



Enhanced performance of mesostructured perovskite solar cells in ambient conditions with a composite $\text{TiO}_2\text{--In}_2\text{O}_3$ electron transport layer



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ABSTRACT

Perovskite solar cells (PSCs) with a composite mesoporous $\text{TiO}_2\text{--In}_2\text{O}_3$ electron transport layer were constructed and tested. For the formation of the In_2O_3 overlayer three different molar concentrations of the precursor indium(III) acetylacetonate were investigated ($\text{TiO}_2\text{--In}_2\text{O}_3$ (1) 46 mM, $\text{TiO}_2\text{--In}_2\text{O}_3$ (2) 50 mM and $\text{TiO}_2\text{--In}_2\text{O}_3$ (3) 56 mM). In_2O_3 was chosen because of its slightly higher conduction band placement compared with TiO_2 which could facilitate the charge transport of the photogenerated electrons from the conduction band edge of the perovskite to the TiO_2 , reducing the recombination rate. The electron transport films were characterized structurally and morphologically, while the electrical properties of the PSCs with the $\text{TiO}_2\text{--In}_2\text{O}_3$ bilayer were examined in detail and compared against cells with pristine TiO_2 . All layers were deposited by the spin coating method and at ambient conditions of temperature and humidity. The structure that was used for the mesostructured PSCs was FTO/c- TiO_2 /mp- TiO_2 /(In_2O_3)/ $\text{CH}_3\text{NH}_3\text{I}_{3-x}\text{Cl}_x$ /P3HT/Au. The optimum device performance was observed for PSCs with the $\text{TiO}_2\text{--In}_2\text{O}_3$ (2) electron layer, having an improved short-circuit current density and open-circuit voltage, with an overall power conversion efficiency of almost 12.9%.

1. Introduction

Perovskites are compounds employed in a wide range of applications; specifically in photovoltaic applications, they are used as light absorbers in one of the emerging third generation photovoltaics known as perovskite solar cells (PSCs). They are direct bandgap semiconductors displaying unique properties, such as fascinating optical absorption from the visible to the near infrared (NIR) region, ambipolar carrier transport, long electron–hole diffusion lengths and high dielectric constant leading to low exciton binding energy [1–4]. Typical solution based methods used for the deposition of the perovskite film are the one-step deposition or the two-step sequential deposition [5,6]. Alternatively, vapor based deposition techniques can also be applied for uniform perovskite films [7].

PSCs can be easily fabricated through simple solution processes following either the planar (thin film) cell configuration or the mesoscopic (sensitized) cell structure, where the perovskite is confined between the electron transport layer (ETL) and the hole transport layer

(HTL). In the planar structure, if the incident light reaches first the ETL the corresponding cell has the regular n-i-p structure, while if it reaches the HTL the cell has the inverted p-i-n structure [7–9]. However, both planar and mesoscopic configurations are faced with some challenges; although planar PSCs usually have lower power conversion efficiencies compared with their mesoscopic counterparts [10], the latter require metal oxide films with high mesoporosity while the former consist of compact layers. Results have been presented showing moderate or even negligible hysteresis for both structures [10,11]. A vital component employed in PSCs is the ETL; several ETLs have been tested such as TiO_2 , SnO_2 , ZnO and even Al_2O_3 scaffolds [12–14]. Due to its properties TiO_2 has been extensively used in photocatalysis, dye-sensitized solar cells (DSSCs) and PSCs [15,16]. However, its film formation requires sintering at high temperature which increases the cost and confines its use on only rigid substrates, limiting the PSCs' applications. SnO_2 is also a wide band-gap semiconductor with exceptional optical and electrical properties, presenting a perfect band alignment at the SnO_2 /perovskite interface [17]. It is well

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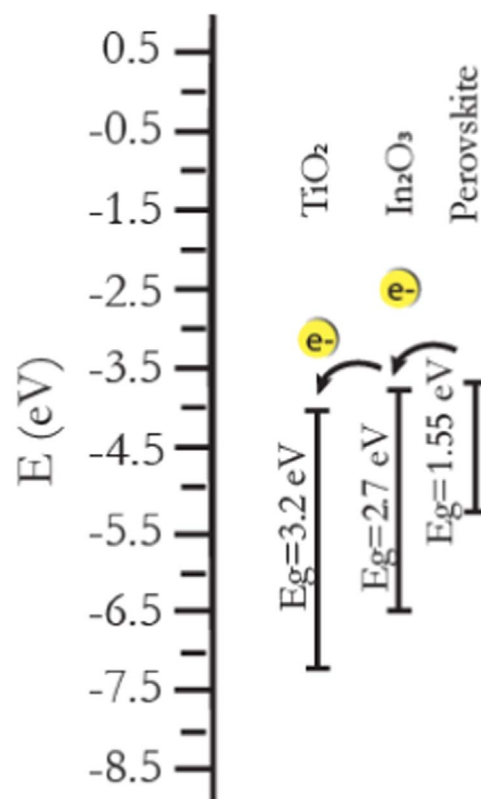
known that ZnO has two orders of magnitude higher conductivity than TiO₂ favoring the electron mobility and that ZnO films can be grown with low-temperature processes enabling the use of flexible substrates [18,19]. Nevertheless, PSCs employing ZnO as electron transport layer are unstable, forcing the researchers to use other electron conductors [20,21]. Notably, it has been established that PSCs with insulating Al₂O₃ as a scaffold over a compact TiO₂, where the perovskite film acts as both a light absorber and an electron transport media, exhibit faster charge collection and higher open-circuit voltages [14].

Organic–inorganic halide perovskites comprise the general formula ABX₃, where A is a cation, usually CH₃NH₃⁺, CH(NH₂)₂⁺, Cs⁺ or a mixture of them, B is divalent or trivalent metal, usually Pb⁺², Sn⁺², Bi⁺³, Sb⁺³ and X is a halogen anion, usually I⁻, Br⁻, Cl⁻ or a combination of them [22–27]. Combining different halogen elements at the perovskite has manifold advantages at its properties by increasing the stability and the electron–hole diffusion length and by tuning the band gap for broad light absorption and improved open-circuit voltages [1,3,14,28]. The highest overall power conversion efficiency reached so far for PSCs is 22.1%, which was achieved by focusing on the fabrication of an extremely uniform perovskite layer [29].

Traditionally, HTMs are used for extracting the holes created at the perovskite layer to the cathode. While perovskites can effectively transport electrons and holes, which makes the use of HTMs optional, PSCs without HTMs are exhibiting lower power conversion efficiencies (PCEs) than devices employing a hole transport layer (HTL) [30,31]. Although, spiro-MeOTAD (N²,N²,N²,N²,N⁷,N⁷,N⁷,N⁷-octakis(4-methoxyphenyl)-9,9'-spirobi[9 H-fluorene]-2,2',7,7'-tetramine), PTAA (Poly[bis(4-phenyl)(2,4,6-trimethylphenyl) amine]) and P3HT (Poly(3-hexylthiophene-2,5-diyl) regioregular) are three of the frequently employed HTMs, additives are regularly employed in their solutions (lithium Bis(trifluoromethane)sulfonimide) (Li-TFSI)- and 4-tert butylpyridine (4-TBP), which increases even more their already high cost [10,32,33]. Researchers have also investigated the use of alternative low cost HTMs, such as copper phthalocyanine, copper iodide, copper thiocyanate and nickel oxide, but the PSCs displayed lower efficiencies [34–37].

One of the main factors reducing the overall efficiency of the PSCs is the recombination of the electrons generated at the perovskite with the holes. Doping the ETL through the incorporation of a metal ion, such as Nb⁵⁺, Y³⁺, In²⁺, can alter its electrical properties by moving the conduction band (CB) of the semiconductor and limiting the recombination and energy losses [38–40]. Alternatively, combining two semiconductors as ETLs with appropriate CB edges can provide a better energy level alignment with the CB of the perovskite, facilitating the electron transport, minimizing the recombination rate and consequently improving the power conversion efficiency of the PSC. At first, composite semiconductors have been introduced in dye-sensitized solar cells to reduce the recombination processes [41,42]. In perovskite solar cells, double electron transport layers have been used to boost the cells' performance mainly by enhancing the open-circuit potential. In particular, a complex mesoporous layer of TiO₂/SrTiO₃ or BaTiO₃ has been employed, which in both cases improved the PSCs' overall efficiency [43,44].

In this paper for the first time, atop of the mesoporous TiO₂ layers of the mesoscopic PSCs, an indium oxide layer is deposited prior to the mixed halide perovskite (CH₃NH₃I_{3-x}Cl_x), for aligning the energy levels. In₂O₃ is a semiconductor with an indirect band gap of 2.7 eV and a slightly higher conduction band placement than TiO₂, leading to a composite ETL for efficiently transferring the photogenerated electrons (Scheme 1). The composite inorganic films were characterized with microscopy techniques and porosimetry to examine their structural properties, X-ray diffraction measurements to certify the crystallinity of both oxides, while the electrical properties of the PSCs were examined in detail.



Scheme 1. Energy level diagram.

2. Experimental methods

2.1. Materials

Titanium diisopropoxide bis(acetylacetonate) (75 wt% in isopropanol, Aldrich), titanium(IV) butoxide (97%, Aldrich), glacial acetic acid (99.8+%, Chem-Lab), pluronic P123 (5.800 g/mol, Aldrich), indium(III) acetylacetonate (Aldrich), triton X-100 (646.86 g/mol, Fisher Scientific), hexamethylenetetramine (HMT, ≥99%, Aldrich), hydroiodic acid (55%, Aldrich), anhydrous dimethylformamide (DMF, 0.03% water, Carlo Erba reagents), lead(II) chloride (Acros organics), chlorobenzene (>99.5%, Sds votre partenaire chimie), regioregular poly(3-hexylthiophene-2,5-diyl) (P3HT, 95.5%, 94.000 g/mol, Ossila), lithium Bis(trifluoromethane)sulfonimide (Li-TFSI, 99.95%, Aldrich), 4-tert butylpyridine (4-TBP, 96% Aldrich) and all solvents were used as received. Gold wire was sublimated to form the cathode.

2.2. Fabrication of the perovskite solar cells

Fluorine-doped tin oxide (FTO) glasses, with a sheet resistance of 8 Ω/□ (Pilkington), were used as substrates for the fabrication of the monolithic mesoscopic PSCs. A part of the FTO glasses was chemically etched with zinc powder and HCl aqueous solution, in order to prevent any short-circuit phenomenon. Afterwards, the FTO glasses were cleaned with detergent, deionized water and acetone. The TiO₂ solution for the formation of the compact layer (c-TiO₂), which consisted of titanium diisopropoxide bis(acetylacetonate) diluted in isopropanol (10% v/v), was deposited on the FTO glass by spin-coating at 2000 rpm for 10 s. The mesoporous TiO₂ layers (mp-TiO₂) were formed by spin-coating (1200 rpm for 20 s) a solution that was previously prepared by dissolving 0.37 mL Titanium(IV) butoxide, 0.4 mL glacial acetic acid and 0.5g Pluronic P123 in 4 mL isopropanol. The procedure was repeated until the desired thickness was obtained.

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