

Highly stable organic-inorganic perovskite solar cells

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Abstract

We show that the introduction of benzoquinone (BQ) into an organic-inorganic perovskite film greatly enhances both power conversion efficiency and stability of solar cells. The morphology and crystal quality of the perovskite films were improved because intermolecular interaction between methylammonium iodide and BQ slowed the rate of perovskite crystal formation. Further, electron transfer from perovskite to BQ reduces charge recombination losses, and the oxidizing ability of BQ effectively suppresses the formation of metallic lead, a source of carrier traps, under continuous solar irradiation. Through BQ addition, the conversion efficiency was enhanced from 10.7% to 15.6% and lifetime extended about twenty-six times.

Finding new strategies for cheap, clean energy is a major challenge facing humanity. Organic-inorganic halide perovskite solar cells (PSCs) are an innovative, fast-growing field of research at the intersection of chemistry, materials science, physics, and device engineering. Although the reported power conversion efficiencies (η) of PSCs have recently surpassed 20%, the reproducible fabrication of efficient, stable PSCs remains challenging. Several methods, such as vacuum thermal evaporation, sequential deposition, solvent-induced precipitation, and additive-assisted deposition, have been developed to produce flat perovskite surfaces with good substrate coverage for high η , and solution-based processes for perovskite film fabrication from a precursor solution are especially promising for their simplicity.

Control of perovskite crystal formation by utilizing an intermediate phase of PbI_2 and *N,N*-dimethyl sulfoxide (DMSO) in the precursor solution led to a certified η of 20.1%. A phosphonic acid ammonium additive as a crosslinking agent was introduced between perovskite crystals, enhancing efficiency and extending device stability under 10% full sun intensity. However, while carrier traps are known to easily form in methylammonium lead triiodide ($\text{CH}_3\text{NH}_3\text{PbI}_3$) perovskite absorbers and degrade performance, the intrinsic source of traps and methods to suppress trap formation are poorly understood. Recently, we showed that the inclusion of water molecules in $\text{CH}_3\text{NH}_3\text{PbI}_3$ perovskite films is one source of hole trap formation and degradation in PSCs [1].

Eliminating water inclusion during PSC fabrication significantly improved the long-term stability of our PSCs.

In this study, we introduce the photochemically active additive BQ into a precursor solution of methylammonium iodide (MAI) and lead iodide (PbI_2) used for the fabrication of perovskite films to improve both morphology and stability [2]. Molecular interactions between MAI and BQ molecules reduce the speed of $\text{CH}_3\text{NH}_3\text{PbI}_3$ crystal formation, resulting in uniform, flat perovskite films with larger grains. The improved film formation along with reduced carrier recombination and efficient electron transfer from perovskite to BQ lead to an enhancement of η from 10.7% to 15.6% by BQ addition. More importantly, the addition of BQ effectively suppresses the formation of metallic lead, which we confirm to be a source of carrier traps, and improves device stability measured under standard laboratory weathering testing (ISOS-L-1 Laboratory) with a light intensity of 100 mW cm^{-2} without using a UV filter.

The pristine $\text{CH}_3\text{NH}_3\text{PbI}_3$ perovskite films were hazy and brown with a poly-dispersed small crystal island structure having poor surface morphology because of the relatively fast crystallization. Flat, uniform perovskite surfaces with large grains were obtained from the precursor solution containing

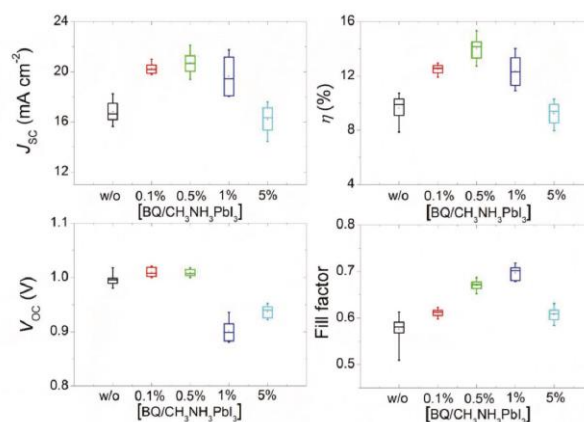


Fig. 1. Dependence of J_{sc} , V_{oc} , FF, and η on the BQ concentration of PSCs measured under one sun condition (100 mW cm^{-2} , AM1.5G) [2].

0.5% BQ. Similar morphologies were observed in films containing 0.1% and 1% BQ. When the molar ratio of BQ increased to 5%, the grains of $\text{CH}_3\text{NH}_3\text{PbI}_3$ became large with tall peaks, and grooves around the grains form, which might contribute to the formation of charge recombination centers and reduce performance.

The PSCs without BQ measured under a forward bias scan had a short-circuit current density (J_{SC}) of 18.23 mA cm^{-2} , an open-circuit voltage (V_{OC}) of 1.02 V, a fill factor (FF) of 0.58, and an η of 10.73% (Fig. 1). Adding 0.1% BQ improved η to 12.94% mainly because of an enhanced J_{SC} (20.64 mA cm^{-2}), which is in part due to the better film morphology induced by BQ additive. At the best BQ concentration of 0.5%, we obtained $J_{\text{SC}} = 21.55 \text{ mA cm}^{-2}$, $V_{\text{OC}} = 1.02 \text{ V}$, FF = 0.67, and $\eta = 14.69\%$. Further increasing the BQ concentration resulted in η dropping to 10.20% (5% BQ), which is lower than that of the PSCs without BQ, because of a decrease of V_{OC} .

The J_{SC} , V_{OC} , FF, and η of the best performing PSC containing 0.5% BQ measured under a forward bias scan were 21.32 mA cm^{-2} , 1.02 V, 0.71, and 15.56%, respectively, and were similar to the values obtained for a reverse bias scan with the same scan rate of 50 mV s^{-1} and an initial delay time of 2 s, indicating negligible hysteresis. On the contrary, PSCs without the BQ additive exhibited strong hysteresis, which should be mainly caused by a rough surface and small grain size. High efficiency with little hysteresis suggests that charges are extracted efficiently before recombination and that the trap density is low, which should also benefit device stability.

Figure 2 displays the effect of BQ on the degradation of η under continuous light irradiation using different light sources. Most reports of device stability in the literature are for devices containing mesoporous and/or compact TiO_2 layer, and UV-filtered solar light or white light-emitting diodes (WLEDs), which do not emit UV light, are often used as the light source to avoid UV-induced oxygen degassing and material degradation and obtain better long-term stability. We

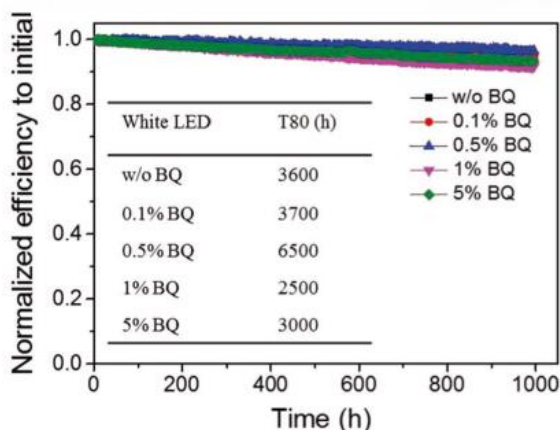


Fig. 2. Evolution of normalized η of PSCs with and without BQ additive under continuous irradiation from WLEDs with a light intensity of 100 mW cm^{-2} [2].

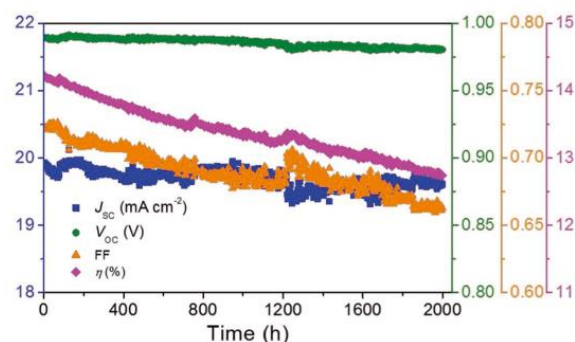


Fig. 3. Evolution curves of J_{SC} , V_{OC} , FF, and η for the device containing 0.5% BQ with the longest lifetime under continuous one sun solar irradiation (100 mW cm^{-2} , AM1.5G) without a UV cut filter at open-circuit conditions [2].

firstly examined the stability under WLEDs with a light intensity of 100 mW cm^{-2} . As shown in Fig. 2, our PSCs demonstrated excellent lifetimes that are extended with the addition of 0.1% and 0.5% BQ. The linearly extrapolated time until η reaches 80% of the initial value (T80) was 6500 h for the PSC with 0.5% BQ, which is much longer than 3600 h for our PSCs without BQ and published results. The T80 was shorter for larger BQ concentrations of 1% and 5%. Results of thermally stimulated current measurement, time-of-flight secondary ion mass spectrometry, X-ray photoelectron spectroscopy revealed that the improved stability arises from the fact that the formation of metallic lead, a source of carrier traps, during the continuous solar irradiation is suppressed.

The evolution of the solar cell parameters for the 0.5% BQ device with the longest lifetime as a function of exposure to light (ISOS-L-1 Laboratory) is shown in Figure 3. After 2000 h under solar irradiation at 100 mW cm^{-2} without a UV cut filter, η (12.74%) was still 90% of its initial value (14.21%). By fitting the time-dependent η with a straight line, T80 was estimated to be 4000 h. Note that the main decrease of η originates from a decrease of FF, which suggests that the efficiency and stability could be further improved by systematic interface engineering.

In conclusion, we have demonstrated that BQ addition into a precursor solution for the fabrication of perovskite films increases conversion efficiency through improved perovskite morphology and crystal quality while also significantly extending device lifetime by reducing the formation of carrier traps during solar irradiation. Another important finding in our study is that the carrier traps originate from metallic lead formed during continuous light irradiation. We believe that the present findings offer insight to help obtain efficient, stable organic-inorganic hybrid perovskite solar cells for future applications.

References

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