CEST 2023

18th International Conference on Environmental Science and Technology

Athens, Greece, 30 August to 2 September 2023

Interface engineering approaches for efficient and robust perovskite solar cells

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Abstract The field of perovskite photovoltaics has been witnessing a surge of interest over the past few years across the breadth of advanced nanomaterials, nanoscience and nanotechnology. Intensive research activity focuses on the development of hybrid perovskite absorbers with controlled nanomorphology where we can modify intrinsically different properties (crystallinity, defects, grain boundaries) and optimize charge transport in the bulk structures at the corresponding interfaces. The perovskite absorber and its interfaces with the electron transport layer (ETL) and the hole transporting material (HTM) play a pivotal role in obtaining perovskite solar cells (PSCs) with high power conversion efficiency (PCE) and enhanced stability. This contribution deals with advanced engineering strategies developed by our group focusing on the optimization of perovskite interfaces to regulate the geometric, structural and electronic properties of the solar cell basic components.

Keywords: Perovskite solar cells; interface engineering; water resistance; efficiency; stability.

1. Introduction

During the last decade the discovery of perovskite solar cells (PSCs) revolutionized the field of third generation photovoltaics with significant progress in materials science and engineering which led to the development of innovative devices outperforming the silicon based alternatives. Despite the spectacular increase in power conversion efficiencies (PCEs) where certified values exceeding 25% were achieved, a number of key issues prevent the industrialization and commercialization of the new technology. The perovskite degradation under harsh environmental conditions (humidity, oxygen, UV light) remains one of the main concerns, without neglecting the negative impact of hysteresis phenomena and lead toxicity.

2. Materials and Methods

$APbX_3$ lead-based perovskites [A = Cs, CH_3NH_3-MA, NH_2CHNH_2-FA, $(CH_3)_3S$, HMIm and X = Cl, Br, I] were prepared by reacting equimolecular quantities (1 mol/L) of lead(II) halides with the corresponding organic halide salts in DMSO (in anhydrous conditions and in the absence of oxygen) under vigorous stirring for 1 h at 60 °C and their structural and optoelectronic properties were characterized by XRD, Raman and UV-Vis (Niemann et al., 2016; Kaltzoglou et al., 2018; Kontos et al., 2020). Using the corresponding tin halide precursors and similar controlled conditions, synthetic approaches and characterization techniques, a series of tin-based $ASnX_3$ and A_2SnX_6 perovskite materials were also prepared and characterized (Bounos et al., 2020). The tin-based defect perovskites were incorporated as HTMs in dye-sensitized solar cells (DSSCs) of the type: FTO/TiO_2-Dye/Perovskite/Pt/FTO (Elsenety, Antoniadou, Kaltzoglou et al., 2020). In addition, the perovskites were used as absorbers and interlayers in PSCs of the following architecture: FTO/ETL//Perovskite//HTM/Ag (Balis et al., 2018; Zaky et al., 2021; Elsenety et al., 2023). The solar cells were characterized under simulated solar light (1 sun, 100 W cm^{-2}, AM 1.5G) from a Xenon 300 W source, analyzing the current density-voltage (J-V) characteristics recorded using linear sweep voltammetry on the Autolab PGSTAT-30 potentiostat.

3. Results and discussion

Adopting improved processing protocols, P. Falaras et *al.* have synthesized (in solution and/or solid state reacting conditions) a series of novel lead (ABX_3:[$CH_3NH_3PbI_{3-x}Cl_x$, $CH_3NH_3PbX_3$ (X = Cl, Br, I), $FAPbI_3$, $(CH_3)_3SPbI_3$, $(CH_3)_3SPbI_{3-x}Br_x$ and $(CH_3)_3SPbI_{3-x}Cl_x$ (x = 1,2,3), $CsMAFAPbI_{3-x}Br_x$, $CsMAFAPbI_{3-x}Br_x/(CH_3)_3SPbI_3$, $(NH_2CHNH_2)PbX_3$ (X = Cl, Br, I), $FAPbBr_3$, $(HMIm)PbI_3$ and $CsMAFAPbI_{3-x}Br_x/(HMIm)PbI_3$]) (Niemann et al., 2016; Kaltzoglou et al., 2017; Kaltzoglou et al., 2018; Kontos et al., 2020) and tin-based (ABX_3 and A_2BX_6: [$Cs_2SnI_3Br_3$, Cs_2SnX_6(X = Cl, Br, I), $CsSnBr_3$, Cs_2SnI_6, $CsSnI_3$, Cs_2SnX_6 (X = Cl, Br, I), $((CH_3)_3S)_2SnX_6$ (X = Cl, Br, I), $ASnI_3$ (A = Cs^+, $CH_3NH_3^+$ and NH_2-CH=NH_2^+), $(CH_3)_3SSnI_3$, $((CH_3)_3S)_2SnI_{6-n}Cln$ and $((CH_3)_3S)_2SnI_{6-n}Br_n$ (n=1, 2), $(CH_3)_3SSnCl_3$]) (Bounos et al., 2020; Kontos et al., 2018; Belessiotis et al., 2021) perovskite materials with different chemical composition, symmetry and dimensionality (0D, 1D and 3D) that meet fabrication processing and environmental requirements for applications in solar cells. The materials were mainly prepared using appropriate precursors of low toxicity and

their structural and optoelectronic properties were thoroughly characterized by powder X-ray diffraction with Rietveld refinement, UV−vis diffuse reflectance, photoluminescence spectroscopies, multitemperature Raman spectroscopy and differential scanning calorimetry. Advanced characterization was also performed under harsh conditions including the investigation of temperature and pressure effects on structural evolution and vibrational properties. These perovskites were used as main components (active light absorbers, additives, interlayers and hole transporters) in a number of device architectures developed at the laboratory.

The energy level alignment at the interfaces of solar cells is an essential factor that determines their efficiency and stability. An interface capable of appropriate energy band bending, minimal defects and good contacts is the key issue to obtain highly performing and stable solar energy conversion devices. It has been demonstrated that the tin-based (Sn^{4+}) defect perovskites present high structural stability and dispose exceptional optoelectronic properties. Thus, the M_2SnX_6 defect perovskite materials have been incorporated with in solid-state dye-sensitized solar cells and the obtained results confirmed that they can effectively act as hole transporters (Kaltzoglou et al., 2015; Kalzoglou, Antoniadou, Kontos et al., 2016; Kaltzoglou, Perganti et al. 2016; Elsenety et al., 2018; Elsenety, Antoniadou, Kaltzoglou et al., 2020).

In addition, a number of engineering strategies involve the incorporation of multipurpose materials (dyes, squaraines, thiols, polymers, 1D perovskites, transition metals, rGO and g-C_3N_4] between the perovskite and the electron transport layer and the hole transporter in PSCs (Figure 1). These functional compounds contain active groups and characteristics that permit the development of inorganic or organic interlayers which are able to adjust the energy levels at the corresponding interfaces. Dye sensitization of the electron transport layer, perovskite passivation and dimensionality engineering employing ID $(CH_3)_3SPbI_3$ and $(HMIm)PbI_3$ perovskites on top of the main 3D absorber have been demonstrated to be the most effective strategies for this purpose (Balis et al., 2018; Zaky et al., 2021; Elsenety, Antoniadou, Balis et al., 2020; He et al., 2021; Niu et al., 2022; Elsenety et al., 2023).

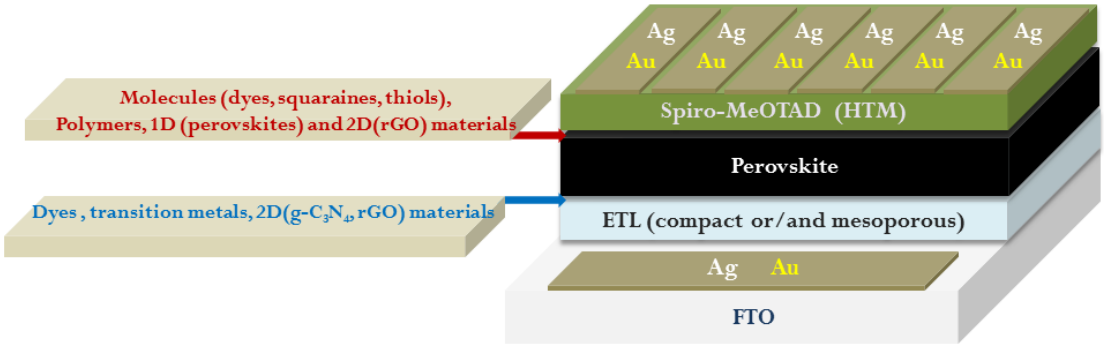

Figure 1. Engineering strategies involving the incorporation of multipurpose modifiers in PSCs

The adoption of such advanced engineering approaches leads to improved crystallization of the perovskite which is at the origin of increased short circuit photocurrent. The improvement of fill factor and open circuit potential can be attributed to the passivation of the perovskite absorber. The presence of interlayers affects the perovskite growth and reduces its crystallization rate, leading to large grains with low number of defects and high crystallinity. In addition, the extraction of electrons and holes at the corresponding interfaces was enhanced, their recombination was suppressed and the power conversion efficiency of the PSCs was increased. Moreover, the modified interfaces have an ultra-hydrophobic character, which protects the perovskite from moisture and leads to excellent device stability. Such innovative interface engineering approaches provide important guidance for further development of functional interlayers incorporated into efficient and robust perovskite modules and panels. toxicity.

4. Conclusions

Novel synthetic approaches led to the development of innovative perovskite (lead-based and lead-free) materials with exceptional structural and optoelectronic properties that make them ideal candidates for application in photovoltaic technologies. Dye sensitization, defect passivation and dimensionality engineering are among the most promising strategies that have been successfully employed to fine tune and optimize the ETL/absorber and absorber/HTM interfaces, leading to the development of robust devices retaining high efficiency and enhanced stability after accelerating aging in harsh environmental conditions. toxicity.

Acknowledgements: The author acknowledges funding of this work by European Union's Horizon 2020 Marie Curie Innovative Training Network 764787 "MAESTRO" project and additional support by the Hellenic Foundation for Research and Innovation (H.F.R.I.) under the "2nd Call for H.F.R.I. Research Projects (PVLumo-KA 80635).

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