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Diego Di Girolamo, Guillaume Vidon, Jessica Barichello, Francesco Di Giacomo, Farshad Jafarzadeh, et al.. Breaking 1.7 V Open Circuit Voltage in Large Area Transparent Perovskite Solar Cells Using Interfaces Passivation. *Advanced Energy Materials*, 2024, 10.1002/aenm.202400663 . hal-04595571

HAL Id: hal-04595571

<https://hal.science/hal-04595571v1>

Submitted on 14 Nov 2024

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Breaking 1.7 V open circuit voltage in large area transparent perovskite solar cells using interfaces passivation.

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Abstract

Efficient semi-transparent solar cells can extend the adoption of photovoltaics beyond standard utility-scale, commercial, or residential applications. Halide perovskites are particularly suitable in this respect owing to their tunable bandgap. The main drawbacks in the development of transparent perovskite solar cells are the high open-circuit voltage (Voc) deficit and the difficulties in depositing high-quality thin films over large area substrates, given the low solubility of bromide and chloride precursors. In this work, passivation strategies are developed for the high bandgap Br perovskite able to reduce charge recombination and consequently improve the Voc. The study demonstrates 1 cm² perovskite solar cells with Voc up to 1.73 V (1.83 eV Quasi Fermi Level Splitting) and a PCE of 8.1%. The average visible transmittance (AVT) exceeds 70% by means of a bifacial light management and a record light utilization efficiency (LUE) of 5.72 is achieved. Moreover, the potential use of the technology is evaluated toward Internet of Things (IoT) application owing to a bifaciality factor of 87% along with 17% PCE under indoor lighting. Finally, the up-scaling is demonstrated by fabricating 20 cm² active area modules with PCE of 7.3% and Voc per cell up to 1.65 V.

Introduction

Photovoltaic technology has become the most cost-effective source of energy, having recently surpassed the significant milestone of 1 TWp of cumulated installed capacity^{1,2}. There are optimistic forecasts indicating that this figure will continue to grow, with an expectation that the annual installed capacity will enter the TWp range within the next decade^{1,2}. The market for photovoltaics is primarily dominated by crystalline silicon photovoltaics, which is a mature technology that has benefited greatly from continuous development over several decades³. However, there is potential for novel materials to facilitate a wider adoption of photovoltaics, particularly in sectors where it may not be optimal to implement silicon. Metal halide perovskites, in particular, are a promising example⁴. Owing to the tunable bandgap⁵ along with low costs of fabrication⁶ and excellent optoelectronic properties^{7,8} this class of materials is ideal for many applications, ranging from indoor PV^{9,10} to tandem with silicon PV^{11,12}. A particular application enabled by the tunable bandgap of halide perovskite is transparent photovoltaics. The advancement of solar cell technology that offers customizable aesthetic features and consistent energy output has the potential to revolutionize the field of building-integrated photovoltaics. Photovoltaic windows are a clear example of this, where the majority of visible light must be allowed to pass through the solar cells while efficiently converting infrared and/or ultraviolet light into electrical energy. This technology would be especially advantageous in urban areas, where the available rooftop space for conventional photovoltaics is limited. The ability to utilize vertical surfaces such as windows or entire facades could provide up to 40 % of the required electrical energy demand¹³.

Solar cells that are transparent to visible light are capable of functioning with infrared (IR) and/or ultraviolet (UV) light. Organic semiconductors and molecules utilized in for organic photovoltaics (OPV)¹⁴ or as sensitizers in dye-sensitized solar cells (DSSCs) can be designed to absorb IR light with wavelength beyond 780 nm¹⁵. Conversely, conventional semiconductors, including metal halide perovskites absorb the light below a certain wavelength threshold^{16,17}, making them appropriate for utilizing the UV range (< 380 nm).

There are three key figure of merits for transparent photovoltaics: the power conversion efficiency (PCE), the average visible transmittance (AVT) calculated considering the photopic response of the human eye, the transmittance of the cell and the solar spectrum, and the light utilization efficiency (LUE), that is the product of PCE and AVT¹³. So far, the highest LUE value demonstrated for perovskite solar cells (PSCs) is 5.12 % (PCE = 7.50 %, AVT= 68.2 %) by the group of Alex Jen¹⁸, employing a double step deposited mixed chloride/bromide $\text{FAPbBr}_{3-x}\text{Cl}_x$. The majority of literature

reports LUE values below 4%, indicating significant challenges in achieving efficient ST-PSCs with AVT exceeding 50%¹⁹. The primary impediment to achieving a high PCE for wide bandgap PSCs is the substantial voltage loss, which is typically above 0.8V (corresponding to a 1.53 V Voc for a 2.36 eV bandgap) in ST-PSCs²⁰ and 0.65 V for opaque PSCs²¹. As we shown in our previous work, the alloying of chloride into bromide perovskites exacerbates the Voc losses issue²⁰. Although the precise mechanism remains the subject of ongoing investigation, this observation suggests that the bromide-chloride alloy may introduce detrimental defect chemistry into the perovskite lattice. Conversely, to achieve high AVT with bromide perovskites it is imperative to restrict the film thickness to below a certain threshold. However, casting a <200 nm perovskite layer on large area substrates presents a formidable challenge, partly due to the faster precipitation of bromide perovskites compared to their iodide-based counterparts. In fact, most published works exploit double step deposition, where the uniformity of the deposited layer is largely contingent on the deposition of PbBr₂, or necessitate post-synthesis treatments aimed at improving the morphology. Reducing the perovskite layer thickness typically increases surface area / volume ratio of the perovskite layer. This will emphasize the impact of the grain boundaries defects on the performance of the cell, thus an advanced passivation strategy is required for thin perovskite cells.

In this work, we develop several passivation strategies based on the use of bulky cations from chloride salts, namely neo-Pentylammonium chloride and iso-Pentylammonium chloride to passivate defects and to form a two-dimensional large gap perovskite layer on a bulk FAPbBr₃. The surface passivation of the perovskite by means of these bulky ammonium cations delivered a Voc 1.73 V along with Quasi-Fermi Level Splitting (QFLS) above 1.83 eV. Thinning down the perovskite film along with careful light management enables PCE=8.1 %, AVT > 70 % and LUE > 5.7 % in large area (1cm²) PSCs. Notably, the reduction of the thickness is not sufficient to achieve high Color Rendering Index (CRI) value (60.4 in average). For this reason, hereafter we will define our devices as “colored”.

Moreover, the solvent-antisolvent strategy used for perovskite deposition comprises only DMSO and Ethyl Acetate, representing one of the best alternatives for the solution processing of metal halide perovskites in terms of hazard, health, and environmental impact²²⁻²⁴. A bifaciality factor of 87 % along with a PCE of 17 % under indoor low-light illumination highlight the potential of this colored PV technology as an element for next generation architecture. Finally, the upscaling has been demonstrated fabricating semi-transparent perovskite solar module (ST-PSM) with PCE up to 7.3 % and LUE= 4.74 %.

Results and discussion

To address the Voc deficit issue of bromide perovskites we sought for the introduction of bulky cations from chloride salts, namely the neo-Pentylammonium chloride (hereafter called NEO) and/or iso-Pentylammonium chloride (hereafter called ISO), on the surface of 150 nm thick FAPbBr₃ perovskite film, as presented in **Figure 1a**. In fact, NEO has previously been demonstrated as an effective 2D passivator capable of significantly increase the Voc of devices based on MAPbBr₃, resulting in Voc values as high as 1.65 V²¹. We also investigated an isomer to NEO, namely the iso-Pentylammonium chloride that has not yet employed in PSC technology. In order to grasp information about the role of the introduction of both bulky chloride cations, morphological characterizations such like Scanning Electron Microscopy (SEM) and Atomic Force Microscopy (AFM) planar images have been performed. Low and High magnification SEM images (**Figure S1**) confirmed morphological changes at the surfaces of the polycrystalline layer using both NEO and ISO bulky chloride cations. It is confirmed from AFM images (**Figure S2**), where it is also found an increase of the surface roughness in NEO (18 nm) samples with respect to the REF and ISO samples (12 nm). A combined low and high angle XRD investigation was performed, offering an important insight into the effects of NEO and ISO on the structural properties of the perovskite layers. As visible in **Figure 1b** pure α -phase perovskite (see reported Miller indexes) was detected for all samples. Moreover, crystallographic FTO signature labelled accordingly to ICDD card nr. 00-003-1114 was also observed. In the inset of **Figure 1b**, the low angular region of the patterns is reported, allowing to observe the eventual formation of 2D perovskites. Indeed, only ISO-containing samples evidences the presence of 2D perovskite, adopting the Ruddlesden–Popper type crystal structure. The (020) reflection, and subsequent multiple [0k0] orientations, was identified at $2\theta = 4.0^\circ$, corresponding to the formation of the [PbI₆]⁴⁻ octahedral layers. Such orientations and inter-planar spacing typically correspond to a number of 2D layers $n = 2$ ²⁵⁻²⁷. These experimental evidences therefore suggest that ISO can form a 3D-2D interface while NEO is expected to be simply adsorbed on the perovskite surface.

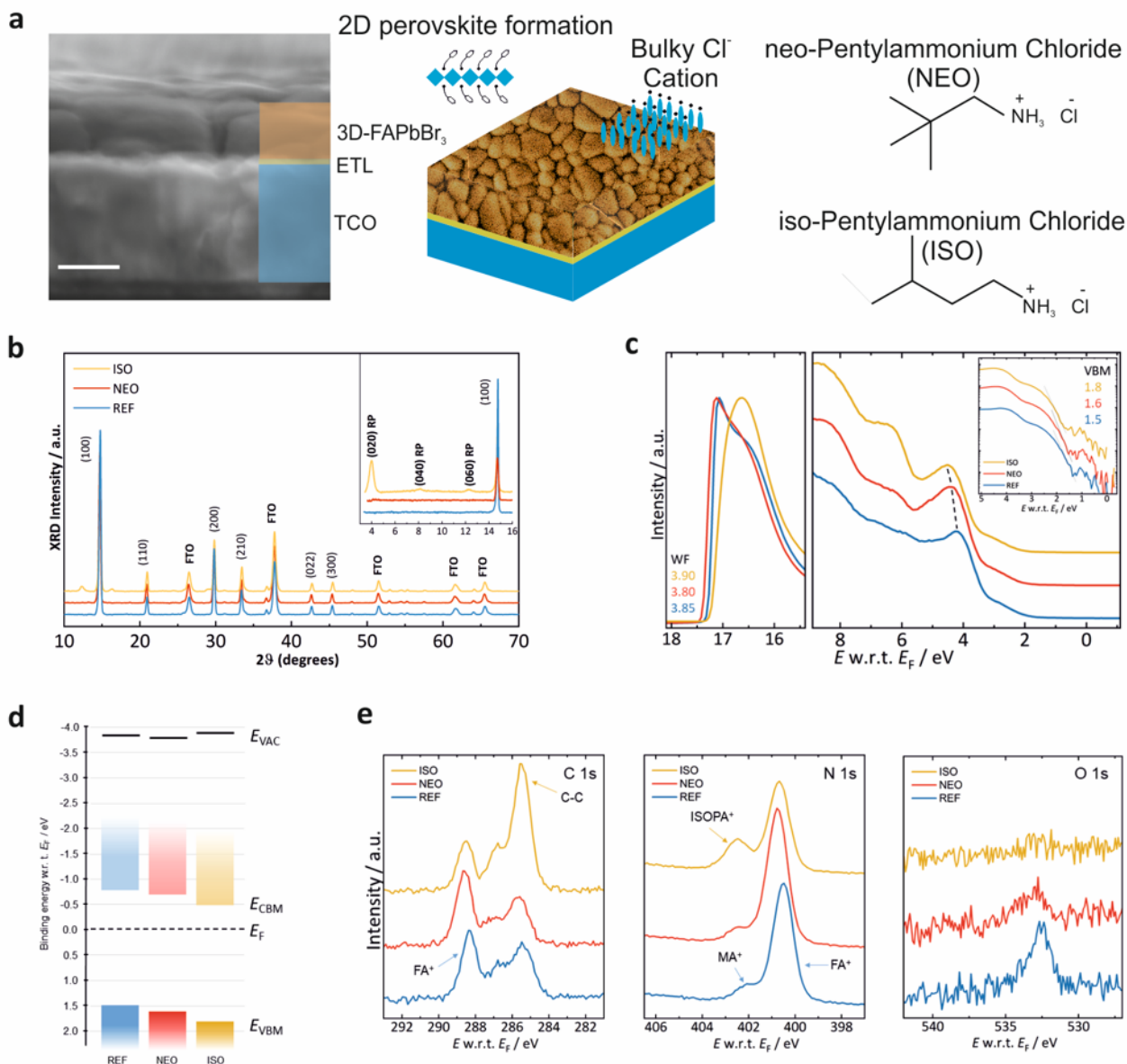


Figure 1. XRD and Photoemission spectroscopy data of the free surfaces of the REF, NEO and ISO samples. *a.* Passivation scheme using bulky chloride cations showing the chemical formula of NEO and ISO molecules. *b.* XRD patterns collected upon the REF, NEO, ISO samples. Perovskite structure is labelled with Miller indexes corresponding to the cubic phase. In the inset low angular XRD patterns are shown evidencing the presence of PVK 2D structure detected from ISO. *c.* UPS spectra with secondary electron cut-off for the work function (WF) determination and valence band region for the determination of the valence band maximum (VBM) from the semi-log plot (inset). The dashed lines in the valence band region indicate the shift of the spectra with surface modifiers. *d.* Energy level diagram showing the VBM and vacuum level position as well as the shift of the Fermi level in the gap towards the (projected) position of the conduction band minimum with NEO and ISO

modification with respect to the pristine perovskite surface. e. XPS core level scans for the reference and surfactant-treated FAPbBr₃ films showing the C 1s, N 1s and O 1s spectral regions. The spectral signatures for the C and N species in FA+ are indicated as well as the ammonium content due to MA+ and ISOPA+.

We employ photoemission spectroscopy measurements to examine the chemical changes at the surface of the FAPbBr₃ film due to the treatment with the NEO and ISO surfactants, respectively, as well as the corresponding effect on the surface energetics, which denote critical parameters for the functionality of the perovskite interface²⁸. In this regard, we determined the work function of the perovskite films by analyzing the secondary electron cutoff of the ultraviolet photoemission spectra (UPS). Our findings indicate that the addition of NEO and ISO treatments has only a minor impact on the vacuum level position of the perovskite film (as shown in Figure 1c). Specifically, while the initial work function of the REF sample was measured at 3.85 eV, it increased to 3.9 eV for the NEO sample and decreased to 3.8 eV for the ISO sample. Notably, although no clear trend emerges for the work function, the valence band spectra of the perovskite reveal a significant shift following treatment with the molecules. While the initial valence band onset was observed at 1.5 eV relative to the Fermi level, this value increased to 1.6 eV for the NEO sample and 1.8 eV for the ISO sample. This change suggests that the Fermi level at the surface is shifted further towards the conduction band (**Figure 1d**), which we project from a constant band gap of 2.3 eV in accordance with our photoluminescence data. These findings align with earlier studies showing that the Fermi level at the perovskite surface is close to the conduction band if the perovskite is deposited on an n-type substrate and exhibits a low concentration of defect states in the bulk²⁹. However, the presence of defect states at the surface can introduce a pinning of the Fermi level to the energy level of defect states, as may be the case for the pristine FAPbBr₃ surface. Our data suggests that the molecular treatment, particularly with the ISO molecule, passivates such defect states, thus shifting the Fermi level back towards the conduction band edge. Notably, this shift in the position of the Fermi level closer to the conduction band due to the molecular surfactants is not detrimental for carrier extraction and blocking, as the final energy level alignment changes once the top interface with is formed. In all cases, we do not expect any significant barrier for hole as the ionization energy of the REF, NEO and ISO samples, remain high at 5.4 eV, 5.4 eV and 5.6 eV, respectively, well above the ionization energy of PTAA, which amounts to 5.2 eV³⁰.

The X-ray photoemission spectroscopy (XPS) data, depicted in **Figure 1e**, indicates that the carbon and nitrogen signal of the REF and NEO do not show any marked difference. In contrast to that, the ISO sample exhibits an increase of the contribution of C-C bonds and a concomitant decrease of contribution of the C-N-C bonds to the C 1s spectra. This goes along with an increase of the ammonium content located at 402.4 eV in the N 1s spectra and a concomitant decrease of formamidinium. Similarly, we exclusively find traces of chlorine (not shown here) in the ISO samples. Thus, only for the ISO sample a significant amount of new molecular species is found at the surface. Of further note, the oxygen content is not negligible for the bare FAPbBr₃ surface, but the amount decreases after NEO treatment, whereas in case of the ISO treatment we find no more oxygen at the surface. These results are in agreement with the suggestion of reduced surface defect densities and changed surface energetics via the NEO and ISO treatments. The Pb and Br core levels do not exhibit any significant changes and in particular, we observe the absence of metallic lead (**Figure S4**).

We consider the following reference ST-PSC stack based on nip architecture: Glass/FTO/TiO₂:SnO₂/FAPbBr₃/PTAA/ITO (**Figure 2a**). Cells have an active area of 1 cm² where the thickness of the each layer are measured by cross-sectional SEM image in **Figure S4**. The compact TiO₂ film deposited through spray pyrolysis is doped with Nb (2 % atomic ratio to Ti in the precursor solution) to enhance its electrical conductivity³¹. An ultra-thin layer of SnO₂ nanoparticles was used to decorate the surface of the Nb:TiO₂ layer, as recently reported by Kim and coauthors³². This approach enabled us to benefit from the good electron extraction of SnO₂ yet maintaining the structural robustness of the TiO₂ layer. The choice of PTAA, which is doped with LiTFSI and 4-tBP, stems from its better compatibility with the sputtering of ITO top electrode with respect to Spiro-OMeTAD¹². No buffer layers are introduced between PTAA and ITO as results of the optimization of the sputtering parameters (power density, pressure and temperature) for ITO layer as shown in our previous studies^{12,20,33}. In **Figure 2b** we report the PCE box charts measured at Maximum Power Point (MPP) and 1Sun AM1.5G illumination conditions performed after the measurements of the J-V characteristics (see **Figure S5-S8** and S.I. explanations for details on measurement conditions and the results measured from independent laboratories).

The reference devices performed quite poorly with an average PCE around 4.2 % and a broad distribution of PCEs between 3.5 % and 5 %. As shown in **Figure 2c**, the Voc of the REF PSCs is well below 1.5 V even for the most performing devices. This represent a voltage loss above 0.8 V with respect to the FAPbBr₃ bandgap (2.30 eV). In order to overcome this limitation, both NEO and ISO

passivators have been introduced for the ultra-thin ST-PSC. By utilizing both salts as passivation agents, we successfully achieved Voc values above 1.6 V, albeit with notable differences between the two. Firstly, we observed that the PTAA solution in toluene exhibited limited spreading over the perovskite surface treated with ISO, whereas NEO had a negligible effect on this parameter. It is noteworthy that at higher concentrations, both salts induced poor PTAA wettability, but at the concentration of 1mg/mL, this phenomenon was only observed with ISO.

We achieved a high Voc (up to 1.65 V) using NEO, which served as an excellent starting point, but was unfortunately accompanied by a degree of hysteresis and/or instability during the initial J-V scans (**Figure S9**). In contrast, the ISO-passivated devices showed slightly lower average Voc likely due to poorer PTAA coverage but higher Jsc and FF values when compared to NEO (**Figure S10**). In addition, the devices passivated with ISO show a more stable electrical output across different J-V scans interspersed with a MPP tracking of two minutes as shown in **Figure S11**. The first and second J-V scan of the ISO devices practically overlap and the Maximum Power Point Tracking (MPPT) yields an even higher efficiency than the J-V curve. On the other hand, the first J-V curve from NEO devices usually show a marked hysteresis, although this was ameliorated in the second scan, , and a MPP tracking showing a bump in the firsts seconds followed by plateau with a slightly negative slope (**Figure S11**). Interestingly, after monitoring the tracking of the Voc under 1 Sun light exposure, we observe that NEO passivated device show a trend similar to the MPPT: a Voltage overshoot in the first 50s (reaching a Voc of 1.65 V) with a subsequent stabilization at lower values. On the opposite, the ISO devices reach a Voc plateau of 1.65V within the same time span (**Figure S12**).

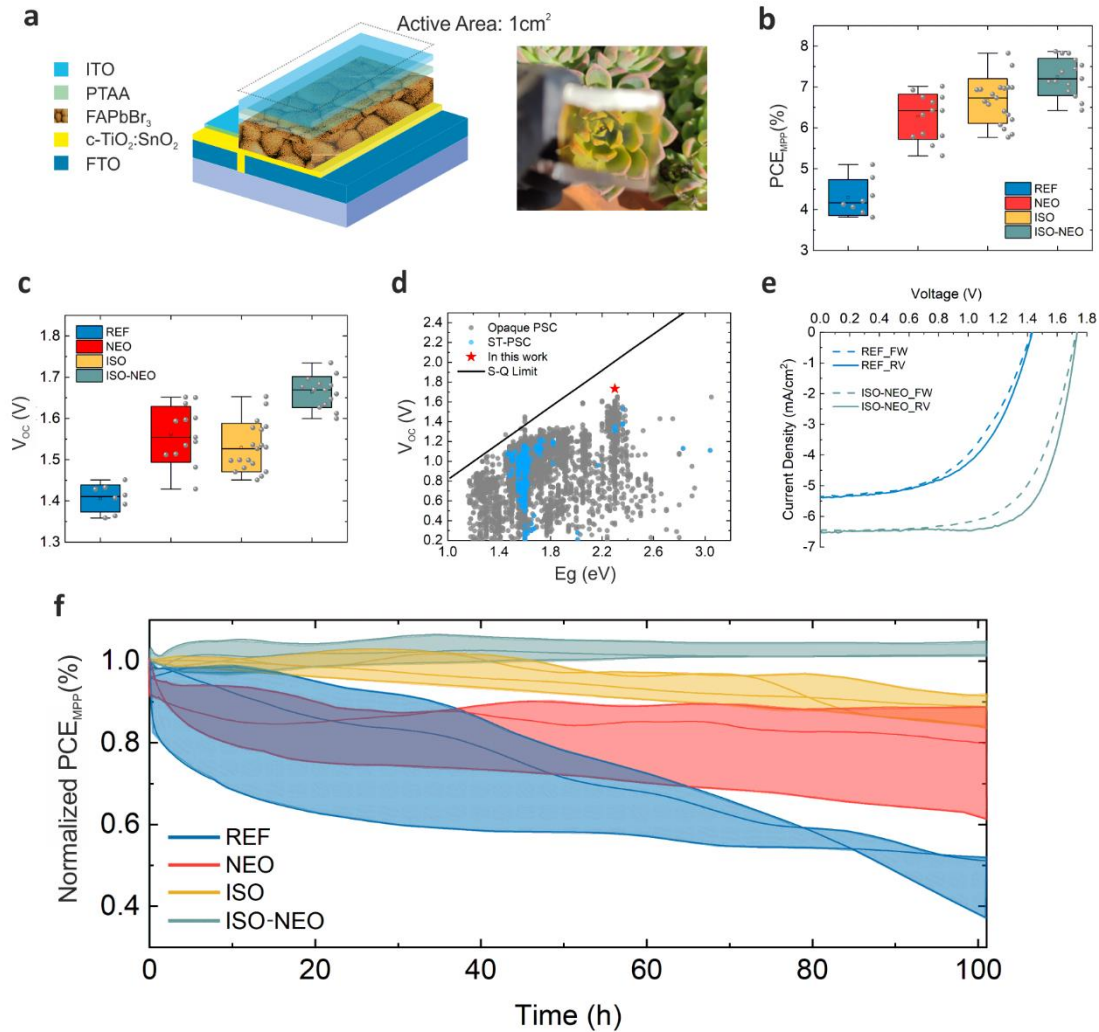


Figure 2. Photovoltaic performance of ST-PSC using different passivation scheme. **a.** Sketch and picture of the 1cm^2 -sized ST-PSC using planar NIP architecture. **b-c.** Box Charts graph for PCE (**b**) and Voc (**c**) values measured in a batch of n samples using REF ($n=8$), NEO ($n=13$), ISO ($n=19$) and ISO-NEO ($n=14$) passivation schemes. The left and right sides of the box are Mean \pm Standard Deviation (SE). The vertical line that split the box in two is the median. The whiskers represent the range within 1.5IQR where IQR is the interquartile interval. **d.** State-of-art Voc for opaque PSC (grey circles) and ST-PSC (blue circles) cells by varying the perovskite band gap. The data set was carried out from the perovskite database reported by Jacobsson et al.³⁴ The red star represent the best Voc value measured in our work. **e.** J-V characteristics of representative ST-PSCs using REF and ISO-NEO passivation scheme measured at 1 Sun AM1.5G illumination condition under forward and reverse scan directions. The scan rate of the J-V was 130mV/s . **f.** Light soaking test at maximum power point (MPP) performed in air at 50°C using LED-based lamp. The device are encapsulated with laminated adhesive kapton tape. The data are grouped using different colors representing the results of three

cells for each passivation strategies. The initial averaged PCE values are: $4.2 \pm 0.1\%$ for REF, $6.4 \pm 0.2\%$ for NEO, $6.8 \pm 0.1\%$ for ISO, $7.4 \pm 0.2\%$ for ISO-NEO.

Considering the complementarity of the two passivation strategies we operate both passivation at the same time (hereafter called ISO-NEO). In ISO-NEO passivation the ISO and NEO stock solution are mixed 1:1 v/v prior to use. Interestingly, with the ISO-NEO samples we obtained a higher efficiency compared to the single passivation schemes. ISO-NEO passivation helped to solve the issue related to the poor surface wettability of PTAA solution improving all the average PV parameters (and shrinking the box chart distribution) showing an outstanding maximum Voc of 1.73 V (**Figure 2c**). From our knowledge, this represent the best Voc value ever reported for PSC technology using opaque or semi-transparent device stacks (**Figure 2d**). The Voc tracking and the J-V/MPPT characterizations are in agreement with ISO results but showing huge improvement of all the PV parameters with respect to the REF sample (**Figure 2e**). As intermediate results, ISONEO based ST-PSC device reached a maximum tracked PCE_{MPP} equal to 7.9 %, AVT equal to 64 % for a LUE of 5.056 %. We remark that the improved PCE in ISO-NEO devices is mainly ascribed to the increase of Voc reaching average value of 1.66 V in a batch of 32 devices (**Figure S13**). Furthermore, XRD confirm the presence of 2D perovskite also for the ISO-NEO sample (**Figure S14**).

Notably, all devices, including the REF sample, exhibited excellent shelf-life stability when stored without encapsulation in ambient air, with their performances even improving during the first two to four weeks of storage (**Figure S15**). However, when testing their MPPT stability in air, the ISO-NEO passivation exhibited superior performance compared to the PSCs using only REF, NEO, and ISO (**Figure 2f**). The REF and NEO devices exhibited a clear burn-in within the first 10-20 hours before stabilizing their power output to approximately 40-50 % for REF and 60-80 % for NEO devices of the initial efficiency after 100 hours. Conversely, the ISO and ISO-NEO devices retained more than 85% of the initial efficiency, with ISO exhibiting a slow, linear decreasing trend without any evidence of burn-in. The burn-in degradation is a common behavior in perovskite and organic solar cells, for which several mechanisms have been proposed^{35,36}. In our case, the origin of the burn-in is likely to originate at the interface between PTAA and the perovskite, and the introduction of ISO represents a winning strategy in this regard. Prolonged light soaking test at MPP have been performed using ISO-NEO passivation resulting in a negligible relative PCE variation (-6 %) after 400 hours of ageing (**Figure S16**).

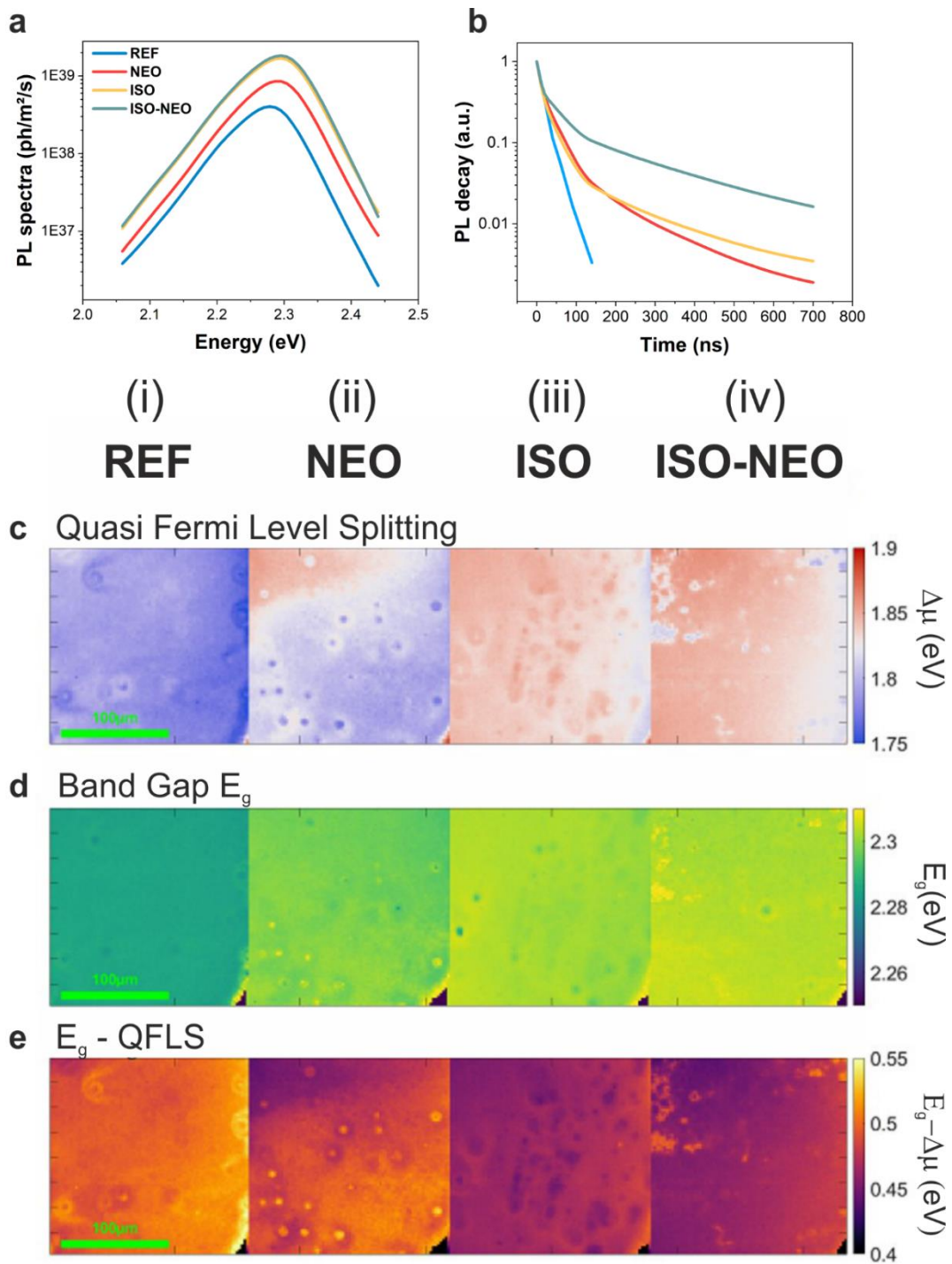


Figure 3. Photoluminescence analysis of full stacks (FTO/Nb:TiO₂/SnO₂/FAPbBr₃/PTAA) including reference, ISO, NEO and ISO-NEO samples. Column (i) to (iv) correspond to the addition (or not) of the cation at the absorber/HTL interface. **a. Averaged PL spectra **b.** Averaged PL decays **c.** Quasi-Fermi Level Splitting (QFLS) maps. **d.** Energy gap maps; **e.** Difference between the Energy gap and the QFLS.**

The impact of the bulky cations at the perovskite/HTL interface on the optoelectronic and transport properties of full stacks (FTO/Nb:TiO₂/SnO₂/FAPbBr₃/PTAA) has been investigated by two spatially³⁷ resolved multidimensional imaging systems: a Time-Resolved Fluorescence Imaging (TR-FLIM) set-up³⁸ and a spectrally resolved Hyperspectral Imager. In particular, we aimed at investigating the level of coverage of the perovskite absorber by the passivating agents and their impact on quantitative parameters related to the main photovoltaic figures of merits such as quasi-Fermi level splitting and carrier decay times. The Quasi-Fermi Level Splitting ($\Delta\mu$) represents the maximum work that can be extracted from electron-hole (e/h) pairs generated within a semiconductor, generally the absorber layer in a solar cell. Consequently, the highest attainable voltage observed at the electrodes of a solar cell is determined as $\Delta\mu/q$, where q denotes the elementary charge. The primary objective of this analysis is to examine the effect of the introduction of organic cations at the interface between the absorber and the Hole Transport Layer (HTL). This investigation aims at quantifying the reduction in non-radiative losses resulting from the incorporation of these passivation agents, ultimately leading to beneficial effects on the performance of the solar cell, particularly in terms of a significant enhancement of V_{oc} . The difference between the bandgap of the absorber and the QFLS provide a quantitative indicator of such non-radiative losses. We recently used the same approach to investigate carrier recombination dynamics in high efficient inverted PSCs dual passivated by organic cations³⁹.

First, photometrically calibrated and spectrally resolved maps were acquired on a reference sample and on three different stacks with NEO, ISO and ISO-NEO cations added at the interface between the perovskite and the PTAA. We then performed fitting with the generalized Planck's law to obtain local estimates of quasi-Fermi level splitting (QFLS) and band gap energy E_g - details of the physical models employed to determine such parameters are given in the supporting information.

The averaged recorded spectra are reported in **Figure 3a**. We observe a significant increase of the PL maximum intensity of the passivated samples compared to the reference, as well as a slight blue shift. The ISO and ISO-NEO samples exhibit similar PL average spectra. To have further insight on the carrier recombination dynamics we determined the local decay time on the different stacks. The precise methods and details are given in the supporting information. The resulting averaged decays are displayed in **Figure 3b** where we observe an impressive increase of the decay times from the reference sample with a time constant around 27 ns to the ISO at 135 ns, the NEO at 170 ns and at last the ISO-NEO with 230 ns. In **Figure 3c** we display maps of QFLS obtained at 1 Sun equivalent illumination for the different passivation strategies. We observe a gradual improvement from the

reference (i) with a QFLS of 1.79 eV to the NEO (ii) at 1.81 eV and to the ISO-NEO and ISO (iii) and (iv) with a QFLS of around 1.83 eV, leading to a 17 % reduction in voltage loss from the radiative limit of 2.02 eV. This demonstrates the beneficial effect of these compounds in minimising non-radiative losses at the absorber/HTL interface. These findings are in line with the UPS data (**Figure 1c**) which suggested lower defect densities at the surface upon integration of the molecular surfactants with the ISO showing the most pronounced effect. Therefore, the formation of a 2D perovskite layer, that was evidenced by XRD analysis in the case of ISO and ISO-NEO, resulted in a significant improvement of the optoelectronic properties of the stack. The high level of QFLS can be directly related to the record V_{oc} of 1.73 V obtained for a champion device passivated with ISO-NEO. We can notice a significant difference between the V_{oc} and the observed QFLS. Sputtering damage from ITO deposition, which has already been reported for semi-transparent devices⁴⁰, or a not perfect energetic alignment between the perovskite and the selective contacts, which could introduce differences between the QFLS and the actual V_{oc} of the solar cells ⁴¹, could be possible explanations for such discrepancy. The latter hypothesis is corroborated by the reduction of the difference between average QFLS and V_{oc} when the more performant passivation layer is deposited. Indeed, this parameter decreases from 390meV for the reference, to 260 meV for NEO and to about 200 meV for ISO and ISO-NEO compositions, which is nearly half of the value for the reference. Moreover, the J_{sc} significantly increased from an average value of 5.5 mA/cm² for the reference to 6.5 mA/cm² for the ISO-NEO composition, indicating a better carrier extraction in the passivated devices. Furthermore, we plot in **Figure 3d** the maps of fitted bandgap after the cations addition. The bandgap slightly blue shifted from 2.28 eV for the reference device to about 2.30 eV for the passivated devices, suggesting an incorporation of some Cl of the cations in the bulk of the absorber thus increasing the gap. The resulting change in bandgap is small compared to the improvement of the QFLS. To evidence this, we plot in **Figure 3e** the difference of the two quantities. This could be interpreted as the logarithm of the carrier density under operation as we expect:

$$np = N_c N_v \exp\left(-\frac{E_g - \Delta\mu}{kT}\right)$$

We observe that in spite of the slight increase in E_g , the difference $E_g - \Delta\mu$ is significantly more favorable in the passivated devices, as shown in **Figure 3e**. With a decrease of $E_g - \Delta\mu$ of ~ 1 kT, the carrier density under solar operation is increased $\sim 15\%$. These mappings also confirm the fact that the passivation is relatively homogeneous as the dispersion ($6 \times \text{standard deviation}/\text{mean}$) of the

QFLS is 5.6 % for NEO but as low as 3.5 % for ISO-NEO. Moreover, we calculate the Urbach energy from the absorptivity curves, as reported in **Figure S17**. The absorptivity decay is purely mono-exponential, revealing an Urbach absorption below the bandgap with a related energy of 16 meV, confirming the usual low thermal and structural disorder in the perovskite absorbers⁴².

We observe a strong correlation between the electrical data (V_{oc}) and the two optical characterizations described above. In **Figure S18** we plot V_{oc} and E_g -QFLS versus the decay times. These three parameters gradually improve from the reference sample to the single cation case (only ISO or only NEO), until reaching the maximal values for the mixed ISO-NEO passivation. The trends are thus similar, confirming the positive effect of such passivating agents on the optical and electrical properties of the devices. However, as previously reported by Zhu et al.²¹, we can observe a discrepancy between the rise in terms of QFLS (+ 40 meV) and the increase in terms of the V_{oc} (+ 230 mV). This indicates that the improvement in device performance, and in particular of the V_{oc} , is only partially due to interfacial passivation and that the introduction of the cations also improves the electrical behavior of the devices as a result of a possible higher shunt resistance, which is further supported by a significant rise in the FF. In **Figure S19**, we compared our results (in terms of QFLS and V_{oc}) with the other ST-PSCs shown in literature concerning the qV_{oc}/E_g (%) vs. E_g , a figure of merit introduced by Ruhle in 2016⁴³. However, we confirmed that our results represent the state of art for ST-PSC reaching 75.21 % and 79.56 % in terms of V_{oc} and QFLS, respectively.

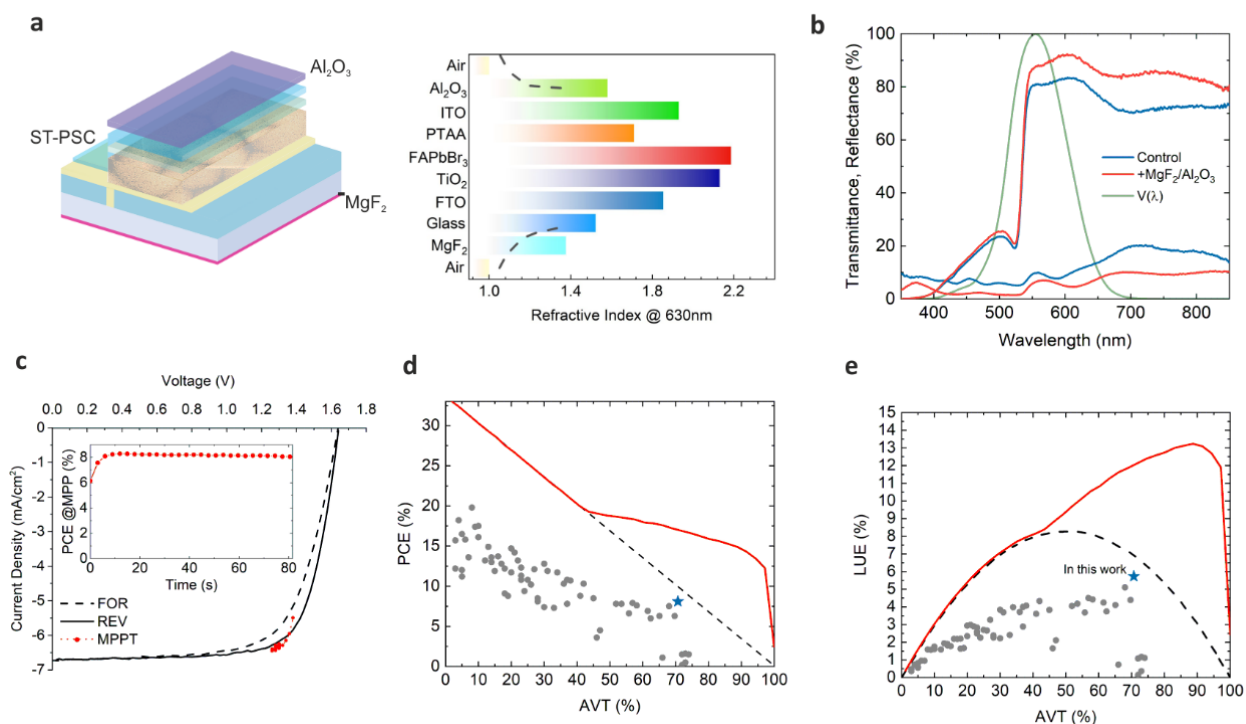


Figure 4. Light Management and key performing parameters of optimized ST-PSC. **a.** Device stack and refractive index profiles using light management tool. **b.** Transmittance and Reflectance spectra of the full ST-PSC with (+MgF₂/Al₂O₃) and without light management (control). **c.** J-V curve and MPPT of the best performing ST-PSC with PCE equal to 8.10 %, 7.54 % and 8.10 % under reverse scan, forward scan and MPPT, respectively. **d-e.** PCE vs. AVT and LUE vs. AVT charts representing the state-of-art results for ST-PSCs. The product of the PCE measured at MPP (panel c) and AVT after applying light management (panel b) results in the LUE shown in the panel e (blue star). Red curves represent the theoretical limits for LUE and PCE parameters by varying the AVT.

The analysis on the passivation supports us in rationalizing the increase in PCE. However, in order to improve the light utilization efficiency ($LUE = AVT \cdot PCE$), it is important to consider also the optical properties of the ST-PSC⁴⁴. The ST-PSCs show an AVT in the range of 61-64 %, with small sample-to-sample variation (**Figure S20**). This value combined with average PCE above 7 % results in average LUE above 4.5 %, at the state of the art of transparent photovoltaics^{19,20}.

Further improvement can be achieved by developing light management routes, minimizing the reflection at air/glass (where the light impinges on the solar cell) and at the ITO/air (where the light exits the device) interfaces^{45,46}. We fabricated another batch of ST-PSC cells applying MgF₂ anti-

reflective coating (ARC) on the glass side and a spin coated Al₂O₃ nanoparticles thin film on the sputtered ITO electrode. In this way, we smoothed the refractive index gradient, thus minimizing the optical reflections (**Figure 4a**). Notably, the AVT of the full device can be increased from 66-67 % with the application of the MgF₂ and further to 70.7 % with the application of Al₂O₃ accompanied by negligible losses in J_{sc} (**Figure S21**). The most performing device delivered a 70.69 % AVT and a PCE of 8.1 % (**Figure 4c**), resulting in a LUE of 5.72 %, overcoming the best results ever reported for semitransparent PSCs, as we show in **Figure 4d**. This result is particularly impressive considering that it is achieved on 1cm² active area solar cells. All the TPV metrics (J-V results, AVT, LUE) with and without the light management are reported in Tab.1 for the best device.

Tab.1 TPV metrics for the best ST-PSC reported in Figure 4c with and without LM.

	J-V Scan Direction	J _{sc} (mA/cm ²)	FF (%)	V _{oc} (V)	PCE (%)	PCE _{MPPT} (%)	AVT (%)	LUE (%)
Without LM	REV	6.75	69.8	1.710	8.06	8.0	64.2	5.136
	FOR	6.70	66.7	1.690	7.55			
With LM	REV	6.68	73.96	1.640	8.10	8.1	70.7	5.727
	FOR	6.72	68.42	1.638	7.54			

The Photon Balance Check (PBC) graphs are reported with and without ARCs in **Figure S22**. We should point out also the reduction of V_{oc} observed for the ST-PSC where ARC is used compared to those without ARC. In **Figure S23**, we reported the PV results obtained in a batch of six cells measured prior and after the light management tool (front and rear ARCs) is applied. The light management reduces the average V_{oc} of about 36 mV without affecting the average PCE (which slightly increases due to the increase in FF value). The V_{oc} reduction can be ascribed to the reduction of the photon recycling in ST-PSC as shown by Bulovic⁴⁷ and Kirchartz⁴⁸.

By comparing the integrated current density from the IPCE measured by illuminating from the two different directions we obtained a bifaciality factor of 87 %. We can clearly observe that the larger current loss occurs at low wavelength, which is mainly related to the parasitic light absorption from PTAA (below 400 nm). Interestingly, the illumination side has practically no effect on the J-V curve confirming that the bifaciality factor approach 90 % also when considering the power conversion efficiency (**Figure S24**). An important feature to consider is the PV performance under artificial

(indoor) light illumination. When integrated in facades as solar windows, or into alternative architectural elements, the semitransparent solar cells could also be illuminated with artificial (indoor) light. While this is negligible when compared to the illumination from the sun, during the night an efficient conversion of indoor lighting could supply low power electronics, as those comprising the IoT paradigm (alarm, sensors). Interestingly, we obtained excellent performances under artificial indoor light, with a PCE between 16-17 % in the range from 200 to 1000 lux (**Figure S25**). Notably, we could achieve a V_{oc} above 1.2 V at 1000 lux and a power density exceeding $60 \mu\text{W}/\text{cm}^2$ ($13 \mu\text{W}/\text{cm}^2$ at 200 lux). The irradiance spectra of the artificial indoor light is shown in **Figure S26** for 200 and 1000 lux, respectively. These results indicate that a 1m^2 colored solar window can reasonably deliver a power exceeding 100mW under indoor lighting, above the requirements for most household devices or wireless communication protocols¹⁰. In **Figure 4d** and **Figure 4e**, we also reported our state-of-art results in LUE (AVT) and PCE (AVT) behavior for ST-PSCs introducing the theoretical limits reported from Bing et al.⁴⁹.

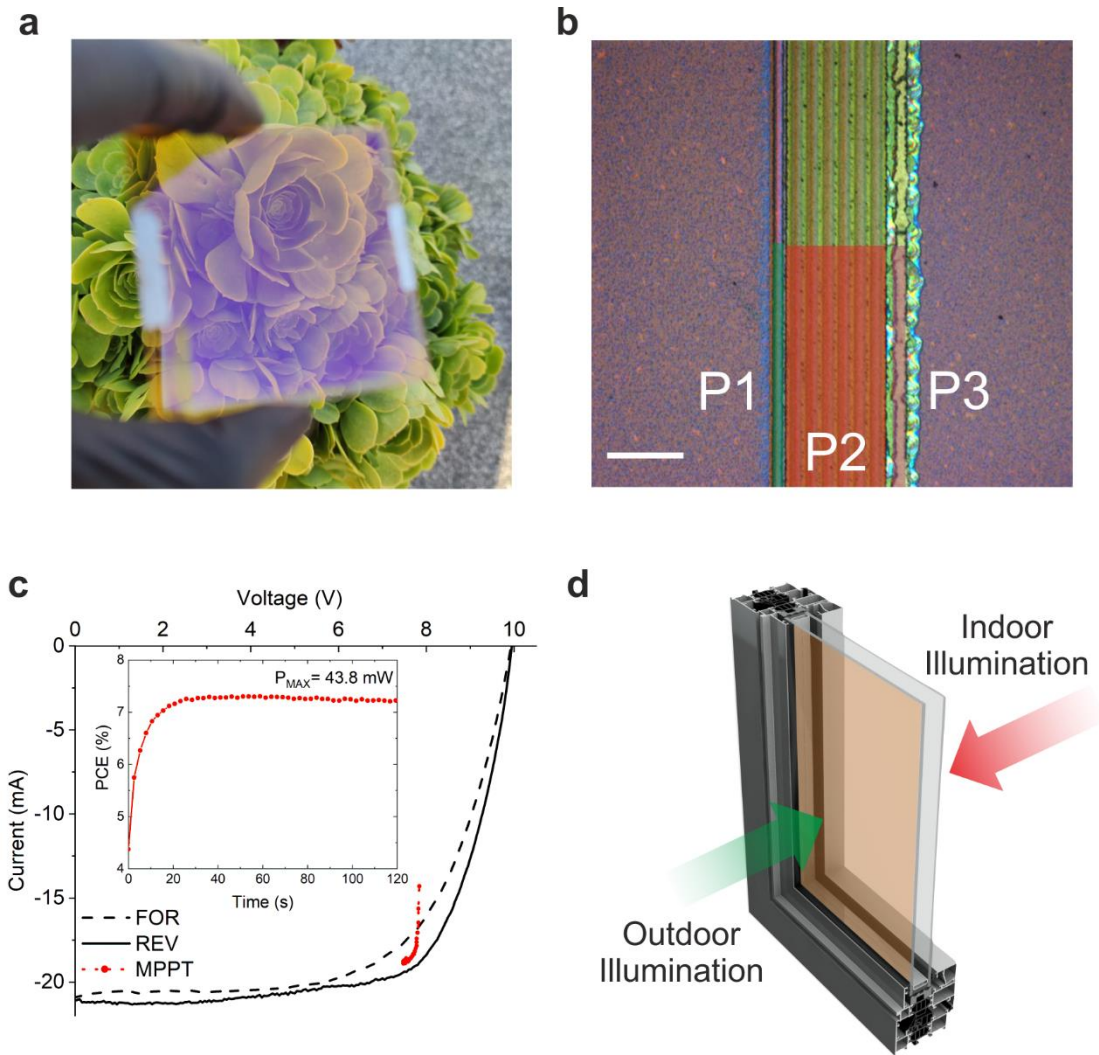


Figure 5. Up scaling process for ST-PSM with high geometrical fill factor. **a.** Image of the $6 \times 6 \text{ cm}^2$ -sized ST-PSM module highlighting its transparency from ITO-side. **b.** Confocal Microscope 20x image of the interconnection area showing P1-P2-P3 laser ablation process. The P1-P2-P3 total width is $160 \mu\text{m}$. The scale bar is $80 \mu\text{m}$. **c.** J-V characteristics of the ST-PSM measured under forward and reverse scan directions at 1 Sun AM1.5G illumination condition. The inset graph show the PCE measured under MPPT of 120 s. **d.** Sketch of the ST-PSM powered BIPV window highlighting the bifacial working operation in outdoor/indoor illumination conditions.

Finally, 20 cm^2 -sized ST-PSMs with high geometrical fill factor (up to 97.83 %) have been fabricated using 3D FAPbBr_3 perovskite and ISO-NEO passivation scheme (**Figure 5a-b**). The results are very encouraging showing a maximum steady-state PCE of 7.3 % (7.1 % in average) after 120 s of MPPT, V_{oc} up to 1.65 V/cell, AVT equal to 65 % (**Figure 5c**) showing a LUE equal to 4.74 % without using LM. Statistical PV parameters measured in a batch of eight ST-PSMs are reported in **Figure S27**.

Finally, we speculate a suitable exploitation of the ST-PSMs when integrated in a BIPV colored window (**Figure 5d**). The sketch highlighted the bifacial working operation of the ST-PSM powered BIPV colored window able to potentially generate electricity switching from outdoor (day) to indoor (at night, with artificial illumination) working conditions. The statistical results on Color Rendering Index (CRI) parameter (60.4 as average) and the CIE coordinates graph are shown in **Figure S28a-b** indicating an orangish color for our ST-PSCs devices. As expected, these results are far from claiming the color neutrality in PSCs for fully bromide PVK absorbers. Nevertheless, our devices are still interesting for environmental design and BIPV products where the coloration represents an additional value⁵⁰. Preliminary results (**Figure S29**) demonstrate the uniform and scalable deposition of the full stack using blade coating deposition technique⁵¹ showing an improved CRI (up to 78.4 %) obtained by adjusting the PVK thickness (<100nm) and its halide composition.

Conclusions

In this work we describe a strategy to develop ST-PSCs with LUE exceeding state-of-art by increasing the open circuit voltage of FAPbBr₃ based perovskites cells. The improved PV performance has been achieved by introducing bulky ammonium cation on the surface of the perovskite. We identified ISO and ISO-NEO able to form a 3D-2D junction improving the performances and the stability of the interface between perovskite and PTAA. A mixed surface passivation comprising ISO and NEO delivered the highest power conversion efficiency, Voc up to 1.73 V with promising environmental and operative stability. Moreover, we show how light management by minimizing the reflection at both air/glass and ITO/air interface enable a high AVT above 70 %. The best solar cell delivers a PCE of 8.1 % which combined to a 70.7 % AVT yields a record high LUE of 5.72 %. This set a new standard for perovskite solar cells, especially considering the difficulties in producing high quality bromide perovskite films for large area (1cm²) substrates by employing a relatively friendly solvent/anti-solvent system. Furthermore, we demonstrated the up-scaling of ST-PSM with 96 % GFF showing PCE up to 7.3 % on 20 cm² active area with outstanding Voc/cell up to 1.65 V and good reproducibility.

In addition, considering the sizeable mismatch between the Voc of the devices and the QFLS, we can further reduce this gap to increase the PCE of ST-PSC technology and to advance the understanding of the Voc loss in wide gap absorber for building integrated photovoltaic field.

We obtained a bifaciality factor of 87 % along with 17 % PCE under low intensity indoor lighting, which make the technology herein developed interesting also for IoT applications, opening new avenues for the application of colored ST-PSMs as smart components of the next generation architectonics such as pixelated PV glass façades, brise soleils, parapets and agri-PV. Future developments will be devoted to the increase of the CRI values obtained from our colored ST-PSCs and ST-PSMs devices in order to tackle the aesthetical issue referred to the FAPbBr₃ absorption cutoff.

Acknowledgments

This project has received funding from the European Union's Horizon 2020 research and innovation programme under Grant Agreement No 101007084 (CITYSOLAR). The authors are grateful to Marco Guaragno (CNR-ISM) for his technical support with X-ray experiments.

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