Supporting Information for:

Size-controlled SiO_2 nanoparticles as scaffold layers in thin-film perovskite solar cells

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((Optional Dedication))

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Experimental

Synthesis of the SiO_2 nanoparticles

Uniform SiO_2 NPs with diameters of 15, 30, 50, 70, and 100 nm were synthesized using a sol-gel method. A mixture of 79 mL of ethanol, 2.8 ml of de-ionized (DI) water, and 7.2 mL of ammonia solution was stirred for 12 hours. Then, 2.9 ml of tetraethyl orthosilicate (TEOS) was added with systematically varied temperatures. The diameter of the SiO_2 NPs was controlled by varying the temperature during the reaction, and increased with the temperature. A colloidal dispersion of the SiO_2 NPs was isolated using a centrifuge at 12000 rpm for 40 min, washed, and re-dispersed in 50 mL of DI water.

Fabrication of the perovskite solar cells

A 30-ml hydroiodic acid (HI) solution (0.227 mol, 57 wt% in water, Aldrich) was added to 27.86 ml of methylamine (0.273 mol, 40 wt% in methanol, Aldrich) and stirred at 0°C for 2 hours in a 250-mL round flask. Crystallization of methylammonium iodide (CH_3NH_3I) was achieved using a rotary evaporator at 50°C for 1 hour, resulting in a brown CH_3NH_3I powder. The CH_3NH_3I was washed three times using diethyl ether, and dried in a vacuum oven for 24 hours. A 60-wt% solution of CH_3NH_3I:$PbCl_2$ (99.9%, Aldrich) with a molar ratio of 3:1 was prepared in dimethyl sulfoxide (DMSO, Aldrich).

Patterned fluorine-doped tin dioxide (FTO) glass electrodes were cleaned by sonication in DI water, acetone, and 2-propanol for 60 min. The compact TiO_2 layer was prepared by coating 0.15-M titanium diisopropoxide bis(acetylacetonate) in 1-butanol solution (75 wt% in isopropanol, Aldrich) onto the FTO glass using a spin-coater at 2500 rpm, followed by heating at 125°C for 5 min on a hot plate. The same steps were repeated twice with a 0.3-M solution of titanium diisopropoxide bis(acetylacetonate) in 1-butanol solution. The coated electrode was heated at 450°C for 30 min. A scaffold layer with a thickness of 230 ± 30 nm was deposited using spin coating. The TiO_2-based scaffold layer was coated using commercially available TiO_2 paste (Dyesol 18NR-T) diluted in terpineol (where the TiO_2:terponeol ratio was 1:2) using a spin-coater. The perovskite precursor solution was infiltrated into the scaffold electrode layer using spin coating at 3000 rpm for 5 min. The electrode was then heated to 100°C for 60 min.

A cholorobenzene solution containing 72.3 mg/mL of Spiro-OMeTAD, 28.8 μL of tert-butylpyridine, and 17.5 μL of a solution composed 520 mg of lithium bis(trifluoromethylsyfonyl)imide salt in 1 mL of acetonirtile was cast onto the perovskite-coated electrode using a spin-coater at 3000 rpm for 45 s. A 60-nm-thick Au layer was then evaporated onto the prepared electrode. The active area of the device was 0.09 cm^2.

Characterization

The morphology of the perovskite infiltrated into scaffold layer was characterized using an FE-SEM (6700; JEOL, Tokyo, Japan) equipped with energy dispersive X-ray spectroscopy (EDS). X-ray diffraction (XRD) data were measured using an M18XHF-SRA (Mac Science Co., Yokohama, Japan) with a Cu Kα radiation source (λ = 1.5406 Å) at 40 kV and 300 mA (12 kW). A Lamda 35 (Perkin-Elmer) was used to provide UV–vis spectra. Photoluminesence (PL) was measured using the PerkinElmer LS 45 Fluorescence spectrometer (excitation at 525 nm)

The capacitance and resistance of devices were measured from 1 MHz to 1 Hz using a Zahner Electrik IM6 analyzer. The photocurrent was characterized under illumination with a 500-W xenon lamp (XIL model 05A50KS source units), which provided an AM 1.5G source with an intensity of 100 W cm^{-2}.

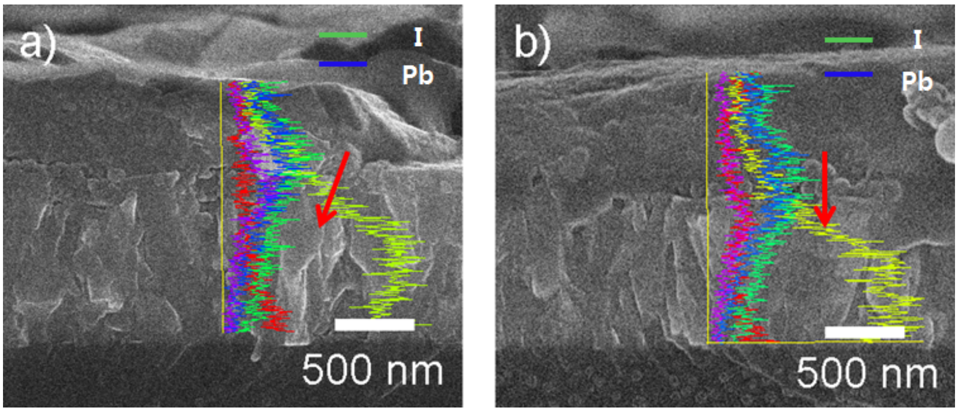

Figure S1. SEM cross-sectional images showing elemental line mapping of the scaffold layer containing SiO_2 NPS with diameters of (a) 15 nm and (b) 100 nm.

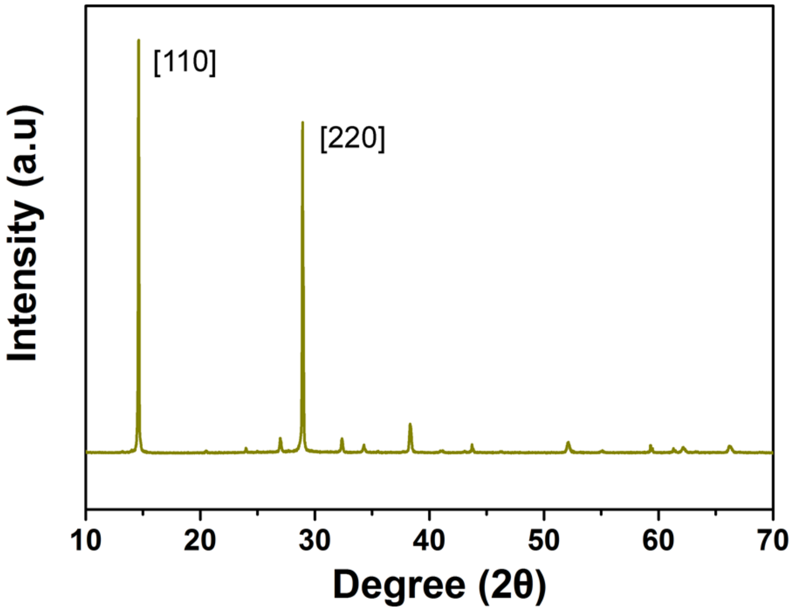

Figure S2. X-ray diffraction pattern of the perovskite layer on a SiO_2-based scaffold layer.

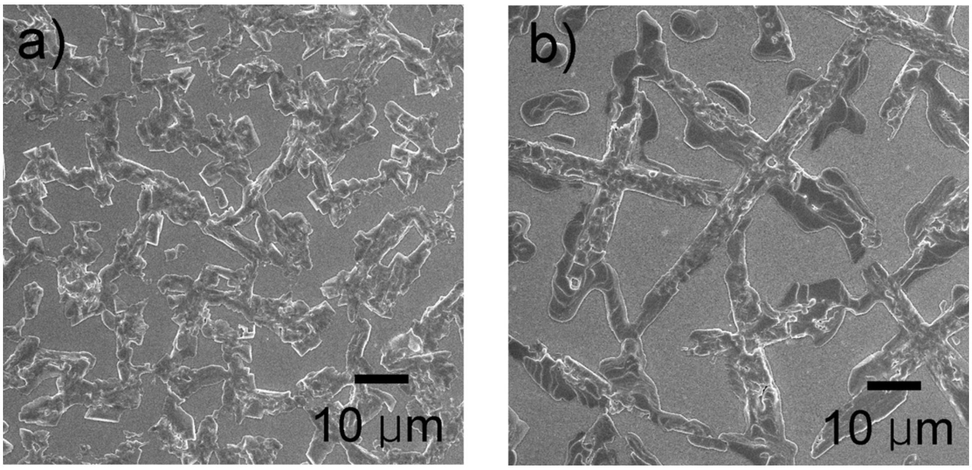

Figure S3. SEM images of the perovskite layers deposited on SiO_2 NPs with diameters of (a) 15 nm and (b) 100 nm**.**

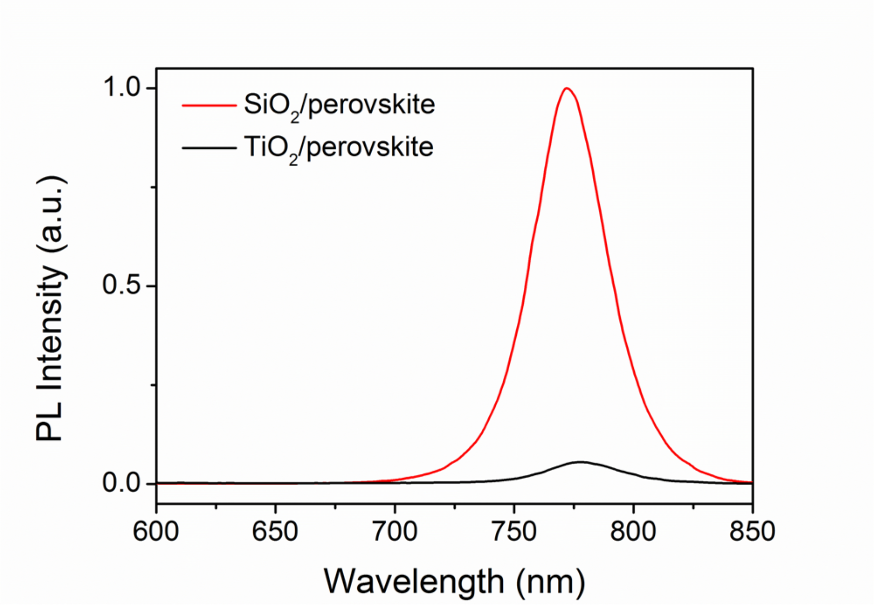

Figure S4. Photoluminesence spectroscopy of SiO$_2$/perovskite film and TiO$_2$/perovskite film photoexcited at 525 nm.