

Supplementary Information

**Stabilizing Molecular Catalyst on Metal Oxide Surfaces Using Molecular
Layer Deposition for Efficient Water Oxidation**

Hong Wang,^{a,#} Jian Li,^{a,#} Ke Liu,^{a, d#} Lei Lei,^{a, b*} Xun Chen,^{c, d*} Degao Wang^{a, b, d*}

^a Zhejiang Key Laboratory of Data-Driven High-Safety Energy Materials and Applications, Ningbo Key Laboratory of Special Energy Materials and Chemistry, Laboratory of Advanced Nuclear Materials, Ningbo Institute of Materials Technology and Engineering (NIMTE) of the Chinese Academy of Sciences (CAS), Ningbo, Zhejiang, 315201, China.

^b Advanced Interdisciplinary Sciences Research (AIR) Center, Ningbo Institute of Materials Technology and Engineering (NIMTE) of the Chinese Academy of Sciences (CAS), Ningbo, Zhejiang, 315201, China.

^c Laboratory of Polymers and Composites, Ningbo Institute of Materials Technology and Engineering (NIMTE) of the Chinese Academy of Sciences (CAS), Ningbo, Zhejiang, 315201, China.

^d University of Chinese Academy of Sciences, Beijing, 100049, China.

These authors contributed equally.

*Corresponding author, Email: leilei0219@nimte.ac.cn, chenxun@nimte.ac.cn, wangdegao@nimte.ac.cn

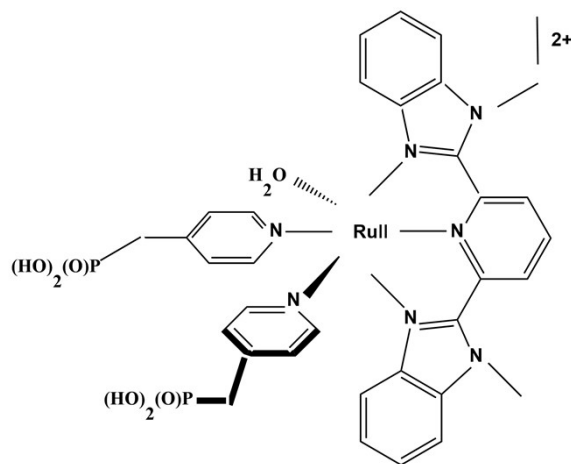


Figure S1. Molecular structures of water oxidation catalyst, $\text{RuCP}(\text{OH}_2)^{2+}$.

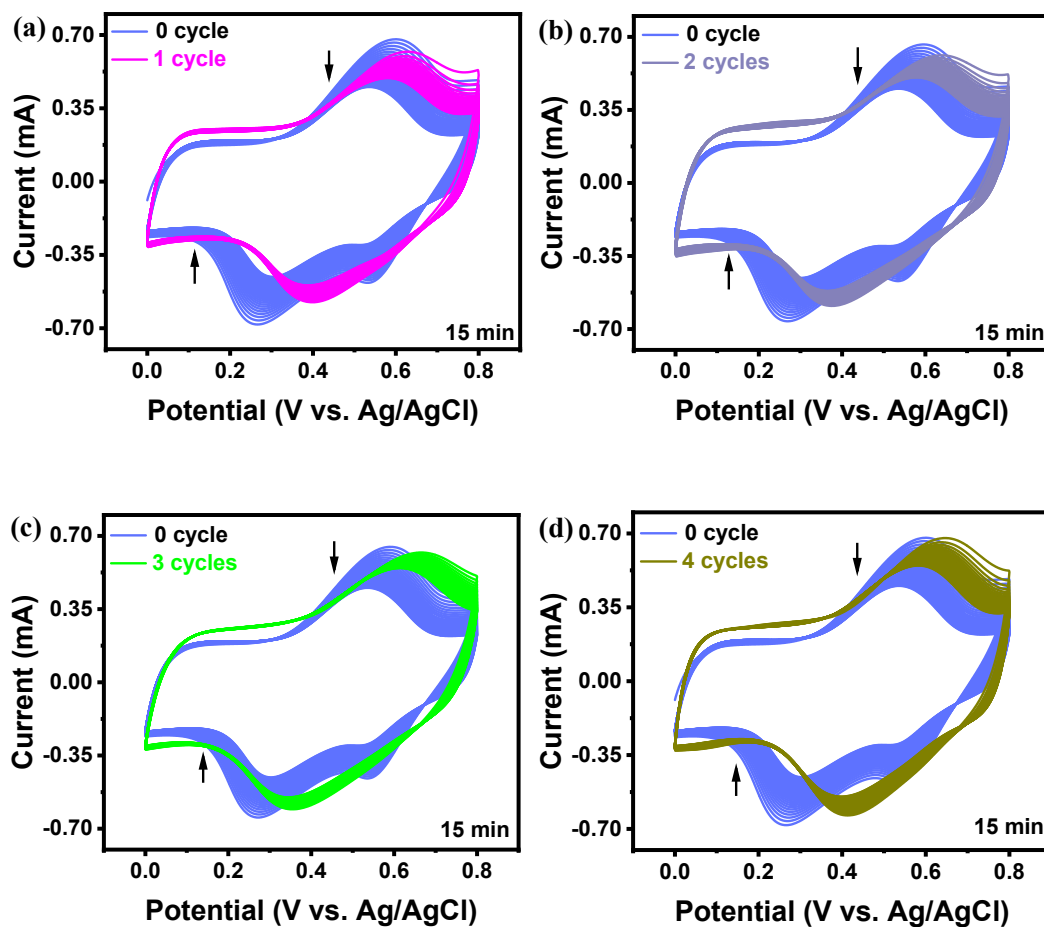


Figure S2. 50 consecutive CV cycles with different cycles of FTO|*nano*ITO-RuCat by MLD-PI at 100 mV/s in 0.1 M phosphate buffer solution (pH~7), here, the FTO|*nano*ITO electrode was soaked in 150 μM RuCat solution for 15 min.

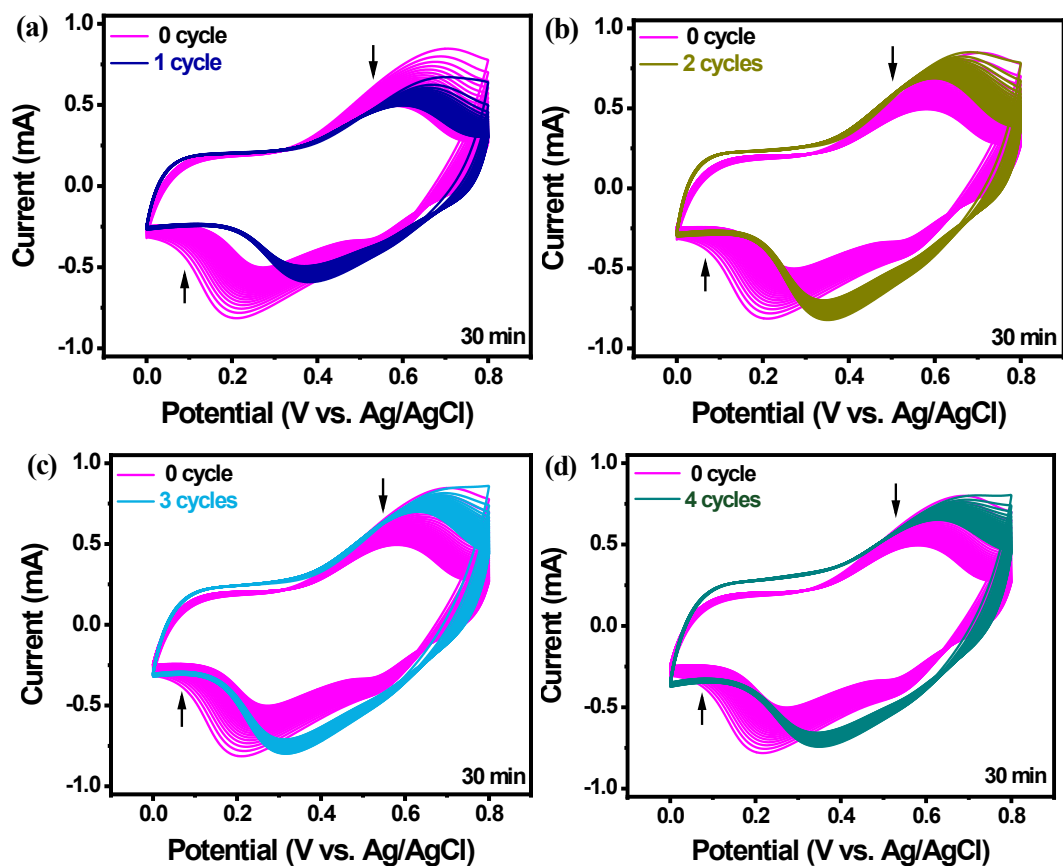


Figure S3. 50 consecutive CV cycles with different cycles of FTO|nanoITO-RuCat by MLD-PI at 100 mV/s in 0.1 M phosphate buffer solution (pH~7), here, the FTO|nanoITO electrode was soaked in 150 μ M RuCat solution for 30 min.

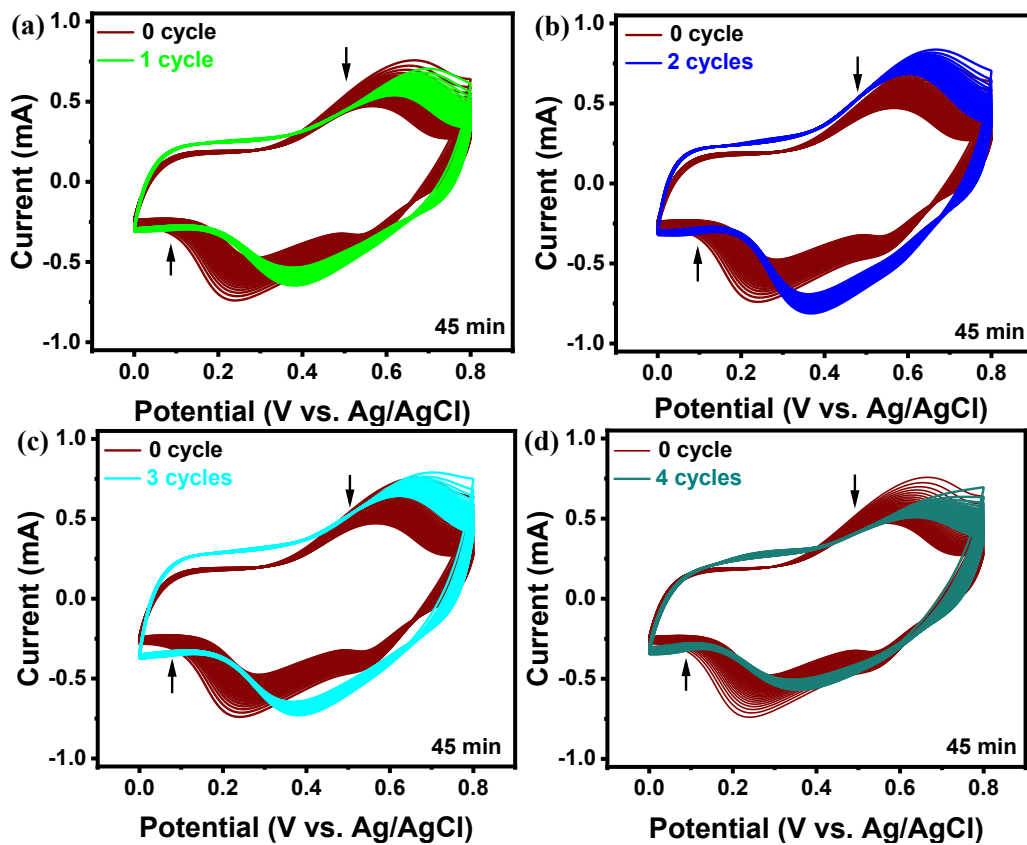


Figure S4. 50 consecutive CV cycles with different cycles of FTO|*nano*ITO-RuCat by MLD-PI at 100 mV/s in 0.1 M phosphate buffer (pH~7), here, the FTO|*nano*ITO electrode was soaked in 150 μ M RuCat solution for 45 min.

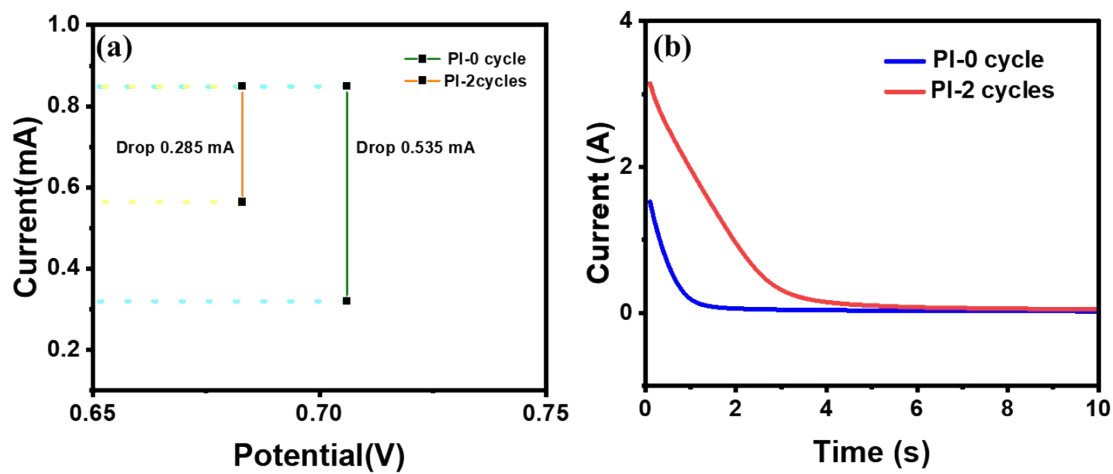


Fig S5. (a) Diagram of redox peak current drop of FTO|*nano*ITO-RuCat and FTO|*nano*ITO-RuCat|PI electrodes after 50 consecutive CV cycles; (b) I-t test of FTO|*nano*ITO-RuCat and FTO|*nano*ITO-RuCat|PI electrodes at 0.8V (vs. Ag/AgCl).

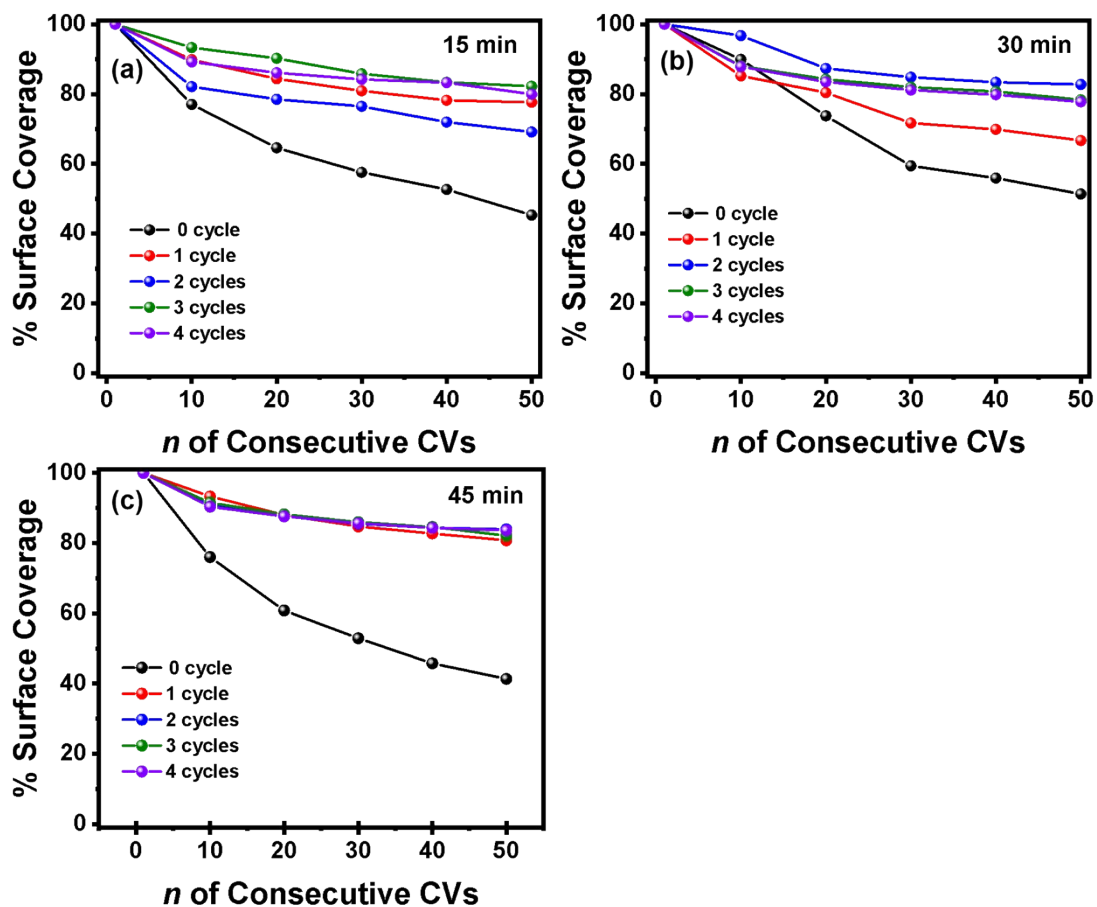


Figure S6. Variation in fractional surface coverage Γ/Γ_0 with number of CV cycles for FTO|*nano*ITO-RuCat by different MLD-deposited PI cycles. The FTO|*nano*ITO electrodes were soaked in 150 μM RuCat solution for (a) 15, (b) 30, (c) and 45 min, respectively.

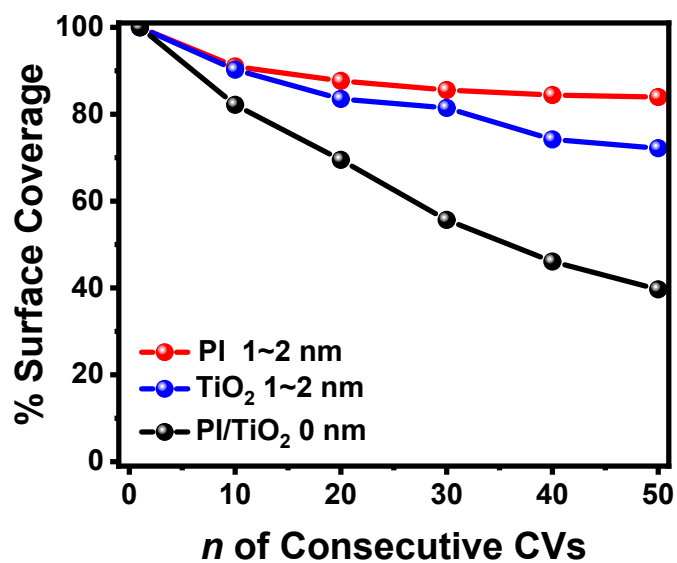


Figure S7. Variation of Γ/Γ_0 with the number of CV cycles for FTO|*nano*ITO-RuCat, FTO|*nano*ITO-RuCat|MLD-PI, and FTO|*nano*ITO-RuCat|ALD-TiO₂.

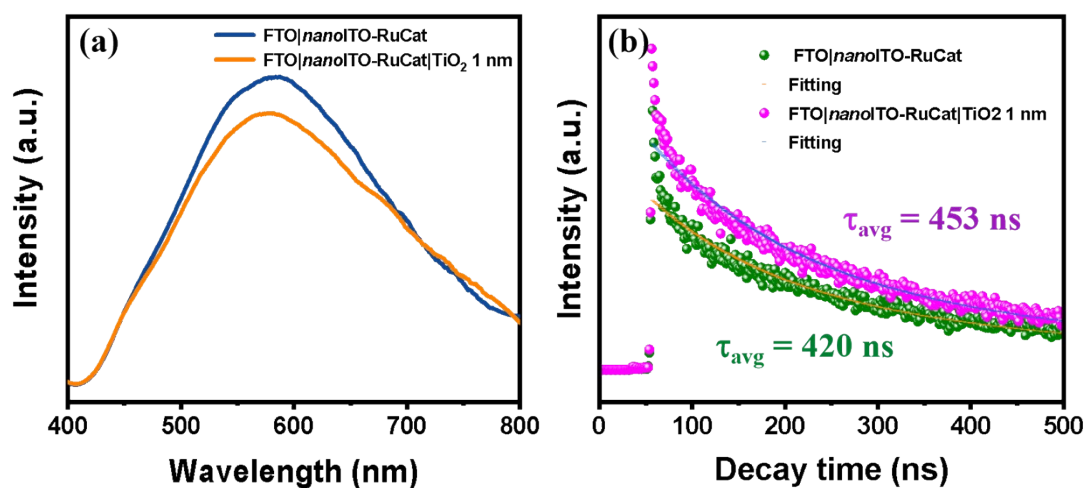


Figure S8. (a) Steady-state fluorescence spectra and (b) Transient fluorescence spectra of FTO/nanoITO-RuCat and FTO/nanoITO-RuCat|TiO₂ electrodes excited using a 375 nm laser.

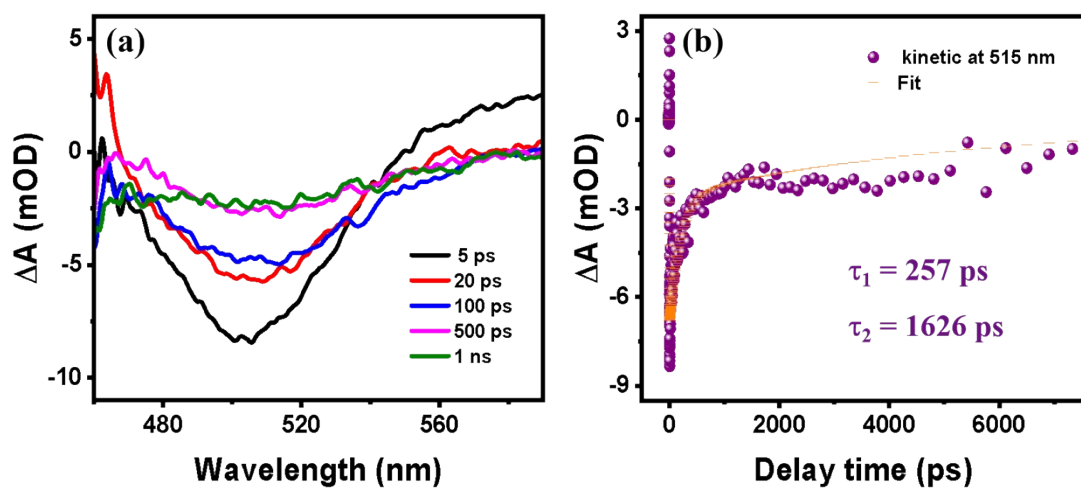


Figure S9. Ultrafast absorption spectra of (a) FTO|*nano*ITO-RuCat|TiO₂; (b) normalized transient time absorption spectra of FTO|*nano*ITO-RuCat|TiO₂.