Supplementary Information

A Bilayer TiO_2/Al_2O_3 as Mesoporous Scaffold for Enhanced Air Stability of Ambient-Processed Perovskite Solar Cells

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Figure S1. Humidity meter showing the relative humidity of over 65% during preparation and storage of perovskite film and perovskite solar cell samples.

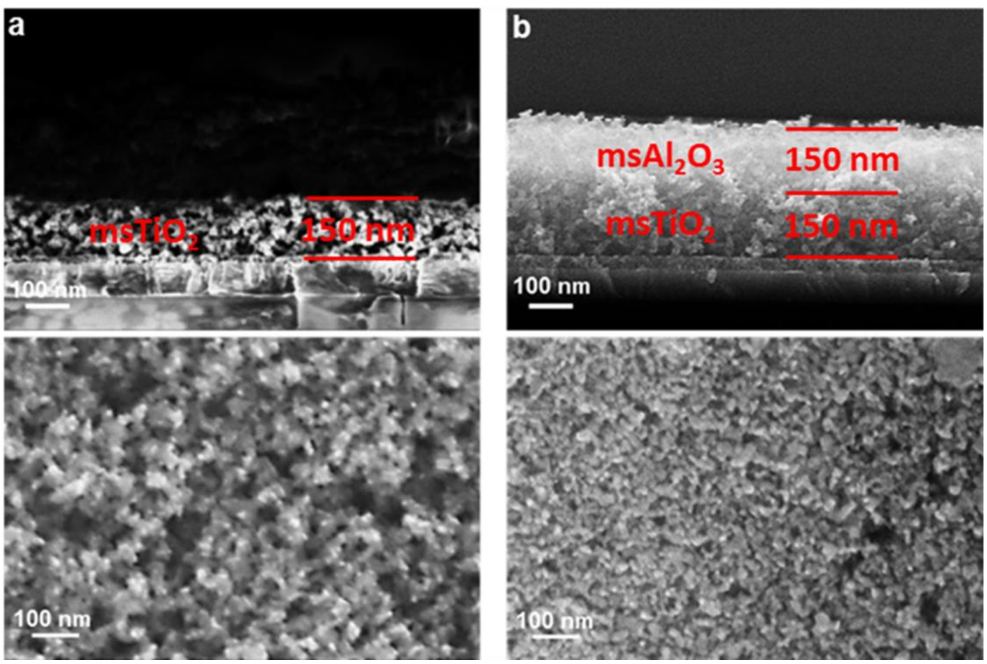

Figure S2. SEM images of (a) mesoporous TiO_2 film and (b) mesoporous TiO_2/Al_2O_3 film. The thicknesses of the single mesoporous TiO_2 and the bilayer mesoporous TiO_2/Al_2O_3 are ~150 nm and 300 nm, respectively.

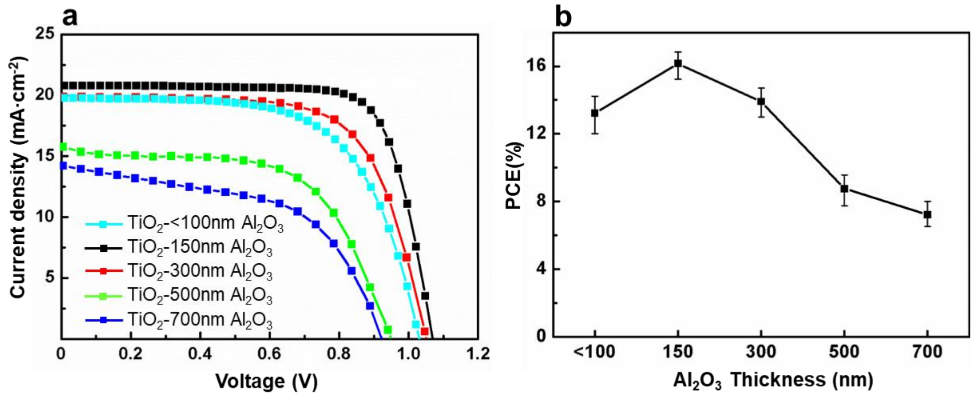

Figure S3. (a) *J-V* curves and (b) PCEs of the bilayer mesoporous TiO_2/Al_2O_3 scaffold-based PSCs with varying Al_2O_3 thicknesses. With the increase of Al_2O_3 thickness, PCE of the bilayer mesoporous TiO_2/Al_2O_3 scaffold-based PSC reaches 16.15% at the point of 150 nm, and finally drops to 7.22%. Each PCE value plotted in Figure S3b is the average over 5 cells in the same counterparts.

Table S1. Photovoltaic parameters [a] of the bilayer mesoporous TiO_2/Al_2O_3 scaffold-based PSCs with varying Al_2O_3 thicknesses.

	PCE (%)	*Jsc* (mA·cm^{-2})	*Voc* (mV)	*FF* (%)
msTiO_2/(50-100) nm ms Al_2O_3	13.21	19.94	1030.5	64.3
msTiO_2/150 nm msAl_2O_3	16.15	20.26	1071.1	73.3
msTiO_2/300 nm msAl_2O_3	13.91	19.74	1047.8	66.5
msTiO_2/500 nm msAl_2O_3	8.76	15.55	948.2	59.1
msTiO_2/700 nm msAl_2O_3	7.22	14.87	918.0	52.7

[a] Each value is the average over 5 cells in the same counterparts.

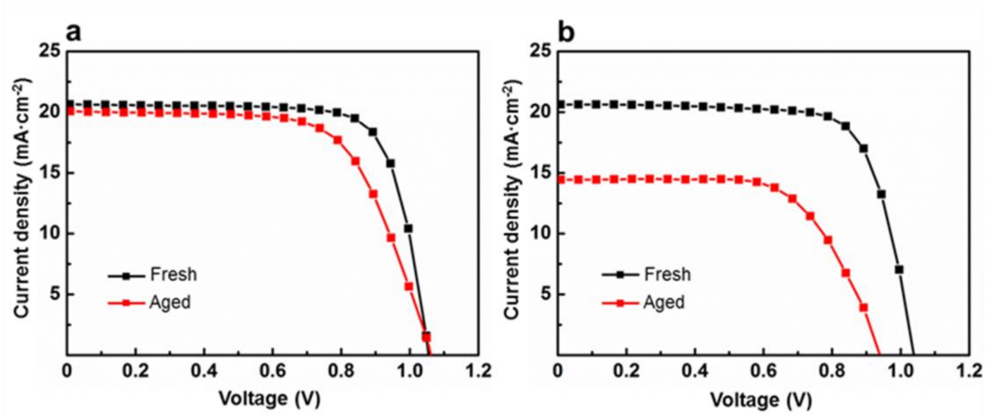

Figure S4. *J-V* curves of the PSCs based on (a) the bilayer mesoporous TiO_2/Al_2O_3 scaffold and (b) the single mesoporous TiO_2 scaffold before and after aging for 12 weeks. All aging tests were carried out in ambient air at >65% relative humidity. PCE of the measured PSC device based on the bilayer mesoporous scaffold decreases from 16.50% to 13.96%, while for the single mesoporous scaffold-based device the PCE falls from 15.83% to 8.78%.

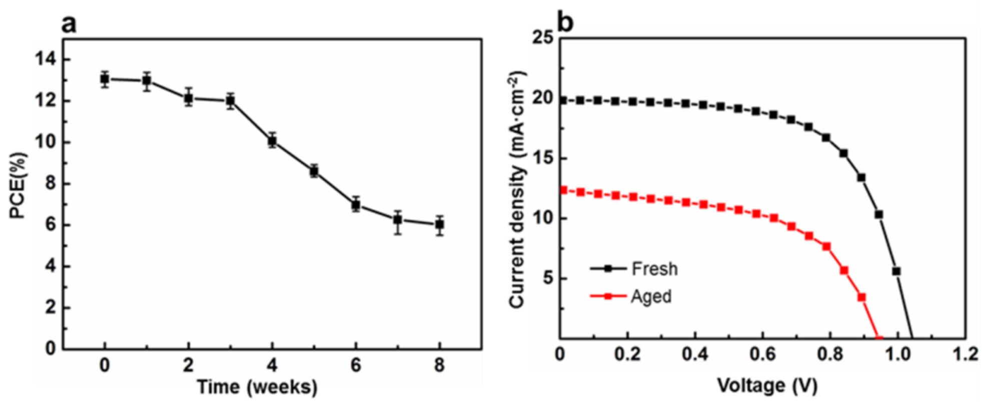

Figure S5. (a) PCE over time of the single mesoporous Al_2O_3 scaffold-based PSC stored in ambient air at >65% relative humidity. Measurement was carried out immediately after fabrication and once a week in the following 8 weeks. Each PCE value is the average over 8 cells in the same counterparts. (b) *J-V* curves of the single mesoporous Al_2O_3 scaffold-based PSC before and after aging for 8 weeks. PCE of the measured PSC device drops from 13.18% to 6.40%.

Table S2. PCE evolution for the PSCs based on the mesoporous TiO_2, the mesoporous Al_2O_3 and the mesoporous TiO_2/Al_2O_3 scaffolds.[a, b]

	Fresh	2 weeks	4 weeks	6 weeks	8 weeks	10 weeks	12 weeks
msTiO_2	15.66	14.84	13.33	10.33	9.09	9.21	8.93
msAl_2O_3	13.06	12.12	10.06	6.97	6.03	-	-
msTiO_2/Al_2O_3	16.04	15.46	14.02	13.85	13.40	13.37	13.16

[a] Each PCE value is the average over 8 cells in the same counterparts (unit: %).

[b] All aging tests were carried out in ambient air at >65% relative humidity.

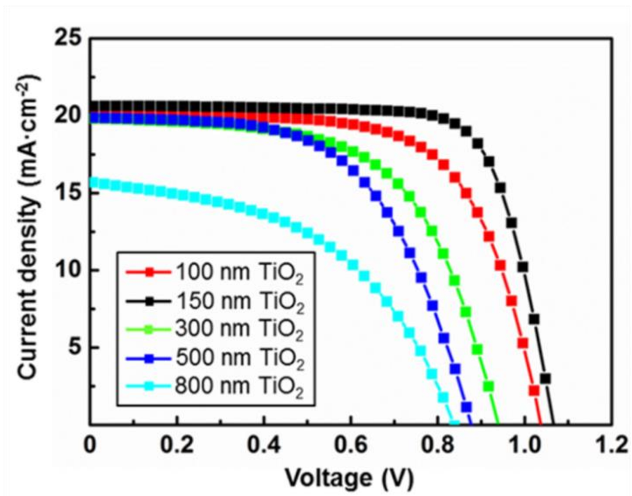

Figure S6. *J-V* curves of the single mesoporous TiO_2 scaffold-based PSCs with varying TiO_2 thicknesses. With the increase of the TiO_2 thickness from 100 nm to 800 nm, PCE of the single mesoporous TiO_2 scaffold-based PSC rises first and reaches 16.43% at the point of 150 nm, then finally drops to 6.36%.

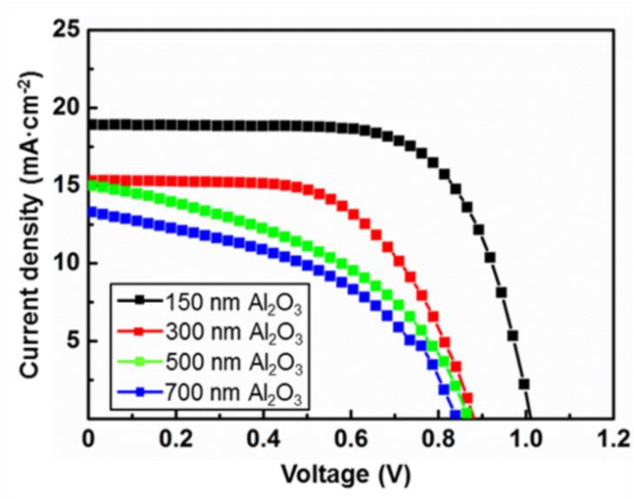

Figure S7. *J-V* curves of the single mesoporous Al_2O_3 scaffold-based PSCs with varying Al_2O_3 thicknesses. With the increase of the Al_2O_3 thickness from 150 nm to 700 nm, PCE of the single mesoporous Al_2O_3 scaffold-based PSC drops from 13.01% to 5.08%.

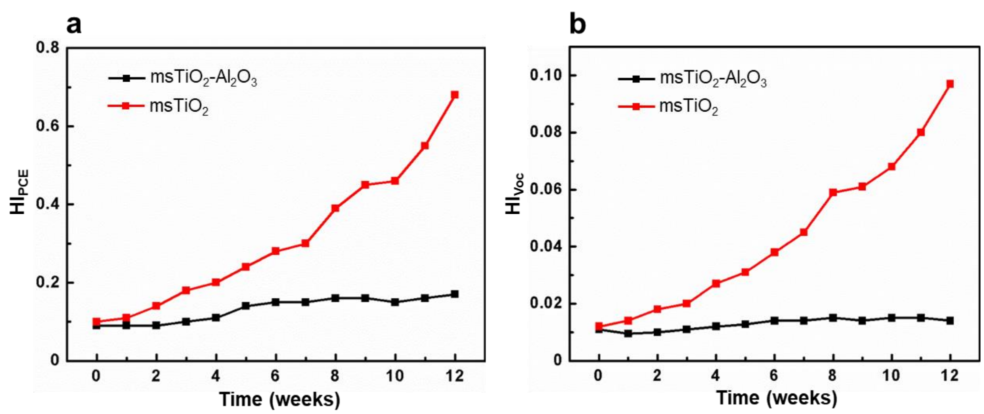

Figure S8. Hysteresis index for (a) PCE and for (b) photovoltage of the PSCs based on different mesoporous scaffolds during the aging test. The devices were stored in ambient air at >65% relative humidity. Measurement was carried out immediately after fabrication and once a week in the following 12 weeks. Each HI value is calculated from the averaged PCE or *Voc* extracted from 8 cells in the same counterparts.

Table S3. Photovoltaic parameters and hysteresis index for PCE and for photovoltage of the PSCs based on the 300 nm TiO_2/Al_2O_3 and 150 nm TiO_2 experiencing different aging times.[a, b]

	Fresh	2 weeks	4 weeks	6 weeks	8 weeks	10 weeks	12 weeks
300 nm TiO_2/Al_2O_3							
PCE (R/F) (%)	16.04/14.60	15.46/14.07	14.02/12.48	13.85/11.77	13.40/11.26	13.37/11.36	13.16/10.92
HI_{PCE}	0.09	0.09	0.11	0.15	0.16	0.15	0.17
Voc (R/F) (mV)	1071/1059	1057/1046	1030/1018	1007/993	1000/985	1004/990	1025/1011
HI_{Voc}	0.011	0.010	0.012	0.014	0.015	0.015	0.014
150 nm TiO_2							
PCE (R/F) (%)	15.66/14.10	14.84/12.72	13.33/10.68	10.33/7.39	9.09/5.58	9.21/4.96	8.93/2.86
HI_{PCE}	0.1	0.14	0.2	0.28	0.39	0.46	0.68
Voc (R/F) (mV)	1073/1060	1038/1019	1009/982	982/945	953/897	959/894	947/855
HI_{Voc}	0.012	0.018	0.027	0.038	0.059	0.068	0.097

[a] All aging tests were carried out in ambient air at >65% relative humidity.

[b] Each PCE or *Voc* value is the average over 8 cells in the same counterparts.

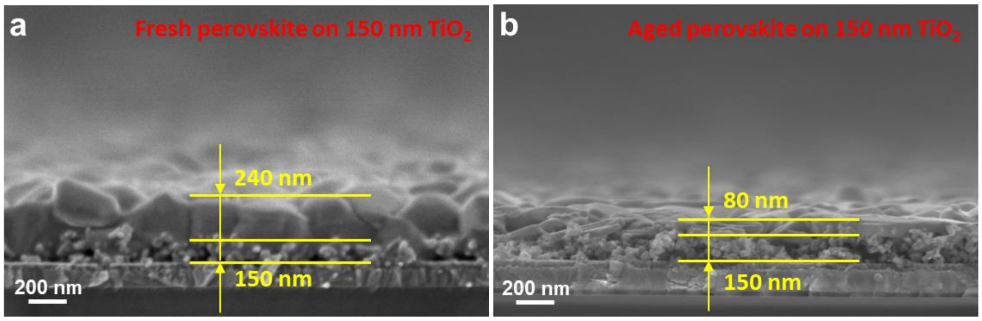

Figure S9. Perovskite film deposited on a 150 nm TiO_2 mesoporous scaffold (a) before and (b) after aging. Aging test was carried out in ambient air at >65% relative humidity. The thickness of the perovskite capping layer or mesoscopic layer is marked in yellow. From the difference of the capping layer's thickness between Figure S9a and b, it can be obtained that TiO_2 mesoporous scaffold is getting close to the external surface of perovskite along with the degradation process, indicating a raised possibility of TiO_2 participating in the interfacial recombination process at the perovskite/HTL interface for the aged devices.