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Crystallization Control in Drop-Cast Quasi-2D Perovskites for Efficient Solar Cells

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Quasi-two dimensional (2D) Ruddlesden–Popper perovskites are attracting increasing attention for use in solar cells due to their significantly higher operational stability compared with 3D perovskites.[1] However, the highest-efficiency quasi-2D perovskite solar cells reported to date have been prepared using a hot spin-casting method,[2] which is unsuitable for upscaling. Recently, we reported a simple, low-waste, and scalable method to prepare quasi-2D perovskite films in which a precursor solution is drop-cast on a heated substrate and then spontaneously spreads to form a smooth, uniform film on drying.[3] In this previous work, focusing on solar cells comprising drop-cast butylammonium-based quasi-2D perovskites, the highest Power Conversion Efficiency (PCE) achieved was 14.9%. This result suggested that the drop-casting method might be a promising basis for producing high-efficiency solar cells, with opportunities to possibly further improve the PCE by using alternative organic cations and optimizing the crystallization process.

In this presentation we describe recent results obtained for drop-cast quasi-2D perovskites produced using a range of organic cations, including butylammonium, iso-butylammonium, and phenylethylammonium. The drop-cast quasi-2D perovskite films display significant differences in morphology, crystallization, and photovoltaic performance. Butylammonium-based quasi-2D perovskites consistently form high-quality drop-cast films, whereas the films formed using iso-butylammonium and phenylethylammonium are very rough and device performance is very low. Strategies, such as use of additives and mixed solvents, were investigated to control crystallization, and, thereby, to improve the quality of films produced using iso-butylammonium or phenylethylammonium cations. Smooth, uniform films of quasi-2D perovskite were successfully obtained using iso-butylammonium after optimizing the preparation process, resulting in PCEs of up to 15.5%, representing the highest PCE reported to date for quasi-2D perovskite solar cells using mono-ammonium cations. The crystallization and crystal orientation obtained using GIWAX will be discussed.

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