their dark I−V curves, respectively. The N_{trap} can be calculated by this equation

\[ N_{\text{traps}} = \frac{2\varepsilon_0\varepsilon_r V_{\text{TRPL}}}{\varepsilon_r L^2} \]

where \( \varepsilon_0 \) and \( \varepsilon_r \) correspond to vacuum permittivity and relative permittivity (\( \varepsilon_r = 9 \)), respectively. \( L \) is the thickness of SnO2 film (22.7 nm, Figure S10). The calculated N_{trap} is 2.43 × 10^{16} cm\(^{-3}\) for SnO2, which is higher than 1.85 × 10^{16} cm\(^{-3}\) of annealing-ZrO2/SnO2 film and 1.54 × 10^{16} cm\(^{-3}\) of UV-ZrO2/SnO2 film. Obviously, the UV-ZrO2/SnO2 film demonstrates the lowest N_{trap} suggesting that UV-ZrO2 is better than annealing-ZrO2. This is consistent with the best performance of UV-PSCs.

In addition, the steady-state PL spectra were carried out to estimate non-radiative recombination, which is related to trap states. Figure 5d shows the steady-state PL of perovskite films on the three different ETLs. The UV-film and annealing-film present lower PL intensity compared to reference-film, suggesting the promoted electron extraction and the reduced carrier recombination in UV-film and annealing-film. The TRPL were measured and shown in Figure 5e. The TRPL curves can be theoretically fitted with biexponential decay eqs 1−3

\[ y = y_0 + B_1 \exp\left(-\frac{x}{\tau_1}\right) + B_2 \exp\left(-\frac{x}{\tau_2}\right) \]

\[ A_i = \frac{B_i\tau_i}{B_1\tau_1 + B_2\tau_2} \quad (i = 1, 2) \]

\[ \tau_{\text{mean}} = \frac{\tau_1 A_1 + \tau_2 A_2}{A_1 + A_2} \]

where \( y_0 \) is the constant; the fast decay (\( \tau_1 \)) originates from the nonradiative recombination. The slow decay (\( \tau_2 \)) is associated with the recombination of free charge, and \( B_1 \) and \( B_2 \) are relative amplitudes of fast component and slow component, respectively. The relative contribution \( A_i \) of the two components and the mean carrier lifetime \( \tau_{\text{mean}} \) can be calculated by the eqs 1 and 2. Table 2 provides the fitting results. The values of \( \tau_{\text{mean}} \) for reference-, UV-, and annealing-films are 76.68, 17.28, and 37.74 ns, respectively. Obviously, the UV- and annealing-film show the shorter lifetime, suggesting a lower electron \( N_{\text{trap}} \) faster electron transfer, and reduced charge recombination rates at the ITO/ZrO2/SnO2/perovskite interface. Figure 5f demonstrates the photovoltage decay curves of PSCs. The photovoltage decay in the UV- and annealing-PSC is slower compared to that of reference-PSC. Photovoltage decay corresponds to the reduced electron density, which is related to charge recombination. The slower photovoltage decay indicates the lower recombination rates in the UV- and annealing-PSC, which contributes to
higher $V_{oc}$ and FF.\textsuperscript{46–48} This is consistent with better photovoltaic parameters in UV- and annealing-PSC.

In addition, electrochemical impedance spectroscopy (EIS) of the three types of PSCs was recorded under illumination. Figure 6a presents the Nyquist curves of reference-, UV-, and annealing-PSCs. The results were fitted with the inserted equivalent circuit model. The recombination resistance ($R_{rec}$) and transport resistance ($R_{tra}$) are obtained from the fitting results, and the fitted parameters are provided in Table 3. The larger $R_{rec}$ values in UV- and annealing-PSC confirm that the inserted ZrO$_2$ film can suppress charge recombination at the ITO/SnO$_2$ interface.\textsuperscript{49–51} Figure 6b lists the histograms of $R_{tra}$ and $R_{rec}$ for the three PSCs. It is noted that the UV- and annealing-PSC demonstrate smaller $R_{tra}$ and bigger $R_{rec}$ values, suggesting that the inserted ZrO$_2$ film at the ITO/SnO$_2$ interface can effectively reduce charge recombination rates.\textsuperscript{2,52–54} Figure 6c demonstrates the dark $J–V$ curves of PSCs. The UV- and annealing-PSC demonstrate the smaller leakage current than the reference-PSC, implying that the inserted ZrO$_2$ layer can effectively decrease the charge recombination.\textsuperscript{2,55} This will contribute to increasing the PCE of UV- and annealing-PSC.

To further understand the recombination kinetics of photogenerated carriers, light intensity ($P_{light}$) dependence of the $J_{oc}$, $V_{oc}$ and FF was measured and shown in Figure 6d–f. The linear relationship of the $V_{oc}$ and $P_{light}$ (natural logarithmic scale) was fitted by the following eq \textsuperscript{(4)} \textsuperscript{56,57}

$$V_{oc} = \frac{n k T}{q} \ln(P_{light}) + \text{constant}$$

where $k$, $T$, and $q$ are Boltzmann constant, absolute temperature, and elementary charge, respectively. It is well known that all photogenerated carriers will recombine in PSCs under an open circuit condition.\textsuperscript{58} For the bimolecular recombination only, the slope of versus $V_{oc}$ ln($P$) is equal to $kT/q$.\textsuperscript{58,59} When the slope is larger than $kT/q$, it means that trap-assisted recombination determines the performance of PSCs.\textsuperscript{56,60} Figure 6d shows the slope of the reference-PSC (1.75$kT/q$), UV-PSC (1.44$kT/q$), and annealing-PSC (1.56$kT/q$). In addition, the $V_{oc}$ of reference-PSC demonstrates the strongest dependence on $P_{light}$ indicating that the ZrO$_2$ modification suppress the trap-assisted charge recombination.\textsuperscript{60} The dependence of $J_{oc}$ versus $P_{light}$ can be described by the eq \textsuperscript{(5)} \textsuperscript{57}

$$J_{oc} \propto P_{light}^\alpha$$

where $\alpha$ corresponds to the exponential factor. At the short circuit, the bimolecular recombination is the minimum for the maximum charge sweep out ($\alpha \approx 1$).\textsuperscript{61} As seen in Figure 6e, the values of $\alpha$ for reference-, UV-, and annealing-PSCs are 0.969, 0.985, and 0.979, respectively. All of them approach 1, suggesting that the bimolecular recombination in these devices is negligible. Figure 6f presents the dependence of FF to $P_{light}$. It is noted that the FF in reference-, UV-, and annealing-PSCs almost remains unchanged at low $P_{light}$ (just under 0.35 Sun). When the $P_{light}$ is further increased, the value of FF in reference-PSC decreases faster compared with those of UV- and annealing-PSC, suggesting the lower recombination rates in the UV- and annealing-PSC.\textsuperscript{61} All of these results reveal that the inserted ZrO$_2$ film at the ITO/SnO$_2$ interface can reduce the charge recombination rates.

### 3. CONCLUSIONS

In summary, the ZrO$_2$ interlayer prepared by UV treatment at room temperature is used to modify the ITO/SnO$_2$ interface. At the optimum process, the UV-PSC achieves a champion PCE of 19.48\% with negligible $J–V$ hysteresis. All experimental results show that the improvement in device performance results from the suppressed charge recombination and the promoted charge extraction at the ITO/SnO$_2$ interface. Our work provides a new method to fabricate PSCs with an excellent performance by modifying the ITO/SnO$_2$ interface at room temperature.

### ASSOCIATED CONTENT

**Supporting Information**

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsaem.9b02356. Spectra for the thickness measurement of ZrO$_2$ and SnO$_2$ films; micrographs of SnO$_2$, UV-ZrO$_2$/SnO$_2$, and annealing-ZrO$_2$/SnO$_2$ films; $J–V$ curves of UV-PSCs and annealing-PSCs with different precursor concentrations and treatment times; $J–V$ curves for the three types of best-performing devices measured in both the reverse and forward scan directions; normalized PCE evolution of devices; transmittance spectra of ITO/SnO$_2$, ITO/UV-ZrO$_2$/SnO$_2$, and ITO/annealing-ZrO$_2$/SnO$_2$ films, and absorption spectra of the corresponding devices; summarized photovoltaic parameters corresponding to the Figures S2–S6; and values of $R_{sh}$, $R_{p}$, and $R_{s}/R_{sh}$ extracted from the $J–V$ measurement of reference-PSCs, UV-PSCs, and annealing-PSCs from 40 PSCs (PDF)

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**Table 3. Fitting Parameters of EIS Data for Reference-PSC, UV-PSC, and Annealing-PSC**

<table>
<thead>
<tr>
<th>Type</th>
<th>$R_s$ ($\Omega$)</th>
<th>$R_{tra}$ ($\Omega$ cm$^2$)</th>
<th>$R_{rec}$ ($\Omega$ cm$^2$)</th>
<th>CPE1-T</th>
<th>CPE1-P</th>
<th>CPE2-T</th>
<th>CPE2-P</th>
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<tr>
<td>reference-PSC</td>
<td>23</td>
<td>63</td>
<td>56</td>
<td>2.79 × 10$^{-4}$</td>
<td>0.83</td>
<td>3.86 × 10$^{-4}$</td>
<td>0.89</td>
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<tr>
<td>UV-PSC</td>
<td>10</td>
<td>52</td>
<td>108</td>
<td>1.04 × 10$^{-7}$</td>
<td>0.76</td>
<td>2.01 × 10$^{-7}$</td>
<td>0.81</td>
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<tr>
<td>annealing-PSC</td>
<td>13</td>
<td>56</td>
<td>97</td>
<td>2.03 × 10$^{-7}$</td>
<td>0.79</td>
<td>2.65 × 10$^{-7}$</td>
<td>0.84</td>
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