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Perovskite Thin Film Formation: An In Situ Investigation of Scalable Processes

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Organic-inorganic hybrid lead halide perovskite semiconductors have attracted a great deal of attention because of their remarkable optoelectronic properties which make them potentially suitable as active materials in photovoltaics, light emission, and photodetection. The key reason for its popularity is that it can yield good semiconductor properties despite being solution processed in ambient conditions and requires no vacuum or excessive heating.

To date, the most efficient perovskite solar cells have been fabricated using spin coating, for which several ink and solvent engineering methods have been developed and perfected. However, this is a wasteful process which cannot be easily scaled up to continuous large area fabrication, where existing solvent engineering methods, such as anti-solvent dripping, are also unlikely to work. Here we compare the ink solidification and film formation mechanisms of $CH_3NH_3PbI_3$ in DMF by spin-coating *versus* blade-coating by utilizing a suite of in situ diagnostic probes including high speed optical microscopy, optical reflectance and absorbance, photoluminescence and grazing incidence wide angle x-ray scattering (GIWAXS), all performed during the coating, to monitor the solution thinning behavior, changes in optical absorbance, and nucleation and growth of crystalline phases of the precursor and perovskite. We have previously reported the existence of ordered intermediate phases during the thin film formation and its impact on final thin film morphology[1].

We show significant differences in the process kinetics and formation of complex intermediate phases between the two processes at room and intermediate temperatures. To overcome these challenges in the context of blade coating, the sample is heated during deposition. We observe high-quality film formation for T > 100^{o}C, namely in conditions which inhibit the formation of the crystalline intermediate complex phases. In doing so, we achieve fast and direct formation of the perovskite phase with solar cells yielding PCE > 17% as shown in Figure 1[2]. We investigate the universality aspect of our findings and explore the co-solvent system of GBL:DMSO. We show that intermediate complexes behave in similar way as in the case of DMF based ink. We observe high quality, pin hole free films when the ink is processed at higher temperatures[3].

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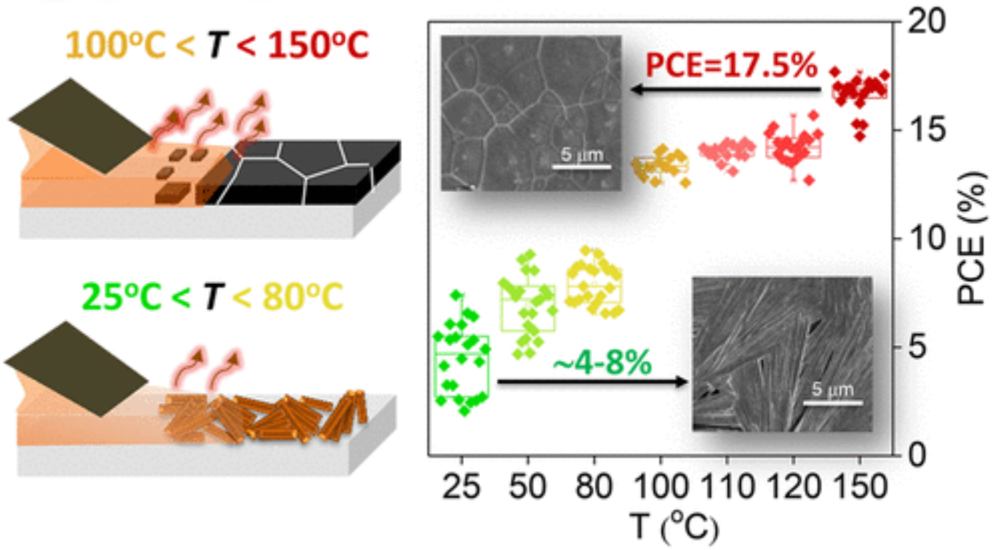

Figure 1 Schematic diagram of blade coating at different temperatures. A plot of blade coating temperature vs. power conversion efficiency of the devices with SEM images as inset. Higher coating temperature results in higher PCE and better reproducibility[2].

References

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