Journal of Materials Chemistry A

PAPER



Cite this: J. Mater. Chem. A, 2018, 6, 23787

Received 26th September 2018 Accepted 1st November 2018

DOI: 10.1039/c8ta09340g

rsc.li/materials-a

1. Introduction

The exceptional photovoltaic properties exhibited by organicinorganic halide perovskites (ABX3, A = Cs/CH3NH3 (MA)/ $HC(NH_2)_2$ (FA), B = Pb, X = Cl/Br/I) in addition to their low processing cost have made them among the most studied in the field of optoelectronics and a disruptive technology in the emerging photovoltaic (PV) market.1-4 Their promising features include broad and tunable band gaps by simple replacement of the cation(s) and/or halide component, high absorption coefficients (~10⁴ to 10⁵ cm⁻¹), low exciton binding energy ($\approx K_{\rm B}T$), ambipolar characteristics, and long charge-carrier diffusion lengths ($\approx 1 \,\mu$ m) and carrier lifetimes ($\approx 100 \text{ ns}$) in conjunction with easy solution processability which enable their direct integration into devices.⁵⁻⁹ In a very short span of time halide perovskite solar cells (PSCs) have seen tremendous development in the past few years since their first report as a visible light sensitizer in 2009 (ref. 10) and recently achieved a record high power conversion efficiency (PCE) of 23.3%.11,12

Hot dipping post treatment for improved efficiency in micro patterned semi-transparent perovskite solar cells[†]

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Perovskite solar cells have emerged as a new semi-transparent PV technology for urban infrastructures that demands an explicit trade-off between power conversion efficiency (PCE) and average visible transparency (AVT) which can be adjusted by various modifications in the absorber layer. Here, we introduce a scalable and facile "one and a half" step deposition route for mixed cation perovskites patterned in a sub-micron sized grid structure for semi-transparent solar cells. The initial perovskite phase is formed in one step using a grid pattern, while the additional step involves dipping of the pre-deposited perovskite grid in a hot solution of formamidinium iodide (FAI) in isopropanol (IPA). Detailed analysis suggests that the additional step increases pore filling, crystal quality, and grain size and lowers the content of residual PbI₂ as well as reveals improved photo physical properties. An average PCE ~10% with an AVT of 28% is attained with a gold contact for the champion semi-transparent solar cell. The proposed deposition route can be generalized for all other types of perovskite based devices to yield better efficiency.

For broader exploitation of the perovskite PV technology they can be integrated directly into building elements, such as walls, roofs and windows (building integrated photovoltaics, BIPV), or can be used as top cells for tandem devices both of which essentially require semi-transparent architectures.13-15 The semitransparency of the PSC will then have to be combined with the benefit of visible light transparency along with high power conversion efficiency. In order to achieve semitransparency, several groups have focused on ultra-thin film fabrication which relies on the spin coating technique or vacuum deposition and achieved a decent combination of efficiency and transparency.¹⁶⁻²⁰ However, the approach is non-scalable with the associated drawback of materials wastage. Moreover, the film thickness limits light absorption. Even the scalable techniques for perovskite deposition such as blade coating, slot-die coating, and spray deposition have poor control over the thickness which is important for obtaining semitransparency.

Thus, the requisite is a scalable architecture that can be easily processed without the waste of precursor solution and can overcome the absorption limiting thin film formulation leading to semitransparency at the same time. Herein, we use a mesh assisted grid deposition technique for mixed cation mixed halide perovskite on mesoporous TiO_2 . This technique has been previously reported by Rahmany *et al.* for MAPbI₃ perovskite cells with the introduction of a wetting agent that allows the quick spreading of perovskite over the active area followed by the slow evaporation of solvents.²¹ Islands of perovskite well-connected across the walls of the mesh are formed. These islands are as large as ~one micron in height, are



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[†] Electronic supplementary information (ESI) available. See DOI: 10.1039/c8ta09340g

capable of efficient light conversion and provide intermittent surface coverage for average transparency. This process ensures the complete usage of materials and is completely scalable once a suitable combination of power conversion efficiency (PCE) and average visible transparency (AVT) is established.

As far as perovskite fabrication is concerned, there are lots of issues reported such as inefficient conversion and control of composition, solvents, annealing temperature, antisolvent treatment.²² While the single step deposition, in which all precursors are dissolved in one solvent, suffers from the incomplete dissolution of Cs and hence suffers from large morphological variation throughout the film, the two step or sequential deposition results in better coverage and pore filling on the TiO₂ scaffold.^{23,24} Though the two step deposition has shown an improvement in morphology and conversion efficiency over the one step process and is widely used to prepare high performance devices, there are other challenges involved in it like lack of control over composition, crystallization kinetics, incomplete conversion, etc. Several groups also suggest the use of additives to retard the crystallization kinetics to boost the conversion from precursor solution to perovskite crystal layer.^{25,26} However, unconverted PbI₂ is a by-product which may form due to improper precursor concentration, air humidity or processing temperature.

In this work, we propose a new approach for perovskite formation which combines the benefits of both the one step and two step/sequential deposition. Here, the first step (one step) is the deposition of the perovskite Cs_{0.20}FA_{0.80}Pb(I_{0.6}Br_{0.4})₃ to form the grid architecture where a proper concentration of cations and anions are combined. An additional process is adopted to engineer the quality of the already fabricated perovskite grid via 1 step where the grid is dipped in FAIisopropanol (IPA) solution for better conversion to the perovskite phase along with improved pore filling. As the initial perovskite phase is formed in 1 step only, the additional step is termed the "half step" and hence the term "one and a half" (or 1.5) step deposition. A detailed investigation of the intermediate optimizations and stability test has been performed along with structural and optoelectronic studies of the grid prepared by the 1.5 step technique. Also, the devices prepared by the 1.5 step process show enhanced PCE as compared to those prepared by the 1 step process owing to their improved crystal quality, grain size, efficient perovskite phase conversion, lower trap density, improved absorbance and improved stability. The obtained AVT values further quantify the semitransparent nature of the devices.

2. Results and discussion

The structure of semi-transparent perovskite solar cells described here is based on a perovskite layer which is sandwiched between mesoporous TiO_2 (electron transport material, ETM) and the hole transport material (HTM) Spiro-MeOTAD and it is fabricated to maintain ~29% of the AVT in order to achieve semi-transparent nature of the complete device. Therefore, the cost effective and scalable "self-assembly" deposition method is adopted. A polymer mesh of 270 µm size is used here as the template of the assembly. The detailed fabrication of the complete device is mentioned in the ESI.† Fig. 1(a) illustrates the "one" and "half" step deposition corresponding to "grid formation" and "dipping in FAI solution", respectively. The primary grid is formed by adding the perovskite solution dropwise over the mesh kept on top of the TiO₂ film followed by the gradual evaporation of solvents and nucleus formation.27 The perovskite solution comprises the mixed cation mixed halide Cs_{0.20}FA_{0.80}Pb(I_{0.6}Br_{0.4})₃ perovskite, which was well studied by McMeekin et al.7 The introduction of a surfactant (here BYK 333) allows instant wetting of the solution along the walls of the screen printing mesh by reducing the surface tension. The wetting allows connected islands of the perovskite (grid) to form along the walls of the mesh with a void region in the centre.²¹ In this bottom-up approach, the assembly of the patterned perovskite seed crystals is formed with the gradual evaporation of solvents. A colour change from yellow to brown further confirms that perovskite nucleation has taken place.

After the grid formation, the mesh is removed gently and the substrates are annealed at 150 °C for the growth of the perovskite crystals (1 step). In the case of 1.5 step deposition, the perovskite grid is dipped in a closed container with a hot solution (hot plate temperature set to 100 °C) of FAI in IPA before annealing. A colour darkening can be observed within 20 seconds of dipping. The semitransparency of 1 step and 1.5 step deposited grids can be assessed by viewing the logo through them (see Fig. 1).

Fig. 1(b) and (c) show top view scanning electron microscopy (SEM) images of the grid fabricated by the 1 and 1.5 step methods, respectively. Better pore filling in the case of the 1.5 step method is evident from the traces of perovskite crystals formed in the void regions. The concentration of the wetting agent is crucial for the AVT and the performance of the cells. Fig. S1(a)[†] shows the increase in the AVT of the perovskite grid with increasing BYK concentration (% by weight). The photoluminescence (PL) peak position and PL intensity as a function of the BYK concentration can be observed in Fig S1(b).† The well grown islands in the case of higher BYK scatter light more than those in the case of lower BYK. This reduces the PL intensity along with a blue shift in the PL peak maxima. The organic entity of BYK may also partially quench the PL at high concentrations. Further, Fig. S1(c)† shows the device performance with different BYK concentrations. Based on these results we choose 3% BYK as the optimum concentration for the rest of the study, owing to its AVT > 33%, decent PL intensity and PCE > 9%.

Fig. 2 shows the detailed visualization of the effect of the additional half step on grid morphology. Fig. 2(a, b, c, and d) and (e, f, g, and h) show the images of the grid pattern viewed under optical and fluorescence microscopes, top view Scanning Electron Microscope (SEM) images and enlarged images of a single island for the 1 and 1.5 step deposited grids, respectively. The effect of the half step treatment is evident as the islands are well grown and connected after dipping (Fig. 2(a) *vs.* 2(e)). Fig. 2(b) and 2(f) also show a clear comparison of the fluorescent perovskite region (red) in the grid pattern. While in



Fig. 1 (a) Schematic illustration of perovskite grid formation by the 1.5 step deposition method. (b) Top view SEM images of the grid prepared by the (b) 1 step and (c) 1.5 step processes.

Fig. 2(b), the size of the islands and grid is random, they are much more organized and of even size in Fig. 2(f). The islands in the SEM image in Fig. 2(c) are not smooth with poor connectivity of the grid lines. The enlarged view of a single island in Fig. 2(d) and its inset also shows the breakage in grid architecture along with some exposed TiO₂. However, in the 1.5 step process, there is better growth as can be seen in Fig. 2(g) in terms of better interconnections between the islands. The inset in Fig. 2(h) shows a sharp spike of the island grown and no

exposed TiO₂ can be observed. Fig. 2(i) shows a cross sectional view of the complete device which corresponds to the schematic architecture shown in Fig. 2(j). The perovskite island is ~1.5 μ m in height with gradually decreasing thickness on either side. Inset II in Fig. 2(i) shows a region with negligible perovskite coverage where the HTM is likely to make contact with the ETM. The encircled region shows the onset of the perovskite island. Clearly, the unavoidable contact between HTM and ETM provides recombination channels which eventually limit device



Fig. 2 Images of the grid under optical and fluorescence microscopes, SEM image and magnified image of the encircled area with inset showing part of the island for the 1 step (a–d) and 1.5 step (e–h) processes. (i) Cross sectional SEM image of the device with inset (I) showing the complete island and (II) showing the perovskite (white dotted line) and non-perovskite region and (j) schematic of the device stack.

performance. This limitation in device performance due to its architecture is compromised to get desired transparency. This contact is reduced in 1.5 step deposition due to better coverage as seen in the top view SEM image in Fig. 1(c). The cross sectional SEM image of the non perovskite region is shown in Fig. S2.[†] There is an obvious enhancement in the grain size and thickness of the perovskite in the non-island areas which suggests that shunting paths are reduced after 1.5 step deposition. This is corroborated by the device study in the following sections. We commence our study by first characterizing the structural stability of the mixed-cation Cs_{0.2}FA_{0.8}Pb(Br_{0.4}I_{0.6})₃ perovskite with varying FAI concentrations in the dipping IPA solution in the 1.5 step process. Fig. 3(a) displays the XRD patterns for the standard untreated grid and three different dipping conditions of the 1.5 step process. All the indexed reflection planes with the common ones at 14.15°, 29.00° and 44.00° corresponding to the (001), (002) and (003) *hkl* planes, respectively, mark the presence of the sharp perovskite cubic α phase peaks.⁷ The zoomed in pattern is also shown for 2θ from 12–15° where the variation in the redundant PbI₂ phase can be observed clearly. For the 0.02 M FAI solution there is no signature of PbI2 at all. This can be attributed to the unconverted PbI2 in the standard grid getting converted to the perovskite phase by reacting with the additional hot FAI solution. There is a gradual reduction in FWHM and increase in intensity owing to better crystal growth and aligned structure due to the 1.5 step process.

The mobile FA⁺ and I⁻ in hot IPA contribute to the conversion of unconverted iodide/bromide phases of lead. This is why the darkening of the grid is observed while dipping. Any possible incorporation of FAI into the lattice, causing the structural change, cannot be completely ruled out. There is a negligible shift of the (001) plane towards a lower 2θ value which can be due to partial substitution of FAI into the lattice. The effect of the slight incorporation of FAI or iodide conversion may be out of the detection limit of XRD. So, the observation is further corroborated by absorption and PL measurements.

UV-Visible absorption spectroscopy is used to calculate the change in AVT and peak absorption of the perovskite as a function of the FAI concentration in the dipping solution of the 1.5 step process. Fig. S3(a)[†] shows a clear red shift in the absorption band at \sim 700 nm as the dipping solution concentration is increased. Also, it is worth noting that the onset of perovskite absorption even without annealing, along with the observed α -phase (with significant δ -phase) in the XRD pattern (see Fig. S3(b)[†]) confirms the hypothesis of nucleation as mentioned earlier. The 0 M condition represents only the hot IPA solution without FAI. The absorption (α) increases in this case with a red shift which means even when there is no FAI, some of the standard perovskite surface is dissolved in hot IPA and recrystallizes upon annealing, yielding better conversion and growth. As the FAI concentration is increased, the band edge becomes steeper indicating enhanced perovskite absorption. The Tauc plot shown in Fig. 3(b) shows that the optical band gap shifts to the low energy side with increasing FAI concentration. Further, the inset in Fig. 3(b) shows the quantitative measurement of AVT and band gap and the values change from 36% to 29% and 1.74 eV to 1.68 eV from the 1 step process to 0.02 M FAI concentration in the 1.5 step process, respectively. The shift in band gap marks the formation of the iodide phase. It should be noted that there is still a possibility that some PbBr₂ is not converted into the α phase. The required iodide for phase conversion is provided by the I⁻ from the hot FAI solution. The other minor contribution may also come from FA⁺ incorporation which results in lattice expansion (since the ionic radius of FA^+ (2.79 Å) > Cs⁺ (1.81 Å)) in accordance with Vegard's law.28

PL and time resolved PL (TRPL) are further performed to analyse the effect of the 1.5 step deposition method on the optical behaviour. Fig. 4(a) shows the PL excitation at 468 nm corresponding to 700 nm emission. The PbI₂ excitation peaks appear at 468 nm. At this excitation wavelength PL is recorded for different grid conditions which are in agreement with the absorption results. Starting from 0 M to 0.02 M FAI the PL shifts



Fig. 3 (a) XRD pattern of the standard 1 step perovskite grid compared to 1.5 step grids with different FAI solution concentrations. The pattern from 12° to 15° is enlarged to highlight the effect on Pbl₂ peak reduction. The peak marked "#" corresponds to the TiO₂ phase. (b) Tauc plot to calculate the energy band gap of the 1 and 1.5 step grid. The inset shows the variation in AVT (pink dots) and band gap (green dots) with different fabrication conditions from (i) to (v).



Fig. 4 (a) PL excitation and emission spectra corresponding to different dipping solution concentrations; (b) FWHM and peak position variation derived from PL; (c) variation in PL intensity with time for the 1 step and 1.5 step methods; (d) recombination kinetics of the grid by the 1 step method, (e) the 1.5 step method for 20 s FAI dipping and (f) the 1.5 step method for 40 s dipping after excitation at 600 nm between 12.0 nJ cm⁻² and 4.8 μ J cm⁻². Solid lines are fits based on the differential equation as described in the main text.

towards longer wavelength with a continuous increase in intensity. The increase in intensity varies inversely to that of full width at half maxima (FWHM). Fig. 4(b) shows the variation of PL maxima and FWHM with respect to the dipping conditions. While the PL shift denotes more and more iodide phase formation, the reduced FWHM and enhanced intensity implies the reduction in surface defects.²⁵ After FAI dipping, the conversion to the perovskite phase leads to phase homogeneity and reduces the defects.

As is well established, the mixed cation mixed halide perovskite suffers from structural and optical instability issues. The addition of Cs induces structural stability by modifying the tolerance factor.²⁹ However, photo instability still remains the major challenge and limits device performance under continuous illumination. Fig. 4(c) shows the variation in PL intensity under continuous excitation of 468 nm for 30 minutes. There is a clear change in the behaviour of intensity reduction from 28% in the 1 step grid deposition to 16% in the 1.5 step grid deposition. The reduction is exponential in both the cases; however, the trend slows down in the 1.5 step grid and tends to achieve saturation faster than the standard 1 step grid.

Clearly, the 1.5 step process minimizes the degrading tendency to a greater extent. In order to rule out any degradation caused by phase segregation, absorption kinetics was also monitored at 700 nm, as shown in Fig. S4.† Almost constant transparency was obtained in both the cases over 30 min. This suggests negligible light induced phase separation.

To further elucidate the effect of dipping in FAI solution, a time resolved photoluminescence study was performed. Fig. 4(d–f) shows the PL decay transients following photoexcitation of the perovskite grid across a range of excitation fluences (12.0 nJ cm⁻² to 4.8 μ J cm⁻², corresponding to an absorbed photon density of approximately 10¹⁴ cm⁻³ to 10¹⁶ cm⁻³). The grids were excited by 600 nm laser pulses at a repetition rate of

1 kHz and PL was collected as a function of increasing pump fluence. To understand the ensuing recombination kinetics behind the growth mechanism of the 1 and 1.5 step processes we chose three conditions of the grid *viz.* Fig. 4(d) the 1 step process, Fig. 4(e) 0.02 M FAI dipping for 20 s in the 1.5 step process and Fig. 4(f) 0.02 M FAI dipping for 40 s in the 1.5 step process, respectively. The monomolecular (arising from trapmediated recombination) and higher-order recombination rate constants were extracted by fitting the PL transients (Fig. 4(d–f)) to a differential equation,

$$\frac{\mathrm{d}n}{\mathrm{d}t} = -k_3n^3 - k_2n^2 - k_1n$$

where *n* is the excited charge carrier density, which is directly related to the absorbed photon density by a factor k_i where i = 3, 2, and 1 are the Auger, bimolecular, and monomolecular recombination rates, respectively. Here, it is worth noting that due to the low carrier densities under solar operating conditions ($<10^{15}$ cm⁻³), low monomolecular recombination is expected to play a dominant role in predicting device performance.³⁰ From our global fitting analysis, the 1 step perovskite grid has a monomolecular recombination rate of $k_1 = 4.39 \times 10^7$ s⁻¹ and a bimolecular recombination rate of $k_2 = 2.12 \times 10^{-8}$ cm³ s⁻¹. As summarised in Table 1, the rates obtained are one order of magnitude higher than the reported values for CsFAPbI₃ and other 3D perovskite thin films, highly likely due to quenching caused by the grid architecture or FTO substrate.³¹

Prior to annealing, dipping in FAI solution maximises perovskite formation and minimises defects. Subsequently, we observe a slight decrease in both the monomolecular and bimolecular recombination rates of the 1.5 step grid compared to those of the 1 step grid. The increase in device performance is corroborated by an overall decrease in both the trap-mediated and bimolecular recombination in the 1.5 step grid dipped for 20 s in FAI solution. However, prolonged dipping (here, 40 s) resulted in a decrease in device performance (shown later), suggesting defect formation or possible re-dissolution of the perovskite in the hot FAI solution. This can be concluded from the increase in the trap-mediated recombination rate. We note that the Auger recombination rate is similar across the samples under different growth conditions, but slightly higher in the 1.5 step/20 s case. However, this may not affect device performance because the carrier densities required for Auger recombination to become significant exceed typical solar operating conditions by two-orders or more. The effective life time increases from 20.8 ns for the 1 step grid to 26.7 ns for the 1.5 step grid. The detailed values corresponding to each fluence for the three cases are tabulated in Table S1.†

Further, the change in colour, structure and optical behaviour effected by the FAI dipping directs us to perform compositional analysis by X-ray photoelectron spectroscopy (XPS). Fig. S5[†] shows the chemical composition in the 1 step and 1.5 step grid for Pb 4f, I 3d, Br 3d and Cs 3d core level spectra. The binding energies are in agreement with those reported previously.22,32-34 The small shoulder peak at 138.8 eV indicates the presence of unsaturated Pb (Pb⁰)²⁶ and is an indication of the existence of iodide deficiencies in the perovskite lattice that act as recombination centres.35 There is a substantial reduction in its intensity in the 1.5 step grid which suggests a lack of unsaturated Pb and its iodide conversion. An analysis of the atomic ratio (see table in Fig. S4[†]) of (I + Br)/Pb reveals that it is nearly 3 in both the 1 step and 1.5 step case (3 : 1 ratio in the perfect stoichiometric crystal). However, as expected, there is a reduction in the Br concentration and enhancement in the I concentration keeping the ratio nearly constant.

After optimizing the dipping solution concentration in the 1.5 step process, we proceed to the device measurement to identify the best dipping time. Fig. S7[†] shows the effect of FAI dipping time on the current density, J_{sc} , at the maximum power point, MPP (J_{MPP}) , as a function of time. It is basically a measure of the operational stability of the device i.e. a voltage at the MPP is applied and the corresponding J_{sc} is measured under continuous illumination with respect to time. The J_{MPP} is quite stable for 20 and 40 s dipping time while it decreases significantly if the dipping time is too short (10 s) or too long (60 s). This gradual reduction is mainly attributed to the ion migration issues.36 In the present case, reducing trend for both 20 and 40 s dipping time are similar where the J_{MPP} tends to stabilize within 3–5 minutes from maximum $J_{
m sc}$ \sim 9.5 mA cm $^{-2}$ to $J_{
m MPP}$ \sim 7.7 mA cm⁻². Fig. S7(a)[†] shows the PL shift and intensity variation for the as-deposited (without annealing) 1 step and 1.5 step grid. The proper conversion is evident from the red shift in PL and maximum intensity corresponding to 20 s dipping which further supports the good device performance. Fig S7(b)† shows the deconvoluted PL peak in case of the as-deposited grid. The two fitted peaks confirm incomplete phase conversion.

To further validate the device performance, Fig. S8[†] shows the statistical variation of 5 devices for each dipping time. There is a threshold governed dependence of PCE on the dipping time and it is best for 20 s dipping time. Therefore, we continued with these conditions (20 s dipping time for 0.02 M concentration) further in this study.

Fig. 5(a) and (b) show the *J*–*V* curves of forward (*F*) and reverse (*R*) scans measured at 30 mV s⁻¹ for the 1.5 step and 1 step champion devices, respectively. The device performance parameters *viz.* open circuit voltage (V_{oc}), short circuit current density (J_{sc}), fill factor (FF) and power conversion efficiency

Table 1	Global-fitted	recombination	rates for the	perovskite o	arids under	various grov	vth conditions	

Type of recombination	1 step	1.5 step/20 s	1.5 step/40 s	
Monomolecular recombination, k_1 (s ⁻¹) Bimolecular recombination, k_2 (cm ³ s ⁻¹) Auger recombination, k_3 (cm ⁶ s ⁻¹)	$egin{array}{l} 4.39 imes 10^7 \ 2.12 imes 10^{-8} \ 1.13 imes 10^{-24} \end{array}$	$egin{array}{l} 3.81 imes 10^7 \ 1.04 imes 10^{-8} \ 1.45 imes 10^{-24} \end{array}$	$6.00 imes 10^7 \ 1.60 imes 10^{-8} \ 1.08 imes 10^{-24}$	

(PCE) from the reverse and forward scan, are reported in the inset table. The best reported PCE is 10.03% for the 1.5 step device with an AVT of 28% as compared to 8.33% for the 1 step device with an AVT of 36%.

Perovskite solar cells encounter a severe issue of hysteresis, *i.e.*, the inconsistency in the performance between two voltagesweeping directions while observing the current–voltage (J–V) measurements. Although the exact mechanism for hysteresis in perovskite solar cells is still unclear, there are some assumptions on its origin like ion migration within the perovskite, charge trapping, interfacial dipoles, *etc.*³⁷ The relative hysteresis can be quantified as³⁸

$$Hysteresis = \frac{PCE(R) - PCE(F)}{PCE(R)}$$

and decreases from ${\sim}22\%$ in the 1 step cell to 2% in the 1.5 step cell.

The accumulation of ions at the TiO_2 /perovskite interface and charge trapping are the widely accepted sources for hysteresis. Both the mechanisms are associated with defects in the perovskite structure. The hysteresis is much smaller in the 1.5 step cell than in the 1 step cell which further confirms a reduction in defects in the case of the 1.5 step cell. In order to further examine the effect of the 1.5 step process on the device performance, we have compared the dark J-Vcharacteristics. Fig. 5(c) shows the semi logarithmic plot of dark J-V characteristics of both the devices. The device processed by the 1.5 step method shows better diode characteristics with a lower leakage current and higher rectification ratio maintaining a lower reverse saturation current as compared to the 1 step device. These factors contribute to the suppressed recombination in the 1.5 step device.³⁹

The external quantum efficiency (EQE) is scanned in the range of 300–800 nm as shown in Fig. 5(d). There is an overall enhancement in EQE in the 1.5 step device. A shift in the band gap is also evident as the edge of the spectrum shifts towards longer wavelength (in accordance with the optical band gap). Also, the conversion is better on the lower wavelength side (350–400 nm) which indicates a reduction in the recombination centres at the TiO₂ interface. The inset in Fig. 5(d) shows an image of a semi-transparent cell with an ITO transparent contact made by 1.5 step deposition. The complete semi-transparent device gives 4.6% PCE with 28% AVT. Future work will aim to modify the transparent contact to minimise losses at the interface and enhance this efficiency value. There is an obvious impact of FAI treatment on all four device



Fig. 5 JV scan (forward and reverse) for the (a) 1.5 step and (b) 1 step device. (c) Semi logarithmic current–voltage curve of the 1 and 1.5 step device under dark conditions. (d) EQE measurement of the 1 and 1.5 step device with the inset showing the semi-transparent device with ITO top contact. (e–h) Statistical variation of 6 devices for the 1.5 and 1 step cells measuring open circuit voltage, fill factor, short circuit current density, and power conversion efficiency.

parameters (Fig. 5(e–h)) resulting in a marked improvement in the mean PCE from 7.4% for the 1 step device to 9.8% for the 1.5 step device.

The ageing performance was monitored for the champion device obtained by the 1.5 step process under repetitive cycles of light and dark (see Fig. 6). For this measurement, the cell was encapsulated. Periodic J-V scans were performed at different time intervals under constant illumination for 25 minutes and then the cell was kept in the dark under open circuit conditions for the next 5 minutes. Three such cycles for light (25 min) and dark (5 min) *I-V* scan were obtained during the course of 90 minutes. The reversible loss in PCE, V_{oc} and FF is worth noticing here. It is associated with the similar defect phenomena giving rise to hysteresis but on a different time scale. Keeping in the dark allows the defects to relax and reduces ion accumulation under open circuit conditions, thus minimizing the inbuilt electric field that induces the carriers to move into their respective contacts.⁴⁰ The J_{sc} values, however, show no recovery probably due to the material degradation during device aging, although a nearly constant J_{sc} value is observed under light which is consistent with Fig. S6.† FF drops in the first 4-5 min which is the stabilization time taken by the device to surpass ion migration and the light soaking effect and after that it attains a constant value. The FF recovers its original value in every subsequent cycle after keeping in the dark. The trend is similar with $V_{\rm oc}$; however, the decrement in every light scan is exponential. Therefore, the major loss in efficiency comes from the



Fig. 6 Ageing effect of the 1.5 step perovskite device performance under three cycles of light (25 min) and dark (5 min, shown by grey column). Open circuit conditions were applied when the cell was under dark conditions.

fall in value of V_{oc} and J_{sc} and hence data points for both follow a simmilar nature. The maximum drop in efficiency for each cycle corresponds to the MPP value which suggest that the cells after treatment are relatively photostable. The device exhibits 7% PCE, with a reduction of ~17% from its initial efficiency, even after rigorous stability measurements under light/dark and applied periodic bias.

3. Conclusion

In this work we demonstrated highly efficient semi-transparent perovskite solar cells made by a simple mesh assisted grid technique which allows for controlling the transparency and has exhibited the potential to be scaled up. We present 1.5 step deposition which improves the perovskite opto-electronic behaviour. It is different from any sequential dipping or post treatment processes where the perovskite phase is formed only after the latter step is performed. Simple mesh assisted screen printing technology has the potential to scale up the semitransparent perovskite architecture. The additional step of dipping in FAI-IPA solution improves the morphology, crystal quality, optical transparency, device performance and stability which was confirmed by detailed physical, optical and photovoltaic measurements. As a result, a PCE of 10.03% with an AVT of 28% is achieved with a gold contact. This result is one of the highest for semi-transparent perovskite solar cells. This 1.5 step method can be generalized to other perovskite solar devices with any composition and deposition variants to boost their efficiency.

Conflicts of interest

There are no conflicts to declare.

Acknowledgements

M. R. would like to acknowledge the support from the National Research Foundation (NRF), Singapore, through the Singapore-HUJ Alliance for Research and Enterprise (SHARE) and Nanomaterials for Energy and Water Management (NEW), NEW-CREATE Phase programme. We are also thankful to Ying Fan Tay for his help in performing XPS measurements and Dr J. M. R. Tan for his constant support and fruitful discussion during the experiments.

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