## Supporting Information

# In-situ growth of an opal-like $\mathbf{T i O}_{\mathbf{2}}$ electron transport layer by atomic 

## layer deposition for perovskite solar cells

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Figure S1. SEM image of PS spheres after annealing with 5 nm ALD TiO 2 thickness.


Figure S2. SEM image of crushed spheres.


Figure S3. Transmission spectra of different $\mathrm{TiO}_{2}$ ETLs.


Figure S4. SEM image of 100 nm PS spheres.


Figure S5. SEM image of PS 500 nm spheres with perovskite layer.


Figure S6. SEM image of one layer, two layers and three layers samples with lowmagnification.


Figure S7. XRD pattern of as-prepared $\mathrm{TiO}_{2}$ sample.


Figure S8. SEM image of perovskite layer on single layer $\mathrm{TiO}_{2}$ ETLs.


Figure S9. The integrated current density of ALD $\mathrm{TiO}_{2}$ and Two layers $\mathrm{TiO}_{2}$ based devices.


Figure S10. The repeatability of 26 devices of ALD and two layers samples.


Figure S11. Forward and reverse scan of two layers and ALD $\mathrm{TiO}_{2}$ devices.


Figure S12. Long time measurement of two layers sample.

| Sample | $\mathrm{V}_{\mathrm{oc}}(\mathrm{V})$ | $\mathrm{J}_{\mathrm{sc}}\left(\mathrm{mA} \mathrm{cm}^{-2}\right)$ | Fill Factor (\%) | Efficiency (\%) |
| :---: | :---: | :---: | :---: | :---: |
| Planar | $0.99 \pm 0.02$ | $23.09 \pm 0.28$ | $63.32 \pm 4.14$ | $14.95 \pm 0.53$ |
| Single | $1.03 \pm 0.02$ | $23.63 \pm 0.57$ | $67 \pm 2.94$ | $16.43 \pm 0.75$ |
| Two | $1.05 \pm 0.01$ | $23.76 \pm 0.16$ | $69.96 \pm 1.65$ | $17.5 \pm 0.55$ |
| Three | $0.91 \pm 0.03$ | $23.77 \pm 0.35$ | $57.60 \pm 5.54$ | $12.47 \pm 1.27$ |

Table S1. The photovoltaic parameters of different $\mathrm{TiO}_{2}$ ETLs based devices.

| Sample | $\mathrm{R}_{\mathrm{s}}(\Omega)$ | $\mathrm{R}_{\mathrm{ct1}}(\Omega)$ | $\mathrm{R}_{\mathrm{ct} 2}(\Omega)$ | $\mathrm{R}_{\mathrm{ct}}=\mathrm{R}_{\mathrm{ct1}}+\mathrm{R}_{\mathrm{ct} 2}(\Omega)$ | $\mathrm{R}_{\mathrm{rec}}(\Omega)$ |
| :---: | :---: | :---: | :---: | :---: | :---: |
| Planar | 65.3 | 152.4 | 178.9 | 331.3 | 1181.5 |
| Single | 52.4 | 155.1 | 235.1 | 390.2 | 1277.1 |
| Two | 49.4 | 132.9 | 121.0 | 253.9 | 2431.6 |
| Three | 22.3 | 208.4 | 64.87 | 273.3 | 11323 |

Table S2. The fitted value of $\mathrm{R}_{\mathrm{ct}}$ and $\mathrm{R}_{\mathrm{rec}}$ of different $\mathrm{TiO}_{2}$ ETLs based devices.

