



VIPERLAB

FULLY CONNECTED VIRTUAL AND PHYSICAL
PEROVSKITE PHOTOVOLTAICS LAB

**D 8.9 The use of
industrially acceptable
solvent systems to obtain
efficient perovskite PV devices**

**DELIVERABLE
REPORT**



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D8.9 THE USE OF INDUSTRIALLY ACCEPTABLE SOLVENT SYSTEMS TO OBTAIN EFFICIENT PEROVSKITE PV DEVICES

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DISCLAIMER

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EXECUTIVE SUMMARY

The VIPERLAB initiative represents a pivotal infrastructure project designed to establish a comprehensive European framework for advancing perovskite photovoltaic (PV) technology. With contributions from 13 partners across Europe, VIPERLAB is committed to facilitating access to both physical and virtual infrastructures, fostering collaboration among researchers and industry stakeholders globally.

Recognizing perovskite PV as the key emerging technology capable of driving EU-based PV production, VIPERLAB is focused on achieving the lowest costs and carbon footprint for future market penetration. The report will explore the various green solvent process routes currently being developed at partners institutions and evaluated within the context of perovskite PV device fabrication. By examining these routes, the report aims to identify the most promising methods for industrialization, considering both environmental impact and scalability. The insights gained from this analysis will contribute to the broader objective of creating a sustainable and competitive European perovskite PV industry, poised to lead the global market with innovative, eco-friendly technologies.

1. INTRODUCTION

The rapid development of perovskite photovoltaic (PV) technology has positioned it as a leading candidate for next-generation solar energy solutions. With its remarkable power conversion efficiencies and potential for low-cost production, perovskite PV has captured significant attention within both the academic and industrial sectors. However, the widespread adoption and commercialization of this technology face critical challenges, particularly concerning the environmental and health impacts associated with its manufacturing processes.

One of the most pressing concerns is the reliance on traditional solvents such as dimethylformamide (DMF), N,N-dimethylacetamide (DMAC) and solvent mixtures derived thereof, in the fabrication of perovskite solar cells. These solvents, while effective in dissolving perovskite precursors and facilitating the formation of high-quality films, are known for their toxicity, and environmental hazards. Because of the risks associated with the use of these solvents, they are currently on the candidate list of Substances of Very High Concern (SVHC).¹ The use of such hazardous substances not only poses risks to human health and safety but also contradicts the broader goals of sustainability and environmental stewardship that underpin the future of the renewable energy sector.

In response to these challenges, the development and implementation of green solvents in perovskite PV fabrication have emerged as critical areas of research. Green solvents, defined by their low toxicity, biodegradability, and derivation from renewable resources, offer a more sustainable alternative to conventional solvents. Their adoption is essential not only for minimizing the environmental footprint of perovskite PV production but also for aligning



with global regulatory frameworks and sustainability initiatives, such as the European Green Deal and the United Nations' Sustainable Development Goals (SDGs).

A detailed literature review of green solvents in the perovskite PV space was conducted in Deliverable 8.2. Therefore, this deliverable will focus on methods developed by VIPERLAB partners and compare these approaches with traditional 'toxic' alternatives. The analysis aims to provide insights into the effectiveness, scalability, and industrial viability of green solvent-based processes, contributing to the broader objective of establishing a sustainable and competitive European perovskite PV industry.

2. RESULTS AND DISCUSSION

Within the consortium, two primary approaches have been pursued to address the challenge of reducing the use of toxic solvents in perovskite PV fabrication. The first approach involves the development of a hybrid coating method (referred to as the "Two step method"), where perovskite deposition is split into two steps. In the first step, the inorganic material is thermally evaporated, followed by the solution deposition of organic compounds, typically using alcohols like ethanol or isopropyl alcohol, to form the perovskite film. The second approach, referred to as the "Single step method", deposits the both the inorganic and organic precursors in a single, wet deposition step.

The following sections will provide a detailed comparison of the green solvent methods developed by each consortium partner against conventional toxic methods. This comparison will highlight the advantages and limitations of each approach, offering insights into the most promising techniques for achieving sustainable perovskite PV manufacturing.

2.1. Swansea University (Single step method)

ACN is generally considered a safer alternative to DMF for PSC fabrication due to its significantly lower toxicity profile and more favourable workplace exposure limits (WEL).^{2,3} In industrial settings, VOCs like DMF and ACN are regulated with specific WELs to protect workers; DMF has a WEL of 5 parts per million (ppm), highlighting its high toxicity and strict handling requirements due to risks like reproductive toxicity, liver damage, and respiratory issues. In contrast, ACN has a WEL of 40 ppm, reflecting its comparatively lower toxicity and allowing for more flexibility in ventilation and handling in manufacturing environments. Additionally, ACN's low viscosity and high volatility facilitate rapid drying and efficient crystallization of the perovskite film at room temperature, reducing the need for high-temperature processing making it compatible for fabrication on plastic substrates. Although ACN is not entirely risk-free due to its flammability and mild toxicity, its use minimizes health and safety concerns compared to DMF making it a more viable choice for large-scale PSC production with fewer stringent safety protocols.

SU has developed a method that substitutes ACN for the conventional DMF-DMSO approach. The following sections will compare the performance and characterization data



of these two solvent systems, demonstrating ACN's potential as a more viable choice for large-scale PSC production with reduced health and safety concerns.

2.1.1 Experiment Details

To compare the two solvent system, NIP architecture with layers Glass/ITO/SnO₂/MAPbI₃/Spiro-OMeTAD/Au. The fabrication of PSCs was conducted through a sequential deposition of layers in a controlled cleanroom environment. Glass substrates coated with indium tin oxide (ITO) were prepared by an extensive cleaning process, including a 2% Helmanex wash, sequential ultrasonic baths in deionized water and isopropanol for 10 minutes each, and a final 10-minute plasma treatment, all completed in a Class 6 cleanroom with visual inspection to confirm cleanliness. Following this, a of tin(IV) oxide (4.2% wt SnO₂) was applied via spin coating at 4000 rpm for 30 seconds using a colloidal dispersion of tin oxide diluted in water. The coated substrate was then annealed at 150°C for 5 minutes. The perovskite layer, consisting of methylammonium lead iodide (MAPI), was spin-coated with a solution of 576 mg PbI₂, 199 mg MAI, 0.8 ml DMF, and 0.2 ml DMSO, stirred at 60°C and filtered with a 0.2 μm PTFE filter for DMF-DMSO solvent system and 230 mg PbI₂, 79 mg MAI was mixed into Methylamine (ca. 9% in ACN) for ACN based formulation. For DMF-DMSO devices during spin coating (4000 rpm for 30 seconds), ethyl acetate was dripped onto the substrate after 15 seconds to enhance crystallization, followed by a 10-minute annealing at 110°C. While for ACN formulation no antisolvent was needed and films were spun at 2000RPM for 60 seconds followed by drying at 110C for 10mins. For the p-type layer, a Spiro-OMeTAD solution (90 mg in chlorobenzene with additives TBP, LiTFSI and FK209) was prepared, stirred at 60°C, filtered, and spin-coated at 4000 rpm for 30 seconds. Finally, a gold electrode was deposited via thermal evaporation to a thickness of 100 nm, completing the PSC assembly.

2.1.2 Results

The JV data in Figure 1a compares the performance of the best (hero) devices fabricated using the DMF-DMSO and ACN-MA solvent systems. For the DMF-DMSO system, the hero device achieved a power conversion efficiency (PCE) of 20.75%, with a short-circuit current density (J_{sc}) of 22.15 mA/cm², an open-circuit voltage (V_{oc}) of 1.09 V, and a fill factor (FF) of 76.48%. In comparison, the best-performing device using the ACN-MA system showed a slightly higher PCE of 21.12%, a J_{sc} of 22.44 mA/cm², a V_{oc} of 1.07 V, and an improved FF of 80.33%. Boxplot analysis (Figure 1c) of the 16 devices in each solvent group shows nearly similar average performance metrics between the two systems, indicating that switching to the industry-friendly, low-toxicity ACN-MA solvent system does not lead to any significant performance drop.

Following the performance analysis, the SEM images and grain size distribution data as shown in Figure 2 provided additional insights into the morphological properties of the perovskite films formed with the DMF-DMSO and ACN-MA solvent systems. The SEM



images reveal that both solvent systems produce well-packed, uniformly distributed grains with a similar appearance in terms of density and compactness. The grain size distribution further supports this observation, with both systems showing a peak frequency in the 150-250 nm range, suggesting that most grains fall within a similar size range. While there are slight variations in grain distribution across different ranges, the overall similarity in average grain size indicates that both solvent systems produce films with comparable grain structures.

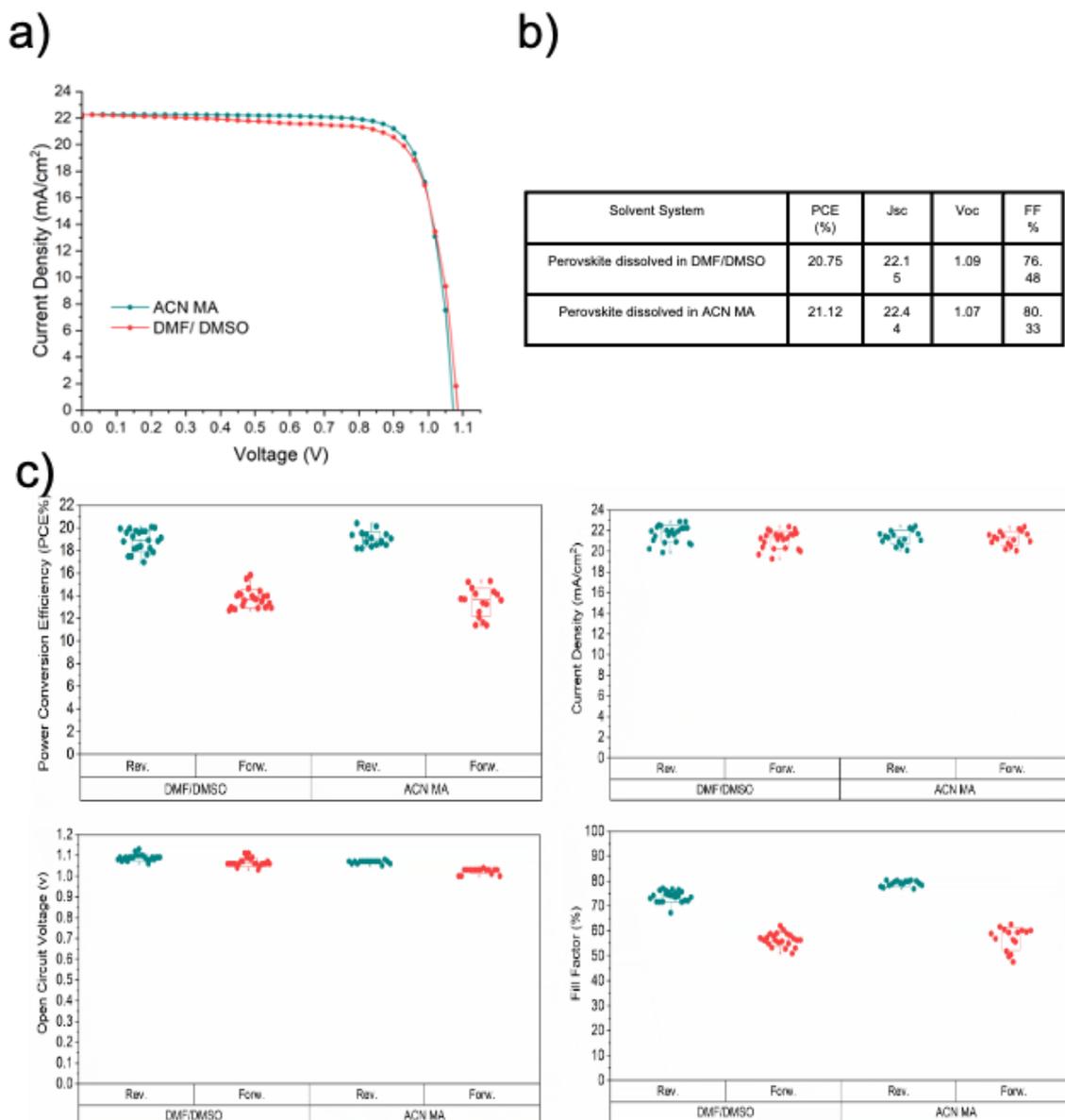


Figure 1. (a) J-V curves comparing the performance of PSCs fabricated with DMF-DMSO and ACN-MA solvent systems, (b) Table displaying the performance parameters for the best-performing devices from each solvent system, (c) Boxplots of PCE, Jsc, Voc, and FF for forward and reverse scans across 16 devices per solvent system.



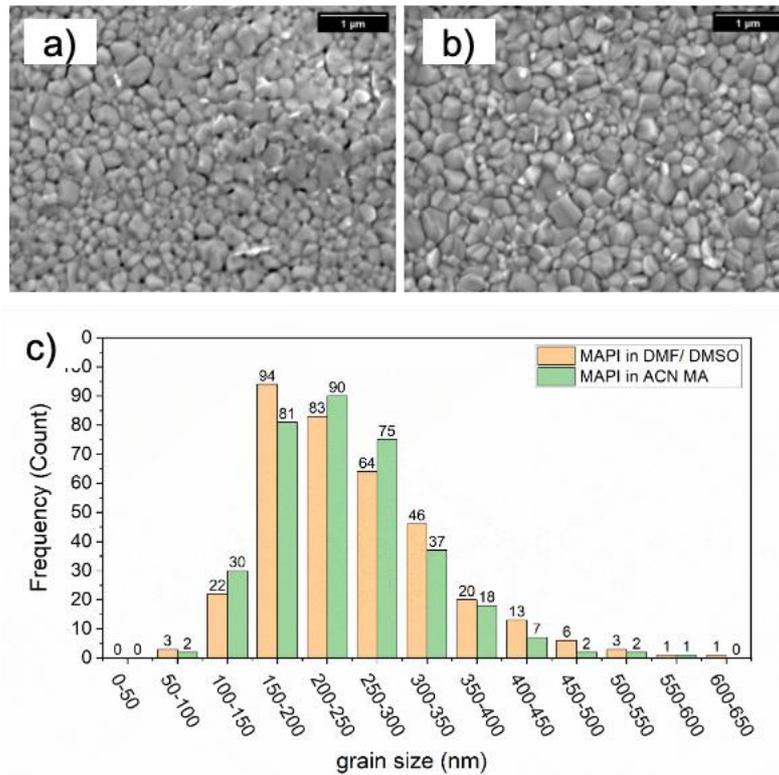


Figure 2. Surface morphology and grain size distribution of perovskite films fabricated with different solvent systems. (a) SEM image of perovskite film using DMF-DMSO solvent system. (b) SEM image of perovskite film using ACN-MA solvent system, illustrating similar grain morphology. (c) Grain size distribution histogram comparing DMF-DMSO (orange) and ACN-MA (green) systems.

Though the performance of PSCs fabricated with the two solvent systems was similar, ensuring the long-term stability of these films is equally critical to prevent compromising device durability. To evaluate stability, XRD spectra were recorded for both the DMF-DMSO and ACN-MA solvent systems on Day 1 and after one week (Day 7). The data is presented in Figure 3. The XRD data show that both solvent systems produced well-crystallized perovskite films, as indicated by sharp and distinct peaks on Day 1, confirming the formation of a high-quality perovskite structure. However, differences in stability are evident by Day 7. In the DMF-DMSO films, a PbI_2 peak appears, signalling the onset of film degradation. This presence of PbI_2 indicates partial decomposition of the perovskite layer, suggesting reduced structural stability over time. In contrast, the ACN-MA films do not exhibit this PbI_2 peak even after a week, indicating minimal degradation and a more stable perovskite structure.

These findings suggest that the ACN-MA solvent system provides enhanced stability, maintaining the integrity of the perovskite layer over time without signs of decomposition. This stability advantage, combined with comparable device performance, highlights ACN-MA as a promising alternative to DMF-DMSO, offering both reduced toxicity and improved film durability for scalable PSC applications.

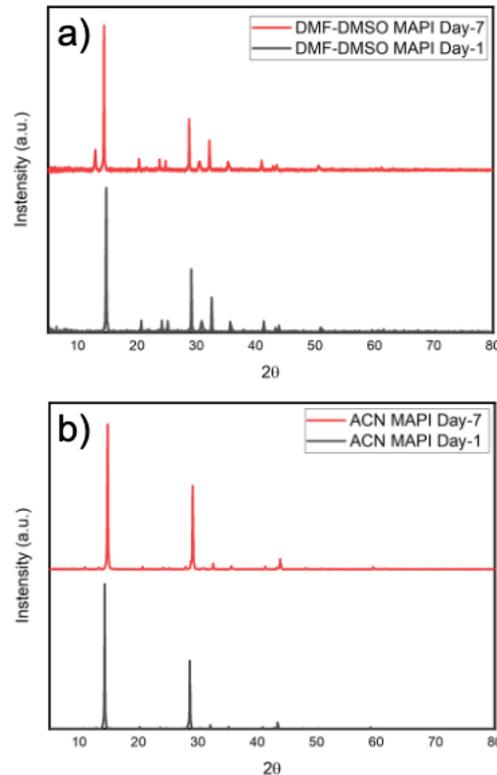


Figure 3. X-ray diffraction (XRD) patterns of perovskite films using different solvent systems over time. (a) XRD patterns of films fabricated with the DMF-DMSO solvent system on Day 1 (black) and Day 7 (red). (b) XRD patterns of films prepared with the ACN-MA solvent system on Day 1 (black) and Day 7 (red).

In summary, the ACN-MA solvent system emerges as a promising, safer alternative to DMF-DMSO for scalable perovskite solar cell production. Its compatibility with high device performance, coupled with improved structural stability, positions ACN-MA as a practical choice for industrial applications, addressing both efficiency and longevity requirements. This shift to a less toxic, industry-friendly solvent supports the advancement of PSC technology toward commercial viability without sacrificing quality or durability.

2.2. CSEM (Single and Two step method)

This section will compare two methods to deposit the perovskite absorber that are under development at the infrastructure shared between EPFL and CSEM.

The first method, denoted the 1-step spin-coating approach, involves spin-coating an ink featuring all the perovskite precursors, an anti-solvent spin-coating step, before then performing an annealing step to fully crystallize the perovskite film. This process is performed in gloveboxes using dimethylformamide, DMF, a toxic solvent with significant environmental and human health impact.¹ The manipulation of large volumes of DMF in an industrial environment will require strict safety measures, measures that are likely to become stricter in the future in Europe to protect the health of workers.⁴ For these reasons, DMF has



been estimated to increase manufacturing costs by >0.03 €/W in literature.⁵ Still, DMF-based processing currently led to best solar cell efficiencies in the laboratory, which explains its widespread use in the research community.

The second method, denoted the 2-step hybrid method, does not involve any DMF or other toxic solvent. The method combines the co-evaporation of the inorganic perovskite precursors in high vacuum conditions, before then spin-coating the organic precursors dissolved in a gentle alcohol (typically ethanol), before then annealing the layer stack to crystallize a perovskite thin film.

These two methods are used at EPFL and CSEM to produce perovskite single-junction and perovskite/Si tandem solar cells, as summarized in the next sections.

2.2.1 Experimental details

1-step spin-coating method

As detailed in Ref.⁶, triple-cation perovskite with a nominal composition of $\text{Cs}_{0.05}(\text{FA}_{0.90}\text{MA}_{0.10})_{0.95}\text{Pb}(\text{I}_{0.80}\text{Br}_{0.20})_3$ were prepared using the four stock solutions detailed below:

- (a) 1.55M FAI (Dyename, $>99.99\%$) and 1.7M PbI_2 (TCI, $>99.99\%$) in DMF:DMSO = 4:1,
- (b) 0.775M FABr (Dyename, $>99.99\%$) and 0.775M MABr (Dyename, $>99.99\%$) and 1.7M PbBr_2 (TCI, $>99.99\%$) in DMF:DMSO = 4:1
- (c) 1.5M CsI (Alfa Aesar, $>99.9\%$) in DMSO and
- (d) 2,3,4,5,6-pentafluoro-benzylphosphonic acid (pFBPA, Sigma Aldrich, 97%) in 28 mg/mL in DMF:DMSO = 4:1.

The solutions were mixed with a volume ratio of a:b:c:d = 80:20:5:X, with the last one varied to evaluate the impact of the pFBPA additive. Inside a N_2 glovebox, the perovskite precursor ink was dropped on the substrate prior to spinning and 400 mL of anisole antisolvent was dropped 20 s after the start of the spin coating at 3500 rpm (total spinning time of 35 s). The samples were then annealed on a hotplate in N_2 at 100 °C for 15 min.

2-step hybrid method

As explained in ref⁶, the first step of the 2-step hybrid method to deposit the perovskite absorber involved depositing 180 nm of PbI_2 ($>99.99\%$, beads from Alfa Aesar) and 18 nm CsBr ($>99.99\%$, Abcr) by co-evaporation in a Lesker mini-Spectros system (at 1 Å/s for PbI_2 and 0.1 Å/s for CsBr). This PbI_2/CsBr template layer was then coated by a mixture of FABr and FAI in various molar ratios in ethanol (0.46 M, precursors from Dyename) using spin coating. Similarly to the 1-step methods, various concentrations of pFBPA were added to the organohalide solution. Dynamic dispensing of 100 µl of the FABr:FAI solution was



performed at a spin-coating speed of 4000 rpm for 20-25 s in a N₂ glovebox. An annealing at 150 °C for 20 min in ambient air then crystallized the Cs_{0.18}FA_{0.82}Pb(I,Br)₃ perovskite film (composition estimated based on energy-dispersive X-ray spectroscopy data).

PSCs were processed in the p-i-n configuration, meaning with the hole transport layer deposited prior to the perovskite and then the electron transport layer. Starting with ITO coated glass, with both 1-step and 2-step perovskites, Me-4PACz was used as hole transport layer. To mitigate the hydrophobicity of the Me-4PACz layer and resulting challenges in coating the perovskite film using the 1-step method on this substrate layer, a partial layer of SiO_x nanoparticles was deposited to improve the wetting of the ink (only for 1-step-processed perovskite films). The perovskite film was then coated by a C₆₀ layer (NanoC, >99.95%) using thermal evaporation. A buffer layer made of SnO_x layer was then added by atomic layer deposition (ALD) at a temperature of 100 °C. Finally, Ag was deposited through a shadow mask by thermal evaporation to complete the cells.

2.2.2 Results

XRD patterns were acquired to evaluate the crystallographic properties of the 1-step and 2-step processed films (Figure 4). The XRD data highlights an improved conversion to a perovskite phase in presence of pFBPA: the fraction of crystalline PbI₂ precursor remaining in the film after annealing decreases with the addition of pFBPA.

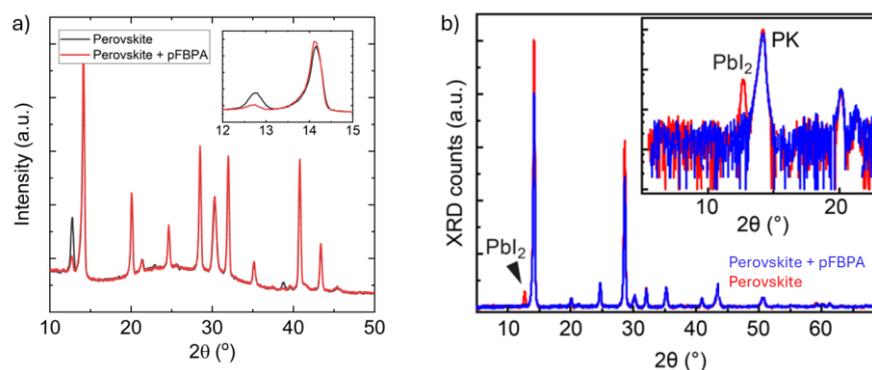


Figure 4: a) XRD pattern of 1-step processed perovskite film with and without pFBPA and b) XRD data of a 2-step hybrid perovskite film with and without pFBPA. Taken from Refs.^{6*}

SEM images of the films were also acquired to evaluate the microstructural properties of the films (Figure 5). A similar domain size is observed whether the perovskite film is processed via a 1-step spin-coating or a 2-step hybrid co-evaporation/spin-coating method. The white clusters in a) are PbI₂ domains, while the dark regions in b) are excess pFBPA molecules that segregate on the top perovskite surface after annealing. It is worth noting that the use of pFBPA is particularly important to improve the microstructural properties of 2-step hybrid films (the domain size is increased).*

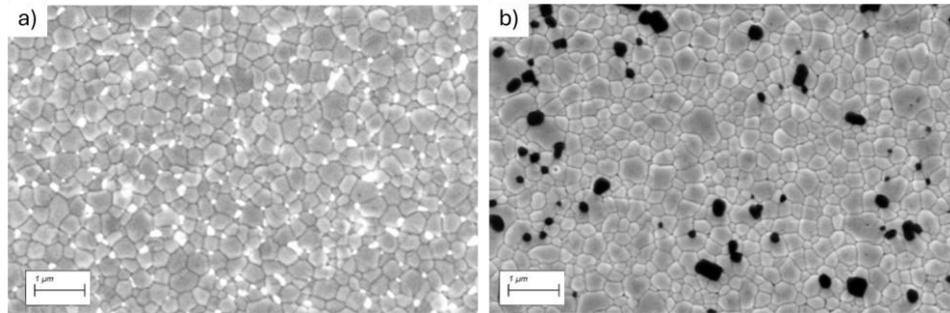


Figure 5: a) SEM image of a 1-step processed perovskite film and b) of a 2-step hybrid film. * The white regions in a) are interpreted to be Pbl₂ clusters, while the dark regions in b) is the excess pFBPA that segregated on the surface of the film.

Finally, devices were produced using a similar architecture, with the exception that 1-step-processed perovskite films required the use of partial layer of SiO_x nanoparticles to yield continuous films.⁶ The current-voltage (J-V) properties acquired in standard test conditions (STC) are shown in Figure 6.

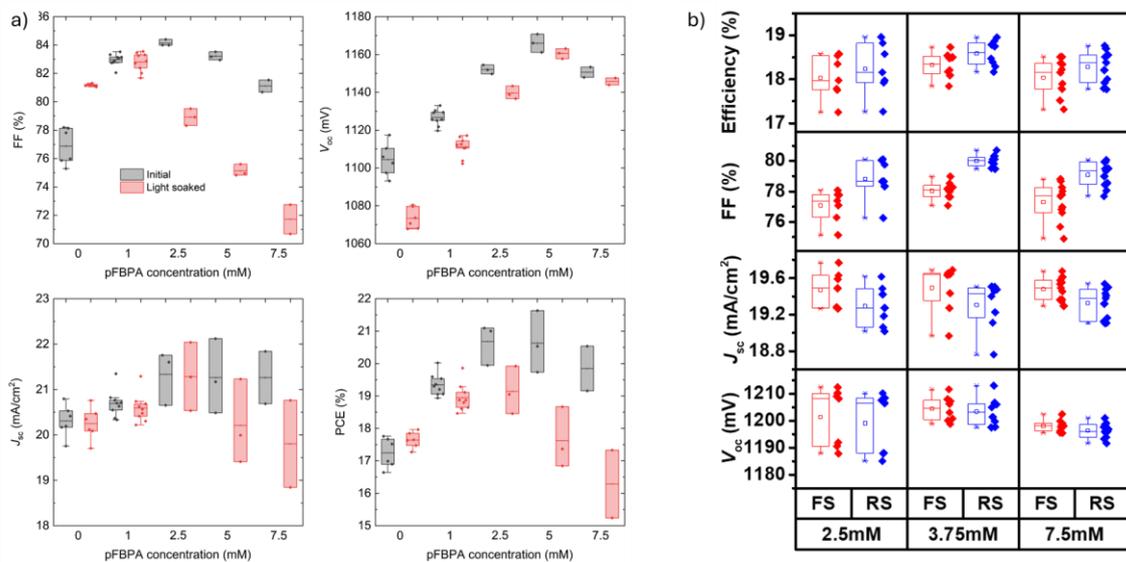


Figure 6: a) J-V data of 1-step processed solar cells (0.1 cm² active area) as function of pFBPA concentration in the perovskite ink, b) J-V data of hybrid 2-step perovskite solar (0.25 cm² active area), again as a function of pFBPA concentration.⁶*

Perovskite solar cells featuring a 1-step spin-coated absorber (with DMF) exhibit a slightly higher power conversion efficiency (PCE), about 1%abs higher, than the perovskite solar cells produced using the hybrid 2-step method (which is DMF free). This difference results from the faster optimization enabled by spin-coating compared to the hybrid 2-step process, where the absorber takes longer to optimize due to long and complex co-evaporation step. In both cases, the use of pFBPA has a beneficial impact on the PCE as it improves the microstructure of the perovskite film but also mitigates voltage losses present at the

perovskite/C60 interface.^{6*} However, it is worth noting that high concentrations of pFBPA (>2.5 mM) make the devices less stable in light soaking conditions, especially for the case of 1-step perovskite films (Figure 6a).

Perovskite absorbers processed with either the 1-step or the 2-step route were then implemented in 2-terminal perovskite/Si tandem solar cells. The devices exhibited similar (certified) PCE, as shown in Figure 7, highlighting that the hybrid method, which is free of any toxic solvent, is a viable route to process high efficiency devices, including record tandem solar cells.

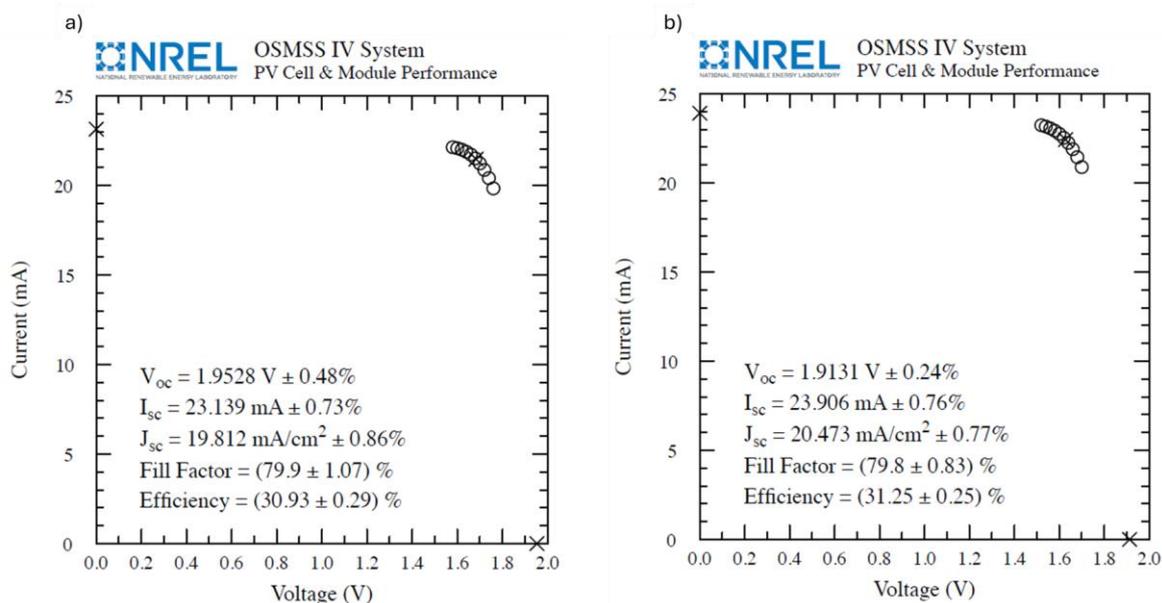


Figure 7: a) Certified J-V data of a perovskite/Si tandem solar cell (1 cm²) featuring a perovskite absorber processed with a 1-step spin-coating method, b) Certified J-V data of a perovskite/Si tandem featuring a perovskite absorber processed with a 2-step hybrid co-evaporation/spin-coating method.^{6*}

This study demonstrates that the use of a hybrid method, which employs only gentle solvents, yields high-efficiency single-junction and, more importantly, perovskite/Si tandem solar cells. The absence of toxic solvent makes this method particularly promising for industrial processing, where large volumes of solvents need to be processed and where toxic solvents such as DMF may prove problematic to handle and costly. The main challenge now to make this hybrid 2-step method compatible with industrial production requirements is to accelerate the co-evaporation step and switch to linear sources (rather than point sources) and to replace the organohalide spin-coating step with a solution-based method compatible with high throughput processing (e.g. slot-die coating or inkjet printing).

2.3. UNITOV (Single step method)

This section will discuss the innovative approach taken by the research team at UNITOV to address the toxicity and environmental concerns associated with DMF, a commonly used



solvent in the conventional fabrication of PSCs as discussed before. DMF, known for its high toxicity and strict handling requirements, poses significant challenges for large-scale, safe, and environmentally friendly PSC production. To mitigate these issues, UNITOV developed an alternative solvent system by replacing DMF with ACN, a less toxic and more industry-compatible solvent, while retaining dimethyl sulfoxide (DMSO) in the formulation.

The two solvent systems compared in this study-DMF-DMSO and DMSO-ACN were evaluated to determine whether ACN could serve as a viable substitute without compromising device performance or stability.

2.3.1 Results:

The experimental data in the Figure 8 illustrates the PCE of PSCs using different SnO₂ dilution levels, represented on the x-axis, and compares the two solvent systems-DMF-DMSO (red background) and the eco-friendlier DMSO-ACN (green background). The SnO₂ dilution levels tested include ratios of 1:6, 1:4 (reference), and 1:1.5, with results presented for both forward (Fwd) and reverse (Rev) scan directions.

In the DMF-DMSO system (red background), the data reveals that the reference 1:4 dilution provides the highest PCE values, with both forward and reverse scans clustered around a relatively consistent performance range. Other dilution levels, such as 1:6 and 1:1.5, show lower PCE, indicating that SnO₂ dilution levels outside the reference may lead to suboptimal electron transport layers in this solvent system.

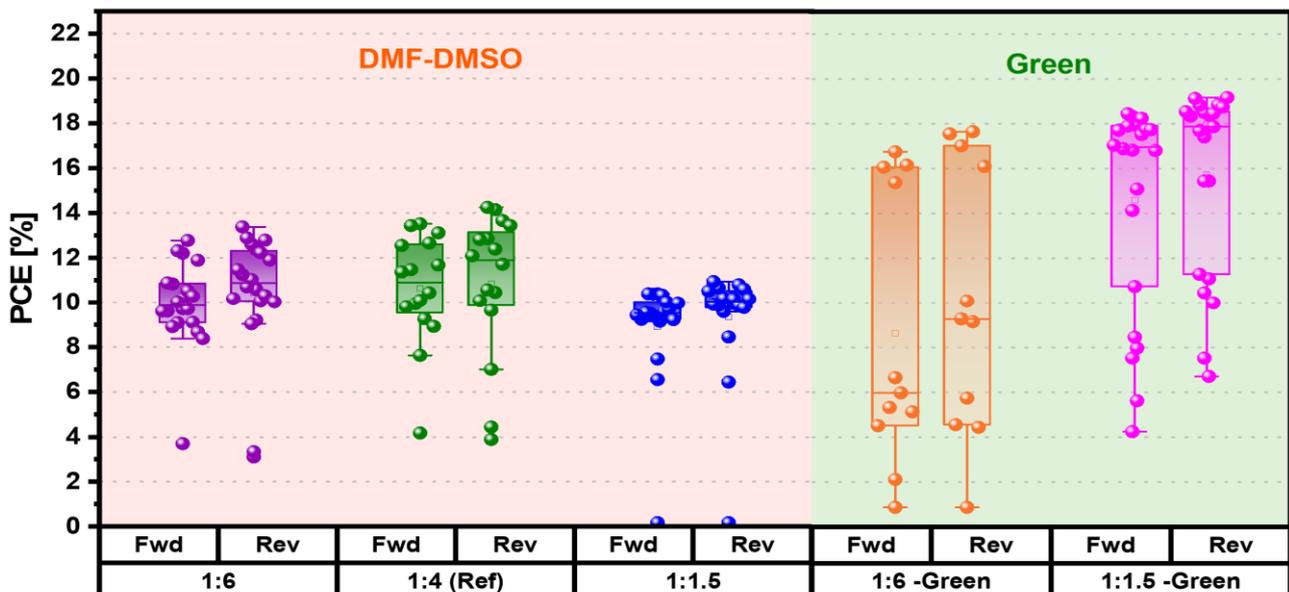


Figure 8. PCE comparison of PSCs fabricated with different SnO₂ dilution levels and solvent systems in forward (Fwd) and reverse (Rev) scan directions. The DMF-DMSO solvent system (red background) includes SnO₂ dilutions of 1:6, 1:4 (reference), and 1:1.5. The DMSO-ACN solvent system (green background), labelled as "Green".

For the DMSO-ACN system (green background), results indicate that the 1:6 and 1:1.5 dilutions performed particularly well in this system, with PCE values consistently higher than those seen with the DMF-DMSO system for the same dilutions but more so for 1:1.5 due to large variation in 1:6 diluted SnO₂. The lower performance consistency at 1:6 could be attributed to the higher dilution resulting in thinner SnO₂ films, which may inadequately cover the bottom electrode. This insufficient coverage could lead to direct contact between the perovskite layer and the bottom electrode, causing increased non-radiative recombination and thus reducing device efficiency.

In summary, the results suggest that while the DMF-DMSO system performs best with the 1:4 SnO₂ dilution, the DMSO-ACN system enables higher efficiency at 1:1.5 dilutions, supporting its versatility and effectiveness as an alternative, less toxic solvent system.

2.4. TNO (Single step method)

This section describes an approach taken by the research team at TNO to focus on the use of a non-toxic solvent for the perovskite precursor ink. We show that with the use of DMSO, we eliminate the use of toxic solvent (mixtures). This prevents exposure risks of researchers and operators to potential harmful solvent vapours. The ink is toxic because of its solutes, not because of the solvent. Therefore, skin contact with the ink should be prevented since the solvent with its toxic solutes may penetrate the epidermis upon direct contact. So, it remains essential to apply proper protective measures by skilled, educated and trained personnel to work safely with the green solvent.

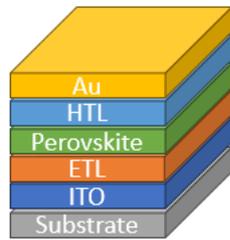
Compared to DMF, DMSO has a lower vapor pressure and higher boiling point. This strongly influences the film drying process and a possible quench step. Here we compare three different process methods to prepare NIP perovskite cells with an active area of 4 mm x 4 mm. After preparation, cells are measured with an aperture area of 3 mm x 3 mm.

The first method is the reference process often applied in a small-scale laboratory setting. Here the often applied DMF:DMSO solvent mixture is applied in a spin coat experiment with a solvent quench step in a N₂ filled glove box. The second method involves the same perovskite precursor composition but dissolved in the non-toxic DMSO. The ink is applied using blade coating under ambient conditions, no quench step is applied. The third method uses the same DMSO based ink. The ink is applied via slot die coating on a flexible substrate. A vacuum quench step is applied to control the crystallization step.

Below, Figure 9, summarizes the device stack and the materials applied throughout this campaign to move from perovskite solar cells processed on rigid glass substrates via spin coating under a Nitrogen atmosphere with a standard DMF based ink, towards flexible perovskite solar cells prepared with a scalable technique (slot die coating) on flexible substrates with a green, industrially relevant solvent under ambient conditions. It is shown



that good device performances can be obtained with or without quench methods.



- ETL: SnO₂ nanoparticle dispersion in water
- Perovskite: 2C perovskite Cs_xFA_{1-x}Pb(I_yBr_{1-y})₃ with a bandgap in the range 1.55-1.6 eV
- HTL: Organic transport layers, PTAA and Spiro OMe-TAD, same dopants, Li:TFSI and TBP in either case
- Top electrode: Au

Figure 9. Summary of the device stack applied in this study.

2.4.1 Results

The first step in this campaign was to prepare reference devices. For this NIP perovskite solar cells were prepared from a DMF:DMSO ink. The device stack is presented in Figure 9. During the drying step, an anti-solvent quench step was applied. The JV-data of the batch are summarized in figure 10. The data reveal a narrow distribution in current density, fill factor and open circuit voltages measured on the devices. The power conversion efficiency as determined from the reverse JV curves is typically between 16 and 17 %.

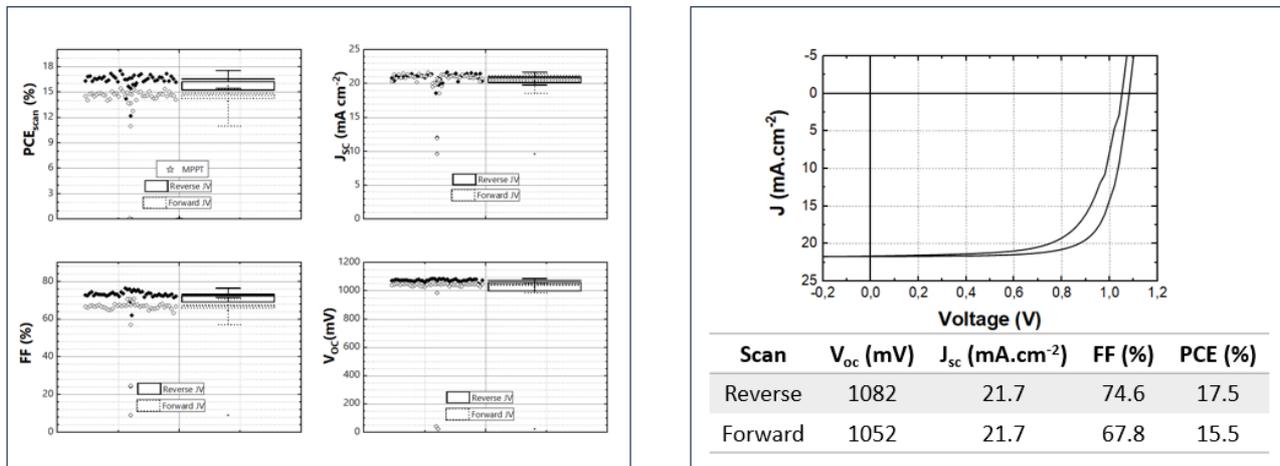


Figure 10. Overview of the JV-parameters (left panel) of NIP perovskite solar cells processed from a DMF:DMSO ink on rigid (glass/ITO) substrates via spin coating. During the drying process, an anti-solvent quench step was applied. The panel on the right shows the reverse and forward JV-curve as well as the MPP-tracked diagram of the best device in the experiment. The table shows the corresponding JV-parameters.

The second step in the campaign involves moving from spin coating to blade coating as an intermediate step towards slot die coating. Samples were prepared on the same glass/ITO substrates with the device stack given in Figure 9. In this second step, the DMF:DMSO ink was replaced by DMSO ink which was used under ambient conditions without quench step. Figures 11 and 12 summarize the results obtained in the batch.

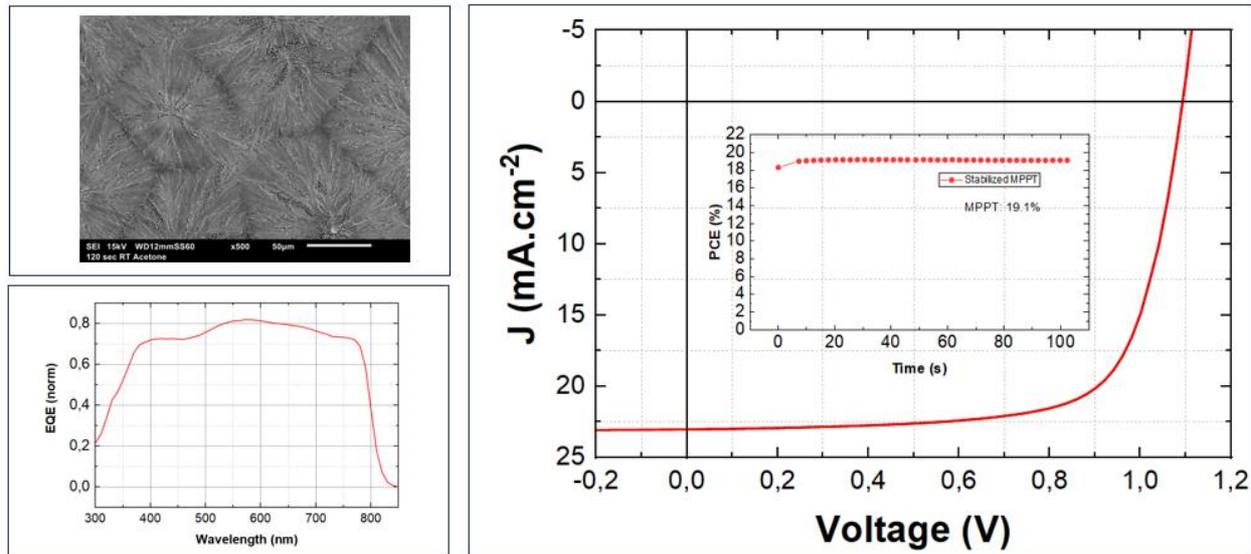


Figure 11. Left upper panel shows a SEM image the perovskite layer formed by blade coating the DMSO ink on a glass/ITO/SnO₂ substrate without quenching method. The right panel presents the JV-curve as well as the MPP-tracked diagram of the best device in the experiment. The lower left panel gives the EQE spectrum of the sample.

As can be seen from the best functioning device, the performance is even slightly higher as compared the best spin coated device using the DMF:DMSO ink. This is a very promising result as it shows well performing devices can be produced a non-toxic solvent under ambient conditions.

The SEM image given in Figure 11 reveals the microstructure of the perovskite layer consisting of relatively large (100 micron) dendritic domains with a fine substructure. XRD measurements on several samples reveal that the perovskite absorber layer consists of almost single phase α -perovskite, with only traces of CsPbI₃ as secondary phase. These results shows that the DMSO ink processed under ambient conditions, without applying a quench process, can lead to a good quality perovskite layer on glass/ITO/SnO₂ substrates leading to good device performance.

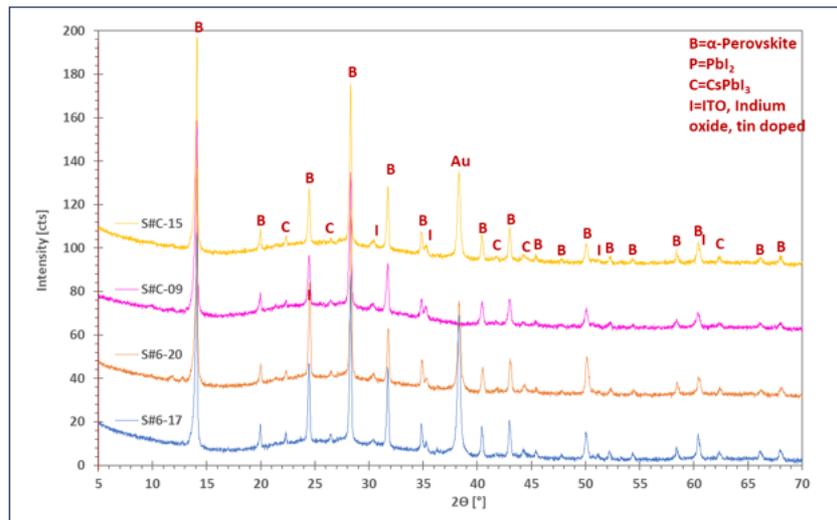


Figure 12. X-ray diffraction (XRD) patterns of several blade coated perovskite layers on glass/ITO/SnO₂. Note, due to the X-ray transparency of the sample also features of the underlying ITO layer are visible.

In the third and final step of the campaign we move from blade coating to slot die coating as an industrial coating method. Further we move from 3 cm x 3 cm rigid glass substrates to flexible 6-inch PET/ITO foils. Similarly to method 2, the DMSO ink was applied under ambient conditions. Also, the device stack, given in Figure 9, remains the same in this batch. Contrary to method 2, a vacuum quench step was applied. Figures 13 and 14 summarize the results obtained in the batch.

Figure 13 presents an optical transmission scan of a slot die coated perovskite layer on a 6 inch PET/ITO/SnO₂ substrate. Besides some pin holes and non-uniformities at the sides of the coating, the coating is uniform.



Figure 13. Optical scan of the perovskite layer slot die coated on a 6-inch PET/ITO substrate. The perovskite layer is coated onto the SnO₂ layer.

After optimizing the perovskite layer deposition via slot die coating on the flexible 6-inch substrates, complete NIP perovskite solar cells were prepared and measured. The results are summarized in Figure 14.

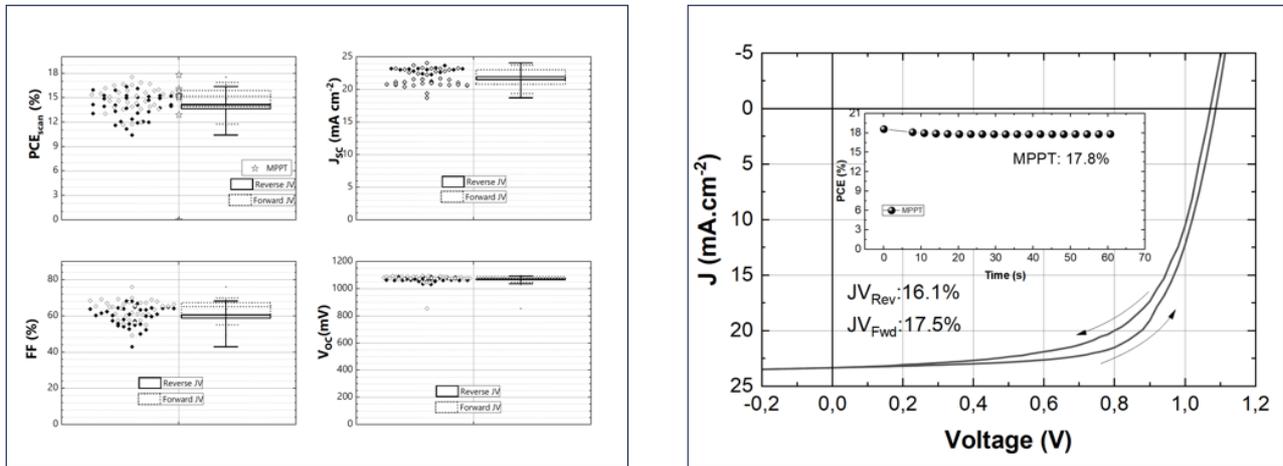


Figure 14. Overview of the JV-parameters (left panel) of flexible NIP perovskite solar cells processed from a DMSO ink on flexible (PET/ITO) substrates via slot die coating. During the drying process, a vacuum quench step was applied. The panel on the right shows the reverse and forward JV-curve as well as the MPP-tracked diagram of the best device in the experiment.

Despite spread in the measured fill factor amongst the characterized devices, good performance for flexible devices were obtained with the ink based on non-toxic DMSO, using an industrially relevant deposition method under ambient conditions.

In conclusion, the reported results show that DMSO, a non-toxic solvent, can be used to prepare perovskite precursor inks that can be applied by blade coating without a quenching process under ambient conditions on a glass substrate with an efficiency of over 19%. Furthermore, the same ink can be applied via slot die coating on flexible substrates under ambient conditions leading to performances of 17.8 %.

3. CONCLUSION

This report highlights important progress in using safer, more environmentally friendly solvents for making efficient perovskite solar cells. By replacing toxic solvents like DMF with greener options like acetonitrile (ACN) and dimethyl sulfoxide (DMSO), we have shown that it is possible to produce solar cells that perform just as well while being safer for both people and the environment.

The research also demonstrated new methods, such as single-step and two-step approaches, that work with these green solvents. These methods were successful in

creating both rigid and flexible solar panels, showing they can be used for large-scale manufacturing.

Overall, this work supports the goals of the VIPERLAB project by providing safer and scalable solutions for making perovskite solar cells. These advancements bring us closer to producing sustainable, high-performing solar technology that can help drive Europe's leadership in renewable energy.

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