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An efficient and simple dual effect by under-layer abduction design for highly flexible NiO_x-based perovskite solar cells

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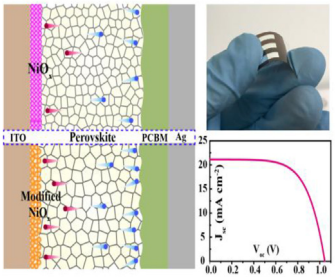
HIGHLIGHTS

- The modified NiO_x is synthesized with low temperature solution processed method.
- The composited HTL reveals an optimization of perovskite crystal nucleation and growth.
- The device exhibits more charge carrier generation and efficient transportation.
- Device based on modified NiO_x shows excellent stability than the pristine one.

GRAPHICAL ABSTRACT



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ABSTRACT

Organic-inorganic hybrid perovskite with large crystal size and high quality are desirable for making high-performance solar cell. However, the expectant film with good surface is normally limited by various factors, such as under-layer, film thickness. Here, an efficient dual effect method on the performance by means of under-layer abduction strategy is developed to improve perovskite crystallization by incorporating nitrogen-doped reduced graphene oxide into nickel oxide as hole transport layer, which significantly improves the electrical conductivity, and endows composite material with encouraging hydrophobic property. In addition, the modified under-layer enhances the space of crystal nucleation and facilitates crystal growth through grain boundary migration, thereby achieving large crystal size and reduced grain boundaries. Consequently, a best device with maximum power conversion efficiency of 18.84% is achieved due to a significant improvement in fill factor and short-circuit current density. Moreover, its flexible application shows an efficiency of 14.11%, retaining > 90% after aging for 1200 h without encapsulation. This work provides a potential platform to fabricate low cost and efficient hole extraction materials for large scale and flexible perovskite solar cell.

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1. Introduction

Since the innovative discovery of perovskite photovoltaics by Miyasaka and co-operation workers in 2009 [1], the power conversion efficiency (PCE) has been approached over 22% [2,3] by incorporating an architecture with mesoscopic or planar TiO_2. However, it has been recognized that, no matter condensed or scaffold TiO_2 normally requires high-temperature to annealing (*ca.* 450 °C), which badly hinders flexible perovskite solar cell (PSC) application [4,5]. Then planar configuration, in which the photo-active layer is sandwiched in charge transport layers, arouses much attention by its simple and low temperature technology, normally using 2,2′,7,7′-tetrakis (N,N-p-dimethoxy-phenylamino)-9,9′-spirobifluor-ene (spiro-OMeTAD), poly (3,4-ethylene dioxythio-phene) polystyrene sulphonate (PEDOT:PSS), poly[bis(4-phenyl) (2,4,6-trimethyl-phenyl)amine] (PTAA) and poly (3-hexylthiophene -2,5-diyl) (P3HT) as hole transport layers (HTL) in flexible PSC [6–13]. However, unfortunately, these organic charge transport materials are significantly inclined to severe degradation in moisture and air, thus causing dramatically damage on the photo-active layer. To this end, more stable inorganic materials such as CuI [14], CuSCN [15,16], NiO_x [17–20] have been developed as HTL to replace organic materials.

Particularly, focus on NiO_x-based perovskite solar cell has achieved great progress due to its large band-gap (E_g), high charge mobility, especially for the simple solution processed property. Despite the promising achievement made using NiO_x as HTL to date, while the low conductivity of NiO_x normally leads to high series resistance [21] and low hole extraction. In this regard, the conductivity can be increased by regulating the stoichiometry of NiO_x or by doping. Common dopant, such as Cu [21–23], Mg [24], Li [25], Ag [26], Cs [27] are considered as an effective way to improve electrical conductivity, then mitigating the losses in short-circuit current density (J_{sc}) and fill factor (*FF*). While these metal doped composite materials were always fabricated via corresponding metal precursor mixing, which is most likely to bring in secondary reaction due to the complicated chemical reaction process. What is more, high temperature annealing is normally required to obtaining high crystalline, then inevitably increase the fabrication cost [22,24,26]. To date, graphene-based 2D materials with high surface area and excellent photoelectric properties were used as modifier, incorporating with different materials for reducing the defect states, which shows a great potential candidate for optoelectronic application [28–30].

Recently, a series of researches on under-layer abduction [31], additive-assisted deposition [32], solution-processed technologies [33], have been reported to prepare high-quality film of perovskite with an objective of high-performance PSC. Particularly, the approach of under-layer abduction, in which it was normally comprised of charge transport materials modification, interface engineering [34] between perovskite and charge transport layer, is widely used due to its simple handling and significant effect. The method can not only optimize the

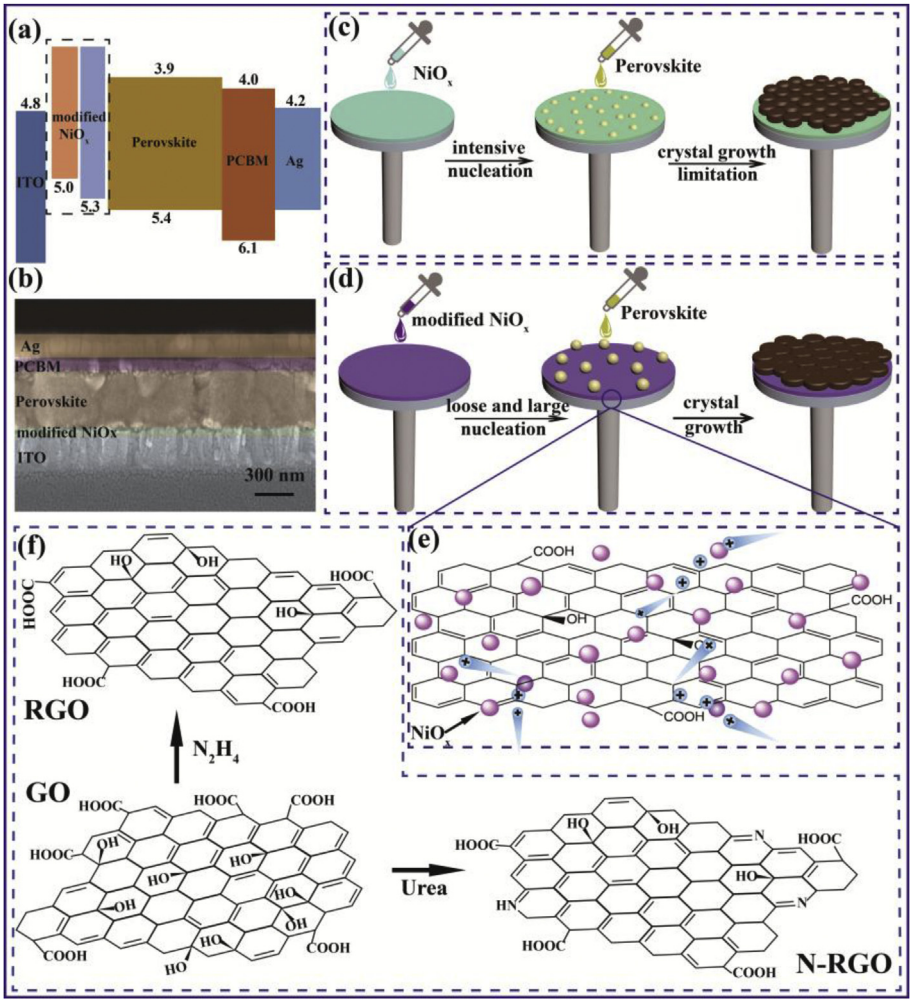

Fig. 1. (a) Schematic energy levels of each layer in PSC. (b) The cross-sectional SEM image of PSC. The schematic illustration of perovskite film fabrication process based on (c) pristine NiO_x. (d) modified NiO_x. (e) The schematic structure of NiO_x with RGO, which demonstrate efficient hole charge carriers transportation in the composite HTL. (f) The chemical structure of GO, RGO and N-RGO.