Composition and Crystallization Engineering of All-Inorganic Wide-Bandgap Perovskites for Inverted Solar Cells

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All-inorganic wide-bandgap perovskites are promising absorbers for single-junction and tandem solar cells because of their tunable optoelectronic properties and excellent thermal stability. However, their performance is often limited by poor film quality, compositional inhomogeneity, and non-radiative recombination. Here, we investigate two complementary strategies based on compositional and crystallization engineering to improve all-inorganic perovskite thin films and inverted solar cells. First, lead-free $CsSn_{1-x}Zn_xBr_3$ films were fabricated by physical vapor co-deposition. At an optimal Zn content of 4%, Zn was successfully incorporated into the $CsSnBr_3$ lattice without pronounced secondary-phase segregation. Zn alloying enlarged the grains, reduced pinholes, enhanced photoluminescence, and improved the energy-level alignment with the charge-transport layers. Consequently, the $CsSn_{0.96}Zn_{0.04}Br_3$ device achieved a power conversion efficiency of 2.59%, compared with 0.76% for pristine $CsSnBr_3$, and retained 83% of its initial efficiency after six days at 25 °C and 60% relative humidity. Second, the crystallization of solution-processed $CsPbI_2Br$ was regulated using 1 mol% $PbCl_2$. Time-resolved X-ray diffraction revealed earlier nucleation during room-temperature resting and slower crystal growth during thermal annealing. This controlled crystallization produced a more homogeneous depth-dependent I/Br distribution, reduced surface-potential fluctuations, and prolonged carrier lifetimes. The optimized $CsPbI_2Br$ device achieved an efficiency of 16.3% with an open-circuit voltage of 1.25 V, while the hysteresis index decreased from 13.6% to 7.0%. These results demonstrate that precise control of B-site composition and precursor crystallization kinetics can effectively improve film morphology, energetic uniformity, carrier dynamics, device efficiency, and stability in all-inorganic inverted perovskite solar cells.