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Effects of Guanidinium and Cesium Addition to $CH_3NH_3PbI_3$ Perovskite Photovoltaic Devices †

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Abstract: $CH_3NH_3PbI_3$ perovskite compounds are unstable in air due to the migration of CH_3NH_3. The purpose of the present work is to investigate the effects of addition of guanidinium $C(NH_2)_3$ (GA) and cesium (Cs) on $CH_3NH_3PbI_3$ perovskite solar cells. The addition of GA/Cs and the insertion of decaphenylpentasilane between the perovskite and hole transport layer improved the external quantum efficiency and short-circuit current density, and the conversion efficiencies were stable. First-principles calculations on the density of states and band structures showed reduction in the total energy by the GA addition.

Keywords: solar cell; guanidinium; cesium; polysilane; first principles calculation; photovoltaic device

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1. Introduction

Although silicon is the most common solar cell material, the fabrication process is expensive. On the other hand, $CH_3NH_3PbI_3$ ($MAPbI_3$) compounds have been widely used for perovskite solar cells [1–5], and the $MAPbI_3$ perovskite compound have tunable band gaps and easy fabrication process with low cost. However, the $MAPbI_3$ compounds are unstable in the ordinary air due to the migration and desorption of CH_3NH_3 (MA) molecules [6–9]. In order to stabilize the perovskite structure, various kinds of cations such as formamidinium ($HC(NH_2)_2$, FA) [10–16], ethylammonium ($CH_3CH_2NH_3$, EA) [17,18], or guanidinium ($C(NH_2)_3$, GA) [19,20], which have larger ionic radii than methylammonium (CH_3NH_3, MA), have been introduced at the MA site, and the fabricated cells were stable to some extent. Introducing alkali metals such as cesium (Cs) [21–23], rubidium (Rb) [24–26], potassium (K) [27–30], and sodium (Na) [31–34], could also be effective because these metal elements do not desorb from the perovskite crystal.

Another approach to improve the stability of the perovskite solar cells is introducing polymeric materials such as polysilane [35–39]. Polysilanes have two important features that are *p*-type semiconductors and are stable at elevated temperatures above 300 °C.

The purpose of the present work is to fabricate $MAPbI_3$ perovskite solar cells added with GA and Cs, and to characterize the devices from experiments and first-principle calculation. In the present work, polysilane was also used both for protection of the perovskite layer and for hole transport [35,37].

2. Experimental Procedures

A fabrication process of photovoltaic devices of the present work is same as those of the previous works [40–42]. A TiO_2 compact layer was formed on the F-doped tin-oxide (FTO) substrates, and a mesoporous TiO_2 layer was formed on the compact TiO_2 layer.

A perovskite layer with desired composition was formed on the mesoporous TiO_2 layer, and decaphenylcyclopentasilane (DPPS), which is a one kind of polysilane, was formed on the perovskite layer [35]. A layer of 2,2′,7,7′-tetrakis-(N,N-di(p-methoxyphenyl)amine)-9,9′-spirobifluorene (spiro-OMeTAD) was formed on the DPPS, and then gold (Au) metal electrodes were formed on the spiro-OMeTAD hole transport layer.

The current density voltage characteristics of the fabricated devices were measured under a solar simulating light source operated at 100 mW cm^{-2}. X-ray diffraction was used to investigate the microstructures of the devices, and first-principles calculation was also carried out to estimate the properties of the perovskite crystals [43–45].

3. Results and Discussion

In order to estimate the structural stability of perovskite compounds, a tolerance factor (t) is used [5,46,47] using the following equation:

$$t = \frac{r_A + r_X}{\sqrt{2}(r_B + r_X)}$$

where r_A, r_B, and r_X are the ionic radii of the A, B, and X ions for ABX_3 perovskite structures, respectively [5]. When the t-value is 1, the perovskite compound has a stable crystal structure with cubic symmetry. From the previous experimental studies on perovskite compounds, the perovskite structure could be formed in the range of $0.813 \leq t \leq 1.107$. Calculated t-factors of perovskite compounds are listed in Table 1. From this calculation, co-addition of GA and Cs could be one of the effective ways to stabilize the $MAPbI_3$ structure.

Table 1. Calculated t-factors of perovskite compounds.

Perovskite	t
$MAPbI_3$	0.912
$GAPbI_3$	1.039
$CsPbI_3$	0.851
$MA_{0.75}GA_{0.125}Cs_{0.125}PbI_3$	0.920
$MA_{0.845}GA_{0.125}Cs_{0.03}PbI_3$	0.926

Figure 1a is a structure model of $MA_{0.75}GA_{0.125}$ $Cs_{0.125}PbI_3$. MA molecules are substituted by GA and Cs, which are located diagonally. Based on this structure model, the physical properties could be predicted. Figure 1b is an electron diffraction pattern of $MA_{0.75}GA_{0.125}$ $Cs_{0.125}PbI_3$ calculated along [111]. Although the usual $MAPbI_3$ has 6-fold symmetry from the [111] incidence, the calculated electron diffraction pattern in Figure 1b shows 2-fold symmetry, which is due to the doped Cs and GA. Therefore, the high symmetry dimension of the space group of $Pm\bar{3}m$ for $MAPbI_3$ is reduced to lower symmetry.

From the first-principle calculations and experimental evaluations, addition of small amount of GA to $MAPbI_3$ is effective to stabilize the perovskite structure [48,49]. Although the addition of GA expands and distorts the crystal lattice of $MAPbI_3$, Cs would reduce the distortion of the lattice, which could lead to the stability of the perovskite crystal.

For the fabricated device with $MA_{0.845}GA_{0.125}Cs_{0.03}PbI_3$ perovskite compound, the high photoconversion efficiency was obtained, which would be in good agreement with the calculated results. The stability of the device at the room temperature was also good. Although several works on $GAPbI_3$ [50] and $CsPbI_3$ [51,52] compounds have been reported, few works have been reported on the co-addition of GA and Cs to $MAPbI_3$. The present work indicated the effectiveness of the co-addition of GA and Cs to the $MAPbI_3$ on the photovoltaic properties for perovskite solar cells.

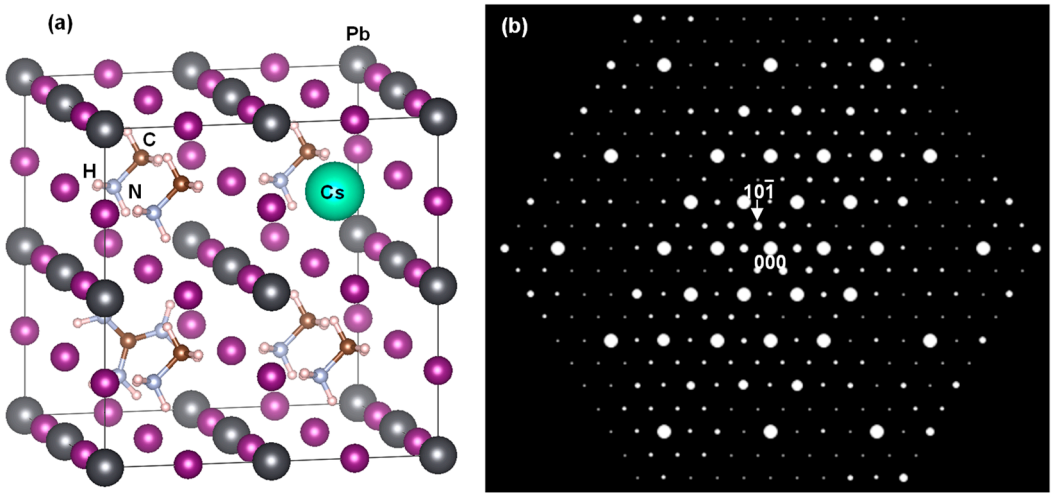

Figure 1. (**a**) Structure model and (**b**) calculated electron diffraction pattern along [111] of $MA_{0.75}GA_{0.125}$ $Cs_{0.125}PbI_3$.

4. Conclusions

The effects of addition of guanidinium GA and Cs on $MAPbI_3$ perovskite solar cells were investigated. The co-addition of GA/Cs and the insertion of DPPS between the perovskite and spiro-OMeTAD improved the EQE, and the conversion efficiencies were stable. The calculated electron diffraction pattern of $MA_{0.75}GA_{0.125}Cs_{0.125}PbI_3$ showed reduction in the structural symmetry. First-principles calculations also showed reduction in the total energy by the GA addition, which indicated the stabilization by the addition.

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