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Nanaji Islavath , S Saroja , K Srinivas Reddy , P C Harikesh , G Veerappan , Shrikant V Joshi , Easwaramoorthi Ramasamy

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Nanaji Islavath, S Saroja, K Srinivas Reddy, P C Harikesh, G Veerappan, Shrikant V. Joshi, Easwaramoorthi Ramasamy*

Centre for Solar Energy Materials, International Advanced Research Centre for Powder Metallurgy and New Materials (ARCI), Hyderabad 500005, India

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Abstract

High-efficiency perovskite solar cells (PSCs) reported hitherto have been mostly prepared in a moisture and oxygen-free glove-box atmosphere, which hampers upscaling and real-time performance assessment of this exciting photovoltaic technology. In this work, we have systematically studied the feasibility of all-ambient-processing of PSCs and evaluated their photovoltaic performance. It has been shown that phase-pure crystalline tetragonal $MAPbI_3$ perovskite films are instantly formed in ambient air at room temperature by a two-step spin coating process, undermining the need for dry atmosphere and post-annealing. All-ambient-processed PSCs with a configuration of $FTO/TiO_2/MAPbI_3$/Spiro-OMeTAD/Au achieve open-circuit voltage (990 mV) and short-circuit current density (20.31 mA/cm^2) comparable to those of best reported glove-box processed devices. Nevertheless, device power conversion efficiency is still constrained at 5% by the unusually low fill-factor of 0.25. Dark current-voltage characteristics reveal poor conductivity of hole-transporting layer caused by lack of oxidized spiro-OMeTAD species, resulting in high series-resistance and decreased fill-factor. The study also establishes that the above limitations can be readily overcome by employing an inorganic p-type semiconductor, copper thiocyanate, as ambient-processable hole-transporting layer to yield a fill-factor of 0.54 and a power conversion efficiency of 7.19%. The present findings can have important implications in industrially viable fabrication of large-area PSCs.