Design of perovskite thermally co-evaporated highly efficient mini-modules with high geometrical fill factors

Li, Jia; Dewi, Herlina Arianita; Wang, Hao; Lew, Jia Haur; Mathews, Nripan; Mhaisalkar, Subodh; Bruno, Annalisa

2020

Li, J., Dewi, H. A., Wang, H., Lew, J. H., Mathews, N., Mhaisalkar, S., & Bruno, A. (2020). Design of perovskite thermally co-evaporated highly efficient mini-modules with high geometrical fill factors. *Solar RRL*, 4(12), 2000473-. doi:10.1002/solr.202000473

<https://hdl.handle.net/10356/146266>

<https://doi.org/10.1002/solr.202000473>

This is the accepted version of the following article: Li, J., Dewi, H. A., Wang, H., Lew, J. H., Mathews, N., Mhaisalkar, S., & Bruno, A. (2020). Design of perovskite thermally co-evaporated highly efficient mini-modules with high geometrical fill factors. *Solar RRL*, 4(12), 2000473-. doi:10.1002/solr.202000473, which has been published in final form at <https://doi.org/10.1002/solr.202000473>. This article may be used for non-commercial purposes in accordance with the Wiley Self-Archiving Policy [<https://authorservices.wiley.com/authorresources/Journal-Authors/licensing/self-archiving.html>].

Design of perovskite thermally co-evaporated highly efficient mini-modules with a high geometrical fill factors

Jia Li, ^a Herlina Arianita Dewi, ^a Hao Wang, ^a Jia Haur Lew, ^a Nripan Mathews, ^{a,b} Subodh Mhaisalkar, ^{a,b*} Annalisa Bruno ^{a*}*

Dr. J. Li, Dr. H. Dewi, Dr. H. Wang, Dr. H. Dewi, Mr. J. Lew, Dr. A. Bruno

Energy Research Institute @ NTU
Nanyang Technological University
Research Techno Plaza, X-Frontier Block, Level 5, 50 Nanyang Drive 637553,
Singapore
E-mail: Annalisa@ntu.edu.sg

Prof. S. Mhaisalkar, Prof. N. Mathews

School of Materials Science and Engineering
Nanyang Technological University
50 Nanyang Avenue 639798, Singapore

Keywords: Co-evaporated perovskite, MAPbI₃, perovskite solar cells, perovskite mini-modules, laser etching

Perovskite solar cells (PSCs) have emerged as a promising technology for next-generation photovoltaics thanks to their high power-conversion-efficiency (PCE). Scaling up PSCs using industrially compatible processes is a key requirement to make them suitable for a variety of applications. Here, we have developed large-area PSCs and perovskite solar modules (PSMs) based on co-evaporated MAPbI₃ using optimized structures and active area designs to enhance PCEs and geometrical fill factors (GFF). Small-area co-evaporated PSCs (0.16 cm²) achieved PCE over 19%. When the PSCs are scaled-up, the thin films high quality allows them to maintain consistent V_{oc} and J_{sc} while their fill factors (FF), which depend on the substrate sheet resistance, are substantially compromised. However, PSCs with active areas from 1.4 cm² to 7 cm² showed a substantially improved FF when rectangular designs with optimized length to width ratios were used. Reasoning these results in the PSM design with optimal subcell' size and for specific dead areas, we demonstrate a 6.4 cm² PSM with a record 18.4% PCE and a GFF of ~ 91%. Combining the high uniformity of the co-evaporation deposition with active areas design it is possible to scale up 40 times the PSCs with PCE losses smaller than 0.7% (absolute value).

1. Introduction

In slightly more than 10 years, perovskite solar cells (PSCs) have achieved record power conversion efficiency (PCE) of 25.2%^[1] presently demonstrated over an active area much smaller than 1 cm² and a continued focus on improving the operational stability^[2-5]. The rapid progress has triggered the interest of transferring the existing technology from small area PSCs into large-area perovskite solar modules (PSMs), necessary for industrial expansion. To meet the requirements of high quality and large area uniformity for perovskite films, several deposition methods based on solution, vacuum, and solution-vacuum hybrid processes have been developed and optimized.^[6-9] The recent works based on solution processes, such as by spin-coating^[6, 10-19], spray-coating^[7], blade coating^[8, 20-23], slot-die coating^[24, 25], softcover^[26, 27] and screen printing^[28] the devices have reached PCEs ranging from 10-18% over areas reaching 100 cm². Some strategies such as solvent optimization^[23], additive engineering^[29], and substrate temperature control^[30] have been adopted to obtain large crystal size and pinhole-free film over a large area and thus significantly improving the PCE in the solution-based processes. Meanwhile, some groups have also explored the solvent-free route^[31] or solution-vacuum hybrid methods, such as the hybrid chemical vapor deposition and cation exchange (HCVD-CE) method^[32, 33]. At the moment, vacuum deposition methods such as co-evaporation^[9, 34, 35] represent the most viable option for integration with the existing industrial processes. The first co-evaporated perovskite solar cells were reported with the PCE over 15% in 2013^[9]. A few years later, a few groups reported a fully evaporated perovskite solar cells based on both nip and pin structure, reaching the PCE around 20%^[34-37]. Very recently, our group also demonstrated the co-evaporated PSCs with a champion PCE of 20.28% for 0.16 cm² and 19% for 1 cm² and the first co-evaporated perovskite solar modules (PSM) with a PCE of 18.1% over an active area of 21 cm², proving the excellent scalability of co-evaporation methods^[34]. These promising results triggered the interest of further studying optimized scaling-up strategy for PSCs with minimum losses of fill factor and fabricating PSMs with high geometrical fill factor (GFF) to maximize the active areas.

The GFF is defined as the ratio between the active area and the total area. High GFF has been previously demonstrated. [38-40]

In this paper, co-evaporated methylammonium lead iodide (MAPbI₃) absorber thickness and PSCs active area sizes and geometry have been optimized, achieving V_{oc} and J_{sc} losses smaller than 2% when the sizes were scaled-up from 0.16 cm² up to 7 cm². We further prove that scaling up the PSCs using a rectangular shape and maintaining a constant width, it is possible to obtain minimal PCE variations. Finally, we show a laser-etched mini-module with a proper sub-cells design reached a PCE of 18.4% over an active area ~6.4 cm² and a GFF with ~90.7%.

2. Results

MAPbI₃ thin films of different thicknesses have been co-evaporated using our optimized growth conditions [34] by controlling the total growth time. Process details are reported in the Material and methods section. The cross-sectional FESEM images three MAPbI₃ films, **Figure S1**, show compact films of 660 nm, 780 nm, and 1000 nm thick, and their respective UV-visible optical absorption spectra are shown in **Figure S2**.

PSCs in a planar n-i-p architecture have been fabricated on fluorine-doped tin oxide (FTO) glass substrates using the three MAPbI₃ active layer thicknesses. Moreover, a SnO₂/PCBM bilayer and a Spiro-OMeTAD layer were used as electron and hole transport layers respectively while an evaporated gold layer acted as the top electrode, **Figure 1a**. The PSCs' performance distributions over 10 PSCs for each condition show their high reproducibility (**Figure 1b-e**) over different batches. IPCE spectra of PSCs with different thickness are reported in **Figure S3**.

PSCs with MAPbI₃ thickness of 780 nm achieve the highest PCE of 19.1% and a consistently higher PCE with an average of 18.6%, **Table 1**, which guarantees the best-operating conditions.

Using the perovskite growth conditions and the device architecture optimized over the small area PSCs (0.16 cm²), we have scaled-up the PSCs by rationally designing their

active areas. Usually, the PSCs' PCEs drop dramatically when their sizes are scaled above 1 cm² [34, 41] due to the high sheet resistance of transparent conductive oxide glass (TCO) substrate (usually 10-15 ohm/square) which cause a substantial drop in the FF. Indeed, large-area PSCs with a square-shape active area show a dramatically drop of FF while J_{sc} and V_{oc} do not vary significantly, **Figure 2**, resulting in PCE dropping from ~ 17% to 10 % when the active area increased from 1 cm² (1 cm x 1 cm) to 6.25 cm² (2.5 cm x 2.5 cm), **Table 2**.

Very interestingly, the very small variation of V_{oc} observed in our PSCs when the sizes are scaled up, is mostly related to the masking procedure as described previously. ⁴² Indeed, the estimated V_{oc} losses are described by

$$\Delta V_{oc} = \frac{nKT}{q} \ln X; \text{ with } X = \frac{A_{Aperture}}{A_{Device}}$$

where n is the diode ideality factor and kT/q is the thermal voltage. $A_{Aperture}$ is defined as the area defined by the mask and A_{Device} is the total device area.

For our PSCs, the estimated ΔV_{oc} values reported in **Table 3** are well in agreement with the V_{oc} variations measured when the PSCs sizes are scaled up from 0.16 cm² to 4 cm², **Table 1**, and **Table 2**.

The best strategy to overcome the limitations of TCO's high sheet resistance is to design the active area which minimizes the device length (L) in the charge transport direction. Rectangular shapes with a high length (L) to width (w) ratio are the best to minimize the total sheet resistance as compared to the square shape counterpart (**Figure 2** and **Figure 3**). ^[43] The TCO (FTO in our case) resistance can be simply expressed as:

$$R_{FTO} = R_{Sheet} \frac{w}{L}$$

R_{sheet} is the sheet resistance of the FTO glass, w is the width in the charge transport direction; L is the length across the orthogonal direction (Figure 2a). The FTO used in this work has a sheet resistance around 15 ohm/square. We kept the PSCs length (L) to 7.0 cm and varied the width (w) from 0.2 to 1.0 cm, **Figure 3a**, achieving active areas ranging from 1.4 cm² to 7 cm². The J-V curves, **Figure 3b**, of the PSCs with different

sizes show that both V_{oc} and J_{sc} are fairly constant during the scaling-up while the FF drops due to the increase of the FTO resistance, R_{FTO} , **Table 4**. The 1.4 cm² and 2.8 cm² PSCs reached a PCE of 18.1% and 17.1% with FF of 71% and 67.6% while the 4.9 cm² and 7 cm² PSCs achieved 15.8% and 15.0% respectively and FF of 63.6 % and 61%.

To further illustrate the drop in FF , another set of experiments included PSCs with a constant width of 0.65 cm and length varying from 2 to 7 cm, **Figure 4a**. The J-V curves, **Figure 4b**, and photovoltaic parameters, **Table 5**, of the PSCs with different sizes show again that J_{sc} varies less than 2% and V_{oc} less than 1%, while this time also the FF just dropped around 4% while L increased. As a consequence, the PCE just varied from 15.9 to 15.4% when the PSCs active area was increased from 1.3 cm² to 4.55 cm². The results confirmed that both the active area design and the high uniformity of co-evaporated perovskite film can guarantee minimal power efficiency losses during scaling up.

To further scale up the PSCs to reach high PCE over larger areas is it necessary to design PSM with several subcells in series. PSMs design needs to consider the formation of series-connected sub-PSCs with a reduced subcells' width size and keeping the dead area between each sub-PSC minimal. A balance between sub-PSCs dimensions, dead areas (the connection area), and the number of series-connections need to be properly assessed to design high-power PSMs with high GFF. Indeed, over a specific area, increasing the number of subcells with small sizes can guarantee a high FF but it will also increase the number of connections thus reducing the GFF. At the same time, minimizing the total dead area can increase the GFF, which can be realized by reducing the number of connections (reducing the number of sub-cells over a specific area) and by reducing the dead area width between every two sub-cells. To find an optimal and suitable sub-cell size to take into account both small subcells' width and a small dead area, we have compared the estimated output power of a model 7x7 cm² PSC with subcells' widths ranging from 0.2 to 2 cm and different dead areas, based on previous results on rectangular-shaped PSCs. Indeed, we have used the subcells' PCEs in **Table 2** (for the 2 cm width PSCs we assumed the same PCE as the 2 cm x 2 cm

PSCs and we have demonstrated that the length, L , will not affect the PCE significantly) and in **Table 4** for all the others width PSCs. The calculated output power as a function of the subcells' width, for dead areas' width varying between 0.04 and 0.25 cm is reported in **Figure 5**. The output power and the PSM parameters are also reported in **Table 6** for the two typical dead area width of a full laser-etched PSM (0.06 cm) and a partially manually etched method (0.25 cm) used in our previous work³⁴. These results show that when the dead area width is 0.06 cm, PSMs delivers the highest output power with a subcells' width of 0.7 cm. On the other hand, when the dead area width is 0.25 cm, the PSMs show the highest output power with a subcells' width of 1 cm. Fully laser-etched PSM (FLE-PSM) can achieve high GFF, maximizing the total active area and by reducing the dead areas.

According to the calculation, we have designed a PSMs with a subcells' width of 0.65 cm since our laser etching scribing process could guarantee a dead area width of 0.06 cm which can achieve a GFF of 90.7%. This is a substantial improvement as compared to our previously demonstrated partially laser-etched PSM which instead reached a GFF of 72%^[34]. The sub-cells were firstly separated by laser scribing the FTO substrates (process 1, P1). Subsequently, the electron transport layer, perovskite, and hole transport layer were deposited and the sub-PSCs were laser-etched again to isolate the interlayers (P2). Finally, after the deposition of the gold electrode, the last laser etching process was performed to delimit the sub-cells (P3). The cross-sectional architecture schematic of the mini-module design was illustrated in **Figure 6a**. The width of P2 and P3 laser etching lines are $\sim 90\ \mu\text{m}$ and $\sim 60\ \mu\text{m}$ which can guarantee a total small dead area as small as $600\ \mu\text{m}$. The optical microscopy image of P1, P2, and P3 lines are shown in **Figure 6b** and the SEM images of P1 and P3 lines are in **Figure S4**. By using this method, an FLE-PSM including 7 sub-cells, with a rectangular shape of $0.59\ \text{cm} \times 1.55\ \text{cm}$, achieved a PCE of 18.4% with over an active area of $6.4\ \text{cm}^2$ and an FF of 75.2%. The PSM J-V curve is reported in **Figure 6c** and the photovoltaic parameters in **Table 7**. This approach shows that 96.3% of PCE can be maintained when the PSCs area is scaled up 40 times, from $0.16\ \text{cm}^2$ to $6.4\ \text{cm}^2$. Moreover, the FLE-PSMs showed that more than 80% of the initial PCE can be retained under

continuous 1 sun illumination at high relative humidity (RH) ~70% over 6 hours, **Figure 6d**. The FLE-PSMs also maintained over 80% of the initial PCE value for over 20 days when stored at room temperature and in a 30% RH environment, **Figure S5**. All the photovoltaic measurements were instead performed at 70% RH.

3. Conclusion

In conclusion, the design of the large area PSCs has been assessed to mitigate the PCE loss due to FTO sheet resistance. Indeed, we have realized co-evaporated MAPbI₃ PSCs with active areas ranging from 0.16 cm² up to 7 cm² achieving PCEs ranging from 19% to 15%. Moreover, using a proper cell and module design based on the optimization of the subcells' size and the dead areas dimensions we prove a PSM with a 6.4 cm² active area, and 90.7% GFF achieved a PCE of ~18.4 %. These results prove that scaling up 40 times the active areas of our thermal evaporated PSCs with a suitable PSM design PCE losses are as low as just 0.7% (absolute). Moreover, further optimization of the laser patterning and interconnection optimization would make it possible to reach high PCE on an even larger area.

4. Experimental Section

Device fabrication

FTO glass (15 ohm/square) were ultra-sonicated in soap (Decon 90), deionized water, and subsequently ethyl alcohol respectively for 15 min and then dried in nitrogen flow. Before use, they are UV-ozone treated (Novascan PSD Pro Series) for 20 min at room temperature. A layer of SnO₂ was then spin-coated on the substrates by using a two steps spin-coating program (1500 r.p.m. for 10 seconds and 5000 r.p.m. for 10 seconds) from anhydrous ethanol solution containing 0.05M SnCl₂·2H₂O (Sigma-Aldrich) and

0.001M InCl_3 . After spin-coating, the substrate was heated at 80°C for 10 min and subsequently heated to 180°C for 60 min to form SnO_2 . The PCBM interlayer was then spin-coated at 3000 r.p.m. from a 3 mg/ml PCBM/chlorobenzene solution. Then the substrates are transferred into the evaporation chamber and pumped down to 10^{-6} torr before evaporating perovskite. The perovskite layer was co-evaporated from PbI_2 (TCI) and MAI (Lumtech), the temperature of PbI_2 and MAI are fixed at 260°C and 100°C respectively. After the co-evaporation finishing, the MAPbI_3 films were heated on a 100°C for 30 minutes. Spiro-OMeTAD was spin-coated at 4000 r.p.m. on perovskite after the samples are cooled down to room temperature. The Spiro-OMeTAD solution is prepared by dissolving 72.3 mg of Spiro-oMeTAD powder (Lumtec) in 1 ml of chlorobenzene and stirring it until it is uniform. Add 28.5 μl of TBP solution and 17.5 μL bis(trifluoromethane)sulfonamide lithium salt solution (520mg/ml Li-TFSI in ACN) and stir for 10min. At last, a 100 nm gold was thermally evaporated on the samples with a shadow mask. For rectangle PSCs, we use a big square substrate to finish all the process and cut them into rectangular shape at last.

For fully laser etching perovskite minimodules (FLE-PSMs), all the etching processes are done by an Nd: Yag fiber laser ($\lambda=1064$ nm, maximum power is 50 W, 100 ns pulse duration). **P1**: After cleaning the FTO glass, they are separated into smaller rectangle strips with a width of 6.5 mm, the laser etching parameter is: 20 kHz, 40 w, 1000 mm/s raster speed. **P2**: After depositing SnO_2 /PCBM/ MAPbI_3 /Spiro-OMeTAD, another laser etching process was done near P1 with the parameter of 30 kHz, 2.5 w, 12500 mm/s raster speed. **P3**: A 100 nm gold was deposited and a final laser etching process was

carried out near P2 line with the parameter of 30 kHz, 4 w, 12500 mm/s raster speed.

For partially laser etching perovskite minimodules (PLE-PSMs), **P1** is the same as FLE-PSMs, **P2** is done by manually scribing, **P3** is done with 0.5 μm wide tape.

Characterization

The film morphology was characterized using Field emission Electron Microscopy (FESEM). Model number: JEJOL JSM-7600F. UV-vis absorption spectra and transmittance spectra were measured using a Hitachi U-3501 ultraviolet/visible/near-infrared spectrophotometer. The current density–voltage (J–V) characteristics curves were measured under simulated AM 1.5 sunlight (Newport 94043A) which is calibrated by a standard silicon solar cell. For the stability test, the devices are stored in a dry box with controlled relative humidity (RH) at around 30% and the J-V measured in RH of 70%

Supporting Information

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

Acknowledgements

This research is supported by the National Research Foundation, Prime Minister's Office, Singapore under Energy Innovation Research Program (Grant numbers: NRF2015EWT-EIRP003-004, NRF-CRP14-2014-03, Solar CRP: S18-1176-SCRP) and Intra-CREATE Collaborative Grant (NRF2018- ITC001-001).

Notes

Two of the authors, N.M. and S.M., are directors of Prominence Photovoltaics Pte Ltd, a perovskite solar cell commercialization company. The other authors declare no competing interests.

References

- [1] NREL. Best Research-Cell Efficiency Chart; U.S. Department of Energy; <https://www.nrel.gov/pv/cell-efficiency.htm>.
- [2] A. Kojima, K. Teshima, Y. Shirai, T. Miyasaka, *J. Am. Chem. Soc.* **2009**, *131*, 6050.
- [3] N. Arora, M. I. Dar, A. Hinderhofer, N. Pellet, F. Schreiber, S. M. Zakeeruddin, M. Grätzel, *Science* **2017**, *358*, 768.
- [4] S. Yang, S. Chen, E. Mosconi, Y. Fang, X. Xiao, C. Wang, Y. Zhou, Z. Yu, J. Zhao, Y. Gao, *Science* **2019**, *365*, 473.
- [5] Y. Wang, T. Wu, J. Barbaud, W. Kong, D. Cui, H. Chen, X. Yang, L. Han, *Science* **2019** *365*, 687.
- [6] T. Bu, X. Liu, Y. Zhou, J. Yi, X. Huang, L. Luo, J. Xiao, Z. Ku, Y. Peng, F. Huang, Y. B. Cheng, J. Zhong, *Energ. Environ. Sci.* **2017**, *10*, 2509.
- [7] J. H. Heo, M. H. Lee, M. H. Jang, S. H. Im, *J. Mater. Chem. A* **2016**, *4*, 17636.
- [8] M. Yang, Z. Li, M. O. Reese, O. G. Reid, D. H. Kim, S. Siol, T. R. Klein, Y. Yan, J. J. Berry, M. F. A. M. van Hest, K. Zhu, *Nat. Energy* **2017**, *2*, 17038.
- [9] M. Liu, M. B. Johnston, H. J. Snaith, *Nature* **2013**, *501*, 395.
- [10] G. S. Han, J. Kim, S. Bae, S.-H. Han, Y. J. Kim, O. Y. Gong, P. Lee, M. J. Ko, H. S. Jung, *ACS Energy Lett.* **2019**, *4*, 1845.
- [11] A. Agresti, S. Pescetelli, A. L. Palma, B. Martin-Garcia, L. Najafi, S. Bellani, I. Moreels, M. Prato, F. Bonaccorso, A. Di Carlo, *ACS Energy Lett.* **2019**, *4*, 1862;
- [12] T. Bu, J. Li, F. Zheng, W. Chen, X. Wen, Z. Ku, Y. Peng, J. Zhong, Y. B. Cheng, F. Huang, *Nat. commun.* **2018**, *9*, 4609.
- [13] J. Kim, J. S. Yun, Y. Cho, D. S. Lee, B. Wilkinson, A. M. Soufiani, X. Deng, J. Zheng, A. Shi, S. Lim, S. Chen, Z. Hameiri, M. Zhang, C. F. J. Lau, S. Huang, M. A. Green, A. W. Y. Ho-Baillie, *ACS Energy Lett.* **2017**, *2*, 1978.
- [14] M. Jaysankar, S. Paetel, E. Ahlswede, U. W. Paetzold, T. Aernouts, R. Gehlhaar, J. Poortmans, *Prog. Photovoltaics* **2019**, *27*, 733.
- [15] E. Calabrò, F. Matteocci, A. L. Palma, L. Vesce, B. Taheri, L. Carlini, I. Pis, S. Nappini, J. Dagar, C. Battocchio, T. M. Brown, A. Di Carlo, *Sol. Energ. Mater. Sol. C.* **2018**, *185*, 136.
- [16] J. Troughton, K. Hooper, T. M. Watson, *Nano Energy* **2017**, *39*, 60.
- [17] T. Bu, S. Shi, J. Li, Y. Liu, J. Shi, L. Chen, X. Liu, J. Qiu, Z. Ku, Y. Peng, J. Zhong, Y. B. Cheng, F. Huang, *ACS Appl. Mater. Inter.* **2018**, *10*, 14922.
- [18] Z. Huang, X. Hu, C. Liu, X. Meng, Z. Huang, J. Yang, X. Duan, J. Long, Z. Zhao, L. Tan, Y. Song, Y. Chen, *Adv. Funct. Mater.* **2019**, *29*, 1902629.
- [19] T. Bu, L. Wu, X. Liu, X. Yang, P. Zhou, X. Yu, T. Qin, J. Shi, S. Wang, S. Li, Z. Ku, Y. Peng, F. Huang, Q. Meng, Y. B. Cheng, J. Zhong, *Adv. Energy Mater.* **2017**, *7*, 1700576.
- [20] Y. Deng, X. Zheng, Y. Bai, Q. Wang, J. Zhao, J. Huang, *Nat. Energy* **2018**, *3*, 560;
- [21] M. Yang, D. H. Kim, T. R. Klein, Z. Li, M. O. Reese, B. J. Tremolet de Villers, J. J. Berry, M. F. A. M. van Hest, K. Zhu, *ACS Energy Lett.* **2018**, *3*, 322;
- [22] S. Razza, F. Di Giacomo, F. Matteocci, L. Cinà, A. L. Palma, S. Casaluci, P. Cameron, A. D'Epifanio, S. Licoccia, A. Reale, T. M. Brown, A. Di Carlo, *J. Power*

Sources **2015**, 277, 286.

- [23] Y. Deng, C. H. Van Brackle, X. Dai, J. Zhao, B. Chen, J. Huang, *Sci. adv.* **2019**, 5, eaax7537.
- [24] F. Di Giacomo, S. Shanmugam, H. Fledderus, B. J. Bruijnaers, W. J. H. Verhees, M. S. Dorenkamper, S. C. Veenstra, W. Qiu, R. Gehlhaar, T. Merckx, T. Aernouts, R. Andriessen, Y. Galagan, *Sol. Energ. Mater. Sol. C* **2018**, 181, 53;
- [25] L. Cai, L. Liang, J. Wu, B. Ding, L. Gao, B. Fan, *J. Semicond.* **2017**, 38, 014006.
- [26] F. Ye, W. Tang, F. Xie, M. Yin, J. He, Y. Wang, H. Chen, Y. Qiang, X. Yang, L. Han, *Adv. Mater.* **2017**, 29, 1701440.
- [27] J. He, E. Bi, W. Tang, Y. Wang, X. Yang, H. Chen, L. Han, *Nano-micro lett.* **2018**, 10, 49.
- [28] Y. Hu, S. Si, A. Mei, Y. Rong, H. Liu, X. Li, H. Han, *Sol. Rrl* **2017**, 1, 1600019.
- [29] Y. Deng, X. Zheng, Y. Bai, Q. Wang, J. Zhao, J. Huang, *Nat. Energy* **2018**, 3, 560.
- [30] H. C. Liao, P. Guo, C.-P. Hsu, M. Lin, B. Wang, L. Zeng, W. Huang, C. M. M. Soe, W.-F. Su, M. J. Bedzyk, M. R. Wasielewski, A. Facchetti, R. P. H. Chang, M. G. Kanatzidis, T. J. Marks, *Adv. Energy Mater.* **2017**, 7, 1601660.
- [31] H. Chen, F. Ye, W. Tang, J. He, M. Yin, Y. Wang, F. Xie, E. Bi, X. Yang, M. Gratzel, L. Han, *Nature* **2017**, 550, 92.
- [32] M. R. Leyden, Y. Jiang, Y. Qi, *J. Mater. Chem. A* **2016**, 4, 13125;
- [33] Y. Jiang, M. R. Leyden, L. Qiu, S. Wang, L. K. Ono, Z. Wu, E. J. Juarez-Perez, Y. Qi, *Adv. Funct. Mater.* **2018**, 28, 1703835.
- [34] J. Li, H. Wang, X. Y. Chin, H. A. Dewi, K. Vergeer, T. W. Goh, J. W. M. Lim, J. H. Lew, K. P. Loh, C. Soci, T. C. Sum, H. J. Bolink, N. Mathews, S. Mhaisalkar, A. Bruno, *Joule* **2020**, 4, 1035;
- [35] C. Momblona, L. Gil-Escrig, E. Bandiello, E. M. Hutter, M. Sessolo, K. Lederer, J. Blochwitz-Nimoth, H. J. Bolink, *Energy Environ. Sci.* **2016**, 9, 3456.
- [36] M. Roß, L. Gil-Escrig, A. Al-Ashouri, P. Tockhorn, M. Jost, B. Rech, S. Albrecht, *ACS Appl. Mater. Inter.* **2020**.
- [37] Y. H. Chiang, M. Anaya, S. D. Stranks, *ACS Energy Lett.* **2020**, 5, 2498.
- [38] S. J. Moon, J.-H. Yum, L. Löfgren, A. Walter, L. Sansonnens, M. Benkhaira, S. Nicolay, J. Bailat, C. Ballif, *IEEE J. Photovolt.* **2015**, 5, 1087.
- [39] L. Rakocevic, R. Gehlhaar, T. Merckx, W. Qiu, U. W. Paetzold, H. Fledderus, J. Poortmans, *IEEE J. Photovolt.* **2016**, 7, 404.
- [40] L. Palma, F. Matteocci, A. Agresti, S. Pescetelli, E. Calabrò, L. Vesce, S. Christiansen, M. Schmidt, A. Di Carlo, *IEEE J. Photovolt.* **2017**, 7, 1674.
- [41] T. Ye, G. Han, A. Surendran, J. Li, T. M. Koh, S. G. Mhaisalkar, W. L. Leong, *Sol Energ. Mat. Sol. C* **2019**, 201, 110113.
- [42] D. Kiermasch, L. Gil-Escrig, H. J. Bolink, K. Tvingstedt, *Joule* **2019**, 3, 16.
- [43] L. Gao, L. Chen, S. Huang, X. Li, G. Yang, *ACS Appl. Energ. Mater.* **2019**, 2, 3851.

Figure 1: Small area MAPbI₃ PSCs photovoltaic performances as a function of the active layer thickness. a) The schematic architecture of the n-i-p PSCs. Photovoltaic parameters statistical distribution and the as a function of the perovskite thickness for b) PCE, c) open-circuit voltage, V_{oc} ; d) short circuit current, J_{sc} , and e) fill factor, FF.

Figure 2: Square-shaped large area PSCs. a) pictures of large area PSCs with active areas of 1 cm^2 ($1 \text{ cm} \times 1 \text{ cm}$), 4 cm^2 ($2 \text{ cm} \times 2 \text{ cm}$) and 6.25 cm^2 ($2.5 \text{ cm} \times 2.5 \text{ cm}$), b) PSCs J-V characteristic curves under 1-sun illumination.

Figure 3: Rectangular-shaped large area PSCs. a) pictures of large area PSCs with a fixed length (L) of 7 cm and width (w) of 0.2 cm (PSCs n.1 active area is 1.4 cm²), 0.4 cm (PSCs n.2 active area is 2.8 cm²), 0.7 cm (PSCs n.3 active area is 4.9 cm²) and 1 cm (PSCs n.4 active area is 7 cm²). b) PSCs J-V characteristic curves under 1-sun illumination.

Figure 4: Rectangular-shaped large area PSCs. a) pictures of large area PSCs with constant width (w) of 0.65 cm and length (L) of 2 cm (n.1 active area is 1.3 cm²), 3 cm (n.2 active area is 1.95 cm²), 5 cm (n.3 active area is 3.25 cm²) and 7 cm (n.4 active area is 4.55 cm²). b) PSCs J-V characteristic curves under 1-sun illumination.

Figure 5. Output module power vs subcells' width. Calculated output power for a $7 \times 7 \text{ cm}^2$ mini-modules with subcells' widths ranging from 0.2 to 2 cm and dead area width varying from 0.04 to 0.25 cm.

Figure 6: FLE-PSMs structure and photovoltaic characteristics. a) Cross-sectional architecture schematics of mini-module. b) Optical microscopy image, c) J-V curves, J_{sc} of 3.05 mA/cm², V_{oc} of 7.7 V and FF of 75.2%, d) light stability under 1 sun, AM 1.5 G continuous illumination of FLE-PSM with an active area of 6.4 cm² and 90.7% GFF.

Table 1: The photovoltaic parameters values champion pure MAPbI₃ PSCs with different perovskite thicknesses. The average PCE is calculated based on 10 devices.

Thickness	J_{sc}	V_{oc}	FF	PCE	PCE _{average}
(nm)	(mA/cm ²)	(V)	(%)	(%)	(%)
660	21.5	1.11	77.9	18.6	18.0
780	22.3	1.12	76.4	19.1	18.6
1000	21.9	1.11	75.7	18.4	17.6

Table 2: The photovoltaic parameters of PSCs with different square-shaped active areas.

PSCs n.	Area (cm ²)	w (cm)	L (cm)	J_{sc} (mA/cm ²)	V_{oc} (v)	FF (%)	PCE (%)
1	1	1	1	21.4	1.125	70.6	17.0
2	4	2	2	21.3	1.141	45.8	11.1
3	6.25	2.5	2.5	20.7	1.138	41.7	9.8

Table 3: PSCs total area and device aperture reported together the V_{oc} variation

$A_{Aperture}$ (cm ²)	A_{Device} (cm ²)	$A_{Aperture} / A_{Device}$	ΔV_{oc} (mV)
0.16	0.25	0.64	-22.6
1	1.32	0.758	-14.0
4	4.62	0.866	-7.3
6.25	7.02	0.890	-5.9

Table 4: Photovoltaic and geometrical parameters of rectangular-shaped PSCs with fixed length (L) of 7 cm and width (w) varying from 0.2 cm to 1 cm.

PSCs n.	Area (cm ²)	w (cm)	L (cm)	J_{sc} (mA/cm ²)	V_{oc} (v)	FF (%)	PCE (%)
1	1.4	0.2	7	22.3	1.138	71.4	18.1
2	2.8	0.4	7	22.2	1.139	67.6	17.1
3	4.9	0.7	7	21.6	1.140	63.6	15.7
4	7.0	1	7	21.6	1.140	61.0	15.0

Table 5: Photovoltaic and geometrical parameters of rectangular-shaped PSCs with constant width (w) of 0.65 cm and length (L) varying from of 2 cm to 7 cm

PSCs n.	Area (cm ²)	w (cm)	L (cm)	J_{sc} (mA/cm ²)	V_{oc} (v)	FF (%)	PCE (%)
---------	-------------------------	----------	----------	--------------------------------	--------------	--------	---------

1	1.3	0.65	2	21.5	1.125	65.9	15.9
2	1.95	0.65	3	21.9	1.129	64.1	15.8
3	3.25	0.65	5	21.7	1.127	64.5	15.8
4	4.55	0.65	7	21.7	1.123	63.2	15.4

Table 6. Calculated output power of the 7 cm x 7 cm PSM with different subcells' widths of 0.2 to 2 cm and different dead area width of 0.06 and 0.25 cm. GFF=photoactive area / total area = (subcells' width-dead area width) / subcells' width.

Subcells' width (cm)	Dead area width (cm)	Subcells' number	Subcells' PCE (%)	GFF (%)	Output power (mW)
0.2	0.06	35	18.1	70.0	621
0.4		17	17.1	85.0	692
0.7		10	15.7	91.4	703
1		7	15.0	94.0	691
2		3	11.1	97.0	452
0.2	0.25	-	18.1	-	-
0.4		17	17.1	37.5	305
0.7		10	15.7	64.3	495
1		7	15.0	75.0	551
2		3	11.1	87.5	408

Table 7: Photovoltaic parameters of the champion FLE-PSMs

Subcells' number	Area (cm ²)	GFF (%)	w (cm)	L (cm)	Dead area width (cm)	J _{sc} (mA/cm ²)	V _{oc} (V)	FF (%)	PCE (%)
7	6.4	90.7	0.65	1.55	0.06	3.15	7.779	75.2	18.4

Supplementary information

Design of perovskite thermally co-evaporated highly efficient mini-modules with high geometrical fill factors

Jia Li,^a Herlina Arianita Dewi,^a Hao Wang,^a Jia Haur Lew,^a Nripan Mathews,^{a,b}

Subodh Mhaisalkar,^{a,b} Annalisa Bruno^{a}*

^a. Energy Research Institute @ NTU (ERI@N), Nanyang Technological University,

Singapore 637553.

^b School of Materials Science & Engineering, Nanyang Technological University,

Singapore, 639798.

Corresponding authors: nripan@ntu.edu.sg

subodh@ntu.edu.sg, annalisa@ntu.edu.sg

Figure S1. Cross-sectional SEM images of MAPbI₃ perovskite thin- films with different thicknesses: a) 660 nm, b) 780 nm and c) 1000 nm.

Figure S2. Absorption spectra of different thick perovskite, 660 nm, 780nm and 1000 nm, respectively.

Figure S3. IPCE spectra of PSCs with different thick perovskite, 660 nm, 780nm and 1000 nm, respectively, the integrated J_{sc} are 20.59, 21.12 and 20.95 mA/cm², respectively.

Figure S4. SEM images of a) P2 Laser etching line and b) P3 Laser etching line.

Figure S5. Shelf stability of the full laser-etched perovskite mini-modules (FLE-PSM). Normalized a) PCE, b) V_{oc} , c) J_{sc} , d) FF values. The devices are stored in a dry box with humidity around 30% and measured in the atmosphere with humidity near 70%.