DELIVERABLE REPORT

VALHALLA

Develop a highly efficient (>26%) cell technology which surpasses the stability thresholds which can be transferred to WP3 and 4 for further development in the project

Deliverable D2.2
JUNE-2025

PREPARED BY
UOXF
COORDINATED BY
UVEG

Funded by
the European Union

VALHALLA is a 3-year research and innovation project funded by the European Commission through the Horizon Europe Research and Innovation Action (RIA) grant N°101082176, responding to the call for a "*Sustainable, secure and competitive energy supply*" (HORIZON-CL5-2021-D3-03).

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.

Project info	101082176 – VALHALLA – (HORIZON-CL5-2021-D3-03)
Deliverable Title	D2.2 : Develop a highly efficient (>26%) cell technology which surpasses the stability thresholds which can be transferred to WP3 and 4 for further development in the project
Lead Beneficiary	UOXF
Authors	Henry Snaith, Barkha Tyagi, Lidon Gil-Escrig, Manuel Piot, Michele Sessolo and Henk J. Bolink
Approved by	Annamaria Petrozza
Dissemination level	Public
Due date	30th June 2025
Submission date	16 of July 2025
Version	2
Linked to WP - task	WP3 and WP4 – all tasks

Funded by the European Union

This project has received funding from the European Union's Horizon Europe research and innovation programme under grant agreement No. 101082176

VALHALLA Consortium

Organization name	Short name	Country
UNIVERSITAT DE VALENCIA	UVEG	ES
KAUNO TECHNOLOGIJOS UNIVERSITETAS	KTU	LT
RIJKSUNIVERSITEIT GRONINGEN	RUG	NL
CONSIGLIO NAZIONALE DELLE RICERCHE	CNR	IT
FONDAZIONE ISTITUTO ITALIANO DI TECNOLOGIA	IIT	IT
UNIVERSITE DE LIEGE	ULiège	BE
THE CHANCELLOR, MASTERS AND SCHOLARS OF THE UNIVERSITY OF OXFORD	UOXF	UK
3SUN SRL	3SUN	IT
TEKNOLOGIAN TUTKIMUSKESKUS VTT OY	VTT	FI
ICARES CONSULTING/BECQUEREL INSTITUTE FRANCE	BI/BI F	BE/FR
ARK METRICA LTD	ARKM	UK
CSEM CENTRE SUISSE D'ELECTRONIQUE ET DE MICROTECHNIQUE SA - RECHERCHE ET DEVELOPPEMENT	CSEM	CH

How to Cite
Henry Snaith, Barkha Tyagi, Lidon Gil-Escrig, Manuel Piot, Michele Sessolo and Henk J. Bolink (2025). Deliverable 2.2 Report: Develop a highly efficient (>26%) cell technology which surpasses the stability thresholds which can be transferred to WP3 and 4 for further development in the project. in Project VALHALLA: Perovskite Solar Cells with Enhanced Stability and Applicability (No. 101082176). European Union. PUBLIC [Communicate on 16/07/2025].

Funded by the European Union
This project has received funding from the European Union's Horizon Europe research and innovation programme under grant agreement No. 101082176

Table of Content

Funded by the European Union

This project has received funding from the European Union's Horizon Europe research and innovation programme under grant agreement No. 101082176

Abbreviations and acronyms list

Abbreviation	Meaning	Abbreviation	Meaning
PCE	Power conversion efficiency	Cs	Cesium
FA	Formamidinium	ITO	Indium Tin Oxide
PV	Photovoltaic	TCO	Transparent Conductive Oxide
MA	Methylammonium	Eg	Bandgap
eV	Electron volt	FF	Fill Factor
Voc	Open circuit voltage	Jsc	Short circuit current

1. Executive Summary

1.1 Description of the deliverable content and purpose

This document describes the achieved performances of solar cells containing perovskites fabricated using two strategies: co-evaporation and sequential evaporation. In co-evaporation the perovskites are prepared by simultaneous sublimation of the precursors, whereas in sequential evaporation first the inorganic precursors are sublimed into a thin film which is then converted into a perovskite film when the organic precursor is sublimed on it, followed by an annealing step. The studied solar cells contained perovskites with different combinations of precursors, hence with different A, and X materials in the ABX_3 perovskite structure. We compare the achieved performances and stabilities using both the mentioned strategies.
From this analysis we conclude the most promising strategies, devices structures, and fabrication conditions to reach the targeted power conversion efficiency of 26%. Additionally, we outline the roadmap for technical improvements that would enable us to reach that target.

The purpose of this deliverable is to describe the efforts in VALHALLA to achieve highly efficient and stable solar cells based on vacuum deposited perovskite films. The best performing solar cell architectures have been transferred to WP3 and WP4 for fabrication of encapsulated mini-modules and for more extensive stability tests and energy yield assessment. In this deliverable the efficiency of some of the best solar cells are described including some of their stability data obtained under laboratory based accelerated stress conditions. As we described in D4.1, the correlation between outdoor and indoor obtained stability data is not yet conclusive due to a lack of degradation observed under outdoor conditions.

The challenge is to the find the optimum device architecture that can achieve high energy yield and stability. To do so, we have fabricated devices under different annealing conditions and employed various device structures. This approach provides a simple yet effective way to identify the limiting factors and which component of the device architecture and materials deserve further optimization.

Funded by the European Union
This project has received funding from the European Union's Horizon Europe research and innovation programme under grant agreement No. 101082176

2. Results

2.1 Intermediate bandgap (Eg around 1.55 eV) Formamidinium and methylammonium-based sequentially evaporated perovskite solar cells .

At UOXF the perovskite solar cells that we investigate in this study are fabricated with an all thermally evaporated device stack of: glass/indium-tin-oxide (ITO)/2,2′,7,7′-tetrakis(*N*,*N*′-di-*p*-methylphenylamino)–9,9′-spirobifluorene (Spiro-TTB)/perovskite/C_{60}/bathocuproine (BCP)/silver (Ag), where Spiro-TTB is the hole transport layer (HTL) and C_{60} is the electron transport layer (ETL).

The sequentially evaporated perovskite films were then integrated into these stacks, as seen in Fig. 1(A). The perovskite layers were post-annealed in two distinct environments: under a nitrogen glovebox environment (0% RH) and in ambient conditions with a controlled 35% relative humidity (35% RH). To assess the photovoltaic characteristics of these solar cells, we conducted current density–voltage (J–V) scans, maximum power point (MPP) tracking, as well as steady-state open-circuit voltage and short-circuit current measurements, as illustrated in Fig. 1(B) and (C). Our 0% RH annealed champion device with an estimated bandgap of 1.56 eV from EQE exhibited an open-circuit voltage (Voc) of 1.02 V, a current density (Jsc) of 21.6 $mAcm^{-2}$, a MPP efficiency of 18.0%, and a corresponding calculated “fill factor” (FF) of 0.82. Encouragingly, the champion device containing a perovskite film annealed under 35% RH with an estimated bandgap of 1.55 eV from EQE had an improved MPP efficiency of 21.0 %. This was the result of a Voc of 1.05 V, a Jsc of 24.3 $mAcm^{-2}$ and a calculated FF of 0.83. In Fig. 1(D) we show the statistical distribution of the MPP efficiency for a batch of 15 devices of each type. When annealed in controlled humidity the mean efficiency increases from 14% to 20%, with the distribution significantly narrowing. The external quantum efficiency (EQE) spectra for a representative 0% RH and 35% RH annealed device are shown in Fig. 1(E), where the integrated current density of 21.3 mA cm^{-2} and 24.1 $mAcm^{-2}$ for the solar cell fabricated under 0% RH and 35% RH closely matched the device's current density of 21.6 mA cm^{-2} and 24.3 $mAcm^{-2}$, measured with the solar simulator. These results highlight the strong impact of controlled relative humidity during perovskite film annealing upon the photovoltaic performance. Importantly, the statistics we show in Fig. 1(D) also highlight the importance of this processing step upon improving the reproducibility and narrowing the performance spread in the devices.

To test the stability of our all-vacuum processed perovskite solar cells under elevated temperature (85 °C – as measured with a black standard temperature sensor located in the plane of the test cells in the aging box) and full spectrum simulated sun light, held under open-circuit using cells with SnOx/Cr/Au top contacts (as opposed to BCP/Ag), as we illustrate in Fig. 1(A). We show the mpp efficiency as a function of ageing time in Fig. 1(F), where we observe that both sets of devices exhibited an initial “burn-in” phase, leading to a significant early decrease in median performance, which has also been observed in other studies. The set of devices annealed under nitrogen, experienced a substantial performance drop of ∼7% in absolute efficiency under this initial burn-in phase, whereas those fabricated at 35% RH, showed a smaller decline of ∼4% in absolute efficiency. After 200 hours of ageing, the nitrogen-annealed devices showed a 92% relative drop in performance, whereas the devices annealed under 35% RH only dropped by 32% relative to their initial median performance.

Funded by the European Union
This project has received funding from the European Union’s Horizon Europe research and innovation programme under grant agreement No. 101082176

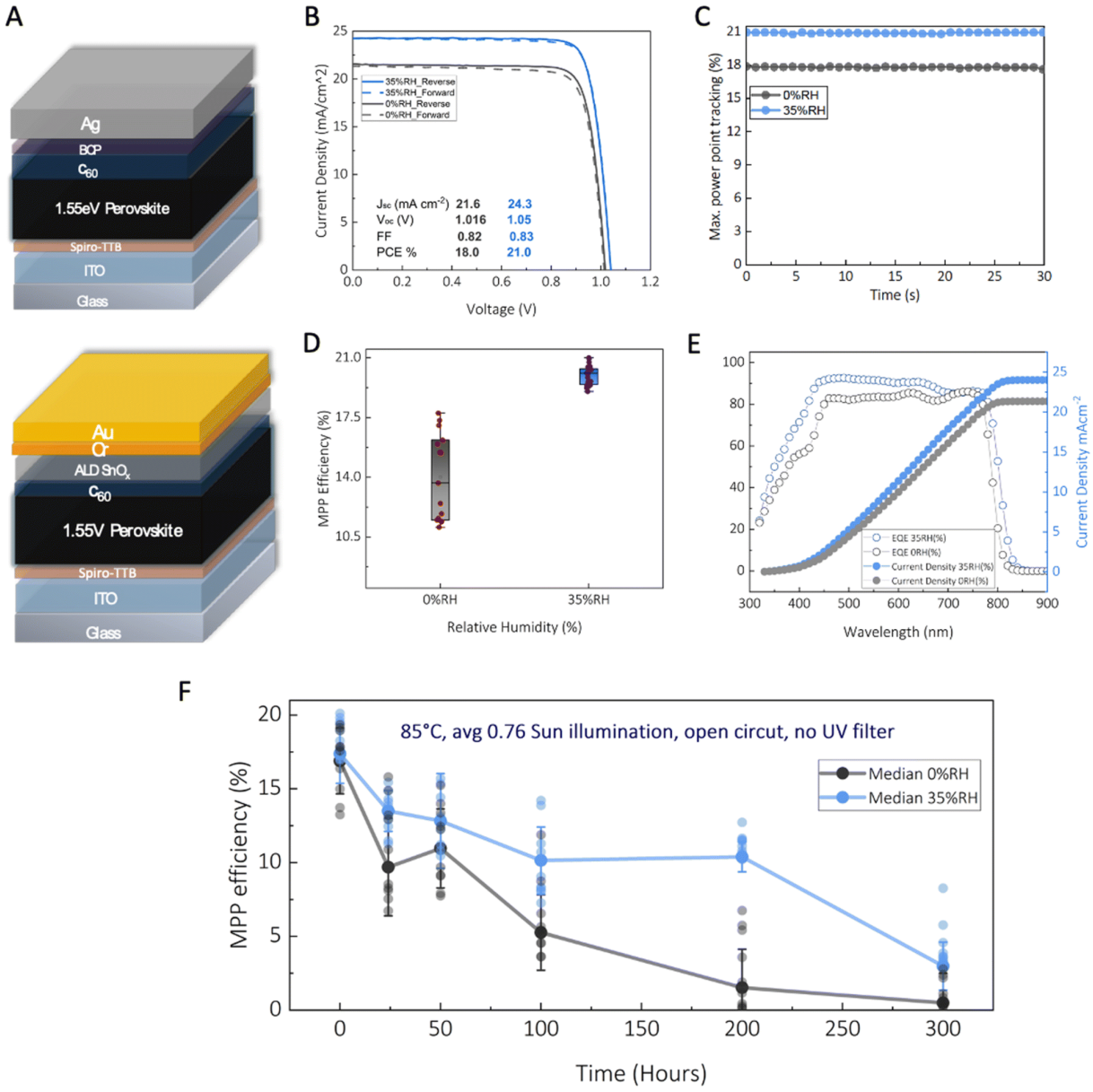

Figure 1: (A) fully vacuum deposited device stack using BCP/Silver or SnO_x/Cr/Au top contacts. (B) JV curves under simulated sunlight of devices fabricated where annealing is done in presence of 0% RH (nitrogen) and 35% RH (controlled humidity air). (C) Maximum power point (mpp) tracking of champion devices obtained from nitrogen (0% RH) and 35% RH annealing, respectively. (D) Statistical comparison of mpp efficiency obtained from mpp tracking. (E) External quantum efficiency of a device where the perovskite is annealed under 0% RH and 35% RH. (F) MPP efficiency as a function of time for cells aged under open-circuit at elevated temperature under light (85 °C, 0.76 suns illumination from a xenon lamp aging box with no UV filter) ~ISOS-L-2, where the perovskite was annealed under 0% RH in nitrogen and 35% RH in air. The cells were occasionally removed from the ageing rig, allowed to cool to room temperature, and measured under AM1.5 100 mW cm^{-2} simulated sun light to determine the photovoltaic performance parameters, including the mpp efficiency. The error bars represent the standard deviation about the median (solid dark symbol). The lighter symbols are data points.

Funded by the European Union

This project has received funding from the European Union's Horizon Europe research and innovation programme under grant agreement No. 101082176

2.2 Methylammonium-based sequentially evaporated perovskite solar cells.

At UVEG, sequentially evaporated perovskites were also studied. The device stack used was as follows: glass/indium-tin-oxide (ITO)/a hole transport layer/perovskite followed by passivation/C_{60}/Tin oxide (SnO2)/Copper (Cu), where C_{60} is the electron transport layer (ETL) as depicted in the schematic diagram in Fig 2 (A). The perovskite we focussed on is methylammonium lead iodide ($MAPbI_3$) with a bandgap around 1.55 eV. solar cells that we investigate in this study are fabricated with an all thermally evaporated layer stack of: Using a passivation approach we were able to achieve PCE>18% as can be seen from the current vs voltage curve plotted in Fig 2 (B). In Fig. 2C the EQE spectra of the solar cells is depicted, where the integrated current density of the devices was calculated to be 19.32 mA/cm2 which agrees with the Jsc values obtained from the J-V curves. Fig 2(D) shows statistical distribution of devices parameters, from where we can conclude excellent device reproducibility and yield over 24 devices.

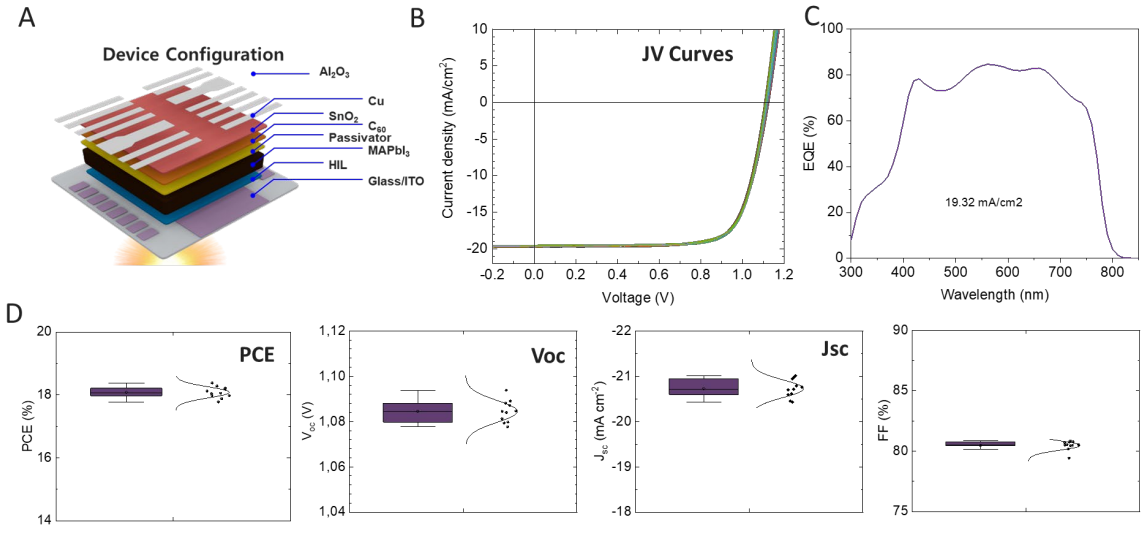

Figure 2 (A) Fully vacuum deposited device stack using Cu as top electrode. (B) JV curves under AM1.5 100mWcm^{-2} simulated sunlight of the solar cells containing the sequentially sublimed $MAPbI_3$ perovskites. (C) External quantum efficiency of a device. (D) Statistical comparison of the device parameters.

These $MAPbI_3$ based solar cells prepared using sequential sublimation of the PbI_2 and MAI were firstly capped with an ALD deposited layer of alumina, and then encapsulated using a UV curable resin and a top glass plate. The pixels were contacted using soldered copper wires. These cells were than stressed at 75 °C and under 1 sun equivalent illumination while maintaining the cells under maximum power point. These cells exhibited excellent stability with a gradual decline, with 90 % of the initial power efficiency maintained after 1000 hours of operation, as we show in Figure 3.

Funded by the European Union

This project has received funding from the European Union's Horizon Europe research and innovation programme under grant agreement No. 101082176

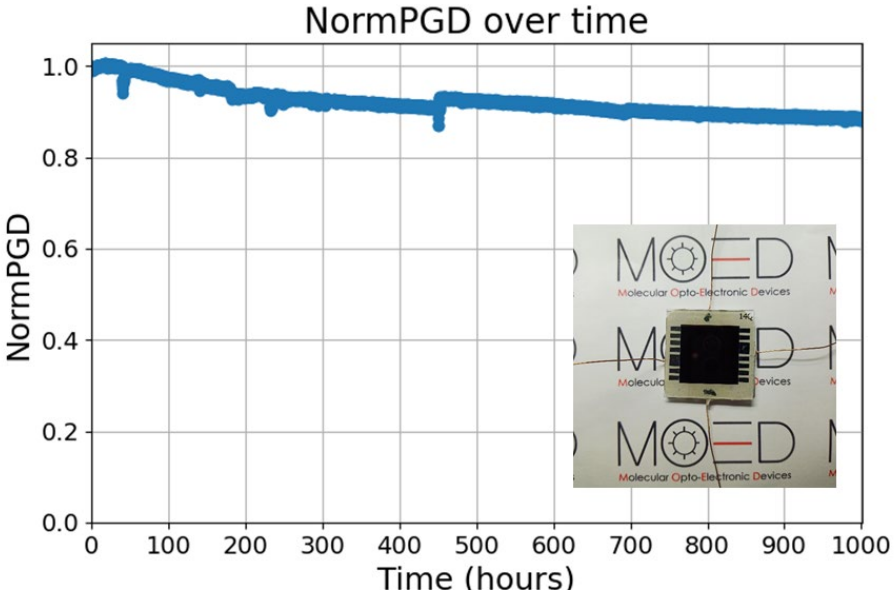

Figure 3 Normalised power generation density (PGD) of the solar cell over a time period of 1000 hours while exposed to 1-sun irradiance at 75C, inset shows an image of the encapsulated device.

2.3 Formamidinium and methylammonium-based co-evaporated perovskite solar cells

At UVEG formamidinium based co-evaporated perovskite were developed. The device structure is similar to the one in Fig 2(A), except the perovskite used is $FAMAPbI_3$. This perovskite was prepared by simultaneous (co-) sublimation of the MAI, FAI and PbI_2 perovskite precursors using three separate sublimation sources. The J-V curves in Fig 4(A) present an outstanding efficiency of 23.24%, with a Voc of 1.12 V, Jsc of 25.5 mA/cm^2, and a FF of 81.6%. The Figure 4(B) shows the EQE spectra of the champion device. Fig 4(C) shown statistical distribution of devices parameters, from where we can conclude excellent device reproducibility and yield.

Funded by the European Union

This project has received funding from the European Union's Horizon Europe research and innovation programme under grant agreement No. 101082176

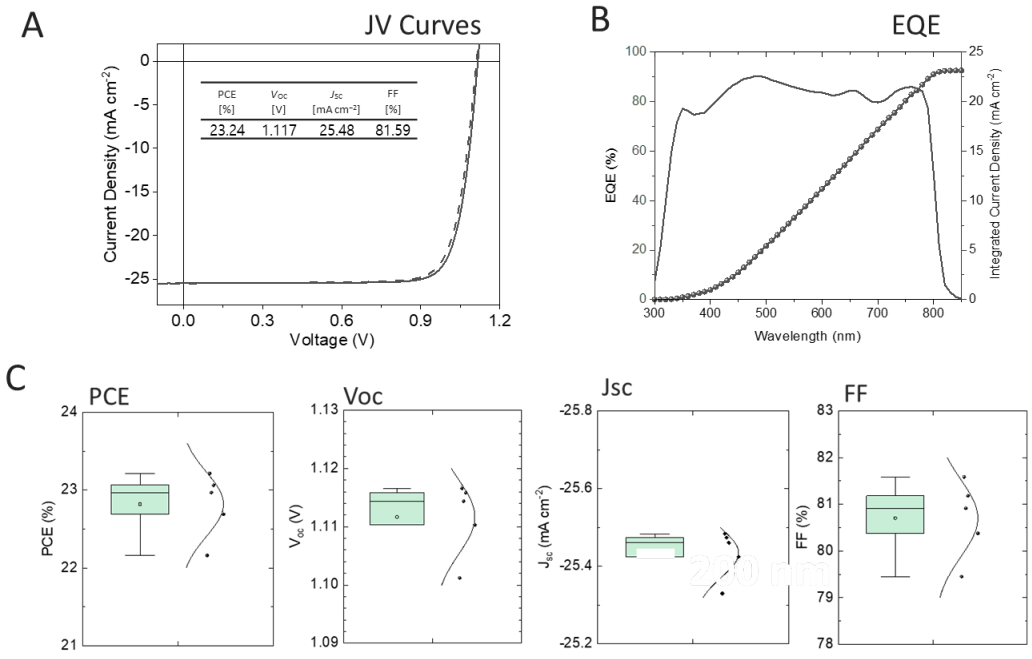

Figure 4 (A) J-V curves under simulated sunlight of $FAMAPbI_3$ prepared by co-sublimation based devices fabricated under simulated 1.5. (C) External quantum efficiency of the device. (D) Statistical comparison of the device parameters.

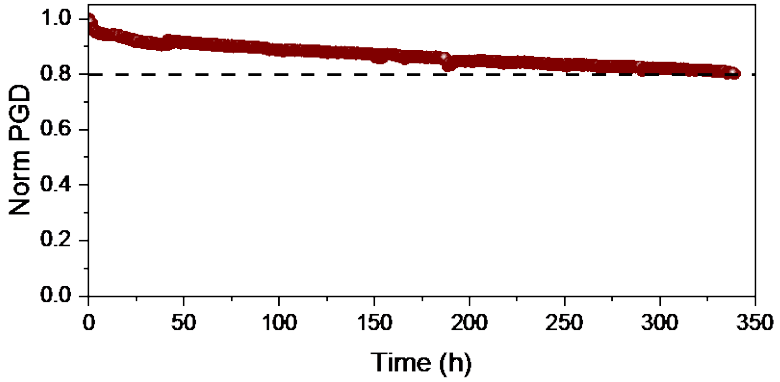

Figure 5 Normalised PGD of the devices over time period of 350 hours under 1-sun irradiance at 75C.

2.4 Methylammonium-free co-evaporated perovskite solar cells.

At UOXF, co-evaporated perovskite was developed using FA and Cs based perovskites. All thermally evaporated layer stack of: glass/indium-tin-oxide (ITO)/2,2′,7,7′-tetrakis(N,N′-di-p-methylphenylamino)–9,9′-spirobifluorene (Spiro-TTB)/perovskite/passivation/LiF/C_{60}/Tin oxide (SnO2)/silver (Ag), as shown in Fig 6(A). Here, LiF is used as a buffer layer for better energetic alignment at the perovskite and C60 interface and reduced non-radiative recombination. The perovskite was annealed in air relative humidity >28% RH. New ammonium salt-based materials were explored for evaporability and one was found to be effective in passivating the perovskite surface and improving the device performance.

Funded by the European Union

This project has received funding from the European Union's Horizon Europe research and innovation programme under grant agreement No. 101082176

The champion devices yielded Jsc of 22.5 mA/cm2, Voc of 1.05 V, and FF of 81.6% achieving PCE of 19.27% as displayed in the J-V curve in Fig 6(B). The device also displayed MPP efficiency values of 19% over a time period of 30 sec (Figure 6 (C)).

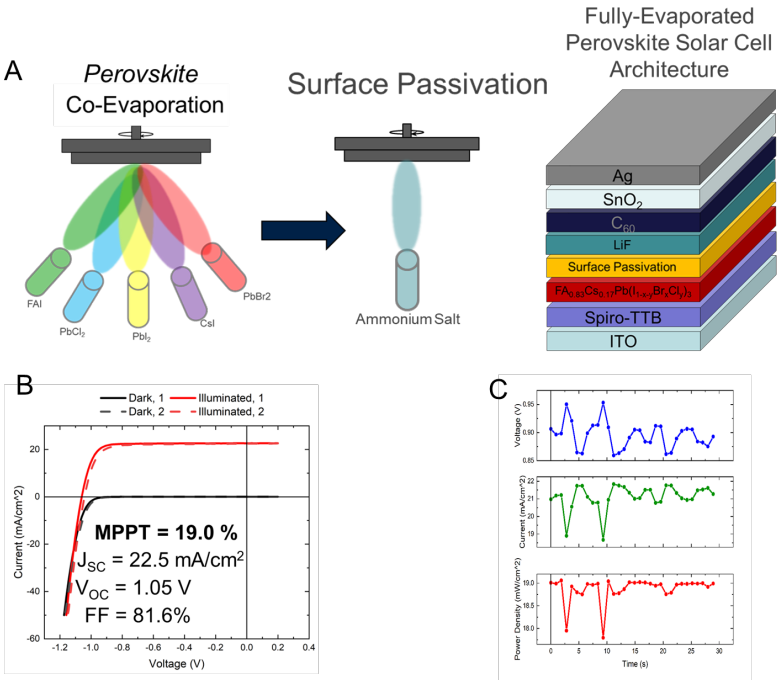

Figure 6 (A) Schematic illustration of the perovskite co-evaporation scheme followed by surface passivation and the device structure of the FACs based device's structure. (B) JV curves of FACsPb($I_{1-x-y}Br_xCl_y$)$_3$ based devices fabricated. (C) mpp tracking of champion devices.

2.5 Relation with other activities in the project

The results described in this deliverable have a strong link with WP1, where the materials were developed that are used to prepare the solar cells. Additionally, these best performing (in terms of efficiency and stability) devices have been transferred to WP3 and WP4 for integration into mini-modules and for more extensive stability evaluation both indoor and outdoor.

3. Conclusions

This deliverable shows that the solar cells employing vacuum deposited metal halide perovskite can have high efficiency and substantial stability. Two different deposition methods have been evaluated, namely sequential and co-sublimation of the precursors. At this stage it is not clear which of the two methods leads to better performance. The highest power conversion efficiency of 23.2% was achieved for a cell employing perovskite prepared by co-sublimation of the three precursors. However, for the methylammonium-free compositions, the so-far best performance was obtained for solar cells that employ a perovskite prepared using sequential deposition.

We also demonstrate that post deposition treatments such as annealing and exposure to specific environments affect the performance of the solar cells. In the remainder of the project we will aim to

Funded by the European Union
This project has received funding from the European Union's Horizon Europe research and innovation programme under grant agreement No. 101082176

increase the device efficiency by implementing bottom, top and bulk passivating molecules as one of the device main bottlenecks is related with the limited open circuit voltage.

As far as stability goes, we appear to obtain slightly better results for the co-evaporated perovskite cells, than the sequentially evaporated cells. We have also obtained a surprise result, that when encapsulated the $MAPbI_3$ cells are surprisingly stable under elevated temperature and sun light when aged at mpp. Despite being slightly lower efficiency, the device design that we will focus on for the minimodule development is the co-evaporated FA/Cs based cells, reported in Fig 6.

The relationship between accelerated (indoor) stressing and real-life outdoor operation is at this stage unclear due to a very slow outdoor degradation of the solar cells, by the end of the project we hope that a clearer correlation will emerge.

Funded by the European Union

This project has received funding from the European Union's Horizon Europe research and innovation programme under grant agreement No. 101082176