



Dependence of halide composition on the stability of highly efficient all-inorganic cesium lead halide perovskite quantum dot solar cells



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ABSTRACT

All-inorganic perovskites suffer from low temperature phase instability and degradation under moisture and polar solvents. Here we have synthesized organic ligand assisted stable perovskite quantum dots (QDs) with uniform size ~ 10 nm. Varying the composition in $\text{CsPbBr}_x\text{I}_{3-x}$ it was observed that $\text{CsPbBr}_{1.5}\text{I}_{1.5}$ could be an alternative solution for stable perovskite QDs. Photoluminescence (PL) decay showed comparable average lifetime useful for photovoltaic (PV) application. The photovoltaic devices made from $\text{CsPbBr}_{1.5}\text{I}_{1.5}$ showed power conversion efficiency (PCE) $\sim 7.94\%$ with open circuit voltage ~ 1.0 V, fill factor 0.70 and long term stability. This PCE is also the highest as compared to other PV reports for QDs with $\text{CsPbBr}_{1.5}\text{I}_{1.5}$ composition. Moreover this composition offers nearly hysteresis free perovskite device.

1. Introduction

Trihalide perovskites of the formula ABX_3 (where $\text{A} = \text{Cs}^+$, CH_3NH_3^+ , $\text{HC}(\text{NH}_2)_2^+$; $\text{B} = \text{Pb}^{2+}$, Sn^{2+} ; $\text{X} = \text{Br}^-$, I^- , Cl^-) have been potentially the key material for solution processed optoelectronic applications like light-emitting diodes, laser, photodetectors and PV research within just 8 years due to their superior properties such as high optical absorption, tunable bandgap, intense emission and high carrier mobility [1–7]. Since the first report by Kojima et al. of 3.8% PCE in 2009 [1], halide perovskites have received tremendous interests in PV research with its current PCE of 22.7% [8], competitive with the conventional solar cells based on crystalline Si [9], CdTe [10] and CIGS [11]. Several modifications in architecture with compositional variations and fabrication processes have been attempted to reach such high efficiencies [12–15] but major problems linger with the organo-halide counterparts in terms of phase instability, poor stability under moist air and photo degradation [16–19]. Fabrication of large area devices by solution processing routes, retaining high efficiency similar to the small area lab-scale devices with proficient stability, is another concern that remains to be solved from commercialization prospects.

In this regard, colloidal all-inorganic perovskite QDs such as that of CsPbI_3 apprehended the attention from bulk organo-halide perovskites [20], due to their extraordinary PL quantum yield (PLQY), sharp emission characteristics, negligible electron/hole trapping and precise control over the size and shape of QDs [21–24]. With a coating of

formamidinium iodide on CsPbI_3 QD films, a significant improvement in PCE $\sim 13.4\%$ has been reported recently [25] asserting their presence alongside bulk organo-halide counterparts. Apart from PV applications, halide perovskite QDs have generated significant interest in light emitting diodes [26] and luminescent solar concentrators [27] due to their high PLQY reaching $\sim 100\%$ [28]. Notwithstanding the superior characteristics, CsPbX_3 perovskites too suffer from phase instability and degradation under moisture and polar solvents [29]. CsPbI_3 in particular is best suited for PV applications due to its appropriate bandgap (~ 1.73 eV) for solar energy absorption, but is equally unstable. CsPbI_3 can exist in its cubic α -phase (Fig. 1a) at comparatively high temperature (> 315 °C) known as “black phase” due to its dark colour. At low temperatures, this metastable α -phase transforms into the orthorhombic δ -phase (Fig. 1b) also known as “yellow phase” due to its colour. The δ -phase although is thermodynamically stable at room temperature, it is PL-inactive which prevents its use in PV applications [30].

Various master plans have been adapted to stabilize α - CsPbX_3 . Eperon et al. [31] was among the pioneers to report the stable cubic α -phase at moderately low temperature (100 °C) using hydroiodic acid as an additive to the precursor solution prior to spin-coating. Exceptional air and photo-stability of CsPbBr_3 QDs was reported by Pan et al. [32] using an inorganic–organic hybrid ion pair as the capping ligand resulting in high PLQY. Dastidar et al. [33] reported that chloride doping near the solubility limit of chloride in CsPbI_3 can stabilize the

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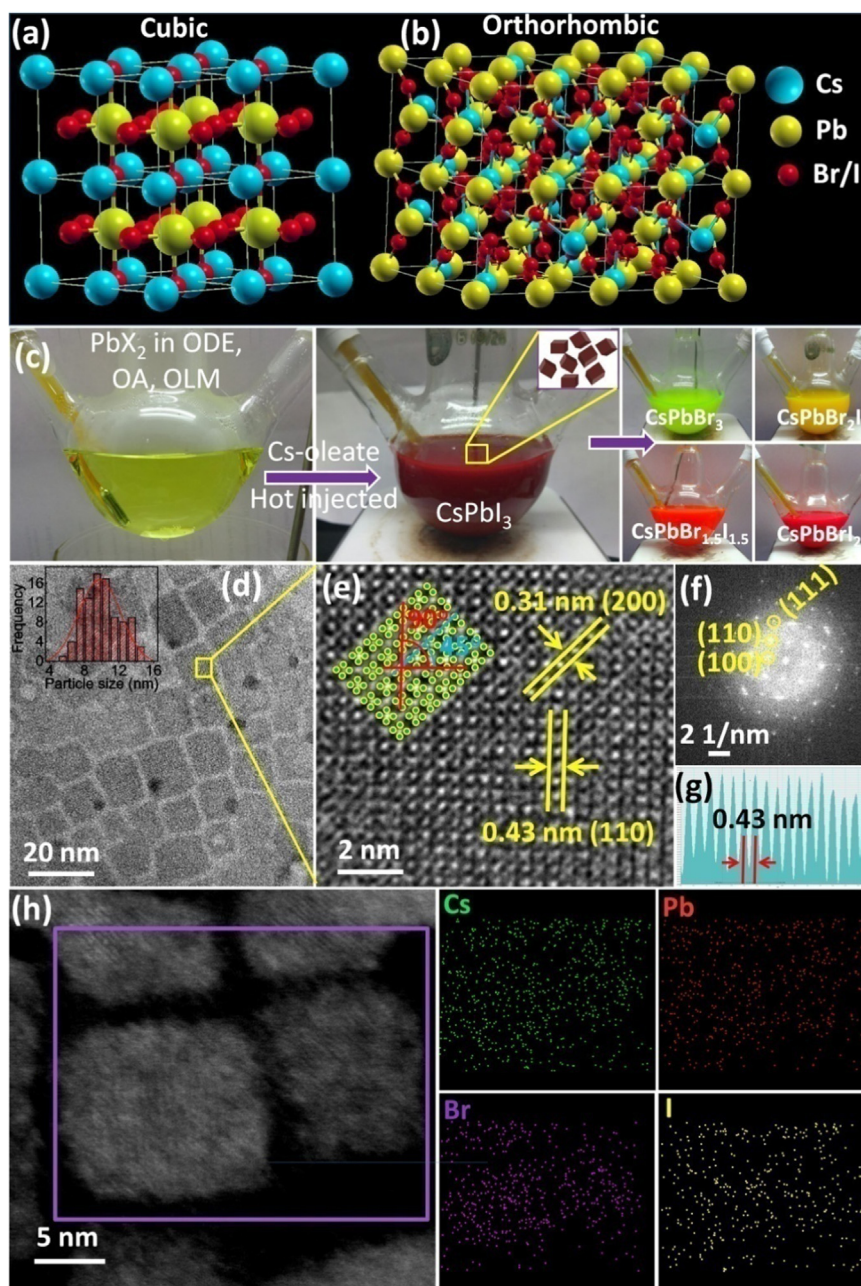


Fig. 1. (a) Cubic and (b) orthorhombic crystal structures of $\text{CsPbBr}_{3-x}\text{I}_x$. (c) The synthesis protocol with digital images of the as-synthesized perovskite QD dispersions. (d) TEM image of the representative $\text{CsPbBr}_{1.5}\text{I}_{1.5}$ QDs with inset showing the particle size distribution. (e) HRTEM image representing different lattice spacing and angle between planes. (f) SAED pattern and (g) simulated image of lattice planes from HRTEM image. (h) STEM-HAADF image and the corresponding elemental mapping.

α -phase. QD-induced phase stabilization of α - CsPbI_3 was documented by Swarnkar et al. [34] using oleic acid and oleylamine as ligands. Wang et al. [35] showed bis-(2,2,4-trimethylpentyl)phosphinic acid (TMPPA) as an oleic acid alternative in the synthesis of α - CsPbI_3 . In spite of this rich literature, it is difficult to stabilize α - CsPbI_3 particularly from the device point of view. Removal of surface ligand through chemical processes is necessary from the device perspective but this step results in phase degradation. On the contrary, the α -phase of CsPbBr_3 , a less attractive absorber, is essentially stable.

Apart from stability issues, the presence of toxic Pb in the perovskite solar cells is a major environmental concern which impedes their implementation for large scale applications [36]. Applying alternative metals such as Sn results in highly unstable perovskites. A systematic comparison between the environmental impacts of Pb- and Sn-based

perovskites was performed using zebrafish as the model organism [36]. Pb-based perovskite decomposes into $\text{Pb}(\text{OH})\text{I}$ complex and HI which is the possible route to its toxic character. Moreover migration of Γ^- ions in perovskite decomposes the organo-halides faster. Theoretical studies show that half substitution of Γ^- by Br^- ions raises the diffusion barrier and stabilizes the perovskite [37]. Therefore mixed halide compositions will likely stabilize the perovskite structure against decomposition and thereby reduce the probability of environmental exposure.

Herein we have shown that in $\text{CsPbBr}_x\text{I}_{3-x}$ ($x = 3-1$) perovskite QDs, halide composition is one of the key players which can create an optimal balance between the PV device efficiency and its stability. As compared to CsPbI_3 , the $\text{CsPbBr}_{1.5}\text{I}_{1.5}$ QD based PV devices have the advantage of improved stability under air at the cost of a slightly compromised PCE. After removal of surface ligands, CsPbI_3 QDs

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