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Perovskite Solar Cells on Polymer-Coated Smooth and Rough Steel Substrates

Benjamin T. Feleki, Ricardo K. M. Bouwer, Martijn M. Wienk, and René A. J. Janssen*

Fabricating efficient perovskite solar cells on steel substrates could enable easy building integration of this photovoltaic technology. Herein, an n−i−p perovskite solar cell is developed on steel substrates for top illumination. The optimized stack uses a Ti bottom electrode, covered with an indium tin oxide (ITO) interlayer and a SnO₂ electron transport layer passivated by [6,6]-phenyl-C₆₁-butyric acid. The active layer is a triple-cation perovskite. A thermally evaporated tris(4-carbazolyl-9-ylphenyl)amine/MoO₃ bilayer acts as hole transport layer. The transparent top contact consists of ITO with a MgF₂ antireflective coating. Optical analysis shows small parasitic absorption and reflectance losses for this stack, which provides 15.9% power conversion efficiency when fabricated on glass. On steel, covered with a polyamide imide planarization coating to moderate the surface roughness (Rₛ), the highest efficiency is 15.2% for high-gloss steel (Rₛ ≈ 200 nm), 14.9% for battery steel (Rₛ ≈ 500 nm), 14.2% for packaging steel (Rₛ ≈ 1500 nm), and 13.8% for construction steel (Rₛ ≈ 2500 nm). While the short-circuit current density and open-circuit voltage are invariant, the fill factor decreases with increasing Rₛ due to increasing series resistance and decreasing shunt resistance. The yield of working devices remain high, also for the roughest substrates.

1. Introduction

Building integration of perovskite solar cells could 1 day become feasible because of their low cost, aesthetics, lightweight, and impressive power conversion efficiency (PCE).[1-5] When focusing on potential substrate materials compatible with the building industry, coated steel offers an interesting perspective because it is one of the most common architectural materials, especially in industrial buildings. Steel is a cheap (substrate) material and offers excellent mechanical, heat resistance, and barrier properties against oxygen and humidity.[6-7] Combining perovskite solar cells with steel can give added value to this commonly employed building material. One of the challenges to tackle when fabricating solar cells directly on steel substrates is the higher surface roughness as compared to glass or polymer film which can be fatal for thin-film solar cells. Using smooth steel substrates would add to the cost due to the extra surface polishing steps. The cost can be reduced when combining rough steel substrates with an additional planarization layer.[8]

Fabricating perovskite solar cells on rough substrates may reduce device performance and yield, due to irregularities such as spike-like protrusions, valleys, and peaks. To investigate the impact of surface roughness on the photovoltaic performance, we developed a substrate-configuration n−i−p solar cell for coated steel substrates (Figure 1). Fabrication of perovskite solar cells on rough substrates has been mostly studied in superstrate-configuration single-junction and top-illuminated perovskite Si monolithic tandem solar cells.[9-15] In several studies on tandem solar cell applications a rough pyramidal-textured Si substrate has been used. To achieve a conformal coverage of the perovskite active layer, the perovskite layer needs to be sufficiently thick, or it needs to be deposited via co-evaporation[9-11] or a hybrid evaporation/spin coating deposition method.[12-14] Tockhorn et al. demonstrated conformal coating of the perovskite active layer in single-junction superstrate-configuration perovskite solar cells by employing a self-assembled [2-(9H-carbazol-9-yl)ethyl]phosphonic acid (2PACz) monolayer hole transport layer (HTL) on a nanotextured indium tin oxide (ITO) glass substrate providing 19.7% efficiency.[15] In substrate-configuration perovskite solar cells, most devices have been fabricated on polished Ti foils, reaching efficiencies up to 15%.[16] Although most studies on substrate-configuration perovskite solar cells use polished Ti...
foils, the impact of substrate surface roughness as an important parameter for future upscaling was not investigated. Herein, we have developed a substrate-conﬁguration n–i–p perovskite solar cell for integration with polymer-coated steel substrates and investigate the in‡uence of the substrate surface roughness on the photovoltaic parameters. We start from a semitransparent n–i–p perovskite solar cell on an ITO-covered glass substrate having a transparent ITO top electrode coated with an antirefective coating. In a stepwise fashion we transformed this cell into an optimized substrate-conﬁguration device stack with an opaque Ti bottom electrode. Fabricated on glass, this opaque cell gave 15.9% PCE with top illumination, and the PCE dropped only slightly when fabricated on different surface quality polymer-coated steel substrates to reach PCEs between 15.2% for high gloss steel and 13.8% for construction steel. The increase in surface roughness of the substrate was found to mainly affect the ﬁll factor (FF) of the cells but not the short-circuit current density (Jsc), open-circuit voltage (Voc), or fabrication yield of working devices.

2. Results and Discussion

The modiﬁcation steps in transforming a semitransparent solar cell fabricated on glass, via a top-illuminated device with an opaque metal bottom electrode, into a cell that can be fabricated on steel are shown in Figure 1. The different layer thicknesses in these stacks are collected in Table S1, Supporting Information and the cells are discussed in detail in the next subsections.

2.1. Semitransparent Cells

A key aspect of an efﬁcient substrate-conﬁguration perovskite solar cell is the use of a transparent top electrode that enables in-coupling of the light that is to be absorbed by the perovskite semiconductor. In a n–i–p conﬁguration, this not only requires the use of a transparent top electrode, but also a weakly absorbing, transparent HTL. We have recently shown that thin (7–10 nm), pristine, thermally evaporated organic HTLs can signiﬁcantly reduce the parasitic absorption by the HTL compared to the more commonly used thick and doped Spiro-OMeTAD layers.[17] Here, we used tris(4-carbazoyl-9-ylphenyl)amine (TCTA). To protect the thin TCTA layer against damaging, it was covered with a layer of thermally-evaporated MoO3 before depositing ITO by sputtering. To reduce re‡ection of light caused by the high-refractive index ITO top contact, we employed a thermally evaporated MgF2 anti-re‡ective coating. To test the performance of the TCTA/MoO3/ITO/MgF2 top contact, we fabricated a semitransparent perovskite solar cell (Figure 1, stack A). In this cell, the bottom contact consists of an ITO-covered glass substrate with a SnO2 electron transport layer (ETL) deposited from an aqueous SnO2 nanoparticle dispersion and passivated with [6,6]-phenyl-C61-butyric acid methyl ester (PCBM). As active layer, we used a triple-cation Cs0.05(MA0.17FA0.83)0.95Pb(I0.83Br0.17)3 perovskite (MA is methylammonium, FA is formamidinium).[18] The current density–voltage (J–V) characteristics for this cell recorded under simulated AM1.5G (air mass 1.5 global) solar light provided a stabilized PCE of 16.1% when illuminated from the bottom side (glass/ITO). This cell provided a Jsc,EQE of 19.7 mA cm−2, obtained via integration of the external quantum efﬁciency (EQE) spectrum with the standard AM1.5G solar spectrum, a Voc of 1.17 V, and a FF of 0.70 (Figure 2a and Table 1). With top-side (ITO/MgF2) illumination the same device reached a nearly identical PCE of 16.7% with very similar Jsc,EQE (19.5 mA cm−2), Voc (1.17 V), and FF (0.73) (Table 1). Differences in the shape and magnitude of the EQE spectra between the two illumination directions are minimal (Figure 2b). The reduced EQE for wavelengths below 400 nm with top-side illumination are caused by the parasitic absorption of light...
by the sputtered amorphous ITO top electrode. Concomitantly, the higher EQE values with top-side illumination for wavelengths above 500 nm are due to a reduced reflection owing to the antireflective MgF2 coating. From these results, we conclude that the use of a thin TCTA/MoO3 HTL in combination with an ITO/MgF2 electrode results in minimal optical losses with top illumination compared to bottom illumination from via the glass/ITO/SnO2/PcBa layers.

2.2. Opaque Substrate-Configuration Cells on Glass

In the next steps (A→BII, BIII, and BIII), we fabricated opaque substrate-configuration solar cells on glass substrates. The prime reason to use glass in this step of the optimization is to first exclude effects of surface roughness introduced by the steel substrates. We started with a highly reflective opaque Au bottom electrode, which was covered with a thin (10 nm) sputtered ITO interlayer to significantly improve the wetting of the aqueous SnO2 nanoparticle dispersion used for depositing the ETL on the hydrophobic Au bottom electrode via spin coating. The SnO2 ETL was passivated with PCBM (B1 stack) or with [6,6]-phenyl-C61-butyric acid (PCBA) (BIII stack). By binding to the SnO2, PCBA is better retained during processing of the Cso.05(MA0.17FA0.83)0.95Pbl0.83Br0.171 absorber layer on top.[19] While passivation of SnO2 with PCBA does not increase the maximum PCE compared to PCBM, it dramatically increases the yield of efficient devices and reduces the spread as shown in Figure 3a,c. Under slow sweep conditions the best PCBA-passivated (BIII stack) cell reached 16.1% efficiency, compared to 17.4% for the best cell with PCBM-passivation (B1 stack) (Table 2). The much higher yield of well-performing BIII-stack devices is a consequence of a significantly better wetting of the perovskite precursor solution on the PCBA-passivated SnO2 than on PCBM-passivated SnO2 (Figure S1, Supporting Information), which leads to a significant reduction of shorted devices and much less spread in the device performance (Figure 3c). The lower Jsc,EQE for the best BIII device compared to the best B1 cell is due to a slight reduction in the EQE values over the relevant wavelength regime (Figure 3b) and can be explained by variations in layer thicknesses. The second modification (BII→BIII) involved replacing the Au bottom electrode with a Ti bottom electrode. Compared to Au, Ti is more cost effective and is commonly used as bottom electrode in substrate-configuration n–i–p perovskite solar cells. Ti did not alter the J–V characteristics significantly and the cell reproducibility remained good (Figure 3a,c). The best performing BIII substrate cell reached a PCE of 15.9% with lower Jsc,EQE (19.5 mA cm−2) but slightly higher Voc (1.15 V) and FF (0.70) compared to best Au-based (BII) cell (Table 2). The reduced Jsc,EQE for the Ti-based devices is due to the slightly lower EQE above 550 nm (Figure 3b), owing to reduced reflection from the Ti bottom electrode compared to Au.

The simulated optical absorption and reflection caused by the individual layers in stack BII and BIII cells with Au and Ti bottom electrodes are visualized in Figure 4. The figure shows the product of the absorbance of the various layers (or cell reflectance) with the AM1.5G photon flux [Φ] as function of wavelength. Table 3 summarizes the corresponding contributions to and losses of photocurrent as the product of the elementary charge q [C] and Φ that is reflected, transmitted, or absorbed after integration over the relevant spectral range of the two cells.

The simulated total optical loss in the Ti-based BIII stack is 2.8 mA cm−2, compared to 2.2 mA cm−2 for the Au-based BIII stack (Table 3). In both solar cells, the main optical losses are caused by reflection of light from the top MgF2 coating (0.8–0.9 mA cm−2, red areas in Figure 4), absorption of light by the ITO top electrode (1.0 mA cm−2, yellow areas in Figure 4), and the MoO3 buffer layer (0.2 mA cm−2, blue areas in Figure 4). The main difference between the two stacks is the increased absorption of light by the Ti bottom electrode.
(0.8 mA cm⁻²) compared to (0.1 mA cm⁻²) for the Au bottom electrode (cyan areas in Figure 4). As a consequence, the estimated maximum photocurrent generated by the perovskite later is 22.0 mA cm⁻² for the Ti-based B III stack compared to 23.0 mA cm⁻² for the Au-based B II stack. Comparison with the experimental values of 19.9 mA cm⁻² for B III and 20.5 mA cm⁻² for B II, provides estimates for the AM1.5G-averaged internal quantum efficiency (IQE) of 90% and 89%, respectively. The analysis shows that the absorption of photons by this Ti limits the photocurrent to some extent. Other cost-effective, but more reflective metals such as Cu are frequently used together with an ITO diffusion barrier in crystalline-Si solar cells and could possibly enhance the maximum attainable photocurrents to the level of a B II stack with a Au bottom electrode.⁹⁻¹¹

### 2.3. Cells on Polymer-Coated Smooth and Rough Steel Substrates

In a final step (B III→C III), perovskite solar cells were fabricated on polymer-coated steel substrates. To assess the impact of surface roughness on the photovoltaic parameters, steel samples with widely different surface roughness were used. We selected a Ni-plated high-gloss steel (R1), Ni-plated battery steel (R2), Cr-plated packaging steel (R3), and Zn-coated construction steel (R4). For future integration of perovskite solar cells in steel buildings, the devices will eventually be deposited directly on the polymer-coated steel itself. A typical coating for such steels consists of a primer layer followed by a topcoat layer both with a thickness range in the order of 10⁻²₀₀ μm. These coatings typically consist of commodity polymers like polyester for the lower end of the product portfolio to polyvinylidene fluoride (PVDF) and polyurethane (PU) for more demanding, higher end, applications. These coated products often come with lifetime guarantees of 15 up to 40 years of corrosion protection and offer excellent resistance to ambient conditions in various environments. However, to avoid any incompatibilities with the processing of the perovskite solar cells, a more inert polyimide-amide (Torlon) coating was chosen as planarization layer to reduce roughness and insulate the substrate from the bottom electrode. Electrical insulation is necessary if multiple cells are interconnected on a substrate and for building integration.
The surface roughness, represented by the maximum profile peak height above the mean line ($R_p$), determined with surface profilometry of the glass/ITO and PAI-coated steel substrates varied between roughly between 200 and 3000 nm (Table 4 and Figure 5). The substrates exhibit very different macroscopic surface textures depending on their surface finishing. While the glass/ITO and R1 substrates show fairly smooth surface textures with only small protrusions, R2 and R3 substrates yield a macroscopic surface texture with a linear orientation. The R4 substrate is very different, and its texture can be described with randomly appearing macroscopic hills and valleys. To evaluate the microscopic surface roughness, AFM measurements were performed (Figure 6). The microscopic root-mean-squared surface roughness ($R_q$) of the glass/ITO substrate is 2.2 nm with a $R_p$ of 10 nm (Table 4). All PAI-coated steel substrates gave consistently lower microscopic $R_q$ values between 1.0 and 1.4 nm (Table 4). The corresponding $R_p$ values were between 7 and 40 nm and are due to the presence of small protrusions visible as white spots in the AFM images. Differences in $R_p$ values between the two profilometry measurements are related to the significantly smaller scanning area ($5 \times 5 \mu m^2$) in AFM compared to the surface profiler ($2 \times 2 mm^2$).

Interestingly, the photovoltaic performance and efficiency of Ti-based coated steel substrate cells (CIII) decreased only slightly with increasing surface roughness (Figure 7a, Table 5) and the reproducibility in the performance and the yield of properly working cells remained virtually unaffected (Figure 8, Table 5). Cells on steel substrates show stable operation under maximum power point tracking for 5 min. as shown in Figure S2, Supporting Information. The best CIII cell on steel gave a PCE of 15.2%, which is only slightly less than the 15.9% for the best BIII device on glass/ITO (Table 5). The stabilized efficiencies of the best CIII cells dropped from 15.2% for R1 to 13.8% for R4 substrate with increasing roughness as measured with the profilometer. The efficiency drop is mainly caused by a decrease in FF which decreased from 0.70 (glass/ITO) and 0.67 (R1) to 0.60 (R4). The $J$–$V$ characteristics (Figure 7a) show that the reduction in FF is due to a combination of an increased series resistance and decreased shunt resistance which both evolve progressively going from R1 to R4. The reduced shunt resistance is expected when the perovskite layer thickness drops locally, while the increased series resistance may originate from local poor or absent electrical contacts between adjacent layers. For R4 substrates, some cells gave S-shaped $J$–$V$ characteristics, which could be related to such a poor contact with an energetic

Table 3. Integrated AM1.5 G photocurrent absorbed or reflected by each layer in opaque substrate-configuration cells with Au (BII) and Ti (BIII) bottom electrodes.

<table>
<thead>
<tr>
<th>Layer</th>
<th>Thickness [nm]</th>
<th>$q\Phi$ [mA cm$^{-2}$]</th>
<th>$q\Phi$ [mA cm$^{-2}$]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Air (reflected)</td>
<td>$\infty$</td>
<td>0.9</td>
<td>0.8</td>
</tr>
<tr>
<td>MgF$_2$</td>
<td>90</td>
<td>1.0</td>
<td>1.0</td>
</tr>
<tr>
<td>ITO top electrode</td>
<td>180</td>
<td>0.2</td>
<td>0.2</td>
</tr>
<tr>
<td>TCTA</td>
<td>10</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Cs$<em>{0.25}$(MA$</em>{0.75}$F$<em>{0.25}$)$</em>{0.97}$Pb$<em>{0.03}$I$</em>{3.07}$</td>
<td>600</td>
<td>23.0</td>
<td>22.0</td>
</tr>
<tr>
<td>PCBA</td>
<td>1</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>SnO$_2$</td>
<td>85</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>ITO</td>
<td>10</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Au/Ti bottom electrode</td>
<td>120</td>
<td>0.1</td>
<td>0.8</td>
</tr>
</tbody>
</table>

$^a$Integration from 300 to 755 nm, for all layers except for the perovskite layer where integration was up to 800 nm.

Figure 4. The AM1.5G photon flux reflected, transmitted, or absorbed by each individual layer in the a) BII and b) BIII substrate-configuration cells with Au and Ti bottom electrodes, respectively.

Table 4. $R_p$ and $R_q$ roughness of the glass/ITO and PAI-coated steel substrates R1 to R4.

<table>
<thead>
<tr>
<th>Substrate type</th>
<th>$R_p$ [nm]</th>
<th>$R_q$ [nm]</th>
<th>$R_q$ [nm]</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>AFM</td>
<td>AFM</td>
<td>Profilometer</td>
</tr>
<tr>
<td>Glass/ITO</td>
<td>2.2</td>
<td>10</td>
<td>260</td>
</tr>
<tr>
<td>Ni-plated high gloss steel (R1)</td>
<td>1.0</td>
<td>40</td>
<td>220</td>
</tr>
<tr>
<td>Ni-plated battery steel (R2)</td>
<td>1.4</td>
<td>10</td>
<td>510</td>
</tr>
<tr>
<td>Cr-plated packaging steel (R3)</td>
<td>1.0</td>
<td>10</td>
<td>1500</td>
</tr>
<tr>
<td>Zn-coated construction steel (R4)</td>
<td>1.3</td>
<td>20</td>
<td>2600</td>
</tr>
</tbody>
</table>
barrier. While the $R_p$ roughness affected the FF it had a negligible impact on the $J_{sc, EQE} \approx 20 \text{ mA cm}^{-2}$ and on the shape of the EQE spectra (Figure 7b). Similarly, the $V_{oc}$ was almost identical ($\approx 1.15 \text{ V}$) for all the substrate variations.

Charge recombination in BIII and CIII cells on glass/ITO and PAI-coated steel substrates was investigated by measuring the $V_{oc}$ as function of photon flux (Figure 7c). The ideality factor determined from the slope of the semilogarithmic plot was estimated to be $n = 1.64–1.73$ on coated steel substrates with a CIII stack, compared to $n = 1.75$ for the best BIII cell on the glass/ITO substrate. With increasing surface roughness the ideality factor increased but at 1 sun light intensity the $V_{oc}$ values are virtually identical. The increase of $n$ with increasing $R_p$ could originate from small shunts that are more pronounced in cells fabricated on rougher substrates.\(^{[23]}\)

It is worth mentioning that the best device on a PAI-coated steel substrate gave a slightly higher PCE than the best Ti-based single-junction cell reported so far in the literature with 15%.\(^{[16]}\) Although the best device with a Ti-bottom electrode gave significantly higher $V_{oc}$ and $J_{sc, EQE}$ values, the FF leaves room for further improvement. This can possibly be achieved by reducing resistive losses in the n–i–p substrate cell by improving interlayer or top contacts.

Figure 5. 3D surface height maps (2 × 2 mm$^2$) of a) glass/ITO and b–e) PAI-coated steel substrates R1–R4. The color bars represent different height scales. a) 360 nm. b) 420 nm. c) 810 nm. d) 3150 nm. e) 5900 nm.

Figure 6. AFM height images (5 × 5 $\mu$m$^2$) measured on a) glass/ITO and b–e) PAI-coated steel substrates R1–R4.
3. Conclusions

In summary, we have developed an optimized n–i–p device stack for substrate-conﬁguration perovskite solar cells on polymer-coated steel substrates and investigated the impact of surface roughness on the J–V characteristics. The optimized stack has an opaque Ti/ITO/SnO2/PCBA bottom contact for electron collection and a TCTA/MoO3/ITO/MgF2 transparent top contact for hole collection. When deposited on a glass substrate, the cell reached 15.9% PCE, which was only slightly limited by absorption of light by the Ti bottom electrode, while other photon losses were mainly due to reﬂection and parasitic absorption by the top ITO contact. The impact of the substrate surface roughness on the photovoltaic performance was investigated by fabricating the same device stack on four different planarization layer coated steel substrates with increasing macroscopic surface roughness. The cells reached stabilized PCEs of 15.2% on a high gloss steel \((R_p/\alpha) = 200\) nm, 14.9% on battery steel \((R_p/\alpha) = 500\) nm, 14.1% on packaging steel \((R_p/\alpha) = 1500\) nm, and 13.8% on a construction steel \((R_p/\alpha) = 2500\) nm. With increasing surface roughness the FF showed a gradual decrease as a result of increasing series resistance and a reduced shunt resistance, while the \(J_{sc}\) and \(V_{oc}\) remained nearly unchanged. Also the yield of working devices and the statistics of the photovoltaic parameters was not really affected by the surface roughness. This demonstrates that developed device stack is fairly robust against the roughness of the substrate.

The reduced FF with increasing surface roughness seems to originate from an increased series resistance and a lower shunt resistance. The latter is not unexpected and can be understood by considering thickness variations and nonconformal or local absence of layer coverage by small protrusions. The increased series resistance could find its origin in localized regions where

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**Table 5.** Stabilized J–V parameters of the optimized substrate-conﬁguration perovskite solar cells with a Ti bottom electrode on glass/ITO \((B^{III})\) and PAI-coated steel \((C^{III},\ R1–R4)\).

<table>
<thead>
<tr>
<th>Substrate</th>
<th>(J_{sc}) ([mA cm^{-2}])</th>
<th>(J_{sc,EQE}) ([mA cm^{-2}])</th>
<th>(V_{oc}) ([V])</th>
<th>FF</th>
<th>PCE ([%])</th>
<th>PCE_{E QE} ([%])</th>
<th>Yield (^{a})</th>
</tr>
</thead>
<tbody>
<tr>
<td>Glass/ITO</td>
<td>19.9</td>
<td>19.9</td>
<td>1.15</td>
<td>0.70</td>
<td>15.9</td>
<td>15.9</td>
<td>8/8</td>
</tr>
<tr>
<td>R1</td>
<td>19.8</td>
<td>19.8</td>
<td>1.16</td>
<td>0.67</td>
<td>15.2</td>
<td>15.2</td>
<td>11/12</td>
</tr>
<tr>
<td>R2</td>
<td>20.3</td>
<td>20.2</td>
<td>1.15</td>
<td>0.64</td>
<td>15.0</td>
<td>14.9</td>
<td>6/8</td>
</tr>
<tr>
<td>R3</td>
<td>20.3</td>
<td>20.1</td>
<td>1.15</td>
<td>0.61</td>
<td>14.2</td>
<td>14.1</td>
<td>7/8</td>
</tr>
<tr>
<td>R4</td>
<td>20.0</td>
<td>20.0</td>
<td>1.15</td>
<td>0.60</td>
<td>13.8</td>
<td>13.8</td>
<td>7/8</td>
</tr>
</tbody>
</table>

\(^{a}\)The number of properly working cells/the total number of cells.

---

**Figure 7.** a) Stabilized J–V characteristics of substrate perovskite solar cells with an ITO interlayer coated opaque Ti bottom electrode on glass/ITO substrate \((B^{III})\) and coated steel substrates \((C^{III},\ R1–R4)\), illuminated with simulated AM1.5G light \((100 mW cm^{-2})\). b) Corresponding EQE spectra. c) Corresponding light intensity dependence of \(V_{oc}\). d) Photograph of the four best \(C^{III}\) devices on a R1 coated steel substrate.
Solutions were stirred at 60 °C overnight before the spin coating, unless stated otherwise. The starting point was a semitransparent n-i-p cell (A stack) with an ITO bottom electrode and a MgF₂-coated ITO transparent top electrode. For the ETL, a commercial Ti-based substrate-configuration solar cell (A stack) with an ITO bottom electrode (R1, HILAN, Tata Steel, R1), Ni-plated battery steel (HILUMIN, Tata Steel, R2), Cr-plated packaging steel (TCCT, Tata Steel, R3), and Zn-coated construction steel (MagiZinc Tata Steel, R4) substrates were cleaned in 2-propanol and blow dried with nitrogen. For all the steel substrates, a wire-bar-coated polyamide-imide (PAI, Torlon Al-10, Solvay) was used as planarization layer. The planarization layer was cured in air at 265 °C for 15 min. and cut to 3 × 3 cm² samples for further use. Prior to the bottom electrode deposition, the samples were sonicated in isopropanol for 15 min and blow dried with N₂. The solar cell fabrication on the PAI-coated R1–R4 steel substrates (stack CIII) was identical to the fabrication of stack BIII. For the Au-based substrate-configuration devices (stack B, CIII), a 120 nm patterned Au bottom electrode was deposited (1 Å s⁻¹) onto the ITO glass substrate via thermal evaporation. For the Ti-based substrate-configuration devices (stack B, CIII), a 200 nm patterned Ti bottom electrode was deposited (2 Å s⁻¹) onto the ITO glass substrate via electron-beam deposition. For all opaque substrate-configuration solar cells, a 10 nm patterned ITO interlayer was deposited (0.3 Å s⁻¹) via radio frequency magnetron sputtering under Ar/O₂ flow.
The SnO₂ dispersion was spin coated onto the ITO interlayer coated metal bottom electrodes on different substrates, at 2800 rpm (with a 2000 rpm s⁻¹ acceleration) for 60 s and heat treated at 150 °C for 30 min in ambient atmosphere. The SnO₂ (85 nm) film was then treated with UV–ozone (10 min.) and immediately transferred into a nitrogen-filled glovebox. To passivate the SnO₂ ETL, the PCBM or PCBA solution was spin coated onto the SnO₂ coated substrate at 2000 rpm (with a 2000 rpm s⁻¹ acceleration) for 30 s to leave a ≈1 nm thin passivation layer. For PCBM, the samples were annealed at 100 °C (30 min.) and cooled to room temperature. For PCBA no annealing was required. The C50.05[MA0.17FA0.83]0.95Pb(I0.13Br0.87)₃ perovskite film (≈600 nm thick) was processed using a ramped spin-coating deposition. The perovskite precursor solution was deposited statically onto the passivated ETL at 1000 rpm (200 rpm s⁻¹) for 10 s, which was followed by a ramp to 6000 rpm (2000 rpm s⁻¹). 10 s prior to the end of the spin-coating program 200 μL chlorobenzene was deposited. Prior depositing the HTL, the sample was annealed in the glovebox at 100 °C for 60 min. and cooled to room temperature. Thermally evaporated TCTA HTLs were deposited (2 Å s⁻¹) onto the perovskite films. The ITO top electrode (180 nm) was deposited (≈0.3 Å s⁻¹) using radio frequency sputtering under Ar/O₂ flow. For the devices with Au bottom electrode, MgF₂ was deposited via thermal evaporation. The active area (0.09 or 0.16 cm²) was determined by the overlap of the ITO, Au or Ti bottom electrode and the transparent ITO top electrode.

Device Characterization: All samples were stored and measured in a nitrogen-filled glovebox without any further exposure to air or any preconditioning, unless stated otherwise. The current–density–voltage (J–V) characteristics were measured by a Keithley 2400 source meter. During the J–V measurements, light from a tungsten-halogen lamp was filtered by a Schott GG385 UV filter and a Hoya L8120 daylight filter to mimic the AM1.5G spectrum (100 mW cm⁻²). For top ITO top electrode side illumination of solar cells, a black shadow mask with an aperture area of 0.0676 or 0.1296 cm² was employed to define the illuminated cell area. During the fast forward sweep measurements, the source meter swept the voltage from 1.5 V to −0.5 V (reverse scan) or from −0.5 V to +1.5 V (forward scan) at a scan rate of 0.25 V s⁻¹. Light soaking preconditioning of the solar cells were performed by exposing the cell area to continuous illumination of simulated AM1.5G (100 mW cm⁻²) light for a given time, followed by a fast sweep measurement. For the stabilized J–V measurement (slow sweep measurements), the Voc of the solar cell was first tracked for 5 min. under constant illumination and then a reverse sweep from Voc + 0.04 V to −0.04 V was performed with a step size of 0.04 V; the acquisition time of the current density at each voltage step was 5 s. External quantum efficiency (EQE) measurements were performed in a nitrogen atmosphere. The probe light was generated by a 50 W tungsten-halogen lamp (Philips Focusline), which was modulated with a mechanical chopper (Stanford Research, SR 540) before passing through a monochromator (Oriel, Cornerstone 130). The spectral response of the device was recorded as a voltage from a pre-amplifier (Stanford Research, SR 570) using a lock-in amplifier (Stanford Research, SR 830), and was calibrated by a reference silicon cell. To accurately determine the short-circuit current density (Jsc), a green LED (530 nm, Thorlabs MS510L3, driven by a DC4104 driver) was utilized as a light bias during the EQE measurement to provide the solar cell with approximately one sun equivalent illumination intensity.

Film Characterization: 3D surface profile maps for the planarization layer coated steel substrate were measured using a Dektak surface profiler (Bruker). AFM surface topologies were measured with a Veeco Dimension 3100 AFM in tapping mode. The root-mean-square roughness (Ra) and maximum profile peak height (Rpk) values were estimated from these measurements.

Optical Simulation: Optical simulations were performed using the transfer matrix method with Setfos 5.0 (Fluxim AG). The wavelength dependent refractive indices for TCTA, sputtered ITO, triple-cation perovskite active layer and the Ti bottom electrode (n) and extinction coefficients (k) are depicted in Figure S3 (Supporting Information). The wavelength dependent n and k for the other layers can be found in our previous work from our group.²⁴

Supporting Information
Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest
The authors declare no conflict of interest.

Data Availability Statement
The data that support the findings of this study are available from the corresponding author upon reasonable request.

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metal-halide perovskites, optical modeling, solar cells, steel substrates

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