Supporting Information

CaI_2: A More Effective Passivator of Perovskite Films than PbI_2 for High Efficiency and Long-term Stability of Perovskite Solar Cells

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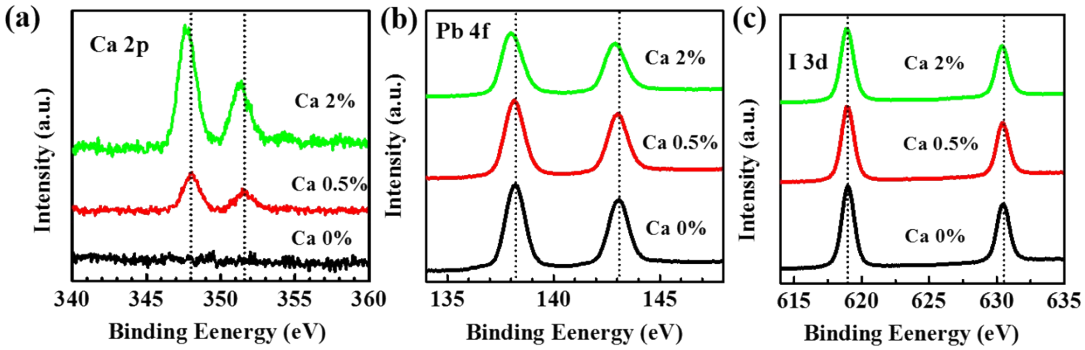

Figure S1. XPS spectra of (a) Ca 2p, (b) Pb 4f and (c) I 3d at the surface of the $MAPbI_3$, $MAPbI_3(CaI_2)_{0.005}$, $MAPbI_3(CaI_2)_{0.02}$ films, respectively.

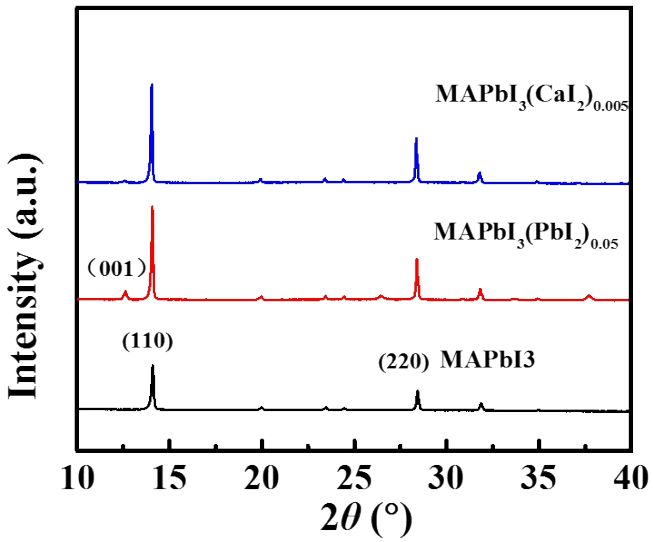

Figure S2. XRD patterns of the pristine $MAPbI_3$, $MAPbI_3(PbI_2)_{0.05}$ and $MAPbI_3(CaI_2)_{0.005}$ films, respectively.

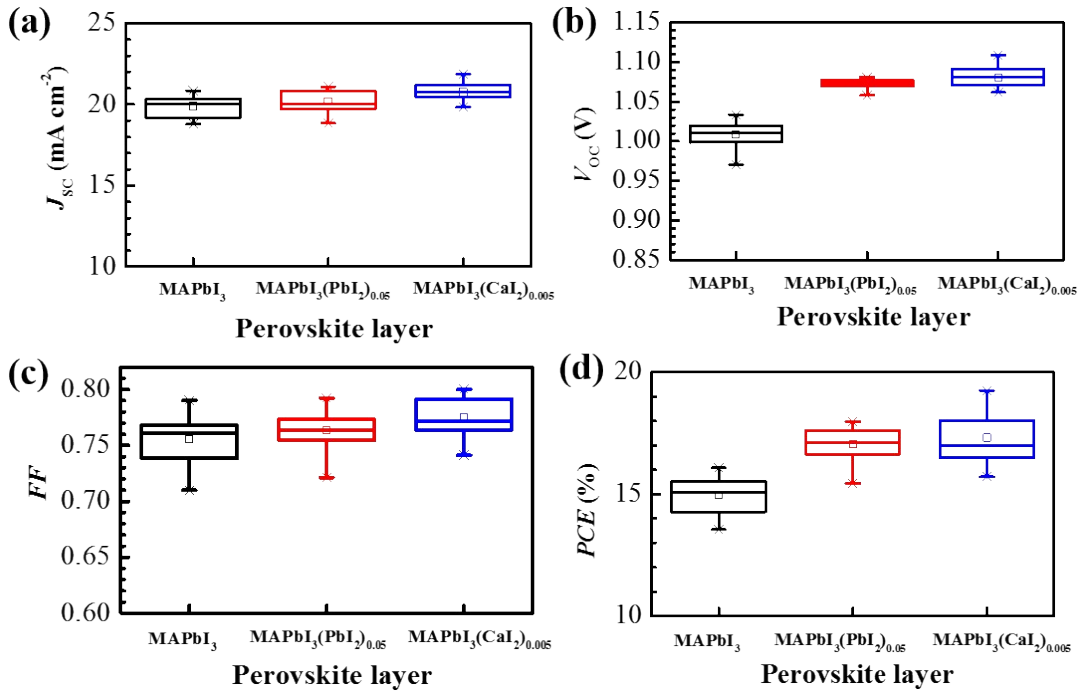

Figure S3. Statistic on the performance parameters of 16 pieces solar cells based on the pristine $MAPbI_3$, $MAPbI_3(PbI_2)_{0.05}$ and $MAPbI_3(CaI_2)_{0.005}$ films: (a) J_{SC}, (b) V_{OC}, (c) *FF*, (d) *PCE*.

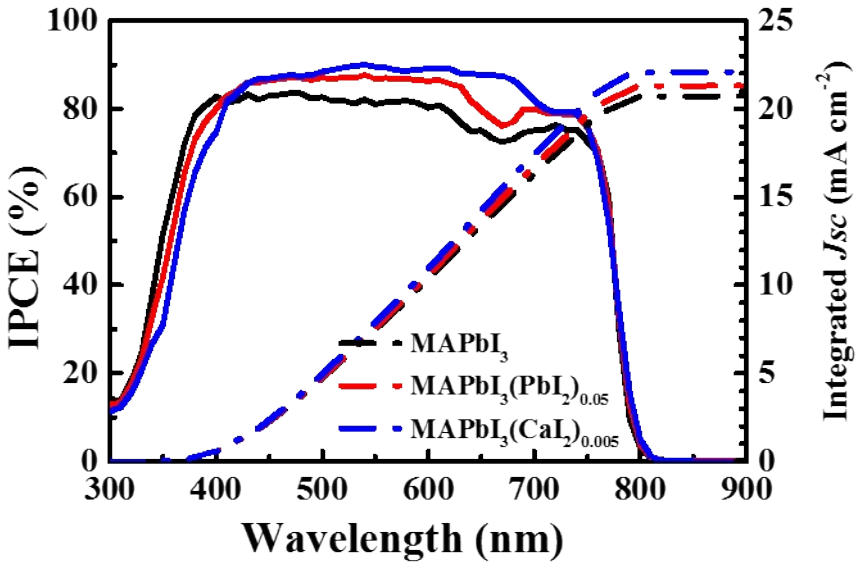

Figure S4. IPCE spectra (solid lines) and the integrated photocurrent density (dashed lines) of the devices based on the pristine $MAPbI_3$, $MAPbI_3(PbI_2)_{0.05}$ and $MAPbI_3(CaI_2)_{0.005}$ films, respectively.

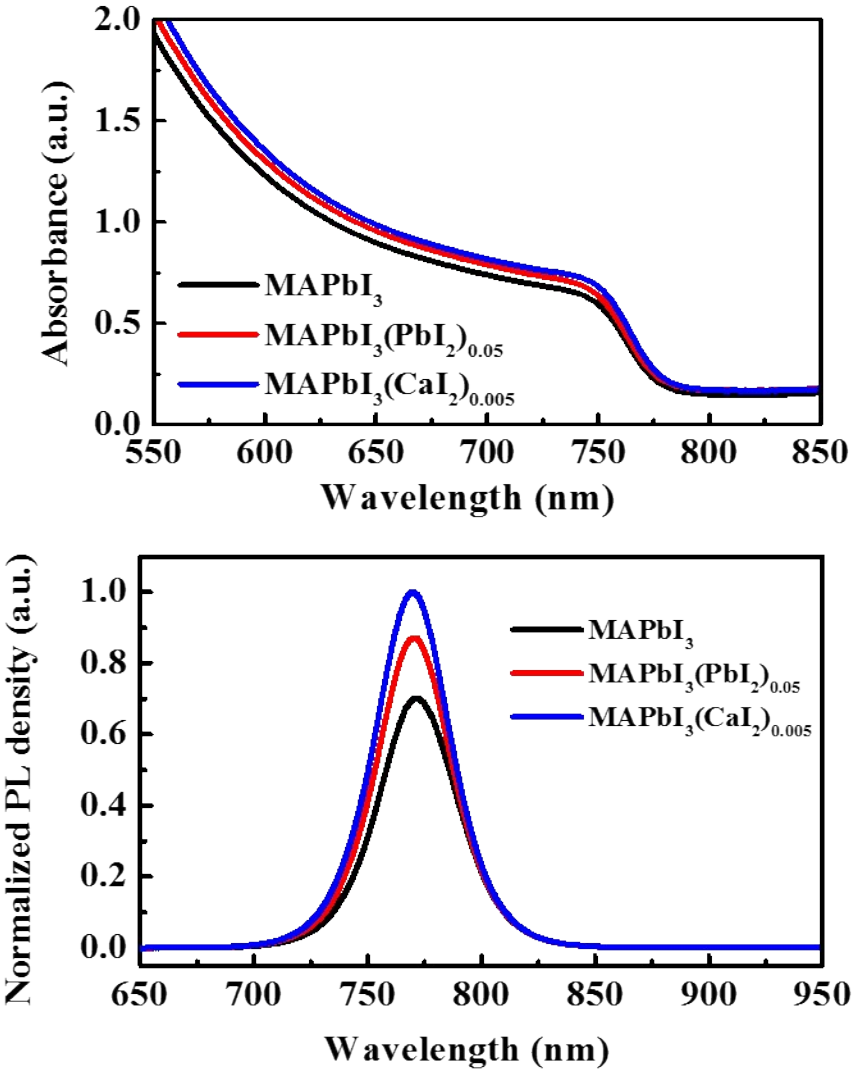

Figure S5. (a) UV–vis absorption spectra of the pristine $MAPbI_3$, $MAPbI_3(PbI_2)_{0.05}$ and $MAPbI_3(CaI_2)_{0.005}$ films on glass substrates. (b) Steady-state PL spectra of the compared three films on glass substrates.

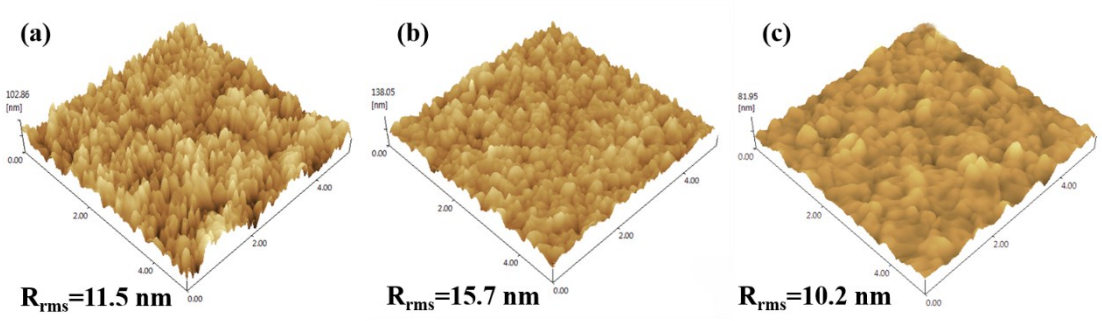

Figure S6. AFM three-dimensional views of (a) the pristine $MAPbI_3$, (b) $MAPbI_3(PbI_2)_{0.05}$ and (c) $MAPbI_3(CaI_2)_{0.005}$ films; their root mean square roughness (R_{rms}) are 11.5 nm, 15.7 nm and 10.2 nm, respectively.

Table S1. Performance parameters of the champion cells based on $MAPbI_3(CaI_2)_x$ perovskite layers measured in the forward and reverse scan directions at a scan rate of 0.05 V s^{-1} under simulated solar light (AM 1.5G, 100 mW cm^{-2}).

$MAPbI_3(CaI_2)x$	Scan direction	V_{OC} (V)	J_{SC} (mA cm^{-2})	*FF*	*PCE* (%)	R_S Ω cm^2	R_{SH} KΩ cm^2
x=0	Forward	1.028	20.57	0.760	16.07	1.86	1.55
	Reverse	1.039	20.31	0.752	15.85	2.02	1.50
x=0.0025	Forward	1.059	21.35	0.791	17.88	1.37	2.72
	Reverse	1.055	21.33	0.786	17.68	1.51	3.11
x=0.005	Forward	1.109	21.87	0.794	19.25	1.08	12.44
	Reverse	1.099	21.88	0.797	19.17	1.21	11.41
x=0.01	Forward	1.049	13.85	0.705	10.25	3.54	0.63
	Reverse	1.038	13.62	0.695	9.83	5.72	0.59
x=0.02	Forward	0.958	5.956	0.528	3.01	9.54	0.52
	Reverse	0.968	5.792	0.518	2.91	10.57	0.50

Table S2. Performance parameters of the champion solar cells based on the pristine $MAPbI_3$, $MAPbI_3(PbI_2)_{0.05}$ and $MAPbI_3(CaI_2)_{0.005}$ films, respectively, measured at a rate of 50 mV s^{-1} under simulated solar light (AM 1.5G, 100 mW cm^{-2}).

Perovskite layer	**Scan direction**	V_{OC} **(V)**	J_{SC} **(mA cm^{-2})**	***FF***	***PCE* (%)**	R_S **Ω cm^2**	R_{SH} **KΩ cm^2**
$MAPbI_3$	**Forward**	**1.028**	**20.57**	**0.760**	**16.07**	**1.86**	**1.55**
	Reverse	**1.039**	**20.31**	**0.752**	**15.85**	**2.02**	**1.50**
$MAPbI_3(PbI_2)_{0.05}$	**Forward**	**1.080**	**21.07**	**0.790**	**17.98**	**1.58**	**2.53**
	Reverse	**1.079**	**21.20**	**0.767**	**17.54**	**1.64**	**2.43**
$MAPbI_3(CaI_2)_{0.005}$	**Forward**	**1.109**	**21.87**	**0.794**	**19.25**	**1.08**	**12.44**
	Reverse	**1.099**	**21.88**	**0.797**	**19.17**	**1.21**	**11.05**