Supporting Information

A Novel Transformation Route from PbS into $CH_3NH_3PbI_3$ for Fabricating Curve and Large-Area Perovskite Films

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1. Experimental details

1.1 Preparation of PbS films by CBD method

PbS films were prepared by a typical CBD method. Firstly, the reaction bath solutions were prepared in a 250 mL beaker by the sequential addition of 100ml deionized water, 20 mL 0.015 M $Pb(NO)_2$, 10 mL 1.5 M $SC(NH_2)_2$ and 26 mL ammonium hydroxide, and then adding deionized water to 200 ml. Secondly, the substrate was putted into the bath solution beaker, and then leaded in a water bath pot. After reaction at 65 ^oC for 10 minutes, the shining black films were deposited. Finally, the as-prepared films were washed with deionized water and dried, and the desired PbS films were obtained.

1.2 Preparation of $CH_3NH_3PbI_3$ absorbers by CVD method

The CH_3NH_3I powder and TiO_2/FTO glass substrate were prepared as our previously reported method.[1] The $CH_3NH_3PbI_3$ absorbers were fabricated by a typical tubular chemical vapor deposition (CVD) method. In brief, the obtained PbS films, surrounding by 2 g CH_3NH_3I powder without physically touching the films, were placed into the quartz tube with a dimension of 60 mm$\times$1000 mm. After rushing by N_2 for three times, the quartz chamber is sealed and pumped by mechanic pump for 10 minutes, then slowly heated to 145 ^oC with a heating rate of 2.4 ^oC/minute and reacted for 100 minutes, and finally natural cooling to room temperature. As such, the black PbS films transformed into and dark brown $CH_3NH_3PbI_3$ absorbers.

1.3 Preparation of PSCs

The PSCs with a structure of FTO/C-TiO2/$CH_3NH_3PbI_3$/Spiro-MeOTAD/Ag were sequentially

fabricated by our previous reported procedures.[1,2] The first batch of 5 cells was successfully prepared in this work and their active area is 0.12 cm^2.

1.4 Characterizations of the as-perpared films by VACBD method

X-ray diffraction (XRD) was measured by using X-ray diffractometer (X'Pert PRO MPD) with Cu Kα as the radiation source. Raman spectroscopy was measured by a Focal Laser MicroRaman Spectrometer (LABRAM-HR) with an excitation 514.5nm. UV-Vis absorption was measured on UV-visible spectrophotometer (CARY 5000) with a wavelength ranging from 350 to 1500 nm. The morphology of the PbS and $CH_3NH_3PbI_3$ films were investigated by the field-emission scanning electron microscope (FESEM, Sirion200) and atomic force microscope (AFM, Dimension Fast Scan). XPS analysis was carried out using the VG ESCALAB250 surface analysis system equipped with a monochromatic Al X-ray (1486.6 eV) source. *J-V* curves (reverse scan from 1.2V to -0.1V, with a step voltage of 10 mV) were detected with a Keithley 2400 source meter under illumination of 100mW/cm^2, AM1.5 by a solar simulator (Newport, Oriel Sol3A 94023A) in air ambient.

References:

1. P. Luo, Z. Liu, W. Xia, C. Yuan, J. Cheng and Y. Lu, *ACS Appl. Mater. Interfaces*, 2015, **7**, 2708-2714.
2. P. Luo, Z. Liu, W. Xia, C. Yuan, J. Cheng and Y. Lu, *J. Mater. Chem. A,* 2015, **3**, 22949-22959.

2. Tab. S1.

Tab. S1. Device performance of PSCs.

Device	V_{oc} (V)	J_{sc} (mA/cm^2)	FF	PCE(%)
*$CH_3NH_3PbI_3$	0.677±0.066	17.058±2.847	27.84±3.64	3.28±1.41

*Each value represents the average from 5 cells.

3. Fig. S1-S10

Fig. S1. Our industrial pilot-scale CBD equipment (Deposition area: 30cm×30cm, Singulus Stangl Solar GmbH).

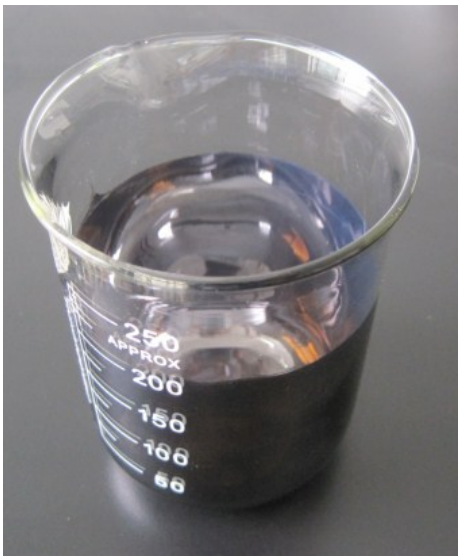

Fig. S2. The photograph of PbS films deposited on beaker by CBD method. The shining black films are uniformly grown on a 250 mL beaker, which indicates that CBD method could deposit the desired PbS precursor films on substrate with all kinds of shape.

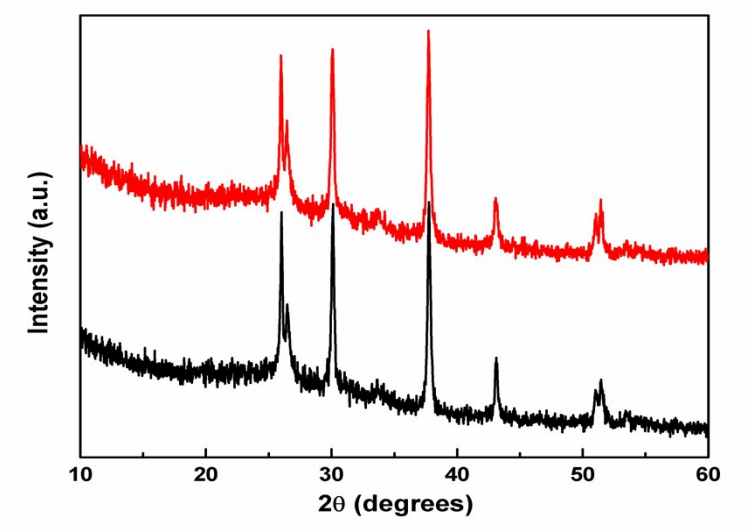

Fig. S3. XRD patterns of the PbS films (dark line) and the PbS films after dipping in a solution of CH_3NH_3I in 2-propanol (10mg/ml) (red line). There is no change in the character of the main peaks of PbS films after dipping treatment, and the (111), (200), (210) and (311) planes still separately locate at 26.02°, 30.12 °, 43.12°, and 51.02°, which means PbS films by CBD cannot react with CH_3NH_3I solution at room temperature. Inversely, the spin-coated PbI precursor can rapidly react with it.

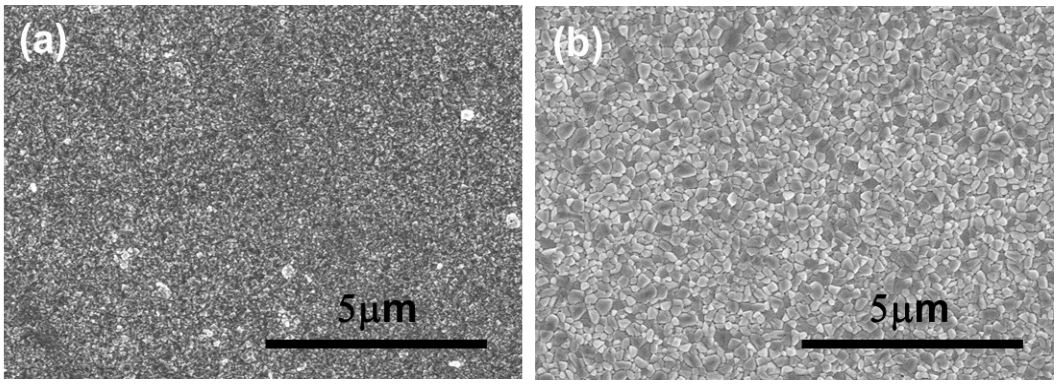

Fig. S4. The low-magnification SEM image of (a) PbS films and (b) $CH_3NH_3PbI_3$ absorbers. From above images we can see that we obtain smooth, dense, and uniform PbS films and $CH_3NH_3PbI_3$ absorbers via VACBD method.

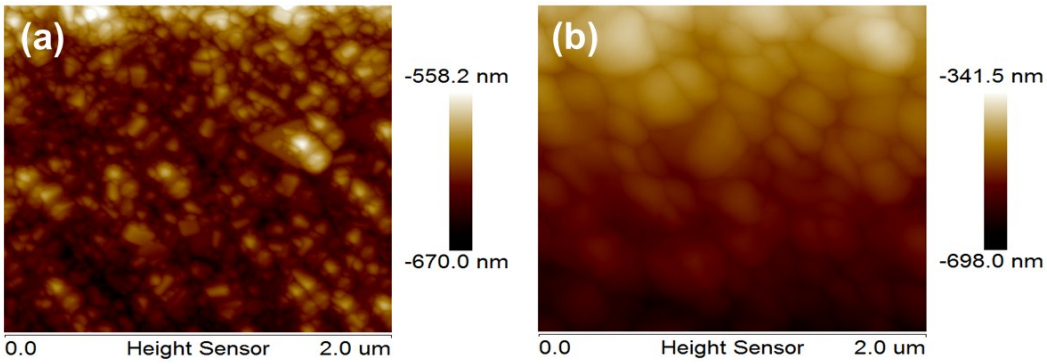

Fig. S5. The 2-D AFM height image of (a) PbS films and (b) $CH_3NH_3PbI_3$ absorbers. The uniform PbS grains with a size of ~100 nm grow up to above 300 nm after CVD process under CH_3NH_3I atmosphere.

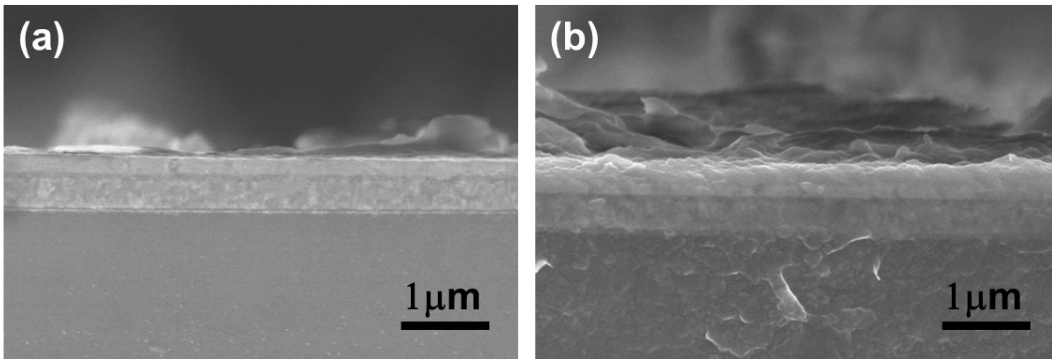

Fig. S6. The cross-sectional SEM image of (a) PbS films and (b) $CH_3NH_3PbI_3$ absorbers. We can observe that uniform PbS and $CH_3NH_3PbI_3$ films are deposited on the TiO_2/FTO glass substrate, and the film thickness tremendously expands up from ~250 nm to ~500 nm after the transformation.

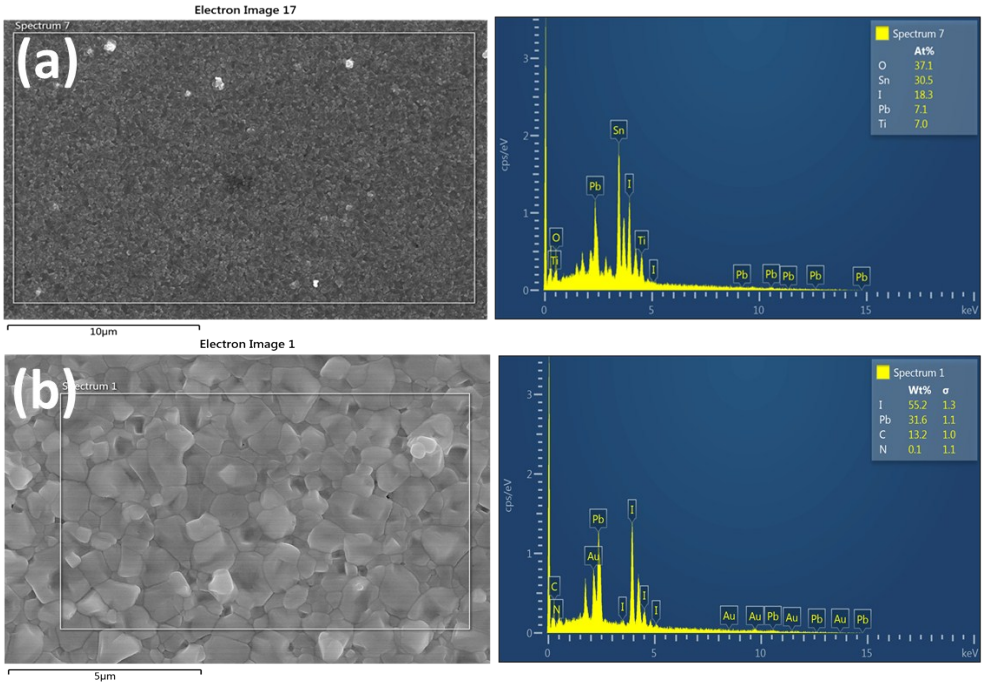

Fig. S7. SEM-EDS images of the final $CH_3NH_3PbI_3$ absorbers. The I/Pb ratios of $CH_3NH_3PbI_3$ absorbers are separately 2.58 : 1 (a: large area) and 2.85 : 1 (b: small area), with a little I-poor composition compared to the stoichiometry 3 : 1, which should be the decomposition of perovskites to PbI_2. Meanwhile, we could not find the peak of S element in the final $CH_3NH_3PbI_3$ films.

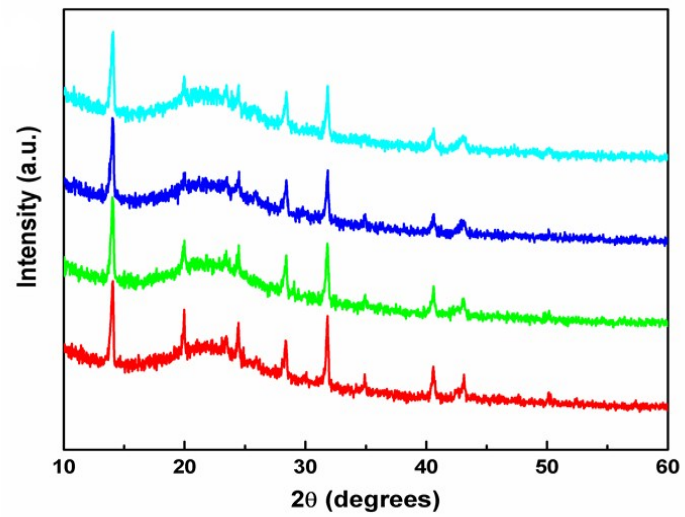

Fig. S8. XRD patterns of curve and large-area perovskite films (4 cm × 3 cm) measured at 4 different points. Here we can see that all the patterns show the same perovskite structure and there is no obviously difference among the four patterns, which exhibits a good uniformity of crystallization.

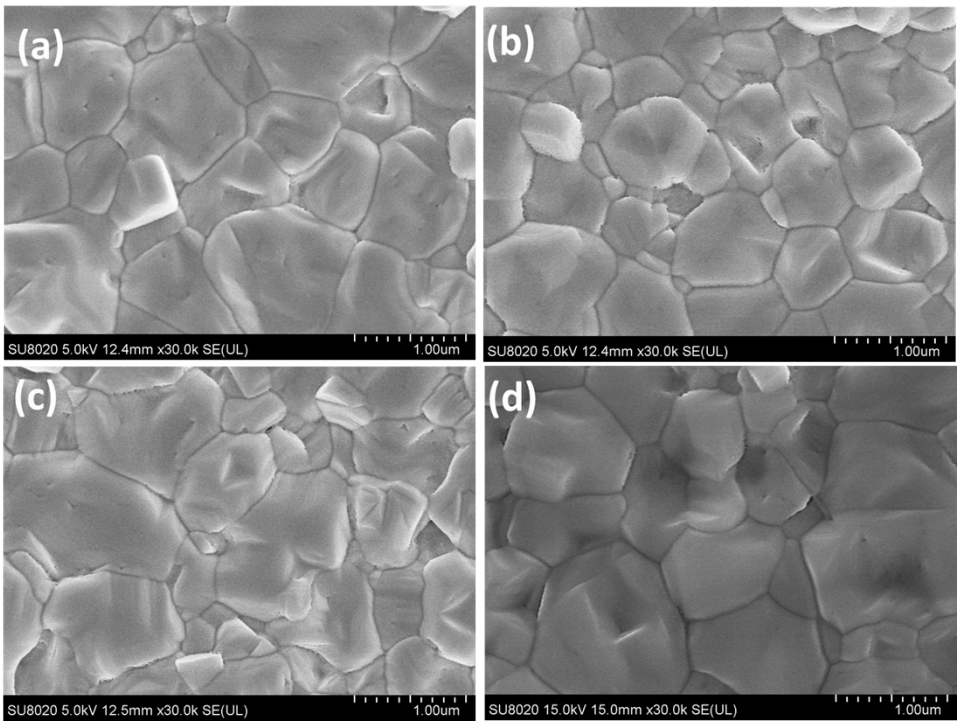

Fig. S9. Planar SEM images of curve and large-area perovskite films (4 cm × 3 cm) measured at 4 different points. Here we can observe that $CH_3NH_3PbI_3$ absorbers fabricated by VACBD demonstrate a smooth and compact appearance in large area.

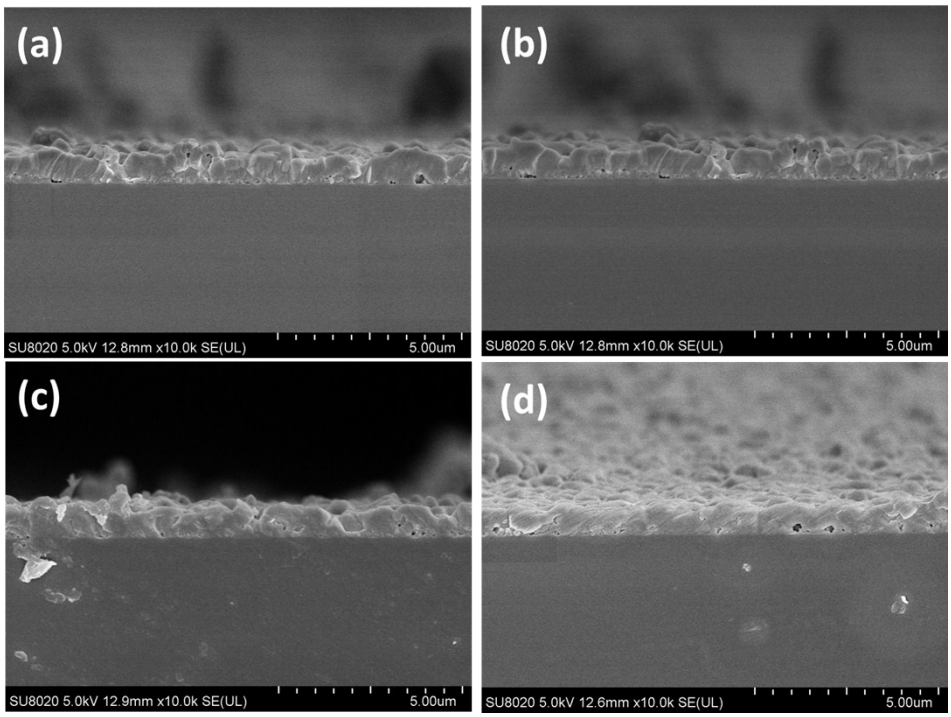

Fig. S10. Cross-sectiona SEM images of curve and large-area perovskite films (4 cm × 3 cm) measured at 4 different points. It is clear that $CH_3NH_3PbI_3$ absorbers fabricated by VACBD almost have the same thickness.