

## Supporting Information

### Large-area grown ultrathin molybdenum oxide for label-free sensitive biomarker detection

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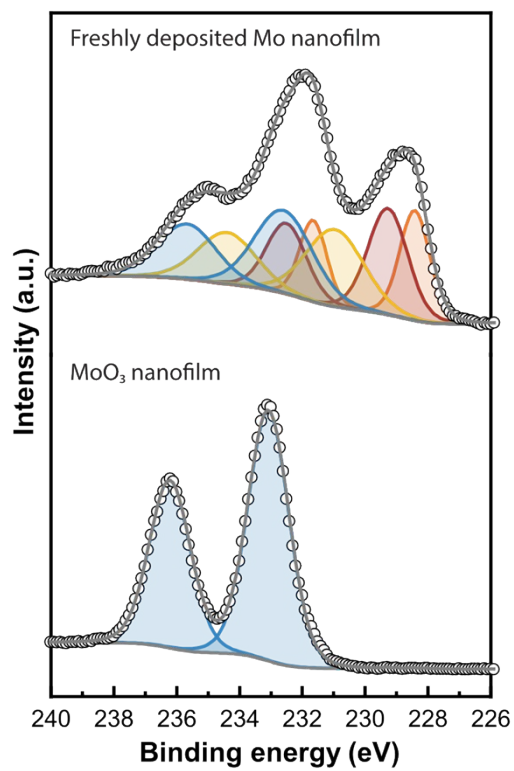
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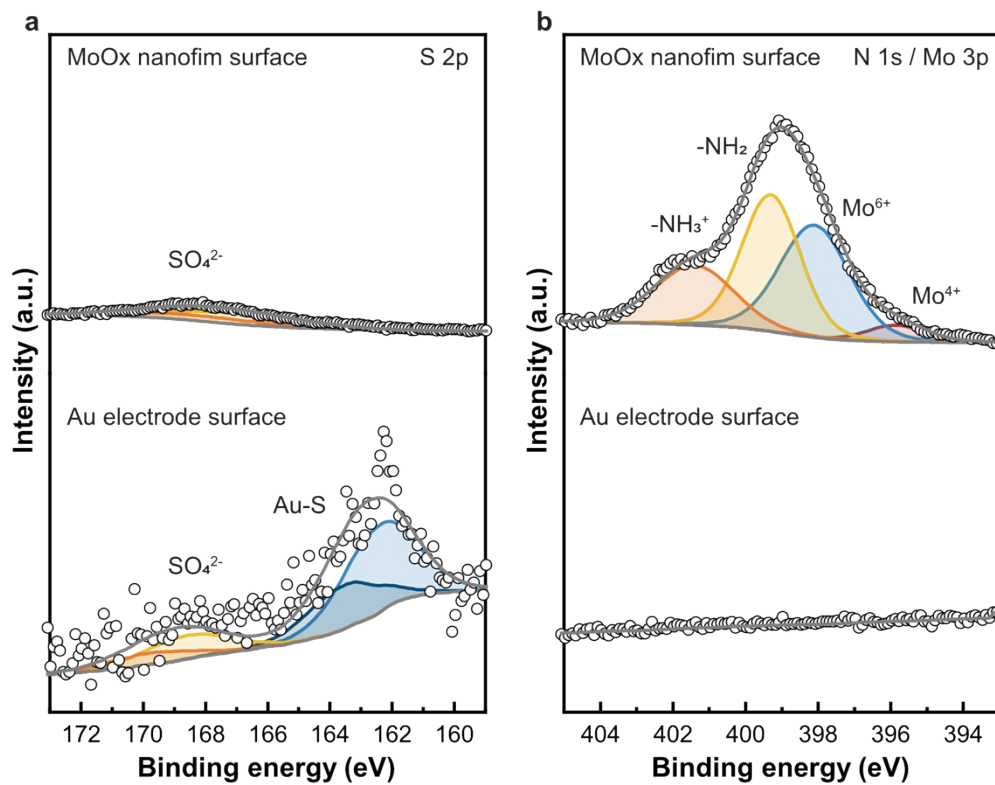
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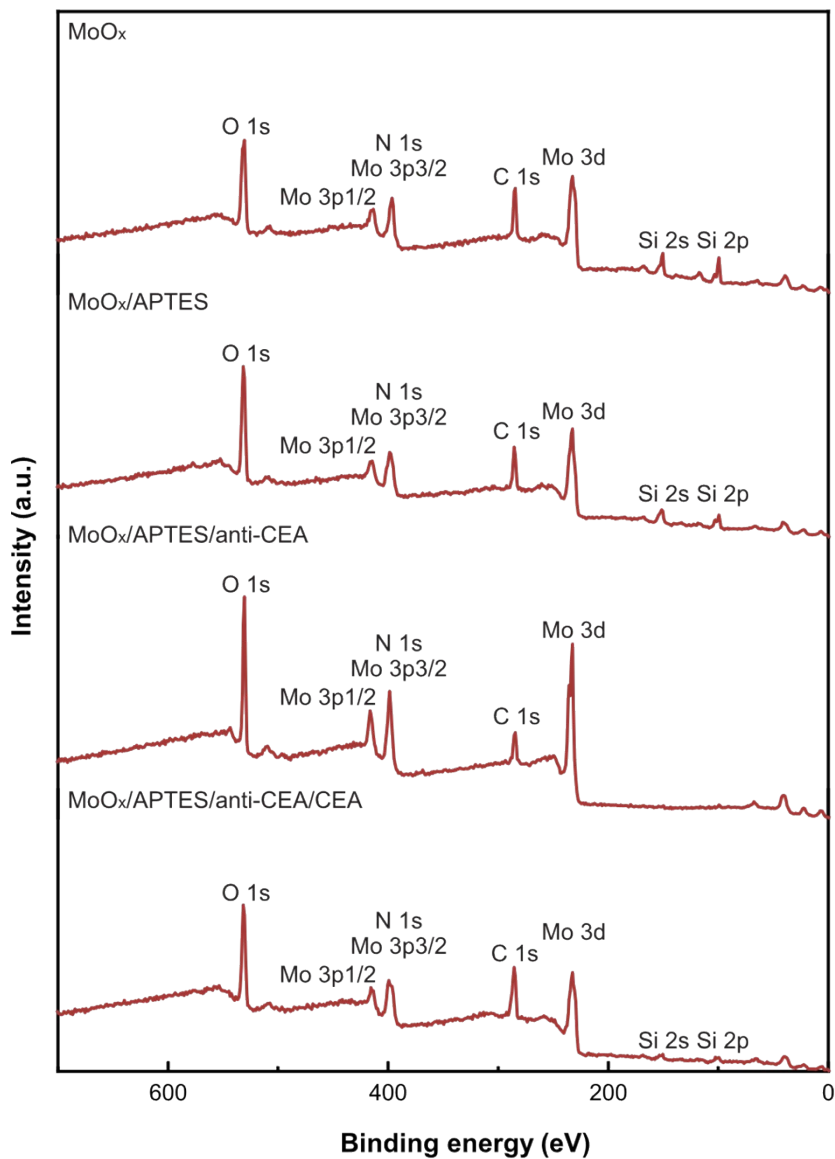
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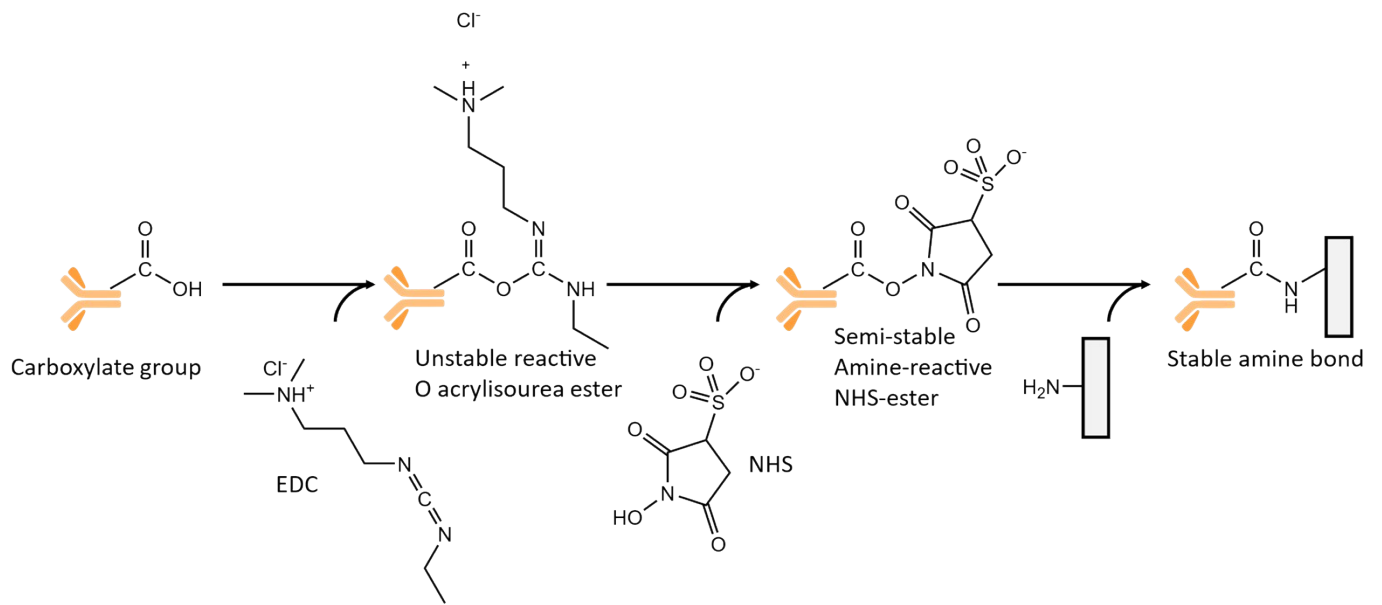
**Figure S1.** The XPS spectra of freshly deposited ultrathin Mo film (top) and MoO<sub>3</sub> nanofilm after oxidation process (bottom). The fitted molybdenum metal peaks (228.4 eV and 231.6 eV), Mo<sup>4+</sup> peaks (229.2 eV and 232.4 eV), Mo<sup>5+</sup> peaks (231.0 eV and 234.2 eV), and Mo<sup>6+</sup> peaks (232.7 eV and 235.8 eV) are indicated in the graph.



**Figure S2.** Comparison of high resolution XPS spectra of MoO<sub>x</sub> nanofilm surface (top) and Au electrode surface (bottom) after functionalization with APTES for (a) N 1s; (b) S 2p. The low signal-to-noise ratio of S2p is due to the dual factors of sole S atom in each 1-Dodecanethiol molecule<sup>1</sup> and low concentration (10 mM) of 1-Dodecanethiol molecule incubated in ethanol solution [1, 2].



**Figure S3.** XPS survey for MoO<sub>x</sub>, MoO<sub>x</sub>/APTES, MoO<sub>x</sub>/APTES/anti-CEA, and MoO<sub>x</sub>/APTES/anti-CEA/CEA.



**Figure S4.** Immobilization of anti-CEA on an amine group functionalized surface mediated by EDC and NHS coupling.

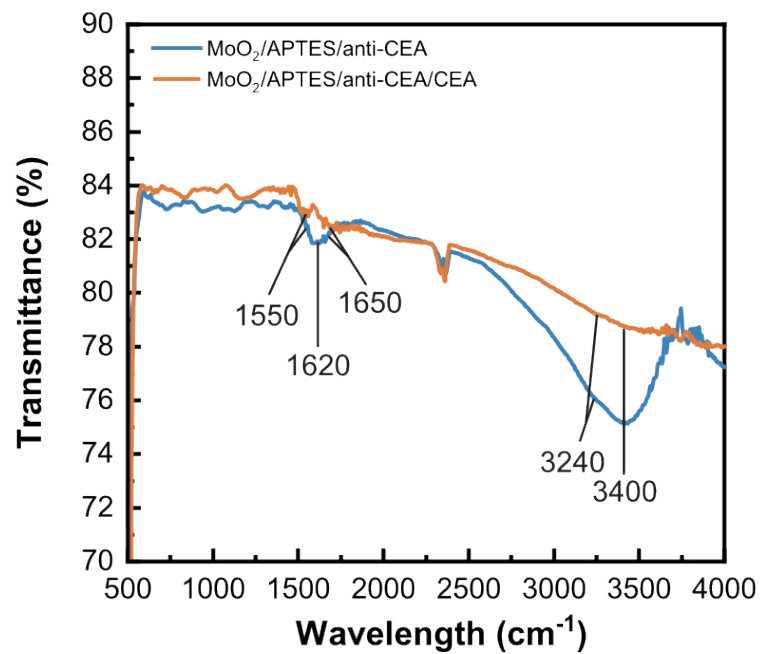
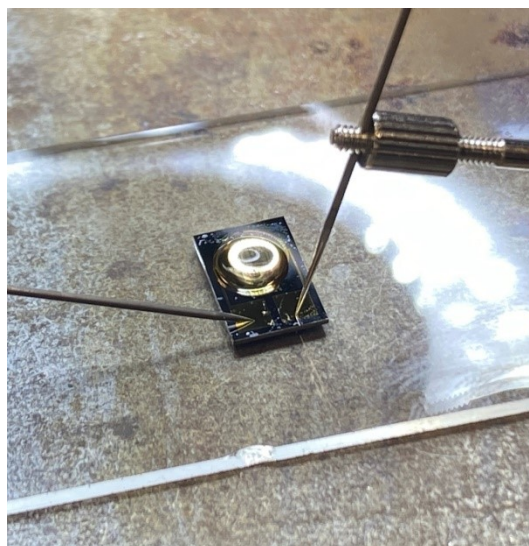


Figure S5. FTIR spectra of MoO<sub>x</sub>/APTES/anti-CEA, and MoO<sub>x</sub>/APTES/anti-CEA/CEA.

**Table S1.** Detailed composition of ultrathin MoO<sub>x</sub> film in different functionalization stage calculated from the XPS

	Ref	MoO <sub>x</sub> /APTES	MoO <sub>x</sub> /APTES/anti-CEA	MoO <sub>x</sub> /APTES/anti-CEA/CEA
Mo <sup>6+</sup> [%]	16.55	25.83	12.94	11.01
Mo <sup>5+</sup> [%]	28.64	21.77	24.55	25.12
Mo <sup>4+</sup> [%]	54.80	52.4	62.52	63.87
x	2.31	2.37	2.25	2.24



**Figure S6.** Electrical measurements on the developed  $\text{MoO}_x$ -IDE biosensing chip with output characteristic at a fixed 0.1 V source voltage.



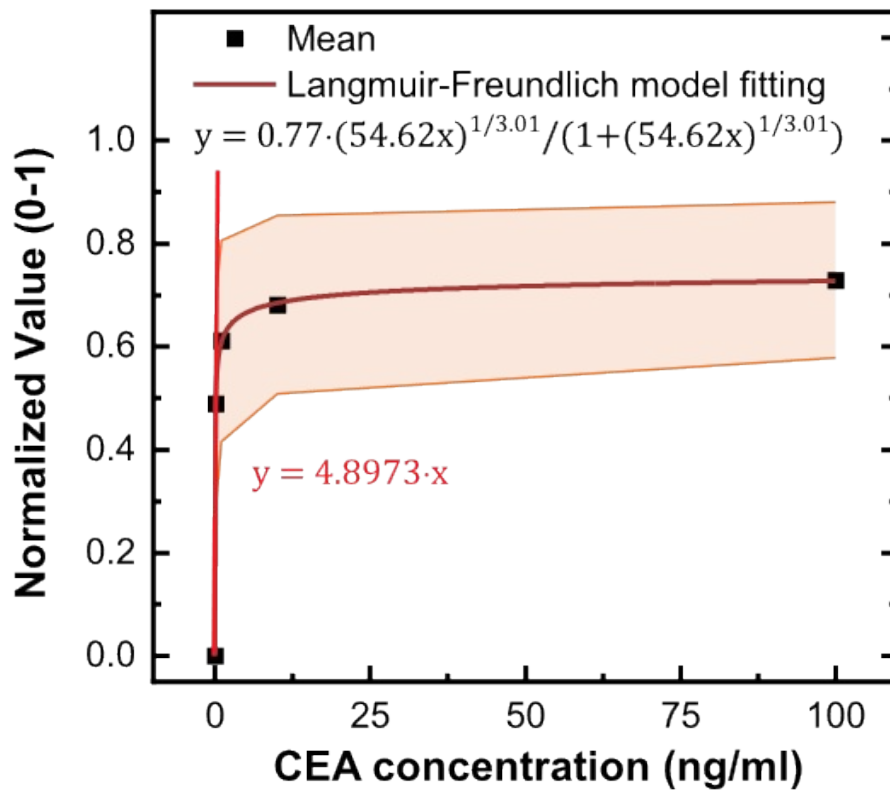


Figure S7. the slope of the linear region of the Langmuir-Freundlich model fitted curve.

**Table S2.** Comparisons of the sensing performance of developed electronic biosensing chip with other methods for the detection of CEA biomarker.

Device	Detection Type	Linear range (ng/ mL)	Detection limit (ng/ mL)	Reference
anti-CEA/ADA-COOH/APTES/SiO <sub>2</sub> /IDE	Labeled	0.1 - 1000	0.007	[3]
Si <sub>3</sub> N <sub>4</sub> /Au/IDE	Label-free	0.0001 - 10	0.0001	[4]
anti-CEA/PRY-NHS/Graphene/FET	Label-free	0.1 - 100	0.1	[5]
anti-CEA/denatured-BSA/Graphene/FET	Labeled	0.01 - 100	0.00034	[6]
anti-CEA/MoS <sub>2</sub> -Au/GCE	Labeled	0.001 - 50	0.00027	[7]
Anti-CEA/APTES/MoO <sub>x</sub> /IDE	Label-free	0.1-100	0.015	This work

## Reference

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