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Corresponding author(s): Stolterfoht, Martin

Solar Cells Reporting Summary

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Experimental design

Please check: are the following details reported in the manuscript?

1. Dimensions

Area of the tested solar cells	[x] Yes [ ] No	The fast-hysteresis (FH) measurements were performed on 6 mm2 large cells as defined by the overlap of ITO and Cu.
Method used to determine the device area	[x] Yes [ ] No	The intended device area was confirmed with an optical microscope.

2. Current-voltage characterization

Current density-voltage (J-V) plots in both forward and backward direction	[x] Yes [ ] No	Figure 2 and 4, and throughout SI (Figure S4-S7, S9-S10, S12, S13, S14, S20, S23-S25, S29, S31, S32).
Voltage scan conditions *For instance: scan direction, speed, dwell times*	[x] Yes [ ] No	For fast-hysteresis (FH) measurements the scan speed was varied from 10 mV or 100 mV/s to over 2000 V/s using a triangular voltage pulse in backward and forward direction as explained in the Methods section. In order to cross-check the FH results at slow scan speeds (10-100 mV/s), standard JV measurements were performed with a Keithley 2400 on the same cells which resulted in essentially identical performance metrics as obtained with the FH setup. Specified in Methods, Supporting Information (SI).
Test environment *For instance: characterization temperature, in air or in glove box*	[x] Yes [ ] No	Unless specified otherwise, the temperature of the cell was fixed to 25 °C during the JV-scan (also for measurements with the fast-hysteresis setup) in a nitrogen-filled glovebox using a sample holder with built-in Peltier element. Specified in Methods.
Protocol for preconditioning of the device before its characterization	[x] Yes [ ] No	As described in the main text, in the fast-hysteresis measurement, the cell is hold slightly (~ 20 mV) above the VOC (prebias), followed by a reverse and forward sweep with variable frequency or scan speed. The duration of the prebias was 5x longer than the total scan time of the voltage sweep at the respective frequency.
Stability of the J-V characteristic *Verified with time evolution of the maximum power point or with the photocurrent at maximum power point; see ref. 7 for details.*	[ ] Yes [x] No	We note for the reference "83:17" triple cation cells previously showed that the stabilized efficiency (under maximum power point tracking) is essentially equal to the PCE at slow scan speeds where we observe no apparent hysteresis (10.1002/solr.202100772, https://doi.org/10.1038/s41560-018-0219-8). We believe that this is valid for all tested cells in this paper.

3. Hysteresis or any other unusual behaviour

Description of the unusual behaviour observed during the characterization	[x] Yes [ ] No	The hysteresis as a function of scan-speed is subject of this paper and is analyzed throughout the paper, e.g. in Figure 2.
Related experimental data	[x] Yes [ ] No	Experimental data shown in Figure 2 and 4, Figure S4-S7, S9-S10, S12, S13, S14, S20, S23-S25, S29, S31, S32

4. Efficiency

External quantum efficiency (EQE) or incident photons to current efficiency (IPCE)	[ ] Yes [x] No	We note, the EQE data is available for all cells presented in this manuscript, however, the data is not provided with this submission as the manuscript does not present conceptually new devices with exceptional device performances but focuses on the analysis of the perovskite device stability.
A comparison between the integrated response under the standard reference spectrum and the response measure under the simulator	[ ] Yes [x] No	The integrated EQE-Jsc comparison is available for all cells presented in this manuscript, however, the data is not provided as the manuscript focuses on the analysis of the device stability. As an example, for the reference 83:17 triple cation cell, we previously determined the integrated current from the EQE to match the Jsc from the sun simulator (e.g. https://doi.org/10.1038/s41560-018-0219-8).

For tandem solar cells, the bias illumination and bias voltage used for each subcell

☐ Yes ☒ No

N/A

5. Calibration

Light source and reference cell or sensor used for the characterization

☒ Yes ☐ No

An Oriel class AAA Xenon lamp-based sun simulator was used for illumination providing approximately 100 mWcm-2 of simulated AMI.5G irradiation as calibrated using a KG3 filtered Si reference certified from Fraunhofer ISE. The intensity was further monitored simultaneously with a Si photodiode in order to use the exact illumination intensity for efficiency calculations. Specified in Methods, SI.

Confirmation that the reference cell was calibrated and certified

☒ Yes ☐ No

The simulator was recently calibrated with a KG3 filtered silicon solar cell (certified by Fraunhofer ISE). Specified in Methods, SI.

Calculation of spectral mismatch between the reference cell and the devices under test

☒ Yes ☐ No

A spectral mismatch calculation was performed for the reference 83:17 triple cation cell based on the spectral irradiance of the solar simulator, the EQE of the reference silicon solar cell and 3 typical EQEs of the cells. This resulted in 3 mismatch factors of M = 0.9949, 0.9996 and 0.9976. Given the small deviation from unity the measured JSC was not corrected by the factor 1/M. Specified in Methods, SI.

6. Mask/aperture

Size of the mask/aperture used during testing

☐ Yes ☒ No

No mask was used for the fast-hysteresis measurements as these measurements were primarily conducted to analyze the stability of the cells. However, we note that in order to cross-check the FH results at slow scan speeds (10-100 mV/s), standard JV measurements were performed with a Keithley 2400 on the same (6 mm2 sized) cells using a 4.32 mm2 mask, which provided essentially the same results as the FH measurements.

Variation of the measured short-circuit current density with the mask/aperture area

☐ Yes ☒ No

No variation was observed.

7. Performance certification

Identity of the independent certification laboratory that confirmed the photovoltaic performance

☒ Yes ☐ No

This manuscript focuses analysis of the perovskite device stability, certified cells were tested in the revised version. Certified power conversion efficiency of interlayer modified 95:5 TH devices shown in Figure S12 were certified with a PCE of 24.1% in reverse scan and 23.7% in forward scan. Cells shown in Figure S13 were previously certified as reported in ref. (DOI:10.1126/science.adg3755). The certification was performed by the Shanghai Institute of Microsystem and Information Technology (SIMIT).

A copy of any certificate(s)
Provide in Supplementary Information

☐ Yes ☒ No

The interlayer that led to the improved 95:5 TH devices (Figure S12) will be discussed in a separate work. Cells shown in Figure S13 were previously certified as reported in ref. (DOI:10.1126/science.adg3755).

8. Statistics

Number of solar cells tested

☐ Yes ☒ No

We note the aging measurements on the reference 83:17 devices presented in Figure 1 and 2 were repeated multiple times (8x) resulting in qualitatively similar losses and the same trends as reported in the manuscript. Also, the results on the other devices presented in Figure S4-S7 were repeated 2-4x each resulting in very similar losses and the same trends. Similarly, the PLQY (Figure 5) and BACE (Figure 6) measurements were repeated resulting in similar values and same trends as reported.

Statistical analysis of the device performance

☐ Yes ☒ No

No analysis of the device performance is reported as this manuscript but can be provided upon request.

9. Long-term stability analysis

Type of analysis, bias conditions and environmental conditions
For instance: illumination type, temperature, atmosphere humidity, encapsulation method, preconditioning temperature

☒ Yes ☐ No

The cells tested in this study were were subjected to different stability tests: Light aging (Figure 1, Figure 2 and 3a,b, Figure S4-S7, S9, S10, S31), voltage and temperature aging (Figure S23 and S24), stabilized power output (SPO) tracking (Figure 4) and maximum power point (MPP) tracking (Figure S8, S12, S13, S24). The experimental details of all aging tests is provided in the Methods section of the Supplementary Information.

Further reading

1. Shrotriya, V. *et al*. Accurate measurement and characterization of organic solar cells. *Adv. Funct. Mater.* **16**, 2016–2023 (2006).
2. Dennler, G. *et al*. The value of values. *Mat. Today* **10**, 56 (2007).
3. Cravino, A., Schilinsky, P. & Brabec, C. J. Characterization of organic solar cells: the importance of device layout. *Adv. Funct. Mater.* **17**, 3906–3910 (2007).
4. Reese, M. O. *et al.* Consensus stability testing protocols for organic photovoltaic materials and devices. *Sol. Energ. Mat. Sol. C* **95**, 1253–1267 (2011).
5. Snaith H. .J. The perils of solar cell efficiency measurements. *Nat. Photon*. **6**, 337–340 (2012).
6. Luber, E. J. & Buriak, J. M. Reporting performance in organic photovoltaic devices. *ACS Nano* **7**, 4708–4714 (2013).
7. Snaith, H. J. *et al*. Anomalous hysteresis in perovskite solar cells. *J. Phys. Chem. Lett.* **5**, 1511–1515 (2014).
8. Grätzel M. The light and shade of perovskite solar cells. *Nat. Mat*. **13**, 838–842 (2014).
9. Zimmermann E. *et al*. Erroneous efficiency reports harm organic solar cell research. *Nat. Photon*. **8**, 669–672 (2014).
10. Beard M.C., Luther J.M. & Nozik A.J. The promise and challenge of nanostructured solar cells. *Nat. Nanotech*. **9**, 951–954 (2014).
11. Timmreck, R. *et al*. Characterization of tandem organic solar cells. *Nat. Photon.* **9**, 478–479 (2015).

A number of international committees develop industry standards on the characterization of photovoltaic technologies (for example ASTM-E44 and IEC-TC 82), which can provide guidance for academic research.