Electronic Supplementary Information

Highly flexible, high-performance perovskite solar cells with adhesion promoted $AuCl_3$-doped graphene electrodes

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Experimental Section

Preparation of $AuCl_3$ doped graphene ($AuCl_3$-GR), and $AuCl_3$-GR/APTES.

Single layer graphene sheets were grown on Cu foils by chemical vapor deposition and subsequently transferred onto PET and APTES/PET substrates. The APTES solution was deposited on the PET substrate prior to transferring the graphene under 3000 rpm for 60 s and then dried at room temperature. Subsequently, APTES/PET substrate was washed with methanol. The graphene/substrate stack was prepared by conventional transfer method. Gold chloride ($AuCl_3$) powder was dissolved in nitromethane to prepare $AuCl_3$ solution at 7.5 mM. For doping of graphene, the solution was dropped on the whole surface of the graphene sheet, and after 1 min elapsed, it was spin-coated at 2500 rpm for 1 min.

Characterization of electrodes.

Hall-effect measurements were performed in an apparatus (Ecopia model HEM-2000) by using the van der Pauw method for ITO, $AuCl_3$-GR, and $AuCl_3$-GR/APTES electrodes. All Hall measurements were done in a dark room. The transmittance and sheet resistance of three electrodes were measured by ultraviolet-visible-near IR optical spectrometer (Agilent Model) and the 4 probe van der Pauw method and its surface morphology was analyzed by a SEM. (Carl Zeiss model merlin) The atomic bonding states of the graphene/APTES and graphite/APTES were characterized by XPS and FTIR. The mechanical properties of the three electrodes samples were evaluated using a specially designed inner and outer bending system.

The change in resistance of the three films due to substrate bending can be expressed as $\Delta R = (R-R_0)/R_0$, where R_0 is the initial measured resistance, and R is the resistance measured under substrate bending.

Fabrication of perovskite solar cells.

To fabricate the inverted $MAPbI_3$ and $FAPbI_{3-x}Br_x$ perovskite planar hybrid solar cells, poly- (3,4-ethylenedioxythiophene):poly(styrenesulfonic acid) (PEDOT:PSS, Clevios, Al4083)/ methanol (1 : 1 v : v) (PEDOT:PSS) was spin coated on an $AuCl_3$-Gr/APTES/glass substrate at 3000rpm for 60 and then annealed at 150 ^oC for 20 min. For $MAPbI_3$ perovskite layer, we prepared the 40 wt% $MAPbI_3$ perovskite solution by mixing the methylammonium iodide powder (MAI, DS LOGICS CO., LTD.) and lead (II) iodide (PbI_2, Aldrich) (1:1 mol:mol) in *N,N*-dimethylformamide (DMF, Aldrich) at 60 ^oC for 30 min, and 100 μL hydriodic acid (HI, Aldrich) was added in 1mL $MAPbI_3$ perovskite solution at room temperature. The 40wt% $MAPbI_3$ perovskite solution was spin-coated on the PEDOT:PSS/$AuCl_3$-Gr/APTES/glass substrate at 3000 rpm for 120 s , and then was dried on the hot plate at 100 ^oC for 2 min. For $FAPbI_{3-x}Br_x$ perovskite film, we prepared $PbI_2(DMSO)_2$ complex by dissolving 50 g PbI_2 in 150 mL dimetylsulfoxide (DMSO, Aldrich) at 60 ^oC for 30 min, and then 350 mL toluene slowly dropped into the PbI_2 solution. The white precipitate was filtered and was annealed in vacuum oven at 60 ^oC for 5h to obtain PbI_2(DMSO) complex. 1 M PbI_2(DMSO) complex was dissolved in DMF at 60 ^oC for 5 min. The PbI_2(DMSO) complex solution was coated by spin coating at 3000 rpm for 30 s followed by spin-coating of 0.5 M FAI and MABr (0.85:0.15 mol:mol) mixture solution in *iso*-propanol (IPA, Aldrich) at 5000 rpm for 30 s. The film tuned from clear to dark brown during spin-coating was dried on the hot plate at 150 ^oC 20 min. After preparing the perovskite film, a phenyl-C61-butyric acid methyl ester (PCBM, nano-C)/1,2-dichlorobenzen (1,2-DCB) solution (20 mg/1 mL) or NDI-BN/1,2-DCB solution (15 mg/ 1mL) was coated on the perovskite film/PEDOT:PSS/ITO substrate by spin-coating at 3000 rpm for 60 s. Finally, Al counter electrode was deposited by thermal evaporation. The active area was fixed 0.16 cm^2. All device fabrication was conducted below relative humidity of 25%

Characterization of solar cells.

The photovoltaic properties of the cells were measured using a solar simulator (Peccell, PEC-L01) under illumination of 1 Sun (100 $mWcm^{-2}$ AM 1.5G) and a calibrated Si reference cell certificated by Japanese Industrial Standards. To measure the hysteresis of J-V curves, the forward and reverse scan rate was set to 10 mV·200 ms^{-1} as a standard condition. The J-V curves of all devices were measured by masking the active area with metal mask of 0.096 cm^2. The external quantum efficiency (EQE) was measured by a power source (ABET 150 W xenon lamp, 13014) with a monochromator (DongWoo Optron, MonoRa-500i).

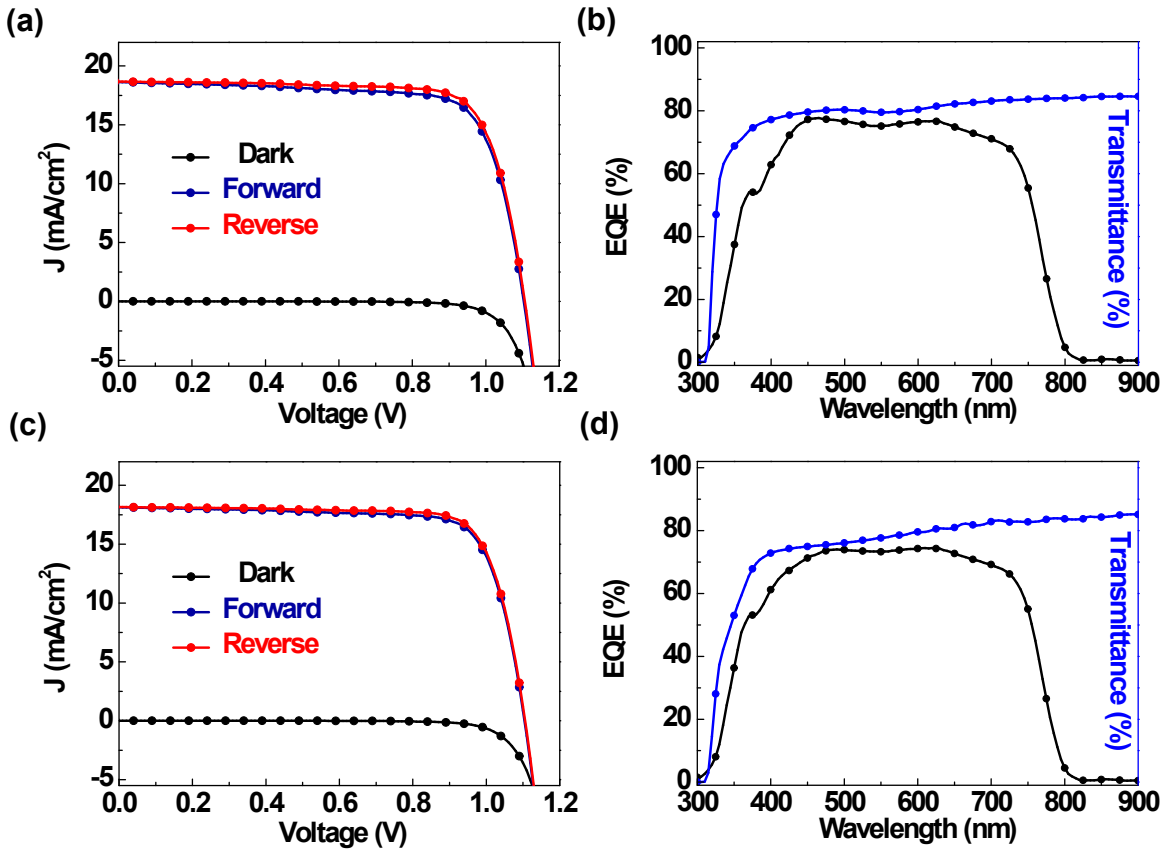


Figure S1. The J-V curves of $MAPbI_3$ perovskite solar cells with (a) $AuCl_3$-GR and (c) ITO TCE under dark and illumination, respectively; EQE and transmittance spectra of the solar cells for (b) $AuCl_3$-GR and (d) ITO TCE.

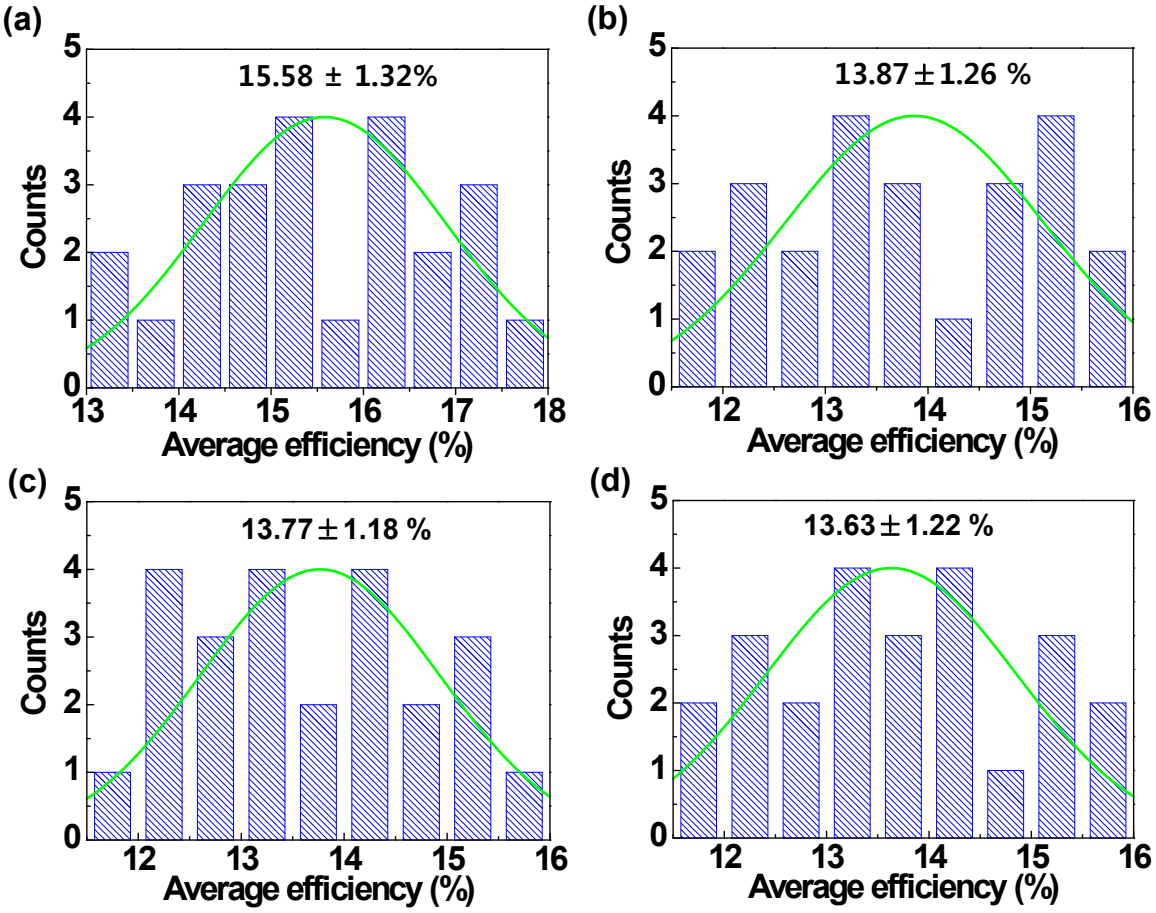


Figure S2. Statistical deviations of the average PCE for 24 solar cell devices with (a) $FAPbI_{3-x}Br_x$ and (b) $MAPbI_3$ with $AuCl_3$-GR/APTES, (c)$AuCl_3$-GR, (d) and ITO TCEs.

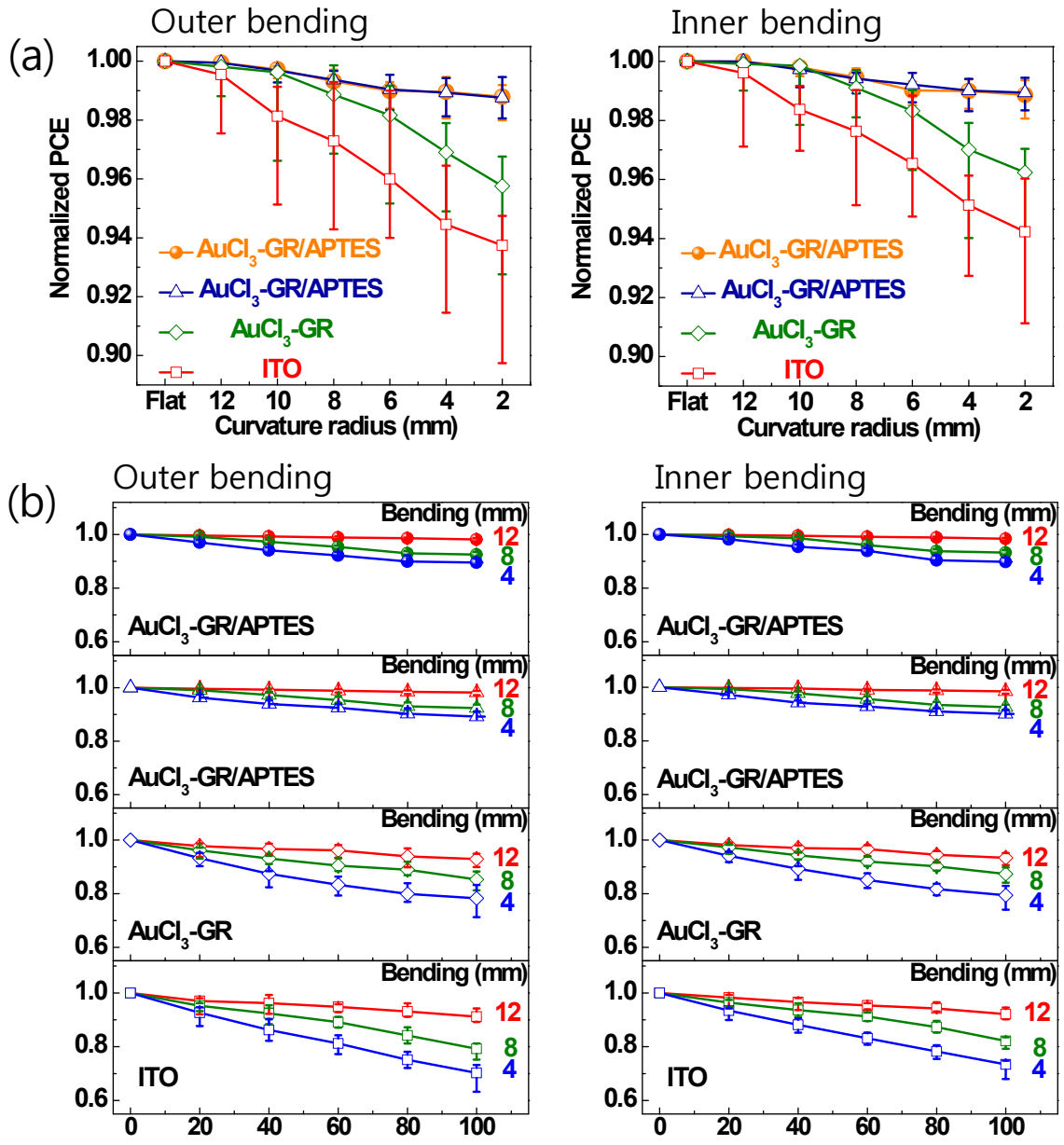


Figure S3. (a, b) Normalized PCEs of the flexible solar cells with respect to (a) the bending curvature and (b) bending cycles at curvature radius = 12, 8, and 4 mm for ITO, $AuCl_3$-GR, and $AuCl_3$ GR/APTES TCE on a PET substrate under outer and inner bending condition: ●: $FAPbI_{3-x}Br_x$ and △,◇,□: $MAPbI_3$.

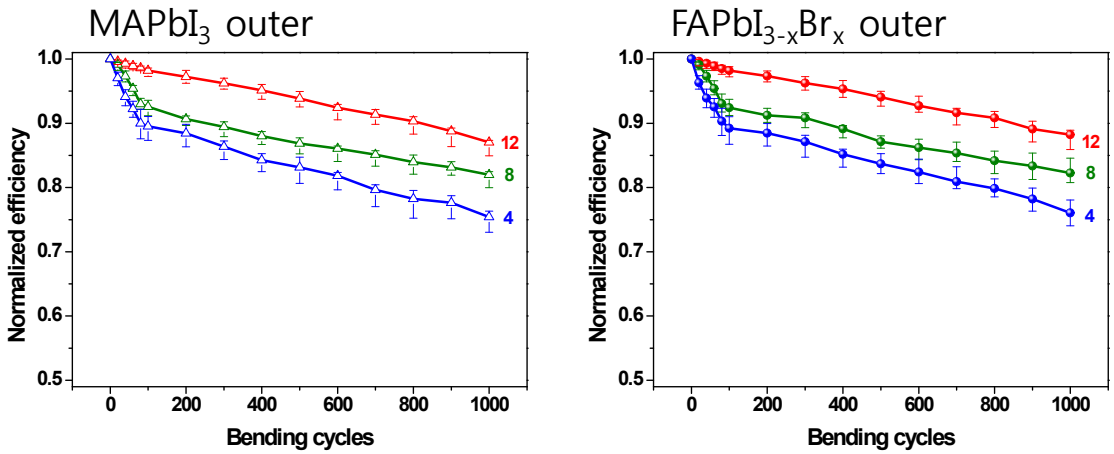
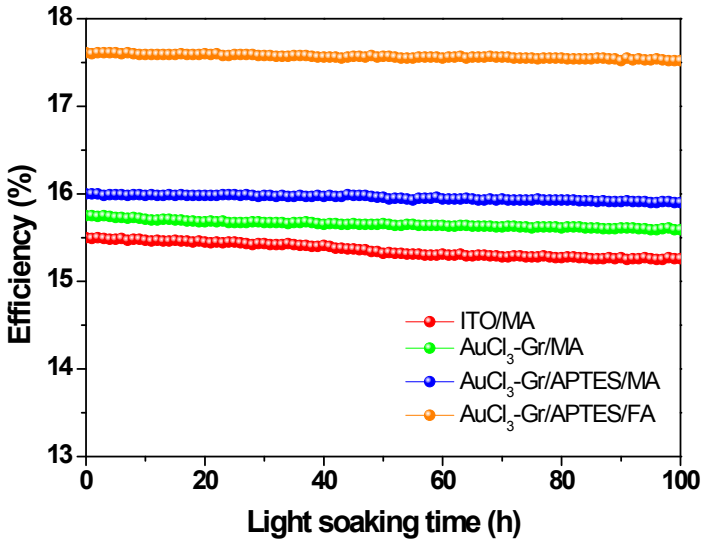


Figure S4. Normalized PCEs of the flexible solar cells with respect to the bending cycles at curvature radius = 12, 8, and 4 mm for ITO, $AuCl_3$-GR, and $AuCl_3$ GR/APTES TCE on a PET substrate under outer and inner bending condition: ●: $FAPbI_{3-x}Br_x$ and △,◇,□: $MAPbI_3$.



Figyre S5. Stability of ITO/$MAPbI_3$, $AuCl_3$-GR/$MAPbI_3$, $AuCl_3$-GR/APTES/$MAPbI_3$, and $AuCl_3$-GR/APTES/$FAPbI_{3-x}Br_x$ based unencapsulated inverted perovskite solar cells under relative humidity of 50 % at 1Sun illumination for 100 h.