Supplementary information

Highly efficient perovskite solar cells with a compositionally engineered perovskite/hole transporting material interface

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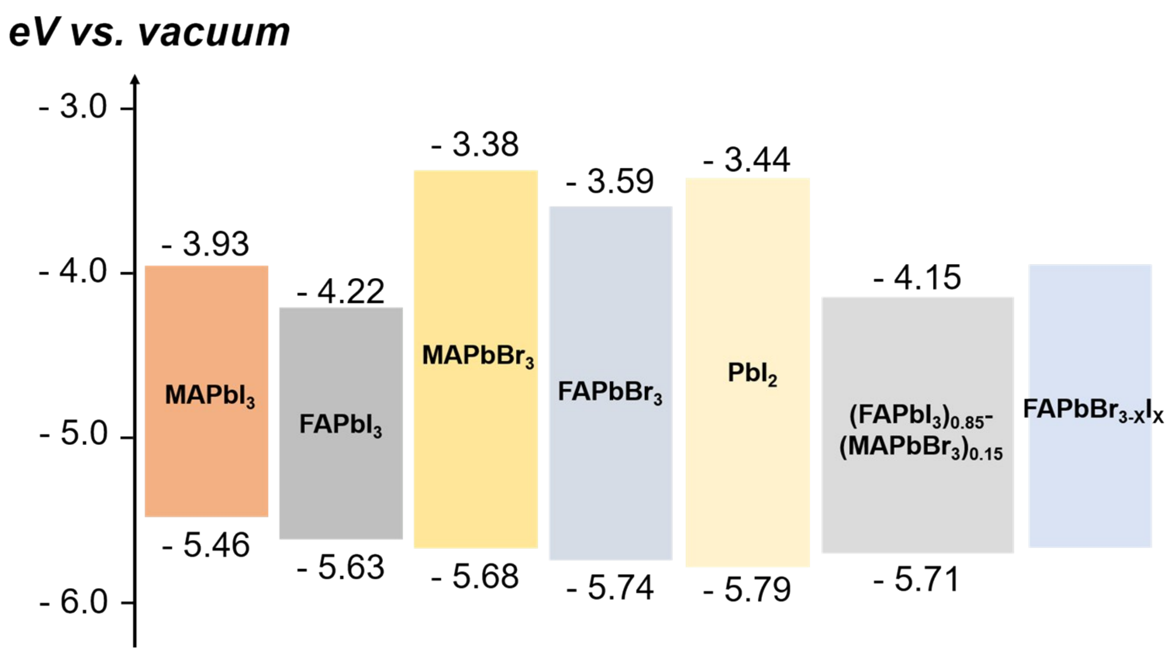


Figure S1. Estimated bandgap edge position of $(FAPbI_3)_{0.85}(MAPbBr_3)_{0.15}$ and $FAPbBr_{3-x}I_x$. Energy levels for $FAPbI_3$, $FAPbBr_3$, $MAPbBr_3$ and PbI_2 are referred from previous papers using photoelectron spectroscopy in air (PESA)[1-3] and $(FAPbI_3)_{0.85}(MAPbBr_3)_{0.15}$ is calculated approximately by a reference from ultraviolet photoelectron spectroscopy[4].

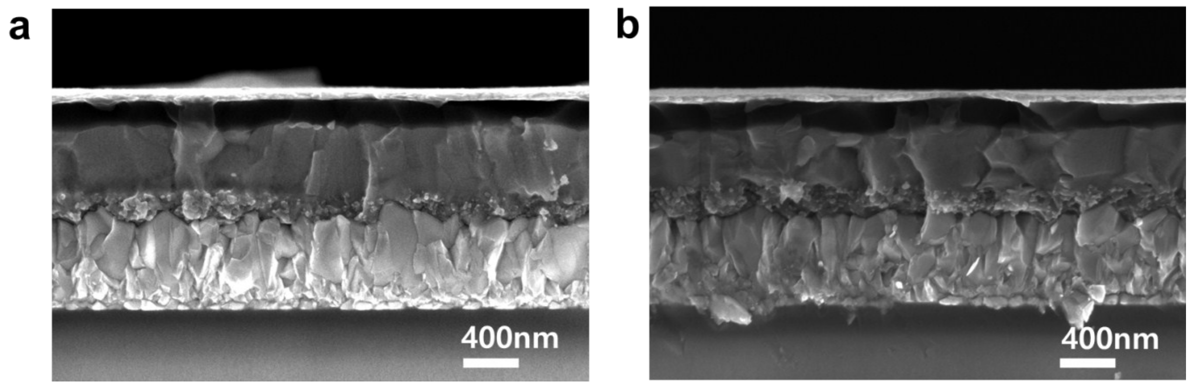


Figure S2. Cross-sectional SEM images of the complete perovskite solar cells. a, From an as-prepared perovskite film. **b**, From a passivated perovskite film. No obvious difference was observed between devices, which indicates that the passivation layer affects exclusively the surface of perovskite film.

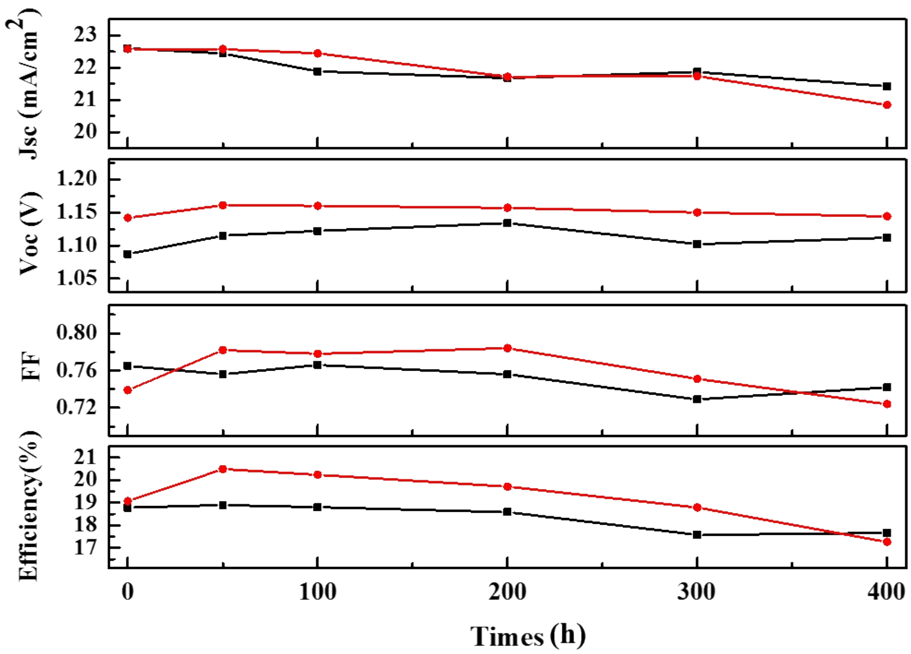


Figure S3. Stability of J_{SC}, V_{OC}, FF and PCE in devices with the as-prepared (black line) and passivated (red line) perovskite film, measured in air without any encapsulation for 400 h.

These devices were stored in dark at a relative humidity of 10%.

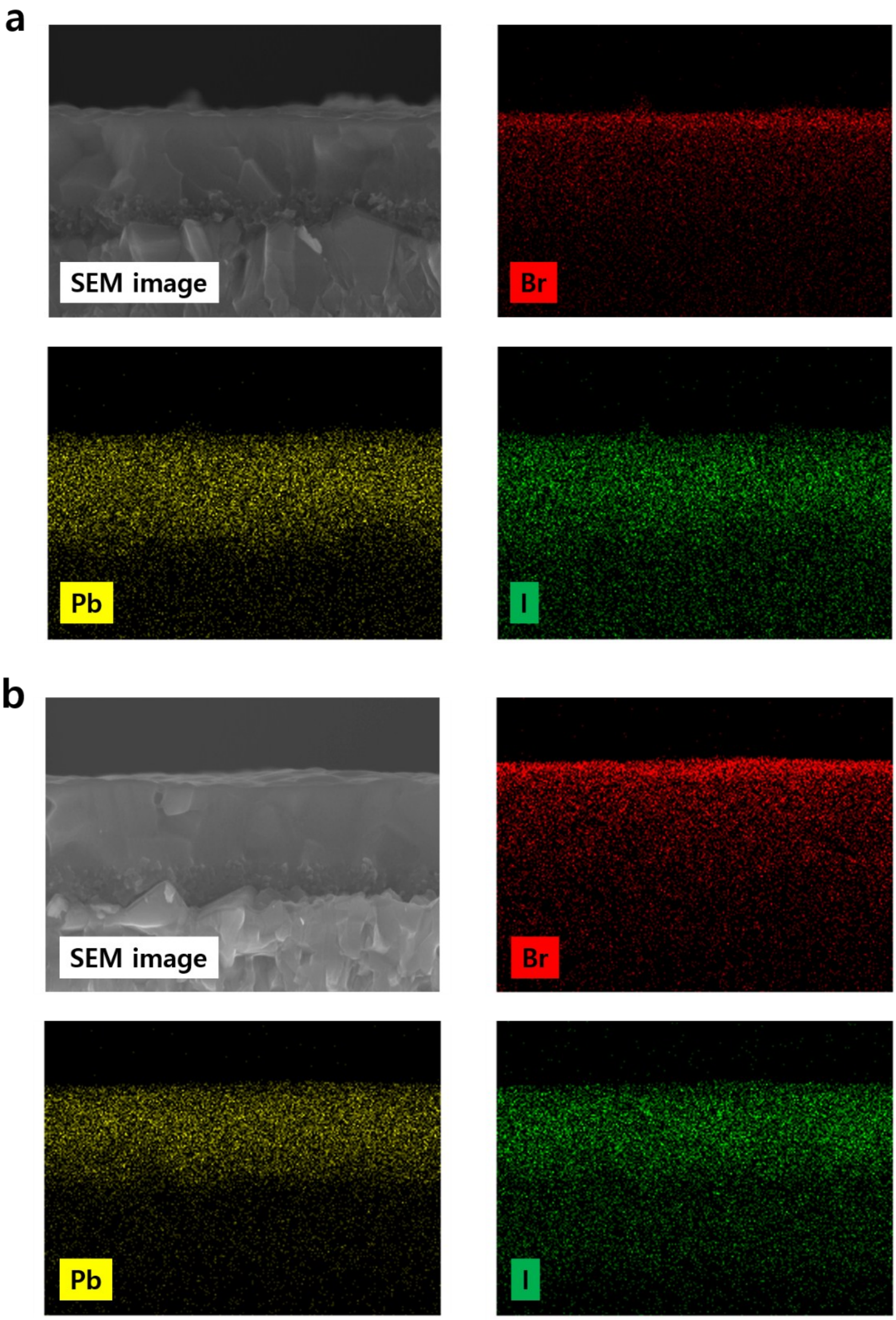
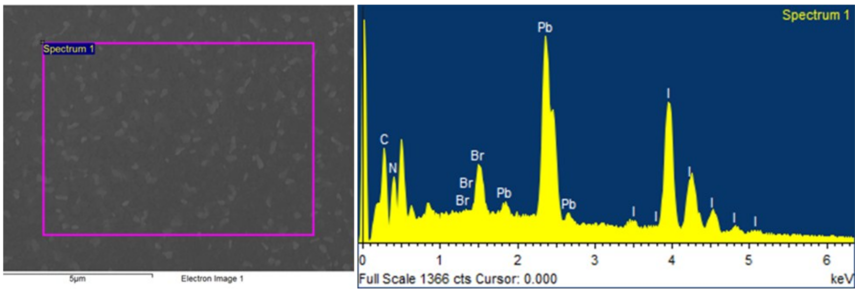


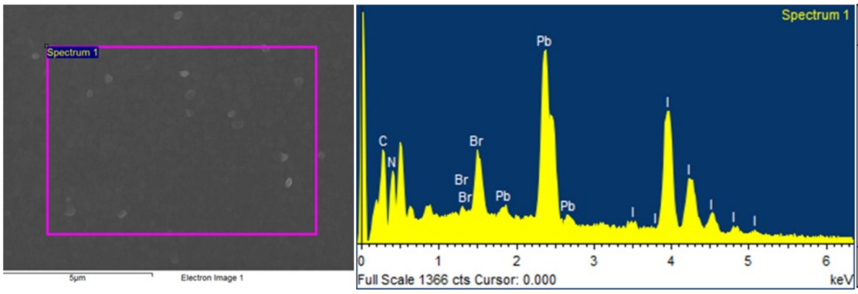
Figure S4. Cross-sectional SEM images and corresponding EDX mapping images of Br, Pb, and I component for as-prepared (a) and passivated (b) perovskite films.

a



Element	wt. %	at. %
C	5.32	29.41
N	6.20	29.38
Br	3.76	3.13
I	54.07	28.27
Pb	30.64	9.81
Total	100.00	100.00

b



Element	wt. %	at. %
C	5.42	28.72
N	7.00	31.81
Br	4.68	3.73
I	52.77	26.48
Pb	30.14	9.27
Total	100.00	100.00

Figure S5. Energy dispersive X-ray spectroscopy (EDS) spectra of perovskite films. a, As-prepared. **b**, Passivated. Measured atomic percentages of the main elements (C, N, Pb, I and Br) in the annealed perovskite are summarized in tables. Br component can be re-calculated to 7.59 % and 9.45 % (considering Pb, I, and Br) in the pristine and passivated perovskite films, respectively. An increased amount of Br element after spin coating FABr and post annealing is verified.

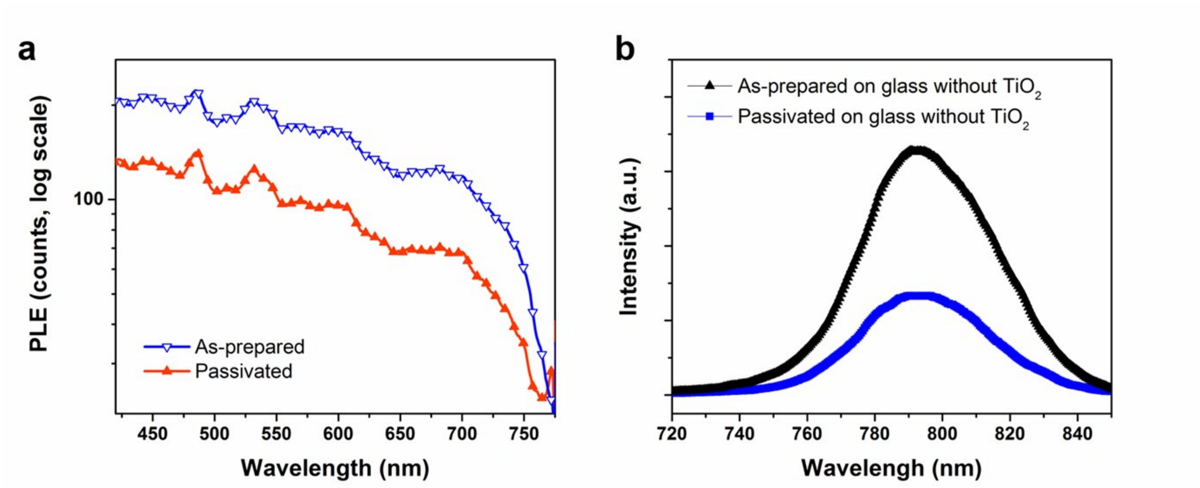


Figure S6. a. Photoluminescence excitation (PLE) spectra of as-prepared and passivated perovskite films deposited on mesoporous TiO_2. **b.** Photoluminescence (PL) spectrum of as-prepared and passivated perovskite films on the glasses without FTO and TiO_2.

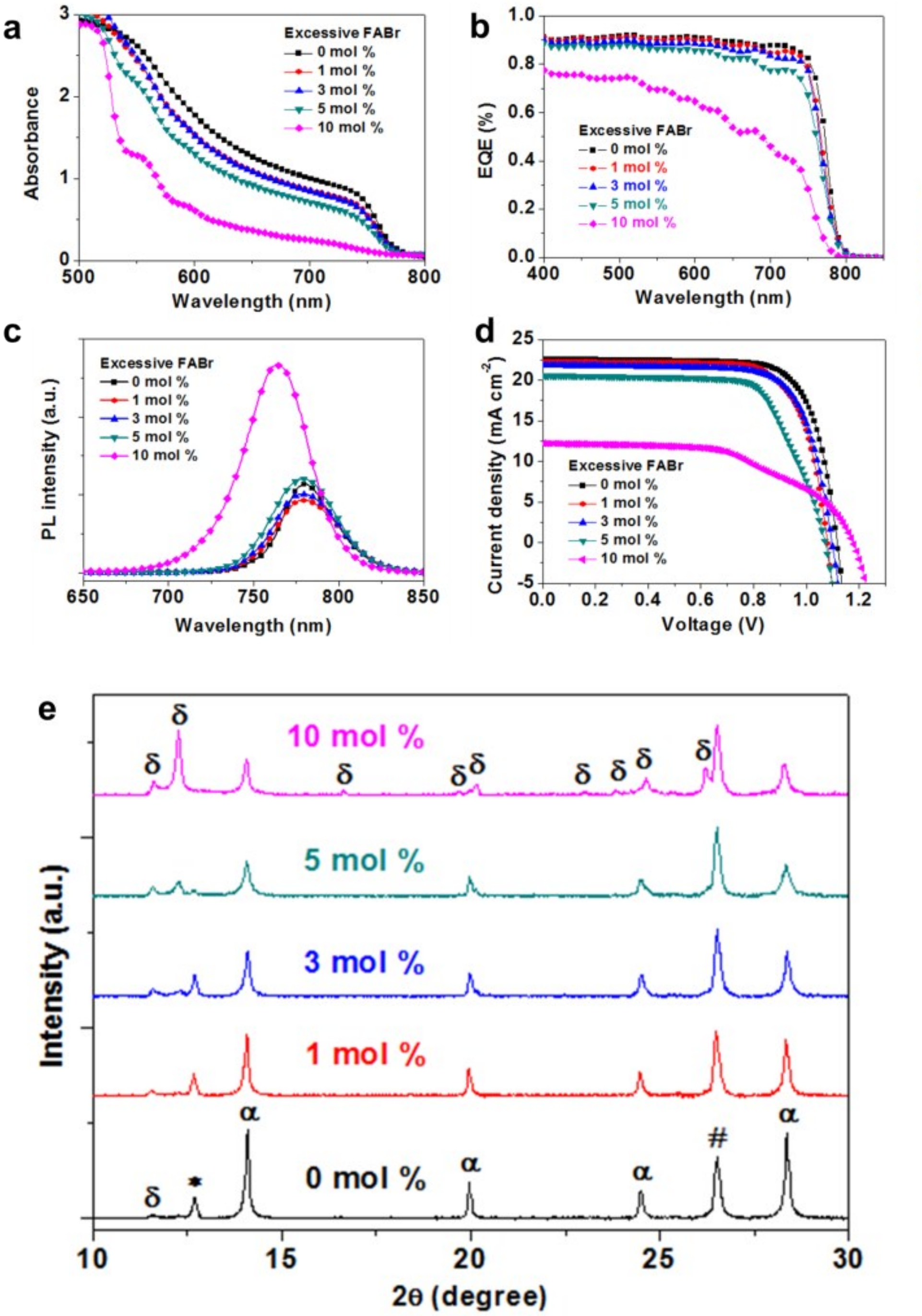


Figure S7. Photo-physical properties and photovoltaic performance observed from the perovskite films treated with FABr solutions (0, 1, 3, 5, and 10 mol %) (**a**) UV-Vis absorption; (**b**) EQE and (**c**) PL spectra for the as prepared samples; **(d)** *J-V* curves obtained from devices prepared with the corresponding perovskite films; (e) XRD patterns of the perovskite films. α, δ, * and # denote the diffraction peak of α-$FAPbI_3$ phase, δ-$FAPbI_3$ phase, PbI_2 and FTO, respectively.

Reference

1. Zhumekenov, A. A.; Saidaminov, M. I.; Haque, M. A.; Alarousu, E.; Sarmah, S. P.; Murali, B.; Dursun, I.; Miao, X.-H.; Abdelhady, A. L.; Wu, T. Formamidinium Lead Halide Perovskite Crystals with Unprecedented Long Carrier Dynamics and Diffusion Length *ACS Energy Lett.* **1**, 32– 37 (2016).

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