Electrode metallization for scaled perovskite/silicon tandem solar cells: challenges and opportunities

Atteq ur Rehman, Emmanuel P. Van Kerschaver, Erkan Aydin, Waseem Raja, Thomas G. Allen and Stefaan De Wolf

KAUST Solar Center (KSC), Physical Sciences and Engineering Division (PSE), King Abdullah University of Science and Technology (KAUST), Thuwal, 23955-6900, Saudi Arabia

Abstract:

Monolithic perovskite/silicon tandem solar cells have the potential to reach very high power conversion (PCE) efficiencies in a cost-effective manner. In the last decade, significant technological advancements have been made for lab-scale devices (~1 cm²), with PCEs now higher than the theoretical PCE limit of single-junction silicon solar cells. For market entry of such tandems, the involved processing steps need to be scaled to industrial wafer dimensions, typically >244 cm², which also mandates the development of adequate electrode-metallization strategies. Here we discuss challenges and opportunities related to this latter point, including the required properties of the front metal grid for perovskite/silicon tandems, as well as key motivations and challenges in adopting screen-printed metallization, which is the current standard for mainstream silicon solar cells. We give a cost estimation for the front metal grid by considering the cost of low-temperature metal pastes that are compatible with the thermal budget limitations imposed by the perovskite top cell. We also consider opportunities to employ alternative metallization schemes that arise from the reduction in current density in tandem solar cells compared to single-junction devices. Lastly, we discuss possible routes to replace or minimize the silver content in costly silver-based metallization for industrial applications.

Keywords: Perovskite/silicon tandems, Photovoltaics, Metallization, Metal contacts, Screen-printing, Cost analysis
1. INTRODUCTION

The photovoltaics (PV) market is dominated by crystalline silicon-based (c-Si) technologies, owing to the low fabrication cost, excellent performance and high reliability offered by their materials and manufacturing processes [1]. In recent years, the cost of PV cells has continued to decrease significantly, making the balance-of-system (non-module) and the installation costs now the major contributors to the price of PV energy installed in both residential and utility settings [2]. In this scenario, improving the PCE of PV panels – combined with mitigating parasitic module heating – is an increasingly important and promising technical direction to further drive down the levelized cost of electricity (LCOE) from PV [3, 4].

Currently, the most efficient commercial c-Si PV module PCEs are about 20–22% [5], whereas the certified record module PCE is 24.1% [6]. At the cell level (before packaging), the current record PCE is 26.7% [7, 8]. However, with the fundamental limit PCE of c-Si at 29.5%, further improvements in cell performance are increasingly challenging [9]. Minimizing the thermalization losses by stacking multiple absorbers with appropriate band gaps in a tandem cell configuration extends the theoretical limit beyond 40% [10, 11]. Here, the key challenge is the successful integration of the sub-cells at low cost. In this regards, polycrystalline metal halide-based perovskite solar cells have received significant attention for their potential application as a top cell in tandems, paired with conventional c-Si PV technologies, due to their high-efficiency potential, tunable bandgap, and promise for cost-effective fabrication methods [12-15]. Owing to such appealing properties, perovskite/Si tandems have been extensively studied in recent years [16-22], and remarkable progress has been reported with record lab-scale PCEs >29% (monolithic) [23] and >28% (four-terminal) [24].

Currently, the metallization techniques used in lab-scale tandem devices, such as thermal evaporation, are slow and expensive compared to the ubiquitous screen-printed metallization used in large-volume manufacturing of c-Si solar cells. Considering that PV modules soon will be manufactured at the terawatt scale, and given the necessity for constant cost reductions [25], adapting the existing cost-efficient metallization technologies to perovskite/Si tandem solar cells and searching for alternative cost-efficient methods and materials is of high urgency. Scaled perovskite/Si tandem devices require a well optimized transparent electrode (usually transparent conductive oxides, TCOs), as additional electrical and optical losses are introduced with scale up [26]. More importantly, an electrically and optically tuned metal grid is needed for efficient
extraction of the current to an external circuit (or interconnection to another cell in a module). We note here that in a tandem device, compared to a single-junction silicon solar cell, the short-circuit current density ($J_{SC}$) is halved, whereas the open-circuit voltage ($V_{OC}$) is more than doubled. Therefore, for a given fill factor ($FF$), equivalent current losses will translate in a $PCE$ loss more than twice in a tandem than in a single-junction device (as $PCE = J_{SC} \times V_{OC} \times FF$), underlining the importance of optical device and materials design for tandems.

Adopting screen-printed contacts on perovskite/Si tandems necessitates curing and sintering the metal paste (processes needed to achieve the desired conductivity of the metal grid) within the low temperature window imposed by the thermal resilience of the perovskite top cell. However, the device structure of monolithic perovskite/Si tandems has the advantage to tolerate a higher line resistance (relative to single-junction Si cells) in the grid metallization, due to their halved $J_{SC}$, compared to single-junction devices. Moreover, the transition to larger areas also requires an optimum grid design to balance all the losses (TCO, metal line/contact resistance and grid shading) at the sun-facing side of the perovskite/Si tandem cell. In this article, we give a detailed perspective regarding scale-up of the front metallization of perovskite/Si tandems by optimizing the metal grid design, the feasibility to adopt the existing screen-printed technique and alternative routes for cost effective metal contacts for industrial applications.

2. State-of-the-art c-Si, perovskite and perovskite/Si tandem cells and modules

The potential of perovskite/Si tandems was demonstrated in recent years by several research groups with $PCE$s now close to 30%. However, since the main research focus has been the demonstration of high efficiencies, most of the reported lab-scale single-junction and tandem cells utilizing perovskites are still fabricated over very small device areas (typically 0.1-1.5 cm$^2$). For their large-scale counterparts, only a limited number of reports are available. Fig. 1 summarizes published results for Si and perovskite (single junction and tandems) solar cells and modules, with limited data points for perovskite-based devices in the upper right-hand quadrant (large area/high efficiency). To date, these small-area devices have only low rated power values, as can be observed in the lower left quadrant of Fig. 2. An important step to potential commercialization will be scaling the technology from the rated power values reported (2.8mWp) to values comparable to commercial products (>400Wp at the module level). For monolithic 2-terminal perovskite/Si tandem cells, one path towards commercialization is forecasted where the perovskite (top) cell
would be manufactured as an add-on to high-efficiency c-Si cells manufactured with minimal process changes in the existing industrial production lines for the bottom cell [27]. Most c-Si cells originating from these modern production lines currently have an area of (at least) 244 cm$^2$, and nowadays the industry is moving to larger wafer sizes, implying that all top cell processing will have to be compatible with a similar area.

![Graph showing PCE vs. Solar Cell or Module Area (cm$^2$)](image)

**Fig 1.** PCE’s of lab-scale, large area cells and modules for single junction Si solar cells [23] and modules [28-33], perovskite [34-37] and perovskite/Si (monolithic & 4T) tandems [18, 19, 23, 24, 38-49].
In general, PV modules based on multi-junction solar cells will only be preferable to single-junction products if their LCOE is lower. Several of the processing steps for manufacturing highly efficient, lab-scale perovskite/Si tandems reported in the literature thus far, especially the reported metallization processes, are unlikely to be scalable to production volumes at an acceptable cost. Several groups have reported efforts for upscaling the deposition of perovskite layers, either in single-junction module or in perovskite-based tandems, with various techniques including vacuum-based evaporation, blade/slot-die coating, and spray coating [18, 21, 50]. However, the transition to larger cell areas is still challenging, as point defects in the perovskite film cause significant losses in the form of leakage current and shorting between the electrodes, reducing the FF and Voc [51]. Another important factor is the presence of highly mobile ions or volatile components in perovskite layers that can migrate through the transparent conductive oxide (TCO).
layer and even in the metallic electrodes causing these layers to degrade. There have been many reports of significant conductivity loss in the metallic contacts due to their reaction with iodine anions from the perovskite [52, 53]. Indeed, there is still much work to be done to improve the stability of perovskite layer to prevent the degradation of the metallic contacts.

3. Transparent conductive oxides for tandem solar cells

Transparent electrodes are a vital component of state-of-the-art perovskite/Si tandem solar cells [54]. For silicon heterojunction (SHJ) bottom cell-based tandems, the most successful silicon bottom cell so far, 4T perovskite/silicon tandems typically require more (and more conductive) transparent conductive oxide (TCO) layers, than the monolithic 2T concept; the higher the number of TCOs, the higher the probability for parasitic absorption losses. [55] In the future, with the adoption of new bottom cell technologies such as poly-silicon passivating contact devices, the number of TCO layers might be reduced further. Also, utilizing nanocrystalline silicon-based, or metal oxide-based RJs [56] can eliminate the use of TCOs at the recombination junction [11, 18]. Therefore, rather than focusing on the recombination junction and rear reflector TCOs, here we focus on the front TCO electrode, which is significant due to its direct influence on the current generation of both sub cells. Here, we again emphasize the point that optical losses in tandem solar cells incur a more severe performance loss compared to single-junction devices due to the high operating voltage of tandems (typically 1.7-1.9 V).

Ideally, the front TCOs should have (i) broadband transparency (enabled by a wide bandgap) with low parasitic absorption (enabled by a high carrier mobility, allowing a reduction in free carrier density), (ii) high lateral conductivity to transport the collected charge carriers with minimal resistive losses, and (iii) low contact resistivity with the overlying metal contacts and underlying ETL or HTL. Several TCOs have been applied successfully to solar cells such as tin doped indium oxide (ITO), H-doped indium oxide (In$_2$O$_3$:H), [57] Zr-doped In$_2$O$_3$ (IZRO), [58] W-doped In$_2$O$_3$ (IWO), [59] and indium zinc oxide (IZO). Among these examples, IZO is currently preferred in perovskite/Si tandem solar cells due its high conductivity and high near-infrared transparency with room-temperature processing, without the need for post-deposition annealing. The higher annealing temperature (>160°C) of typical In$_2$O$_3$-based TCOs, which is needed to ensure the crystallinity and overall improved mobility and transparency, is undesirable for metal-halide perovskite layers due to the deprotonation of the system’s A-site organic cation (mostly MA$^+$) via evaporation. On the other hand, the main drawback of IZO is its strong blue absorption
within the ~250-440 nm range due to its blurry band edge owing to its amorphous nature (Fig 3a). Replacing amorphous IZO with crystalline TCOs (as shown in SEM top-view images in Fig. 3b-e) may provide an increase in the current density of up to ~2 mA/cm². Lastly, the low contact resistivity of TCOs with metal contacts is quite critical to achieve high FF values.

So far, all lab-scale tandems have been utilizing thermally evaporated silver contacts. However, considering the scaling-up of such devices, the implementation of fast, cost-efficient electrodes is a critical step for commercialization. Here, as discussed in Section 4, screen-printed contacts, as a heritage of c-Si technology, seems the most suitable technique for large area, high throughput manufacturing. However, the widespread applications of these contacts is still to be adopted for tandem devices; very few cases have been reported so far in literature.[60] The potential challenges of incorporating screen-printed contacts with the top TCO might be a high contact resistivity due to the restricted annealing temperature of the tandem devices (usually below 150 °C), which will be discussed in detail in Section 4.4. In the future, MA⁺-free or fully inorganic perovskites may overcome the limitation to utilize alternative crystalline TCOs, since these perovskite variants allow annealing at higher temperatures (>150 °C). Although, MA⁺ free perovskite/Si tandems have been shown with PCEs >25%, for fully inorganic tandems, still more efforts are needed.[61]

Fig. 3. Absorptance data of the some commonly used TCOs for perovskite/silicon tandem solar cells, with our in-house data. The EQE graph was taken from [11] and represents the highest J_sc tandems in the literature with IZO front electrode. In the given EQE example, the used TCO was IZO. SEM top-view images of the b, IZO [62] c, ITO,[63] d, IO:H,[64] and, e, IZRO[58] thin films
reported in literature. SEM figures reprinted with permission from References.

4. Front metallization for large area perovskite/Si tandems

4.1 Background

The dual-cell design of 2T tandem devices restricts the photocurrent to a value of about ~20 mA/cm², which is approximately half that of a typical single-junction c-Si solar cell (~40 mA/cm²). However, the voltage across the tandem devices is more than double to that of a typical single junction cell. Let’s consider the current-voltage [I(V)] characteristics derived from the one-diode model of a solar cell [65].

\[
I = I_{ph} - I_0 \left( \exp \left( \frac{V + IR_{s,tot}}{nV_t} \right) - 1 \right) - \frac{V + IR_{s,tot}}{R_{sh,tot}} \tag{1}
\]

Here, \( R_{s,tot} = R_s + R_{s,grid} \) is the series resistance, and \( R_{sh,tot} \) is the shunt resistance of the diode. The total current \( I \) is a superposition of the photogenerated \( (I_{ph}) \) current, the diode current and the shunt leakage current. Other parameters in the equation are the thermal voltage \( (V_t) \), and diode ideality factor \( (n) \). The equation depicts the impact of series and shunt resistances on the characteristics of the diode. \( R_{s,grid} \), representing the effective resistance of the front electrode, is an important characteristic for metal grid electrodes and is composed of discrete resistive components such as busbar resistance \( (R_b) \), finger resistance \( (R_f) \), contact resistance between metal/conductive electrode, and the lateral resistance of the top TCO. The total series resistance contribution of the top contact layers of monolithic perovskite/Si tandem cells can be obtained by adding all the resistances (series connection) in Fig. 4.
Fig 4. Cross-sectional diagram of monolithic perovskite/Si tandem solar cell showing different series resistance contributors. The $\rho_c(\text{front})$ is an important parameter for designing the front metallization of the cell and defined by the metal/TCO and TCO/perovskite interfaces.

For many of the small cells (<2 cm$^2$) a metal grid is largely cosmetic, mainly used to simplify probing and is usually located outside of the active area of the device. As such, PCEs over 27% have been reported with a 2-3$\mu$m thin layer of silver evaporated around the active area of the tandem device [41]. As the area of the devices increases, the conductivity of the front TCO layer is no longer sufficiently low to carry the generated current to the edges without significant resistive losses and a more conductive metal grid will be required. For some of the early scaling efforts, the use of thermal evaporation of the metal grid through a shadow mask has been employed (see Table 1).

Thermal evaporation of metal grids is conveniently used for small-area tandems, as the technique is already used for several other layers in the top cell construction and has a minimal thermal impact on the device during processing. The line resistivity of the evaporated grid is close to the intrinsic bulk resistivity of the metal used, and a line-width definition down to 10$\mu$m is achievable. However, this approach of front metal contact formation is not a practical choice to be applied to large-area tandems intended for industrial production. One reason is the low material utilization of evaporated metal grids, with more than 97% of the evaporated materials covered on shadow mask or the vacuum chamber walls. Besides, evaporation is generally a thin-film deposition technique and a very slow process (several Angstrom/s) performed at low pressure which makes the deposition of thick metal layers impractical and thus results in a relatively high line resistance.
relative to the line width. For industrially manufactureable large area devices, a metallization technique with better line resistivity values, lower material costs and high throughput is required.

**Table 1:** Annual progress of monolithic perovskite/Si tandems utilizing evaporated contacts with an active area \( \geq 1 \text{ cm}^2 \). PCE along with \( V_{OC}, J_{SC}, \) and \( FF \) of the devices are stated.

<table>
<thead>
<tr>
<th>Year</th>
<th>Metal-thickness (nm)</th>
<th>Area (cm(^2))</th>
<th>( J_{SC} ) (mA/cm(^2))</th>
<th>( V_{OC} ) (V)</th>
<th>( FF ) (%)</th>
<th>( PCE ) (%)</th>
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<td>23.6</td>
<td>[17]</td>
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4.2 **Grid design**

On the research level, monolithic tandem devices are in general processed on substrates larger than the device-active area which is usually defined by a shadow mask at the time of depositing the top transparent electrode and metal grid, and an aperture employed at the time of measurement (Fig. 5a). Because of the limited dimensions of these cells \( \text{W} \times \text{L} < 2 \text{cm}^2 \), and the
relatively low resistivity of the TCO, there is no need for any metal to be deposited in the active area. The photo-generated current in the light-exposed areas \( (J_{mpp}) \) will flow laterally to the collecting busbars and be transported to the probing pad(s). For such a structure the power loss linked to the current flowing in the TCO relative to the power generated by the device can be expressed as:

\[
P_{loss, TCO} = \frac{1}{12} \cdot \rho_{TCO} \cdot \frac{J_{mpp}}{V_{mpp}} \cdot W_D^2
\]  

(2)

For a TCO sheet resistance \( (\rho_{TCO}) \) of 50\( \Omega/\square \), this value remains below 0.5\% up to a device width (WD) of 0.5 cm. The loss however, increases quadratically with the device dimension and already approaches 2\% when doubling the width to 1 cm.

As such, as the area of the devices is scaled up, the addition of metal “fingers” assisting in a low-loss transfer of collected carriers to the busbars is required. For relatively small devices that can be a pattern inspired by concentrator cells but for relevant (non-concentrator) commercial cell sizes, an H-type pattern, as employed for traditional c-Si cells, will have to be adopted. The H-pattern design (illustrated in Fig. 5b) is composed of a grid connecting multiple finger lines and perpendicular busbars to extract the current from the TCO. This, however, introduces additional shadowing loss at the active area of the solar cell. Wilkinson et. al. presented the design rules to calculate the optimum grid parameters for high efficiency on scaled single junction perovskite solar cells [73]. In their simulated cell designs — a cell with no metal grid, a cell with bus bars only and a cell with a metal grid comprising of bus bars and fingers — a higher PCE (up to 25\%) on an area >200 cm\(^2\) was demonstrated from cell with grid consisting of bus bars and fingers. This demonstrates that an optimum metal grid covering the TCO is important in increasing the PCE of large-area cells and modules. Fig. 6 shows a range of metal grids adopted for small-area (\( \leq 1\)cm\(^2\)) and large-area devices, as reported in literature. The metal grid designs evolved with larger cell areas in a way to optimally cover the front TCO of the device.
Fig 5. Illustrations of the current flow transport at the front electrode of small-area devices (left). Here current is transported towards the metal busbars at the edges and the active area is exposed during the measurements. However, large-area devices require metal grids covering all the active area for efficient current extraction. Traditional H-pattern metal grids (right) are used to contact such large area devices. In this case the metal (busbars and fingers) covering the active area introduces an additional loss in form of shadowing the active area of the solar cell.
Fig 6. Front metal grids adopted for monolithic-perovskite/Si tandems. In case of small area (a) KAUST used a rectangular design with an active area of 0.83 cm$^2$ [11], (b) HZB adopted a square design for their record cell efficiency over an area of 1.059 cm$^2$ [72]. For large area devices (c) UNSW used multiple fingers covered by a busbar over 16 cm$^2$ area [46]. (d) The efficiency of UNSW’s 16 cm$^2$ cell was further improved by modifying the front metal grid increasing the FF from 71 to 78% [45] (e) EPFL metal design for a 12.96 cm$^2$ device [47], and (f) the large area screen printed metal grid (57.6 cm$^2$) of EPFL [49].

In H-type patterns, the photo generated current flows through the top TCO towards the metal fingers that transport the current to wider busbars that are used as external contacts to the device in module fabrication. The resistive losses to be considered relative to the front grid design are now the resistive power loss through the TCO ($P_{\text{loss, TCO}}$), similar to the small-area devices but now using the finger spacing ($d_f$) as a parameter; the resistive loss linked to the current through the fingers ($P_{\text{loss, grid}}$), which is quadratically dependent on the busbar spacing ($d_{BB}$); and finally, the power loss related to the contact resistance between the TCO and the grid fingers $P_{\text{loss, contact}}$. 
The loss in the busbars is typically neglected, since in module fabrication highly conductive Cu-based tabs are attached for the series/parallel connection of the individual devices. The different components of the power loss can be expressed as:

\[ P_{\text{loss,TCO}} = \frac{1}{12} \cdot \rho_{\text{TCO}} \cdot \frac{I_{\text{mpp}}}{V_{\text{mpp}}} \cdot d_f^2 \]  

(3)

\[ P_{\text{loss,grid}} = \frac{1}{12} \cdot \rho_{\text{finger}} \cdot \frac{I_{\text{mpp}}}{V_{\text{mpp}}} \cdot d_{BB}^2 \cdot d_f \]  

(4)

\[ P_{\text{loss,contact}} = \frac{1}{2} \cdot \rho_{\text{contact}} \cdot \frac{I_{\text{mpp}}}{V_{\text{mpp}}} \cdot d_{BB} \cdot \frac{d_f}{W_f} \]  

(5)

All increase super linearly with increasing spacing of the fingers and busbars inviting to keep those as small as possible. However, in the areas covered with the metal grid, the incoming light will be blocked and, at least in the first order, these shaded areas can be regarded as lost for power generation. The relative power loss related to the shading of the finger grid can be expressed as:

\[ P_{\text{shading,fingers}} = \frac{W_f}{d_f} \]  

(6)

and for the busbars as:

\[ P_{\text{shading,busbars}} = \frac{W_{BB}}{d_{BB}} \]  

(7)
Fig 7: (a) Optimization for the total power loss ($P_{\text{loss}}$) of metallization as a function of $\rho_{\text{TCO}}$ and $d_f$ for perovskite/Si tandem solar cells. Here the $J_{\text{mpp}}$ and $V_{\text{mpp}}$ values are considered from (a) state of the art tandem reported in Ref [74] and (b) ideal tandem cell. (c) The comparison of the total power loss for (a) and (b) at sheet resistivity value of 50 $\Omega/\square$.

Narrow and widely spaced fingers and busbars are obviously preferred with respect to shading, even though they do have to be balanced with the intrinsic increases in resistive power losses. To minimize the power losses in a solar cell the finger spacing needs to be optimized for the finger widths. Fig. 7 (a, b) shows the modelling of power losses at the front for the state-of-the-art high efficiency (in-house electrical data) and ideal perovskite/Si tandem solar cells. Here 800 $\mu$m busbars along with 50 $\mu$m finger widths were considered. Since our focus is only metallization at the front, the optical losses caused by parasitic absorption are ignored, and a fixed sheet resistivity of (50 $\Omega/\square$) is considered. The minimum power loss can be observed for finger spacing in the range of 3 – 3.5 mm (Fig. 7c). This loss is much lower than that for state-of-the-art high efficiency SHJ solar cells. This is because of lower shading losses resulting from wider finger spacing, a consequence of the reduced current density in the tandem cell design. Additionally, since
the current in the perovskite/Si tandem is halved (compared to a Si cell), the overall power loss from resistance goes down by a factor of 4 \((P=I^2R)\). This opens an opportunity to consider using relatively resistive materials as an alternative to silver metallization. **Fig. 8** shows the percentage power losses with variation in finger resistivity for perovskite/Si tandem solar cell. The losses are lower than in single-junction Si cells, even for resistive materials such as Al or thin ink-jet printed silver, which are beneficial in terms of cost and material utilization. Section 3.5 and 3.6 discuss these cost benefits in detail. Finger width and grid resistivity will largely depend on the capabilities of the metallization technology selected and the processing window imposed by the underlying cells, in particular the limitations imposed by the perovskite top cell.

![Fig 8. Power loss analysis of monolithic-perovskite/Si tandem with 50 \(\Omega/\square\) front TCO layer. The finger resistivity vs. finger spacing suggests that relatively resistive materials can be used as a front metal contact as perovskite/Si tandems offers lower power losses in comparison to single-junction Si solar cells. The values for the line resistances (40 – 50 \(\mu m\) wides finger lines) are calculated from the bulk resistivity of the material/paste and references [75-79].](image)

**4.3 Perspectives of screen-printed front contacts for tandem devices**

To understand how to translate screen-printed contacts to perovskite/silicon tandem solar cells, we first need to recall the key processing steps of the screen-printing process. In this
technique, a paste filled with small Ag-particles (among other components, such organic binders) is pushed by a squeegee over a fine mesh covered by a thin emulsion layer (Fig. 9a). The mesh is characterised by its mesh number (number of wires per inch), wire diameter, mesh thickness, and emulsion-over-mesh (EOM) thickness as illustrated in Fig. 9(b). The pattern to be printed is defined by creating the required openings in the emulsion, which mostly determines the width of the printed electrode. Additionally, the shape of the electrode is also influenced by the printing parameters. The most important are print speed, squeegee pressure and snap-off distance (distance between screen and substrate). An optimized squeegee pressure is required as a too high pressure will decrease the height of the printed electrode, while at too low pressure, the squeegee cannot push the paste through the screen opening. Moreover, the optimized snap-off distance helps to perform a uniform printing process with continuous finger lines over the solar cell. A final curing step is performed to drive out the solvents and binders in the paste to achieve the desired bulk resistivity of the printed electrodes.

Fig 9. Schematic of screen-printing process to realize the metallization grid and (b) cross-section of mesh and screen opening.
4.4 Limitation of screen-printed metallization for tandem solar cells

The replacement of evaporated contacts with screen-printed metallization for perovskite/Si tandems is not an easy task and doing so will affect the $FF$, $V_{OC}$, and $J_{SC}$, even for small-area devices. Kamino et al. observed a clear drop in $FF$ and $V_{OC}$ of the devices, mainly attributed to a higher contact resistivity ($60 \text{ m}\Omega \text{.cm}^2$) between the printed contacts and ITO, which is several orders of magnitude higher than the contact resistance between ITO and evaporated silver ($\sim 0.02 \text{ m}\Omega \text{.cm}^2$) when both are annealed at 130 °C. [49]. In addition, a decrease in $J_{SC}$ of the devices was also observed which is due to the increased shading losses for the screen-printed (3%) compared to an evaporated (1.25%) grid. This clearly illustrates the issues with scaling up the screen-printed metallization for perovskite/Si tandems. The parameters of the screen-printing technology should meet the criteria listed in Table 2 in order to meet the challenges to adopt this technology on large-area devices. The busbars and finger widths here are of prime importance, as they introduce shading of light that tends to reduce the $J_{SC}$ of the device. There are limits to reduce the metal coverage in screen printed metallization, as the thinner busbars and finger widths increase the series resistance, thereby adversely affecting the $FF$ and hence the $PCE$.

As mentioned above, an additional challenge for printed silver metal grids for perovskite/Si tandems is the limited temperature the perovskite can withstand, mostly below $<150 \degree C$ [17]. Higher curing temperatures of the printed pastes give superior paste compaction, which results in lower line- and bulk-resistivity [80, 81]. Curing of the silver pastes at a lower temperature leads to an elevated bulk resistivity of the metal and hence an increased series resistance of the device. In SHJ devices, where the maximum processing temperature is limited to about 200°C, the bulk resistivity is about 3-6 times higher compared to that of the silver pastes cured at higher temperature ($\sim 800\degree C$) [82]. Obtaining a higher conductivity for metal grid lines at lower temperature requires the developments of a paste that can be cured at temperature $<150 \degree C$. The compatibility of such pastes with TCO’s in terms of contact resistance needs to be investigated to incorporate such metallization schemes.
Table 2: Expectations of the geometrical and electrical parameters for 5-BB front side structure with screen-printed (SP) perovskite/Si tandems (area: 156 x 156 cm²).

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Unit</th>
<th>Screen printed front contacts</th>
</tr>
</thead>
<tbody>
<tr>
<td>Busbar width ((w_b))</td>
<td>µm</td>
<td>500 – 800</td>
</tr>
<tr>
<td>Finger width ((w_f))</td>
<td>µm</td>
<td>30 – 50</td>
</tr>
<tr>
<td>Average finger height ((h_f))</td>
<td>µm</td>
<td>10 – 15</td>
</tr>
<tr>
<td>Aspect ratio (height to width)</td>
<td>-</td>
<td>0.2 – 0.8</td>
</tr>
<tr>
<td>Finger spacing ((S))</td>
<td>mm</td>
<td>3 – 3.5</td>
</tr>
<tr>
<td>Metal coverage (finger + bus bar)</td>
<td>%</td>
<td>4 – 5</td>
</tr>
<tr>
<td>Finger resistivity ((\rho_f))</td>
<td>Ω/cm</td>
<td>2 - 3</td>
</tr>
<tr>
<td>Contact resistivity ((\rho_c))</td>
<td>mΩ·cm²</td>
<td>≤ 1</td>
</tr>
<tr>
<td>TCO sheet resistance (R_{sh})</td>
<td>Ω/sq</td>
<td>~ 50</td>
</tr>
<tr>
<td>Temperature of contact formation ((T))</td>
<td>ºC</td>
<td>Low (&lt;150)</td>
</tr>
</tbody>
</table>

4.5 Cost considerations

Silver prices are quite volatile, as prices ranged from 500 USD/kg to 1800 USD/kg over the last 10 years, with a current (as of September 2021) price of about 840 USD/kg (see Fig.10) [83, 84]. This silver-price fluctuation directly affects the cost of metallization, as it increased from ~13% to ~60% of the cell fabrication cost (excluding the c-Si wafer cost) between 2019 and 2020 [85, 86]. Current silver paste costs are conservatively estimated at 1300 USD/kg and 1700 USD/kg for high-temperature and low-temperature pastes, respectively. That is why the amount of silver used per cell or Watt peak (g/cell or g/Wp) is an important factor and the metallization price, specifically for SHJ cells that uses low temperature pastes, which is therefore sensitive to deviations in paste prices [87]. However, lower metallization costs are expected in perovskite/Si tandems as these cells will require a comparatively lower amount of paste since a wider finger spacing is possible (See Fig. 7). For instance, a 5BB H-pattern grid on a 244 cm² cell requires ~91 finger lines (typically 1.7 mm spacing) on SHJ solar cells, which can be decreased to as low as 47 finger lines (3.25 mm spacing) on perovskite/Si tandems (assuming for the same line and contact resistance values). This allows for a big step in reducing the silver consumption per wafer and can reduce the cost per Wp. To compare the metallization cost of perovskite/Si tandems with SHJ
devices, let us assume that the line and contact resistances of the printed finger lines annealed at lower temperature (≤ 150 °C) remain the same as the paste cured at 200°C (typical annealing condition for SHJ). Roughly 100 mg of silver paste is required for printing a grid with a 5BB design on standard-sized SHJ devices (6 inch × 6 inch) [85]. Assuming the current estimated price of the paste (1700 USD/kg), the material cost amounts to 0.17 USD/wafer only for the front side metallization of a SHJ solar cell. In contrast, for a tandem device, the paste requirement is 40% less (60 mg of paste) and hence will cost 0.102 USD/wafer. This decrease in material cost per wafer is even lower than that of the high temperature pastes that is estimated to be 0.12 USD/wafer by assuming its current price from Fig. 10. If we consider the price in USD/Wp, then the front metallization cost for a 6 inch 24%-efficient SHJ PV module will be approximately 0.029 USD/Wp, while for a 30% perovskite/Si tandem, a realistic anticipated efficiency value in production, about the cost is calculated to be only 0.013 USD/Wp.
The metallization cost may be further reduced by using cheaper materials as the low current values negate the need for highly conductive fingers. In Fig. 11, we show the impact of an increased finger resistivity (relative to Ag) on the metallisation related power loss. To compensate for this additional loss and end up with an equal or overall lower $/Wp value of the cells, the higher resistance has to be offset by a lower cost. The graph on the left-hand side shows the required cost reduction compared to silver to attain a break-even in the overall cell cost.
Fig 11. (a) Power losses for perovskite/Si tandems with an increase in line resistance. (b) Relative cost to silver paste to maintain the cell cost as the finger line resistance increases for lower cost materials. Line resistance value is relative to the silver finger's resistance.

Historically, the amount of silver used per cell (and \( W_p \)) has been decreasing year by year, and is expected to decrease further in the coming years [85]. Even though the silver used per cell is inhibited by the minimal conductivity requirements [88], it is predicted to decrease further from 80 mg to 40 mg by 2025 [85]. The effect of this reduction in silver use per cell would be substantial as perovskite/Si already requires considerably lower amount (40% less) of silver paste per cell.

4.6 Alternative Metallization Routes

Further reduction in metallization cost can be achieved by either substituting silver with a cheaper material or adopting techniques that can more efficiently utilize the metal usage per cell. Copper (Cu) is a metal that can be used as a low-cost substitute for Ag, and has shown a great potential to fabricate a high-efficiency, “silver-free” c-Si solar cells [89]. Electro-plated Cu lines exhibit a bulk resistivity as low as \( 2 \times 10^{-6} \, \Omega \cdot \text{cm} \), which is almost equal to the resistivity offered by pure bulk copper \( (1.7 \times 10^{-6} \Omega \cdot \text{cm}) \) [90]. One good way to adopt Cu metals would be the use of low-temperature curable Cu pastes. Most Cu pastes are inherently “low temperature” owing to the fact that copper easily oxidises at high temperature. Recent results show that contact curing can be performed with minimal oxidation of copper in an inert gas or low-oxygen atmosphere [91, 92] or even in normal atmospheres by means of specially developed Cu paste [77]. Moreover, pastes composed of silver-coated copper particles as a replacement to silver paste can also be used to
reduce the cost of the metallization. The silver coating helps to prevent the copper particles from oxidation and comparable results (0.4% cell efficiency difference) with pure silver paste have been achieved [78]. Moreover, these silver-coated copper pastes have 30% lower silver content, potentially translating to substantial cost reductions [79]. The cost of a pure Cu metal front contact on a perovskite/Si tandem device with 30% PCE is estimated as low as 0.000127 USD/Wp which is about 100 times lower than that of silver implying possible cost savings for Cu-based metallization schemes [83, 84].

Another potential metallization approach can be an alternative deposition technique that more efficiently utilizes the metal to reduce the total amount of metal per cell. For instance, the finger height variation of typical screen-printed contacts is relatively high (~5µm) [93], which is significantly higher than the theoretical requirement and thus consumes a larger amount of metal that does not contribute to the overall finger conductivity. Alternatively, electroplated contact formation is one extensively studied technique as a replacement for screen-printed metallization and can efficiently utilize the materials as area-selective deposition can be done [89] Metal line widths of 20 µm and a line resistance as low as 1 Ω/cm can be realized with such selective deposition techniques [90]. However, the process involves the immersion (or floating) of the cell in a wet chemical solution, rendering the integration of such a method into perovskite/Si tandem cell fabrication may be quite challenging, due to the solubility of the perovskite device stack [94]. Inkjet printing is another promising technique that combines high material utilization, cost-effectiveness, and flexibility. In this process, a chemical ink consisting metal acetate reacts during the metallization process to form the metal grid. The method is an attractive approach as bulk-like resistivity can be realized at a significantly lower temperature[95]. In addition, inkjet printing can precisely control the amount of materials deposited, and the thickness of the contact, which results in extremely low metal consumption (<10 mg of Ag/cell). A mini module (6-inch HJT cells) with an inkjet-printed finger height of 3 µm has demonstrated an efficiency of 19.7%, a similar performance as reference screen-printing based mini-modules. These cells have finger widths of 50µm and consumed 6 and 40 mg of silver for inkjet and screen-printed metallization silver for inkjet and screen-printed metallization, respectively. Moreover, ink-jet printing can be very useful to metallize the perovskite-based PV devices as low-temperature (curable at 80 °C), reactive silver inks (RSI) can be realized with low bulk resistivity values, comparable to that of commercially available screen-printed paste [96]. The silver used per cell can be further reduced to a value as
low as $0.3 \pm 0.1$ mg of silver nanoparticles for a single side of a 6-inch bifacial cell with a conversion efficiency of 23.7% by a novel Flex-Trail-printing technique, as introduced by Fraunhofer ISE [79]. This enables an opportunity to print thin finger widths ($16 \pm 1 \mu m$) with such Flex-Trail printing technique utilizing a nanoparticle silver ink, resulting in very low silver consumption.

The advantages and disadvantages of available metallization techniques that could be adopted to perovskite/Si tandems are summarised in Fig. 12. Evaporated and electroplated contacts give excellent contact resistivity values but come with the proven difficulty to be adopted on an industrial scale. Screen printing is a good choice; however, due to the efficient metal utilization inkjet printing may be preferable.

<table>
<thead>
<tr>
<th></th>
<th>Evaporated</th>
<th>Electroplating</th>
<th>Inkjet printing</th>
<th>Screen printing</th>
</tr>
</thead>
<tbody>
<tr>
<td>Contact resistivity</td>
<td>★★★★★★</td>
<td>★★★★★★</td>
<td>★★★★★</td>
<td>★★★★★</td>
</tr>
<tr>
<td>Conductivity</td>
<td>★★★★★★</td>
<td>★★★★★★</td>
<td>★★★★★</td>
<td>★★★★★</td>
</tr>
<tr>
<td>Adaptability</td>
<td>★★★★★★</td>
<td>★★★★★★</td>
<td>★★★★★</td>
<td>★★★★★</td>
</tr>
<tr>
<td>Ease of processing</td>
<td>★★★★★★</td>
<td>★★★★★★</td>
<td>★★★★★</td>
<td>★★★★★</td>
</tr>
<tr>
<td>Throughput</td>
<td>★★★★★★</td>
<td>★★★★★★</td>
<td>★★★★★</td>
<td>★★★★★</td>
</tr>
<tr>
<td>Material utilization</td>
<td>★★★★★★</td>
<td>★★★★★★</td>
<td>★★★★★</td>
<td>★★★★★</td>
</tr>
<tr>
<td>Overall cost</td>
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<td>★★★★★★</td>
<td>★★★★★</td>
<td>★★★★★</td>
</tr>
</tbody>
</table>

**Annotation**

| ★★★★★★ | Excellent |
| ☆☆☆☆☆☆ | Poor      |

**Fig 12.** Comparison of various metallization techniques in terms of different parameters with considerably acceptable parameters of the metal grid design such as, contact resistivity ($<1 \text{ m}\Omega\cdot\text{cm}^2$), metal bulk conductivity ($\sim 2-3 \times 10^{-6} \text{ \Omega}\cdot\text{cm}$), material utilization ($<100$ mg per 6-inch wafer), with high throughput and lower cost (less than 0.1 USD/wafer).
5 Summary & Conclusions

We summarized the recent developments in perovskite/Si tandems from lab-scale (~1cm²) to industry scale devices. While the currently reported efficiencies with rated power values of 2.8 ~ 2.9 mWp are orders of magnitude lower than commercially available Si modules (>400Wp), there is considerable scope for the development of industrially applicable large-area front metallization schemes for monolithic perovskite/Si tandems. The front sun-facing side TCO is an important component of the perovskite/Si tandem device, with IZO being the primary candidate due to its high electrical conductivity and high near-infrared transparency, combined with room-temperature processing. However, as perovskite compositions become more thermally stable, via the reduction or elimination of MA⁺ (or similar organic cations), crystalline TCOs with higher transparency and mobilities can be utilized.

To understand the possible power losses from typical H-pattern metallization schemes, we modelled the front metallization grid for monolithic-perovskite/Si tandem solar cells. We observed that an optimized grid can result in a relative power loss of less than 5%, as wider finger spacing results in much lower shading losses in comparison to conventional single junction Si cells. Moreover, the lower currents in perovskite/Si tandems enable the opportunity to adopt relatively resistive metals such as Al with 6% metallization loss relative to losses in SHJ solar cell with a potentially significant impact on metallization cost.

Industrially applicable screen-printed metallization is an important route to realize large area perovskite/Si tandems. So far minimal progress has been shown with only one 22.6% (57.6 cm²) device having been reported. The key challenges in adopting screen printed metallization are the low annealing temperatures (<150 °C) as the perovskite layer is temperature sensitive, which results in low FF’s due to an increased series resistance of the cells coming from the higher contact resistance of metals (as the pastes are not fully cured). Development of low temperature sintering with increased conductivity of the printed metal lines by engineering the mix of metallic flakes and spheres is required for the pastes annealed at lower temperatures. Moreover, the TCO’s need to be optimized in such a way to improve the contact resistance to improve the performance of screen-printed perovskite/Si tandems.
Finally, we discussed the metallization cost, which is of prime importance in the PV industry and comprises one of the main cost contributing elements in the module's overall cost. Our front grid design with the potential of using relatively lower amount of silver paste suggests that a metallization cost of about 0.013 USD/Wp for 6 inch 30% efficient perovskite/Si tandems is achievable, which is more than 50% lower than the reference 24% single junction SHJ solar cell (0.077 USD/Wp). The price can be reduced further by reducing the silver consumption by adopting alternate metallization techniques such as inkjet and novel Flex-trail printing techniques. Another possible way of reducing the metallization cost is to replace silver with less expensive metals such as copper (e.g. via silver coated copper paste) or another material with a higher resistivity, for example, aluminium, keeping in mind the minimum conductivity required by tandems.

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