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Effects of the Addition of Copper Chloride and Potassium Iodide to Methylammonium-Based Perovskite Solar Cells †

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Abstract: Organic–inorganic hybrid perovskite solar cells have the advantage of being able to implement a high conversion efficiency, easy fabrication process and low cost to commercial products of photovoltaic devices. The perovskite solar cells have a photovoltaic performance with reduced durability due to the volatility of organic cations and toxic lead in the perovskite crystal as an active layer. The purpose of this research is to investigate the effect of additives such as cupper chloride and potassium iodide in the perovskite crystal on the photovoltaic properties and electronic structure. The distribution of 3d orbital of cupper ion conjugated with 5p orbital of iodine ion at the valence band, and 6p orbital of lead ion at the conduction band, influences the charge generation and transfer, as well as the carrier mobility with a narrowing band gap. The addition of potassium iodide delocalizes the charge distribution near the cupper, iodide, and lead ions, which promotes the charge generation and carrier diffusion, yielding an increase in the short circuit current density relating to the conversion efficiency.

Keywords: perovskite; solar cell; photovoltaic device; copper; methylammonium; potassium; polysilane; decaphenylcyclopentasilane

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1. Introduction

Lead halide perovskite semiconductors attracted attention as the active layer of electroluminescence in the 1990s [1]. After the first application of a $CH_3NH_3PbI_3$ compound to solar cells [2], the lead halide perovskites have been actively researched worldwide [3–7]. Since $CH_3NH_3PbI_3$ perovskite solar cells have a high sensitivity to visible light and an easy production process, it is expected that they will be next-generation solar cells. However, toxicity problems in including Pb perovskite solar cells have impeded their wholescale commercial application. Moreover, the long-term instability caused by the decomposition of perovskite crystal has still not been resolved. The contamination of soil and water by Pb^{2+} ions is permanent. When organisms take in Pb, it is not eliminated from the body and causes serious adverse effects. It enters the human body and causes dysfunction in the nervous, digestive, and blood systems. It is mandatory to provide safe and environmentally friendly products. From previous studies, less toxic ions such as Sn^{2+}, Ge^{2+}, Co^{2+}, and Cu^{2+} are expected to be alternative elements [8–12]. Among them, the environmentally friendly transition metal Cu^{2+} has been examined as a candidate for Pb^{2+} replacement, but there are few reported cases [13–15]. In addition, the durability of perovskite solar cell is caused mainly by the decomposition of the perovskite crystals due to methylammonium (MA) desorption. To solve these problems, attempts to introduce additives into the perovskite layer to improve the electronic properties have been studied [16–27]. Previous studies have reported that the substitution of potassium (K) can inhibit MA desorption,

resulting in improved performance and long-term stability [28–33]. The aim of this work is to fabricate and characterize the perovskite solar cell doped with cupper chloride and potassium iodide. The photovoltaic properties, morphologies, and crystal structure were investigated via the substitution of Cu^{2+} and K^+ ions. The stability of the performance was measured. In addition, first-principle calculations were performed and compared with the experimental results.

2. Materials and Methods

The present perovskite solar cells were prepared according to the literature [34–38]. For preparing the perovskite compound, a mixture of CH_3NH_3I (MAI, Tokyo Chemical Industry, 2.4 M), $PbCl_2$ (0.8 M, Sigma-Aldrich, Tokyo, Japan), copper chloride ($CuCl_2$, Sigma Aldrich), and potassium iodide (KI, Wako Pure Chemical Corporation, Osaka, Japan) with the desired molar ratio in *N*,*N*-dimethylformamide (DMF, NacalaiTesque, Kyoto, Japan, 0.5 mL), was stirred at 60 °C for 24 h. As a standard recipe, the mole of MAI and $PbCl_2$ in DMF was adjusted to be 2.4 M (190.8 mg) and 0.8 M (111.2 mg). In the doped case of 2% $CuCl_2$, the mole of MAI, $PbCl_2$, and $CuCl_2$ was adjusted to be 2.4 M (190.8 mg), 0.78 M (109.0 mg), and 0.02 M (1.07 mg). In the doped case of 2% $CuCl_2$ and 2% KI, the mole of MAI, $PbCl_2$, KI, and $CuCl_2$ was adjusted to be 2.35 M (186.9 mg), 0.78 M (109 mg), 0.01 M (0.93 mg), and 0.02 M (1.07 mg). The perovskite solutions were spin-coated on TiO_2 with air flow at three times. A solution of decaphenylcyclopentasilane (DPPS, Osaka Gas Chemical, OGSOL SI-30-15, 10 mg) was prepared in chlorobenzene (0.5 mL) and dropped onto the perovskite layer during the last stage of the spin-coating process. DPPS was used as a hole-transporting material, with the cell being protected from moisture and oxygen. Annealing process was performed at 200 °C. All procedures were performed in air atmosphere. A gold (Au) electrode was deposited to serve as the top electrode. The structure of the solar cells is denoted as FTO/TiO_2/perovskite/DPPS/spiro-OMeTAD/Au. The prepared cells were stored at temperature of 22 °C and with humidity below 30%.

The electronic structures of the Cu-, and K-doped perovskite crystal were single-point and calculated using ab initio quantum calculation [39–45] based on the restricted Hartree–Fock method and hybrid density functional theory (DFT) using restricted B3LYP with LANL2MB as the basis set (Gaussian 09). The $MAPbI_3$ perovskite crystals with supercells of $2 \times 2 \times 2$ as cluster model were formed on the basis of the experimental results using X-ray diffraction data.

3. Results and Discussion

3.1. First-Principles Calculation

Electron density distributions at the highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) as well as the electrostatic potential (ESP) and partial charge of MAPb(Cu)I_3 and MA(K)Pb(Cu)I_3 perovskite cubic crystals with $2 \times 2 \times 2$ supercells are shown in Figure 1a,b, respectively. The electron density distributions of the MAPb(Cu)I_3 perovskite demonstrated that the 6p orbitals of the Pb atom dominated at the LUMO. The 3d orbitals of Cu^{2+} ion and the 5p orbitals of the I^- ion were delocalized at the HOMO. The charge transfer between the 3d orbitals of the Cu^{2+} ion and the 5p orbital of the I^- ion would promote the carrier generation and carrier diffusion.

The electron density distributions of the MA(K)Pb(Cu)I_3 perovskite are shown in Figure 1b. The 6p orbitals of the Pb^{2+} ion and the 4s orbitals of the Cu^{2+} ion were formed in the LUMO. The 3d orbitals of the Cu^{2+} ion and the 5p orbitals of the I^- ion were dominated in the HOMO. The addition of K^+ caused 4s orbitals of Cu^{2+} conjugated with 6p orbital of Pb ion in the LUMO, promoting charge transfer between 4s orbital and 6p orbital in the coordination band. For the ESP and partial charge of MAPb(Cu)I_3 and MA(K)Pb(Cu)I_3 perovskite, the charges of Cu^{2+} and I^- ion were delocalized by the positive charge of K^+, which would promote the carrier diffusion and an increase in short circuit current density.

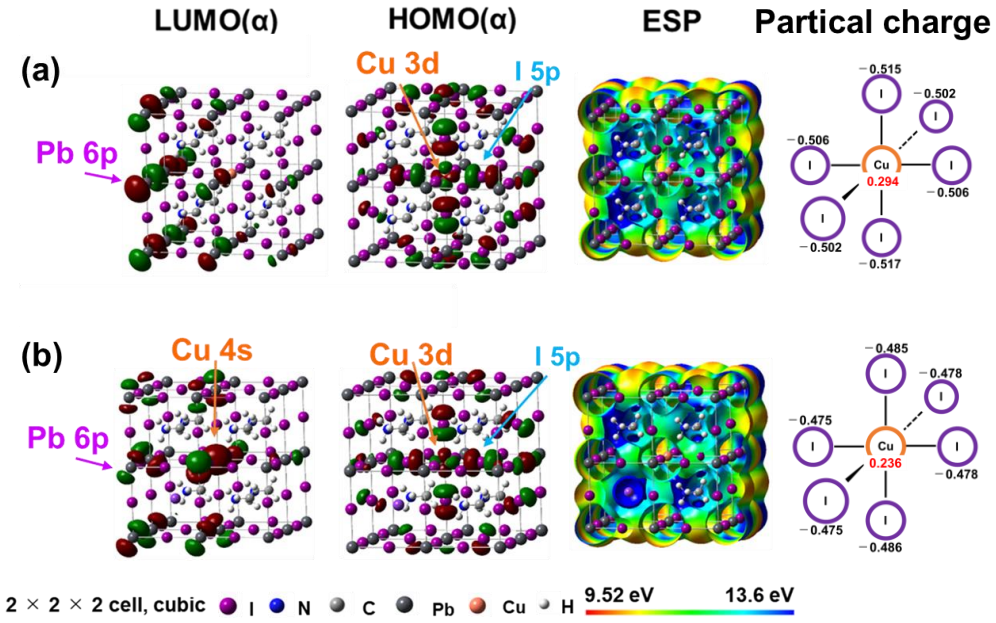

Figure 1. The electron density distributions at the highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO), electrostatic potential (ESP) and partial charge of (**a**) $MAPb_{0.963}Cu_{.0.037}I_3$ and (**b**) $MA_{0.875}K_{0.125}Pb_{0.963}Cu_{0.037}I_3$ perovskite cubic crystals with 2 × 2 × 2 supercells.

3.2. Device Characterization

The photovoltaic parameters of open-circuit voltage (V_{OC}), short-circuit current density (J_{SC}), fill factor (FF), conversion efficiency (η), and band gap (E_g) are listed in Table 1. The conversion efficiencies of the Cu- and K-doped perovskite solar cells were found to be 10.59%, which were higher than that of the Cu-added solar cell. The addition of Cu^{2+} and K^+ ions supported the photovoltaic performance with an increase in J_{SC} related to η, due to the enhancement of the carrier transfer in the perovskite crystal.

Table 1. Photovoltaic parameters of present perovskite photovoltaic devices.

Devices	J_{SC} (mA cm^{-2})	V_{OC} (V)	FF	η (%)	η_{ave} (%)	E_g (eV)
$MAPbI_3$	21.6	0.822	0.622	11.03	9.00	1.56
$+Cu^{2+}$ 2%	18.5	0.800	0.627	9.26	8.47	1.56
$+Cu^{2+}$ 2%, K^+ 2%	21.4	0.837	0.590	10.59	8.99	1.56

4. Conclusions

The fabrication and characterization of Cu- and K-doped $MAPbI_3$ perovskite solar cells was performed. The photovoltaic properties and electronic structures were investigated. The 2% Cu- and 2% K-doped perovskite solar cells had the photovoltaic performance of conversion efficiency with increases in the Jsc values. The charge transfer between the 3d orbital of the Cu^{2+} ion and the 5p orbitals of the I^- ion would influence the carrier generation and diffusion in the cubic $MAPb(Cu)I_3$ perovskite. Addition of K^+ into $MAPb(Cu)I_3$ perovskite caused the 3d orbital of the Cu^{2+} ion in the HOMO and LUMO. The addition of K^+ delocalized the 3d and 5p orbitals of the Cu^{2+} and I^- ions near the HOMO as well as the 4s and 6p orbital of the Cu^{2+} and Pb^{2+} ions near the LUMO with a wide charge distribution, which promoted the carrier generation and charge transfer, yielding an increase in J_{SC} related to η.

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