



Short communication

Study on cobalt doped tin based perovskite material with enhanced air stability

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ABSTRACT

Although the power conversion efficiency of the lead-halide perovskite solar cells boost to > 20%, the toxicity of lead hinders their large-scale implementation. Tin based perovskite have less toxicity, however its air stability is a big challenge. In this study, the air stability of tin based perovskite is improved by doping cobalt. The cobalt doped tin based perovskite thin films ($\text{GASn}_{1-x}\text{Co}_x\text{I}_{2-2x}\text{Cl}_{1+2x}$) with cubic structure are prepared by one-step spin coating method. All $\text{GASn}_{1-x}\text{Co}_x\text{I}_{2-2x}\text{Cl}_{1+2x}$ ($0.025 \leq x \leq 0.15$) perovskite materials exhibit strong absorption in the visible light spectrum range. According to voltammetric measurement, the highest occupied molecule orbit (HOMO) and the lowest unoccupied molecule orbit (LUMO) of $\text{GASn}_{1-x}\text{Co}_x\text{I}_{2-2x}\text{Cl}_{1+2x}$ ($x=0.15$) are -5.45 and -3.99 eV, respectively, and the bandgap is calculated to be 1.46 eV. The binding energy increase of Sn3d and I3d for the cobalt doped tin based perovskite thin film might be the reason for enhanced air stability.

1. Introduction

Due to their higher power conversion efficiency (PCE), silicon based solar cells dominate the photovoltaic market at present. However, their high cost and heavily polluted manufacturing procedure decrease the feasibility of widespread use [1]. Therefore there have been intensive efforts to develop the alternatives to silicon based solar cells [2,3].

An innovative photovoltaic material, lead based organic-inorganic hybrid perovskite ($\text{CH}_3\text{NH}_3\text{PbX}_3$ ($X=\text{I}, \text{Br}, \text{Cl}$)), emerged in 2009, and the PCE of perovskite solar cells boost to > 20% in 2015 via solvent engineering, interface engineering and composition engineering [3–5]. Owing to their lower cost and their comparable efficiency to silicon based solar cells, perovskite solar cells are expected to replace silicon based solar cells in future [3,4]. Nevertheless, the toxicity of lead in the perovskite will hinder their practical application. Replacing the lead element in the perovskite with less toxic metal elements is a big challenge. Tin based perovskite solar cells have been developed with the initial PCE of over 6% under 1 sun illumination, but their stability is a serious problem: their PCE decreases by 64.0% in 24 h when the solar cell devices are stored in nitrogen glove box after careful sealing [6,7]. It is because Sn^{2+} ion will be rapidly oxidized to its more stable Sn^{4+} analogue in the atmosphere environment. Although other non-toxic perovskite materials, eg. $\text{Cs}_2\text{AgBiBr}_6$, have been explored recently,

the materials with similar photoelectronic properties have not yet developed [8–10].

In order to inhibit the oxidation of Sn^{2+} ion, Co^{2+} or Fe^{2+} is usually introduced to the electroplating solution in electroplating industry [11]. Inspired by this, it is expected that the oxidation of Sn^{2+} in tin based perovskite materials in air is inhibited by doping Co^{2+} .

Methylammonium (MA) is usually used as the organic cation in lead or tin based perovskite; however, it is reported that the replacement of MA by guanidinium (GA) leads to suppressed hysteresis, higher device open voltage and PCE, owing to a nearly zero dipole moment of GA cation and suppressed nonradiative carrier loss in perovskites [12,13]. According to the empirical Goldschmidt tolerance factor, it is predicted that GA cation is too large to be incorporated into the lead or tin iodide cavity [14,15]. However, since all of three NH_2 groups in GA can form hydrogen bonds with I atoms, the lead or tin iodide cavity will be compressed, leading to higher stability of perovskites with GA [13]. Therefore, GA is used as the cation in tin based perovskite materials in this study.

2. Experimental

The perovskite thin films were prepared by one-step spin coating method. SnI_2 , CoCl_2 and guanidine hydrochloride were dissolved in N,

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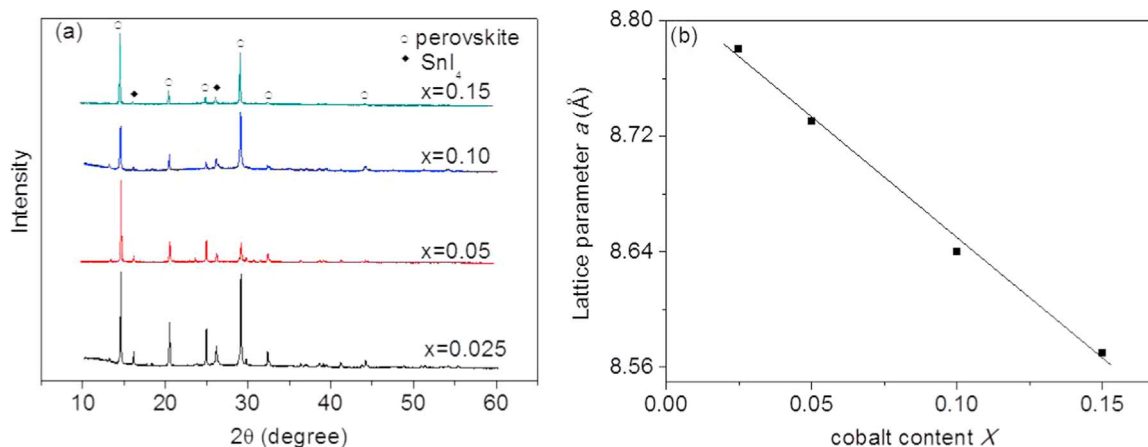


Fig. 1. (a) XRD patterns of the perovskite thin films with various cobalt content x . (b) Lattice parameters of the perovskite thin films as a function of cobalt content x .

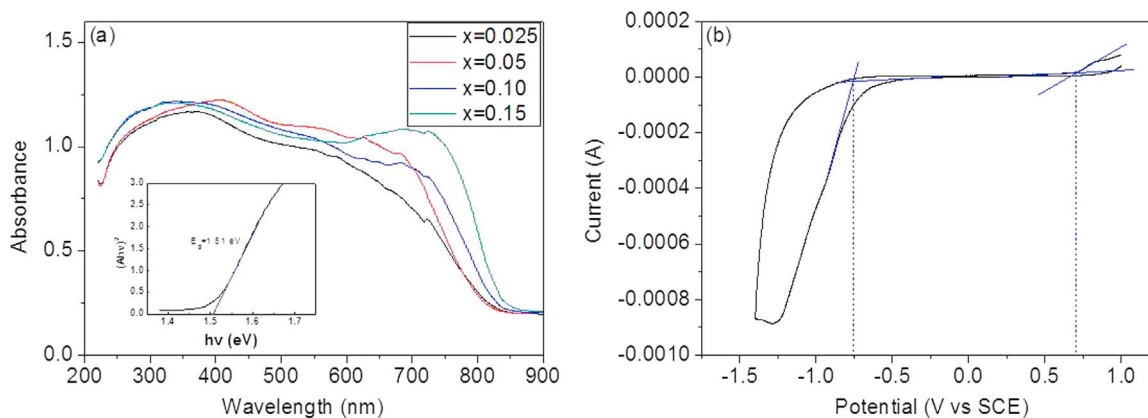


Fig. 2. (a) UV-visible absorbance spectra of the perovskite thin films with various cobalt content x . Inset: Tauc plot showing the characteristics of bandgap. (b) Cyclic voltammetric curve of the $\text{GASn}_{1-x}\text{Co}_x\text{I}_{2-2x}\text{Cl}_{1+2x}$ ($x=0.15$) perovskite thin film.

N' -dimethylformamide (DMF) and stirred at 60 °C for 12 h. The total concentration of SnI_2 and CoI_2 was 0.5 M, and the concentration of GA was 0.5 M. The precursor solution was spin-coated on soda-lime glass at 2000 rpm for 30 s, and the resulting thin films were annealed at 100 °C for 30 min on the hot plate, thus the dark brown, cobalt doped tin based perovskite ($\text{GASn}_{1-x}\text{Co}_x\text{I}_{2-2x}\text{Cl}_{1+2x}$) materials were obtained. All of the fabrication process was conducted in the atmosphere environment, not in the glove box filling nitrogen.

The crystal structure of thin films was determined by X-ray diffractometry (XRD, D8 Advance, Bruker, Germany). The thin films of $\text{GASn}_{1-x}\text{Co}_x\text{I}_{2-2x}\text{Cl}_{1+2x}$ perovskite were stored in the drier for 60 days, and the XRD patterns of the as-prepared and stored samples were compared.

The optical absorption spectra were measured by the ultraviolet-visible spectrophotometer (UV-3600, Shimadzu, Japan), and the scanning wavelength was ranged from 220 to 900 nm. The x-ray photoelectron spectrum (XPS) was measured by ESCALAB 250 (ThermoFisher Scientific, USA) equipped with x-ray source of twin anode Al K α 300 W, and all binding energies were calibrated to C1s at 284.6 eV.

The cyclic voltammetric measurement was conducted in a three-electrode cell, and the potential scan rate was 0.01 V s⁻¹. In the three-electrode cell, the $\text{GASn}_{1-x}\text{Co}_x\text{I}_{2-2x}\text{Cl}_{1+2x}$ perovskite thin film on indium tin oxide (ITO) conductive glass, platinum foil and saturated calomel electrode (SCE) were used as the working electrode, counter electrode and reference electrode, respectively. The photocurrent was measured in a quartz glass beaker containing acetonitrile with 100 mM

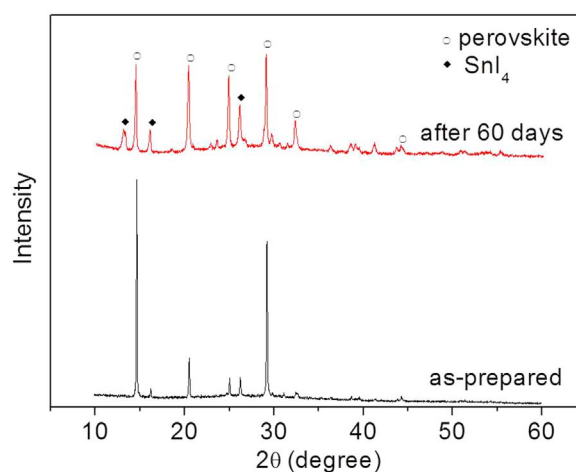


Fig. 3. XRD patterns of the as-prepared and stored perovskite thin films ($x=0.15$). The diffraction peaks of SnI_4 become stronger for the stored sample.

NaClO_4 . The visible light irradiation was emitted from xenon lamp (AULITT, Beijing, P. R. China) with the irradiation intensity of 100 mW cm⁻² on the thin film.

3. Results and discussion

$\text{CH}_3\text{NH}_3\text{SnX}_3$ ($X=\text{Br}, \text{I}$) perovskite thin films fade within seconds when exposed in atmosphere environment [6,7]; in contrast, there is

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