Supporting Information

Interfacial Modification via Aniline Molecules with Multiple Active Sites for Performance Enhancement of n-i-p Perovskite Solar Cells

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First-principle calculations:

In this work, Device Studio program provides a number of functions for performing visualization, modeling and simulation. And based on density functional theory (DFT), all our calculations are performed using DS-PAW software integrated in Device Studio program[1, 2]. The generalized gradient approximation (GGA) of the Perdew-Burke-Ernzerhof (PBE) functional is used as the exchange-correlation functional[3]. A vacuum thickness of 20 Å is built to avoid the interactions between adjacent layers. Slab models of three-layered $PbI_2/C_9H_{10}N_2O_4$ and two-layered PbI_2/ $C_9H_{10}N_2O_4$ and their junction were constructed for simulation[4]. The Mon-khorst-Pack k-point meshes of 2 × 1 × 4 is adopted for geometrical optimization and electronic structure calculations in unit cell of PbI_2/ $C_9H_{10}N_2O_4$. The cutoff factor is set as 1. During optimization, all the structures were fully relaxed with the force and total energy convergence criteria of 0.883 eV Å and 1e-4eV, respectively. During adsorption and diffusion possesses, the DFT-D2 method with Grimme correction is adopted to describe the long-range van der Waals interactions[5, 6]. Besides, three-layered of lead iodide and two-layered of lead iodide are used as substrates, and the k-points is set as 2 × 1 × 4.

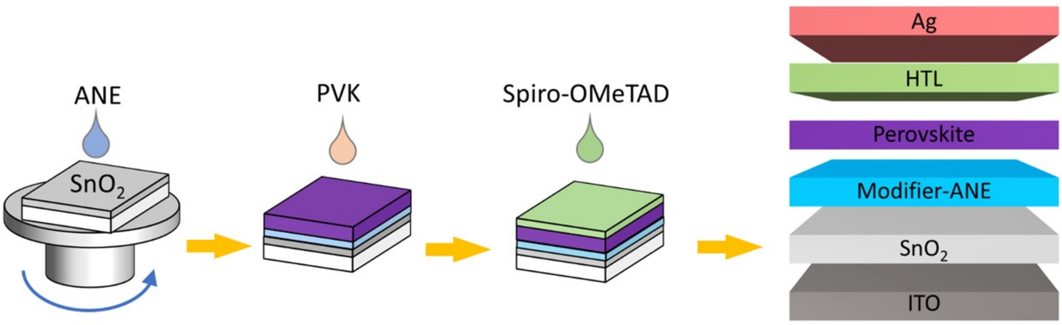

Fig. S1. The preparation process of the PSCs used in this work.

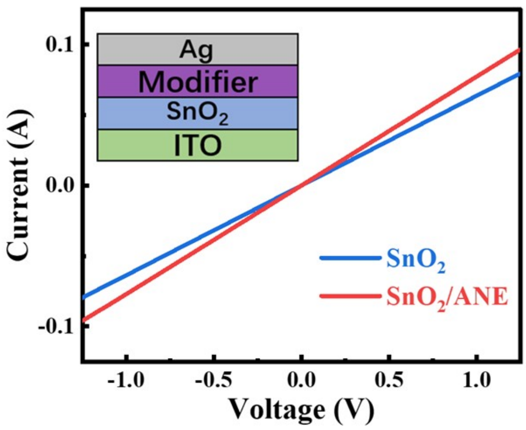

Fig. S2. I-V curves of the SnO_2 films without and with ANE.

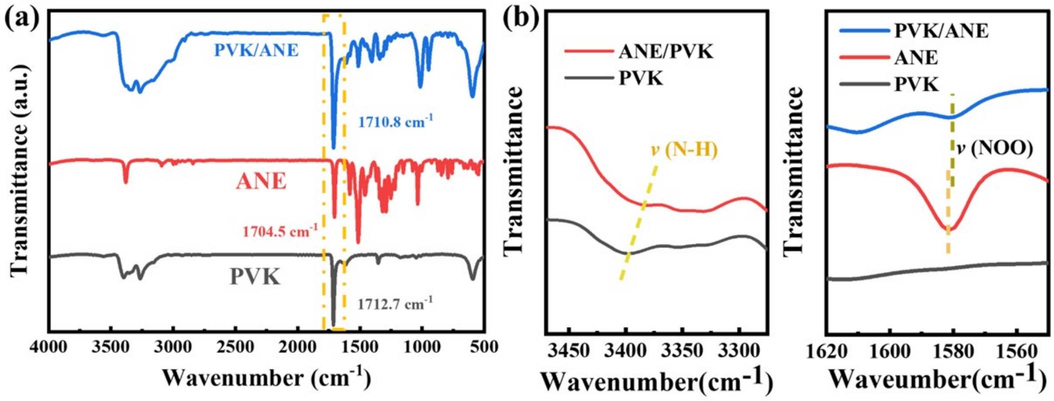

Fig. S3. FTIR spectra of PVK, ANE and PVK modified with ANE.

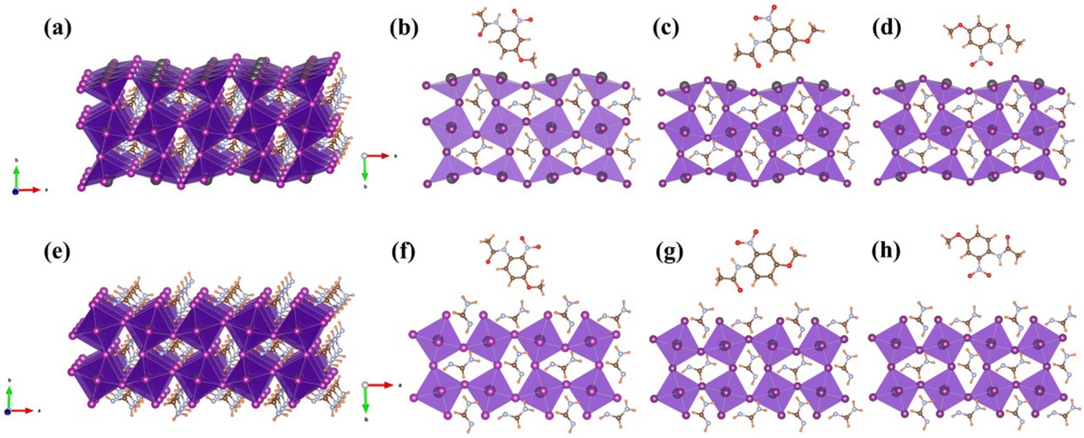

Fig. S4. Structure diagrams of unadsorbed substrate for (a) PbI_2 and (e) FAI terminations. The interfaces for the inorganic PbI_2 termination in contact with the different sites of ANE molecules (b-d), and the models were C-O-C, C=O and NOO sites, respectively. The interfaces for the organic FAI termination in contact with the different sites of ANE molecules (f-h), and the models were C-O-C, C=O and NOO sites, respectively.

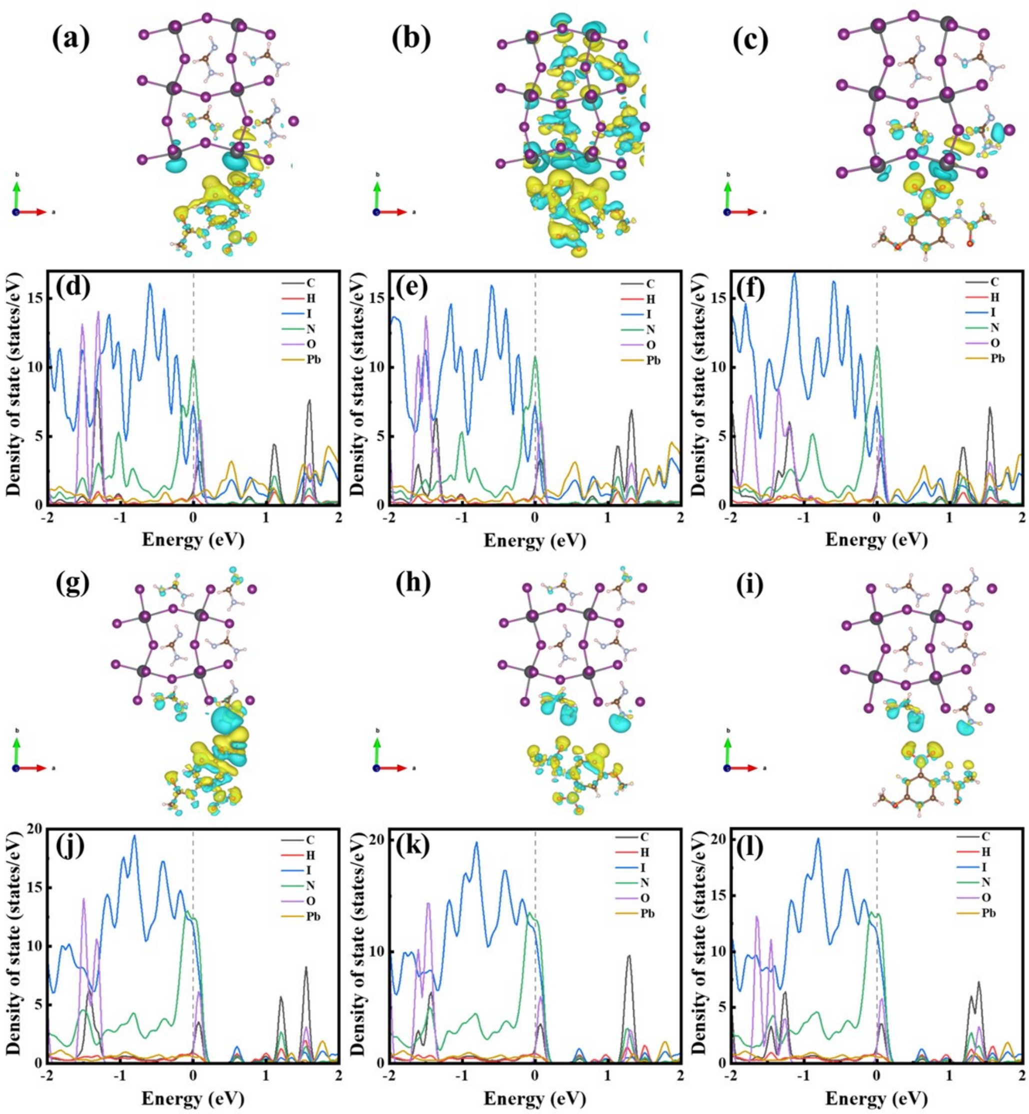

Fig. S5. Differential charge density and corresponding DOS diagram of contact between different active sites of ANE and PbI_2 and FAI terminations.

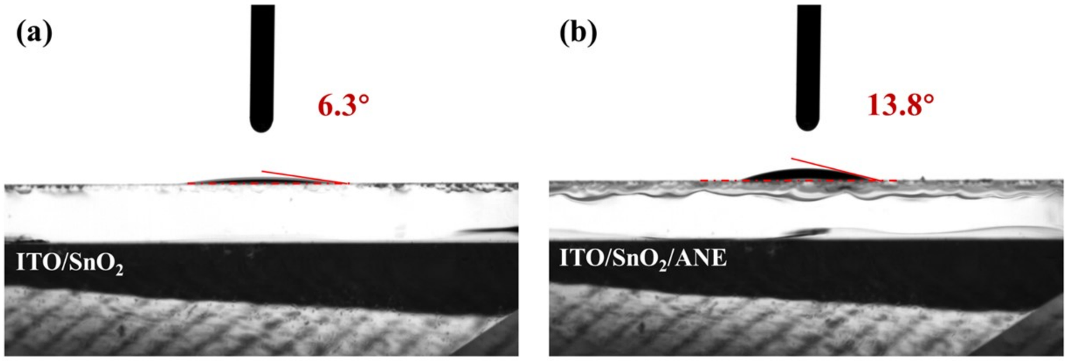

Fig. S6. Contact angle measurements of the (a) control and (b) modified SnO_2 films with ANE.

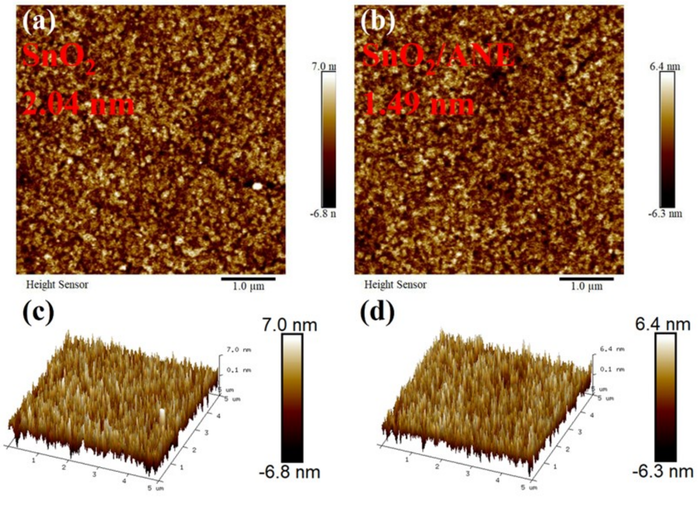

Fig. S7. AFM images of (a) the pristine SnO_2 and (b) SnO_2/ANE films. 3D AFM images of (c) SnO_2 and (d) SnO_2/ANE films.

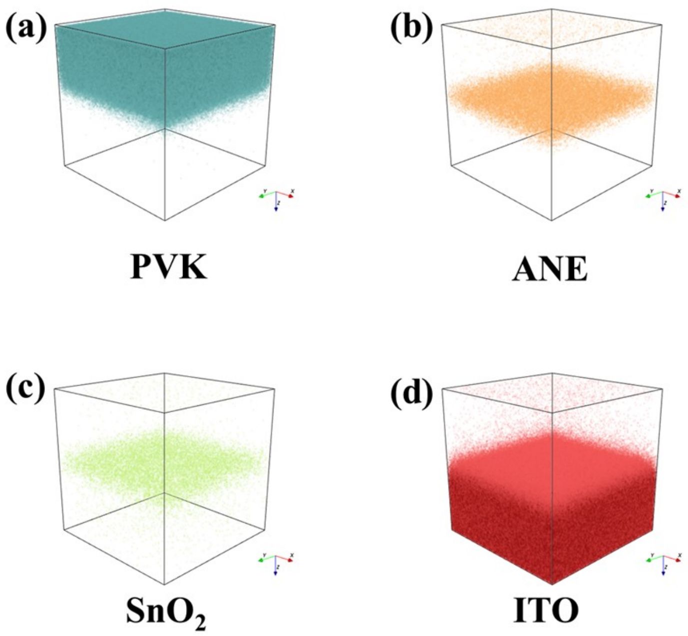

Fig. S8. Tof-SIMS 3D-depth profiles of the target devices with the structure of ITO/SnO_2/ANE/perovskite.

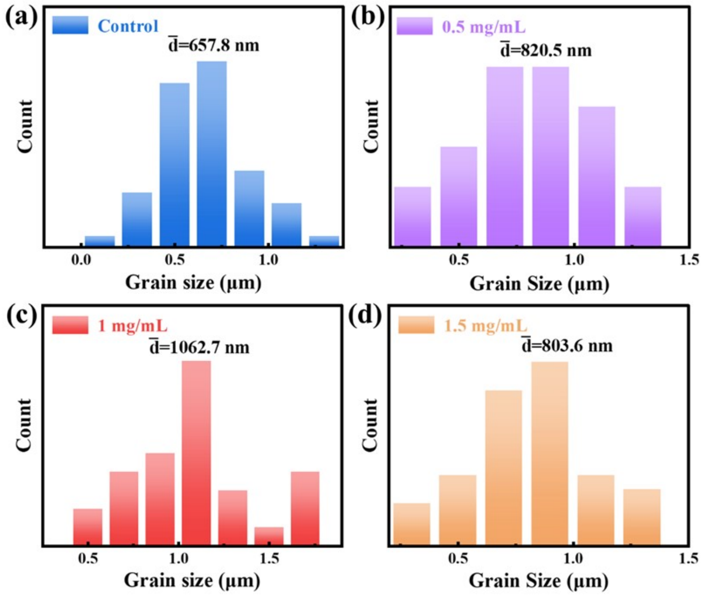

Fig. S9. Statistical distribution of PVK grains modified with different concentration of ANE.

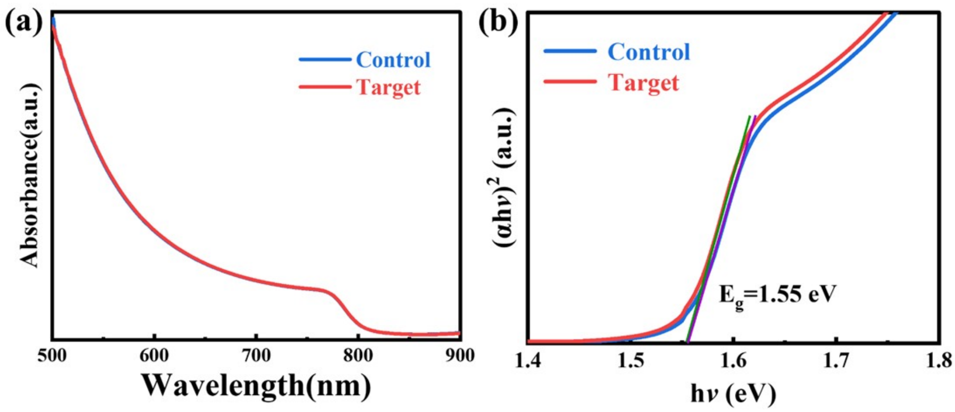

Fig. S10. (a) UV-Vis absorption spectra of the perovskite films without and with ANE passivation. The corresponding Tauc plot is shown in (b).

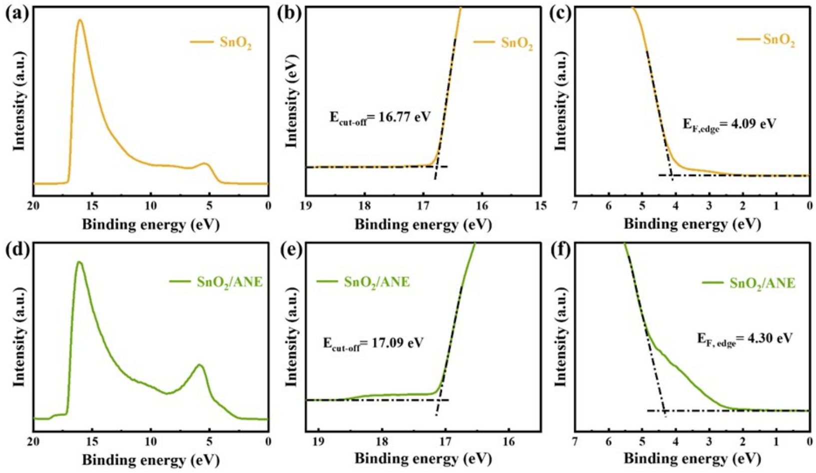

Fig. S11. (a) and (d) UPS full spectra. (b) and (e) UPS spectra describing the cut-off energy ($E_{cut-off}$). (c) and (f) UPS spectra describing Fermi edge ($E_{F,\ edge}$).

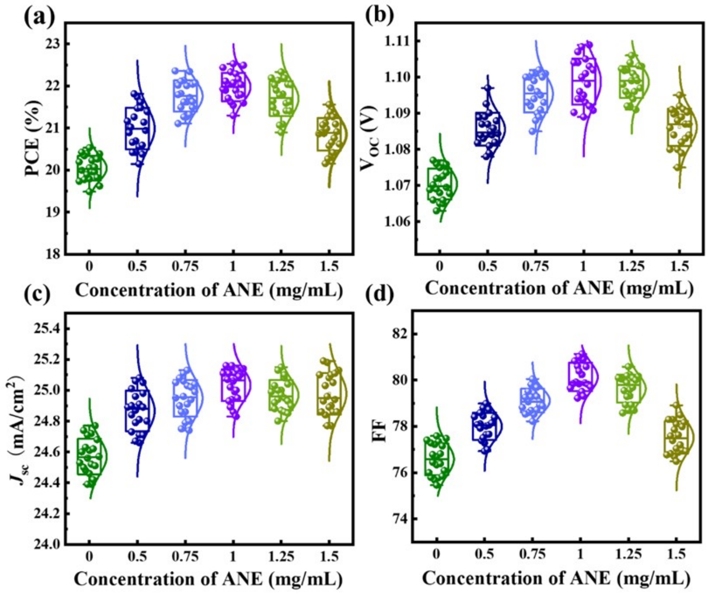

Fig. S12. Statistics of (a) PCE, (b) V_{OC}, (c) J_{SC}, and (d) FF of PSCs based on SnO_2 ETL modified by different concentrations of ANE.

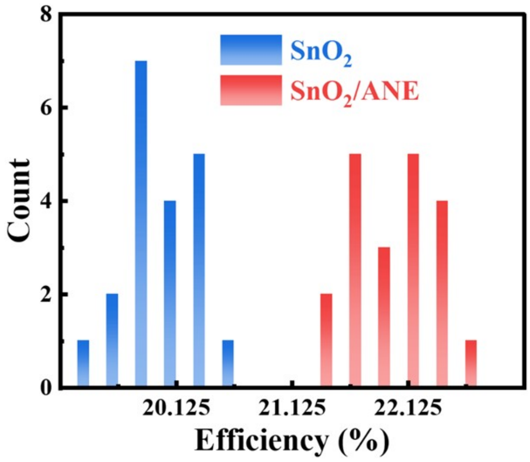

Fig. S13. Statistical distribution of PCEs of the PSCs based on the bared and modified SnO_2 by ANE. The statistical data were obtained from 30 cells for each group.

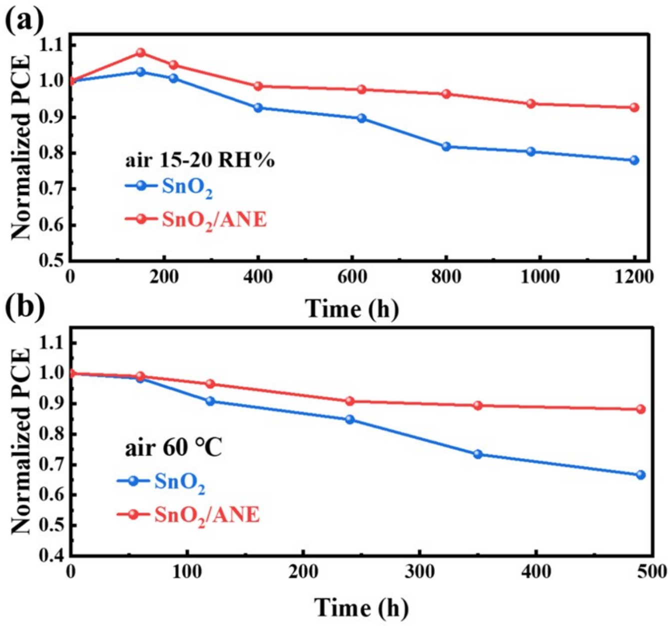

Fig. S14. (a) Moisture stability of the unsealed control and modified PSCs under an ambient condition with a relative humidity of 15–20% under dark. (b) Thermal stability of the unencapsulated control and modified devices at 60 ℃ in the air, under dark.

Table S1. Fitted results of TRPL curves of the perovskite films deposited on glass substrates with or without modifiers measured from perovskite side. The PVK represents the perovskite layer.

Samples	τ_1 (ns)	A_1 (%)	τ_2 (ns)	A_2 (%)	τ_{ave} (ns)
Glass/PVK	147.94	18.36	451.97	81.64	431.12
Glass/ANE/PVK	265.30	22.74	763.08	77.26	716.87

Table S2. Fitted results of TRPL curves of the perovskite films deposited on SnO_2 substrates with or without modifiers measured from perovskite side. The PVK represents the perovskite layer.

Samples	τ_1 (ns)	A_1 (%)	τ_2 (ns)	A_2 (%)	τ_{ave} (ns)
ITO/SnO_2/PVK	144.69	6.03	507.67	93.97	501.15
ITO/SnO_2/ANE/PVK	115.81	22.03	351.32	77.97	331.25

Table S3. The fitted EIS parameters of the devices based on SnO_2 and SnO_2/ANE ETLs.

ETL	R_s (Ω)	R_{ct} (Ω)	R_{rec} (Ω)
SnO_2	29.05	11806	49720
SnO_2/ANE	43.01	7583	78330

Table S4. Photovoltaic parameters of the average performing control and target devices measured in RS and FS.

Device		J_{SC} (mA/cm^2)	V_{OC} (V)	FF	PCE (%)	R_s (Ω)	R_{sh} (Ω)
Control	FS	24.52	1.068	0.7595	19.88	4.12	6173.58
	RS	24.53	1.072	0.7682	20.20	4.07	7052.25
Target	FS	25.12	1.106	0.8104	22.51	3.30	9034.97
	RS	25.14	1.108	0.8110	22.59	3.11	11084.62

References

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