



Carbon-based, all-inorganic, lead-free Ag_2BiI_5 rudorffite solar cells with high photovoltages

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ABSTRACT

Lead-free Ag-Bi-I rudorffite materials, such as Ag_2BiI_5 , have gained ever-increasing attention as potential alternatives to lead halide perovskites for solar cell applications. However, nearly all of rudorffite solar cells are built with a mesoporous structure, which contain instable and expensive hole-transport-layers (HTLs) along with noble metal-electrodes. Herein, carbon-based, all-inorganic Ag_2BiI_5 rudorffite solar cell that is free of HTL and noble metal-electrode is reported for the first time. By improving the quality of Ag_2BiI_5 film with air blowing recipe, the solar cell with much enhanced efficiency of 0.71% is realized. In particular, it yields an open-circuit voltage (V_{oc}) of 0.770 V, standing the highest value among Ag_2BiI_5 solar cells reported so far. Hence, our work opens the door for developing rudorffite solar cells with simplified fabrication procedures and low cost.

1. Introduction

Since the advent of perovskite solar cells (PSCs) in 2009, their power conversion efficiencies (PCEs) have soared to 25.5% until now [1]. However, there are still some fundamental obstacles that prevent them from commercialization [2,3], such as the issue of toxic Pb^{2+} cations and inferior thermo/photo/hydro-stability of lead halide perovskites. These substantial obstacles motivate great efforts to explore lead-free analogues [4-9]. Primarily, tin-based halide perovskites have been investigated; [8,10] yet, the stability of resultant solar cells is impeded by the oxidation of Sn^{2+} cations [11].

In this case, the Ag-Bi-I families that are known as rudorffites are proposed [5]. In particular, AgBi_2I_7 with the bandgap (E_g) of 1.87 eV was demonstrated as the light absorber of solar cells by Kim et al. [12], which yielded a PCE of 1.22%. By use of the dynamic hot casting method, the PCEs of AgBiI_4 and Ag_2BiI_5 solar cells were boosted to 2.1% and 2.6%, respectively [13]. Turkevych et al. [14] reported the solar cells with a structure of FTO/c-TiO₂/m-TiO₂/Ag₃BiI₆/PTAA/Au, which exhibited a superior PCE of 4.3%. Pai et al. [15] reported the $\text{Ag}_3\text{BiI}_{5.92}\text{S}_{0.04}$ solar cells with the best PCE of 5.56% by the Ar-assisted method, where S incorporation increased the valence band edge. By

using SnO_2 as an electron transport layer (ETL) and adding Li-TFSI, Zhang et al. [16] obtained a high open-circuit voltage (V_{oc}) approaching 0.9 V for AgBiI_4 solar cells. Hu et al. [5] reported the bulk heterojunction bismuth-based solar cells with the photoactive layer consisting of in-situ phase-separated $\text{Cs}_3\text{Bi}_2\text{I}_9$ and $\text{Ag}_3\text{Bi}_2\text{I}_9$ components, achieving a high PCE of approximate 3.6% and an unprecedented V_{oc} reaching 0.89 V. These results demonstrate the great promising of Ag-Bi-I rudorffites for photovoltaic applications. However, all of the rudorffite solar cells are constructed with a mesoporous structure. The instable, expensive hole transport layers (HTLs) and noble metal electrodes are involved in them inevitably, which not only burden their production costs, but also cause new stability issues.

In terms of PSCs based on lead halide perovskites, there have some reports on the ones with simplified structures, benefiting from the ambipolar carrier transport of lead halide perovskites. Typically, carbon-based, all-inorganic PSCs that are free of HTLs and noble metal electrodes were reported to exhibit better stability, lower cost, and simpler fabrication [17,18]. However, as for Ag-Bi-I rudorffites, there is no any demonstration of solar cells with a simplified configuration. Thus, it is interesting to clarify whether the rudorffite solar cells could be fabricated with a simplified structure, as the carbon-based, all-inorganic

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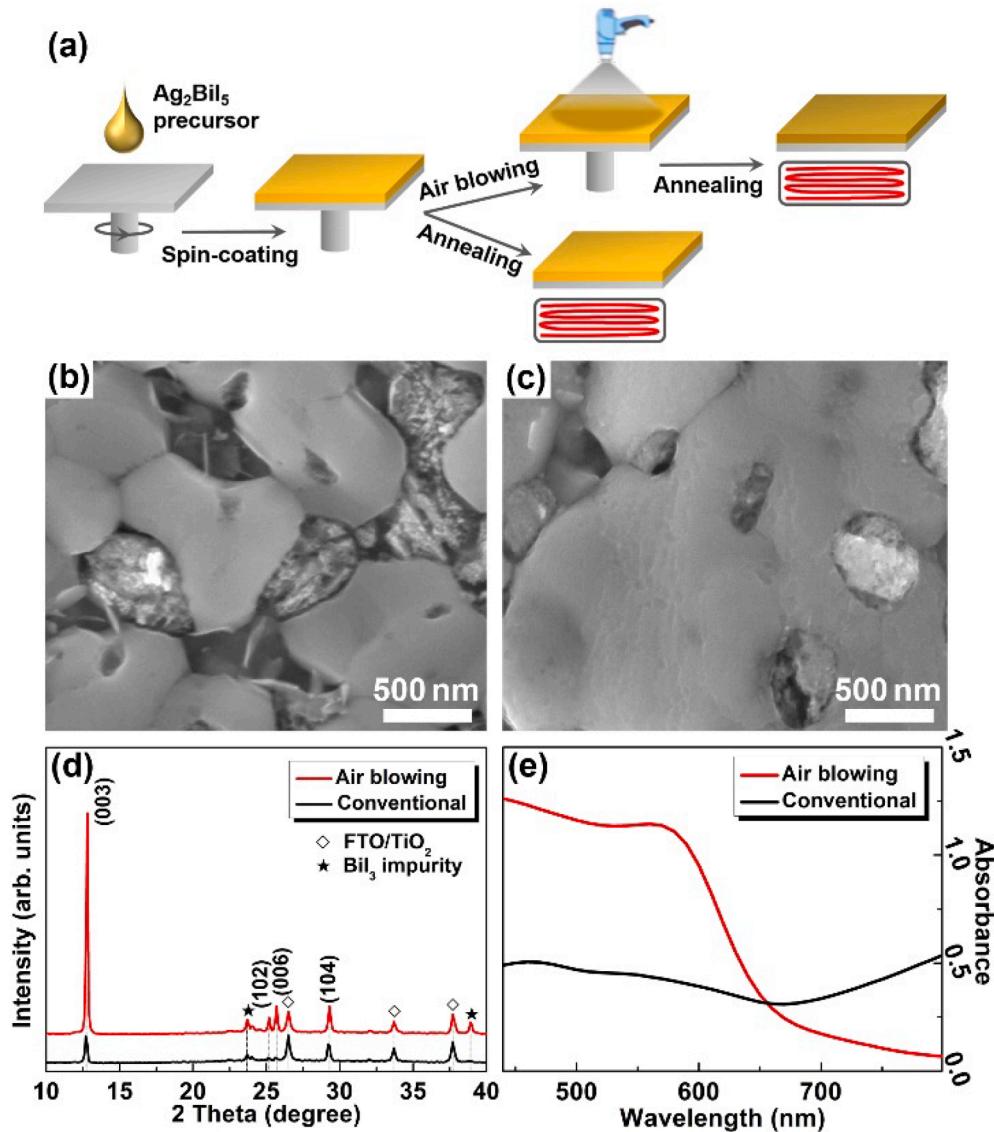


Fig. 1. (a) Schematic preparation procedures of Ag_2BiI_5 films prepared without and with air blowing modification, respectively. (b-c) Top-view SEM images of Ag_2BiI_5 films prepared (b) without and (c) with air blowing modification, respectively. (d) XRD patterns and (e) UV-vis absorption spectra of as-prepared Ag_2BiI_5 films.

PSCs.

Herein, we report for the first time the carbon-based, all-inorganic, lead-free solar cell based on Ag_2BiI_5 , which yields a PCE of 0.36% and an open-circuit voltages (V_{oc}) of 0.595 V. Such sluggish photovoltaic performance primarily arises from the poor quality of one-step spin-coated Ag_2BiI_5 film. Therefore, further modification of one-step method with air blowing recipe is performed. Because of improved coverage and crystallinity of Ag_2BiI_5 film, an optimized PCE of 0.71% and a record-high V_{oc} of 0.770 V are obtained. Hence, our work uncovers the feasibility to construct ruddorfite solar cells with a simplified structure.

2. Experimental details

FTO/c-TiO₂ substrate was prepared according to our previous work [17]. Ag_2BiI_5 precursor solution was prepared by dissolving 281 mg AgI and 354 mg BiI_3 into 1 mL mixed solvent of dimethyl sulfoxide (DMSO) and N,N-dimethylformamide (DMF) with volume ratio of 1:4, wherein 70 μL concentrated hydrochloric acid was added to promote the dissolution of precursors. Then, Ag_2BiI_5 films were prepared by one-step spin-coating method without and with air blowing modification,

respectively. For the former, 60 μL Ag_2BiI_5 precursor solution was spin-coated on FTO/c-TiO₂ substrate at 4500 rpm for 45 s, followed by annealing at 160 °C for 15 min. And, for the latter, the sample after spin-coating process was treated with continuous stream of air gas for 10 min. After that, the film was undergone similar annealing process. Finally, carbon back-electrode (0.09 cm^2) was deposited by screen-printing method, followed by a heating treatment at 110 °C for 15 min. A detailed description for cell measurements can be found in our previous work [17].

3. Results and discussion

Fig. 1(a) illustrates the preparation procedures of Ag_2BiI_5 films by one-step method without and with air blowing modification, respectively. Since air blowing can extract some DMSO and DMF molecules from the fresh Ag_2BiI_5 precursor film [19,20], some crystallized Ag_2BiI_5 species form in the film before annealing, which can be supported by its deepened color after air blowing (**Fig. S1**). Such Ag_2BiI_5 species could act as nucleation sites for Ag_2BiI_5 grains, thus contributing to large-grained Ag_2BiI_5 film with improved surficial coverage. Hereinafter,

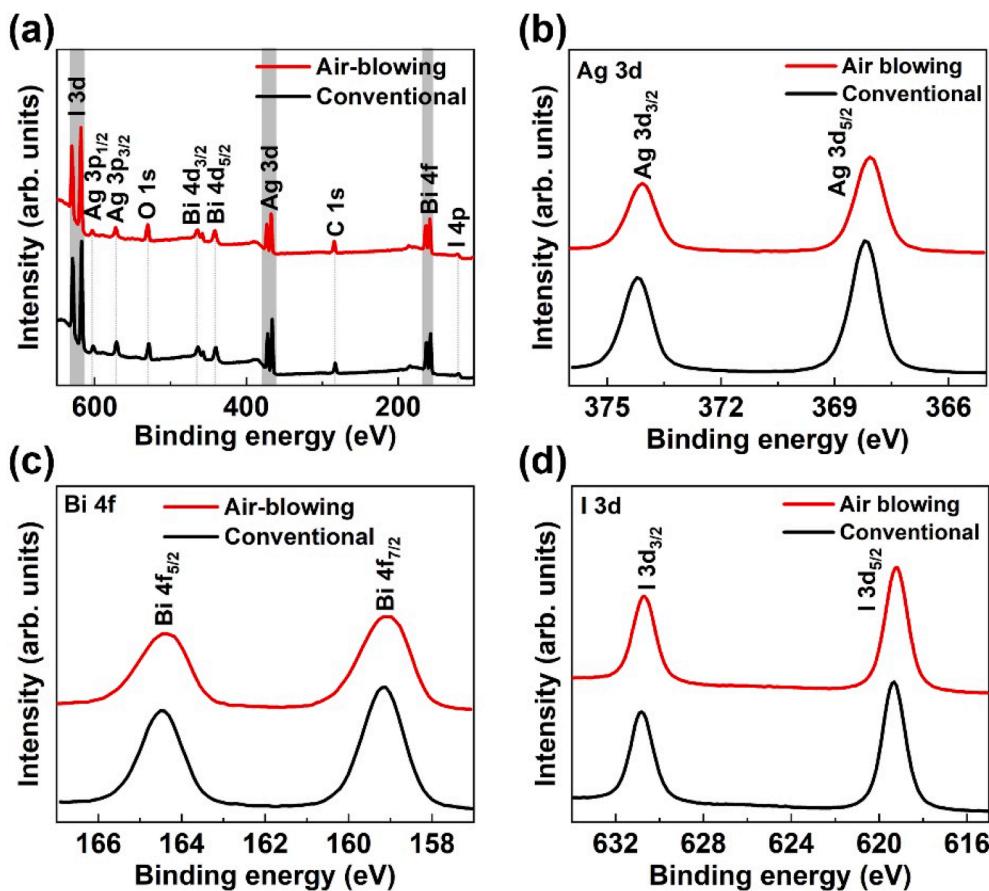


Fig. 2. (a) XPS survey spectra for Ag_2BiI_5 films prepared without and with air blowing modification, respectively. (b-d) Core-level Ag 3d, Bi 4f, and I 3d XPS results for as-studied Ag_2BiI_5 films.

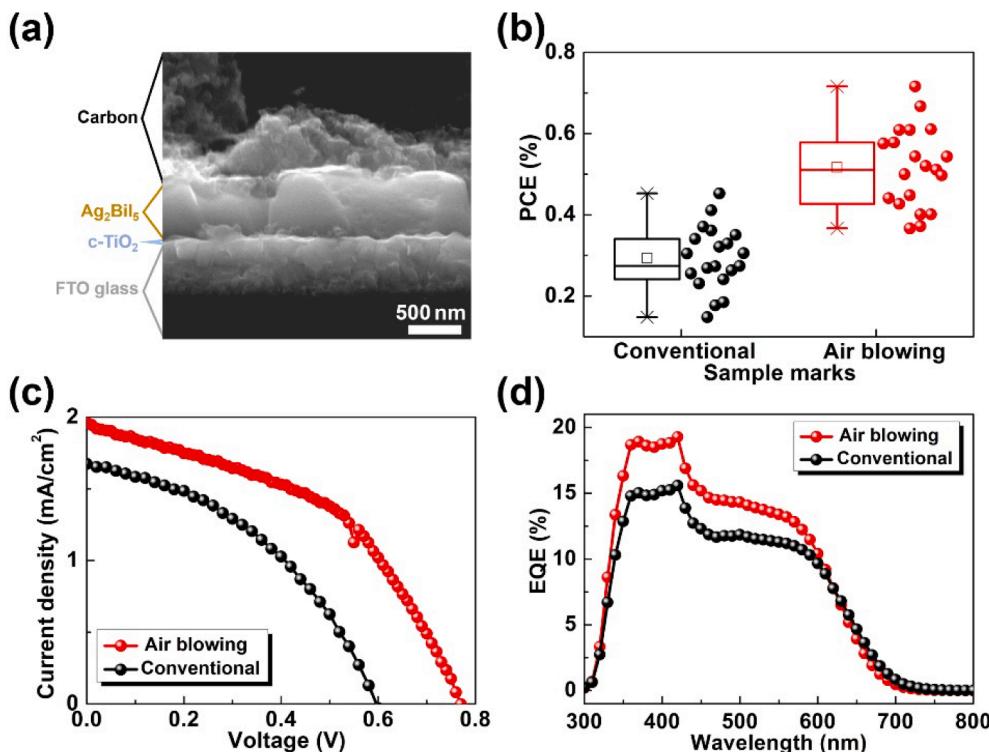


Fig. 3. (a) Cross-sectional SEM image of one typical carbon-based, all-inorganic, lead-free Ag_2BiI_5 ruddruffite solar cell. (b) Statistic PCEs of 20 independent cells fabricated without and with air blowing modification, respectively. (c) J-V curves and (d) EQE spectra of the champion cells.

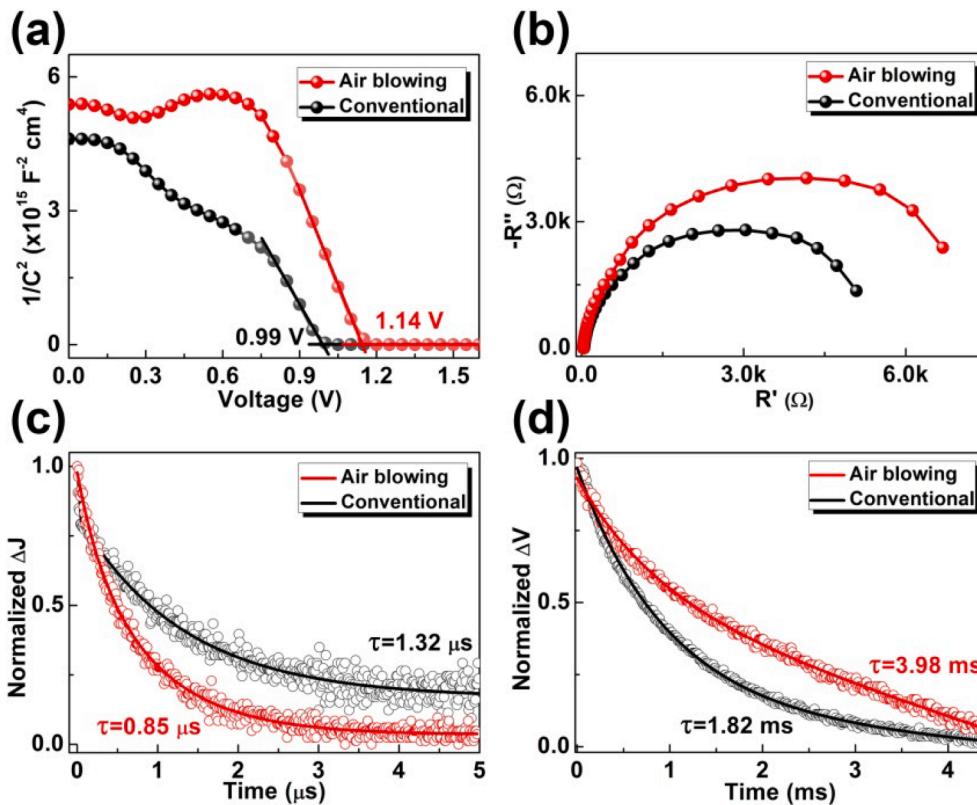


Fig. 4. (a) $1/C^2$ -V results, (b) Nyquist plots, (c) TPC curves, and (d) TPV curves for the best-performing Ag_2BiI_5 solar cells fabricated without and with air blowing modification, respectively.

Ag_2BiI_5 film and solar cell prepared without air blowing modification are labeled as “conventional”, while the ones prepared with air blowing modification are marked as “air blowing” for clarity.

Top-view scanning electron microscope (SEM) images of as-obtained Ag_2BiI_5 films are given in Fig. 1(b-c). The grains with smooth surface are assigned to Ag_2BiI_5 , while the rough regions correspond to impurity phase or pinholes. Therefore, one can see that both films contain some undesirable impurity phase and pinholes. However, they are suppressed substantially in the Ag_2BiI_5 film prepared with air blowing modification. In addition, the grains in it have much larger sizes over micron scale, indicative of the less grain boundaries in Ag_2BiI_5 film prepared through air blowing.

Fig. 1(d) presents X-ray diffraction (XRD) patterns of as-prepared Ag_2BiI_5 films. The main diffraction peaks could be indexed to hexagonal-phase Ag_2BiI_5 ruddruffite [14,21,22], wherein the diffraction peaks corresponding to BiI_3 impurity appear for both films, in accordance with the SEM results. Moreover, it is observed that XRD peak of (003) plane is much higher for Ag_2BiI_5 film prepared with air blowing modification. And, the full-width-at-half-maximum (FWHM) values of (003) diffraction peaks were estimated to be 0.185° and 0.135° for the Ag_2BiI_5 films obtained without and with air blowing modification, respectively. These results signify that the Ag_2BiI_5 film prepared with air blowing modification possesses better crystallinity. Fig. 1(e) provides ultraviolet-visible (UV-vis) absorption spectra of the films. The one prepared without air blowing modification yields low absorption intensities, and its absorption onset is difficult to identify. By contrast, Ag_2BiI_5 film prepared with air blowing modification displays much higher absorption intensities, probably due to reduced pinholes and impurity phase in it as well as improved crystallinity of it. In addition, the absorption onset of $\sim 690 \text{ nm}$ can be identified for the film. As a whole, the above investigations indicate that Ag_2BiI_5 film is formed by one-step method. And, the film’s quality could be further improved by air blowing modification.

Meanwhile, the composition of Ag_2BiI_5 films prepared without and with air blowing modification was studied by X-ray photoelectron spectroscopy (XPS). As shown in Fig. 2(a), the clear signals corresponding to Ag, Bi, and I elements can be detected from both films. Fig. 2(b) indicates that the binding energies of Ag 3d_{3/2} and

Ag 3d_{5/2} are 374.1 and 368.0 eV, respectively. The core-level Bi 4f XPS peaks (Fig. 2(c)) can be resolved into two individual peaks at 164.5 and 159.1 eV, which can be assigned to corresponding to Bi 4f_{5/2} and Bi 4f_{7/2}, respectively. And, one can see from Fig. 2(d) that the core-level I 3d_{3/2} and I 3d_{5/2} XPS peaks locate at 630.9 and 620.3 eV. The above measured binding emerges of Ag 3d, Bi 4f, and I 3d are basically consistent with the Ag_2BiI_5 films reported earlier [15,23]. That is to say, both the as-investigated films are mainly composed of Ag_2BiI_5 . In addition, based on Fig. 2(b-d), the compositional molar ratios of $[\text{Ag} + \text{I}] / [\text{Bi} + 3\text{I}]$ are estimated to be 1.87 and 1.94 for Ag_2BiI_5 films prepared without and with air blowing modification, respectively, which signifies the fewer BiI_3 impurity in the latter one, in accordance with the SEM and XRD results above.

Next, Ag_2BiI_5 films are used for carbon-based, all-inorganic, lead-free ruddruffite solar cells with the simple configuration of FTO/c-TiO₂/ Ag_2BiI_5 /carbon. This configuration avoids use of HTLs and noble-metal electrodes, which not only reduces production costs but also endows reliable stability of the solar cells. Fig. 3(a) gives the cross-sectional SEM image of one typical device, from which each of the functional layers can be identified clearly. Fig. 3(b) provides statistic PCEs of 20 independent cells fabricated without and with air blowing modification. Average PCEs are estimated to 0.30% and 0.52% for the former ones and the latter ones, respectively. These results indicate that carbon-based, all-inorganic solar cells can be realized based on Ag_2BiI_5 , as similar with lead halide perovskites. Namely, Ag_2BiI_5 film could serve as absorber layer and HTL simultaneously. Moreover, the performance of resulting cells could be further improved by boosting the quality of Ag_2BiI_5 films.

Fig. 3(c) gives current density versus voltage (J-V) curves for the

champion cells fabricated without and with air blowing modification, respectively. The former yields a short-circuit current density (J_{sc}) of 1.67 mA cm^{-2} , a V_{oc} of 0.595 V , a fill factor (FF) of 0.42, corresponding to a PCE of 0.41%. By comparison, the J_{sc} , V_{oc} , and FF values for the one fabricated with air blowing modification are improved to 1.96 mA cm^{-2} , 0.770 V , and 0.47, amounting a much enhanced PCE of 0.71%. It is noted that the V_{oc} achieved herein exceeds nearly all of Ag_2BiI_5 rudorffite solar cells reported previously [14,16,20,22]. Their external quantum efficiency (EQE) spectra are shown in Fig. 3(d). The cells exhibit the same response cutoff wavelength of $\sim 690 \text{ nm}$. Similarly, the cell prepared with air blowing modification yields higher EQEs over the whole response ranges, revealing its better photoelectric conversion capability again.

Finally, the Ag_2BiI_5 rudorffite solar cells are analyzed by Mott-Schottky (M-S), electrochemical impedance spectroscopy (EIS), and transient photocurrent/photovoltage (TPC/TPV) technologies, respectively [17,24,25]. Fig. 4(a) gives their $1/C^2$ -V plots, wherein the built-in voltage (V_{bi}) values are fitted to 0.99 and 1.14 V for solar cells fabricated without and with air blowing modification, respectively [25]. The formation of build-in fields accounts for their photovoltaic property, wherein the larger V_{bi} for the one fabricated with air blowing modification explains its higher PCE. Fig. 4(b) provides Nyquist plots of the solar cells. Obvious semicircles are assigned to carrier recombination at interface of $\text{TiO}_2/\text{Ag}_2\text{BiI}_5$, reflecting the recombination resistance of R_{rec} [17,26,27]. Larger R_{rec} for the solar cell fabricated with air blowing modification reveals weaker carrier recombination in it. Fig. 4(c-d) present TPC/TPV curves for as-fabricated solar cells. Faster TPC and slower TPV decays are identified for the one prepared with air blowing modification, further consolidating improved carrier extraction and suppressed carrier recombination in it [17,27,28]. Overall, one can make sense that Ag_2BiI_5 solar cells could be attained based on a simplified carbon-based, all-inorganic configuration. Moreover, their performance may be further boosted by improving the quality of Ag_2BiI_5 films.

4. Conclusions

Lead-free Ag_2BiI_5 rudorffite is applicable for building carbon-based, all-inorganic solar cell that is free of HTL and noble-metal electrode. By improving the quality of Ag_2BiI_5 film with air blowing recipe, the cell's PCE is improved from 0.41% to 0.71%; V_{oc} is boosted from 0.595 V to the record-high value of 0.770 V , because of enhanced carrier extraction and reduced carrier recombination. Although the PCE achieved herein is lower than those of state-of-the-art Ag_2BiI_5 solar cells, the low-cost, simple fabrication, and non-toxic traits make carbon-based, all-inorganic, lead-free Ag_2BiI_5 rudorffite solar cells promising in developing advanced photovoltaic platforms.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.sse.2020.107950>.

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