Supporting Information

CsAg₂Sb₂I₉ Solar Cells

Zhimin Fang,^{‡ab} Shizhe Wang,^{‡bc} Shangfeng Yang^{*a} and Liming Ding^{*b}

^aHefei National Laboratory for Physical Sciences at Microscale, Key Laboratory of Materials for Energy Conversion (CAS), University of Science and Technology of China, Hefei 230026, China. E-mail: sfyang@ustc.edu.cn ^bCenter for Excellence in Nanoscience (CAS), Key Laboratory of Nanosystem and Hierarchical Fabrication (CAS), National Center for Nanoscience and Technology, Beijing 100190, China. E-mail: ding@nanoctr.cn ^cUniversity of Chinese Academy of Sciences, Beijing 100049, China.

‡ Z. Fang and S. Wang contributed equally to this work.

Experimental section

Preparation of the precursor solution

The precursor solution was prepared by dissolving CsI, AgI and SbI₃ in DMF, and keeping AgI and SbI₃ with a concentration of 0.1 M. The solution was stirred at room temperature overnight inside a nitrogen glovebox. All the solutions were filtered with a 0.45 μ m polytetrafluoroethylene (PTFE) filter.

Materials characterization

Absorption spectra for perovskite films on glass were recorded on a Shimadzu UV-1800 spectrophotometer. Wide angle X-ray diffraction (XRD) patterns were obtained on D/MAX-TTRIII (CBO) with Cu K α radiation ($\lambda = 1.542$ Å) operating at 40 kV and 200 mA. Field emission scanning electron microscopy (FESEM) was performed on Hitachi SU-8220 operating at 5.0 kV. Atomic force microscopy (AFM) was performed on a Multimode microscope (Veeco) using tapping mode and a XE-7 scanning probe microscope in noncontact mode (Park Systems, Korea). Film thicknesses were measured with a profilometer (KLA Tencor D-120). XPS was measured on ESCALAB 250Xi.

Device fabrication and measurements

Patterned ITO glass with a sheet resistance of 15 Ω sq⁻¹ was cleaned by ultrasonics in detergent, deionized water, acetone, isopropanol sequentially and then treated with UV-ozone for 10 min. The poly(3,4-ethylenedioxythiophene)-polystyrene sulfonate (PEDOT:PSS, CleviosTM PVP Al 4083) layer was formed by spin coating an aqueous dispersion onto ITO glass (4000 rpm for 30 s). PEDOT:PSS coated substrates were dried at 150 °C for 10 min, and then the substrates were transferred into a N₂ glovebox. Then the perovskite precursor solution was spin-coated onto PEDOT substrates at 3000 rpm for 30 s, and without post-annealing. PC₆₁BM solution (20 mg/mL in chlorobenzene) was then spin-coated onto CsAg₂Sb₂I₉ layer at 1500 rpm for 30 s. Finally, aluminum (100 nm) was deposited onto PC₆₁BM layer through a

shadow mask under vacuum (ca. 10⁻⁴ Pa). The effective area for the devices is 4 mm². *J-V* curves were measured by using a computerized Keithley 2400 SourceMeter and a Xenon-lamp-based solar simulator (Enli Tech, AM 1.5G, 100 mW/cm²). The illumination intensity of solar simulator was determined by using a monocrystalline silicon solar cell (Enli SRC2020, 2 cm×2 cm) calibrated by NIM. The external quantum efficiency (EQE) was measured by using a QE-R3011 measurement system (Enli Tech).



Fig. S1 Tauc plot for CsAg₂Sb₂I₉ film.



Fig. S2 XRD patterns for CsAg₂Sb₂I₉ films prepared with different molar ratios of CsI:AgI:SbI₃.



Fig. S3 XPS spectrum of CsAg₂Sb₂I₉ film.



Fig. S4 SEM images for $CsAg_2Sb_2I_9$ films prepared with different molar ratios of $CsI:AgI:SbI_3$: (a) 0.4:2:2, (b) 0.8:2:2, (c) 1:2:2, (d) 1.2:2:2, (e) 1.6:2:2, (f) 2:2:2, (g) 4:2:2.



Fig. S5 AFM images for $CsAg_2Sb_2I_9$ films prepared with different molar ratios of $CsI:AgI:SbI_3$: (a) 0.4:2:2, (b) 0.8:2:2, (c) 1:2:2, (d) 1.2:2:2, (e) 1.6:2:2, (f) 2:2:2, (g) 4:2:2.

Thickness	$V_{\rm oc}$	$J_{ m sc}$	FF	PCE
[nm]	[V]	[mA/cm ²]	[%]	[%]
122	0.81	1.53	63.94	$0.79 (0.74)^a$
104	0.77	1.91	67.10	0.99 (0.92)
94	0.79	1.72	66.41	0.90 (0.86)
88	0.79	1.62	65.95	0.84 (0.81)

Table S1 Effect of thickness on performance of CsAg₂Sb₂I₉ solar cells.

^aData in parentheses stand for the average PCEs for 10 cells.

Table S2 Effect of annealing temperature on performance of $CsAg_2Sb_2I_9$ solar cells.

Temperature	$V_{\rm oc}$	$J_{ m sc}$	FF	PCE
[°C]	[V]	[mA/cm ²]	[%]	[%]
RT	0.77	1.91	67.10	$0.99 (0.92)^a$
60	0.78	1.83	64.18	0.92 (0.86)
70	0.79	1.78	62.85	0.88 (0.81)
80	0.77	1.74	64.21	0.86 (0.78)
90	0.77	1.61	62.56	0.78 (0.73)

^aData in parentheses stand for the average PCEs for 10 cells.



Fig. S6 *J-V* curves of CsAg₂Sb₂I₉ solar cell under different scans.



Fig. S7 Stable PCE for CsAg₂Sb₂I₉ solar cell.



Fig. S8 The stability of $CsAg_2Sb_2I_9$ solar cell in a N_2 glovebox.