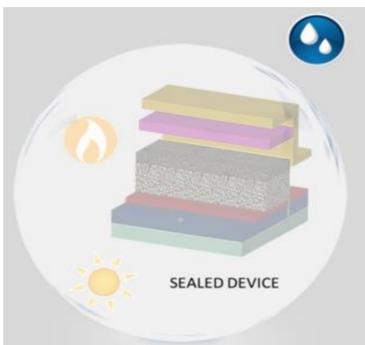


Investigation on the intrinsic stability of large area perovskite solar cell using an optimized sealing procedure

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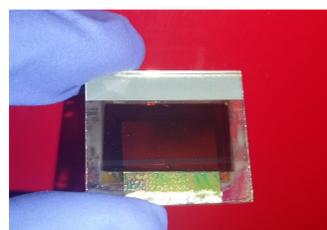
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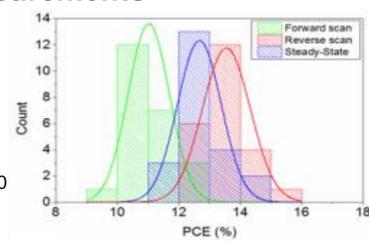
In the booming sector of perovskite solar cells (PSCs), there is a lot of concern about long-term stability of organic-inorganic hybrid materials, and the degradation processes are typically studied often considering only the extrinsic factor, such as the moisture effect and/or the environment during the storage of the unsealed device. In this work, we demonstrate that an efficient sealing strategy is extremely required to avoid the cross talking between extrinsic and intrinsic factors. In this work, we study the intrinsic stability of high efficient mesoscopic $\text{CH}_3\text{NH}_3\text{PbI}_3$ -based PSCs under several stress conditions. Furthermore, the optimized sealing was further evaluated performing accelerated ageing test such as damp-heat, thermal and light soaking. The results show the intrinsic stability of the $\text{CH}_3\text{NH}_3\text{PbI}_3$ perovskite structure and doped Spiro-OMeTAD. The devices maintain more than 70% of the initial efficiency after Shelf life, Humidity Test, Thermal Test at 60°C and Light Soaking Test.

Large area PSC: Materials and I-V measurements



I-V measurement:

- Scan Rate: 32mV/s
- AM1.5G 1 Sun illumination conditions
- Scan direction: reverse and forward scan directions.
- Steady-State: MPP T for 180 s.



PCE Dispersion graphs

Active Area: 1.05 cm²

c-TiO₂: Spray pyrolysis (50nm)

n-TiO₂: Spin coating (180nm)

Perovskite: $\text{CH}_3\text{NH}_3\text{PbI}_3$, solvent engineering method

Antisolvent: Toluene

HTM: doped Spiro-OMeTAD

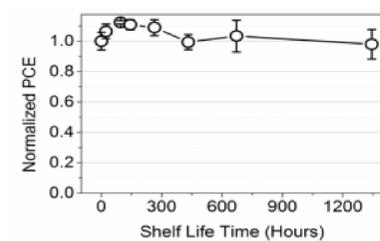
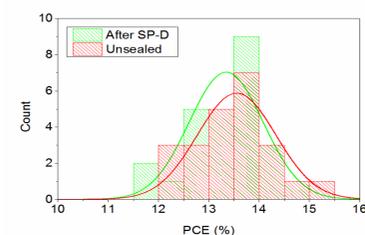
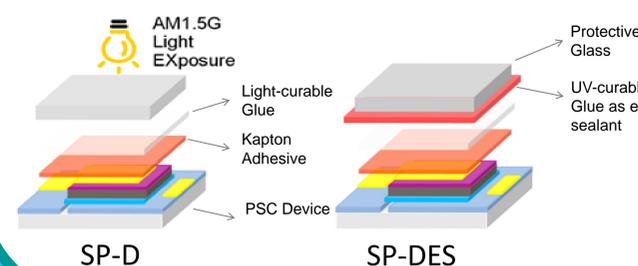
Back Contact: Au Evaporated (80nm)

Active Area	LV measurement	V _{oc} [V]	J _{sc} [mA/cm ²]	FF (%)	PCE (%)
1.05cm ²	RS	0.984 ± 0.014	-20.13 ± 0.66	68.43 ± 2.85	13.55 ± 0.78
	FS	0.988 ± 0.015	-20.78 ± 0.73	53.66 ± 1.74	11.03 ± 0.68
	MPP Tracking	V _{MPP} [V]	J _{MPP} [mA/cm ²]	PCE _{MPP} (%)	
		0.715 ± 0.019	17.70 ± 0.65	12.68 ± 0.71	

Average photovoltaic parameters of 23 large area PSCs measured under AM1.5G 1 Sun illumination conditions under reverse/forward scan directions. The average values of V_{MPP}, J_{MPP} and PCE_{MPP} are also reported.

Sealing Procedure

- No effect of the sealing on the initial PV performance
- Kapton adhesive film was laminated on the device as primary sealing then a protective glass was sealed to the device using a methacrylate glue.
- SP-D was tested under shelf life condition (30%RH in dark) for 1350h without showing detrimental effect on the PV performance.
- An edge sealant was used to protect the edge of the protective glass to limit the moisture entrance.

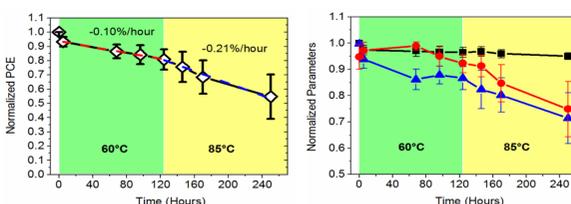


Damp Heat Test

- Tested Devices: Unsealed, SP-D and SP-DES.
- Test conditions: 95% RH @ 40-50°C for 104hours.
- Beneficial effect of the SP-D sealing with an edge sealing (SP-DES).
- Increase of the series resistance (lower FF) due to the heat stress
- No evident perovskite degradation



PCE behavior and images during the damp-heat test with and without sealing process

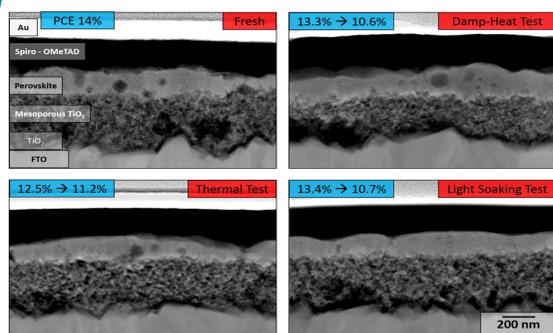


Evolution of the photovoltaic parameters during the thermal test at 60°C (green) and 85°C (yellow)

Thermal Test

- Tested Devices: SP-DES sealing
- Test conditions: in dark @60°C for 124h and then @ 85°C until 250h.
- Linear decrease of the PCE with different slopes: -0.1%/hours @60°C and -0.21 %/hours @85°C.
- Increase of the series resistance (lower FF) due to the detrimental effect of the heat stress on the transport properties of the Spiro-OMeTAD as HTM [1].

TEM investigation

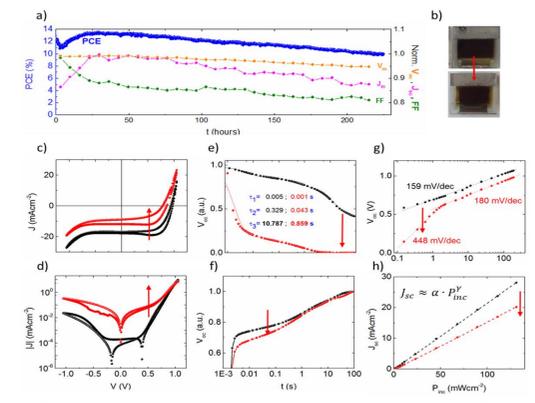


TEM images prior and after the ageing tests

- Scanning Transmission Electron Microscopy (STEM) is performed in order to investigate changes under different stress conditions on four cells sealed with the SP-D method.
- The morphology of the perovskite layer does not exhibit significant changes after 100h damp-heat, 100h thermal and 220h light soaking, confirming the results of the photovoltaic characterization.
- The perovskite capping layer appears quite regular and smooth in the four devices. The infiltration in the titania mesoporous layer is incomplete in some areas due to the the up-scaling of the solvent engineering method to large area cells.
- STEM images do not show Spiro-OMeTAD degradation, sometimes visible as voids induced by air exposure [2].

Light Soaking

- Tested Devices: SP-DES.
- Test conditions: White LED lamp @ 1 Sun illumination condition (I_{LED}=350mA)
- MPP tracking: The dynamic MPP value is acquired using a commercial tool (ARKEO, Cicci Research srl). The I-V characteristic is measured each twenty minutes of the MPP measurement.
- Light stability of the $\text{CH}_3\text{NH}_3\text{PbI}_3$ perovskite without considering the degradation processes induced from UV and IR components.
- The light exposure improves the formation of photo-induced trapping sites in the Perovskite structure as confirmed by transient photovoltage measurements.
- Evident perovskite degradation



MPP tracked Light soaking Test, Images and Transient photovoltage measurement prior and after the light soaking test

Conclusions

The aim of this work is to develop such encapsulation strategy by testing several sealing procedures and comparing them by using accelerated life tests on encapsulated cells. Considering the importance of sealing protocols for the scaling-up of the perovskite technology, we only consider large area cells with an active area of 1.05cm² fabricated by solvent engineering, with an average PCE of 13.6% (batch of 23 PSCs) with a maximum PCE of 15.4%.

The SP-DES is then exploited to assess the intrinsic stability of PSCs under different accelerated life time tests, comprising damp-heat, dry-heat and light soaking. Thanks to the edge sealing, the humidity is not affecting the integrity of the cell and the observed PCE reduction is mainly related to the temperature (40-50 °C). In fact, long time temperature stress is affecting the efficiency of PCE. We find a reduction of PCE of 0.1%/hour at 60 °C and 0.21% at 85 °C, mainly related to the degradation of Spiro-OMeTAD HTL. For both damp heat and thermal tests, there is no visible degradation of the $\text{CH}_3\text{NH}_3\text{PbI}_3$ layer, while, on the opposite, light soaking test induces visible intrinsic degradation of the $\text{CH}_3\text{NH}_3\text{PbI}_3$. A relative reduction of PCE of 0.14%/hour is found for the light-soaking test with a T₈₀ (time to reduce the efficiency to 80% of the initial value) of 140 h. We point out that the proposed cost-effective sealing technique is suitable for the industrial development of perovskite-based optoelectronics devices such as solar cells, led emitting diodes and photo-detectors.

Acknowledgements

F.M., L.C. and A.D.C. acknowledge funding from EU FP7 ITN "Destiny", the European Union's Horizon 2020 framework program for research and innovation under grant agreement No. 653296 (CHEOPS). G.D., S.C. and C.D. acknowledge funding from ERC under grant number 259619 76 PHOTO EM. G.D., P.A.M. and C.D. acknowledges financial support from the EU under grant number 77 312483 ESTEEM2.

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