nature portfolio

Peer Review File

Efficient and stable perovskite mini-module via high quality homogeneous perovskite crystallization and improved interconnect

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REVIEWER COMMENTS

Reviewer #1 (Remarks to the Author):

The authors improved the crystallization orientation, carrier lifetime, and morphological quality of perovskite films through a simple low thermal field growth under a scalable process, and obtained high-quality, uniform, and large-area perovskite films. The championship efficiencies of small-area device (0.0737cm2) and large-area module (14.6cm2) are 24.93% and 23.2% respectively, showing very low efficiency loss with area amplification. The author also invented the P1.5 to build a lateral barrier layer to address the problem of lateral diffusion degradation near the interconnection area in the module, thereby improving the stability of the device. After operating for 1,000 hours at the maximum power point of continuous illumination at around 40 degrees Celsius, the efficiency of the module device remains above 90% of its initial efficiency. The certification efficiency of the module was 22.7% (14.6cm2). The results achieved in this manuscript are very impressive. The manuscript should be considered for publication in Nature Communication after addressing following suggestions/comments:

1. The author points out that when the substrate temperature is 18 oC, the peak intensity ratio of 110/310 is the highest, indicating the existence of preferred orientation. But it's unclear what the specific orientation of 110 means, and what properties of the reinforced material this will benefit? The 110 peak of perovskite for 18 degree shifted to smaller angle, what might be the reason?
2. The PL of the perovskite film under different substrate temperatures is not seen in the manuscript, and the corresponding peak position change results need to be given.
3. Figure 3d and supporting material Figure 13 should be listed to give more information, such as device structure/area/performance, etc.
4. The detailed calculation procedure for the geometric fill factor (GFF) in the manuscript should be given.
5. Using P1.5 improves the stability of the module, but it still drops by about 10% near the maximum power point. Please provide the IV curve after MPP as much as possible, count the decline of key parameters, and give possible optimization suggestions.
6. Does the "ambient temperature" mean the vacuum chamber temperature or the spin-coating chamber temperature? or the annealing chamber temperature?
7. Please give a microscopic image of a P1.5.

Reviewer #2 (Remarks to the Author):

In this manuscript (efficient and stable perovskite mini-module via high quality homogeneous perovskite crystallization and improved interconnect), Zhou et al. proposed a physical method (lowering the substrate temperature) to control the nucleation growth of perovskite intermediates. This method is very helpful in formation of high quality and homogeneous perovskite film, and this method is independent on the environment condition, which is very important for the industry fabrication. Meanwhile, the author also proposed a simple solution of constructing a diffusion barrier layer (called P1.5), which lead to significant suppression of the lateral diffusion. With these two strategies, the author developed a mini module with a certified efficiency of 22.7%, which is indeed a leading certified efficiency result at present; moreover, the large-area device also shows good operational stability. There is no doubt that these inventions and creations will be helpful to the area expansion of perovskite cells. Overall, after clarifying the following issues, I recommend it for publication in Nature Communications. Minor revisions:

The authors have shown that the perovskite film quality is independent on the environment temperature from XRD and SEM results, the reviewer suggest the authors add the device performance in the supplementary information.

The efficiency loss between small-area devices and large-scale devices is already pretty low, which is encouraging. There is still a gap between the small size device (0.1 cm2) and minimodule, it is better to discuss the which is the limitation at present in this manuscript.

P1.5 was performed without surface protection of the perovskite. Will this affect the stability of the perovskite? Moreover, in the dead area, in addition to the vicinity of P2, the vicinity of P3 will also affect stability. Are there any relevant improvements to this problem?

The experimental part should disclose more details, such as the experimental parameters of ALD

tin oxide, and the different details of the preparation of functional layers of large-area and small-area devices.

Reviewer #3 (Remarks to the Author):

The manuscript reports on high-performance perovskite mini-modules achieved by optimizing the temperature for vacuum-assisted drying of the perovskite film. Additionally, diffusion barrier layers (DFLs) are introduced in the mini-modules, and the effects of DFLs on the lifetime of the mini-modules are studied. While the efficiency of the modules is impressive, I do not recommend publication in Nature Communications due to a lack of novelty.
The manuscript claims two key novelties. The first is low thermal field growth (LTFG). While they demonstrate a clear temperature effect on the quality of the perovskite film, I do not agree that it constitutes a novel technique. The optimized temperature is more likely within the typical range of temperatures found in air-conditioned labs. If devices were fabricated at a known optimum ambient temperature (18-20 °C), the results would likely be similar to those of the optimized samples. I would say that their trials for further improvement by increasing/decreasing drying plate temperature did not work rather than calling it LTFG technique for the optimum temperature of 18 °C. Yet, I appreciate the clear demonstration of the temperature effect which will guide the researchers working on the technology. In addition, I am not sure where the "field" is from.

The second novelty is the DFLs. While the approach may be novel, its applicability is limited. I believe that such a barrier effect can only be achieved by a pinhole-free metal oxide layer, which can be fabricated only by the atomic layer deposition (ALD) technique, as the authors used in this work. The costly nature of this process restricts the adoptability of the technology. Moreover, the approach may not be useful for commonly used Au electrodes or solution-processed carbon electrodes.

Therefore, I do not recommend publication of this manuscript in Nature Communications; however, it may be reconsidered if the authors can demonstrate the general applicability of the DFLs by showing their effect with various electrodes and (preferably solution-processed) electron transport layer (ETL) materials.

In case the authors submit a revised manuscript, I suggest addressing the following points:

- Can the authors provide photocurrent maps (LBIC data) of modules depending on the aging time for both air and N2 aged samples? This data would provide comprehensive information on the degradation pathway, and the effects of DFLs could be clearly seen.
- Only power conversion efficiency (PCE) is provided for the stability test. It would be useful to have Jsc, FF, and Voc curves for the stability test for a better understanding of the degradation mechanism.
- The slot die setup should be shown in the manuscript or supplementary information.

List of point-to-point response of reviewers' comments

NCOMMS-24-13983-T

We really appreciated the Reviewers' great comments and suggestions, which really helped us to further improve the manuscript. We have tried to address all the concerns mentioned by the reviewers and we hope the Reviewers will find their previous comments addressed to a satisfactory level by our responses below.

Reviewer #1

"The authors improved the crystallization orientation, carrier lifetime, and morphological quality of perovskite films through a simple low thermal field growth under a scalable process, and obtained high-quality, uniform, and large-area perovskite films. The championship efficiencies of small-area device (0.0737 cm^2) and large-area module (14.6 cm^2) are 24.93% and 23.2% respectively, showing very low efficiency loss with area amplification. The author also invented the P1.5 to build a lateral barrier layer to address the problem of lateral diffusion degradation near the interconnection area in the module, thereby improving the stability of the device. After operating for 1,000 hours at the maximum power point of continuous illumination at around 40 degrees Celsius, the efficiency of the module device remains above 90% of its initial efficiency. The certification efficiency of the module was 22.7% (14.6 cm^2). The results achieved in this manuscript are very impressive. The manuscript should be considered for publication in Nature Communication after addressing following suggestions/comments:"

- **Our Response:** We thank the reviewer for their positive feedback and for their constructive comments to improve our manuscript. Detailed responses to each point are provided below.

1. "The author points out that when the substrate temperature is 18 ^oC, the peak intensity ratio of 110/310 is the highest, indicating the existence of preferred orientation. But it's unclear what the specific orientation of 110 means, and what properties of the reinforced material this will benefit? The 110 peak of perovskite for 18 ^oC shifted to smaller angle, what might be the reason?"

- **Our Response:** Thanks for the Reviewer's comment. The improvement in the orientation could increase carrier mobility and reducing defect densities (Refs. 27-29).

- **In page 6**, a sentence "It is reported that the preferred orientation of the diffraction peak near 14º is beneficial to increasing carrier mobility and reducing defect density[27-29]."
- We enlarged the diffraction peak of (110) for different substrate temperatures, we found the diffraction is the same at 10–26 ºC, while there is a 0.03º shift to large diffraction peak while processing at 30 ºC (Fig. R1), this could be due to the existence of strain in perovskite layer. (*Sci. Adv.*, 2017, 3, eaao5616; *Nat. Commun.*, 2020, 11, 1514).

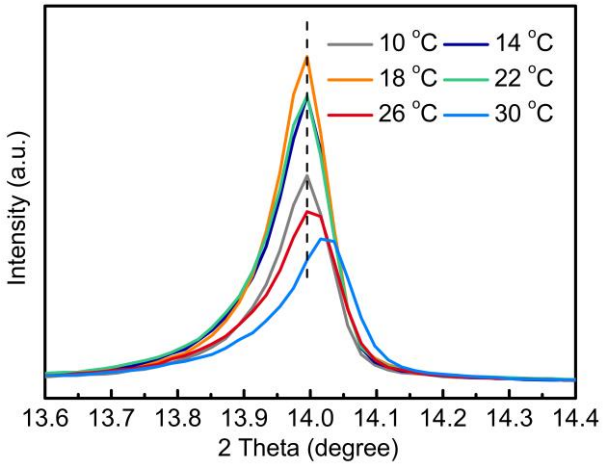


Figure R1 *XRD of perovskite films obtained at different substrate temperatures.*

2. The PL of the perovskite film under different substrate temperatures is not seen in the manuscript, and the corresponding peak position change results need to be given.

- **Our Response:** We have shown the PL results in Figure R2, this result has been added as Supplementary Fig. 3 in the revised Supporting Information.
- **In page 6**, a sentence "Photoluminescence (PL) results (Supplementary Fig. 3) show that the perovskite film fabricated with a LTSG-18 ºC has the strongest PL peak, indicating that the film has the lowest defect state density. In addition, it was also found that as the LTSG temperature increased to 30 ºC, the PL peak position red-shifted from 797 nm to 809 nm, which was speculated to be related to defects."

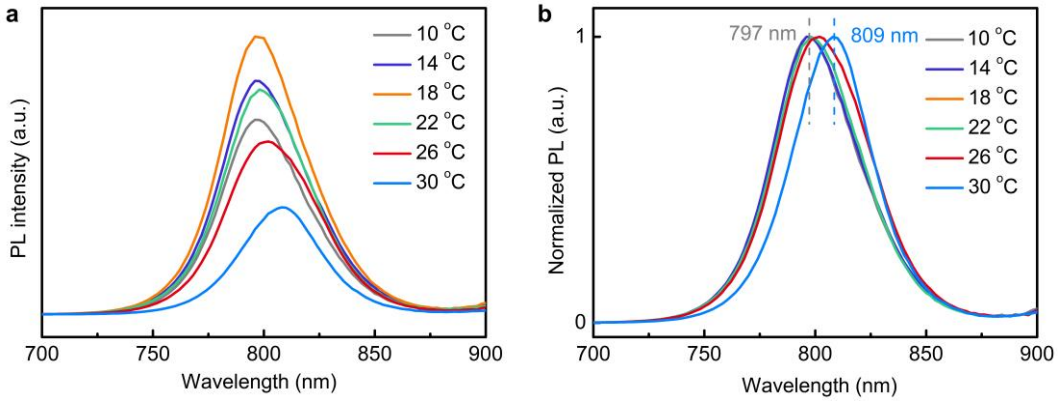


Figure R2 *Steady-state PL spectrum of a perovskite films (a) and its normalized representation (b).*

3. Figure 3d and supporting material Figure 13 should be listed to give more information, such as device structure/area/performance, etc.

- **Our Response:** We appreciate this constructive suggestion. We detailed the device structure, aperture area, and power conversion efficiency (PCE) (please see Table R1 and Table R2), we have added these details as supplementary Table 2 and Table 4 in the revised Supplementary Information.

Table R1 *Structure, aperture area, and PCE of reported small-area and large-area devices.*

Device structure	Aperture area (cm^2)	Aperture area PCE (%)	Ref.
ITO/SnO_2/perovskite/spiro-OMeTAD/Ag or Au	0.06	25	*Nature* 620, 323-327 (2023)
	27.83	21.4	
ITO/SnO_2/perovskite/Organic HTL/Ag or Au	1	23.5	*Science* 379, 288-294 (2023)
	17.1	21.4	
ITO/PTAA/perovskite/C_{60}/BCP/Cu	0.07	23.8	*Science* 373, 902-907 (2021)
	17.9	20.1	
	50.1	19.7	
FTO/TiO_2/SnO_2/perovskite/spiro-MeOTAD/Au	0.085	25.09	*Nat. Sustainability* 6, 1465-1473 (2023)
	12.25	20.75	
FTO/SnO_2/perovskite/spiro-OMeTAD/Au	0.16	24.02	*Nat. Energy* 7, 528-536 (2022)
	22.4	20.5	
ITO/PTAA/perovskite/C_{60}/BCP/Cu	0.08	24.6	*Science* 380, 823-829 (2023)
	26.9	21.8	
ITO/PTAA/Al_2O_3/perovskite/C_{60}/SnO_2/Ag	0.09	23.21	*Adv. Mater.* 36, 2309310 (2024)
	12.84	20.88	
ITO/SnO_2/perovskite/spiro-OMeTAD/Au	0.09	21.8	*Nat. Energy* 5, 596-604 (2020)
	22.4	16.6	
ITO/NiO_X/PTAA/perovskite/PCBM/BCP/Ag	0.05979	24.7	*Energy Environ. Sci.* 16, 557-564 (2023)
	19.3	21.6	
ITO/PTAA/perovskite/C_{60}/BCP/metal	0.08	21.3	*Sci. Adv.* 5, eaax7537 (2019)

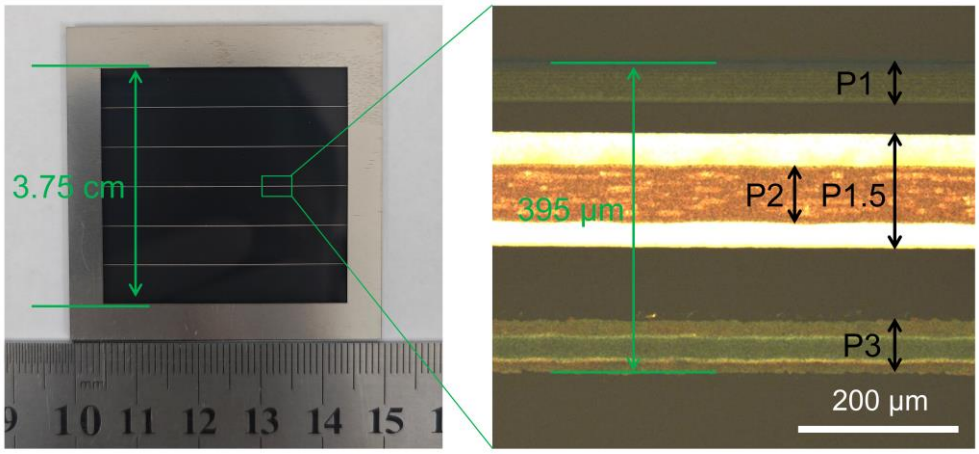
	63.7	16.9	
FTO/TiO_2/SnO_2/perovskite/spiro-MeOTAD/Au	0.0803	25.7	*Science* 375, 302-306 (2022)
	1	23.3	
	20.92	20.75	
	66.95	19.7	
FTO/NiO_X/Me-4PACz/perovskite/PCBM/SnO_2/Cu	0.0737	24.93	**This work**
	14.625	23.2	

Table R2 *Structure, certified steady-state PCE, and aperture area of the reported mini-modules.*

Device structure	**Aperture area (cm^2)**	**Certified steady-state PCE (%)**	**Ref.**
ITO/PTAA/perovskite/C_{60}/BCP/metal	63.7	16.37	*Sci. Adv.* 5, eaax7537 (2019)
ITO/SnO_2/perovskite/spiro-OMeTAD/Au	22.26	13.88	*Nat. Energy* 5, 596-604 (2020)
ITO/PTAA/perovskite/C_{60}/BCP/Cu	30	18.6	*Nat. Energy* 6, 633-641 (2021)
ITO/PTAA/perovskite/C_{60}/BCP/Cu	18.1	19.3	*Science* 373, 902-907 (2021)
	50	19.2	
FTO/TiO_2/perovskite/Spiro/Au	31	17.53	*Joule* 5, 2420-2436 (2021)
ITO/PTAA/Al_2O_3/perovskite/C_{60}/SnO_2/Ag	12.84	20.1	*Adv. Energy Mater.* 12, 2202287 (2022)
ITO/NiO_X/PTAA/perovskite/PCBM/BCP/Ag	18.52	20.35	*Energy Environ. Sci.* 16, 557-564 (2023)
ITO/PTAA/perovskite/C_{60}/BCP/Cu	26.9	21.1	*Science* 380, 823-829 (2023)
ITO/PTAA/Al_2O_3/perovskite/C_{60}/SnO_2/Ag	12.84	20.56	*Adv. Mater.* 36, 2309310 (2024)
FTO/NiO_X/Me-4PACz/perovskite/PCBM/SnO_2/Cu	14.61	22.73	**This work**

4. The detailed calculation procedure for the geometric fill factor (GFF) in the manuscript should be given.

- **Our Response:** The GFF calculation process is shown in Figure R3 as follows, which has been added in Supplementary Fig. 15 in the revised Supplementary Information.



$$GFF = \frac{3.75\ cm - 5 \times 0.0395\ cm}{3.75\ cm} = 94.7\%$$

Figure R3 *Calculation process of geometric fill factor (GFF).*

5. Using P1.5 improves the stability of the module, but it still drops by about 10% near the maximum power point. Please provide the IV curve after MPP as much as possible, count the decline of key parameters, and give possible optimization suggestions.

- **Our Response:** We have provided the *I-V* curves and performance parameters (Figure R4) of the P1.5 module before and after MPPT, and the results are added in Supplementary Fig. 23 in the revised Supporting Information.
- **In page 11**, a sentence "We noticed that the loss of PCE of P1.5 module after MPPT mainly comes from the decrease of open circuit voltage (Supplementary Fig. 23), which could be due to the passivation layer degraded."

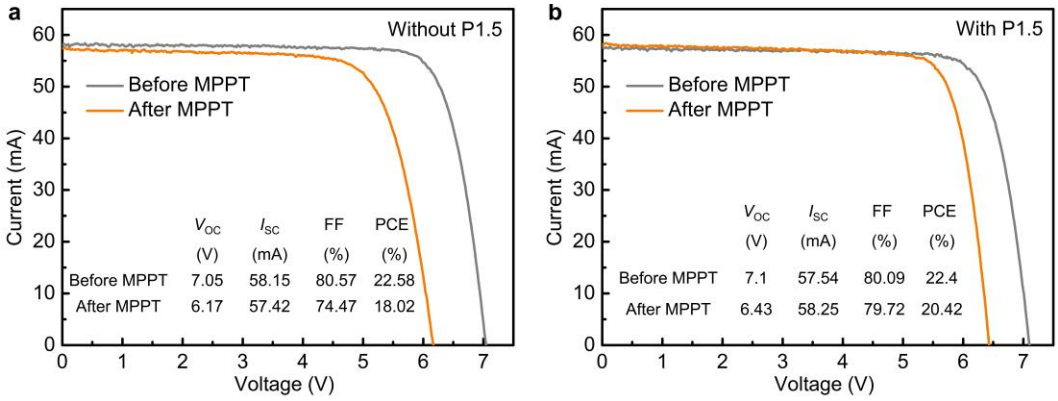


Figure R4 *I-V curves and performance parameters of perovskite modules before and after (a) 600 hours maximum power point tracking (MPPT), and (b) 1000 hours maximum power point tracking (MPPT).*

6. Does the "ambient temperature" mean the vacuum chamber temperature or the spin-coating chamber temperature? or the annealing chamber temperature?

- **Our Response:** We thank the reviewer for pointing out this unclearly described information. The ambient temperature in this manuscript refers to the environment temperature while we carried out spin coating, vacuum pumping/flash and annealing.
- **In page 3**, we have added the explanation "the ambient temperature (environment temperature during spin-coating/vacuum chamber/annealing)" in the revised manuscript.

7. Please give a microscopic image of a P1.5.

- **Our Response:** We have included microscopic images of P1.5 (Figure R5), which has been added in Supplementary Fig. 12 in the revised Supporting Information. It can be clearly seen that the laser induced pattern is very sharp.

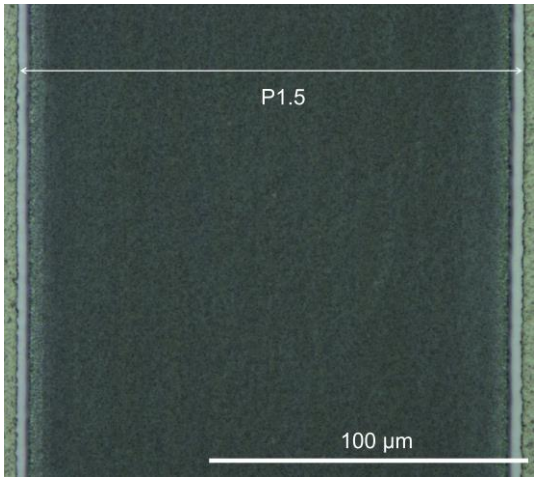


Figure R5 *Optical microscope image of P1.5.*

Reviewer #2

"In this manuscript (efficient and stable perovskite mini-module via high quality homogeneous perovskite crystallization and improved interconnect), Zhou et al. proposed a physical method (lowering the substrate temperature) to control the nucleation growth of perovskite intermediates. This method is very helpful in formation of high quality and homogeneous perovskite film, and this method is independent on the environment condition, which is very important for the industry fabrication. Meanwhile, the author also proposed a simple solution of constructing

a diffusion barrier layer (called P1.5), which lead to significant suppression of the lateral diffusion. With these two strategies, the author developed a mini module with a certified efficiency of 22.7%, which is indeed a leading certified efficiency result at present; moreover, the large-area device also shows good operational stability. There is no doubt that these inventions and creations will be helpful to the area expansion of perovskite cells. Overall, after clarifying the following issues, I recommend it for publication in Nature Communications. Minor revisions:"

- **Our Response:** Thank you for understanding the importance of our work and for your constructive feedback. We carefully considered your comments and revised the manuscript accordingly. Please see our detailed response below.

1. The authors have shown that the perovskite film quality is independent on the environment temperature from XRD and SEM results, the reviewer suggest the authors add the device performance in the supplementary information.

- **Our Response:** We appreciate and thank the reviewers for their suggestions. We have added the *J-V* curves and performance parameters (Figure R6) of devices based on perovskite films prepared at different ambient temperatures in the revised Supporting Information (Supplementary Fig. 5).
- **In page 6**, a sentence "device performance (Supplementary Fig. 5, see below for device structures), indicating the LTSG is insensitive to the surrounding environment."

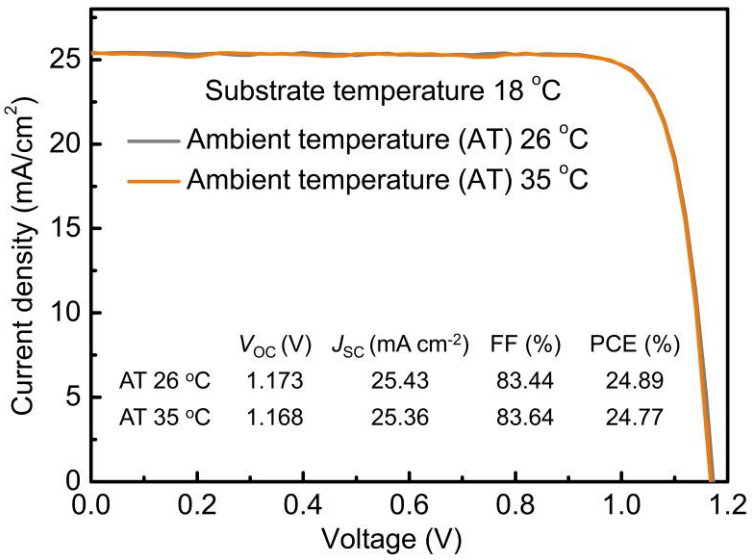


Figure R6 *J-V curves and performance parameters of devices constructed with perovskite films prepared at ambient temperatures of 26 °C and 35 °C respectively.*

2. The efficiency loss between small-area devices and large-scale devices is already pretty low, which is encouraging. There is still a gap between the small size device (0.1 cm^2) and minimodule, it is better to discuss the which is the limitation at present in this manuscript.

- **Our Response:** We appreciate the reviewer's recognition of the small loss in efficiency of our device. We have added relevant parameter comparisons (Table R3) in the revised Supporting Information (Supplementary Table 3).
- **In page 9**, a sentence "By comparing the performance parameters of our champion PCE small-area and large-area devices (Supplementary Table 3), we noticed that the main loss comes from the loss of short circuit current density. Therefore, increasing the geometric fill factor in the future is an important way to further reduce efficiency losses."

Table R3 *Open circuit voltage, short circuit current density, fill factor and PCE of our small and large area devices.*

	V_{OC} (V)	J_{SC} (mA cm^{-2})	FF (%)	PCE (%)
Small area device	1.187	25.36	82.75	24.93
Large area device	7.073/6 = 1.179	(58.47/14.625)*6 = 23.99	82.03	23.2

3. P1.5 was performed without surface protection of the perovskite. Will this affect the stability of the perovskite? Moreover, in the dead area, in addition to the vicinity of P2, the vicinity of P3 will also affect stability. Are there any relevant improvements to this problem?

- **Our Response:** It can be concluded from our experimental results that performing P1.5 when exposing perovskite to air did not deteriorate the properties of the perovskite. For example, Fig. 3c in the manuscript shows similar efficiency and low hysteresis whether or not P1.5 is executed; Fig. 4 in the manuscript shows better stability of the module implementing P1.5.
- We agree that P3 is also a key factor affecting module stability. In our current manuscript, we have not taken steps to investigate the impact of P3 on stability. However, we believe that constructing P3 by mask evaporating metal electrodes may be an effective improvement method.

4. The experimental part should disclose more details, such as the experimental parameters of ALD tin oxide, and the different details of the preparation of functional layers of large-area and small-area devices.

- **Our Response:** Thanks for the Reviewer's suggestion. We have added experimental details in the revised manuscript.
- **In Page 13**, the sentences "The substrate was then transferred to the atomic layer deposition (ALD) system for SnO_2 layer deposition, the deposition parameter can be found in elsewhere[37]" and "For the mini-module device fabrication, the procedures are similar as the small size device, the only one difference is the amout of solution used" have been added in the experimental.

Reviewer #3

"The manuscript reports on high-performance perovskite mini-modules achieved by optimizing the temperature for vacuum-assisted drying of the perovskite film. Additionally, diffusion barrier layers (DFLs) are introduced in the mini-modules, and the effects of DFLs on the lifetime of the mini-modules are studied. While the efficiency of the modules is impressive, I do not recommend publication in Nature Communications due to a lack of novelty."

- **Our Response:** Thanks for the reviewer's positive comments on our minimodule device performance. And we also appreciated the Reviewer's critical comments on our manuscript, which will help us to further improve the manuscript.

1. The manuscript claims two key novelties. The first is low thermal field growth (LTFG). While they demonstrate a clear temperature effect on the quality of the perovskite film, I do not agree that it constitutes a novel technique. The optimized temperature is more likely within the typical range of temperatures found in air-conditioned labs. If devices were fabricated at a known optimum ambient temperature (18-20 ^oC), the results would likely be similar to those of the optimized samples. I would say that their trials for further improvement by increasing/decreasing drying plate temperature did not work rather than calling it LTFG technique for the optimum temperature of 18 ^oC. Yet, I appreciate the clear demonstration of the temperature effect which will guide the researchers working on the technology. In addition, I am not sure where the "field" is from.

- **Our Response:** We thank the reviewers for recognizing that we have clearly demonstrated temperature effects and that our work will be helpful to related researchers. We agreed that if we can keep the ambient environment on the optimized temperature such as 18 ^oC, which will show the similar results. However, it will consume huge amounts of energy for controlling the temperature

in a large space, which undoubtedly increases production costs and reduces market competitiveness. Therefore, our low-temperature substrate solution may be a low-cost candidate in the actual production process. Furthermore, our solution may be a way to improve the tolerance to ambient temperature changes in real production.

- As the reviewer's suggested, to avoid misunderstanding, we changed “low thermal field growth (LTFG)” in the original manuscript to “low temperature substrate growth (LTSG).”

2. The second novelty is the DFLs. While the approach may be novel, its applicability is limited. I believe that such a barrier effect can only be achieved by a pinhole-free metal oxide layer, which can be fabricated only by the atomic layer deposition (ALD) technique, as the authors used in this work. The costly nature of this process restricts the adoptability of the technology. Moreover, the approach may not be useful for commonly used Au electrodes or solution-processed carbon electrodes. Therefore, I do not recommend publication of this manuscript in Nature Communications; however, it may be reconsidered if the authors can demonstrate the general applicability of the DFLs by showing their effect with various electrodes and (preferably solution-processed) electron transport layer (ETL) materials.

- **Our Response:** We thank that the reviewer’s appreciation of novelty of our P1.5 method.
- As the Reviewer suggested, we have constructed thermal evaporation deposited C_{60} and solution deposited PCBM as DBLs *via* our P1.5 method, respectively. C_{60} and PCBM are clearly spread and covered on the side of the perovskite (Figure R7 and R8), which well isolates the direct contact between the perovskite and the metal (Au) electrode.
- We also built modules using these electron transport layers (ETLs) and Au electrodes, and found that modules with P1.5 (DBLs) showed better thermal (85 ^{o}C) aging stability (Figure R9 and R10).
- In addition, unlike metal electrodes used to prepare high-efficiency solar cells, carbon electrodes usually do not have strong diffusion behavior with perovskites and can be in direct contact with perovskites during use (*Science*, 2024, 383, 1198 – 1204). Therefore, we only conducted relevant studies on modules with metal electrodes.

- In summary, these results indicate that our solution (P1.5) for constructing DBLs is compatible with a variety of electron transport layer materials (SnO_2, C_{60} and PCBM, etc.) and their deposition methods (ALD, thermal evaporation and solution methods, etc.), as well as metal electrode materials (Cu and Au etc.).

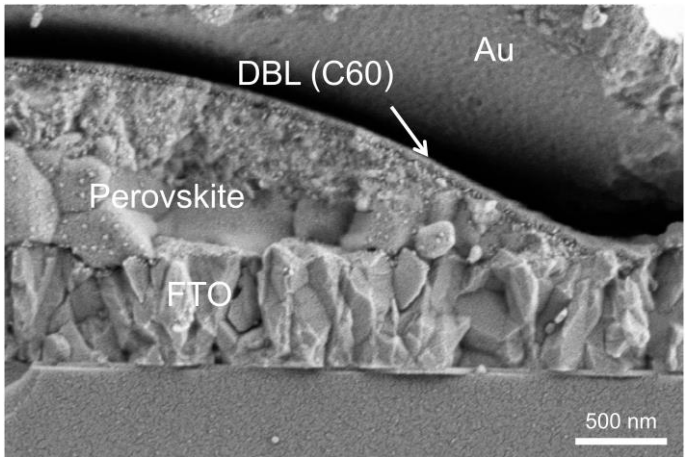


Figure R7 *Through the P1.5 method, C60 (thermal evaporation deposition) covers the sides of the perovskite.*

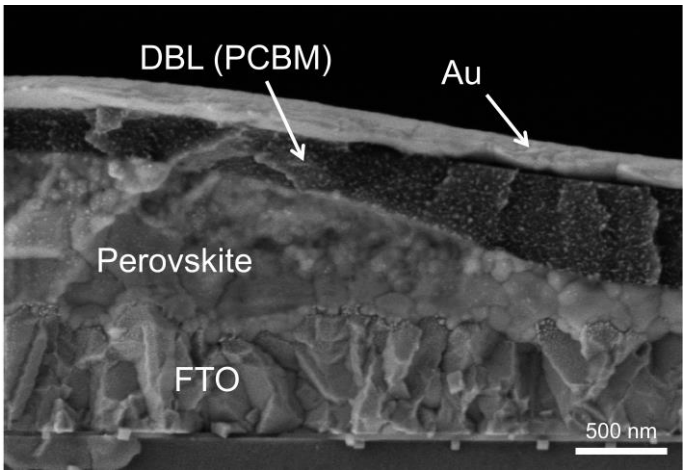


Figure R8 *Through the method of P1.5, PCBM (solution deposition) is well covered on the side of the perovskite.*

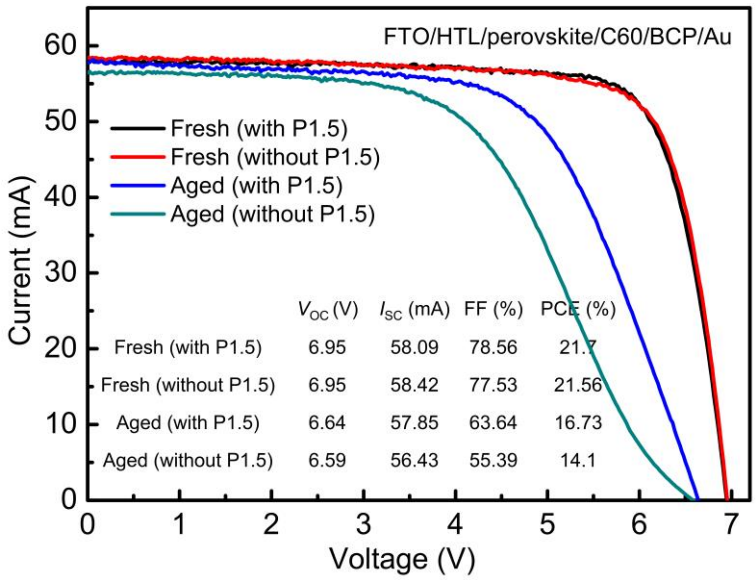


Figure R9 *FTO/HTL/perovskite/C60/BCP/Au. I-V curves and performance parameters of modules containing C_{60} before and after thermal aging (85 °C, 305 hours).*

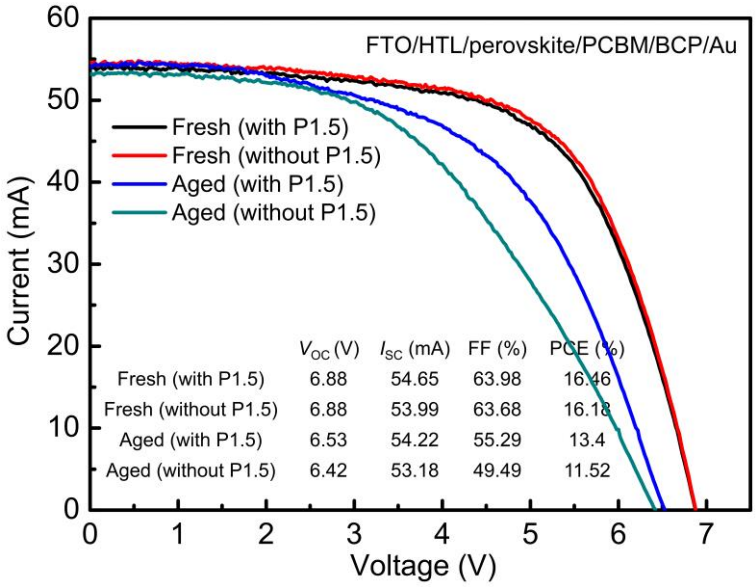


Figure R10 *FTO/HTL/perovskite/PCBM/BCP/Au. I-V curves and performance parameters of modules containing PCBM before and after thermal aging (85 °C, 262 hours).*

In case the authors submit a revised manuscript, I suggest addressing the following points:

3. Can the authors provide photocurrent maps (LBIC data) of modules depending on the aging time for both air and N_2 aged samples? This data would provide

comprehensive information on the degradation pathway, and the effects of DFLs could be clearly seen.

- **Our Response:** We appreciated the reviewer's very important suggestion, we have carried out light beam induced current (LBIC) measurement, please see Figure R11 and Supplementary Fig. 20.
- **In page 10**, a sentence "We have carried out light beam induced current (LBIC) measurement for the device with and without P1.5, we found that after air aging, the P1.5 device has a more uniform, bright, and clear response image compared to the non P1.5 device (Supplementary Fig. 20), further demonstrating the anti-degradation effect of P1.5 (DBLs)."

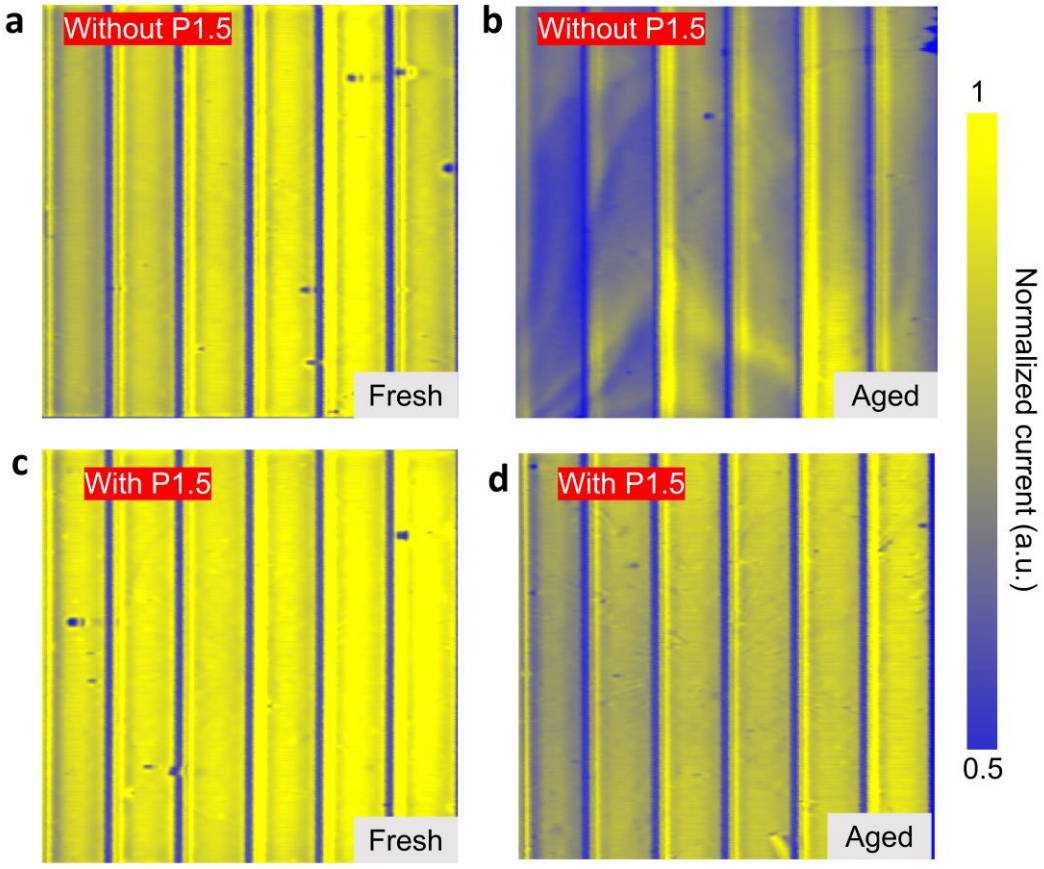


Figure R11 *Light (532 nm) beam induced current (LBIC) mapping images of perovskite solar modules with and without P1.5 (DBL) before and after air ambient aging.*

4. Only power conversion efficiency (PCE) is provided for the stability test. It would be useful to have J_{SC}, FF, and V_{OC} curves for the stability test for a better understanding of the degradation mechanism.

- **Our Response:** Thanks for the Reviewer's suggestion. We have added the photovoltaic parameters variation such as I_{SC}, FF, and V_{OC} during aging in air

(Fig. R12, Supplementary Fig. 19), heating at 85 °C (Fig. R13, Supplementary Fig. 21) and MPPT (Fig. R14, Supplementary Fig. 23).

- **In page 9**, a sentence "After aging for 328 hours (Fig. 4a and Supplementary Fig. 19), we found that the module without P1.5 decreased to 60% of the initial PCE, with the FF, I_{SC}, and V_{OC} loss are 29.6%, 9.9%, and 5.2%, respectively, the module with P1.5 still maintains 82% of the initial PCE, with FF, I_{SC}, and V_{OC} loss are 12.2%, 4.9%, and 1.9%, respectively."

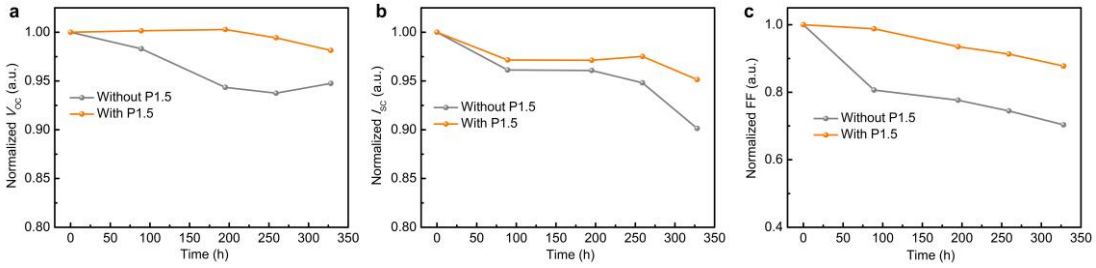


Figure R12 *Performance of perovskite modules in air aging experiments at 25 °C and 30-40% relative humidity.*

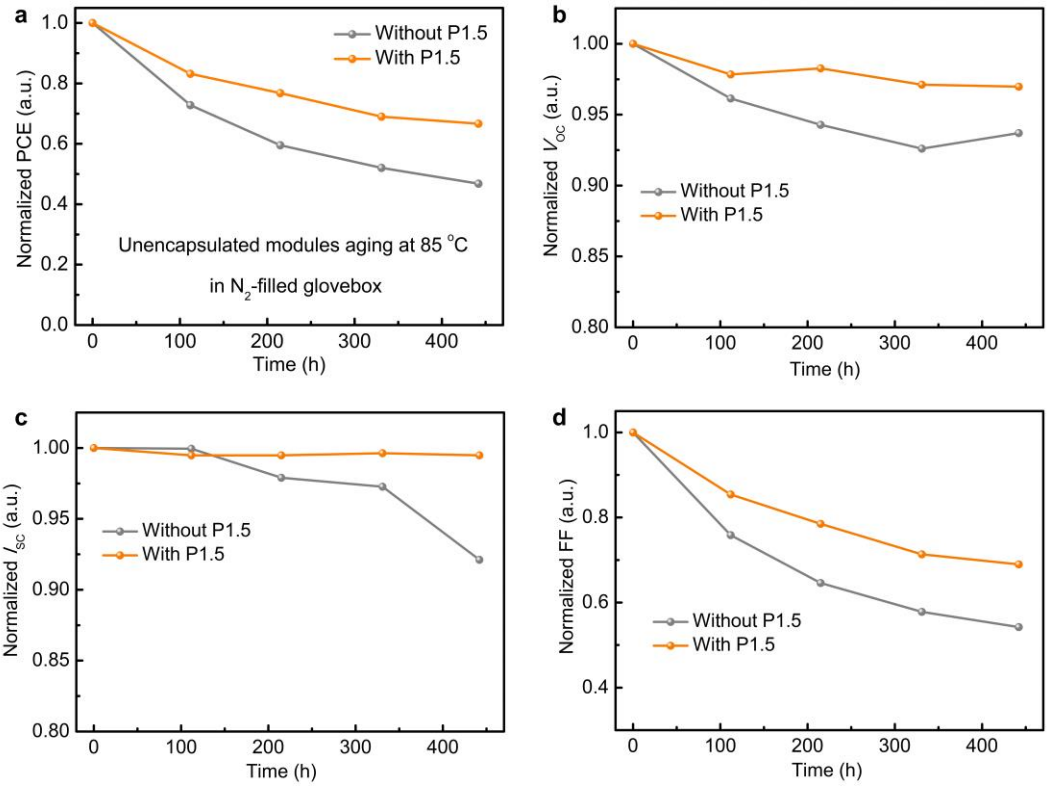


Figure R13 *Thermal stability tracking of the modules heated on an 85 °C hotplate in a N_2 glove box.*

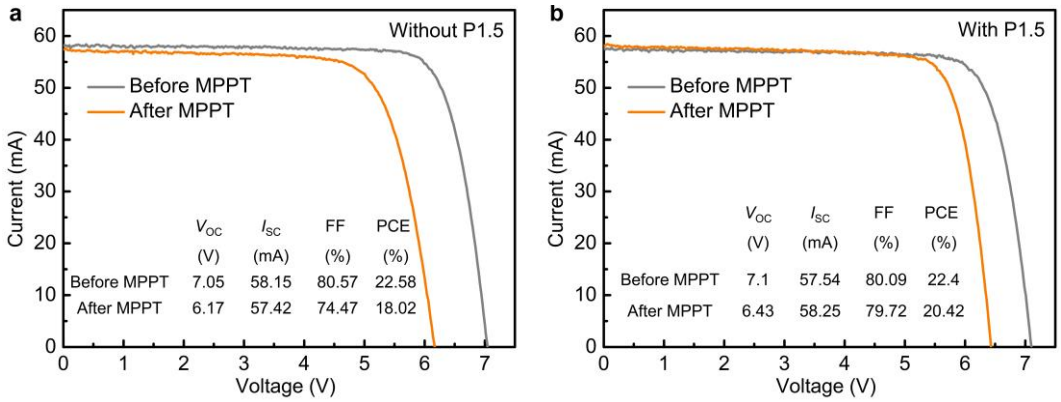


Figure R14 *I-V curves and performance parameters of perovskite modules before and after (a) 600 hours maximum power point tracking (MPPT), and (b) 1000 hours maximum power point tracking (MPPT).*

5. The slot die setup should be shown in the manuscript or supplementary information.

- **Our Response:** As the Reviewer suggested, we have added the slot die setup in Supplementary Fig. 18b (Also in Figure R15b) in the revised supplementary information.

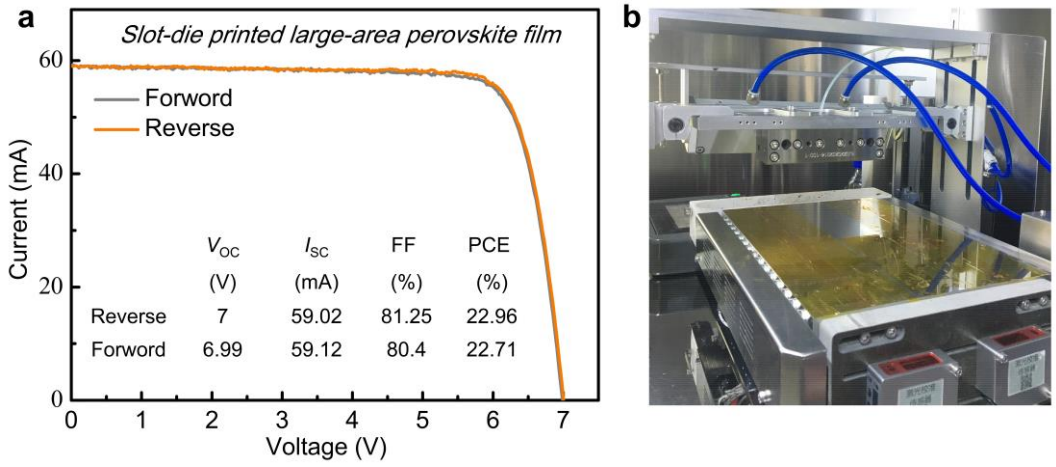


Figure R15 a, *I-V curves and performance parameters of large-area module (aperture area 14.625 cm^2) fabricated by slot-die printed perovskite precursor wet films, combined with our LTSG and P1.5 solutions.* ***b,*** *Slot-die system (purchased from Datamaker).*

REVIEWERS' COMMENTS

Reviewer #1 (Remarks to the Author):

The authors have clearly responded to my concerns. And I considered it could be accepted now.

Reviewer #2 (Remarks to the Author):

I am satisfied with the response from the authors and have no further questions. I recommend the publication of this manuscript in Nature Communications.

Reviewer #3 (Remarks to the Author):

The authors have responded well, and the manuscript has been significantly improved. So, I am happy to recommend the publication of the manuscript in Nature Communications.