

Efficient and Stable Large-area Perovskite Solar Cells

Liyuan Han

Photovoltaic Materials Unit, National Institute for Materials Science
Tsukuba, Japan
apvc@nims.go.jp

Abstract The recent dramatic rise in power conversion efficiencies (PCEs) of perovskite solar cells (PSCs) has triggered intense research worldwide. However, high PCE values have often been reached with poor stability at an illuminated area of typically less than 0.1 square centimeter. We used heavily doped inorganic charge extraction layers in planar PSCs to achieve very rapid carrier extraction, even with 10- to 20- nanometer-thick layers, avoiding pinholes and eliminating local structural defects over large areas. The robust inorganic nature of the layers allowed for the fabrication of PSCs with an aperture area >1 square centimeter that have a PCE >15%, as certified by an accredited photovoltaic calibration laboratory. Hysteresis in the current-voltage characteristics was eliminated; the PSCs were stable, with >90% of the initial PCE remaining after 1000 hours of light soaking.

Keywords—Perovskite solar cell; large area; certified; inorganic charge extraction layers; stability

I. INTRODUCTION

Organic-inorganic metal halide perovskite solar cells (PSCs) have attracted attention as a result of the meteoric rise in their solar-to-electric power conversion efficiencies (PCEs) over the past few years [1-2]. However, all high efficiency PSCs thus far have had small areas, with device sizes often 1 cm² is required for certified PCEs to be recorded in the standard solar cell efficiency tables edited by public test centers, such as the National Renewable Energy Laboratory in the United States and the National Institute of Advanced Industrial Science and Technology (AIST) in Japan. The poor reproducibility and lack of uniformity of PSCs make it challenging to obtain high efficiencies with large devices. It is difficult to control the formation of cracks and pinholes in the selective carrier extraction layers over large areas. The dilemma with optimizing such charge carrier extraction layers in solar cells is that the film should be thin to minimize resistive losses, while at the same time, it should cover the entire collector area in a contiguous and uniform manner. [3] Meeting these requirements becomes increasingly difficult as the device area increases.

Here we present a strategy that addresses simultaneously the scale-up and stability issues facing current PSC embodiments.[4] We developed heavily p-doped (p⁺) Ni_xMg_{1-x}O and n-doped (n⁺) TiO_x contacts to extract selectively photogenerated charge carriers from an inverted planar MAPbI₃-PCBM film architecture, as shown in **Fig 1**. We implemented the p- and n-doping by substituting Ni(Mg)²⁺ ions and Ti⁴⁺ ions on the Ni_xMg_{1-x}O lattice and TiO_x matrix with Li⁺ and Nb⁵⁺ ions, respectively. The resulting dramatic increase in the electrical conductivity enabled 10- to 20-nm-thick oxide layers to be used for selective extraction of one type of charge carrier, while improving the electronic blocking effect for the other type by reducing the density of pinholes and cracks over large areas. Accordingly, the series resistance (R_s) of the oxides decreased and the shunt resistance (R_{sh}) greatly increased with respect to the undoped layers, leading to an excellent fill factor (FF), with values exceeding 0.8, and hysteresis-free behavior. With this strategy, we successfully fabricated large-size (>1 cm²) PSCs with an efficiency of up to 16.2%, as shown in **Fig 2**. A PCE of 15% was certified by a public test center (Calibration, Standards and Measurement Team at the Research Center for Photovoltaics, AIST). This is listed as the first official efficiency entry for PSCs in the most recent edition of the solar cell efficiency tables. [5]

Moreover, the devices based on these stable p⁺ and n⁺ inorganic metal-oxide charge extraction layers show high stability, rendering them attractive for future practical deployment of PSCs. **Fig 3** shows the long-term stability of PSCs that use the optimized inorganic charge extraction layers. The silver back contact was protected by a covering glass, which was separated from the front FTO glass by a UV-activated glue used as a sealant. The cells maintained 97% of their initial PCE after being kept in the dark for 1000 hours. Exposing the cells for 1000 hours in short-circuit conditions to full sunlight from a solar simulator resulted in a PCE degradation of less than 10%. During this time, an electric charge of around 72,000 C (4.49 × 10²³ electrons) passed through the device. Thus, the planar cell structure and the metal oxide extraction layers, as well as the organometal halide perovskite material, are robust enough to sustain continued current flow under light exposure for 1000 hours. A further increase in the PCE without sacrificing stability could be obtained by varying the composition of the perovskite, replacing, for example, part of the methylammonium cations in the MAPbI₃ perovskite with formamidinium and a small fraction of the iodide anions with bromide.

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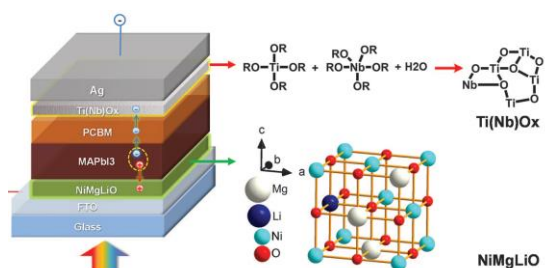


Fig. 1. Structure of the PSC, Diagram of the cell configuration highlighting the doped charge carrier extraction layers. The right panels show the composition of Ti(Nb)Ox and the crystal structure of Li⁺-doped NixMg1-xO, denoted as NiMg(Li)O

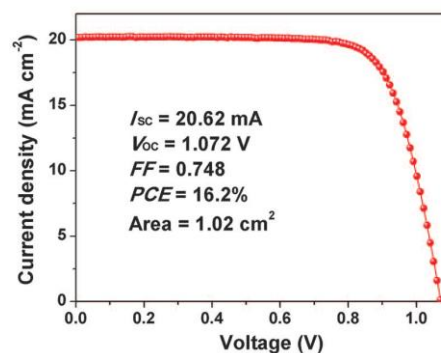


Fig. 2. Performance and stability of large-size cells. J-V curve of the best large cell endowed with antireflection film.

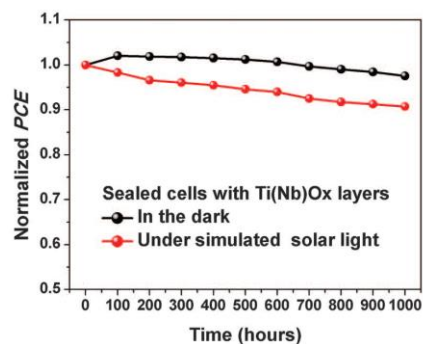


Fig. 3. Stability of large-size cells. Stability of sealed cells kept in the dark or under simulated solar light (AM 1.5; 100 mW cm⁻²; using a 420-nm UV light cut-off filter; surface temperature of the cell, 45° to 50°C; bias potential, 0 V).