A Novel 2D Perovskite as Surface "Patches" for Efficient Flexible Perovskite Solar Cells

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Figure S1 Cross-section SEM image of the different PFAI-treated PSCs: (a) control perovskite, (b) 2-PFA, (c) 6-PFA, (d) 10-PFA, (e) 14-PFA, (f) control film with postannealing, (g) 2-PFA-H, (h) 6-PFA-H, (i) 10-PFA-H, (j) 14-PFA-H.

Perovskite/PFAI	Voc(V)	$Jsc(mA/cm^2)$	FF (%)	PCE (%)
Control	1.059±0.005	24.07±0.12	78.82±0.6	20.09±0.04
Champion device	1.056	24.16	78.98	20.15
2 mg/mL PFAI	1.087±0.004	24.04±0.04	79.90±0.28	20.88±0.12
Champion device	1.096	24.10	80.10	21.16
6 mg/mL PFAI	1.089±0.003	23.97±0.05	79.96±0.69	20.87±0.15
Champion device	1.089	23.92	81.18	21.15
10 mg/mL PFAI	1.095±0.004	24.03±0.14	80.46±0.36	21.17±0.12
Champion device	1.098	24.11	80.77	21.38
14 mg/mL PFAI	1.072±0.006	23.94±0.11	78.18±0.83	20.06±0.20
Champion device	1.074	24.00	78.80	20.31

Table S1 Photovoltaic performance parameters the PSCs based on the PFAI treatment from J-V measurements under standard AM 1.5 illumination (100 mW cm⁻²). *



Figure S2 J-V curves of the champion devices based on the PFAI treatment and the combination of the PFAI treatment and post-annealing at backward scan and forward scan conditions.

Table S2 Photovoltaic performance parameters of the PSCs based on the PFAI treatment with post-annealing from J-V measurements under standard AM 1.5 illumination (100 mW cm⁻²). *

Perovskite/PFAI	Voc(V)	Jsc (mA/cm ²)	FF (%)	PCE (%)
Control	1.061±0.005	24.09±0.08	78.58±0.45	20.08±0.07
Champion device	1.052	24.07	79.67	20.17
2 mg/mL PFAI	1.090±0.003	23.97±0.08	79.71±0.25	20.83±0.08
Champion device	1.094	24.09	79.50	20.95
6 mg/mL PFAI	1.095±0.007	24.02±0.07	79.54±0.36	20.92±0.15
Champion device	1.097	24.04	80.23	21.16
10 mg/mL PFAI	1.121±0.007	23.99±0.13	77.43±0.21	20.82±0.19
Champion device	1.131	24.17	77.58	21.20
14 mg/mL PFAI	1.059±0.011	23.71±0.04	69.71±1.15	17.50±0.43
Champion device	1.076	23.67	71.39	18.18

Table S3 Photovoltaic performance parameters of the PSCs based on the PFAI treatment, BAI treatment, OAI treatment, and PEAI treatment from J-V measurements under standard AM 1.5 illumination (100 mW cm⁻²). *

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Perovskite/PFAI	Voc(V)	Jsc (mA/cm ²)	FF (%)	PCE (%)
Control	1.061±0.005	24.09±0.08	78.58±0.45	20.09±0.07
Champion device	1.052	24.07	79.67	20.18
With BAI, 100 °C	1.077±0.003	24.13±0.060	79.19±0.47	20.58±0.11
Champion device	1.080	24.13	79.90	20.82
With OAI, 100 °C	1.080 ± 0.002	24.19±0.07	79.69±0.35	20.82±0.11
Champion device	1.080	24.25	80.22	21.01
With PEAI, RT	1.092±0.002	24.24±0.09	79.30±0.34	21.00±0.12
Champion device	1.104	24.08	80.01	21.27
With PEAI, 85 °C	1.073±0.011	24.22±0.10	76.92±0.80	19.99±0.39
Champion device	1.088	24.35	77.78	20.61
With PFAI, RT	1.095±0.004	24.03±0.14	80.46±0.36	21.17±0.12
Champion device	1.098	24.11	80.75	21.38
With PFAI, 100 °C	1.119±0.005	23.91±0.10	78.19±0.23	20.92±0.09
Champion device	1.126	23.93	78.21	21.07



Figure S3 EQE curve and the integrated Jsc curve of the 10-PFA-H based PSC.



Figure S4 Change of the device PCE with the different post-annealing time of the 10-PFA-H perovskite layer.



Figure S5 Change of the device PCE with the different post-annealing temperature of the 10-PFA-H perovskite layer.



Figure S6 (a) XRD patterns of the 2D perovskite films resulting from the PFAI and PbI₂ (2:1) precursor solutions containing different FA cations (denoted as 15% FAI-2:1, 25% FAI-2:1, and 35% FAI-2:1) and XRD pattern of the 2D perovskite film resulting from the PFAI and PbI₂ (1:1) precursor solution (denoted as 25% FAI-1:1). (The percentages 15%, 25%, and 35%, are the molar ratios of FAI salt to PbI₂, respectively.) (b) XRD patterns of the 2D perovskite films resulting from the PFAI and PbI₂ (2:1) precursor solutions containing different metal/MA⁺ halides.



Figure S7 Topographic images and corresponding current images of (a) 10-PFA, (b) 10-PFA-H, (c) 14-PFA, and (d) 14-PFA-H. The configuration of the devices for the catomic force microscopy (c-AFM) measurements is perovskite/SnO₂/FTO. ORCA mode with a bias of 1.0 V was applied on an AFM machine (Cypher S, Asylum Research, USA) in air.

Figures S7a and S7b shows the c-AFM images and corresponding images of 10-PFA and 10-PFA-H perovskite films. The average currents of 10-PFA and 10-PFA-H are 115 and 126 pA, respectively. It can be concluded that the post-annealing treatment can enhance the conductivity of surface 2D perovskite. However, the post-annealing treatment also results in a rougher surface caused by the improved crystallization, as shown in Figure S7a and S7b. Similar results can be concluded from samples 14-PFA and 14-PFA-H.



Figure S8 Absorption spectra of the perovskite films based on different PFAItreatment with or without post-annealing.



Figure S9 (a) Electrochemical impedance spectra and (b) fitted recombination resistances of the devices based on the control perovskite films, the 2-PFA film, the 6-PFA film, and the 10-PFA film at different bias voltages in the dark.



Figure S10 SEM images of (a) the surface morphology of the PFA-10 film on the flexible substrate and (b) the cross-section morphology of the flexible PSC based on

the PFA-10 film.



Figure S11 J-V curves of the champion flexible PSCs based on (a) the 10-PFA film and (b) the 10-PFA-H film under forward and backward scan directions under AM 1.5G standard illumination. (c) Histogram distribution of PCEs belonging to the flexible PSCs based on the 10-PFA film.



Figure S12 Determination the absorption threshold of the perovskite absorber by (a)

differential of EQE, (b) UV-vis absorption spectrum, and (c) UV-vis diffuse

absorbance spectrum.



Figure S13 Dependence of the PCE of the flexible PSC based on the PFA-10 film on

the bending cycles at a bending radius of 10 mm.