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# Multilayer Blade-Coating Fabrication of Methylammonium-Free Perovskite Photovoltaic Modules with 66 cm<sup>2</sup> Active Area

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Solar cells based on hybrid organic/inorganic perovskites have shown an astonishing efficiency development in the past years, having peaked in power conversion efficiencies of >25% for small-area single-junction devices. To pave the way for future commercialization, however, high power conversion efficiencies also have to be demonstrated on areas multiple orders of magnitude larger. Herein, methylammonium-free perovskite photovoltaic modules with an active area of 66 cm<sup>2</sup> are presented. All functional layers processable from solution are deposited by blade coating without the use of an antisolvent, demonstrating the feasibility of this approach for large-area module fabrication. The coating process is analyzed in detail and a model based on the Landau-Levich problem is developed for the blade-coating setup. The perovskite crystallinity can be improved by the addition of lead(II) thiocyanate, which results in increased crystallite size as judged by Williamson-Hall's analysis of X-ray diffraction data and corresponding scanning electron microscopy images. The homogeneity of the final modules is investigated with dark lock-in thermography and electroluminescence imaging, indicating only few shunts in the module area. Modules are made up of 15 serially interconnected solar cells and reveal a stabilized efficiency of 12.6% under maximum power point tracking.

## 1. Introduction

Solar cells based on hybrid perovskite photoabsorbers have shown the steepest learning curve of all photovoltaic (PV) technologies with respect to their power conversion efficiency. While perovskite layers can successfully be deposited by physical vapor deposition techniques<sup>[1,2]</sup> and hybrid routes,<sup>[3]</sup> solar cells with highest reported power conversion efficiencies most frequently include processing from a solution of precursor salts, only recently demonstrating an efficiency of 25.6%.<sup>[4]</sup> Active areas of record solar cells, however, are usually very small and in the mm<sup>2</sup> range. Besides stable long-term performance, high power conversion efficiencies also have to be demonstrated for device areas multiple orders of magnitude larger to enable market entrance of the technology. Perovskite PV module efficiency has now reached a record value of >20% on an active area of  $\approx$ 27 cm<sup>2</sup>,<sup>[5]</sup> with several results for larger-area mod-

ules presented in literature (for a review, see the study by Shalan<sup>[6]</sup>) or press releases<sup>[7–9]</sup> without details on the fabrication process. Even though perovskites containing the

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methylammonium (MA) cation at the A-site are suspected to result in inferior device stability,<sup>[10]</sup> most of the published module results are still based on methylammonium lead triiodide (MAPbI<sub>3</sub>), even more so when the perovskite layer was deposited by methods other than nonscalable spin coating. More recent examples in which perovskites with no or reduced MA content are deposited by scalable methods include work by Deng et al. (FA0.92Cs0.08PbI3 absorber. PCE = 20.2%, active area:  $\approx 27 \text{ cm}^2$ ),<sup>[5]</sup> Vesce et al. (Cs<sub>0.05</sub>(MA<sub>0.17</sub>FA<sub>0.83</sub>)<sub>0.95</sub>Pb(I<sub>0.83</sub>Br<sub>0.17</sub>)<sub>3</sub> absorber,  $\label{eq:PCE} PCE > 16\%, \quad active \quad area: \quad 10.05 \ cm^2), \end{tabular}^{[11]} \quad Yang \quad et \quad al.$  $(FA_{0.83}Cs_{0.17}PbI_{2.83}Br_{0.17} absorber, PCE = 16.63\%$ , active area  $20.77 \text{ cm}^2$ ),<sup>[12]</sup> and Lim et al. (FA<sub>0.875</sub>Cs<sub>0.125</sub>Pb(I<sub>0.875</sub>Br<sub>0.125</sub>)<sub>3</sub> absorber, PCE = 17.01%, active area:  $18.66 \text{ cm}^2$ ).<sup>[13]</sup>

Even though it is straightforward to understand that due to the involved crystallization processes, coating of the perovskite absorber layer is more challenging in comparison with other functional layers in the device, it is worth noting that the majority of published research work is relying on spin coating or physical vapor deposition of electron and/or hole transport materials which are not scalable or likely to compete with solution processing from an energetic footprint point of view.

To deposit high-quality perovskite layers from solution, it is crucial to enable fast nucleation of perovskite crystallites, followed by slow crystal growth.<sup>[14]</sup> Fast nucleation is achieved by rapid reduction of the solubility of the perovskite intermediate phase in solution, for example via addition of an antisolvent,<sup>[15]</sup> quick removal of solvents by vacuum<sup>[16]</sup> or gas quenching,<sup>[17]</sup> or heat treatment.<sup>[18]</sup> While addition of antisolvents is investigated with regard to scalable fabrication of perovskite PV devices,<sup>[11]</sup> gas quenching is an established technique<sup>[19–21]</sup> and likely to be used in future processes due to its ease of use and the avoidance of potentially critical antisolvents.

In this work, methylammonium-free perovskite modules with an active area of  $66 \text{ cm}^2$ , based on the absorber  $FA_{0.83}Cs_{0.17}PbI_3$ , are presented. All layers beside the front and back electrodes are deposited under nitrogen atmosphere from solution by blade-coating processes, using an air knife for rapid drying and crystallization of the transport and perovskite layers, respectively.

To develop a coating process, several methylammonium-free perovskite compositions are investigated, targeting a low bandgap to enable efficient single-junction devices. Also, a model based on the Landau-Levich problem is developed and applied to the perovskite and PCBM coating process, for which a good agreement between theory and experiment is found. Furthermore, the effect of adding lead(II) thiocyanate Pb(SCN)<sub>2</sub> to the perovskite coating solution on the crystallinity of resulting layers is investigated. It is found that Pb(SCN)<sub>2</sub> addition increases crystallite size within the perovskite layer, as judged by Williamson-Hall's analysis of X-ray diffraction (XRD) data and scanning electron microscopy (SEM), and correspondingly affects solar cell performance. Optimized modules yield a power conversion efficiency of 12.6% under 1 sun illumination. Dark lock-in thermography (DLIT) is used to investigate the coating quality and strategies to improve it are discussed.

### 2. Results and Discussion

#### 2.1. Perovskite Composition and Solar Cell Results

For single-junction perovskite solar cells, FAPbI<sub>3</sub> can be regarded as an optimal material due to its low bandgap and as it is methylammonium free. However, even in spin-coated devices, the desired cubic  $\alpha$ -phase is often not formed or instable, due to an unfavorable Goldschmidt tolerance factor and a more stable, nonphotoactive hexagonal  $\delta$ -phase.<sup>[22]</sup> To identify potentially more stable perovskite stoichiometries, the tolerance factors of various compositions of FA<sub>1-x</sub>Cs<sub>x</sub>PbI<sub>3</sub> were calculated (Figure 1).

It is worth to note that with an increasing Cs content, the perovskite bandgap increases.<sup>[23]</sup> Correspondingly, the Cs content should be kept low for optimal performance of singlejunction devices. Perovskite compositions with x = 0.04, 0.09, and 0.17 were chosen for blade-coating trials. Even though a black perovskite phase was formed in all cases, only for x = 0.17homogeneous perovskite layers could be obtained with the used parameter set. This is attributed to the potentially smaller processing window for blade coating in comparison to, for example, spin coating. With adjusted blade-coating parameters, it is likely that perovskites with higher or lower Cs content can be formed as well. The bandgap of the perovskite with optimized composition was determined from the signal peak in photoluminescence spectra to be 1.56 eV (Figure S1, Supporting Information). Throughout the remaining experiments, the same composition (FA083Cs017PbI3) dissolved in DMF and NMP was used. With this, small-area  $(0.09 \text{ cm}^2)$  solar cells fabricated by conventional spin-coating methods yielded a power conversion efficiency of 18.1% (Figure 2).

#### 2.2. Establishment of a Blade-Coating Model

To understand the blade-coating process in more detail and to rationalize coating parameters for module fabrication, a model based on the Landau–Levich problem was developed for the used blade-coating setup, following a theory established by Le Berre



**Figure 1.** Goldschmidt tolerance factors for perovskites with stoichiometry  $FA_{1-x}Cs_xPbI_3$  and MAPbI<sub>3</sub>. Perovskites with x = 0.04, 0.09, and 0.17 were used for blade-coating trials in this work.





Figure 2. Figures of merit of spin-coated perovskite solar cells with an active area of 9 mm<sup>2</sup>, based on the  $FA_{0.83}Cs_{0.17}PbI_3$  absorber.

et al.<sup>[24]</sup> and Davis et al.<sup>[25]</sup> In short, the wet film thickness  $t_{\text{wet}}$  is determined by the viscosity of the coating solution  $\mu$ , its surface tension  $\gamma$ , the blade movement speed *U*, a constant  $c_1$ , and the characteristic length scale *L* 

$$t_{\rm wet} = c_1 \times L \times \left(\frac{\mu U}{\gamma}\right)^{2/3} \tag{1}$$

The characteristic length scale *L* is given as

$$L = \frac{H}{\cos(\theta_1) + \cos(\theta_2) - \frac{H^2}{2 \cdot l_{cap}}}$$
(2)

where *H* is the height of the meniscus,  $\theta_1$  and  $\theta_2$  the upper- and lower-back meniscus wetting angles, and  $l_{cap}$  is the capillary length, equal to  $\gamma/\rho g$ , with  $\rho$  being the density of the liquid and *g* the gravitational acceleration. The resulting dry film thickness  $t_{dry}$  is then given by

$$t_{\rm dry} = t_{\rm wet} \times \frac{\frac{w_{\rm pero}}{\rho_{\rm pero}}}{\frac{w_{\rm solvent}}{\rho_{\rm pero}} + \frac{w_{\rm solvent}}{\rho_{\rm solvent}}}$$
(3)

where  $\rho_{\text{pero/solvent}}$  and  $w_{\text{pero/solvent}}$  are the density and mass fraction of the perovskite and solvent, respectively.

For implementation of this theoretical description, it needs to be considered that H is a function of the applied volume V, the blade dimensions, as well as coating gap G. To simplify its calculation, the applied solution is assumed to be distributed over the area underneath the blade and two adjacent triangular symmetrical prisms forming at the sides. With that, Equation (4) can be derived, indicating that H is constant and defined by the gap size G for small volumes of solution because the meniscus is forming only under the flat part of the blade.

$$H(V) = \begin{cases} \sqrt{G^2 + \tan(\alpha) \times \left(\frac{V}{B_1} - GB_2\right)} & \text{for } \frac{V}{B_1} > G \times B_2 \\ G & \text{for } \frac{V}{B_1} \le G \times B_2 \end{cases}$$
(4)

where  $B_1$  is the coating width and  $B_2$  and  $\alpha$  are parameters of the blade geometry, namely, the length of the flat part of the blade and the angle of the two slopes above the up- and downstream meniscus.

Equation (1)–(4) combined give an expression for the dry layer thickness depending on the controllable coating parameters such as coating speed *U*, volume *V*, and coating gap *G*. Density, viscosity, and surface tension of the coating solutions are assumed to be similar to the solvents, whose values were used in the calculations. An error in these assumptions does not affect the curvature of the theoretical data but leads to an offset between experimental and theoretical data, which can be accommodated by adjusting the constant  $c_1$  in Equation (1).

A close-up photography reveals that the menisci formed under the symmetrical blade have identical up- and downstream wetting angles (Figure S2, Supporting Information). The value of the wetting angles, however, could not precisely be determined from these pictures. Thus, contact angles of the solutions on glass were determined and then used as meniscus wetting angles according to Davis et al.,<sup>[25]</sup> assuming  $\theta_1$  and  $\theta_2$  to be equal (Figure S3, Supporting Information). A summary of the fixed parameters used for the blade-coating model is shown in



**Table 1.** Fixed parameters used for the blade-coating model, used to describe the PCBM and perovskite coating process. Variable coating parameters are detailed in the text and figures.

Parameter	Symbol	Unit	Values for	
			PCBM	Perovskite
Solvent	-	-	-	-
name(s)	-	-	СВ	DMF:NMP
Ratio	-	1	1	91:9
Viscosity	μ	cPa	0.806	0.9926
Surface tension	γ	$\rm mNm^{-1}$	33.5	37.46
Density	$ ho_{S}$	${\rm gcm^{-3}}$	1.11	0.9524
Solution	-	-	-	-
Concentration	С	${ m mgmL}^{-1}$	15	771.65
Contact angle	$\theta_1 = \theta_2$	degrees	12	28
Dry film density	$ ho_{ m F}$	${\rm gcm^{-3}}$	1.5 <sup>[30]</sup>	4 <sup>[31–33]</sup>
Model constant	<i>C</i> <sub>1</sub>	-	1.53	3.1

**Table 1**. With these, a good correlation between theoretical data and experimentally determined perovskite dry film thickness was observed (**Figure 3**).

The coating solution, when applied into the blade gap, is distributed along the gap and wetting the sides of the blade. Additional solution volume or gap size reduction increases the height H of the outer menisci, resulting in thicker layers for smaller gap sizes, which was experimentally verified. It is worth to note that during the movement of the blade, no change in meniscus height H were observed. The influence of the coating speed on the perovskite dry film thickness could not be investigated experimentally, as only for the smallest possible coating speed of the used coating setup, homogeneous perovskite films could be obtained. Nevertheless, a good agreement between the model and experiment was found for this single data point as well. To fully proof the model also with respect to coating speed influence, it was applied to the deposition of PCBM as well. A good agreement between model and experiment was found for variation of all controllable coating parameters, including a coating speed variation from 5 to  $30 \text{ mm s}^{-1}$  (Figure S4, Supporting Information), highlighting the applicability of the model in realizing homogeneous perovskite and transport layers with desired film thickness.

# 2.3. Blade-Coated Solar Cells and Tuning of the Perovskite Crystallinity

Perovskite solar cells with 1.1 cm<sup>2</sup> active area were fabricated by blade coating of the transport and absorber layers. Before optimization, these cells had power conversion efficiencies of up to 10.4%, significantly lower than their spin-coated, small-area counterparts (**Figure 4**).

Even though homogeneous, mirror-like absorber layers could be obtained, it was suspected that the perovskite layer crystallized differently under gas quenching instead of when an antisolvent step was applied. XRD analysis of spin- and blade-coated perovskite layers revealed a lower crystallinity of the blade-coated layers, as judged by the slightly higher full width at half maximum values of the detected peaks (**Figure 5**).

To further investigate this, Williamson–Hall's analysis<sup>[26]</sup> of the XRD data (Figure S5, Supporting Information) was conducted and revealed a significantly reduced crystallite size in the blade-coated layer (**Figure 6**). At the same time, lattice strain was found to be enhanced in spin-coated layers ( $\varepsilon = 1.4 \times 10^{-3}$ ) in comparison with blade coated layers (lattice strain  $\varepsilon = 0.2 \times 10^{-3}$ ) (Figure S6, Supporting Information), resulting in similar peak shape in the diffractograms. Additional SEM of the perovskite film surface revealed reduced grain size (**Figure 7**) for the blade-coated sample. The observed low solar cell performance was speculated to be caused by the inferior crystallinity, potentially causing increased nonradiative recombination at grain boundaries<sup>[19]</sup> and hindered charge carrier transport.

To improve the crystallinity of blade-coated perovskite films, lead(II) thiocyanate Pb(SCN)<sub>2</sub> was added to the precursor solution in concentrations of 0.5-2.0 mol% with respect to perovskite. This additive was demonstrated in literature for improvement of the crystallinity and, subsequently, the performance of spin-coated perovskite layers.<sup>[27,28]</sup> By addition of Pb(SCN)<sub>2</sub>, the crystallite size in the blade-coated layers could be increased from  $\approx 50$  nm (no Pb(SCN)<sub>2</sub>) to  $\approx 200$  nm (2% Pb(SCN)<sub>2</sub>) (Figure 6). The corresponding SEM images showed an increasing grain size for increasing additive



**Figure 3.** Perovskite dry film thickness as a function of a) applied solution volume, b) coating gap, and c) coating speed. Black curves: Calculated values from the applied blade-coating model. Red data points: Experimentally determined values. If not varied, the volume is 30  $\mu$ l, coating gap is 50  $\mu$ m, and speed 5 mm s<sup>-1</sup>.





Figure 4. Figures of merit of 1.1 cm<sup>2</sup> blade-coated perovskite solar cells as a function of Pb(SCN)<sub>2</sub> concentration in the precursor solution.



**Figure 5.** Diffractograms of spin-coated (dashed line) and blade-coated (solid line) perovskite films processed from precursor solutions containing 0.0–2.0 mol% Pb(SCN)<sub>2</sub>. Spectra were normalized to the first perovskite peak.

concentration (Figure 7). Both the crystallite and grain enlargement were most pronounced when the  $Pb(SCN)_2$  concentration was increased from 0% to 0.5 mol%, but no saturation in size was observed at concentrations as high as 2 mol%.

Upon these results, blade-coated solar cells based on perovskite absorber layers with and without Pb(SCN)<sub>2</sub> were elaborated. It was found that solar cell efficiency was not optimal for the highest crystallite/grain size but peaked at an additive concentration of 1 mol% (Figure 4). XRD analysis of the perovskite layers



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Figure 6. Crystallite size in blade-coated (red) and spin-coated (black)  $FA_{0.83}Cs_{0.17}PbI_3$  layers as determined by Williamson–Hall's analysis as a function of Pb(SCN)<sub>2</sub> concentration in the precursor solution.

revealed a strong presence of the nonphotoactive  $PbI_2$  phase at increasing  $Pb(SCN)_2$  concentration (Figure 5), ultimately limiting device performance. A similar effect was described in literature for spin-coated perovskite layers.<sup>[27]</sup> Optimization of the cell fabrication process with 1 mol% additive concentration resulted in a power conversion efficiency of 16.2% (nonstabilized) and 15.7% (stabilized during maximum power point tracking), respectively, for 1.1 cm<sup>2</sup> solar cells with blade-coated absorber and transport layers (Figure 8).







Figure 7. SEM surface images of a) spin- and b-e) blade-coated perovskite films. Concentrations of Pb(SCN)<sub>2</sub> in the perovskite precursor solution are indicated.



Figure 8. a) Current–voltage curves of an optimized 1.1 cm<sup>2</sup> blade-coated perovskite solar cell. Black: forward scan and red: reverse scan. b) Evolution of the power conversion efficiency of the same cell under maximum power point tracking for 5 min.

#### 2.4. Blade-Coated Perovskite Modules

With the optimized parameter set derived from fabrication of blade-coated solar cells, perovskite modules with 15 cells monolithically connected in series were realized. The applied volume of the blade-coated solutions was increased with respect to the larger coating area. Perovskite modules were first characterized by recording their current–voltage curves and revealed an open-circuit voltage  $V_{OC} = 14.4 \text{ V}$ , short-circuit current  $I_{SC} = 92.4 \text{ mA}$ , and a fill factor FF = 61.2%, resulting in a power conversion efficiency  $\eta = 12.4\%$  on the active area of 66 cm<sup>2</sup> (geometrical fill factor [GFF]: 69%) (Figure 9). It is worth to note that the GFF is relatively low due to the used solvent-assisted mechanical patterning. With laser patterning, for example, significantly higher GFFs can be achieved.<sup>[29]</sup> The fabricated modules featured low hysteresis and were found to be stable during continuous illumination and maximum power point tracking for 5 min, during which the stabilized efficiency increased to  $\eta = 12.6\%$  (Figure 9).

Table 2 and Figure 10 show the results of the best perovskite solar cells and modules fabricated in this work and illustrate the observed losses during upscaling. The open-circuit voltage only slightly decreased when moving from small-area spin-coated cells to larger-area blade-coated modules, indicating that no voltage losses were caused by, for example, reduced parallel resistance due to local-coating defects. Short-circuit current and especially FF, however, were significantly reduced, ultimately limiting device performance. Correspondingly, only a few minor local shunts could be identified in DLIT images under forward bias (Figure 11), whereas the overall current flow is distributed rather inhomogeneously. The dark areas in the DLIT image reveal areas of reduced current flow. During perovskite coating, additional solution was added by a pipette every 5 cm. Especially in these regions, current flow was found to be





Figure 9. a) Current-voltage curves of a blade-coated perovskite module. Black: forward scan and red: reverse scan. b) Evolution of the power conversion efficiency of the same module under maximum power point tracking for 5 min.

**Table 2.** Summary of results of the best perovskite devices fabricated in this work. Values in brackets indicate relative values in comparison with spincoated small-area devices. For better comparison, module  $V_{OC}$  and  $J_{SC}$  values were calculated by division and multiplication of the module measurement values by the number of serially interconnected cells, respectively.

Parameter	0.09 cm <sup>2</sup>	1.1 cm <sup>2</sup>	66 cm <sup>2</sup>
PCE [%]	18.1	16.2%	12.4%
	(100%)	(89.5%)	(68.5%)
V <sub>oc</sub> [V]	0.997	0.993	0.960
	(100%)	(99.6%)	(96.3%)
J <sub>SC</sub> [mA/cm <sup>2</sup> ]	23.6	21.6	21.0
	(100%)	(91.5%)	(89.0%)
FF	0.77	0.76	0.61
	(100%)	(98.7%)	(79.2%)



**Figure 10.** Power conversion efficiencies of spin- (black) and blade (red)coated perovskite devices fabricated in this work. Dots: Stabilized efficiency after maximum power point tracking. Triangles: Efficiency determined from current–voltage curves recorded in forward (open symbols) and reverse (filled symbols) sweep direction.

lower, possibly due to a larger perovskite layer thickness and correspondingly higher resistance.

As the  $J_{SC}$  of the modules, which is limited by the cell with lowest  $J_{SC}$  in the case of series connection, was comparable with (single) blade-coated solar cells, these dark regions could be attributed to module areas with increased series resistance, rather than larger-area defects of individual solar cells. Under illumination and short-circuit conditions, the photogenerated charge carriers can still be extracted in these regions but in the DLIT experiment, the current flow is according to the least resistance and therefore much less homogeneous. This interpretation is also supported by the limited FF of the module (see Table 2). Further improvement of the coating quality is expected by modification of the blade-coating process or for example slot-die coating, in which additional coating solution is continuously fed during the process.

#### 3. Conclusion

In summary, after optimization of a perovskite composition suitable for blade coating with the used setup, methylammoniumfree perovskite modules were fabricated by blade coating all solution-processable layers without the usage of an antisolvent. A good correlation between experimental results and a model for the blade-coating process was found, allowing for prediction of layer thicknesses and thus easing the optimization of the module fabrication process. The modules had a stabilized efficiency of 12.6% on an active area of  $66 \text{ cm}^2$ , in comparison with 18.1% small-area spin-coated devices and larger-area blade-coated solar cells with an efficiency of 16.2%. The observed losses during scale-up can clearly be correlated with decreases in the current density and FF. DLIT imaging correspondingly reveals only few shunts on the module area, but inhomogeneities of the current flow, indicating a suboptimal coating quality not observable by eye. While using a coating process even more suited for largerarea coating from solution (e.g., slot-die coating) is expected to result in better coating quality and module performance, the presented work demonstrates the feasibility of the approach to realize large-area, methylammonium-free perovskite modules from solution, without the need for antisolvents.







Figure 11. a) Photographic image and b) DLIT images of a blade-coated perovskite module.

#### 4. Experimental Section

*Materials*: Cesium iodide (CsI), lead(II) thiocyanate (Pb(SCN)<sub>2</sub>), and bathocuproine (BCP) were purchased from Sigma-Aldrich. [6,6]-phenyl-C61-butyric acid methyl ester (PCBM) was purchased from Solenne BV. [2-(3,6-dimethoxy-9H-carbazol-9-yl)ethyl]phosphonic acid (MeO-2Pacz) and lead(II) iodide (PbI<sub>2</sub>) were purchased from Tokyo Chemical Industry. Formamidinium iodide (FAI) was purchased from Dyenamo. Other chemicals were purchased from Sigma-Aldrich. All chemicals were used as received without further purification.

Fabrication of Solar Cells with Spin-Coated Layers: Prepatterned ITO glass substrates (2.5  $\times$  2.5 cm<sup>2</sup>, sheet resistance 10  $\Omega$ , Kintec Hong Kong) were cleaned twice and successively in acetone, isopropanol, and deionized water under ultrasonic agitation for 5 min, blow dried with nitrogen, and UV-ozone treated for 20 min. All subsequent steps were conducted in a nitrogen-filled glovebox. A thin layer of MeO-2PACz was spin coated from its solution (1 mM in EtOH) for 30 s at 3000 rpm and annealed at 100 °C for 10 min. For the perovskite precursor solution, PbI<sub>2</sub>, FAI, and CsI were mixed stoichiometrically to result in the composition  $FA_{1-x}Cs_xPbI_3$ , with x = 0, 0.04, 0.09, or 0.17. Perovskite layers were deposited by spin coating a 1.3 M solution in DMF:NMP (4:1) at 1500 rpm for 10 s, followed by a second step at 5000 rpm for 20 s. 10 s before the end of the spincoating process, 300 µL of chlorobenzene was dropped onto the substrate and the substrates were annealed at 130 °C for 30 min. A PCBM layer was deposited on top via spin coating at 4000 rpm for 30 s from a 10 mg mL<sup>-1</sup> solution in chloroform. BCP was deposited by spin coating a 0.5 mg  ${\rm mL}^{-1}$ solution in 2-propanol for 60 s at 4000 rpm. The cells were completed by thermal evaporation of 100 nm-thick aluminum back electrodes through a shadow mask, defining the cells' active area of 0.09 cm<sup>2</sup>.

Fabrication of Solar Cells and Modules with Blade-Coated Layers: Prepatterned ITO substrates ( $5 \times 5 \text{ cm}^2$ , sheet resistance  $10 \Omega$ , Kintec Hong Kong) were prepared as described earlier. To reduce series resistance losses, 5 nm chromium and 100 nm gold were thermally evaporated outside of the solar cells' active areas to support the ITO electrode. MeO-2PACz was blade coated from 80 µL solution (0.1 mg mL<sup>-1</sup> in EtOH) using a universal film applicator (MTUA 3000-100, mtv Messtechnik) set to a gap size of 100 µm and moved at a speed of 5 mm s<sup>-1</sup> by a motorized coating table (ZAA 2300, Zehntner) set to a temperature of 30 °C. An air blade (Silent X-Stream, 6", Nexflow) fed with nitrogen at a pressure of 10 kPa was used parallel to the coating. The films were annealed at 100 °C for 10 min. A FA<sub>0.83</sub>C5<sub>0.17</sub>Pbl<sub>3</sub> solution (1.3 M in DMF:NMP 91:9) with added Pb(SCN)<sub>2</sub> (1 mol% with respect to the perovskite concentration) was blade coated with the same setup. The gap size was 100 µm, the solution volume 30  $\mu$ L, the coating speed 5 mm s<sup>-1</sup>, the table temperature 30 °C, and the nitrogen pressure 50 kPa. Substrates were annealed at 130 °C for 30 min, before PCBM was blade coated from a 15 mg mL<sup>-1</sup> solution in chlorobenzene. The gap size was 100  $\mu$ m, the solution volume 100  $\mu$ L, the coating speed 10 mm/sec, the table temperature 40 °C, and the nitrogen pressure 10 kPa. BCP was blade coated from a 0.5 mg mL<sup>-1</sup> solution in 2-propanol with the same parameter set, but the solution volume was 60  $\mu$ L. Finally, 100 nm-thick aluminum was thermally evaporated through a shadow mask, defining the active area of 1.1 cm<sup>2</sup> for single solar cells.

For perovskite modules, identical ITO glass but with dimensions  $21 \times 14 \text{ cm}^2$  was used and treated as describe earlier. The parameters used for blade coating individual layers were identical to the ones used for solar cell fabrication, but the volumes of the coating solutions were adapted to account for the larger-area substrates. The additional solution volume was applied with a pipette every 5 cm along the coating direction.

After coating the BCP layer, the organic and perovskite layers were partially removed by solvent-assisted mechanical scribing (thread wiping) corresponding to the P2 scribe, for subsequent serial interconnection of the individual solar cells. The module was completed by thermal evaporation of a 150 nm-thick aluminum electrode through a shadow mask, during which the 15 solar cells were monolithically interconnected.

Device Characterization: Current–voltage characteristics of solar cells and modules were recorded with a Keithley 2400 source meter by illuminating the samples with a Newport SP94063A sun simulator and a mismatch corrected intensity of "1 sun" to mimic AM1.5 G conditions. *I*–V curves were recorded in both scan directions in the voltage range between -0.1 and +1.5 V for solar cells and between -1 and +17 V for modules, both at a scan rate of 185 mV s<sup>-1</sup>. Using the same measurement setup, stabilized device performance was determined by tracking the maximum power point for 5 min.

Surface roughness and layer thicknesses were determined by profilometry, using a Dektak 150 Surface Profiler from Veeco Instruments.

Photoluminescence spectra were measured using an Andor Shamrock 193i Czerny-Turner spectrograph together with an Andor iDus (DU416A-LDC-DD) silicon array detector. The excitation wavelength of 532 nm was provided by a frequency-doubled Nd-YAG laser from Pegasus laser systems (Pluto, P532.400). The laser beam was widened before the sample, so that full illumination of the sample could be achieved. The illumination intensity was chosen such that a perovskite solar cell in the sample plane showed a short-circuit current density  $J_{SC}$  corresponding to "1 sun" illumination.  $\omega$ -2 $\theta$  scans were conducted in Bragg–Brentano geometry on an Empyrean diffractometer from Malvern Panalytical. Ni-filtered Cu K $\alpha$  radiation, produced from a copper anode powered with 45 kV and 40 mA, was



used for the diffraction experiments. Measurements were carried out in 2 $\theta$  range from 10° to 65° with a step size of 0.055° and a dwell time of 0.235 s. SEM images were recorded using a field-emission scanning microscope (Auriga 60 workstation, Zeiss) equipped with a patented Gemini electron gun. Acceleration voltage was set to 5 kV, and the working distance was  $\approx$ 5 mm. Depending on the magnification level, either an in-lens detector or an Everhart–Thornley detector was used for image generation. DLIT was conducted on a custom-made tool by IRCAM GmbH, Germany. The camera contained an InSb-based midwave infrared focal plane array (MWIR FPA) detector, sensitive to IR radiation in the range of 1.5–5  $\mu$ m, and was actively cooled, resulting in low thermal noise and a temperature resolution limit of <20 mK. The solar cell was subjected to a periodical bias voltage of a Toellner four-quadrant power supply and the IR camera signal was analyzed using a lock-in amplifier to further increase the signal-to-noise ratio.

## **Supporting Information**

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

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### **Conflict of Interest**

The authors declare no conflict of interest.

#### Data Availability Statement

Data available on request from the authors.

#### Keywords

blade coating, cesium, formamidinium, modules, perovskites, upscaling

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