Upscaling Perovskite: Optoelectronics and Morphology of Slot-Die Coated Solar cells

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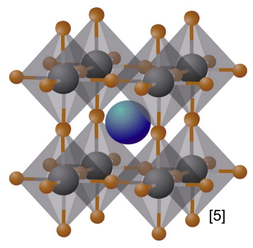
1. Why Perovskite Solar Cells

Image: Panasonic. [2]

+ PCE >20% for large areas cells[1]
+ Solution-based deposition process
+ Low energy payback time
+ Thin and flexible substrates
+ Bandgap tunability
– Long-term stability

- Low-temperature (<100 °C) processing enables deposition onto ultra-thin and flexible plastic substrates unleashing new applications [3]
- Slot-die coating is one of the most promising techniques to upscale perovskite-based solar cells [4]

2. The ABX_3 Perovskite Structure

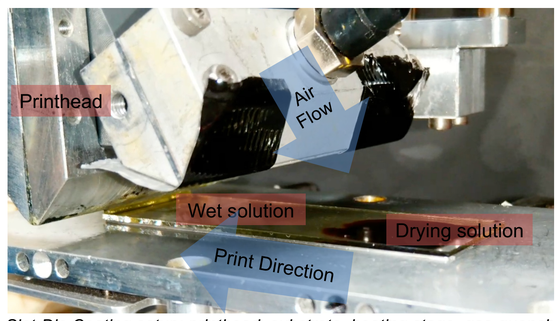


A: $CH_3NH_3^+$ (Methylammonium, MA), $CH(NH_2)_2^+$ (Formamidinium, FA)
B: Pb^{2+} (Lead), Sn^{2+} (Tin)
X: I^- (Iodide), Br^- (Bromide), Cl^- (Cloride)

Perovskite crystallizes in a cubic structure.

- In this work the halide ion (X) is tuned to optimize the film quality and the final cell performances
- **MACl** and **MABr** precursors are used as an additive to obtain a final composition with a molar ratio $MAI:PbI_2:MACl$ (MABr) of 1:1.1:0.15 in 2-Methoxyethanol (2-ME) and DMSO [6]

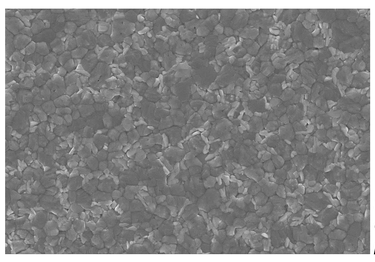
3. Slot-Die Coating



Slot-Die Coating setup: printhead, substrate, heating stage, gas-quench. Not visible: 2 axis motor, solution syringe and controller.

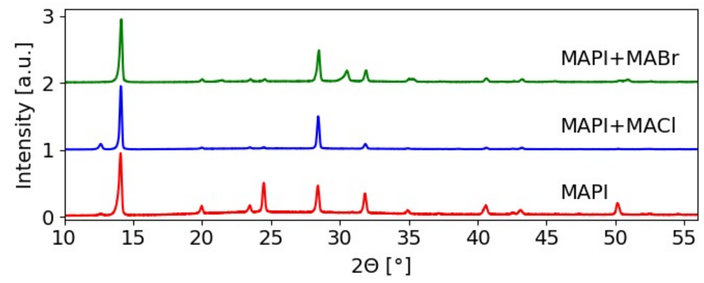
From the SEM surface images we observe:
- Uniform pinhole-free surface
- Large crystal area

- Fast and easily upscalable process
- Deposition of a highly uniform and compact layer
- Thin-films can be processed on both rigid and flexible substrates
- Supreme control on the crystallization process

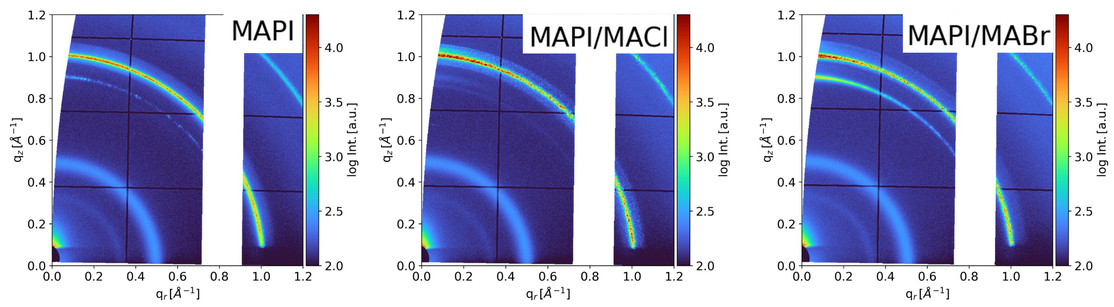


SEM surface image of the printed thin perovskite film.

4. Morphological Properties

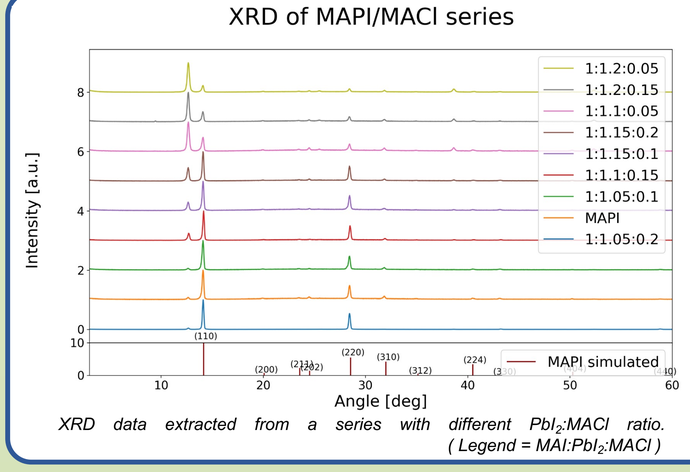


X-Ray Diffraction is a powerful technique to study the structural properties of thin films:
- Good agreement with theoretical MAPI data
- Extra peaks due to lead iodide excess
- Differences in peak intensity ratios on non-parallel directions hint to a dissimilar crystal orientation for the three samples



- GIWAXS data confirms the absence of lead iodide in the MABr formulation
- Increased texture (preferential crystal orientation) in the MAPI/MACl formulation seen in the (110) perovskite peak

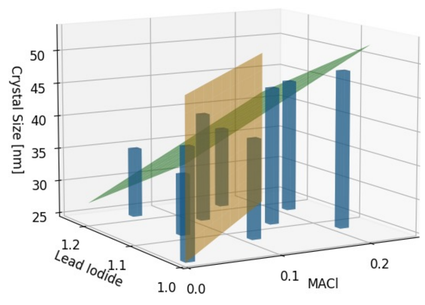
5. MACl additive



XRD data extracted from a series with different PbI_2:MACl ratio. (Legend = $MAI:PbI_2$:MACl)

- MACl concentration has been varied to study orientation phenomena on the most promising additive
- The ratio between PbI_2 (~12°) and perovskite peaks (~14°) decreases with increased MACl

6. Scherrer equation and crystal size

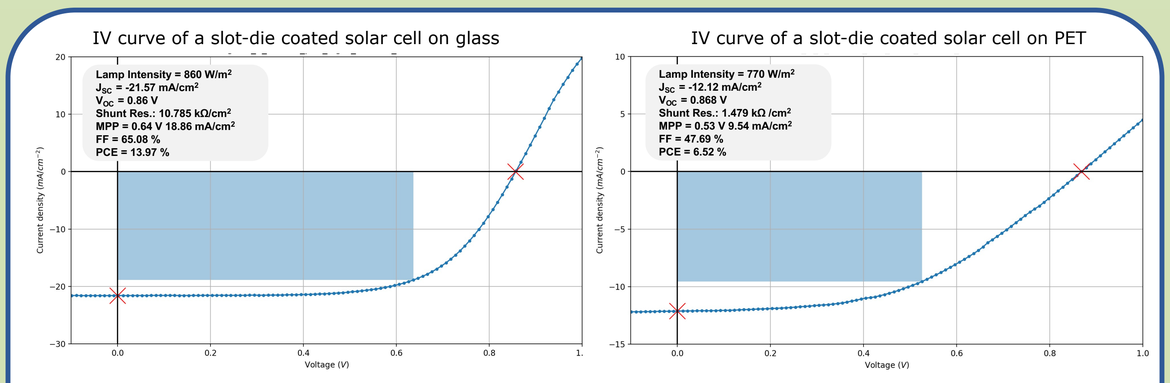


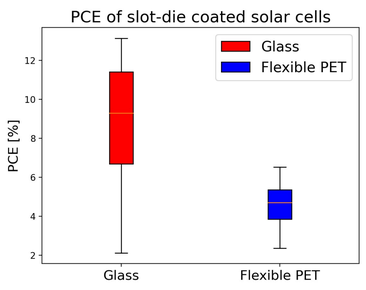
$$D = \frac{K\lambda}{\beta \cos\theta}$$

Where:
D = mean size of crystalline domain
K = ideality factor
λ = X-ray wavelength
β = FWHM peak broadening
ϑ = Bragg angle

- Increasing MACl additive to 20mol%, leads to enlarge crystal size compared to pure MAPI.
- PbI_2 excess decrease the crystal size
- A larger cryst al size could lead to less recombination and thus greater PCE

7. Electrical Characterization



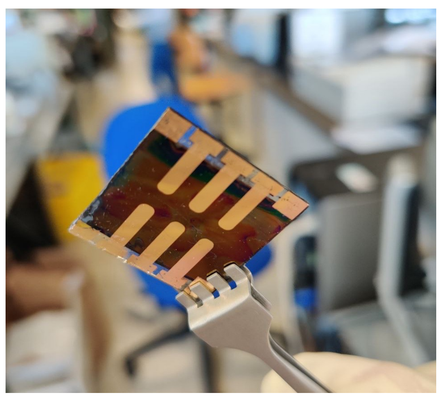


- The **PCE** of the champion slot-die coated MAPI/MACl solar cell is almost **14%** and a statistical analysis shows good reproducibility.
- The lower PCE of the solar cells printed on flexible PET is partially explained by an higher surface resistance (60 Ω/sq. vs. 8 Ω/sq. for rigid substrates). This leads to a high series resistance and a low FF.

8. Summary and Outlook

- Additives changing the composition of the halide ion result in strong differences in the crystal orientation
- MACl addition results in increased texture of the (110) perovskite peak and larger crystal size
- Preliminary results obtained for flexible substrates are a proof of concept of the capabilities of perovskite solar cells

Outlook: Optimize the manufacturing process to achieve high homogeneity and to maximize the SCs performances on flexible substrates.



Slot-die coated perovskite solar cell after electrical measurements.

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