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Facile Recrystallization Process for Efficient Triple-Cation Mixed-Halide Planar-Structure Perovskite Solar Cells

Jacob Tse-Wei Wang[†1], Liangyou Lin[† 1,b], Timothy W. Jones[1], Mihaela Grigore[3], Andre.Cook[1], Dane W. deQuilettes[4], Roberto Brenes[4], Benjamin C. Duck[1], Kenrick F. Anderson[1], Noel W. Duffy[5], Vladimir Bulović[4], Jian Pu[2], Jian Li[2], Bo Chi[*2], Gregory J. Wilson[*1]

[1]CSIRO Energy, Newcastle Energy Centre, 10 Murray Dwyer Cct, Mayfield West, NSW 2304, Australia.
[2]Center for Fuel Cell Innovation, School of Materials Science and Engineering, Huazhong University of Science &Technology, Wuhan 430074, China
[3]CSIRO Energy, 11 Julius Avenue, North Ryde 2113, Australia
[4]Research Laboratory of Electronics, Massachusetts Institute of Technology, 77 Massachusetts Avenue, Cambridge, Massachusetts 02139, USA
[5]CSIRO Energy Clayton Laboratories, Clayton, Vic 3168, Australia.
†These authors contributed equally to this work.
E-mail: jacob.wang@csiro.au

The state-of-the-art PSCs use mixed-cation and mixed-halide perovskite compositions due to the superior qualities over the archetypal $MAPbI_3$[1]. Adding Cs^+ to perovskite has been shown to improve qualities including thermal stability, reproducibility, and suppression of phase segregation.[2-4] In general, controlling the precise stoichiometry in a perovskite is critical for meeting its desired properties. However, it has been found purposely introduced excess PbI_2 in the perovskite causes significant influence on the intrinsic perovskite qualities. The few percent excess PbI_2 predominantly resides at grain boundaries, providing benefits of prolonged carrier lifetime, enhanced charge carrier injection, suppressed ion migrations and hysteresis, leading to improved overall device performance.[5-8] Theories have been proposed to explain these beneficial effects, including the better energy level alignments, grain boundary passivation, and superior crystal quality and crystallinity.[7-8] Nevertheless, there are also reports showing the residual PbI_2 can lead to degradation and fragile film structure with long-term device stability concerns.[9-11] Nonetheless, with most of the best performing cells still incorporating slight excess PbI_2, it is important to investigate this indefinite property function for further improving device performance and stability.

In this study, we use the $Cs_{0.05}(FA_{0.85}MA_{0.15})_{0.95}Pb(Br_{0.15}I_{0.85})_3$ stoichiometric perovskite structure as a platform to demonstrate a simple yet effective recrystallization method for PbI_2-rich perovskite crystals with a series of recrystallization agents. Surprisingly, for the first time, through the Grazing Incidence X-ray Diffraction (GIXRD), we found not only the excess PbI_2 in the film can be successfully removed, the perovskite had a profound recrystallization throughout the bulk, showing enhanced PL emission/lifetime, charge collection efficiency, reduced defects at grain boundaries (GBs) as well as superior device performance over 20% PCE.

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