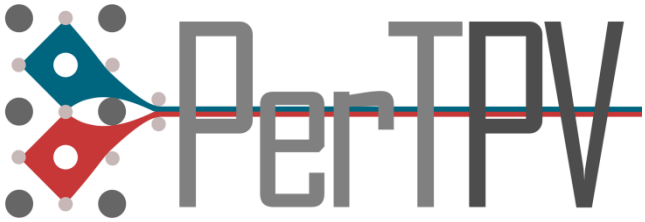

PerTPV – Perovskite thin film photovoltaics

Grant agreement 763977

Deliverable 3.7

1cm^2 Efficient Tandem Solar Cell

WP3

Lead beneficiary: Oxford PV
Authors: CSEM, OXPV, UVEG, UOXF
Delivery date: 31 August 2021
Confidentiality level: PUBLIC

The PerTPV project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No 763977.

Revision History

Author Name, Partner short name	Description	Date
Matt Klug, OXPV	Draft deliverable	30/08/2021
Henk Bolink, UVEG	Revision 1	03/09/2021
Matt Klug, OXPV	Revision 2	29/09/2021
Henry Snaith, UOXF	Final version	07/10/2021

Contents

1. Objective

The objective of D3.7 is to demonstrate an efficient tandem solar cell having a power conversion efficiency above 30% with an open-circuit voltage of 2.25 V, fill factor of 83%, and a short-circuit current density of 16.3 mA/cm^2 or greater. To work towards this goal, the consortium set up a framework that used parallel efforts to assess different processes and variations of the device stack and architecture and the perovskite composition.

2. Introduction

The overarching goal of PerTPV is to deliver high performing all-perovskite multi-junctions and build-up routes that are compatible with future commercialisation. To work towards this goal, tandem device development proceeded along multiple avenues of design in order to maximise output. This includes four terminal (4T) and two terminal

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(2T) tandem architectures, superstrate and substrate device configurations, and perovskite processing using solution-based and vacuum-based deposition methods.

To avoid the known thermal instability of methylammonium cations, perovskite compositions were selected that use formamidinium and caesium ions exclusively at the A-site whenever possible.

The first published embodiment of a perovskite-perovskite tandem[3] utilised a superstrate device architecture, where the wide bandgap perovskite was first deposited on a substrate coated with a transparent conductive oxide and then the narrow bandgap perovskite was deposited on top with a suitable recombination layer sandwiched between them. When in solar operation, the device is flipped upside-down and illuminated through the substrate glass so that sunlight first passes through the wide bandgap perovskite top-cell before any filtered low energy photons strike the narrow bandgap perovskite bottom-cell below. As the archetypal perovskite-perovskite 2T tandem build, this was the natural starting point for the PerTPV project, especially given the experience of many project partners in constructing single-junction devices, which in principle could be combined into a tandem device. We also explored 4T architectures, since these enabled separating out the processing requirements of the two junction. Finally, we developed a superstrate 2T architecture.

3. Results

3.1 2T Superstrate tandem architecture

We first started with a combination of wide band gap perovskite cells solution processed at CSEM, capped with narrow gap solution processed perovskite cells processed at UOXF. The result was operational tandems, but there existed significant challenge in retaining layer integrity during processing of the low gap cell. The resultant devices often contained “shunts”, as exemplified in Figure 1 below.

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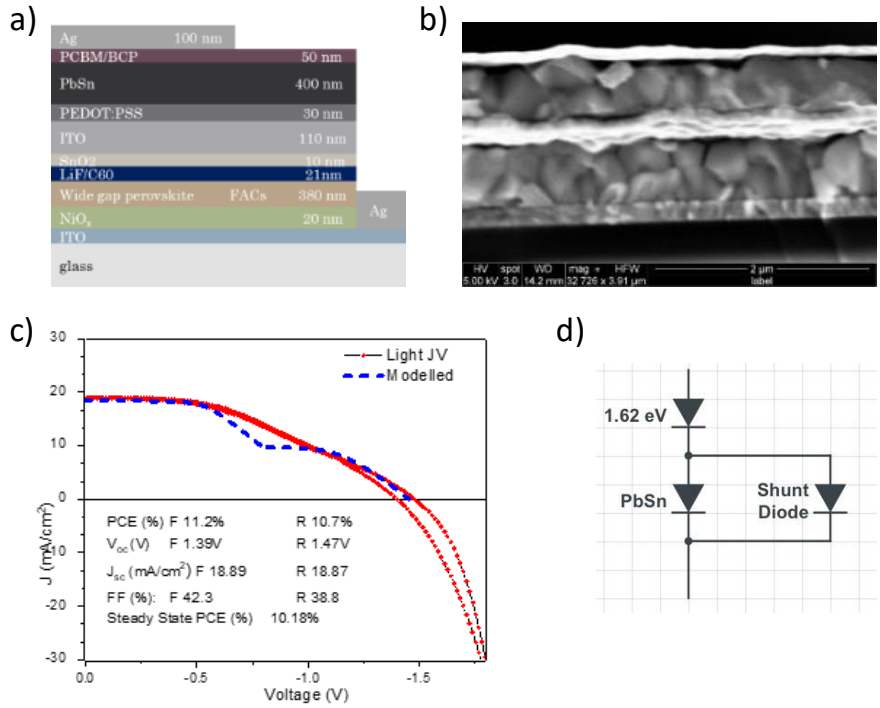
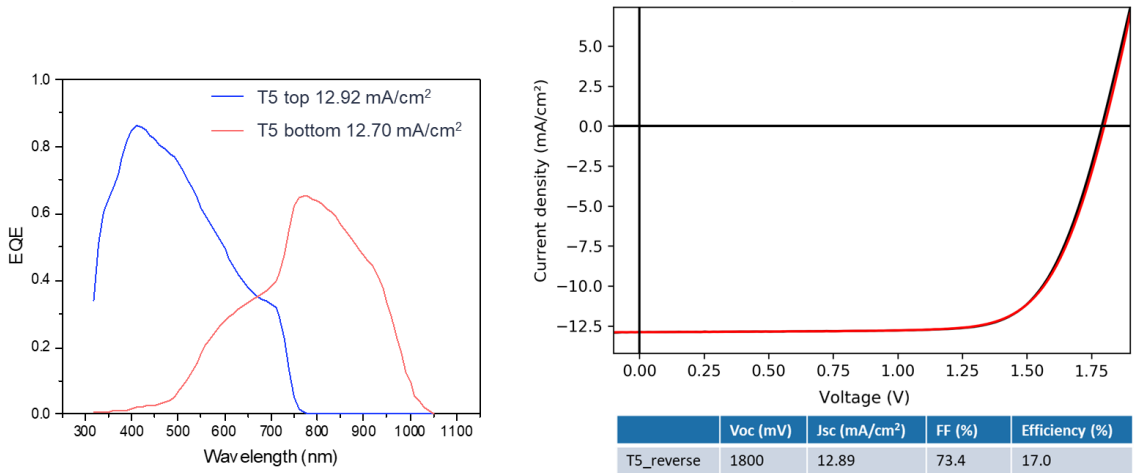


4.

5. **Figure 1**. (a) Superstrate-based 2T device stack. (b) Cross-sectional SEM image of a representative device. (c) As-measured and modelled current-voltage characteristic. The modelled JV was produced through circuit analysis, assuming a diode-like shunt characteristic and varying all diode parameters to obtain the best fit. (d) Circuit model used for analysis.

Further work was performed at CSEM optimising the wide band gap cell and developing the low band gap cell and interconnecting layers. This resulted in improved tandem cell operation and efficiency. A tandem cell EQE spectrum and JV curves are shown in Figure 2.



	Voc (mV)	Jsc (mA/cm²)	FF (%)	Efficiency (%)
T5_reverse	1800	12.89	73.4	17.0
T5_forward	1790	12.89	73.9	17.1

Figure 2. EQE and IV curve of the best superstrate 2T tandem cell.

3.2 4T Tandem architecture

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Single-junction devices of several perovskite compositions suitable for the top-cell with bandgaps of 1.55 eV, 1.62 eV, and 1.72 eV were fabricated by CSEM using a substrate design where the devices are illuminated through an ITO top electrode. Likewise, single-junction devices of 1.24 eV narrow bandgap perovskite were fabricated by UOXF using a superstrate design where the device is illuminated through the TCO/glass substrate. Four terminal (4T) tandems were then demonstrated by using a thin optical coupling layer of polydimethylsiloxane (PDMS) to connect the glass substrates together, such that simulated sunlight would pass through the semi-transparent wide bandgap cell before continuing through to the narrow bandgap perovskite cell underneath (see 3).

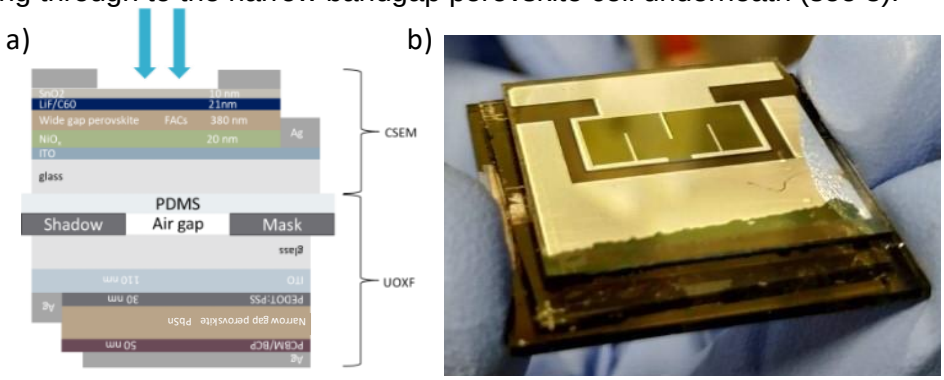

Figure 3. (a) Device stack for a 4T perovskite-perovskite tandem. (b) Photograph of a fabricated 4T tandem complete with PDMS interlayer.

Using this method, the most efficient 4T tandem device achieved 17.5% with the corresponding current-voltage (IV) characteristics and performance metrics reported in Figure .

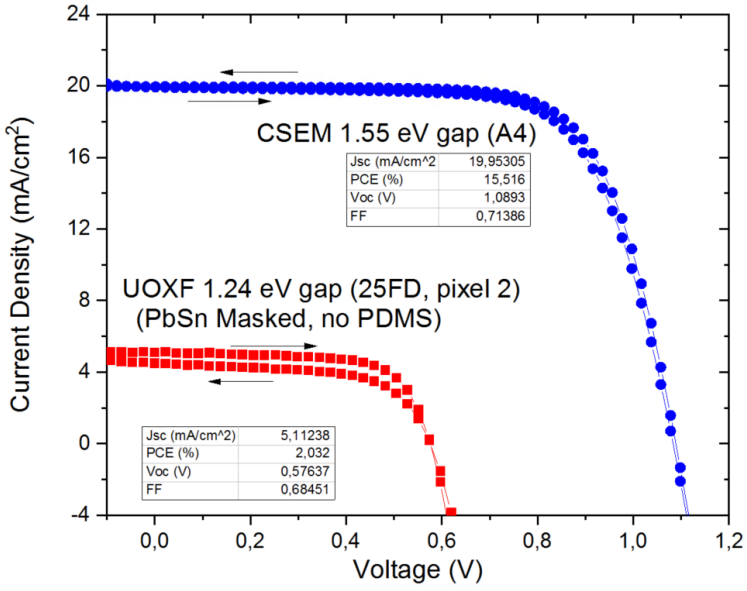

Figure 4. Current-voltage characteristics of a 4T tandem stack consisting of a 1.24 eV perovskite bottom cell masked with a 0.2 cm2 mask and a 1.55 eV top cell.

3.3 2T Substrate Tandem Architecture

We developed a range of different substrate device architectures where the low band gap PbSn perovskite was solution processed first on the substrate, followed by

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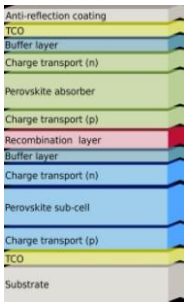
deposition of the charge recombination layers and wide band gap perovskite top cell via vapour phase methods. Two different device architectures are shown in Figure 5. Which shows the partners responsible for the deposition of the different layers in the device stack.



Layer	Nominal Stack	Where
Glass/ITO	10 Ω/□	UOXF
PEDOT:PSS	30 nm	UOXF
$FA_{0.83}Cs_{0.17}Pb_{0.5}Sn_{0.5}I_3$	400 nm (1.24 eV)	UOXF
i-C_{60}	20 nm	UVEG
C_{60}:phlm	25 nm	UVEG
TaTm:F6	25 nm	UVEG
i-TaTM	10 nm	UVEG
$FACsPb(IBr)_3$	550 nm (1.70 eV)	UVEG
i-C_{60}	10 nm	UVEG
C_{60}:phlm	30 nm	UVEG
Window Layers		OxPV
Testing		OxPV

UVEG Organic Recombination Layer

Layer	Nominal Stack	Where
Glass/ITO	10 Ω/□	UOXF
PEDOT:PSS	30 nm	UOXF
$FA_{0.83}Cs_{0.17}Pb_{0.5}Sn_{0.5}I_3$	400 nm (1.24 eV)	UOXF
n-contact		OxPV
Buffer		OxPV
ITO		OxPV
TaTm:F6	25 nm	UVEG
i-TaTM	10 nm	UVEG
$FACsPb(IBr)_3$	550 nm (1.70 eV)	UVEG
i-C_{60}	10 nm	UVEG
C_{60}:phlm	30 nm	UVEG
Window Layers		OxPV
Testing		OxPV

OxPV Inorganic Recombination Layer

Figure 5. Two substrate-based tandem device architectures using either an organic or inorganic recombination junction. Notably, the light illumination direction is from the bottom, in the two specified device stacks shown.

In the first instance, we worked with ITO coated glass as the substrates. However, these devices were semi-transparent, and some loss in current density occurred for the low band gap cell. We therefore proceeded to develop a substrate device architecture with a metal back electrode. We show an example of such a device in Figure 6.

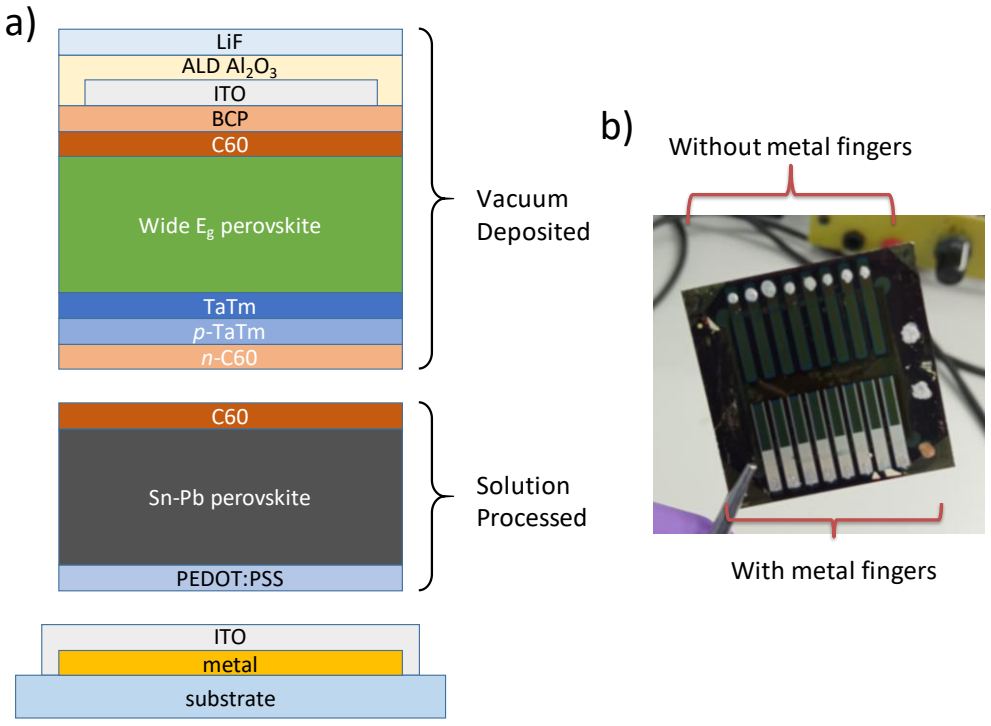


Figure 6. (a) Device layout for a top transparent, bottom metal, 2T perovskite/perovskite tandem solar cell. (b) Photograph of a finished device where 8 pixels have a simple ITO electrode, while the other 8 incorporated lateral metal fingers to help current collection from the cell. The substrate size is 3x3 cm^2, each pixel has an area of 0.0825 cm^2 and is measured with a shadow mask with an aperture area of 0.05 cm^2.

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This metal backed substrate cell structure resulted in working tandem cells. However, we struggled to obtain current densities in excess of 12 mAcm^{-2}. The main problem was the relatively poor performance we achieved with the low band gap Pb:Sn perovskites, as illustrated in the external quantum efficiency (EQE) spectrum shown in Figure 7.

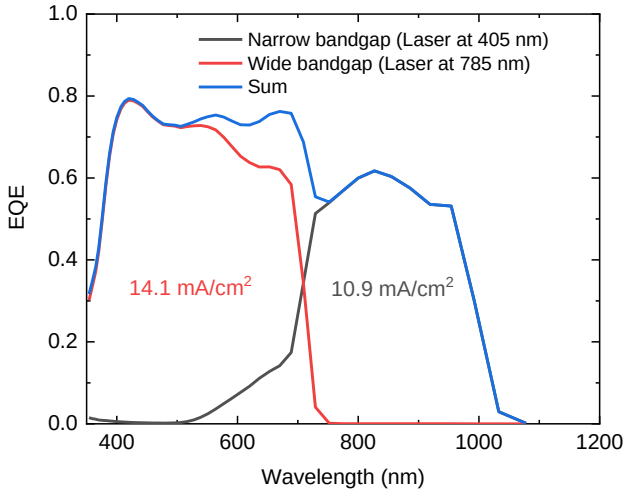

Figure 7. EQE spectra of a small area 2T perovskite-perovskite tandem solar cell. The front (wide bandgap) and rear (narrow bandgap) sub-cells show integrated J_{sc} of 14.1 and 10.9 mA/cm^2.

4. Conclusion and Outlook

The highest efficiencies that were achieved during the project are reported in **Error! Reference source not found.** and fall below the target of 30% and the record efficiencies values reported by the greater community: all-perovskite 4-terminal (4T) of 25% on 0.059 cm^2 and 2-terminals (2T) tandems of 24.8% on 0.049 cm^2 (22% on 1.05 cm^2).[1,2]

Table 1. Champion efficiencies for all-perovskite tandem solar cells produced in PerTPV for this deliverable

Type / Configuration	Size	Power Conversion Efficiency
4-terminal	1 cm^2	17.5%
2-terminal / Superstrate	1 cm^2	17.1%
2-terminal / Substrate	1 cm^2	10.8%
2-terminal / Substrate	0.05 cm^2	14%

In every stage of development, the performance of both 4T and 2T all-perovskite tandems was limited by the quality of the low bandgap PbSn-based perovskites required for the bottom-cell. Whilst UOXF was able to achieve up to 18% power conversion efficiencies at the very beginning of the project, reproducibility became increasingly challenging with time even when the same materials and equipment are used at the same site. Over the course of this project, every research group that developed low bandgap perovskite devices encountered the same issue – regardless of whether the devices were processed via the standard solution methods or with the vacuum deposition routes at UVEG. Despite these challenges, there were many successful outcomes that will be valuable for future all-perovskite tandem development including:

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- Successful development of high-performing, fully vacuum deposited, MA-free $FA_{1-y}Cs_yPb(I_{1-x}Br_x)_3$, wide bandgap perovskite absorber materials with 1.75, 1.77, and 1.81 eV all capable of efficiencies above 14%.
- Successful demonstration of vacuum deposited $FAPb_{0.5}Sn_{0.5}I_3$ low bandgap perovskite with 1.25 eV reaching 14% power conversion efficiency.
- Development of both organic and inorganic recombination junctions, which yielded similar performance in 2T devices.
- The design and validation of a rear-reflector metallic electrode that is inexpensive and compatible with high-throughput manufacturing.
- Successful demonstration of device integration in 4T tandems with a 17.5% power conversion efficiency.
- Successful demonstration of device integration in superstrate-based 2T tandems with open-circuit voltages up to 1.80V in $1cm^2$ devices and 17.1% power conversion efficiency with solution-processed perovskite absorbers at both sub-cells.
- Successful demonstration of device integration in substrate-based 2T tandems with open-circuit voltages up to 1.85V in $1cm^2$ devices, which is very close to the sum of the voltages achieved in single-junctions of each sub-cell (~0.7V for bottom-cell, and ~1.2V for top-cell)

It is expected that once the bottleneck that limits the reproducibility of narrow bandgap PbSn devices is overcome, the device architectures and fabrication routes developed in this project work package can be immediately applied to achieve significant gains in all-perovskite tandem photovoltaic performance.

5. References

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3. Eperon, G. E. *et al.* Perovskite-perovskite tandem photovoltaics with optimized band gaps. *Science* **354**, 861–865 (2016).

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