Strategy for Hole-Transport-Material-Free Perovskite Solar Cells Using Carbon-Based Electrodes (Vol. 3)

Summary

While perovskite solar cells using carbon-based electrodes have been progressively studied, many challenges remain. In this proposal paper a survey result of some research projects has been completed. One study uses only carbon-based electrodes made of electron conductive materials (carbon nanotubes) with low cost and high stability instead of hole transport material (HTM)/Au electrodes. Another research project provides a functional group-introduced material which improved repeatability and stability of cells. It revealed that the method that adds additives to perovskite films and the method that controls perovskite compositions are both effective. An efficiency of 17% or more has already been reported [1]. Additionally, it has also been shown that the key to high efficiency, high durability and low cost of the perovskite solar cells is to form a strong bonding interface between a perovskite film and an electrode.

Proposals for Policy Development

It is required that research be promoted to solve the following issues regarding the perovskite solar cell using carbon-based electrodes.

- Controlling a defect on a perovskite film
  Controlling defects that trap carriers is imperative to obtain more optical current from a photo-excited carrier. Inactivating defects with additives or optimizing perovskite film structures will be more effective measures.
- Optimizing the bonding between perovskite materials and carbon materials
  From the perspective of the hole collection rate, materials and structures of HTM-free carbon-based electrodes need to be considered to shut off the electron transfer to carbon-based electrodes instead of the HTM.
- Investigation of high durability mechanisms
  HTM/Au electrodes deteriorate due to the diffusion of HTM's dopants or gold atoms, while carbon-based electrodes do so due to carbon exfoliation. It is important that those problematic phenomena be quantitatively studied. It is also necessary to consider cell compositions in line with perpetual changeable features of the perovskite instead of usual stability.

1. Improvement of the perovskite layer
   Many studies that use commercially available carbon pastes for solar cells have been reported recently. Some of those report that improved perovskite films or others provide materials with higher efficiencies of 14 to 17% ([2], etc.). Since the perovskite layer has many possible defect structures, passivating a defect with an additive is often used to prevent defect formation. These additives are thought to extend the lifetime of photoexcited carriers as a result of halogen bonding and other interactions that deactivate traps derived from defective structures. Without those additives, inserting a perovskite material $MAPbI_xBr_{3-x}$. between $MAPbI_3$ and carbon also improved the efficiency [1].
2. Junction of perovskite/carbon electrodes
   In the case of carbon electrodes, an efficiency of 17.58% was obtained in a study where an inorganic HTM (CuSCN) was tried instead of an unstable organic HTM[4]. The energy levels of perovskite solar cells that use carbon-based electrodes with or without HTM are shown in Fig. 1. Introduction of the HTMs not only blocks the flow of excited electrons to carbon-based electrodes but possibly allows the hole transfer to occur smoothly, because the HOMO level of the HTM is close to that of the perovskite.
3. Structural control of carbon
   With carbon modification focused, it has been demonstrated that the usage of carbon nanotube electrodes with oxygen-containing functional groups (-COOH, etc.) can provide strong interaction with PbI_2 thin films. The reconstituted perovskite crystals, in which ions easily diffuse, allow the quality of films and bonding interfaces of solar cells with at most 3% of the initial efficiency to be naturally improved after being left for a long period (Fig. 2), converging to efficiency of 11% or more. Other than the bonding improvement of perovskite layers mentioned above or perovskite/carbon-based electrodes, structure control of carbon-based electrodes is indispensable for enhancing the efficiency and durability.

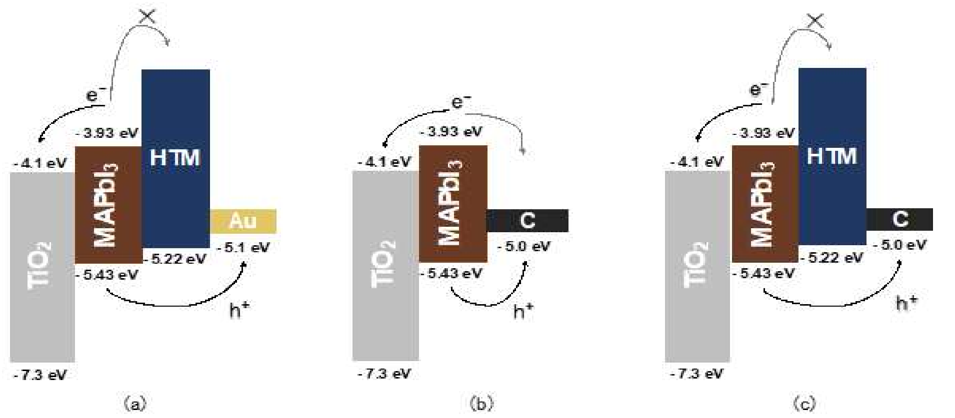

Fig. 1 Energy level of cell constituents

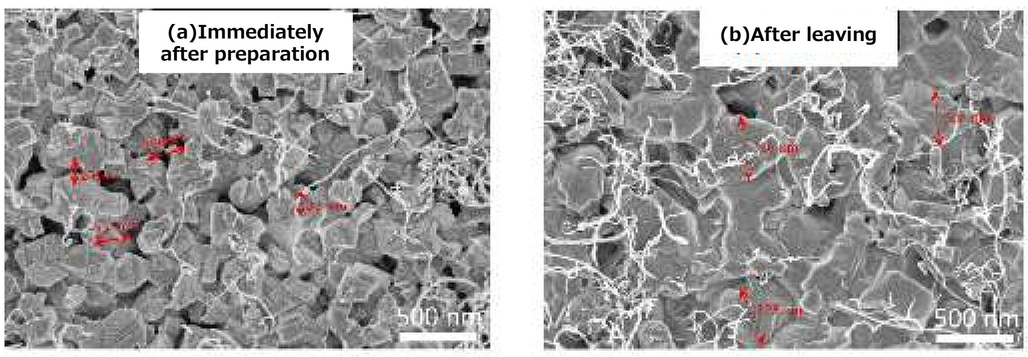

Fig. 2 Electron micrographs of perovskite film surface on the cells using electrodes with oxygen-containing functional groups [3]
(a) Immediately after preparation (b) After leaving

[1] Liu, J. et al. (2019). In situ growth of perovskite stacking layers for high-efficiency carbon-based hole conductor free perovskite solar cells. Journal of Materials Chemistry A, 7(22), 13777-13786.
[2] Liu, C. et al. (2021). Improving the Performance of Perovskite Solar Cells via a Novel Additive of N, 1-Fluoroformamidinium Iodide with Electron withdrawing Fluorine Group. Advanced Functional Materials, 2010603.
[3] Chen, J. et al. (2019). $MAPbI_3$ Self-Recrystallization Induced Performance Improvement for Oxygen-Containing Functional Groups Decorated Carbon Nanotube-Based Perovskite Solar Cells. Solar RRL, 3(12), 1900302.
[4] Wu, X. et al. (2019), Efficient and stable carbon-based perovskite solar cells enabled by the inorganic interface of CuSCN and carbon nanotubes. Journal of Materials Chemistry A, 7(19), 12236-12243.