## Modifying the buried interface with azodicarbonamide for high-efficiency MA-free perovskite solar cells

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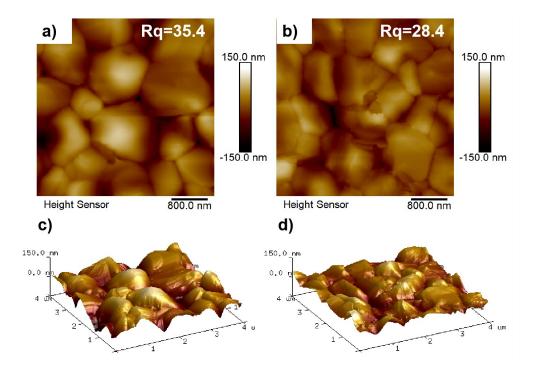


Fig. S1. Two- and three- dimensional AFM images of perovskite films spincoated on (a,c) the  $TiO_2$  and (b,d) ADC-modified  $TiO_2$  substrates.

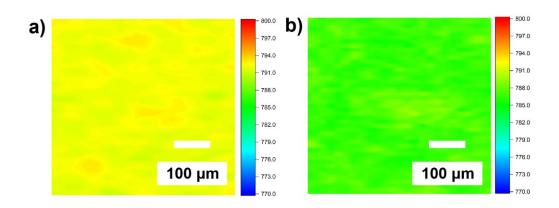


Fig. S2. PL position mapping images of the (a) control and (b) target perovskite film.

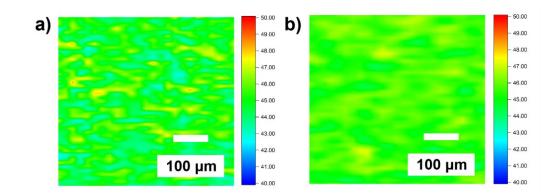


Fig. S3. PL full width at half maxima of the (a) control and (b) target perovskite film.

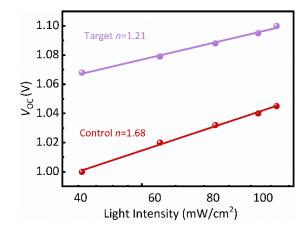


Fig. S4.  $V_{OC}$  versus light intensity for the control and target devices.

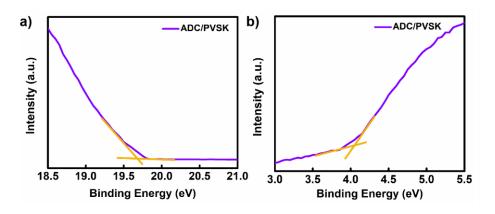


Fig. S5. UPS spectra of ADC/PVSK: (a) secondary electron cutoff regions, and (b) and Fermi edge region.

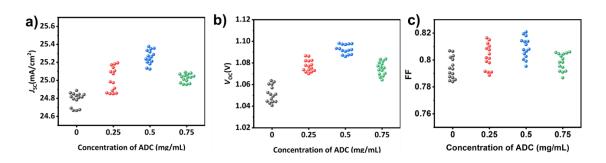


Fig. S6. Statistics of (a)  $J_{SC}$ , (b)  $V_{OC}$ , and (c) FF of PSCs based on TiO<sub>2</sub> modified by different concentrations of ADC.

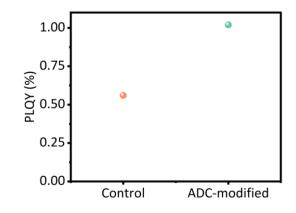


Fig. S7. PLQY for the layer structure  $glass/FTO/TiO_2/(without or with ADC)/perovskite.$ 

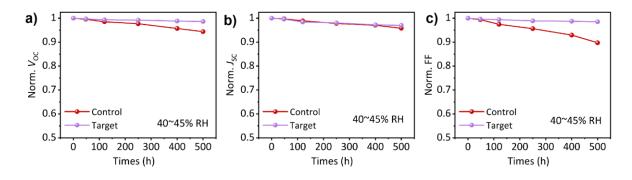


Fig. S8. (a)  $V_{OC}$ , (b)  $J_{SC}$ , and (c) FF as a function of time for the unencapsulated control and target devices aged under a relative humidity of 40-45% at room temperature in the dark.

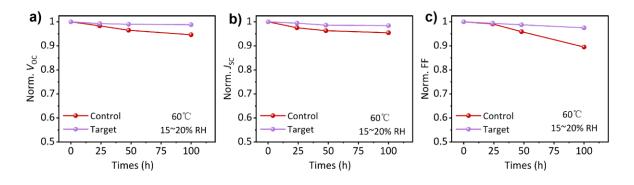


Fig. S9. (a)  $V_{OC}$ , (b)  $J_{SC}$ , and (c) FF as a function of time for the unencapsulated control and target devices aged at 60 °C in the dark where the unencapsulated devices were located in the air condition.

	Glass/PVSK	Glass/ADC/PVSK	
71 (ns)	23.88	18.17 45.5 110.84	
0⁄0	60.2		
$\tau_2$ (ns)	107.63		
0⁄0	39.8	54.5	
$ au_{\rm ave}$ (ns)	86.57	99.68	

Table S1 Fitted results of TRPL curves of the perovskite films deposited on the glass or with glass+ADC.

	FTO/TiO <sub>2</sub> /PVSK	FTO/TiO <sub>2</sub> /ADC/PVSK/	
71 (ns)	5.47	4.40 84.4 19.61 15.6	
%	76.8		
$\tau_2$ (ns)	23.79		
%	23.2		
$ au_{\mathrm{ave}}\left(\mathrm{ns}\right)$	15.85	11.27	

Table S2 Fitted results of TRPL curves of the perovskite films deposited on the substrates with  $TiO_2$  or with  $TiO_2$ +ADC.

			-		
ADC (mg/mL)	Samples	$J_{\rm SC}$ (mA/cm <sup>2</sup> )	V <sub>OC</sub> (V)	FF	PCE (%)
0	Champion	24.67	1.047	0.806	20.82
	Average	24.78±0.099	1.051±0.012	0.793±0.013	20.69±0.146
0.25	Champion	25.01	1.077	0.803	21.61
	Average	25.19±0.184	1.086±0.009	0.816±0.134	21.79±0.171
0.5	Champion	25.38	1.098	0.820	22.52
	Average	25.25±0.118	1.092±0.005	0.809±0.117	22.32±0.196
0.75	Champion	25.09	1.084	0.805	21.69

Table S3. Photovoltaic parameters of the PSCs based on  $TiO_2$  modified with different concentrations of ADC from 0 to 0.75 mg/mL.