Electronic Supplementary Information

Hole-Transport-Material-Free Perovksite Solar Cells Based on Nanoporous Gold Back Electrode

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Materials

Hydroiodic acid (57 wt % in water, 99.99%) and methylamine (33 wt % in absolute ethanol) were purchased from Sigma-Aldrich. N,N-dimethylformamide (DMF, anhydrous 99.5%, Aladdin) and lead iodide (PbI2) (99.999%) were purchased from Aladdin Reagents. Titanium diisopropoxide bis(acetylacetonate) solution (75% in 2-propanol) was purchased from Alfa Aesar. All of the used reagents were analytical grade, without further purification. The 12 carat 100 nm thick white-gold decorative films with 1:1 weight ratio of Ag/Au was purchased from Seep Leaf Products (New York), which used as the raw material of NPG. CH_3NH_3I was synthesized according to a literature.[1]

NPG fabrication

The Ag/Au alloy leaf was first tailored into suitable size. Then, the small leaf was transferred to deionized water to conserve using a glass. Transfer the leaf from deionized water to 68% HNO_3 by a glass and etched in HNO_3 for 20 min. After that,

the NPG was obtained and then transferred to deionized water to remove the residual $HNO_3$for several hours before use.

Solar cells fabrication

The fluorine-doped tin oxide (FTO) glass was etched by Zn powder and 10% HCl. Then the FTO was ultrasonic cleaned with detergent, deionized water, ethanol and acetone. After that the glass was treated in an ultraviolet-ozone cleaner for 30 min. A compact TiO_2 layer was prepared on the cleaned FTO glass by aerosol spray pyrolysis using a commercial titanium diisopropoxide bis (acetylacetonate) solution (75% in 2-propanol) diluted in ethanol (1:25, volume ratio) at 450 ^oC, and annealed at 500 ^oC for 30 min. After cooling down to room temperature, a mesoporous TiO_2 layer was spin-coated at 5,000 r.p.m for 30 s using commercial TiO_2 paste diluted in ethanol at weight ratio 2:7. The film was dried at 125 ^oC for 30 min and then heated at 500 ^oC for 30 min. An Al_2O_3 blacking layer was spin-coated on the mesoporous TiO_2 at 5000 r.p.m for 60 s using a commercial Al_2O_3 paste, the annealing process is the same as mesoporous TiO_2. After that, the NPG was transferred onto the as-prepared substrate by using the substrate instead of the glass to salvage. Before deposition $CH_3NH_3PbI_3$, the substrate was dried for several hours to remove the water completely. $CH_3NH_3PbI_3$ were infiltrated in the pores by different ways. For one-step spin-coating deposition, 1 mol/L $CH_3NH_3PbI_3$ in N,N-dimethylformamide (DMF) was load on the substrate for 60 s, and then spinning at 3000 r.p.m for 30 s. The device was completed after annealing at 90 ^oC for 30 min. For sequential deposition and two-step spin-coating process, 1 mol/L PbI_2 solution in DMF was loaded on the substrate for 30 s, then spinning at 4500 r.p.m for 60 s at 70 ^oC, dried at 90 ^oC for 5 min. After cooling to room temperature, the substrate was dip in 12 mg/ml CH_3NH_3I solution in isopropanol for 2 min or 12 mg/ml CH_3NH_3I solution in isopropanol was loaded on the PbI_2 for 20 s, then spinning at 2000 r.p.m for 30 s, respectively. After that, the sample was annealed at 90 ^oC for 30 min.

Characterization

X-Ray diffraction (XRD) patterns were collected on a Rigaku Ultima Ⅲ X-ray diffractometer (Cu Kα) in the range of 10^{o}-60^{o}. A FEI NOVA NanoSEM230 scanning electron microscopy (SEM) was employed to characterize the top surface morphological of all samples. The J –V testing of the HTM-free perovskite solar cells employed a Keithley 2400 source measure unit under AM 1.5 illumination cast by an Oriel 92251A-1000 sunlight simulator

Notes and references

1. J. H. Im, C. R. Lee, J. W. Lee, S. W. Park and N. G. Park, *Nanoscale*, 2011, **3**, 4088-4093.