

Mechanism of perovskite film formation and the effect of light

Amita Ummadisingu and Michael Grätzel

Laboratory of Photonics and Interfaces (LPI), Institute of Chemical Sciences and Engineering, École Polytechnique Fédérale de Lausanne (EPFL) Station 6, CH-1015 Lausanne, Switzerland
amita.ummadisingu@epfl.ch

In recent years, perovskites have emerged as excellent candidates for use as light harvesters in solar cells. Various deposition methods such as sequential deposition and the anti-solvent methods have been developed for the preparation of perovskite solar cells, with major effort focused on achieving high performance [1]. However, factors controlling the final film morphology in perovskite formation in these deposition methods are little understood, as the fundamental mechanisms are still unclear. This gap in knowledge leads to batch-to-batch inconsistencies in the morphology and thereby the device performance.

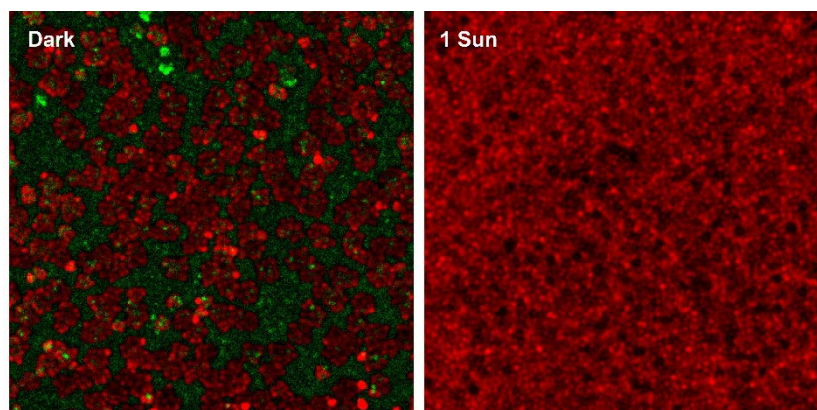


Figure 1: CLSM images (each showing an area of $37 \times 37 \mu\text{m}^2$) of PbI_2 films reacted with $\text{CH}_3\text{NH}_3\text{I}$ solution for 10 s. PbI_2 is emissive between 500 and 550 nm (shown in green) while the $\text{CH}_3\text{NH}_3\text{PbI}_3$ perovskite is emissive between 700 and 800 nm (shown in red). Left: Film prepared in the dark showing the perovskite crystals forming (in red) amidst unconverted PbI_2 (in green). Right: Film prepared under 1 Sun illumination showing the accelerated formation of perovskite (in red).

Using confocal laser scanning fluorescence microscopy (CLSM) and scanning electron microscopy, we identify that illumination during film formation is a major factor in the reaction as it greatly accelerates perovskite formation. We demonstrate the unexpected effect of illumination on the nucleation in the sequential deposition method and unravel the underlying mechanism through photo-electrochemistry. Furthermore, we show that the light effect is present even in the anti-solvent method, the route used for the fabrication of high-efficiency solar cells. Our results establish that illumination is a major factor in various deposition methods and that it should always be considered while preparing perovskite films. We show that light is an efficient and convenient way to control the perovskite morphology for opto-electronic applications [2].

[1] M. Grätzel, *Nature Materials*, **2014**, *13*, 838-842.

[2] A. Ummadisingu, L. Steier, J. Y. Seo, T. Matsui, A. Abate, W. Tress and M. Grätzel, The effect of illumination on the formation of metal halide perovskite films. *Nature*, **2017**, *545*, 208-212.