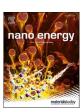


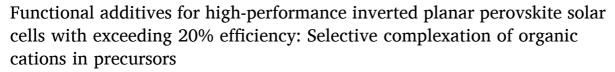
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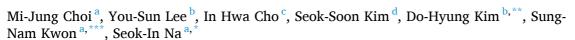
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ABSTRACT

Imperfection originating from the ionic nature of organic inorganic hybrid perovskite (OIHP) is a major factor that degrades perovskite solar cells (PSCs) performance, and mitigating or removing it is essential for the implementation of high-performance PSCs. In this work, 2-hydroxyethyl acrylate (HEA), having both effective C \equiv O and O \equiv H functional groups, is introduced as a functional additive that suppress imperfections by to control the crystallization process and passivate them at the same time, and the role of each function groups have been systematically investigated. We have found that O \equiv H functional groups of HEA strongly interacts with organic cations (i.e., MABr, FAI) to form more stable organic cation-HEA complex selectively, delaying the crystallization process of perovskite. As a result, HEA improves crystallinity and grain size of perovskite and reduces charge trapping density by passivation and/or suppression of defect states. HEA raises average cell-efficiency from 17.37% to 19.01% and the maximum efficiency is reached to 21.01% for the inverted planar PSC. In addition, large-area (1.08 cm 2) PSCs show an excellent efficiency of 20.40%, which is the highest efficiency in large-area inverted planar PSCs to date. PSCs with HEA also show long-term stability and better moisture resistance capabilities in ambient air conditions.

1. Introduction

Recently, perovskite solar cells (PSCs) have received considerable attention as an alternative energy source, and they have shown a rapid efficiency enhancement from 3.8% in 2009 to 25.2% (certified) in 2019 [1–3]. This new class of organic–inorganic hybrid perovskite (OIHP) has an ABX₃ perovskite structure, where A is a monovalent organic/inorganic cation, typically $CH_3NH_3^+$ (i.e., methylammonium cation; MA^+), $HC(NH_2)_2^+$ (i.e., formamidinium cation; FA^+), $C_2H_8N^+$ (i.e., ethylammonium cation; EA^+), EA^+ 0, EA^+ 1, EA^+ 2, EA^+ 3, EA^+ 3, EA^+ 4, EA^+ 5, EA^+ 6, EA^+ 7, EA^+ 8, EA^+ 9, EA^+

as a photoactive layer of solar cells [7], such as high absorption coefficient [9], low exciton binding energy [10], long electron and hole diffusion lengths [11], high charge mobility [12], long charge carrier lifetime [13], tunable band gap [14], and wide absorption range [15], which have allowed OIHP to emerge as one of the most promising materials for next-generation solar cells.

In perovskite solar cells, trapping of charge carriers has a profound effect on device performance. Charging carrier traps are known to be easily produced in perovskite layers by defect sites formed on perovskite surfaces or grain boundaries, thereby degrading device performance [16]. Importantly, the efficiency of PSCs can be determined by the recombination processes of photogenerated charge carriers [17–20].

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