DELIVERABLE REPORT

VALHALLA

Identify accelerated stress conditions that lead to similar degradation as in real life outdoor stress conditions

Deliverable D4.1
MAY-2024

PREPARED BY
CSEM, UOXF, UVEG, 3SUN
COORDINATED BY
UVEG

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VALHALLA aims to develop perovskite solar cells and modules with power conversion efficiencies above 26 % (modules > 23 %) and extrapolated operational lifetime > 25 years, following an eco-design approach: employing harmful-solvent-free perovskite deposition, optimized use of materials, circularity, recyclability, scalable and low-cost manufacturing processes, to create a viable economic pathway for the European commercialization of this sustainable technology.

VALHALLA is formed by a multi-disciplinary consortium: 12 partners from 8 European countries; 3 industrial partners & 9 RTOs, covering the whole value chain of innovation from research centres to technology providers, end-users and market and policies.

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VALHALLA Consortium

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KAUNO TECHNOLOGIJOS UNIVERSITETAS	KTU	LT
RIJKSUNIVERSITEIT GRONINGEN	RUG	NL
CONSIGLIO NAZIONALE DELLE RICERCHE	CNR	IT
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Table of Content

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Abbreviations and acronyms list

Abbreviation	Meaning	Abbreviation	Meaning
PCE	Power conversion efficiency	Co-Evap A	Solar cells with co-evaporated perovskite employing a doped hole transport layer
FA	Formamidinium	Co-Evap B	Solar cells with co-evaporated perovskite employing phosphonic acid (SAM) hole transport layer
PV	Photovoltaic	S-Evap	Solar cells with a sequentially evaporated perovskite.
MA	Methylammonium	Hybrid	Solar cells with a perovskite prepared using the hybrid method

1. Executive Summary

This document contains experimental stability data on four types of perovskite based solar cells. Two employ co-evaporated perovskites in a fully vacuum deposited stack, one uses sequentially evaporated perovskites, and another one is based on hybrid processed perovskites. The latter cell type was added as a comparison. These cells were encapsulated in a glass-glass sealed package and maintained at maximum power point in outdoor conditions in Neuchatel (Switzerland) and at 60 °C and 1 sun illumination using indoor accelerated stress conditions.
As the outdoor tests were performed in the winter to spring period the stress conditions (sunlight and temperature) were mild. Perhaps due to these relatively mild stress conditions, no significant degradation is observed for the cells employing the co-evaporated and the hybrid processed perovskites. Only the sequentially deposited perovskite degraded rapidly in outdoor conditions, which could be due to issues during the encapsulation.
Under indoor accelerated conditions, however, a drop in efficiency is observed for all these cells. The cells containing the co-evaporated and the hybrid perovskites show a drop in efficiency of approximately 10 % per month.
For one of the co-evaporated perovskite cells, it is possible to infer a gradual decrease in efficiency of 5 % over a period of 1 month in outdoor conditions. As we also have the efficiency drop for these cells when stressed using the indoor accelerated conditions, we can get an estimate of a preliminary acceleration factor of 60.
For a more reliable estimation of the acceleration factor the outdoor monitoring should be extended for longer time.

1.1. Description of the deliverable content and purpose

The purpose of this deliverable is to compare the effect of different stress conditions on the performance of perovskite solar cells as a function of stress time. And in particular to derive a correlation between indoor-accelerated stress conditions to real life outdoor conditions.

We have evaluated three different vacuum processed perovskite based solar cells, two employ co-evaporated perovskites and are prepared by UVEG and one employs a sequentially deposited perovskite as prepared by UOXF. To gather more data, we also evaluated a hybrid (inorganic scaffold sublimed and the organic amine salt by solvent processing) processed perovskite based solar cells prepared by CSEM.

1.2. Encapsulation and test conditions and analysis.

To enable a reliable evaluation of the solar cell performance over time it is essential that they are thoroughly encapsulated to eliminate the contact with moisture and oxygen from the air.
For this CSEM has developed a glass-glass lamination process including edge sealing and a ribbon contacting method so that the perovskite cells can be contacted from outside the laminated package (Fig. 1.1)

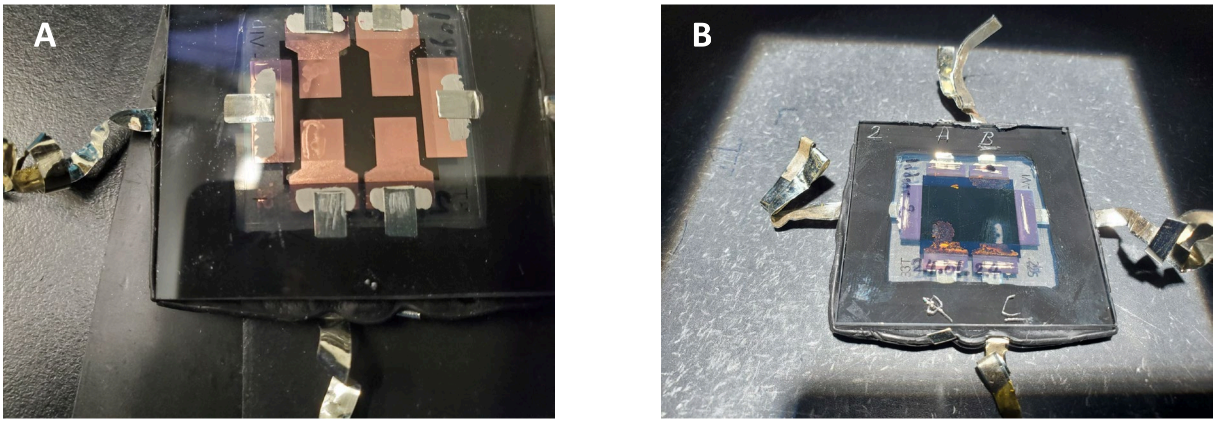

Figure1. Photos of encapsulated solar cells after accelerated degradation (back, A and front, B).

For outdoor testing a continuous monitoring of the irradiance and temperature is performed which is correlated with the power that is generated by the solar cells. The solar cells are maintained at their maximum power point (mpp) using electronic boards and algorithms provided the University of Ljubljana. In figure 2 the irradiances (both the global horizontal irradiance GHI and the plane-of array or global titled irradiance) are shown over several months as obtained in Neuchatel. The temperature on the test rig is also monitored and depicted for the last months in Figure 3. By dividing the generated power of the solar cells by the irradiance data at the same time, the efficiency of the solar cells can be depicted as a function of time.

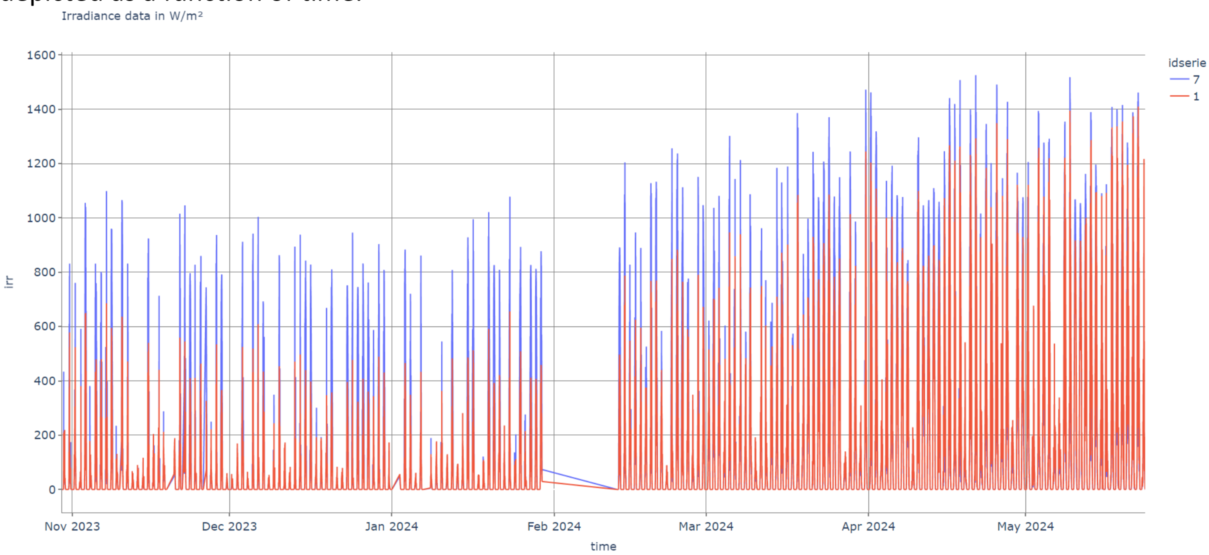

Figure 2. Global horizontal irradiance GHI (red lines) and the plane-of array (aka global titled) irradiance (blue lines) is shown over several months as obtained in Neuchatel.

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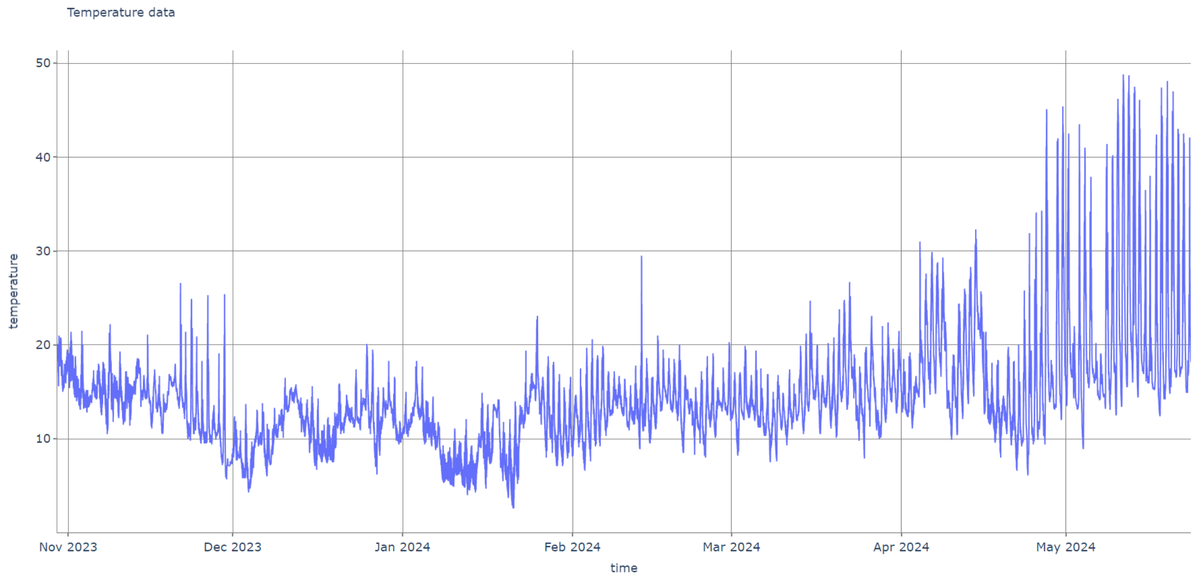

Figure 3. Temperature at the test rig in Neuchatel monitored from November 2023 to May 2024.

Indoor testing is performed using a solar simulator (Lumartix) with an irradiance close to 100 mW/cm^2 and setting the solar cells at a temperature of 60 degrees. Using electronic boards and algorithms provided by Cicci Research, the power that the solar cells generate is monitored over time. From the ratio between the generated power of the solar cells and the irradiance of 100 mW/cm^2 the efficiency of the solar cells is obtained and depicted as a function of time under the accelerated stress conditions (these curves are displayed when the different cells types are discussed below).

1.3. Vacuum processed perovskite solar cells

Bifacial cells prepared by UVEG. Figure 4a shows the stack of the bifacial cells that contain ITO as both bottom and top electrode, with Cu or Ag gridlines on top and employ an organic double layer as the hole selective transporting layer (TaTm) and C60/SnOx bilayer as the electron selective transporting layer. These cells will be referred to in this report as Co-evap type A.

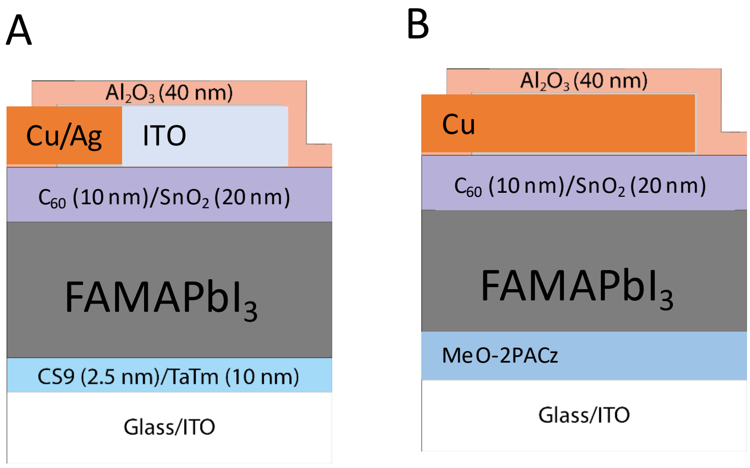

Figure 4. Two device stacks used to evaluate the co-evaporated formamidium-methylamonium lead iodide ($FAMAPbI_3$). A: **Co-Evap A**, a semitransparent cell employing ITO as bottom and top electrode and either Cu or Ag gridlines on the top ITO. The hole selective transport layer is an organic semiconductor bilayer. B: **Co-Evap B**, an opaque cell with an ITO bottom and Cu top electrode. The hole transport layer is a sublimed film of MeO-2PACz.

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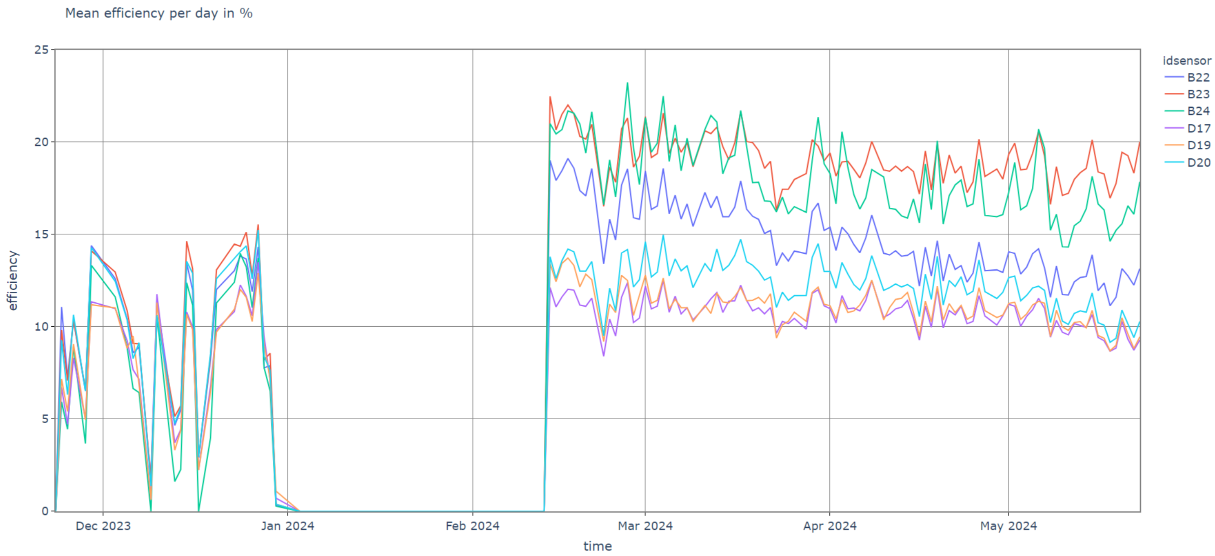

Figure 5. Outdoor performance of the devices Co-Evap A.

During December the Co-evap A cells were occasionally covered in snow and were disconnected from end of December to mid-February. As can be seen from Figure 5, from mid-February to mid-May there is a slow degradation for these types of evaporated cells. The performance is rather noisy and the decrease in efficiency is not very pronounced, yet an estimate can be made, on average showing a decay of -5 % relative per month. We then evaluated the same cells (Co-Evap A) encapsulated using the same glass-glass encapsulation under 1 sun illumination at 60 °C. The evolution of the efficiency over time for these cells is shown in Figure 6 and show the following features; a rapid drop efficiency after turning the devices on which is followed by a more slow but continuous decrease. It takes approximately 24 hours to reach a reduction in efficiency of 10 % relative.

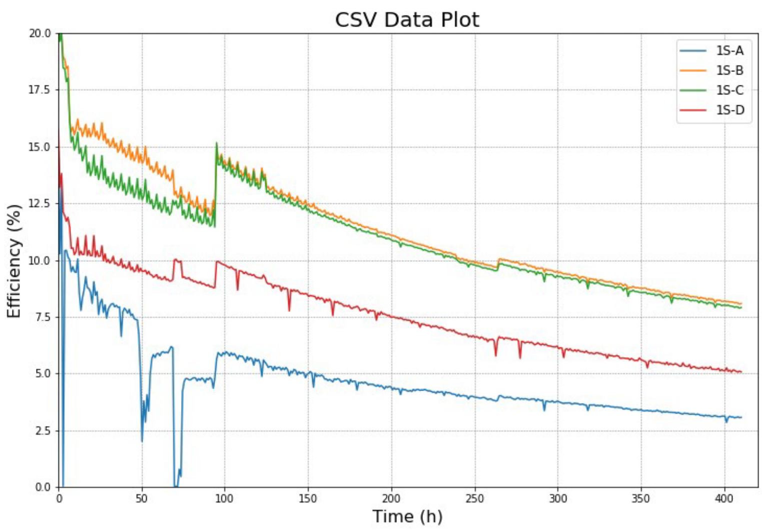

Figure 6. Evolution of the efficiency of solar cells of type Co-Evap A, when kept continuously at mpp under 1 sun illumination at 85 °C.

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Hence, for this type of cells to reduce 10 % relative of efficiency takes 24 hours (1 day) under accelerated stress conditions and approximately 2 months (60 days) under outdoor conditions, this would imply an acceleration factor of 60. However, the performance of this type of devices is atypical as we will comment on now.
We analysed these cells after the first five weeks (from November 2023 to January 2024) we found a pronounced S-shape in the J-V curves which leads to a low FF and therefore a low efficiency (Figure 7). Remark: the encapsulated cells were evaluated without mask which is why the current density is overestimated. Yet the appearance of the cells was unchanged and no discoloration of the perovskite film can be observed.

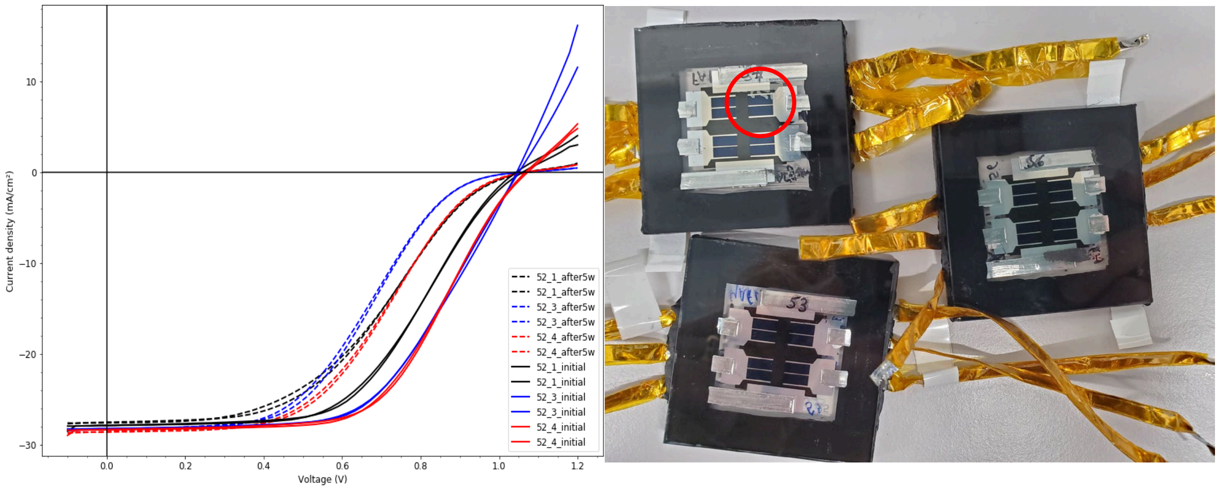

Figure 7, left J-V curves of encapsulated Co-Evap A type cells under 1 sun illumination (tested without an illumination mask, which explains the too high current density) directly after encapsulation and after 5 weeks in outdoor at CSEM test station in Neuchatel. Right photographs of cells after 5 weeks in outdoor at CSEM test station in Neuchatel, no visible sign of degradation except for the delamination of a top electrode in one of the cells (red circle).

We attribute this to the formation of a charge extraction barrier, most likely at the HTL-ITO interface. Nevertheless, these cells where reconnected to the mpp tracking system and tested for another three months. It is rather surprising that under outdoor conditions the cells are still functioning and we were able to collect the efficiency data depicted in Figure 5.
Hence, this needs to be evaluated further, as it may lead to information regarding the degradation mechanism in these devices. Thus, in view of this atypical behaviour the acceleration factor for the devices of type Co-Evap A solar cells is most likely not representative for vacuum deposited perovskite solar cells.

Therefore, we evaluated the second type of co-evaporated perovskite containing solar cells, that employed a different hole extraction layer (MeO-2PACz sublimed to a layer thickness of 1.5 nm) and has a copper top electrode.
The outdoor evolution of the efficiency is depicted in Figure 8, and as can be observed most cells do not show any significant reduction in efficiency over the test period of 3 months (from mid February to mid May). This is different when these cells were stressed indoor at under 1 sun illumination at 60 °C (Figure 9).

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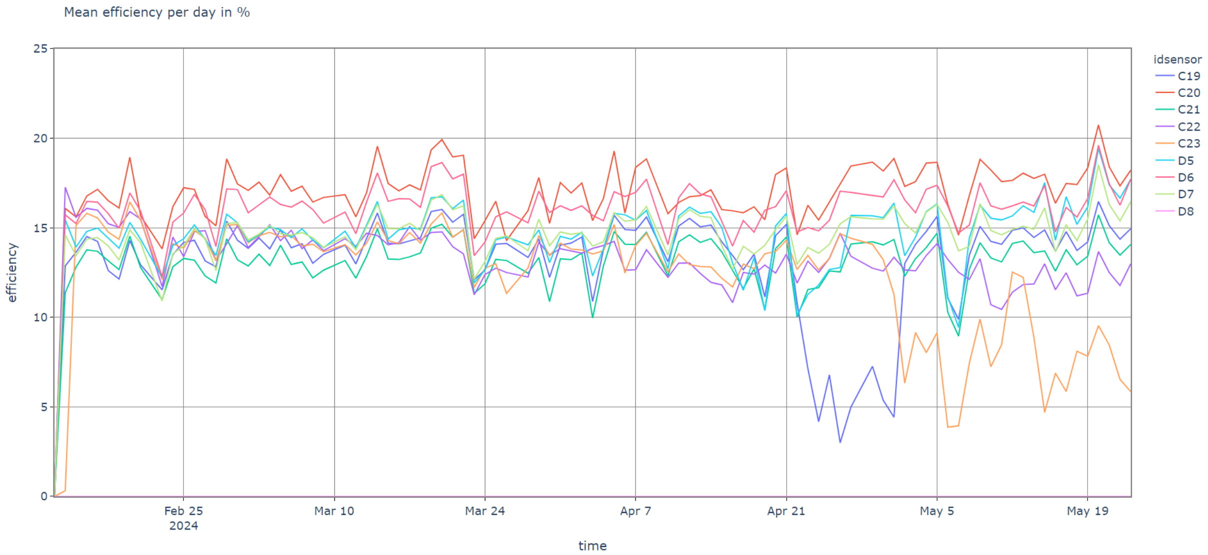

Figure 8. Outdoor performance of the devices Co-Evap B.

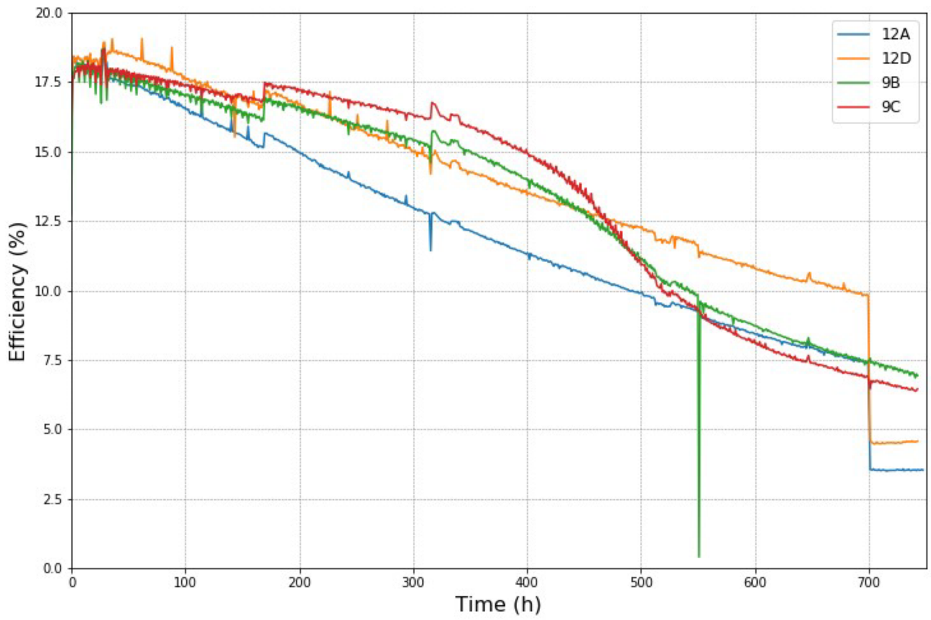

Figure 9. Evolution of the efficiency of solar cells of type Co-Evap B, when kept continuously at mpp under 1 sun illumination at 60 °C.

At UOXF vacuum deposited solar cells were prepared using sequential deposition process (S-Evap) in which first the inorganic precursors are evaporated followed by the organic precursors and an annealing step. This method leads to solar cells with high power conversion efficiencies. The device stack and a schematic of the sublimation procedure is shown in Figure 10.

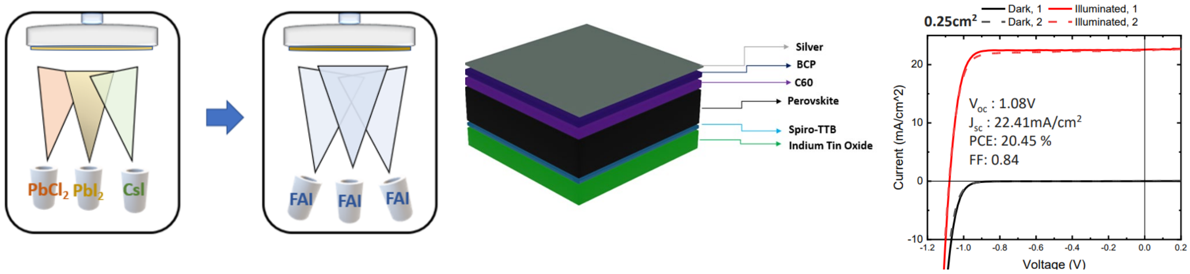

Figure 10. Left. Schematic of the two-step process sublimation process. Middle. Solar cell stack used for the sequentially deposited perovskites (**S-Evap**). Right. J-V curve of the cells directly after their preparation both without and with 1 sun illumination.

Figure 11. Outdoor performance of S-Evap cells from UOXF. three cells degrade in one day, the other three within one week. On the right a photograph of the cells after one week of outdoor stressing. The dark colour, representative of the perovskite layer, is gradually disappearing.

The S-Evap devices, suffered from a rapid decrease in outdoor performance. This is related to the disintegration of the perovskite film as evidenced by the retreating dark coloured regions in the cells (Figure 11 right). This could be related to a not optimized encapsulation protocol as the substrate thickness was different then that used for the Co-Evap and Hybrid solar cells.

1.4. Hybrid processed perovskite solar cells

At CSEM solar cells of type Hybrid were prepared using a sequential approach based on co-evaporation of PbI_2 and CsBr, followed by slot-die coating of FAI and FABr solution in alcohol and an annealing step. 16 pixels cells of 1 cm^2 are fabricated on a 10x10 cm^2 area. The device stack and a schematic of the hybrid fabrication process are shown in Figure 11.

For these devices the outdoor performance has been monitored since about 5 weeks and the cells do not show any signs of degradation yet (Figure 12). The cells stressed indoor under 1 sun and 60°C instead show a degradation of -10% after 150h of testing (Figure 13). After 800h of testing the cells did not show any apparent sign of degradation, despite the loss of -25% efficiency.

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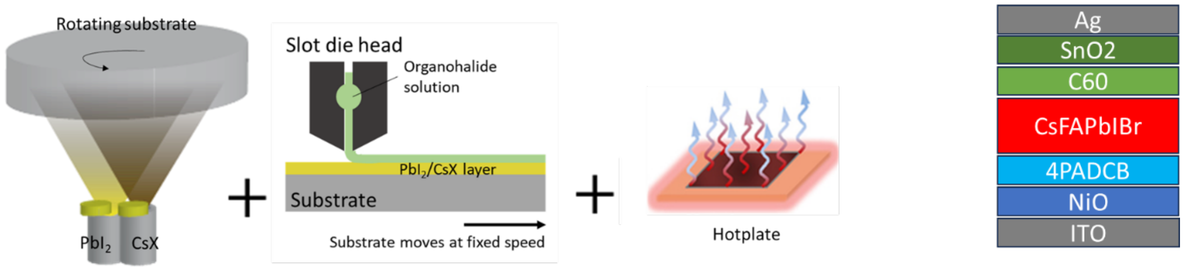

Figure 11. Schematic of the two-step hybrid process used for CSEM cells of type Hybrid (left) and cell stack (right).

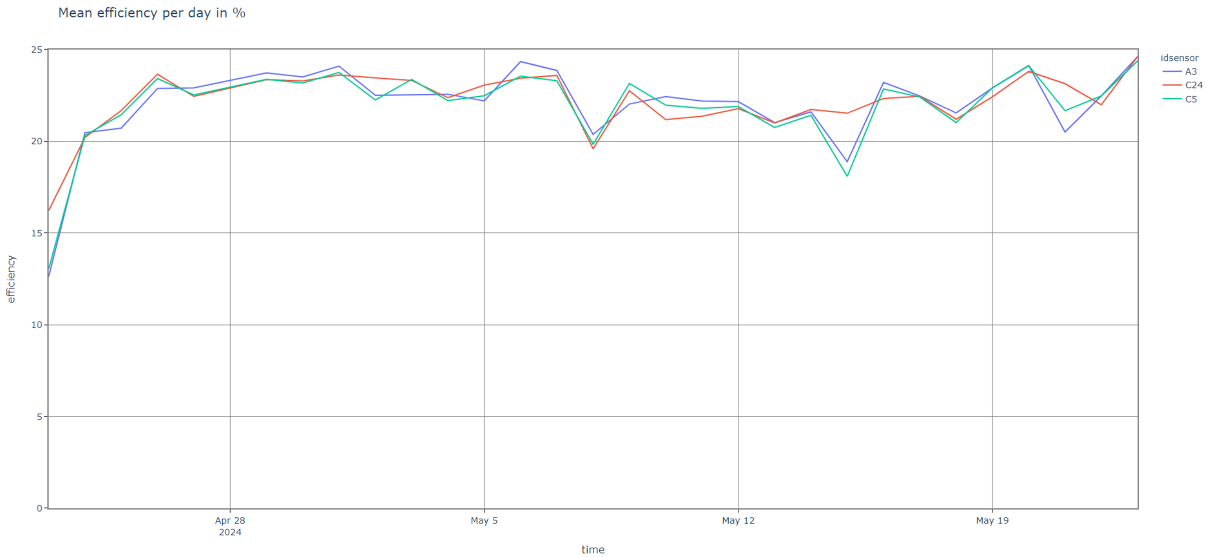

Figure 12. Outdoor performance of the devices of type Hybrid from CSEM.

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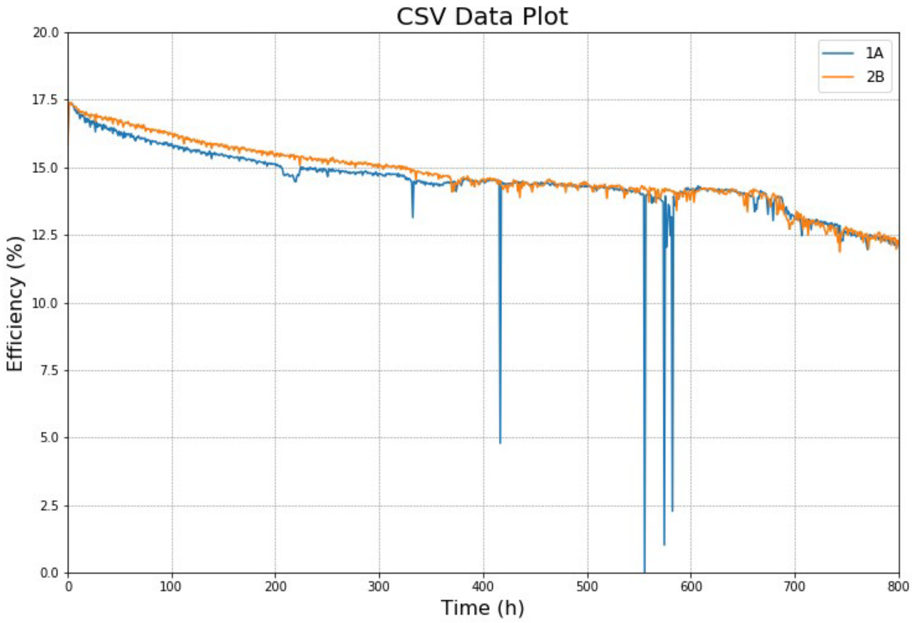

Figure 13. Evolution of the efficiency of solar cells of type Hybrid, when kept continuously at mpp under 1 sun illumination at 60 °C.

1.5. Acceleration factor

To determine a meaningful acceleration factor the solar cells should show a drop in efficiency under both the real world outdoor and the accelerated indoor conditions. From the cell types we evaluated, one degraded very fast (S-evap) probably due to encapsulation issues and is therefore not a representative cell. The only cell type that showed some decrease in efficiency under outdoor conditions was of type Co-evap A, however, for these cells we showed from J-V analysis that the fill factor dropped significantly indicative of a charge extraction barrier. Therefore, the reduction in outdoor performance is most likely related to the formation of an extraction barrier. This is corroborated by the performance of the cells Co-evap B, in which a different hole extraction layer is employed and which did not show a decrease in performance under outdoor conditions.
Thus, Co-evap B and the hybrid cells do not show a decrease in performance under outdoor conditions, which is positive news, yet it also implies that for these cells no acceleration factor can be obtained. Hence, we used the Co-evap A cells to estimate a first value of the acceleration factor. The outdoor efficiency of this type of cells shows a gradual decrease of 5 % over a period of 1 month. As we also have the efficiency drop for these cells when stressed using the indoor accelerated conditions, we can get an estimate of a preliminary acceleration factor of 60.

1.6. Relation with other activities in the project

This deliverable is related to task 4.1 and is meant to provide some guidance on how to stress perovskite solar cells in indoor conditions such that the results on degradation are meaningful to predict the stability of these cells under outdoor real-world conditions. Performance and stability data collected in this report will provide the necessary background information for the following WP4 activities, and they will be fed back to inform the fundamental work done into WP1, 2 and 3.

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2. Conclusions

This deliverable aims to identify optimal accelerated stress conditions that mimic real-world outdoor scenario for perovskite solar cells. With this purpose in mind, the most significant results obtained so far from preliminary outdoor and indoor stress tests performed on four different types of perovskite cells (co-evap A, co-evap B, s-evap, and hybrid) have been collected and compared. Key findings include:

- Cells using both co-evap and hybrid processed perovskites showed minimal degradation under outdoor mpp tracking during the mild winter-to-spring period in Neuchatel. Only the s-evap perovskite cells exhibited rapid degradation, likely due to encapsulation issues.
- On the contrary, selected indoor accelerated stress conditions (60 °C, 1 sun illumination) led to significant efficiency drops across all cell types.
- Co-evap A cells exhibited atypical performance with S-shaped J-V characteristics after spending 5 weeks outdoor, which deserves further investigation.
- Co-evap A cells also showed a gradual 5% efficiency drop over a 2-month period outdoor, and a 10% efficiency decrease after 24 h under indoor stress. Based on these data, a preliminary acceleration factor of 60 has been estimated.

Due to the atypical performance of co-evap A cells, the estimated acceleration factor may not be fully representative of all vacuum deposited perovskite solar cells. Moreover, the lack of significant degradation for the other cell types under outdoor conditions prevented us from identifying a meaningful correlation between outdoor and indoor data. However, it can be stated that for properly encapsulated perovskite solar cells, a stress test of 500 hours in MPPT at 60°C and at 1Sun is representative for a period of time of several months at least. To enhance reliability and refine our estimates, further outdoor stress testing is necessary and will be performed in WP4, particularly by exposing cells to longer and/or more severe outdoor stress conditions (to let them degrade more). These additional tests will provide a more robust assessment of the acceleration factor and guide future design improvements toward robust performance in real-world applications.

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