



Improving the stability of inverted perovskite solar cells under ambient conditions with graphene-based inorganic charge transporting layers



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ARTICLE INFO

Article history:

Received 28 July 2017

Received in revised form

19 September 2017

Accepted 5 October 2017

Available online 5 October 2017

Keywords:

Inverted perovskite solar cells

Li-modified graphene oxide

Graphene oxide

Stability

ABSTRACT

Inverted organometal halide perovskite solar cells of p-i-n architecture allow for the employment of inorganic components that ensure longer time stability than organic charge transporters. This has been demonstrated in the present work where devices were made by employing NiO/GO and Li-modified GO/TiO_x as hole and electron transporters, respectively, in comparison with popular organic components, such as PEDOT:PSS and PCBM. Cells made in the FTO/PEDOT:PSS/Perovskite/PCBM/Al composition were 25% more efficient than cells made in the FTO/NiO/GO/Perovskite/GO-Li/TiO_x/Al composition but the latter was markedly more stable than the former. Emphasis has been presently given to the characterization of the inorganic components, which can thus be employed for the construction of solar cells under ambient conditions with descent 11.2% efficiency. The GO/GO-Li based perovskite solar cell devices showed reasonable stabilized efficiency, high reproducibility and a negligible hysteresis effect.

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1. Introduction

Hybrid organic-inorganic perovskite materials have followed a remarkable route that marked a very popular research field in solar energy conversion. These materials are the main components of perovskite-based solar cells (PSC). They derive their popularity from their strong light harvesting, high electron-hole diffusion lengths [1,2], high charge carrier mobility [3,4], tunable wide band gap range [5] and simple and low-cost fabrication processes [6,7]. PSC is a very young technology but has already been placed at the forefront of solar cell technologies by providing an astounding power conversion efficiency (PCE) of 22.1% [8], which enables commercialization. The standard configuration of PSC is based on an n-i-p architecture, which originated from dye-sensitized solar cells. However, since organic-inorganic hybrid perovskite materials are able to transport electrons and holes themselves with high diffusion lengths [1,2], it encouraged researchers to search for alternative configurations and this lead to inverted devices with p-

i-n architecture. Inverted PSC have some advantages including low temperature processing, easy-fabrication, high throughput and cost-effectiveness for scaling up to large production levels [9]. Organic PSC bearing PEDOT:PSS as hole transporting material (HTM) and PCBM as electron transporting material (ETM) are the most popular inverted PSC [9–12]. PEDOT:PSS, an organic conducting polymer blend, has some advantages as HTM, such as tunable conductivity and high optical transmittance in the visible light wavelength range [13]. However, because of its acidic nature, PEDOT:PSS may corrode both underlying conductive layer and perovskite layer on the top [14]. On the other hand, the major challenge encountered by PCBM as effective ETM is the tradeoff between film thickness and full coverage of the rough perovskite film. ETL should be continuous and thin enough to act as efficient electron transporter, suppress charge recombination and diminish serial resistance. However, thin PCBM films may not be able to cover completely the rough surface of perovskite film due to the low viscosity of its solution and its small-molecule nature [15]. In addition to those problems, the major concern about organic components under illumination is instability in the ambient-air and this makes a drawback for PEDOT:PSS and PCBM as charge transporters and for organic-inorganic hybrid perovskite materials as photo-active layer, for realistic applications [16]. This becomes

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even worse in long-term operation of inverted PSC devices since oxygen and moisture can induce oxidation of the organic compounds, under ambient conditions [17–19]. Degradation of the charge transporting layers has also previously been assigned to their de-doping under illumination [20,21]. As alternatives to the organic components, charge transporters based on inorganic materials are attracting substantial attention, thanks to their better stability [9]. Among such alternative components, carbonaceous materials seem to be well fit to this case. Indeed, moisture has been identified as the major cause of irreversible degradation for perovskite films and the related devices [22,23], due to the presence of the weak Pb–I ionic bonds and the volatile component in perovskite lattice structure [24,25]. Carbon based components may then discourage humidity penetration, thanks to their hydrophobic nature. Interface engineering by employing carbon-based components, may further act as buffer layer to prevent metal-atom penetration into the perovskite underlying layer during ageing [26–28]. Herein based on our previous work [29], we present an interface engineering approach using graphene derivatives as inorganic compounds, applied directly on both lower and upper sides of the perovskite film. These layers could completely cover the perovskite film, leading to substantially seal it from outside humidity conditions. In addition, they protect perovskite from Al-atoms and other cell components which may migrate during their subsequent depositions on the top. In other words, perovskite layer sandwiched between graphene derivatives may be protected against moisture exposure and against harmful component migration. All-inorganic charge transporters of inverted PSC have in the present work been made by employing graphene oxide (GO) combined with NiO as HTL and Li-modified GO combined with TiO_x as ETL. This structure was similar to our previous work [29], with the introduction of an additional layer of NiO, which improved cell performance. In addition and with the purpose to compare the presently made PSC with the most popular inverted PSC, we have also studied cells bearing PEDOT:PSS as HTL and PCBM as ETL. Both types of PSC have been studied under ambient conditions to evaluate and compare their long term stability (see Table 1).

2. Experimental procedures

2.1. Materials

All chemicals were purchased from Sigma-Aldrich, unless otherwise specified, and they were used as received. SnO₂:F transparent conductive electrodes (FTO, Resistance 7 Ω/square) were purchased from Solaronix (TCO22-7). Full details of GO and GO-Li synthesis and TiO_x sol preparation have been reported in our previous works [29,30].

2.2. Solar cell devices construction

In the present work, inverted devices of PSC were constructed with two configurations: (1) FTO/PEDOT:PSS/Perovskite/PCBM/Al, and (2) FTO/NiO/GO/Perovskite/GO-Li/TiO_x/Al. Solar cell devices were constructed in the following sequence: etching and cleaning of FTO substrates; applying the HTL, perovskite photo-active layer and ETL; deposition of aluminum electrode. All of these steps

were the same for the two above configurations, except that the hole and electron transport layers were PEDOT:PSS and PCBM in the former configuration and NiO/GO and Li-GO/TiO_x in the latter configuration. In order to construct the FTO/PEDOT:PSS/Perovskite/PCBM/Al cells, FTO-coated glass substrates were cut in pieces of dimensions 1 cm × 3 cm. Part of the conductive layer was removed using zinc powder and hydrochloric acid. Then they were washed with mild detergent, rinsed several times with distilled water and subsequently with ethanol and acetone in an ultrasonic bath, finally dried under air stream. The patterned and cleaned FTO was washed with ethanol to make it more hydrophilic. HTL was applied using a filtered poly(3,4-ethylenedioxythiophene): poly(styrene sulfonic acid) (PEDOT:PSS)/methanol (1:3 vol:vol.) by spin coating at 3000 rpm for 60 s and dried at 150 °C for 20 min. Active perovskite layer was deposited on the thus prepared PEDOT:PSS film by the following procedure. A precursor solution was made by mixing 253 mg PbCl₂ and 507 mg PbI₂ with 270 mg methyl ammonium iodide in 1.5 mL dimethylsulfoxide (DMSO). The solution was kept under stirring for about 2 h at about 80 °C and then deposited by two consecutive spin-coating steps; first 1000 rpm for 10 s, then 6000 rpm for 30 s. During the second step, 1 mL chlorobenzene, as an anti-solvent, was gently dropped on the spinning substrate. The layer was then heated at 90 °C for about 45 min, which made the sample's color turn from yellow to dark-brown. In order to prepare ETL, 30 mg of [6,6]-phenyl C₆₁-butyric acid methyl ester (PCBM) was dissolved in 1 mL anhydrous chlorobenzene. A thin layer of PCBM was spin-coated on top of the cooled perovskite layer at 3000 rpm for 40 s. The last step for preparing the inverted devices of PSC was the deposition of about 90 nm-thick aluminum electrodes by thermal evaporation under vacuum. These unit devices had an active size of 15 mm² (10 mm × 1.5 mm) as defined by the size of the Al electrodes.

As mentioned earlier, solar cell devices construction procedure was exactly the same for both configurations, with differences in HTL and ETL preparation. In order to apply HTL in the FTO/NiO/GO/Perovskite/GO-Li/TiO_x/Al configuration, first NiO layer was deposited on FTO by spin coating of a NiO solution at 3000 rpm for 40 s followed by firing at 300 °C in air for 40 min. The NiO solution was prepared by diluting a homemade NiO paste with ethanol in a ratio of 1:5. In order to prepare the NiO paste, 1 g of NiO nanopowder was mixed with 30 mL anhydrous ethanol. 5 g of a stock solution of ethyl cellulose in ethanol (111 mg/mL) and 3 g of α -terpineol were successively added under stirring, then placed in an ultrasonic water bath for 30 min to ensure good dispersion. To increase paste viscosity and optimize its rheology, ethanol content was removed by a rotary evaporator. A thin layer of GO was then deposited by spin-coating its dispersion in isopropanol (5 mg/mL) at 2000 rpm for 60 s, followed by heating at 150 °C for 10 min. The deposition/heating process was repeated twice to attain a full-coverage by a uniform film. After deposition and annealing of the perovskite layer as above, GO-Li and TiO_x layers were spin-coated on the top. These layers were deposited from their stock solutions by spin-casting at 2000 rpm for 60s and 2000 rpm for 30 s, respectively, then dried at 100 °C for 10 min. The deposition/drying process of GO-Li layer was also repeated twice in order to fully cover the surface of the perovskite film.

Table 1

Maximum and average photovoltaic parameters of inverted PSC made by different charge-transporting materials in a batch of fifteen devices.

Inverted PSC device configuration	J _{SC} (mA/cm ²)	V _{OC} (V)	FF	η%
PEDOT:PSS/Perovskite/PCBM	22.2 (21.2)	0.89 (0.875)	0.71 (0.69)	14.0 (13.1)
NiO:GO/Perovskite/GO-Li:TiO _x	18.6 (17.7)	0.97 (0.96)	0.62 (0.61)	11.2 (10.4)

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