Supporting Information

Engineering the passivation routes of the perovskite films towards high performance solar cells

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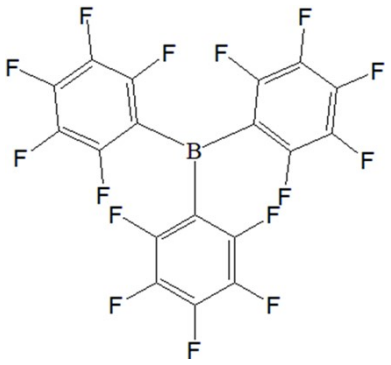

Fig. S1 Molecular structure of tris(pentafluorophenyl)borane (BCF).

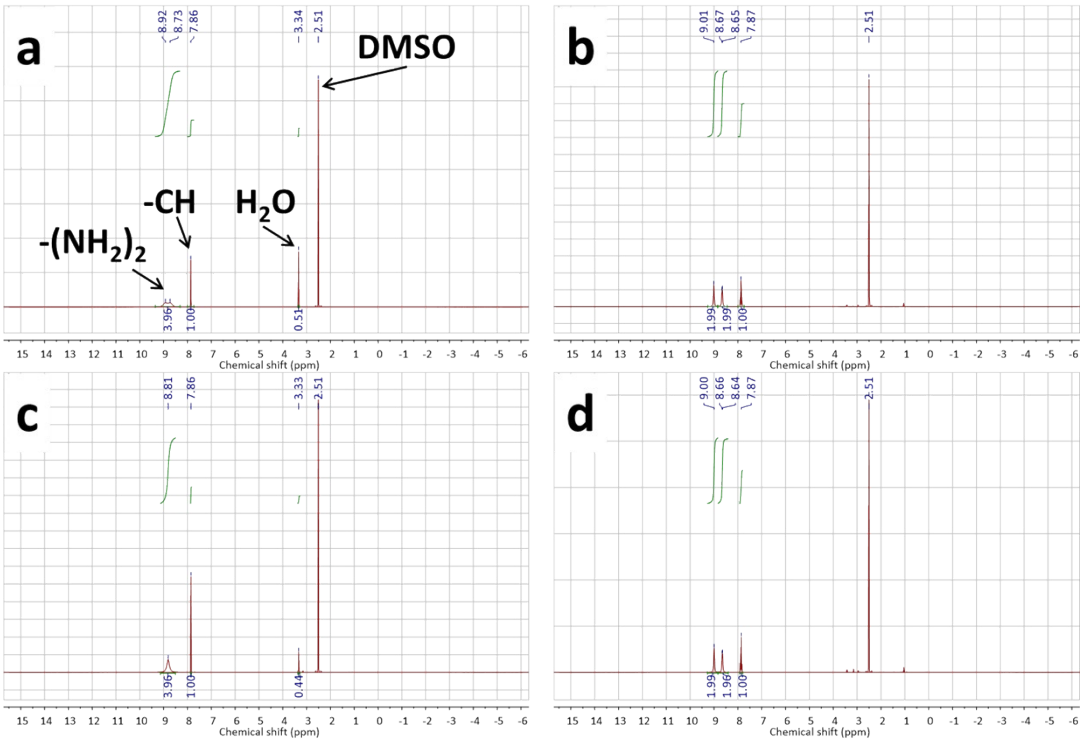

Fig. S2 Liquid-state ^{1}H NMR full spectra (600 MHz, in pure DMSO-d_6) of (a) FAI solution, (b) FAI solution with BCF, (c) perovskite solution and (d) perovskite solution with BCF.

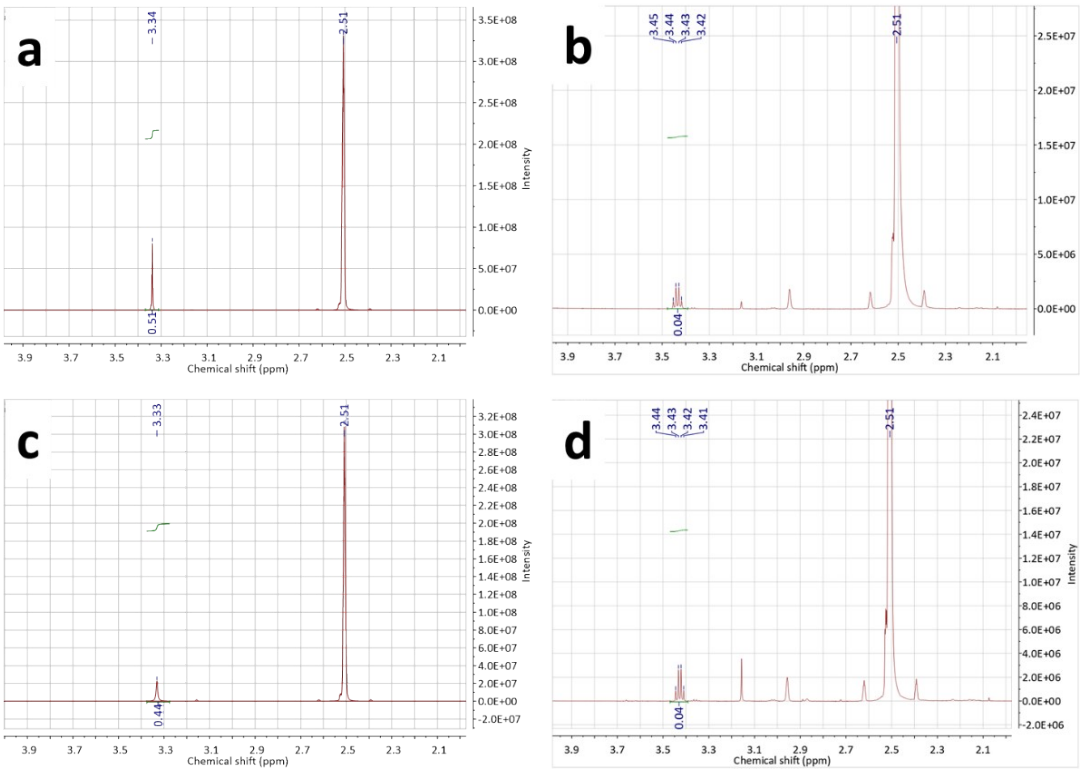

Fig. S3 ^{1}H NMR spectra range from 2 ppm to 4 ppm of (a) FAI solution, (b) FAI solution with BCF, (c) perovskite solution and (d) perovskite solution with BCF. (b) and (d) were magnified about ten times.

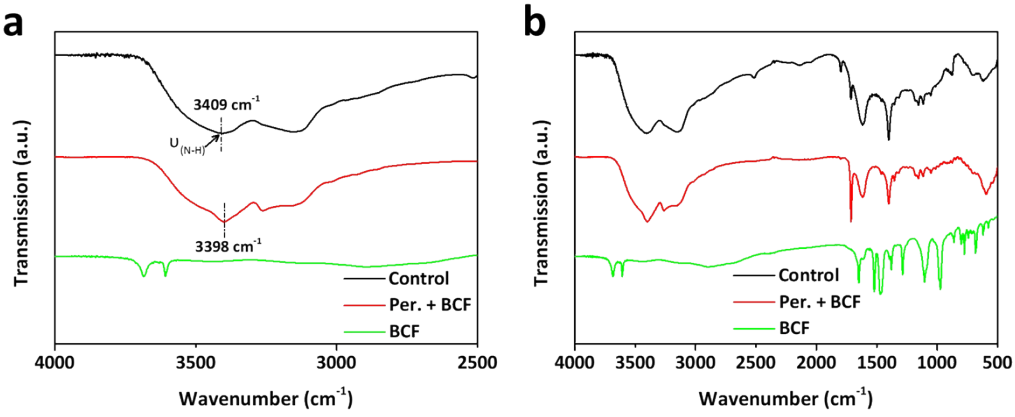

Fig. S4 FTIR spectra of control, BCF passivated perovskite films and pure BCF powders in the range of (a) 4000 - 2500 cm^{-1} and (b) 4000 - 500 cm^{-1}.

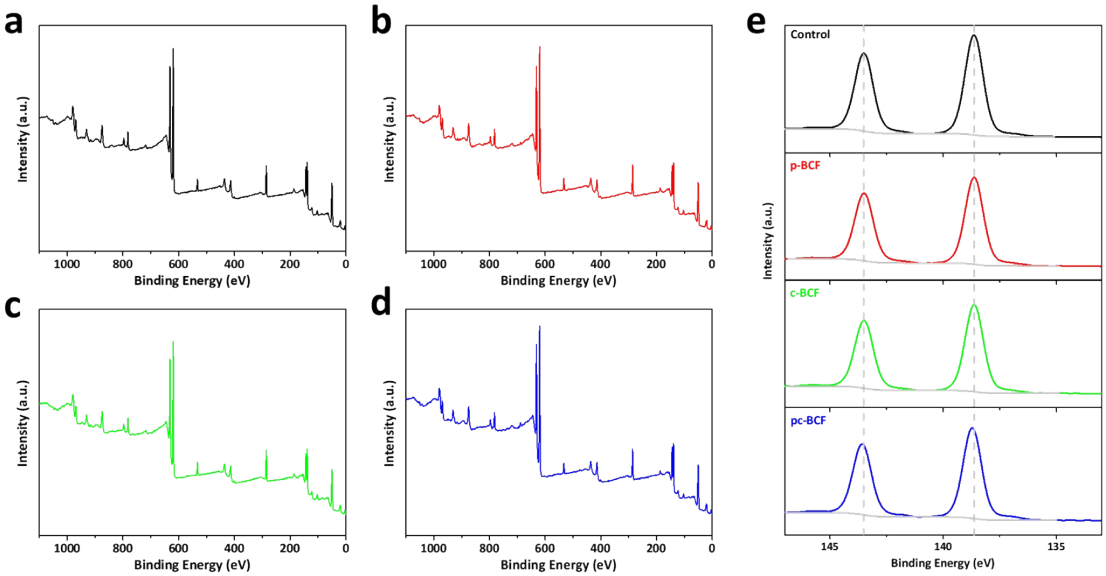

Fig. S5 XPS survey spectra for (a) control, (b) p-BCF, (c) c-BCF and (d) pc-BCF perovskite films. (e) XPS spectra of Pb 4f for the control, p-BCF, c-BCF and pc-BCF perovskite films.

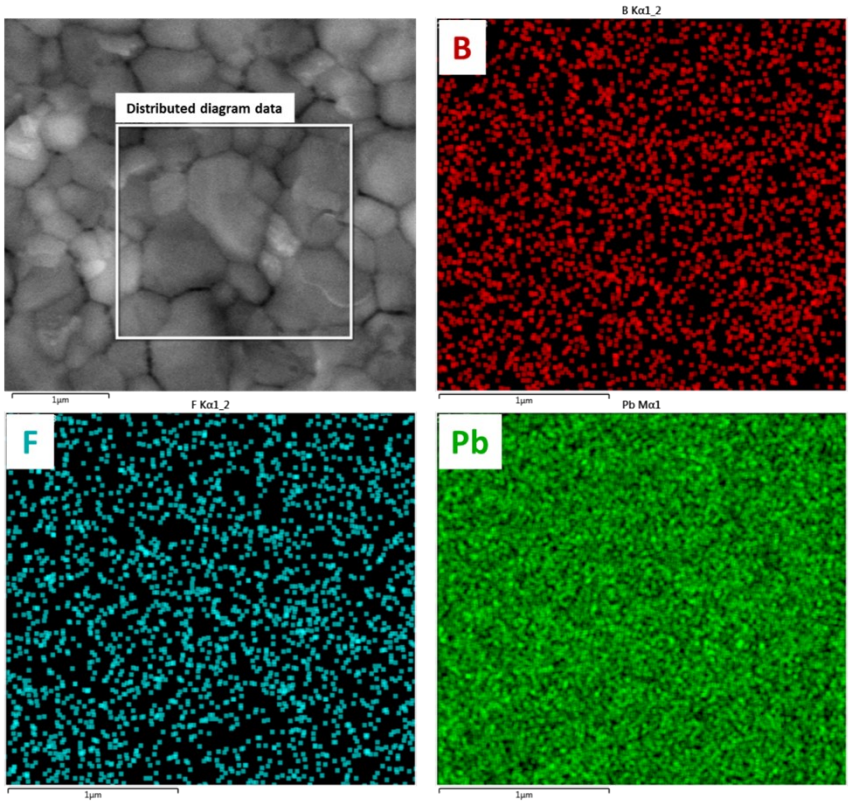

Fig. S6 SEM-EDS mapping of the B, F and Pb components in the p-BCF perovskite film.

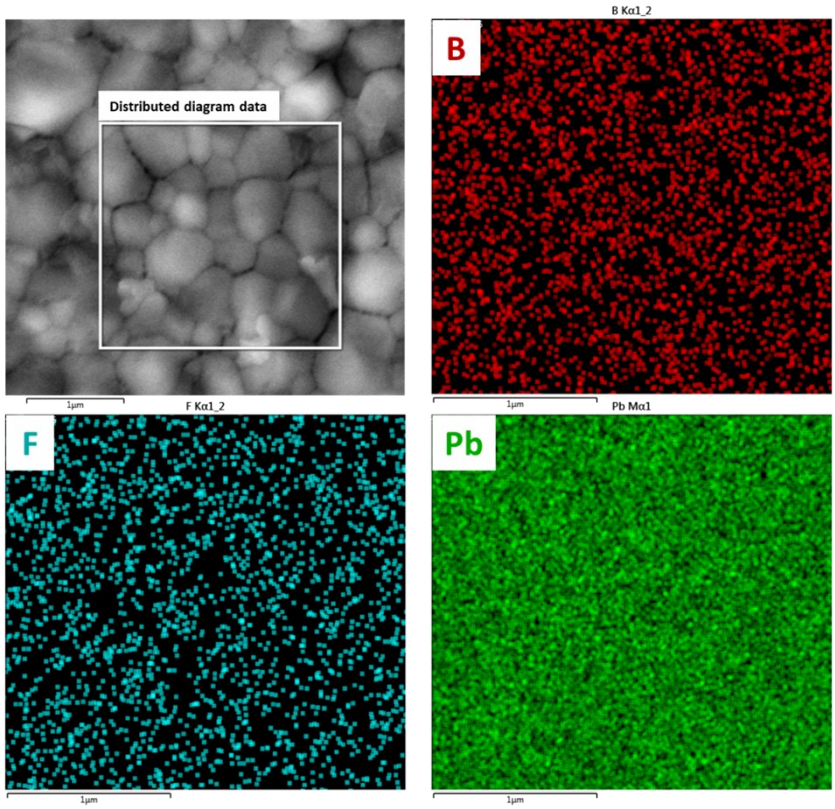

Fig. S7 SEM-EDS mapping of the B, F and Pb components in the c-BCF perovskite film.

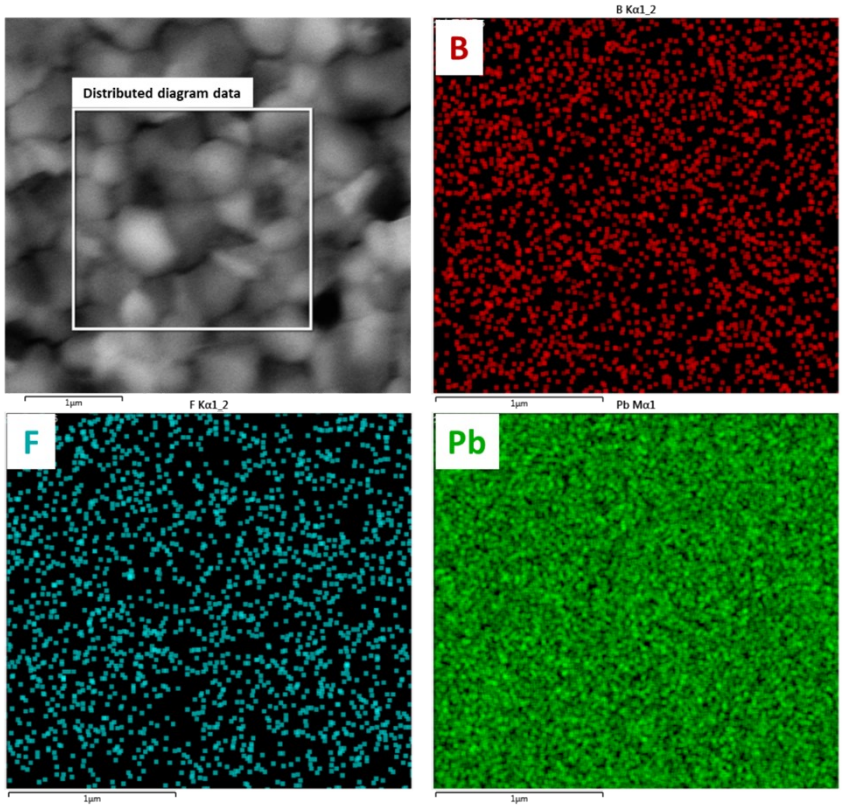

Fig. S8 SEM-EDS mapping of the B, F and Pb components in the pc-BCF perovskite

film.

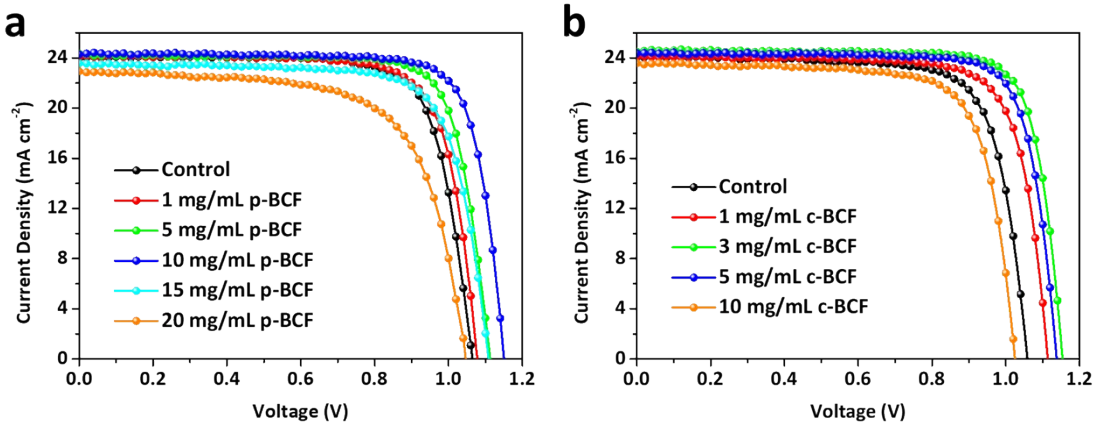

Fig. S9 *J–V* curves of perovskite solar cells with different concentration of BCF by (a) p-BCF treatment and (b) c-BCF treatment.

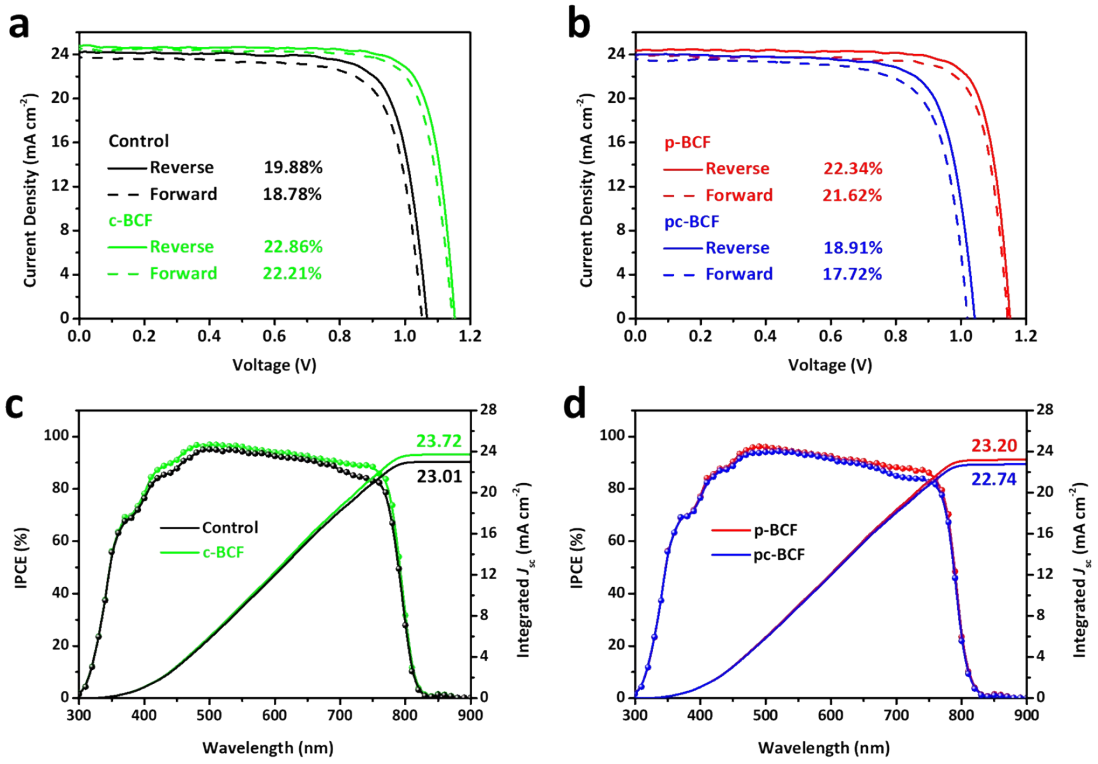

Fig. S10 (a), (b) *J-V* curves under reverse and forward scan directions. (c), (d) Incident photon-to-electron conversion efficiency (IPCE) spectra and integrated J_{SC} of the corresponding devices.

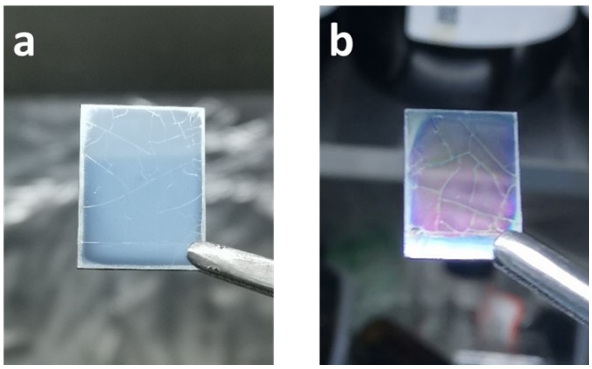

Fig. S11 The pc-BCF perovskite film (a) before and (b) after spiro-OMeTAD coating. After spin coating of HTM, the cracks on the films are still obvious.

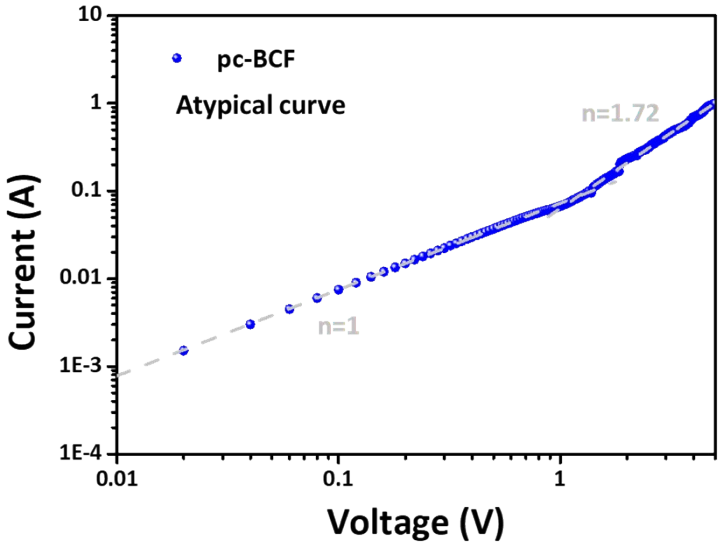

Fig. S12 Dark current–voltage (I–V) curves of hole-only devices of pc-BCF perovskite.

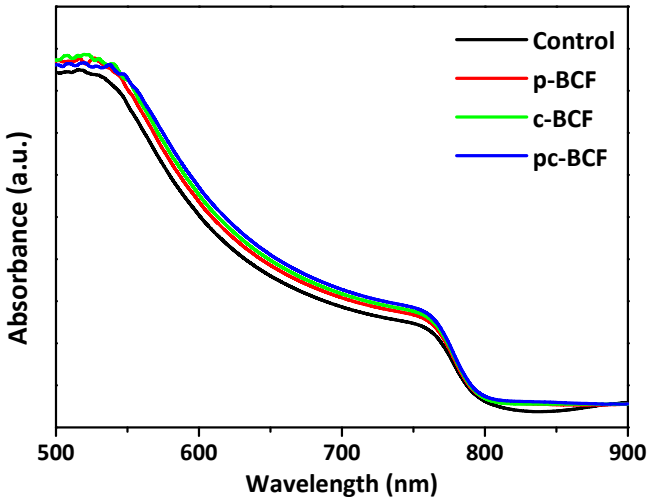

Fig. S13 UV-vis absorption spectra of the control, p-BCF, c-BCF and pc-BCF

perovskite films.

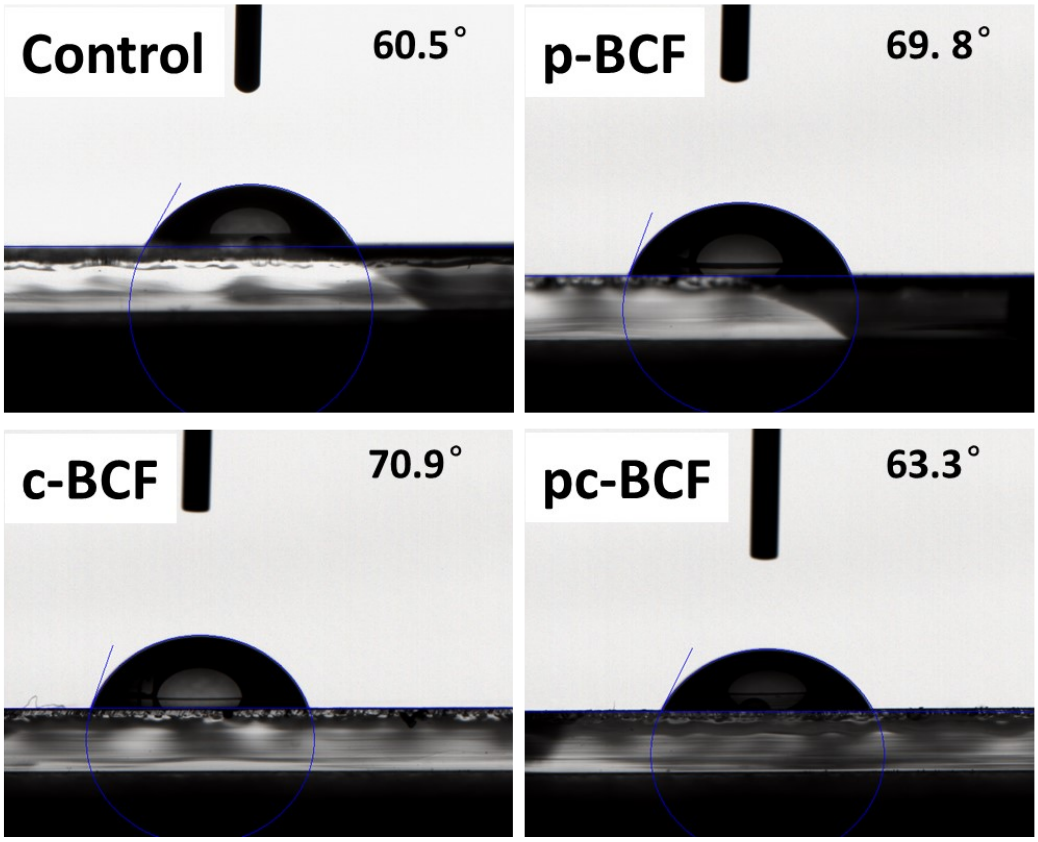

Fig. S14 Water contact angles of perovskite films based on different BCF treatments.

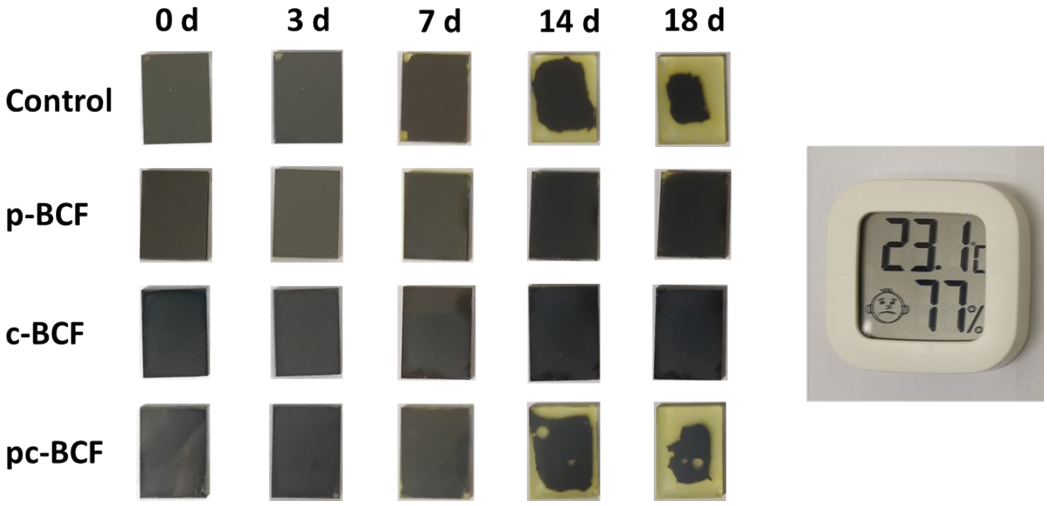

Fig. S15 The photographs of perovskite films based on different BCF treatments after aging under 75±5% RH for 18 days.

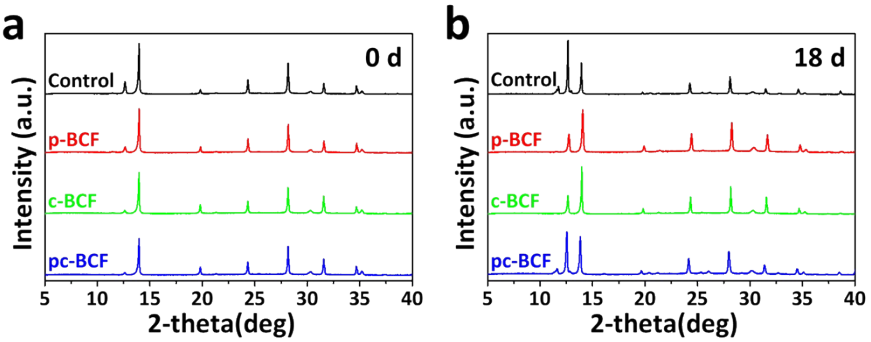

Fig. S16 XRD spectra of the perovskite films based on different treatments (a) before and (b) after aging under 75±5% RH for 18 days.

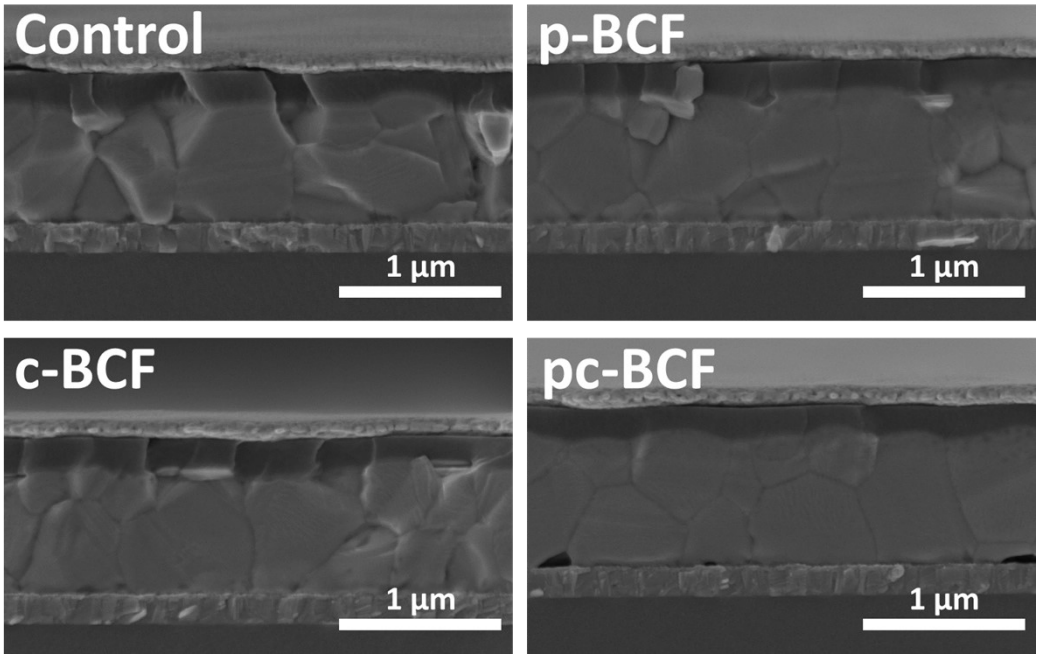

Fig. S17 Cross-sectional SEM images of the PSCs based on different BCF treatments.

Table S1 Detailed device parameters for perovskite solar cells with different concentration of BCF by p-BCF treatment.

Concentration of BCF (mg/mL)	J_{sc} **(mA cm^{-2})**	V_{oc} **(V)**	**FF (%)**	**PCE (%)**
0	24.04	1.06	76.21	19.50
1	24.08	1.08	76.44	19.82

5	24.29	1.11	78.29	21.15
10	24.32	1.15	79.41	22.19
15	23.54	1.11	75.14	19.59
20	22.93	1.04	67.05	16.05

Table S2 Detailed device parameters for perovskite solar cells with different concentration of BCF by c-BCF treatment.

Concentration of BCF (mg/mL)	J_{sc} **(mA cm^{-2})**	V_{oc} **(V)**	**FF (%)**	**PCE (%)**
0	24.07	1.08	76.06	19.73
1	24.13	1.11	77.17	20.71
3	24.53	1.15	80.34	22.71
5	24.42	1.14	79.54	22.08
10	23.51	1.02	74.84	18.02

Table S3 Detailed device parameters for the best control, p-BCF, c-BCF and pc-BCF perovskite solar cells.

Treatment	J_{sc} **(mA cm^{-2})**	V_{oc} **(V)**	**FF (%)**	**PCE (%)**
Control	24.21	1.07	76.98	19.88
p-BCF	24.36	1.15	79.75	22.34
c-BCF	24.78	1.15	80.09	22.86
pc-BCF	24.00	1.04	75.61	18.91

Table S4 The fitted parameters of perovskite with and without BCF treatments deposited on glass substrates from TRPL spectra.

Treatment	τ_1 **(ns)**	τ_2 **(ns)**	A_1	A_2	τ_{ave} **(ns)**
Control	41.56	401.41	1.10	0.33	309.08

p-BCF	52.99	656.51	0.62	0.39	587.88
c-BCF	52.68	761.78	0.59	0.39	694.62
pc-BCF	36.29	522.22	1.31	0.29	406.12

The TRPL curves are fitted via a bi-exponential formula:

$$Y = A_1 e^{\frac{-t}{\tau_1}} + A_2 e^{\frac{-t}{\tau_2}} + Y_0$$

where A_1 and A_2 are the decay amplitudes, τ_1 and τ_2 are the decay lifetimes, Y_0 is a constant for the baseline offset. The fast (τ_1) and slow (τ_2) decay lifetimes are ascribed to the recombination occurring on the surface and in the bulk of perovskite films, respectively.[1] In order to comprehensively describe the recombination lifetime of charge carriers, the average lifetime is introduced according to the equation

$$\tau_{ave} = \frac{A_1\tau_1^2 + A_2\tau_2^2}{A_1\tau_1 + A_2\tau_2}$$[2]

References

1 J. Jeong, M. Kim, J. Seo, H. Lu, P. Ahlawat, A. Mishra, Y. Yang, M. A. Hope, F. T. Eickemeyer, M. Kim, Y. J. Yoon, I. W. Choi, B. P. Darwich, S. J. Choi, Y. Jo, J. H. Lee, B. Walker, S. M. Zakeeruddin, L. Emsley, U. Rothlisberger, A. Hagfeldt, D. S. Kim, M. Gratzel, J. Y. Kim, *Nature*, 2021, **592**, 381-385.

2 M. Du, X. Zhu, L. Wang, H. Wang, J. Feng, X. Jiang, Y. Cao, Y. Sun, L. Duan, Y. Jiao, K. Wang, X. Ren, Z. Yan, S. Pang, S. F. Liu, *Adv. Mater.*, 2020, **32**, e2004979.