Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.


# Enhanced $V_{OC}$ of two-dimensional Ruddlesden–Popper perovskite solar cells using binary synergetic organic spacer cations

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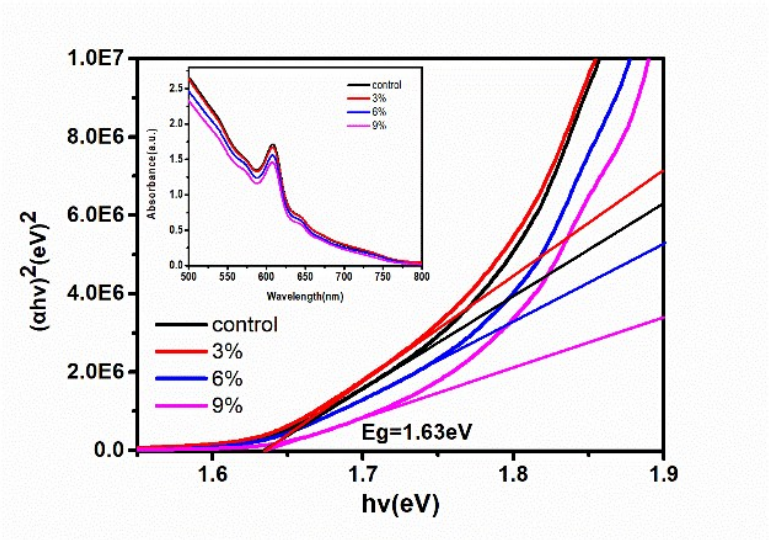


**Fig.S1.** The corresponding optical bandgaps by Tauc plot, and the inset is absorption spectra of the perovskite samples

**Table S1.** Parameters of the TRPL spectroscopy of control film and OA-modified (3%) film on the glass substrate. The excitation is from the substrate side. The average lifetime ($\tau_{ave}$) is calculated using: $\tau_{ave} = (A_1\tau_1^2 + A_2\tau_1^2)/(A_1\tau_1 + A_2\tau_2)$

| samples | $\tau_1$ (ns) | $A_1$ | $\tau_2$ (ns) | $A_2$ |
|---|---|---|---|---|
| **control film** | 12.69 | 9285.95 | 34.15 | 752.26 |
| **OA-modified (3%)** | 23.55 | 9310.17 | 71.45 | 1104.48 |

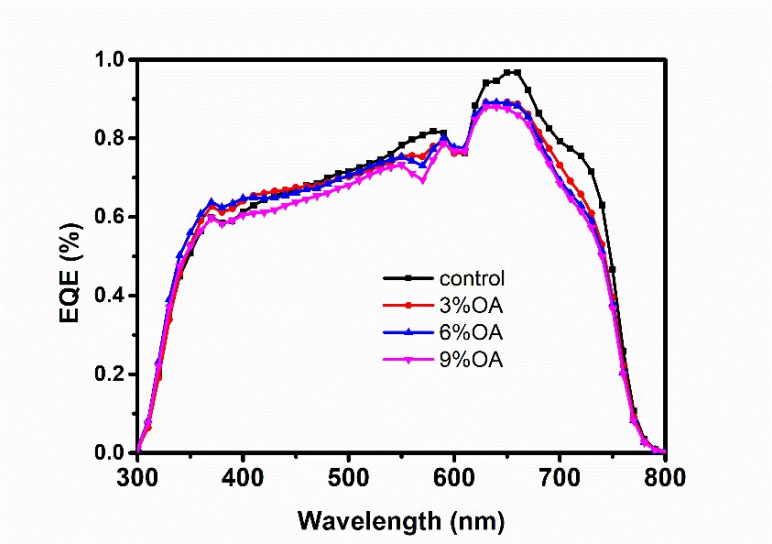


**Fig.S2**. EQE spectra of the PSCs with different molar ratio of OA cations

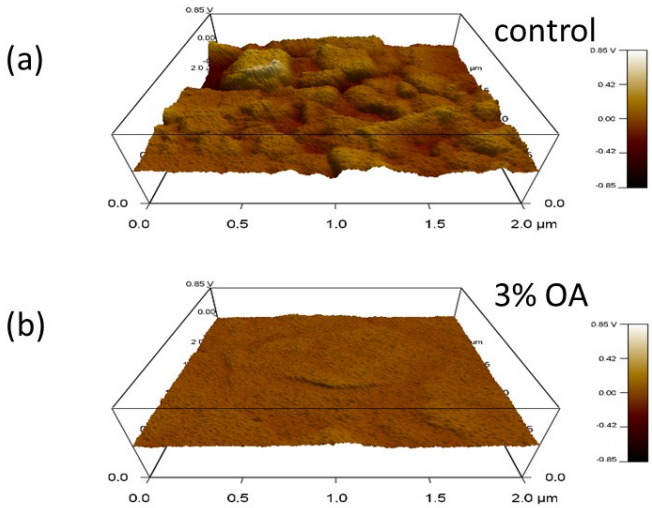


**Fig. S3.** The 3D images of surface potential from **(a)** control film, (**b)** 3% OA additive.

The 3D images of the surface potential are shown in **Fig. S1.** The control film exhibits intense surface potential fluctuations, which will generate more trap states at surface. Compare to the control film, the 2D RPP films with 3% OA additive show more uniform potential distribution across the surface, indicating that 3% OA additive can reduce the trap states at surface[1] and induces the surface passivation of the perovskite films.

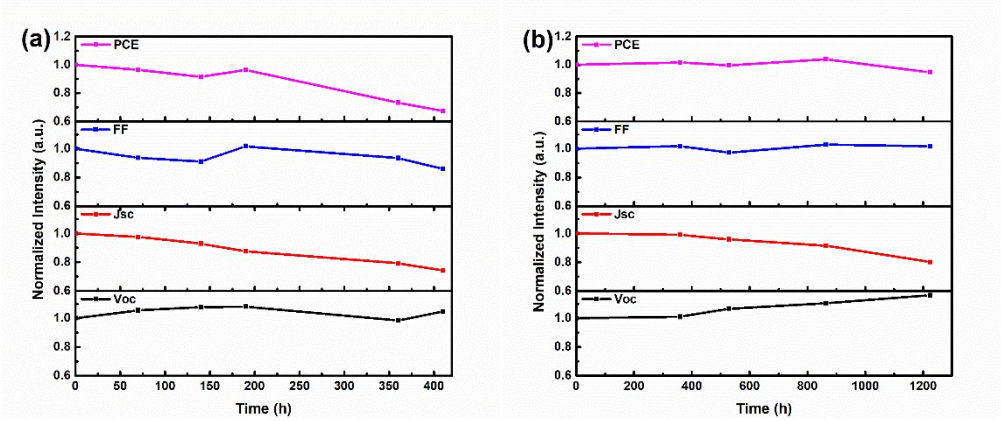


**Fig. S4.** The stability test of unsealed 2D RPPs based on $(BA)_2(MA)3Pb_4I_{13}$ perovskite film under **(a)** air atmosphere with humidity of 30% ± 5% and temperature of 20℃±5℃; **(b)** N2 glove box.

The stability of 2D RPPs based on 100% BA is shown in Fig. S3. The PSCs based on 100% BA remain 67% of its original PCE after keeping in air with humidity of 30% ± 5% and temperature of 20℃±5℃ for 410 h. In condition of stored in N2 glove box, the unencapsulated PSCs with 100% BA remain 94% of its original PCE

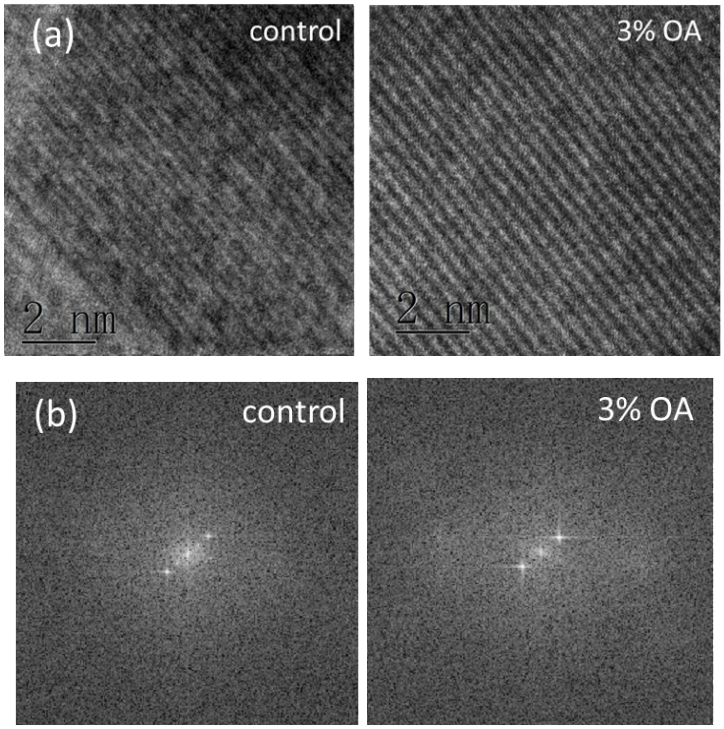


**Fig S5. a)** HRTEM images for control film and OA-modified (3%) film; **b)** images of fast Fourier transform (FFT) of the HRTEM images.

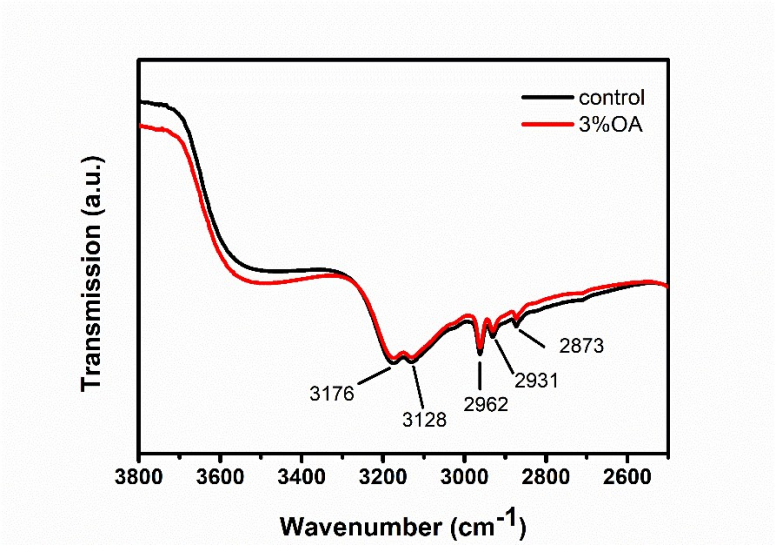


**Fig S6.** The FTIR of control film and OA-modified (3%) film.

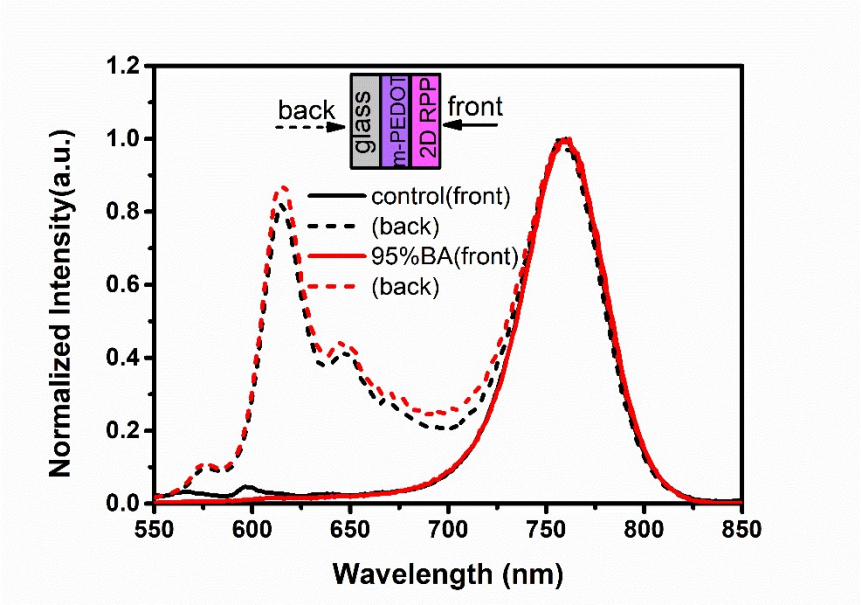


**Fig. S7.** The photoluminescence (PL) spectra of the control film and the film with 95% malar ratio BA cation. The excitation wavelength for PL was at 505 nm