DELIVERABLE REPORT

VALHALLA

Realise a semi-transparent perovskite PV cell with >10% efficiency and >50% ATV

Deliverable D2.3
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PREPARED BY
UVEG
COORDINATED BY
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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.

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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
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CSEM CENTRE SUISSE D'ELECTRONIQUE ET DE MICROTECHNIQUE SA - RECHERCHE ET DEVELOPPEMENT	CSEM	CH

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Table of Content

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Abbreviations and acronyms list

Abbreviation	Meaning	Abbreviation	Meaning
ST-PSC	Semitransparent perovskite solar cell	AVT	Average visible transmittance
		LUE	Light-utilization efficiency
PCE	Power conversion efficiency	PR-he	Photopic response spectrum of the human eye
TCO	Transparent conductive oxides		
ITO	Indium tin oxide	C_{60}	Fullerene
CRI	Color rendering index	PLD	Pulsed laser deposition
TaTm	N,N,N″,N″-Tetra([1,1′-biphenyl]-4-yl)[1,1′:4′,1′‴terphenyl]-4,4‴-diamine	ALD	Atomic layer deposition
		J_{SC}	Short circuit current density
		V_{OC}	Open circuit voltage
HTL	Hole transport layer	FF	Fill factor
ETL	Electron transport layer	EQE	External quantum efficiency
%T	Transmittance	%R	Reflectance

1. Executive Summary

1.1 Description of the deliverable content and purpose

The purpose of this deliverable is to describe the efforts in VALHALLA to achieve technological proof-of-concept of a semi-transparent perovskite solar cells with PCE > 10 % and AVT > 50 % achievable within the project's vacuum-processed device framework, supporting applications such as building-integrated photovoltaics (e.g. power-generating windows).

To meet these targets, the work built upon an ultra-thin absorber layer (<100 nm) of a fully vacuum-deposited low-bandgap $FAMAPbI_3$ perovskite (≈1.55 eV) developed in task 1.1 and a pulsed-laser-deposited ITO layer directly deposited onto the solar cell stack. The challenge was to identify a device architecture that simultaneously delivers high visible transparency while still having a good power conversion efficiency.

Both the perovskite and TCO films were designed and engineered to provide high transparency while maintaining efficient charge generation. The optimisation process focused on fine-tuning absorber thickness, controlling perovskite film morphology, and tailoring the optical and electrical properties of the TCOs to minimise reflectance losses and improve the lLight-utilization efficiency (LUE).

Another key achievement in this deliverable was the realization of a novel approach to neutral colour semitransparency by optimising the TCO rear electrode based on transfer matrix method-based absorptance and 1-R simulations. Specifically, for this approach, in line with simulation results, we diminished the perovskite layer even further and employed thicker ITO without antireflective coatings, resulting in a relatively low variation in the device transmittance in the PR-he spectrum, hence a higher CRI value. Therefore, this strategy successfully leveraged TCO engineering to deliver semi-transparent perovskite solar cells with neutral aesthetics.

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2 Results

2.1 Transfer matrix-based optical simulations for improved LUE.

The perovskite solar cells that we investigate in this study are fabricated with an all vapor-deposited device stack of: LiF/glass/ITO/p-doped TaTm/TaTm/FAMAPbI_3/C_{60}/ALD SnO2/ITO/ALD Al_2O_3/LiF, as seen in Fig. 1a, where TaTm is the HTL and C_{60} is the ETL, Al_2O_3 is an encapsulation layer, and LiF is used as the antireflective coating. We used thermal co-evaporation to deposit the ultra-thin perovskite layer, as this method provides precise thickness control and uniformly pinhole-free films with flat, smooth surfaces (RMS < 10 nm) that enable the use of very thin charge-extraction layers, which suits our goal of reducing parasitic absorption and thereby enhancing both PCE and AVT.

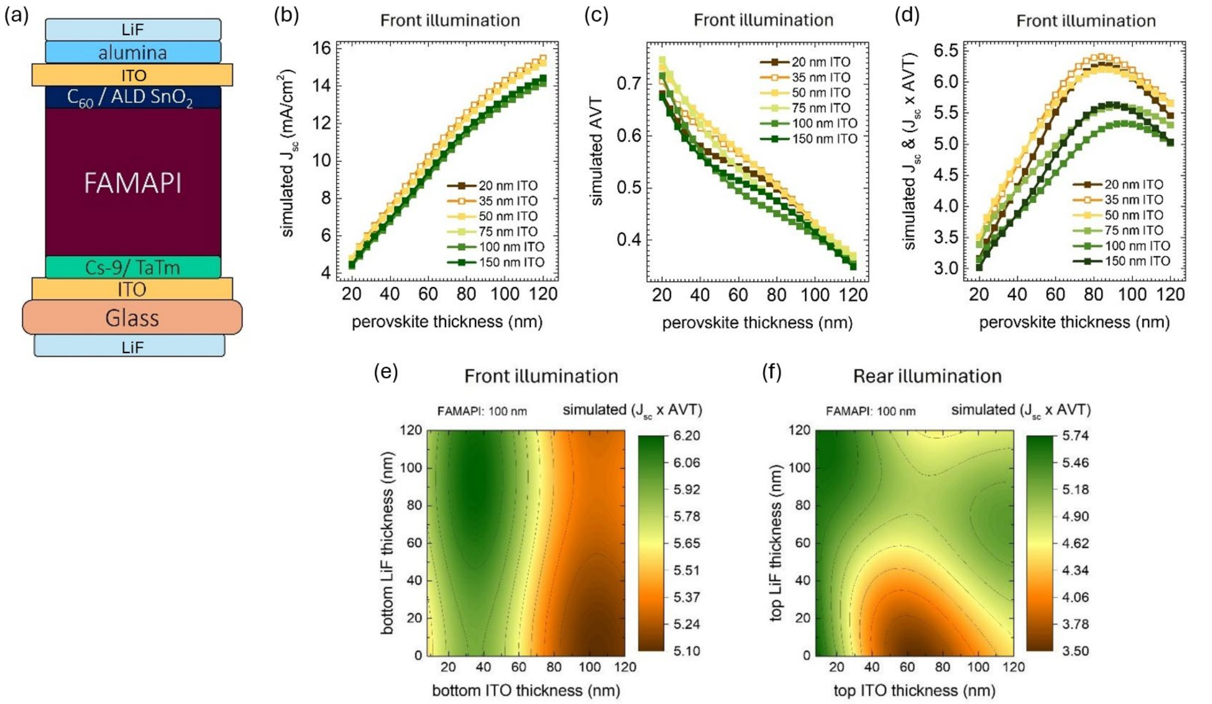

Figure 1: (a) ST-PSC device stack used in the optical simulations for the realization of high LUE values. The illumination of ST-PSC from front (glass side) and rear (thin film side) electrodes correspond to front and rear illumination conditions, respectively. (b-d) Simulated Jsc, AVT and Jsc × AVT values of the ST-PSC under front illumination condition for different front-ITO and FAMAPI layer thicknesses. (e-f) simulated Jsc x AVT values of the ST-PSC having ~100 nm FAMAPI layer under front and rear illumination conditions.

The perovskite layer, along with the TCO and LiF antireflection layers of both the front and rear electrodes, was subjected to transfer-matrix optical simulations in order to find the optimal thicknesses for maximum LUE (defined as the product of PCE x AVT).

To optimize the thickness of the front ITO layer (glass side), we simulated the J_{SC} and AVT values of the ST-PSC stack, for different front-ITO thicknesses over a range of FAMAPI layer thicknesses.
Higher simulated J_{SC} values were observed in the case of ST-PSCs having sub-100 nm front ITO electrodes, as shown in Figure 1b. The simulated AVT values of the ST-PSCs comprising 35 nm and 50 nm thick front ITO electrodes were found to be higher than those of the rest of the ST-PSCs for a large range of FAMAPI layer thicknesses (Figure 1c). Also, the maximum in the Jsc x AVT curve occurs at a FAMAPI thickness range of 70-110 nm, as shown in Figure 1d. When using the 35 nm ITO film front electrode, the optimum thickness of the front LiF antireflection layer was determined as 100 nm as

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shown in Figure 1e. As for rear illumination (thin films side), the optimum thicknesses of the rear ITO and LiF antireflection layers were determined to be 20 nm and 80 nm, respectively for the range of perovskite thickness of 70-110 nm, as shown in Figure 1f.

2.2 Evaporated ST-PSC with improved LUE.

We than proceeded to prepare the perovskite solar cells with the optimized thicknesses identified by transfer matrix-based optical simulations, choosing those that led to the higher theoretical LUE, as shown in Figure 2a. The thin front and rear ITO electrodes were reinforced with evaporated silver gridlines outside the active area to reduce series resistance.

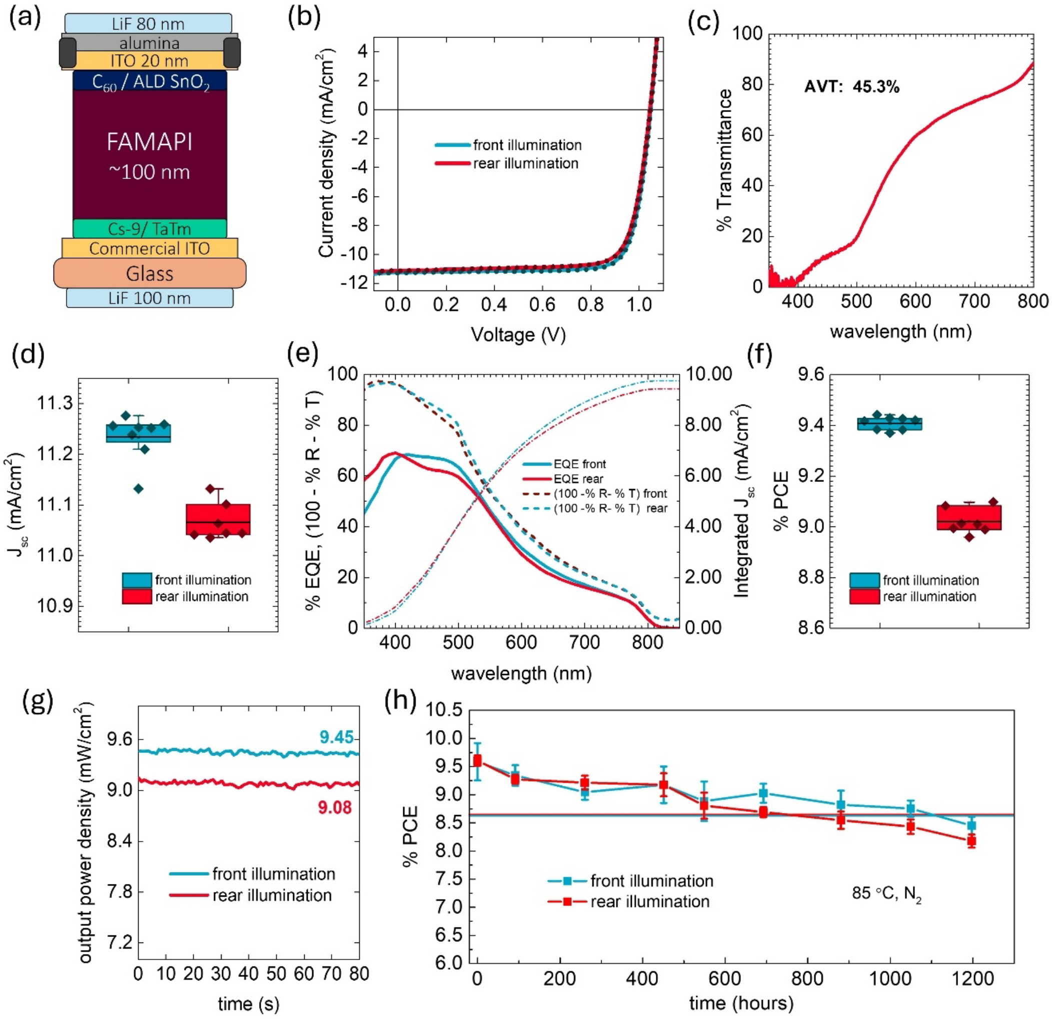

***Figure 2:** (a) Optimized device stack for an improved LUE. (b) champion J-V curves of the fabricated ST-PSCs from front (glass side) and rear (thin film side) illumination. (c) Transmittance spectrum of the ST-PSC along with the corresponding AVT value. (d) Statistics of J_{SC} values from front and rear illumination. (e) EQE spectra and 100-%R-%T spectra (left axis) and integrated J_{SC} values (right axis), (f) statistics of PCE values, and (g) steady state output power-density curves of the ST-PSC from front and rear illumination. (h) Evolution of PCE of ST-PSCs under both front and rear illumination conditions when thermally stressed at 85 °C on a hot plate in N_2 environment. The solid flat lines represent the respective 90% of the initial PCE values.*

Figure 2b and 2c show the J-V curves of the champion ST-PSC from front and rear illumination, and the device transmittance spectrum, respectively. The device exhibited an AVT of 45.3%. Owing to its optically optimal front and rear electrodes, the ST-PSC exhibited high J_{SC} values under both front and rear illumination (Figure 2d), leading to a high bifaciality factor of ~0.98. Comparison of the EQE and '100-%R-%T' spectra revealed that the difference in the light absorption under front and rear illumination in the wavelength range of 350-550 nm is due to parasitic absorption losses (Figure 2e). Moreover, the EQE spectra revealed that the marginally higher integrated J_{SC} from front illumination originated from the corresponding slightly higher EQE in the wavelength range of 600-800 nm (Figure 2e). The device exhibited high FF and V_{OC} values from both front and rear illumination, which indicates the desired presence of high shunt resistance in the device even though the perovskite is only 100 nm thick. Both the higher J_{SC} and slightly higher FF values in front illumination resulted in a slightly higher PCE than rear illumination (Figure 2f). The champion device exhibited PCE values of 9.4% from front illumination, with stable output power generation (Figure 2g). Importantly, the device demonstrated high thermal stability when stressed at 85 ^{o}C in N_2 environment, retaining more than 90% of their initial efficiencies for more than 1000 and 700 hours under front and rear illumination conditions, respectively (Figure 2h).
The champion device exhibited a LUE of 4.28. To our knowledge, this represents the highest reported LUE for low-bandgap ST-PSCs.

2.3 Transfer matrix-based optical simulations for improved CRI

The CRI of an ST-PSC is directly proportional to the 'flatness' of its transmittance across the PR-he spectrum; that is, lower spectral variations in transmittance correspond to higher CRI values. To increase CRI of our ST-PSCs, we modulated the device reflectance through adjustments in perovskite and rear-ITO thicknesses. Namely, increasing the rear ITO thickness from ~20 nm to ~120 nm and removing the rear LiF and alumina layers from the device stack flattened the device transmittance within a portion of the perovskite's main absorption spectrum, as evident from the wider spacing between successive contour lines in Figure 3b as compared to 3a. For sufficiently thin perovskite layers (<~70 nm), the flattened transmittance overlaps more effectively with the PR-he spectrum, enabling higher CRI values (>~80) (Figure 1c). However, this also diminishes the device AVT and the product of Jsc × AVT (Figure 1c).

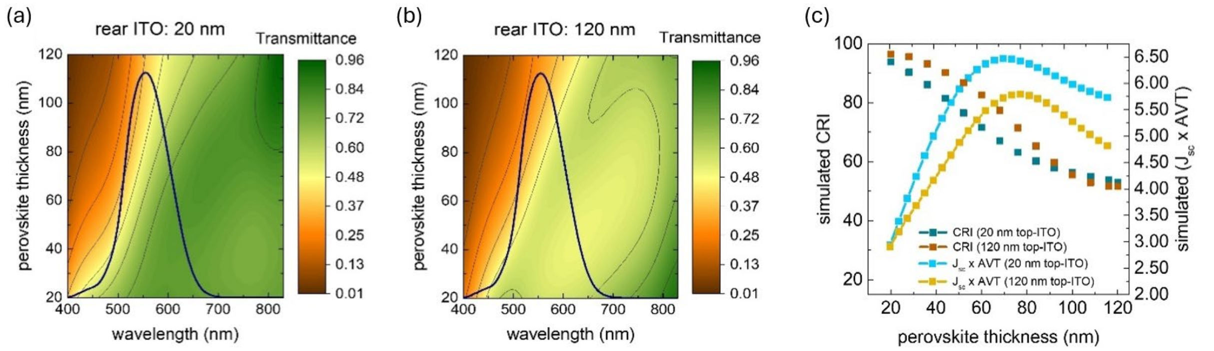

Figure 3: (a-b) Comparison of the spectral variation in the magnitude of transmittance of the two ST-PSCs having thin 20 nm and thick 120 nm rear-ITO electrodes within the PR-he spectrum (dark blue solid line) for a range of FAMAPI layer thicknesses. (c) Comparison of the evolution of simulated CRI and simulated (Jsc × AVT) in the ST-PSCs having 20 and 120 nm rear-ITO electrodes for a range of FAMAPI layer thicknesses.

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2.4 Evaporated ST-PSC with improved CRI.

At UVEG, perovskite solar cells were fabricated having the optimized thicknesses identified in section 2.3, choosing those that led to the higher theoretical CRI. Consistent with the optical simulations, the ST-PSC exhibited a flattened transmittance across the PR-he spectrum (Figure 4a), yielding a high CRI of 82.4 under white-light illumination (CCT: ~5513 K) while maintaining a substantial AVT of 48.5%. This is the highest CRI reported for a low-bandgap ST-PSC at comparable AVT values and outperforms several higher-bandgap perovskite ST-PSCs with even higher AVT. These results confirmed our strategy of optimizing the perovskite and rear TCO thicknesses to attain color neutrality in ST-PSCs that absorb over the entire visible spectrum. The high-CRI ST-PSC reached a PCE of 7.0% (Figure 4b), which translated to a LUE of 3.38. A non-pixelated sample including all device layers was fabricated and the digital photograph in Figure 4c showed the hue of transmitted light in front of a white paper. The object colors viewed directly and through the ST-PSC were compared in Figure 4f to demonstrate the color rendering capability of the ST-PSC.

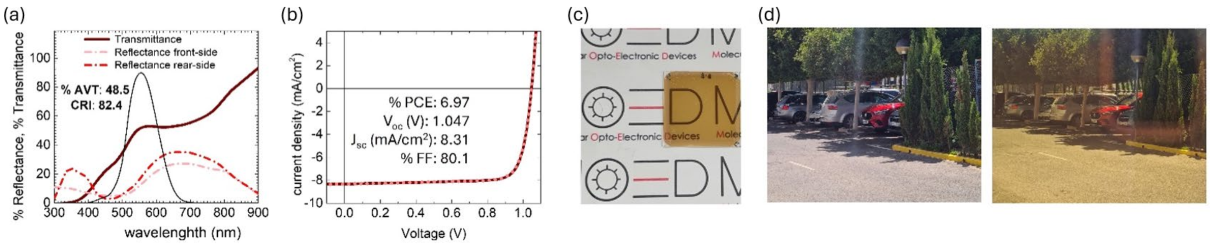

Figure 3: (a) Transmittance and reflectance spectra (from front and rear sides) of the high CRI ST-PSC along with the obtained % AVT and CRI values. (b) champion J–V curve along with the corresponding PCE parameters of the high CRI ST-PSCs. (c) A digital photograph of the high CRI ST-PSC sample on a white paper. (d) Digital photographs of a background when viewed directly and through the high CRI ST-PSC sample.

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.

3 Conclusions

This deliverable shows that semi-transparent perovskite solar cells employing fully vacuum-deposited low-bandgap $FAMAPbI_3$ can achieve both high efficiency and excellent optical performance. Two main optimization strategies were explored: tuning the perovskite and front/rear ITO layer thicknesses to maximize LUE, and adjusting the rear-ITO thickness to flatten the transmittance for improved CRI. The champion LUE reached 4.28 and the highest CRI for comparable AVT values was 82.4.

The champion devices reached PCE values of 9.4% and AVT up to 45.3 %, these results are very close to the targeted values of PCE > 10% and AVT > 50%. Moreover, reducing the perovskite thickness and optizing the TCO's we increased the CRI to above 82 and improved the AVT to 48.5% while still having a PCE of 7 %. And in spite of the very thin perovskite layers the device stability was still very good.
These results demonstrate that precise control over ultra-thin perovskite layers (<100 nm) and TCOs is essential to balance AVT, neutral color, and photovoltaic performance. Devices showed high bifaciality, stable PCE under front and rear illumination, and excellent thermal stability.

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