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Microstructural Control of Hybrid and Inorganic Lead Perovskites Using Sequential Deposition

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The development of high-efficiency lead perovskite solar cells requires precise control of the structural, electrical, chemical and interfacial properties of each layer within these devices. The rapid nucleation and grain growth dynamics of the perovskite during its deposition has created significant challenges in fulfilling these criteria. A number of complimentary deposition approaches have emerged to overcome this fabrication problem, looking at either inducing rapid nucleation or decoupling the nucleation and growth stages from the initial deposition step. This latter method has been termed the sequential deposition method, which provides a facile approach to control the structural properties, while also enabling scaling of these devices. The primary issue in the sequentially deposited perovskite films is the incomplete conversion of the conventionally used, compact PbI_2, which arises because of diffusion limitations within the second step. Any unreacted PbI_2 in the perovskite films arising from this incomplete conversion has the potential to impact stability, performance, and reproducibility of the devices. In this work, we have developed an approach to deposit mesoporous lead iodide (*m*-PbI_2) during the first step by using dimethylsulphoxide (DMSO) as a processing additive. In addition to this molecular additive, the temperature of both the PbI_2 precursor solution and the substrate were determined to be key factors towards achieving microstructural control of the pores in this first step. This porous structure facilitated enhanced diffusion of suitable halide salts, yielding full conversion of the PbI_2 layers into the perovskite films. The presence of DMSO in the mesoporous PbI_2 films was found to facilitate the conversion process by intramolecular exchange, which enabled for the formation of ultra-smooth thin perovskite films with large grains. We have demonstrated the viability of this *m*-PbI_2 in both hybrid and inorganic perovskite solar cells. To date, optimised power conversion efficiencies (PCE) of >15% and >10% have been achieved using this approach within planar heterojunction solar cell configurations in hybrid $MAPbI_3$ perovskite devices and $CsPbBr_2I$ inorganic devices, respectively.

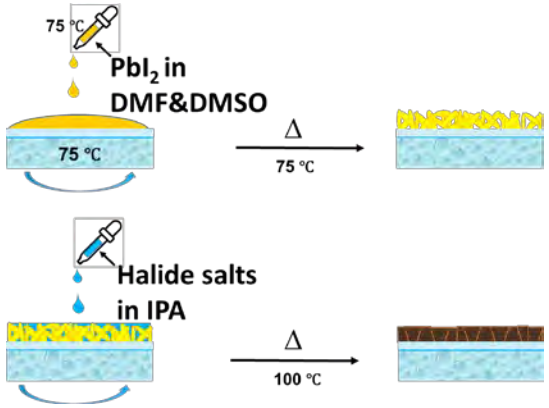

Figure 1. Schematics of two-step sequential deposition processes of fabricating mesoporous PbI_2 films and perovskite film.