Optimizing The Deposition Of Thin Layers Of Organic-Inorganic Hybrid Perovskite Methylammonium Lead Iodide (CH3NH3PbI3) On Large Surfaces Through Their Optical Properties

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INTRODUCTION

The present paper evaluates the deposition of thin films of organic-inorganic hybrid perovskite composed by methylammonium lead iodide (MAPI) on large surfaces (up to 75 mm x 75 mm) via spin coating techniques by considering their optical properties in order to assess their characteristics. Thus, in order to map the deposited area and realize studies about their optical properties, the absorption coefficient, the refractive index and the thickness of the layers were determined by using a high accuracy spectroscopic ellipsometer system based on rotating compensator ellipsometer (RCE) technology.

An Atomic Force Microscopy (AFM) was used to verify uniformity and thicknesses of the layers. Also, the samples were studied with a fluorescence spectroscopic system, in order to evaluate their photoluminescence properties.

METHODOLOGY

Phase I:
- Spin coating on small size substrates.
- Several MAPI weight concentrations & Accelerations.

Phase II:
- Survey of optical properties for PK materials @ literature.
- Measurements with spatial resolution by mapping the substrates.
- Study of the optical properties:
  - Absorption coefficient (α).
  - Refractive index (n).
  - Transmittance & Reflectance.
  - Ideal efficiency.
  - Fluorescence properties.

Phase III:
- Thickness & uniformity assessment.
  - Ellipsometer.
  - AFM.

Phase IV:
- Spin coating on larger substrates.
  - Best recipe from Phase I.

EXPERIMENTAL

Fabrication conditions:
- Clean room (ISO 7).
- Temperature (~21 °C).
- Relative humidity (RH) 10%.

Spatial resolution measurement procedure:

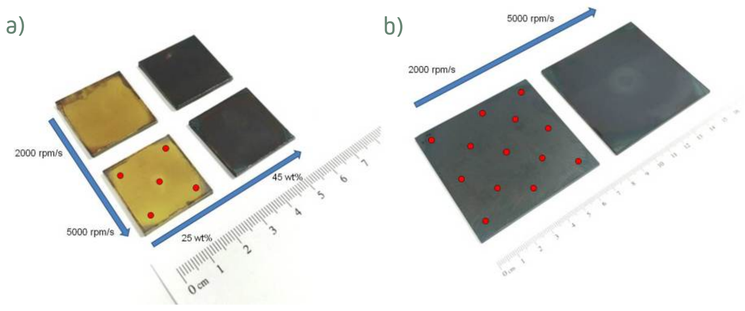

Figure 1: PK thin films deposited on (a) 25 mm × 25 mm substrates and (b) 75 mm × 75 mm substrates, highlighting the measurement points (red dots).

METHOD VALIDATION

- Theoretical fit model derived.
- Optical constants reported as control points.
- Thicknesses measured with spectroscopic ellipsometer and AFM with special samples.

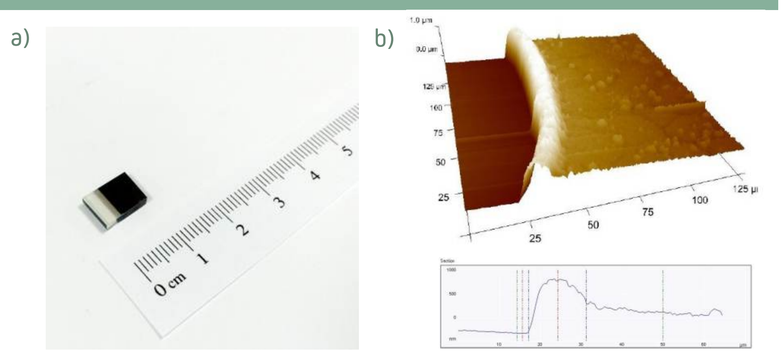

Figure 2: (a) Special sample deposited for measurements and (b) AFM image and cross section, showing the interface between the MAPI-free and MAPI areas in one sample.

RESULTS AND DISCUSSION

SMALL SUBSTRATES

Substrate size: 25 mm x 25mm
MAPI solutions: 25 wt% - 45 wt%
Wafer spinner set up:
- 5000 rpm.
- 2000 rpm/s - 5000 rpm/s

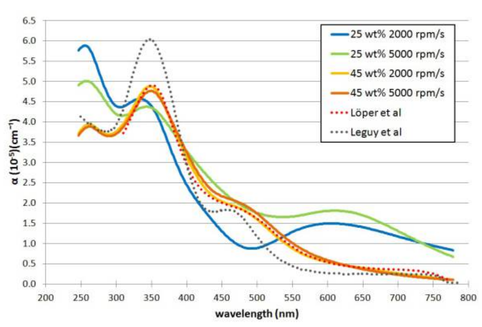

Figure 3: Resulting absorption coefficient (α) & values published in the literature.

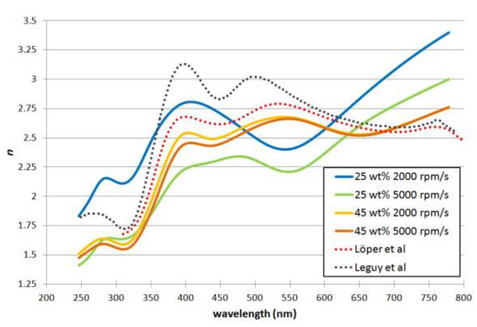

Figure 4: Resulting refractive index (n) & values published in the literature.

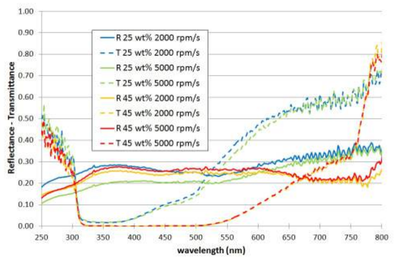

Figure 5: Reflectance and Transmittance versus wavelength

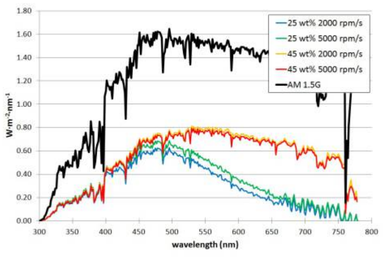

Figure 6: Ideal efficiency as per Xie Ziang et al methodology.

Solution (wt%)	Acceleration (rpm/s)	Substrate size (mm)	Average thickness (nm)	Standard deviation (nm)
25	2000	25 x 25	250.82	43.97
25	5000	25 x 25	262.81	63.16
45	2000	25 x 25	439.07	3.25
45	5000	25 x 25	432.15	4.37

Table 1: Average thickness & standard deviation.

LARGER SUBSTRATES

Substrate size: 75 mm x 75mm
MAPI solution: 45 wt%
Wafer spinner set up:
- 5000 rpm.
- 2000 rpm/s - 5000 rpm/s

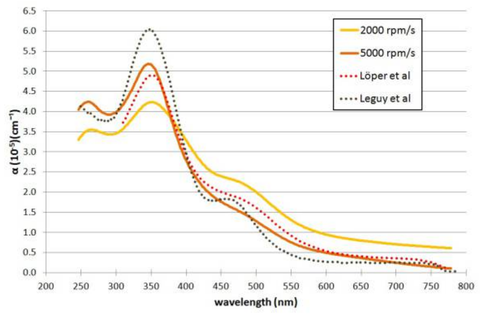

Figure 7: Resulting absorption coefficient (α) & values published in the literature.

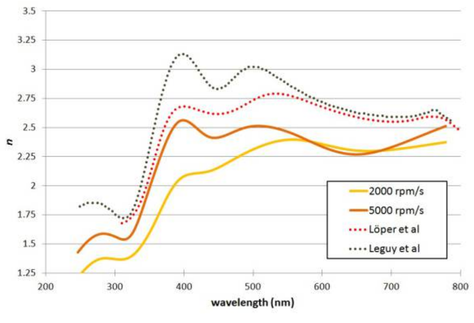

Figure 8: Resulting refractive index (n) & values published in the literature.

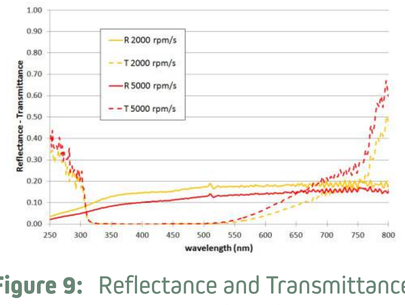

Figure 9: Reflectance and Transmittance versus wavelength

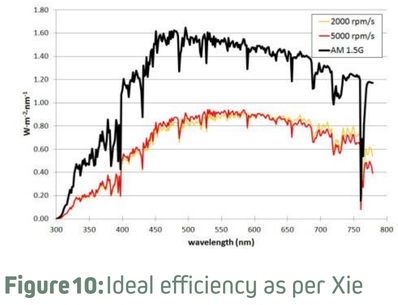

Figure 10: Ideal efficiency as per Xie Ziang et al methodology.

Solution (wt%)	Acceleration (rpm/s)	Substrate size (mm)	Average thickness (nm)	Standard deviation (nm)
45	2000	75 x 75	397.21	141.12
45	5000	75 x 75	478.55	117.05

Table 2: Average thickness & standard deviation.

EXCITATION - EMISSION MAPPING SCAN

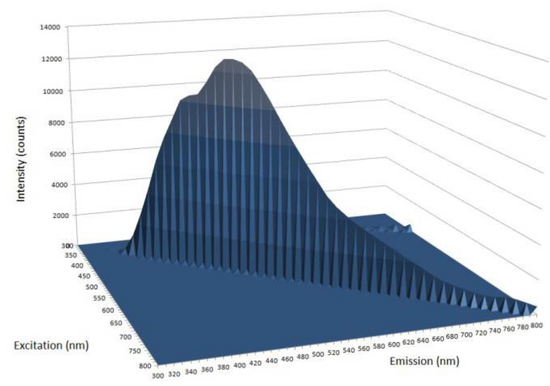

Figure 11: Excitation - Emission Mapping Scan of sample (45wt% MAPI & 5000 rpm/s).

CONVERSION

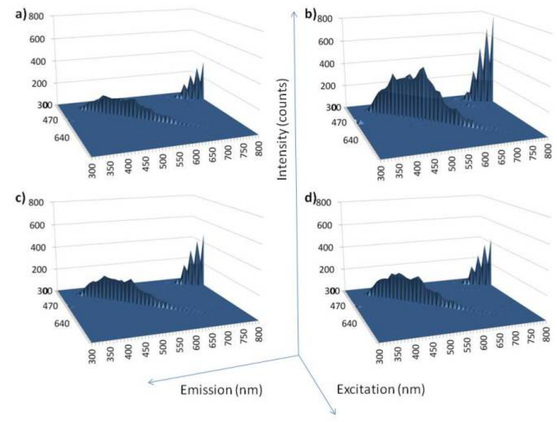

Figure 12: Emission produced at different wavelengths from the excitation: (a) 25 wt% 2000 rpm/s, (b) 25 wt% 5000 rpm/s, (a) 45 wt% 2000 rpm/s and (a) 45wt% 5000 rpm/s. Z axis = intensity in photon counting, X axis = excitation wavelength and Y axis = emission wavelength.

CONCLUSION AND DISCUSSION

- Characterization methodology demonstrated by alternative procedure.
- Theoretical fit model developed for elipsometry measurements.
- 45 wt% MAPI weight concentration solutions as better posed for:
  - Alternative as ARC.
  - Tandem solar cell.
- Supported by optical properties.
- Upsizing substrate achieved (uniformity improvement pending).