



# Effects of halogen replacement on the efficiency of luminescent solar concentrator based on methylammonium lead halide perovskite

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## ABSTRACT

Recently, Organic–inorganic hybrid perovskite compounds are at the forefront of research, due to their unique and exciting optical properties, besides the potential applications in photovoltaic systems. Possessing broad absorption spectrum, excellent photoluminescence quantum yields, large Stokes shifts between their absorption and emission bands and thus low self-absorption loss, high stability under illumination and oxygen exposure and tunable emission spectra, through halogen substitution, are advantages of organic–inorganic hybrid perovskites as active media in luminescent solar concentrators (LSCs). In this work, Methylammonium Lead Halide Perovskites ( $\text{CH}_3\text{NH}_3\text{PbX}_3$ , X = Cl, Br and I), are investigated as the active medium in planar and bulk form of LSCs. Using spectroscopic and photovoltaic measurements, variation of optical properties and device performance, with different halogen sources in the perovskite based LSC precursors, are monitored. Their superior performance is attributed to the high quantum yield of the Halide Perovskites. The obtained results reveal that to find a suitable condition for effective LSC, it is necessary to investigate the appropriate halogen atom in the synthesized perovskite, as well as the concentration of halide perovskite in LSC. It was found that  $\text{CH}_3\text{NH}_3\text{PbCl}_3$  perovskite, with 2.0 wt% perovskite concentration in Polyvinyl Alcohol (PVA), leads to the highest efficiency, with the least dissipative effect in LSC. The efficiency of coated Si PV cell is increased about 90% more, compared to an uncoated cell.

## 1. Introduction

Solar power is the most abundant energy resource on planet Earth. In this regard, investigation of Photovoltaic (PV) devices, has become a subject of intense research over the last years [1–4]. Achieving cost effective, easily processable and efficient solar cells have been a challenge for the scientific community. For photovoltaic energy to become competitive with fossil fuels, it is necessary to reduce the total cost of solar energy or to increase their power conversion efficiencies. One potential solution is the luminescent solar concentrators (LSCs), which can overcome that obstacles by photon-managing devices [5,6]. Luminescent solar concentrator (LSC) is a transparent piece of polymer-based device that has a fluorophore embedded or painted on it. In summary, through the Luminescent Down-Shifting (LDS) process of fluorophore, the sunlight of relatively short wavelength (such as ultraviolet UV) can be shifted to long wavelength light, where the absorption by silicon is optimal [7]. The reemitted light can be concentrated and in the following, can be propagated, through total internal reflection, to the edge of the sheet where the light is absorbed

by a narrow solar cell. Therefore, solar cells attached to LSC can generate more electric power than a conventional solar cell [8–11]. This is a promising technology, since it allows a large collecting area of virtually transparent polymer with a relatively small area of expensive solar cells. Focusing our attention into the case of the Si PV cell that its efficiency is close to the theoretical maximum established by the Shockley–Queisser limit of 32% [12]. One of the major factors limiting this efficiency is the mismatch between the solar spectrum and the Si band gap. One way to cope with the mentioned problems is through the usage of luminescent solar concentrators. There is currently huge invested interest in this technology due to the availability of perfect fluorophore materials. Main problems to be overcome are: limited stability of the luminescent species, low emission quantum efficiency, high self-absorption and poor knowledge of the parameters governing the efficiency [13]. The luminescent materials proposed during the years, as possible optically active centers, in the emissive layer of LSCs can be grouped in three main categories: organic dyes [14–16], quantum dots (QDs) [17,18], transition metals and lanthanide complexes [19,20]. Organic dyes and quantum dots, beside high

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fluorescence quantum yield (FQY), have tunable and wide absorption range and have been thoroughly examined. However, organic dyes and quantum dots undergo photobleaching and self-absorption, caused by the overlapping between their absorption and emission bands [21,22]. Compared with organic dyes and quantum dots, rare earth materials feature large Stokes shifts between their absorption and emission bands and are very promising for applications in LSCs. Besides the benefits, the drawback of lanthanide complexes, that limits their application as LSC centers, is the narrow absorption range. Most lanthanide complexes have absorption only within the UV region. Thus, extending the absorption range is the key problem that needs to be solved [23]. Optical efficiency of 40% has been achieved using a mixture of Lumogen F Red 305 dye in polymer, and power conversion efficiency of 7.1% has been reported using a combination of dyes with GaAs solar cells attached at the edges [24,25]. Higher power conversion efficiency of 14.5% has been achieved using a tandem planar solar concentrator, employing a mixture of dyes in combination with copper indium gallium selenide cells [26]. Bergren et al. demonstrate a record optical efficiency of 8.1%, for a device that transmits ~ 44% visible light by incorporating CuInS<sub>2</sub>/ZnS quantum dots with high quantum yield (> 90%), NIR-emitting, into the polymer interlayer between two sheets of low-iron float glass [27]. Tummeltshammer et al. present fluorophore-doped and flexible LSCs made of Pyrromethene 567 (P567) doped PDMS with a thickness of several millimeters [28]. They also demonstrated that the efficiency remains high while bending the LSC which is essential for flexible LSCs to make an impact on solar energy. Portnoi et al. experimentally verify a model for the optimization of the external optical efficiency of LSCs under varying degrees of curvature [29]. They demonstrate differences between the optimization of flat and bent LSCs and show that optimal fluorophore concentrations can differ by a factor of two. Recently, Nikolaidou et al. investigate the viability of organic–inorganic hybrid perovskite (PVSK) thin films as the active medium in planar LSCs [30]. The results display high optical efficiency in the range 15–29% despite high self-absorption losses, and the devices remain operational even after seven weeks in ambient conditions.

Compared to other types of fluorophores, a new class of phosphors used in luminescent solar concentrators is organic–inorganic perovskite structure, which was introduced in our previous work [7]. Such new halide perovskite structures emit light in the wavelength range of 500–700 nm, which can be efficiently absorbed by solar cells [31,32]. Introduction of such halide perovskite structures gives an enormous flexibility in LSCs' design, allowing enhancement of performance through Down-Shifting. This material combines the excellent chemical stability of inorganic portions with the unique luminescent features of organic ions, such as; (1) a broad absorption bandwidth and efficient tuning of the absorption spectra: to utilize the solar spectrum efficiently, (2) large Stokes shift: to minimize the overlapping of absorption and emission spectra and consequently reduction of self-absorption losses, (3) tuning of the emission spectra through halogen substitution in perovskite structure, as well as high fluorescent quantum efficiency and (4) high stability under illumination and oxygen exposure [33–35]. These features are particularly desirable for producing efficient and durable LSCs. In our previous article, CH<sub>3</sub>NH<sub>3</sub>PbBr<sub>3</sub> perovskite thin films were used to coat a silicon solar cell, to utilize its high absorption and quantum yield in order to improve the solar cell's performance [7]. Here we present the results of two parallel investigations. The first is a study of the hybrid perovskite films in a planar luminescent sheet, placed on top of PV cells. The second is study of the bulk form of traditional LSC configuration with silicon PV cell attached at the edge. We focus on two main aspects in our study; first is optimizing LSC performance, achieved by varying the concentration of hybrid perovskite in a Polyvinyl Alcohol (PVA) solution. Another fundamental issue to be considered is the band gap changes from halide substitution are influenced more by the electronic states of the anion, that is, from Cl to Br to I, the valence band composition changes from 3p to 4–5p, respectively, with a monotonic decrease in the electron binding energy. Substitution

of X = Cl by Br and I in CH<sub>3</sub>NH<sub>3</sub>PbX<sub>3</sub> has been shown to decrease the band gap from 3 to 2.3–1.6 eV, respectively [36]. From this point of view, organic–inorganic lead halide perovskites, with tunable band gaps, are clearly promising systems for down-conversion process in LSCs. The solar spectrum received at Earth's surface covers a broad range of wavelengths from 290 nm to 3790 nm. However, the absorption band of the best PV materials can be found between 400 and 1200 nm, depending on the case. Obviously, there is a large mismatch between the solar spectrum and the absorption properties of the present PV materials. Therefore, much of the available solar energy is not being used, due to this problem. The luminescent solar concentrators based on down-converter lead halide perovskites with a tunable bandgap permit the conversion of the whole UV light region of the solar spectrum (290–400 nm) to a suitable radiation region (visible range) for commercial PV modules (Si, a-Si and GaInP) and those under development (organic solar cells).

In this paper, UV light sensitized CH<sub>3</sub>NH<sub>3</sub>PbX<sub>3</sub>, X = Cl, Br and I, organic–inorganic hybrid perovskites were prepared. Planar luminescent sheet in polymeric matrix was placed on top of PV cells and the bulk form of traditional LSC configuration, with silicon PV cell attached at the edge, to construct the LSCs. Hence, this paper focuses on the optimization and performance characterization of LSCs for use in this system.

## 2. Experimental

### 2.1. Preparation of Materials and LSCs

Lead halide perovskites precursor of different halide sources (CH<sub>3</sub>NH<sub>3</sub>PbX<sub>3</sub>, X = Cl, Br and I) with the proportions of CH<sub>3</sub>NH<sub>2</sub>/PbX<sub>2</sub>: 6/1 M ratio, are prepared by solution chemistry method. In our previous work, we found that CH<sub>3</sub>NH<sub>3</sub>PbBr<sub>3</sub> hybrid with 6/1 M ratio is more effective than others. The experimental details of hybrid preparation have been reported elsewhere [37].

In brief, CH<sub>3</sub>NH<sub>3</sub>X (X = Cl, Br and I) was first prepared by reaction of Methylamine (CH<sub>3</sub>NH<sub>2</sub>, 40% solution in water, merck) with a stoichiometric amount of hydrochloric acid (HCl, 37%, merck) - or hydrobromic acid (HBr, 48%, merck) or hydroiodic (HI, 57%, merck) - aqueous solution in a chilled glass bottle, at 0 °C to remove the reaction temperature. Then, stoichiometric amount of CH<sub>3</sub>NH<sub>3</sub>X was added to the lead halide salt (PbCl<sub>2</sub>, PbBr<sub>2</sub> or PbI<sub>2</sub>, sigma Aldrich). All materials were used without any further purification. Each product was air dried, at room temperature, on a watch glass and then dried at 60 °C for a week, and organic–inorganic lead halide perovskites are formed. In the next step, luminescent solar concentrators were fabricated by dispersing CH<sub>3</sub>NH<sub>3</sub>PbX<sub>3</sub> particles in a Polyvinyl Alcohol (PVA, merck, hydrolysis degree: 87–89%) waveguide, in two forms; Planar luminescent sheet on top of PV cells and bulk form of traditional LSC configuration with silicon PV cell attached at the edge. Schematic diagram of devices are shown in Fig. 1(a), (b). To investigate the effect of the concentration of the luminescence material on the application of the solar cell in the planar luminescence solar concentrator (device A), as shown in Fig. 1(a), the synthesized CH<sub>3</sub>NH<sub>3</sub>PbBr<sub>3</sub> hybrids, was added to the polymer solution as the luminescence material. To prepare a 30% solution of PVA in water, first 4 ml distilled water was put in a beaker and after putting it on hotplate, 1.2 g of PVA polymer was slowly added to the water in order to avoid aggregation. To quicken the solvation of polymer in water, one can increase the temperature up to 65 °C. After 2 h, a clear solution is prepared. Afterwards, the organic–inorganic CH<sub>3</sub>NH<sub>3</sub>PbBr<sub>3</sub> hybrids, were added to the polymer solution as the luminescence material. Weight ratio of the luminescence in the host polymer was chosen as 1.11 wt%, 1.33 wt%, 1.55 wt%, 1.77 wt% and 2.0 wt%. Each one of these solutions, after the evaporation of the solvent, were placed on the 35 × 15 mm commercial silicon solar cell, as the planar luminescent solar concentrator (device A) with the dimensions of 35 × 15 × 0.5 mm. Also, to fabricate the bulk form of

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