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# Supplementary Information (ESI)

**Interfacial $Al_2TiO_5$ at $TiO_2$/Perovskite Interface to Decrease Carrier Losses in Solar Cells**

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**Table S1.** Complete photovoltaic parameters of all cells.

| | Eff.(%) | $J_{SC}$(mA/cm²) | $V_{OC}$(V) | FF |
|---|---|---|---|---|
| **Ref.** | 16.32 | 21.98 | 0.97 | 0.765457 |
| | 16.21 | 22.33 | 0.97 | 0.748381 |
| | 16.15 | 21.73 | 0.97 | 0.766198 |
| | 15.12 | 22.18 | 0.95 | 0.717574 |
| | 15.2 | 22.53 | 0.97 | 0.695522 |
| | 16.32 | 22.29 | 0.99 | 0.739563 |
| | 14 | 22.49 | 0.94 | 0.662233 |
| | 14.4 | 21.35 | 0.97 | 0.695333 |
| | 14.08 | 20.65 | 0.96 | 0.71025 |
| | 15.2 | 22.18 | 0.95 | 0.721371 |
| | 15.89 | 22.44 | 0.96 | 0.737615 |
| **Al[1]-PSC** | 17.34 | 23.53 | 0.97 | 0.759723 |
| | 16.83 | 23.17 | 0.97 | 0.748835 |
| | 17.85 | 23.81 | 0.99 | 0.757258 |
| | 17.76 | 23.89 | 0.98 | 0.758579 |
| | 16.25 | 21.86 | 0.97 | 0.766358 |
| | 16.33 | 22.18 | 0.97 | 0.759019 |
| | 17.55 | 23.66 | 0.98 | 0.756896 |
| | 17.28 | 23.27 | 0.97 | 0.765554 |
| | 16.8 | 22.41 | 0.99 | 0.757238 |
| | 16.72 | 22.48 | 0.98 | 0.758951 |
| **Al-PSC** | 18.72 | 23.66 | 1 | 0.791209 |
| | 17.255 | 23.09 | 0.99 | 0.754842 |
| | 17.82 | 22.98 | 1 | 0.775457 |
| | 18.43 | 23.28 | 1 | 0.791667 |
| | 16.83 | 22.644 | 0.98 | 0.758411 |
| | 17.01 | 23.15 | 0.96 | 0.765389 |

| | | | | |
|---|---|---|---|---|
| | 16.56 | 22.73 | 0.98 | 0.743421 |
| | 16.44 | 23.21 | 0.98 | 0.722771 |
| | 15.96 | 23.08 | 0.98 | 0.70562 |
| | 17.25 | 23.09 | 0.99 | 0.754623 |
| **$Al^3$-PSC** | 16.47 | 23.25 | 0.98 | 0.722844 |
| | 17.55 | 23.67 | 0.99 | 0.748934 |
| | 16.38 | 22.93 | 0.97 | 0.736441 |
| | 16.3 | 22.45 | 0.96 | 0.75631 |
| | 16.34 | 22.75 | 0.98 | 0.7329 |
| | 15.96 | 22.77 | 0.99 | 0.708002 |
| | 16.3 | 22.45 | 0.96 | 0.75631 |
| | 15.1 | 22.43 | 0.96 | 0.701256 |
| | 15.49 | 23.59 | 0.97 | 0.676942 |

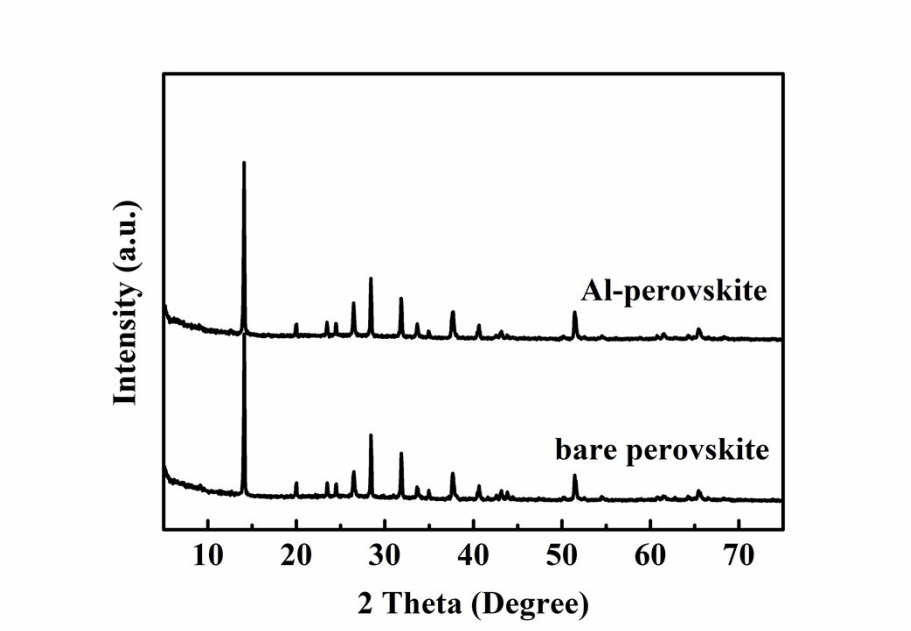


**Figure S1.** X-ray diffraction patterns (XRD) of perovskite films on FTO glass with Al and without Al.

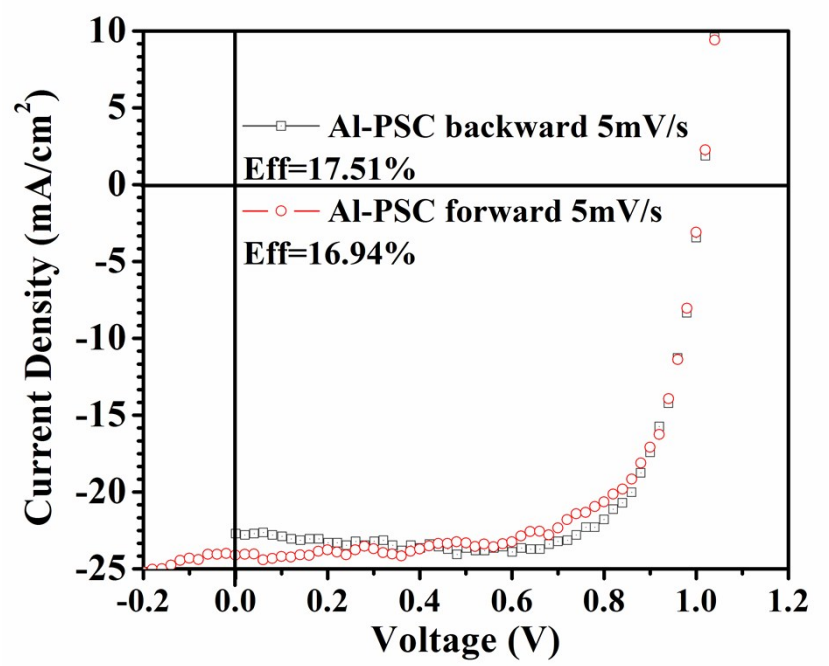


**Figure S2.** J-V curves of Al-PSC measured with forward scan and reverse scan at a scan rate of 5mV/s.

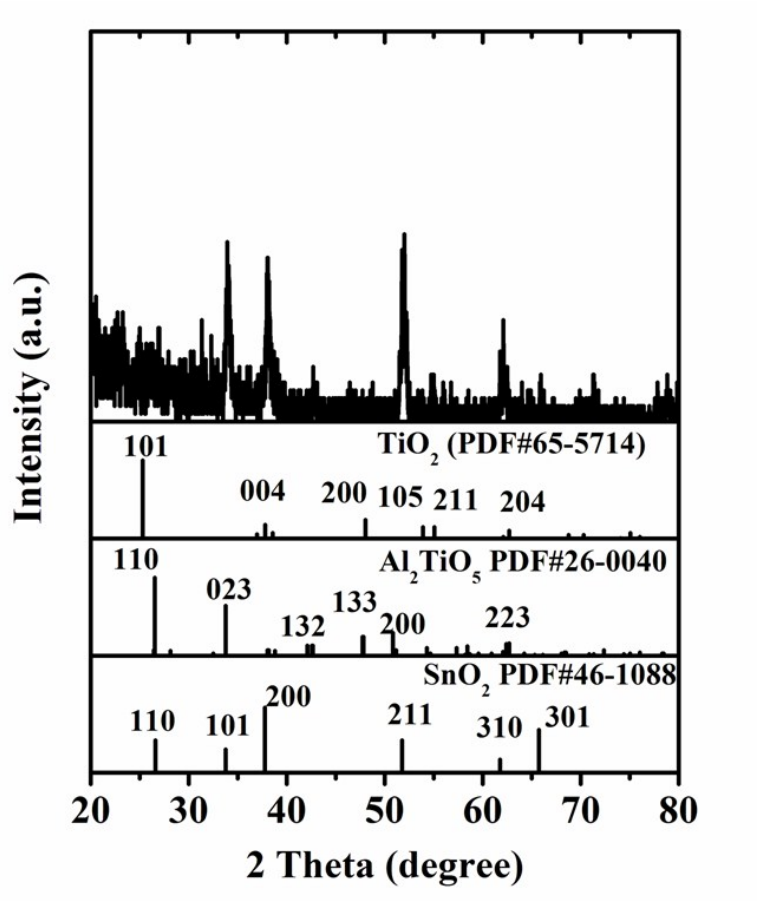


**Figure S3.** Grazing Incidence XRD (GIXRD) patterns of Al-$TiO_2$ film on FTO. The incidence angle for the GIXRD measurement was set as 0.3°.

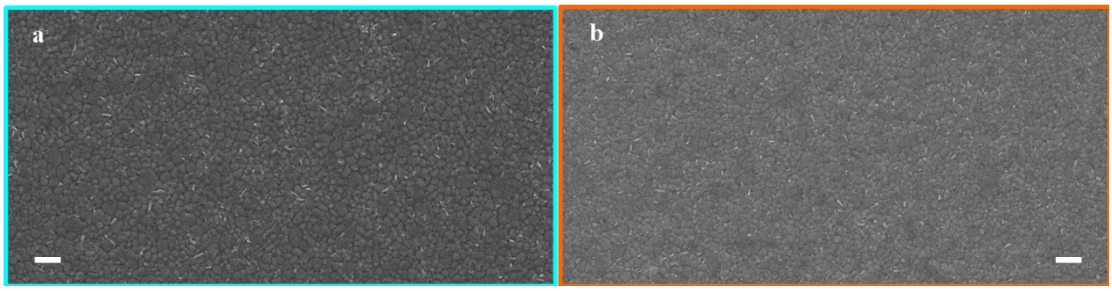


**Figure S4.** Top-view SEM images of bare-perovskite(a) and Al -perovskite(b) film, which shows high quality of perovskite film in large scale.

The morphology of the perovskite thin films was investigated by scanning electron microscopy (SEM). Fig. S5a and S5b indicate that there is no clear difference between the perovskite film and Al-perovskite film. Moreover, SEM images of bare perovskite and Al-perovskite films in large area are shown in Fig. S4, which indicate the uniformity of the $MAPbI_3$ layer extended over large distance. Grain size distributions of the perovskite films are shown in Fig. S5c. The average crystal size of the bare perovskite film is slightly larger compared to that of the Al-perovskite film. This may be induced by the presence of the Al-O, since the properties of perovskite thin films are highly dependent on the underlying layer.[1] The concentration of the Al source used in this work was deliberately controlled to be extremely low (see Experimental Procedure). Therefore, the mere presence of a small quantity of Al at the interface of perovskite/$TiO_2$ film results in slightly smaller perovskite crystals and does not affect the morphology itself. Other workers found that the efficiency of PSCs deteriorated upon decreasing the size of the perovskite crystals; however, our results rather gainsay that conclusion: upon adding $Al^{3+}$ ions the size of the perovskite

crystals decreased slightly and the PSC efficiency improved.

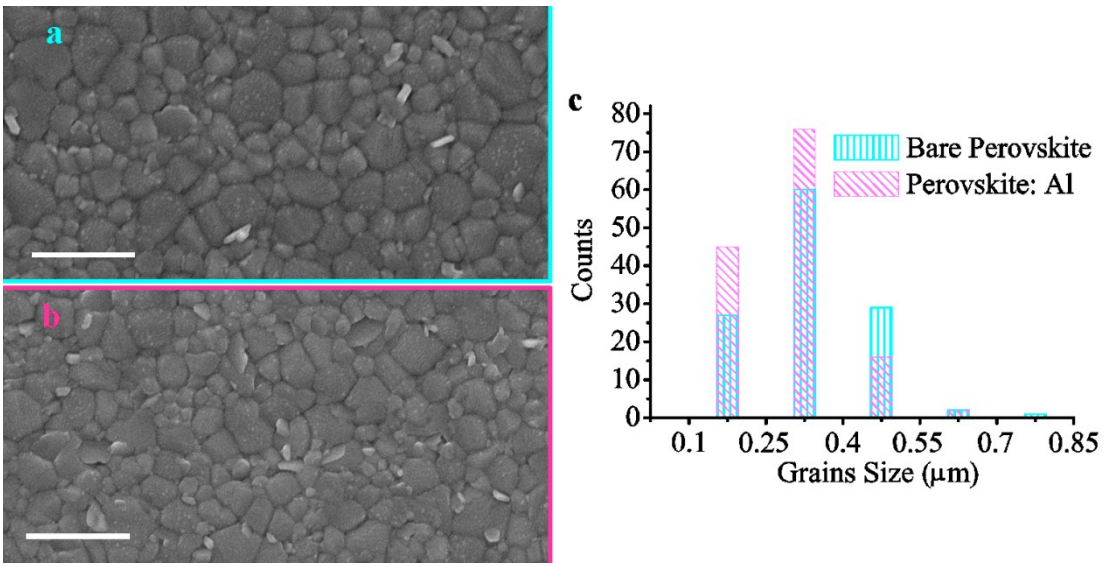


**Figure S5.** Top-view SEM images of (a) perovskite film and (b) Al-perovskite film. The scale bar is 1 μm. (c) The grain size distributions of the perovskite films on $TiO_2$ and Al-$TiO_2$ films.

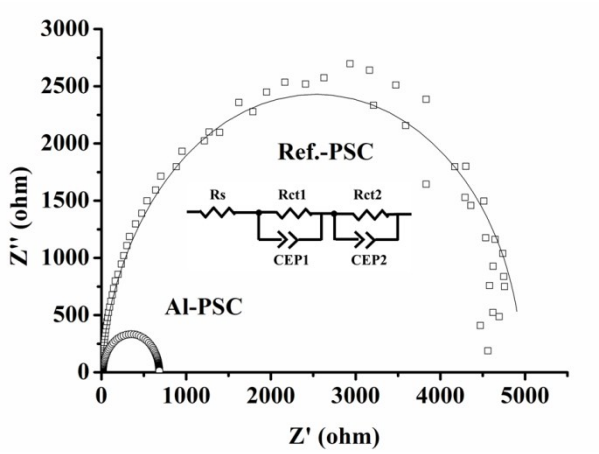


**Figure. S6.** Nyquist plot of the devices measured at a forward bias of 5 mV under simulated solar light of 100 mW/cm$^2$, experimental data are presented in scattered point. Solid lines are the fits obtained using equivalent circuits previously used for solid DSSC with spiro-MeOTAD.[2]

The ref. PSC and Al-PSC have been measured in a frequency range from $10^2$ Hz to $10^6$ Hz. The semicircle of Al-PSC in Figure S6 shows a lower transport resistance.

**Reference:**

1. H. Oga, A. Saeki, Y. Ogomi, S. Hayase and S. Seki, *J Am Chem Soc*, 2014, **136**, 16948-16948.
2. F. Fabregat-Santiago, J. Bisquert, L. Cevey, P. Chen, M. Wang, S. M. Zakeeruddin and M. Gratzel, *J Am Chem Soc*, 2009, **131**, 558-562.