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Gas-assisted coating of Bi-based $(\text{CH}_3\text{NH}_3)_3\text{Bi}_2\text{I}_9$ active layer in perovskite solar cells**Takayuki Okano^a, Yoshikazu Suzuki^{a,b**}**^a Graduate School of Pure and Applied Sciences, University of Tsukuba, Ibaraki 305-8573, Japan^b Faculty of Pure and Applied Sciences, University of Tsukuba, Ibaraki 305-8573, Japan**Abstract**

Methylammonium bismuth iodide, $(\text{CH}_3\text{NH}_3)_3\text{Bi}_2\text{I}_9$, is a promising lead-free perovskite active layer for solar cells. In this study, by using gas-assisted deposition method, we have successfully prepared dense and smooth $(\text{CH}_3\text{NH}_3)_3\text{Bi}_2\text{I}_9$ active layer, resulting in 25% improvement in V_{OC} (from 0.548 V to 0.686 V) and 17% improvement in efficiency (from 0.070% to 0.082%), compared with the conventional 1-step method.

Keywords: Perovskite solar cells; Lead-free; Bi-based; $(\text{CH}_3\text{NH}_3)_3\text{Bi}_2\text{I}_9$; Gas-assisted

1. Introduction

Perovskite solar cells (PSCs) have attracted much attention due to their rapidly-increased and high power conversion efficiency (PCE). Kojima et al. reported the PSCs with conversion efficiency of 3.8 % in 2009 [1]. Within only 7 years, the PCE increased up to 22.1 % [2]. The high efficiency is attributed to excellent optical and electrical properties of $\text{CH}_3\text{NH}_3\text{PbX}_3$ (X=I, Br or Cl) [3-5]. However, the $\text{CH}_3\text{NH}_3\text{PbI}_3$ active layer includes toxic Pb; an alternative lead-free active layer is desired. Sn replacement for Pb has been tried to solve this problem [6,7], but durability is the most serious problem in Sn-based PSCs.

Currently, Bi-based perovskite-type compounds gather much attention [8-14]. Among them, $(\text{CH}_3\text{NH}_3)_3\text{Bi}_2\text{I}_9$ has high stability in air since Bi^{3+} ions are stable, but the typical PCE of $(\text{CH}_3\text{NH}_3)_3\text{Bi}_2\text{I}_9$ PSCs are reported as 0.1-0.2%. Singh et al. have recently reported the 10-week long-term stability of $(\text{CH}_3\text{NH}_3)_3\text{Bi}_2\text{I}_9$ PSCs [10], where the performance degradation was only 25% even in the ambient conditions. This result has demonstrated the high stability of $(\text{CH}_3\text{NH}_3)_3\text{Bi}_2\text{I}_9$ PSCs. Compared with commonly used perovskites having favorable band gaps, *e.g.* $\text{CH}_3\text{NH}_3\text{SnI}_3$ (~1.3 eV [6]) and $\text{CH}_3\text{NH}_3\text{PbI}_3$ (~1.5 eV [15]), $(\text{CH}_3\text{NH}_3)_3\text{Bi}_2\text{I}_9$ has a wider band gap (~2.1 eV [8,13]). $(\text{CH}_3\text{NH}_3)_3\text{Bi}_2\text{I}_9$, however, can be an alternative for $\text{CH}_3\text{NH}_3\text{PbI}_3$ due to the high absorption coefficient ($(\text{CH}_3\text{NH}_3)_3\text{Bi}_2\text{I}_9$: $1.1 \times 10^5 \text{ cm}^{-1}$ at 500 nm [13], $\text{CH}_3\text{NH}_3\text{PbI}_3$:

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