

Electronic supplementary information

Optimization of a Compact Layer of TiO₂ via Atomic-layer Deposition for High-performance Perovskite Solar Cells

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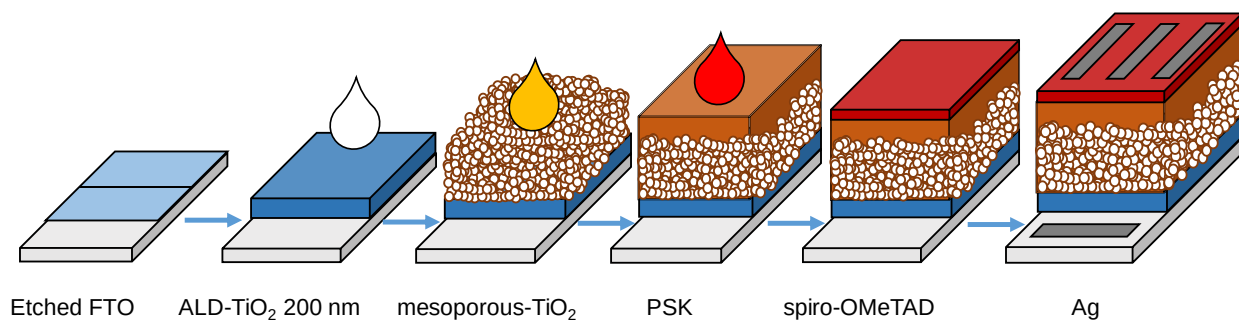


Figure S1. Schematic diagram of the preparation of our PSC layer by layer with configuration glass/FTO/ALD-TiO₂/*m*-TiO₂/MAPbI₃/spiro-OMeTAD/Ag

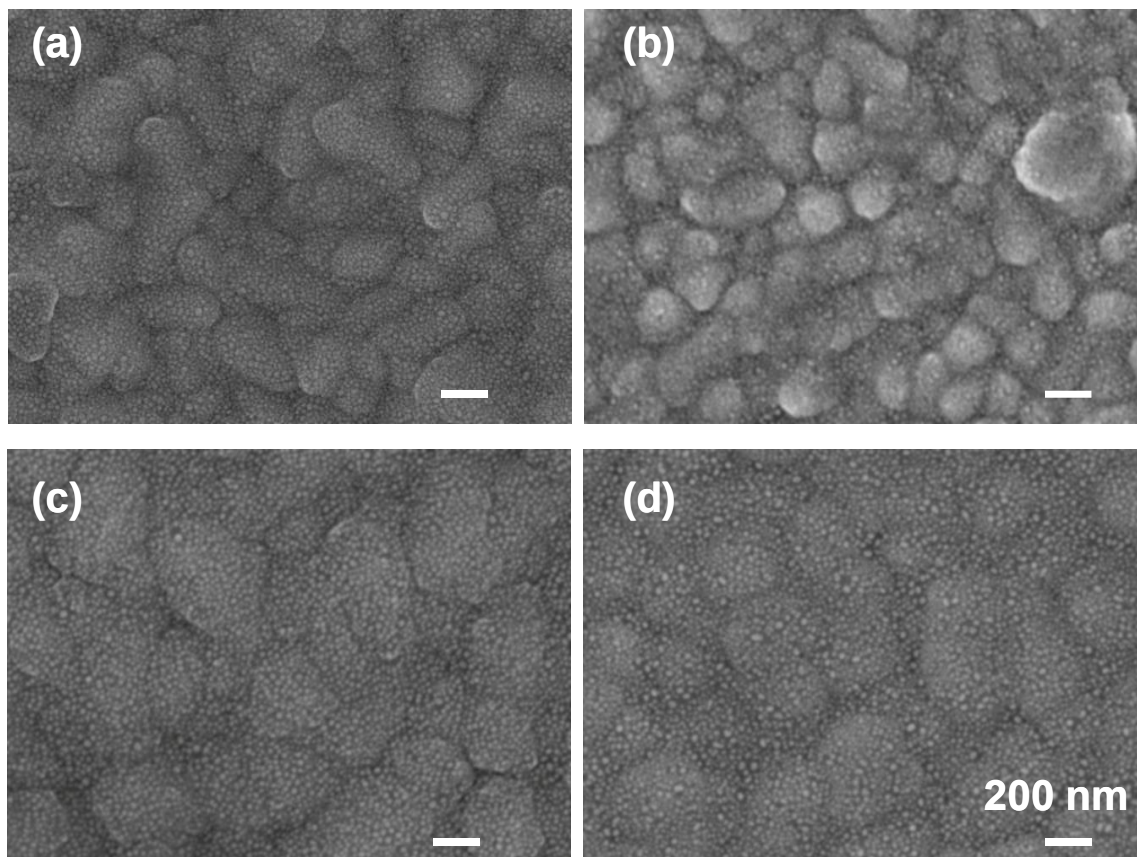


Figure S2. Top-view images of SEM of the ALD-TiO₂ film with the thickness of 50 nm (a), 100 nm (b), 200 nm (c), 400 nm (d) on the FTO substrate, respectively. 3-nm-thick Au was sputtered on the TiO₂ film for the SEM measurement.

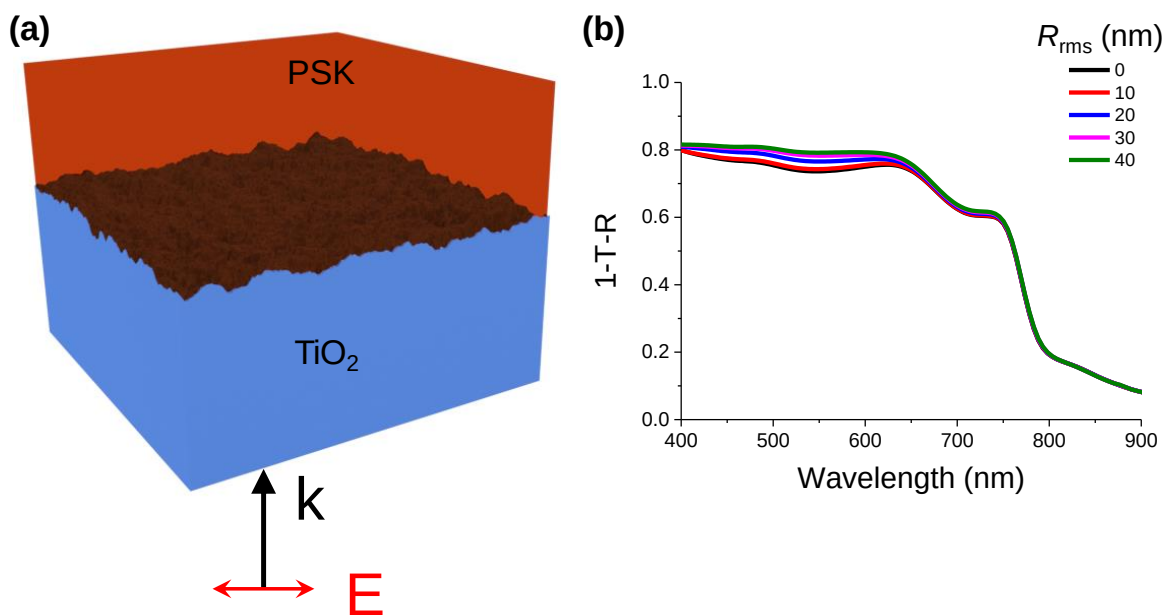


Figure S3. (a) Model for the calculation of the FDTD numerical simulations. (b) 1-T-R spectrum obtained by FDTD numerical simulation. The FDTD simulation was performed with the Lumerical FDTD solution software package on a discrete, non-uniformly spaced mesh with a maximum resolution 2.0 nm. The TiO_2 substrate was assumed to behave as a dielectric with a refractive index $n = 2.5$. In the simulation, a linear polarized plane wave was irradiated onto the structures from TiO_2 at a normal incidence angle; the surface roughness of TiO_2 at the interface of TiO_2 and PSK was varied from 0 to 40 nm as a root-mean-square roughness (R_{rms}). The optical properties of the PSK were obtained with the data from L. J. Phillips et al. [*]

[*] L. J. Phillips, et. al., Dispersion relation data for methylammonium lead triiodide perovskite deposited on a (100) silicon wafer using a two-step vapour-phase reaction process, *Data in Brief*, 5, 926-928 (2015).

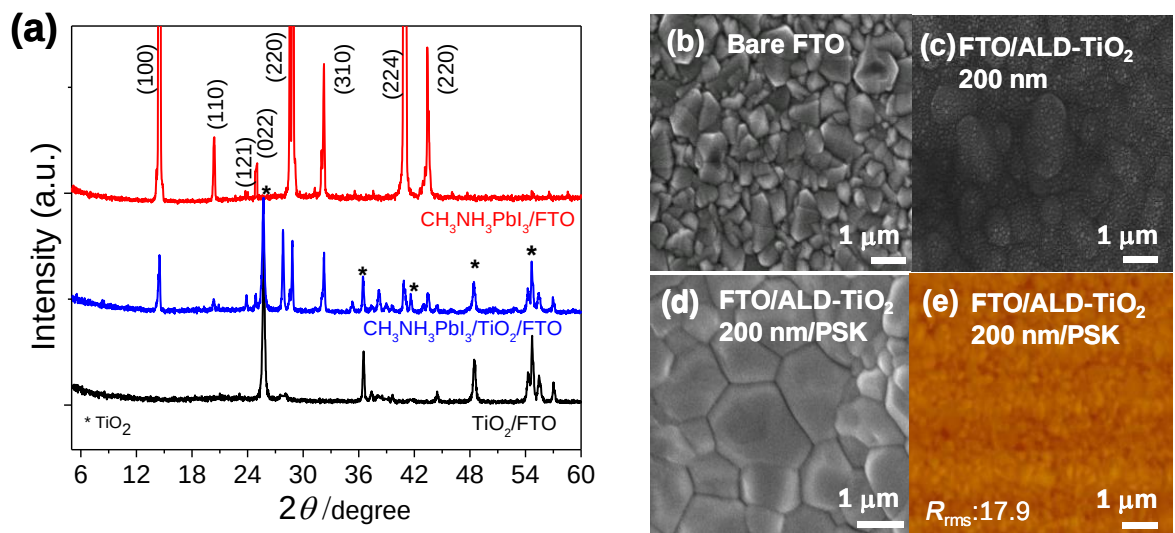


Figure S4. (a) XRD of ALD-TiO₂ on FTO (black line), MAPbI₃ on FTO (red) and FTO/ALD-TiO₂/MAPbI₃ (blue). FESEM of (b) Bare FTO. (c) ALD-TiO₂ 200 nm on FTO. (d) FESEM of FTO/ALD-TiO₂ 200 nm/MAPbI₃. (e) AFM of FTO/ALD-TiO₂ 200 nm/MAPbI₃. The asterisks in the figure refer to the planes of TiO₂ in all samples. The magnification is $\times 50000$; the scale bar is 1 μm for all SEM figures. The magnification is $\times 12500$ and the scale bar is 1 μm for all AFM figures.

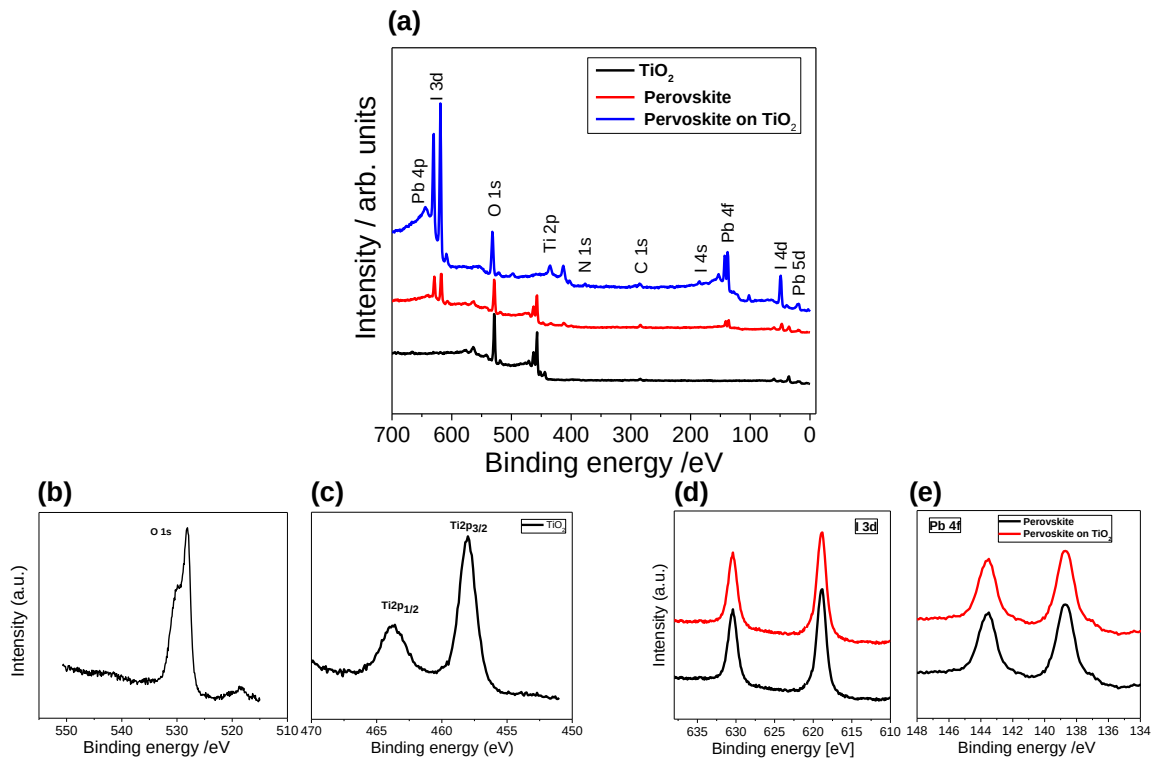


Figure S5. (a) X-ray photoelectron spectra of wide-scan of ALD- TiO_2 , MAPbI₃ and MAPbI₃ on ALD- TiO_2 , respectively. Narrow-range X-ray photoelectron spectra of (b) Ti 2p_{1/2} and Ti 2p_{3/2}, and (c) O 1s for TiO_2 . Narrow-range X-ray photoelectron spectra of (d) Pb 4f and (e) I 3d for MAPbI₃ and MAPbI₃ on ALD- TiO_2 .

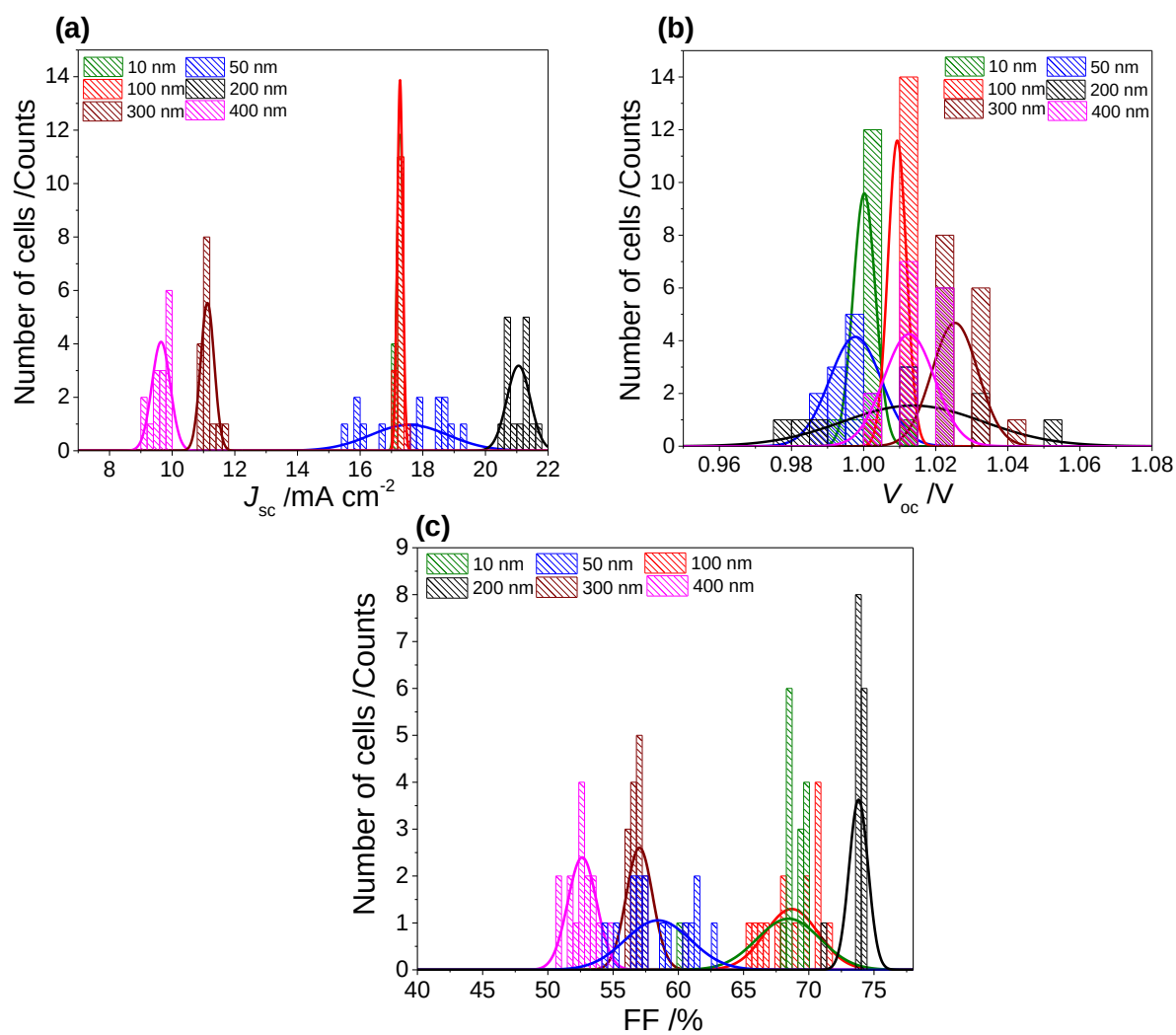


Figure S6. Histograms of J_{sc} , V_{oc} and FF distribution (a, b and c), respectively, of ALD-TiO₂ as HBL with thickness 10, 50, 100, 200, 300 and 400 nm, respectively.

Table S1. Photovoltaic parameters of fifteen PSC devices made of ALD-TiO₂ 10 nm as HBL under simulated AM-1.5G illumination (power density 100 mW cm⁻²) with active area 0.09 cm²

# of cells	$J_{sc}/\text{mA cm}^{-2}$	V_{oc}/V	FF /%	PCE /%
1	17.04	0.99	0.533	9.03
2	17.22	1.00	0.534	9.19
3	17.16	1.00	0.534	9.16
4	17.16	1.00	0.536	9.20
5	17.21	1.00	0.538	9.28
6	17.22	1.00	0.539	9.29
7	17.23	1.00	0.542	9.33
8	17.29	1.00	0.541	9.39
9	17.19	1.00	0.539	9.29
10	17.34	1.00	0.543	9.46
11	17.37	1.00	0.543	9.42
12	17.32	1.00	0.544	9.41
13	17.39	1.01	0.544	9.55
14	17.38	1.00	0.545	9.42
15	17.32	1.00	0.545	9.46
average	17.25±0.1	1.00±0.003	0.54±0.004	9.33±0.14

Table S2. Photovoltaic parameters of fifteen PSC devices made of ALD-TiO₂ 50 nm as HBL under simulated AM-1.5G illumination (power density 100 mW cm⁻²) with active area 0.09 cm²

# of cells	$J_{sc}/\text{mA cm}^{-2}$	V_{oc}/V	FF /%	PCE /%
1	17.74	1.00	0.549	9.74
2	17.80	0.99	0.546	9.64
3	17.86	1.00	0.544	9.67
4	16.06	1.01	0.608	9.86
5	15.59	0.99	0.593	9.14
6	15.96	1.00	0.591	9.40
7	18.89	0.99	0.542	10.17
8	19.20	1.00	0.512	9.79
9	18.53	1.00	0.564	10.45
10	17.52	1.01	0.565	9.97
11	16.61	1.00	0.580	9.60
12	15.91	0.99	0.589	9.30
13	18.53	1.00	0.538	9.94
14	18.65	0.99	0.535	9.87
15	18.67	1.01	0.523	9.86
average	17.56±1.23	0.99±0.01	0.559±0.03	9.76±0.33

Table S3. Photovoltaic parameters of fifteen PSC devices made of ALD-TiO₂ 100 nm as HBL under simulated AM-1.5G illumination (power density 100 mW cm⁻²) with active area 0.09 cm²

# of cells	J_{sc} /mA cm ⁻²	V_{oc} /V	FF /%	PCE /%
1	17.18	1.01	0.733	12.73
2	17.25	1.01	0.734	12.80
3	17.11	1.00	0.703	12.15
4	17.17	1.01	0.732	12.71
5	17.23	1.01	0.734	12.79
6	17.21	1.01	0.733	12.75
7	17.28	1.01	0.733	12.81
8	17.28	1.01	0.735	12.83
9	17.24	1.01	0.734	12.78
10	17.30	1.01	0.734	12.85
11	17.34	1.01	0.735	12.90
12	17.35	1.01	0.734	12.88
13	17.41	1.01	0.736	12.97
14	17.39	1.01	0.736	12.96
15	17.37	1.01	0.735	12.93
average	17.27±0.09	1.01±0.003	0.732±0.008	12.78±0.19

Table S4. Photovoltaic parameters of fifteen PSC devices made of ALD-TiO₂ 200 nm as HBL under simulated AM-1.5G illumination (power density 100 mW cm⁻²) with active area 0.09 cm²

# of cells	$J_{sc}/\text{mA cm}^{-2}$	V_{oc}/V	FF /%	PCE /%
1	20.55	1.05	0.669	14.51
2	20.67	1.02	0.689	14.66
3	20.67	1.02	0.701	14.75
4	20.77	1.02	0.707	15.03
5	21.08	0.98	0.675	13.88
6	20.76	1.02	0.695	14.75
7	20.99	1.01	0.652	13.86
8	21.39	0.98	0.665	13.99
9	21.35	0.99	0.660	13.90
10	21.29	1.02	0.644	13.95
11	21.49	1.01	0.645	14.04
12	21.27	1.03	0.637	13.93
13	21.79	1.03	0.665	14.97
14	21.20	1.02	0.687	14.77
15	20.75	1.01	0.698	14.56
average	21.05±0.37	1.01±0.02	0.673±0.02	14.37±0.44

Table S5. Photovoltaic parameters of fifteen PSC devices made of ALD-TiO₂ 300 nm as HBL under simulated AM-1.5G illumination (power density 100 mW cm⁻²) with active area 0.09 cm².

# of cells	$J_{sc}/\text{mA cm}^{-2}$	V_{oc}/V	FF /%	PCE /%
1	11.78	1.04	0.576	7.07
2	11.42	1.03	0.695	8.18
3	11.08	1.03	0.685	7.80
4	10.95	1.03	0.674	7.57
5	10.96	1.03	0.686	7.71
6	11.05	1.03	0.670	7.62
7	10.94	1.03	0.674	7.56
8	10.98	1.02	0.687	7.73
9	11.02	1.02	0.685	7.72
10	11.09	1.02	0.672	7.61
11	11.21	1.02	0.683	7.83
12	11.19	1.02	0.675	7.70
13	11.07	1.02	0.672	7.60
14	11.09	1.02	0.689	7.81
15	11.02	1.02	0.685	7.72
average	11.12±0.22	1.03±0.01	0.674±0.03	7.68±0.23

Table S6. Photovoltaic parameters of fifteen PSC devices made of ALD-TiO₂ 400 nm as HBL under simulated AM-1.5G illumination (power density 100 mW cm⁻²) with active area 0.09 cm²

# of cells	$J_{sc}/\text{mA cm}^{-2}$	V_{oc}/V	FF /%	PCE /%
1	9.62	1.01	0.494	4.80
2	9.69	1.02	0.485	4.79
3	9.72	1.02	0.481	4.77
4	9.59	1.02	0.491	4.80
5	9.44	1.02	0.493	4.75
6	9.44	1.02	0.494	4.76
7	9.26	1.01	0.499	4.67
8	9.16	1.01	0.499	4.62
9	9.17	1.01	0.503	4.66
10	9.93	1.02	0.518	5.25
11	9.99	1.01	0.504	5.08
12	9.89	1.00	0.505	4.99
13	9.98	1.01	0.474	4.78
14	9.92	1.00	0.473	4.69
15	9.87	1.01	0.481	4.79
average	9.46±0.29	1.01±0.01	0.493±0.01	4.81±0.17