

## Supporting Information

### Oxyanions as dynamic regulators of the oxygen evolution reaction

Yao Wang,<sup>a‡</sup> Haoyu Li,<sup>a‡</sup> Junyuan Duan,<sup>a\*</sup> and Wenbin Wang<sup>b\*</sup>

<sup>a</sup>State Key Laboratory of Green and Efficient Development of Phosphorous Resource, Hubei Key Laboratory of Plasma Chemistry and New Materials, School of Chemistry and Environmental Engineering, School of Materials Science and Engineering, Wuhan Institute of Technology, Wuhan 430205, China.

<sup>b</sup>Key Laboratory of Luminescence Analysis and Molecular Sensing, Ministry of Education, and School of Chemistry and Chemical Engineering, Southwest University, Chongqing 400715, China

E-mail: junyduan@wit.edu.cn, wangwenbin@swu.edu.cn

<sup>‡</sup>These authors contributed equally to this work.

**Table S1. Oxyanions acting as dominant Type I electronic-structure regulators**

Oxyanions	Classification	Key mechanism	Ref.
$\text{TeO}_3^{2-}/\text{TeO}_4^{2-}$	$\text{I}_d + \text{III}_w$	Te and S in CoTe@CoS are oxidized into $\text{TeO}_3^{2-}/\text{TeO}_4^{2-}$ that bind to CoOOH, tuning the Co d-band, enhancing adsorption, improving electron transport, and optimizing OH- distribution.	S1
$\text{SO}_3^{2-}/\text{SO}_4^{2-}$	$\text{I}_d + \text{III}_w$	FeNiS <sub>2</sub> reconstructs, forming $\text{SO}_3^{2-}/\text{SO}_4^{2-}$ bound to NiFeOOH, electrolyte sulfate limits loss, stabilizes Fe sites, enhances OH/OOH adsorption, shifts the d-band, and lowers the reaction energy barrier.	S2
$\text{SO}_4^{2-}$	$\text{I}_d + \text{III}_w$	$\text{SO}_4^{2-}$ formed in-situ on NiFe alloy by pyrolysis shifts the d-band center, weakens metal-oxygen interactions, lowers energy barriers, enhances charge transfer, improves hydrophilicity, and stabilizes adsorption.	S3
$\text{SO}_4^{2-}$	$\text{I}_d + \text{III}_s$	$\text{SO}_4^{2-}$ intercalates into HELDH, forming hydrogen-bonded superlattice that optimizes electronic structure, promotes active phase formation, enhances mass transport, and maintains long-term catalytic stability through strong coordination and a robust porous framework.	S4
$\text{SO}_4^{2-}$	$\text{I}_d + \text{III}_s$	In S-NiMoO <sub>4</sub> , S converts to $\text{SO}_4^{2-}$ during OER, shifting the d-band center toward the Fermi level, enhancing OH adsorption, generating Ni <sup>4+</sup> species, accelerating kinetics, and stabilizing the surface.	S5
$\text{MoO}_4^{2-}$	$\text{I}_d + \text{III}_s$	$\text{MoO}_4^{2-}$ forms in situ during OER and adsorbs on $\gamma$ -NiFeOOH via Ni-O-Mo bond, enhancing charge transfer, tuning Ni electronic structure, lowering barriers, repelling Cl <sup>-</sup> , and improving corrosion resistance.	S6
$\text{NO}_3^-$	$\text{I}_d + \text{III}_w$	$\text{NO}_3^-$ selectively re-adsorbed after in-situ dissolution from NiFeNH, enhances electron transfer, tunes Fe d-band, lowers OER energy barrier, and promotes OH transport and bubble release, boosting stability.	S7
$\text{NO}_3^-$	$\text{I}_d + \text{III}_w$	$\text{NO}_3^-$ is embedded to form FeOOH/Ni <sub>3</sub> (NO <sub>3</sub> ) <sub>2</sub> (OH) <sub>4</sub> heterointerface, enhancing Fe binding, suppressing over-oxidation/dissolution, and improving charge distribution, mass transport, and OER stability.	S8
$\text{CrO}_4^{2-}$	$\text{I}_d + \text{III}_s$	$\text{CrO}_4^{2-}$ enhances Ni-3d electron density, promotes NiOOH formation, lowers OER barriers, shifts mechanism to LOM, and creates environment that favors OH <sup>-</sup> , repels Cl <sup>-</sup> , and improves corrosion resistance.	S9
$\text{PO}_4^{3-}$	$\text{I}_d + \text{III}_w$	$\text{PO}_4^{3-}$ binds CoFe oxide, lengthening Co-O bonds, upshifting Co 3d and O 2p centers, enhancing OH adsorption, enabling LOM, and improving reconstruction, oxygen cycling, and stability	S10
$\text{PO}_4^{3-}$	$\text{I}_d + \text{III}_s$	As electrolyte additive, $\text{PO}_4^{3-}$ bind Ni/Fe sites, modulating electronics, suppressing dissolution and cathode poisoning, maintaining superoleophobicity, and preserving efficient interfacial mass transport.	S11
$\text{BO}_3^{3-}/\text{CO}_3^{2-}$	$\text{I}_d + \text{III}_s$	Their role is the same as the $\text{PO}_4^{3-}$ in ref.S10.	S11
$\text{HCO}_3^-/\text{CO}_3^{2-}$	$\text{I}_d + \text{III}_s$	$\text{CO}_3^{2-}$ removal creates ligand vacancies partly filled by hydroxide, stabilizing Ni(OH) <sub>x</sub> , tuning d orbitals, reducing deprotonation energy, enabling reversible Ni redox, repelling OH, and promoting NOR.	S12
$\text{MoO}_4^{2-}/\text{SO}_4^{2-}$	$\text{I}_d + \text{III}_s$	$\text{MoO}_4^{2-}$ and $\text{SO}_4^{2-}$ formed in situ during sacrificial heterojunction reconstruction, bind to NiOOH, tunes Ni electronic structure and Ni-O bonding, optimizes OER energetics, and suppressing metal dissolution.	S13

**Table S1. (continued)**

Oxyanions	Classification	Key mechanism	Ref.
$\text{SO}_4^{2-}$	$\text{I}_d + \text{III}_s$	$\text{SO}_4^{2-}$ -generated leaching and re-adsorption from NiFeS enhances M-O covalency, strengthens Fe-O/Ni-O bond, suppresses Fe dissolution, and forms a permeable layer that improves mass transfer and hydrophilicity.	S14
$\text{MoO}_4^{2-}$	$\text{I}_d + \text{III}_s$	$\text{MoO}_4^{2-}$ formed during pre-leaching, with minor sulfate from S oxidation, anchors on NiFeO(S)OH via Ni-O-Mo, enhancing charge transfer, strengthening M-S bonds, and improving interfacial kinetics and stability.	S15
$\text{SO}_4^{2-}$ / $\text{SeO}_4^{2-}$	$\text{I}_d + \text{III}_s$	$\text{SO}_4^{2-}$ / $\text{SeO}_4^{2-}$ formed via in situ oxidation of NiFeS(Se), adsorb on NiFeOOH, tune Fe electronic structure, enhances coupling, accelerate PCET, and reorganize interfacial hydrogen bond networks.	S16
$\text{PO}_4^{3-}$	$\text{I}_d + \text{III}_s$	$\text{PO}_4^{3-}$ grown and intercalated into NiFe-LDH, modulating metal valence, electronic structure, and covalency to promote $\gamma$ -NiOOH formation while enhancing hydrophilicity, mass transport, and chloride repulsion.	S17
$\text{PO}_4^{3-}$	$\text{I}_d + \text{III}_s$	$\text{PO}_4^{3-}$ enters NiFe LDH interlayers and adsorbs on surfaces via hydrothermal process, tuning electronic structure, expanding spacing, enhancing mass transport, and increasing active sites, thereby improving catalytic efficiency.	S18
$\text{SO}_4^{2-}$	I	$\text{SO}_4^{2-}$ anchored sulfate formed in situ from $\text{WS}_2$ oxidation binds strongly to $\text{RuO}_2$ via Ru-O-S, alters electronic distribution, weakens Ru-O covalency, and optimizes intermediate adsorption	S19
$\text{SnO}_3^{2-}$	$\text{I}_d + \text{III}_s$	In situ reconstruction generates $\text{SnO}_3^{2-}$ from Sn oxidation, anchoring within NiOOH, enhance adsorption and dehydrogenation, suppress OER, expand layers, and reduce OH <sup>-</sup> competition.	S20
$\text{CO}_3^{2-}$	$\text{I}_d + \text{III}_s$	$\text{CO}_3^{2-}$ formed in situ uniformly adsorbs on NiOOH, electronically couples with Ni, tunes 3d levels, enhances urea activation, stabilizes intermediates, lowers barriers, suppresses OER, and improves selectivity.	S21
$\text{PO}_4^{3-}$ / $\text{SO}_4^{2-}$ / $\text{CO}_3^{2-}$ / $\text{BO}_3^{3-}$	$\text{I}_d + \text{III}_s$	$\text{PO}_4^{3-}$ / $\text{SO}_4^{2-}$ / $\text{CO}_3^{2-}$ / $\text{BO}_3^{3-}$ bind Ni-based catalysts by tuning electronic structure, stabilizing intermediates, enhancing biomass oxidation, suppressing OER, and improving mass transfer.	S22
$\text{PO}_4^{3-}$	$\text{I}_d + \text{III}_s$	$\text{PO}_4^{3-}$ binds to NiFeOOH via interlayer intercalation and surface coordination, tuning the d-band center, enhancing adsorption, ion transport, and lowering the oxygen evolution barrier.	S23
$\text{SO}_4^{2-}$	$\text{I}_d + \text{III}_s$	$\text{SO}_4^{2-}$ formed by in situ electrochemical activation binds transition metal oxyhydroxides, tuning the d-band center, facilitating intermediates, accelerating PCET, and preventing deactivation by repelling harmful ions.	S24
		The electrolyte supplies $\text{PO}_4^{3-}$ that chemisorb on FeOOH, tune	

		its electronic structure, optimize adsorption, accelerate proton transfer, stabilize pH, enhance corrosion resistance, and promote a more active mixed phase.	
$\text{CO}_3^{2-}$	$\text{I}_d + \text{III}_s$	$\text{CO}_3^{2-}$ introduced in situ or through the electrolyte binds $\text{Ni}(\text{OH})_2$ , tuning electronic structure, optimizing intermediate adsorption, enhancing mass transport, expanding active sites, and improving stability at high currents.	S26
$\text{PO}_4^{3-}/\text{SO}_4^{2-}/\text{CO}_3^{2-}$	$\text{I}_d + \text{III}_s$	$\text{PO}_4^{3-}/\text{SO}_4^{2-}/\text{CO}_3^{2-}$ bind to the NiFeCo catalyst, tuning electronic structure and defects, stabilizing the interface and pH, and enabling controlled reconstruction with sustained catalytic activity and durability.	S27

**Table S1. (continued)**

Oxyanions	Classification	Key mechanism	Ref.
$\text{MoO}_4^{2-}$	$\text{I}_d + \text{III}_s$	In situ reconstruction of NiMoO <sub>4</sub> /NiFe LDH releases and anchors $\text{MoO}_4^{2-}$ , tuning electronic structure and OER energetics while expanding spacing, improving hydroxide transport, stabilizing pH, and preventing degradation.	S28
$\text{PO}_4^{3-} + \text{SO}_4^{2-}$	$\text{I}_d + \text{III}_s$	$\text{PO}_4^{3-}$ and $\text{SO}_4^{2-}$ from in situ oxidation and self-reconstruction of Fe-doped Ni thiophosphate, including PO <sub>4</sub> and SO <sub>4</sub> , modulate structure, enhance adsorption, stabilize surfaces, and promote PCET and mass transport.	S29
$\text{SO}_4^{2-}$	$\text{I}_d + \text{III}_s$	$\text{SO}_4^{2-}$ anchored on N-doped $\text{Co}_3\text{O}_4$ nanosheets modulate electronic structure, optimize intermediate adsorption, enhance hydrophilicity, speed mass transfer, and prevent active site deactivation via electrostatic repulsion.	S30
$\text{CrO}_4^{2-}$	$\text{I}_d + \text{III}_s$	$\text{CrO}_4^{2-}$ intercalates into NiFe LDH, repels chloride ions, enhances hydroxide adsorption, improving OER selectivity and stability.	S31
$\text{P}_3\text{O}_{10}^{5-}$	$\text{I}_d + \text{III}_s$	$\text{P}_3\text{O}_{10}^{5-}$ modifies $\text{Ni}(\text{OH})_2$ via CV, raising Ni valence, improving OER activity and selectivity, suppressing CIOR, and enhancing stability through electrostatic repulsion, pH regulation, and surface passivation.	S32
$\text{NO}_3^- / \text{PO}_4^{3-} / \text{SO}_4^{2-} / \text{SeO}_4^{2-}$	$\text{I}_d + \text{III}_w$	$\text{NO}_3^-$ , $\text{PO}_4^{3-}$ , $\text{SO}_4^{2-}$ , or $\text{SeO}_4^{2-}$ are introduced onto NiFeCoOOH via CV, tuning metal 3d and oxygen 2p orbitals. $\text{NO}_3^-$ and $\text{PO}_4^{3-}$ improve stability and activity, while $\text{SO}_4^{2-}$ , and $\text{SeO}_4^{2-}$ reduce performance.	S33
$\text{SO}_4^{2-}$	I	Sulfate ions formed in situ anchor on $\text{RuO}_2$ via barium, creating a Ru-O-S-O-Ba bridge that matches the surface and converts bridging oxygens to three-coordinated sites.	S34
$\text{MoO}_4^{2-}$	$\text{I}_d + \text{III}_s$	Electrodeposition-driven $\text{MoO}_4^{2-}$ intercalation in NiFe-LDH tunes Ni/Fe d band centers, optimizes intermediate adsorption, shifts rate-determining steps, and EDL enrichment accelerates OH desorption and ion migration.	S35
$\text{SO}_4^{2-}$	I	$\text{SO}_4^{2-}$ formed in situ from oxidized NiFeS and anchored on $\text{NiFe}_2\text{O}_4$ modulate adjacent octahedral Ni, converting $\text{Ni}^{2+}$ to OER optimal $\text{Ni}^{3+}$ .	S36
$\text{SO}_4^{2-}$	I	S in $\text{MoS}_2$ oxidizes in air to $\text{SO}_4^{2-}$ , which binds $\text{RuO}_2$ , tunes adsorption, lowers energy barriers, enhances acidic OER activity, and improves stability by suppressing Ru dissolution and structural degradation	S37

I, II, and III denote Type I (electronic-structure regulators), Type II (charge-buffering/electron-relay species), and Type III (microenvironment regulators), respectively. Subscripts d, w, and s indicate dominant, weak, and synergistic roles.

**Table S2. Oxyanions acting as dominant Type II charge-buffering and electron-relay species<sup>1</sup>**

Oxyanions	Classification	Key mechanism	Ref.
$\text{FeO}_4^{2-}$	$\text{II}_d + \text{I}_w$	Surface Fe in NiFe hydroxides dissolves under OER potentials to form $\text{FeO}_4^{2-}$ , cycling via reabsorption, coupling with oxygen intermediates, mediating electron transfer, and enabling O-O formation and self-healing.	S38
$\text{FeO}_4^{2-}$	$\text{II}_d + \text{I}_w$	Electrolyte-derived $\text{FeO}_4^{2-}$ promotes multi-electron transfer, enables dissolution-redeposition to sustain active sites, enhance charge transport, and tune Ni/Co electronic structure for improved oxygen evolution kinetics	S39
$(\text{W}_2\text{O}_7)^{2-}$	II	$\text{CeO}_2$ -modified NiFe LDH intercalated with $(\text{W}_2\text{O}_7)^{2-}$ uses reversible $\text{W}^{5+}/\text{W}^{6+}$ redox buffering; M-O-W bridges and proton coupling enhance transport, reduce barriers, and improve seawater OER performance.	S40
$[\text{PW}_{12}\text{O}_{40}]^{3-}$	$\text{II}_d + \text{I}_w + \text{III}_w$	$[\text{PW}_{12}\text{O}_{40}]^{3-}$ on CoFe LDH as an electron relay, lowering charge resistance, stabilizing Fe against dissolution, and tuning Co sites to favor OH over Cl, enabling long-term efficient seawater OER.	S41
$[\text{PMo}_{12}\text{O}_{40}]^{3-}$	$\text{II}_d + \text{I}_w + \text{III}_w$	The role is the same as $[\text{PW}_{12}\text{O}_{40}]^{3-}$	S41
$[\text{PSi}_{12}\text{O}_{40}]^{4-}$	$\text{II}_d + \text{I}_w + \text{III}_w$	The role is the same as $[\text{PW}_{12}\text{O}_{40}]^{3-}$	S41
$\text{MoO}_4^{2-}$	II	acts as a redox-active species, reversibly cycling between $\text{Mo}^{6+}$ and $\text{Mo}^{5+}$ under OER anodic potentials, enabling dynamic redistribution of interfacial charge.	S42

I, II, and III denote Type I (electronic-structure regulators), Type II (charge-buffering/electron-relay species), and Type III (microenvironment regulators), respectively. Subscripts d, w, and s indicate dominant, weak, and synergistic roles.

**Table S3. Oxyanions acting as dominant Type III microenvironment regulators**

Oxyanions	Classification	Key mechanism	Ref.
$\text{SeO}_3^{2-}/\text{SeO}_4^{2-}$	$\text{III}_d + \text{I}_w$	$\text{SeO}_3^{2-}$ and $\text{SeO}_4^{2-}$ formed from $\text{Ni}_x\text{Se}$ under anodic OER conditions, adsorb on $\text{NiOOH}$ , weakly tuning active sites, optimizing intermediate adsorption, and enhancing activity via subtle interfacial electrostatic effects.	S43
$\text{SO}_4^{2-}$	$\text{III}_d + \text{I}_w$	Under OER conditions, $\text{SO}_4^{2-}$ adsorbs on $\text{NiCoOOH}$ , forming a hydrogen-bond network that enhances water structure, accelerates $\text{OH}^-$ transport, optimizes electronic states, lowers energy barriers, and promotes high-valence active phase formation.	S44
$\text{SO}_4^{2-}/\text{CO}_3^{2-}/\text{NO}_3^-$	III	Externally added $\text{SO}_4^{2-}$ , $\text{CO}_3^{2-}$ , and $\text{NO}_3^-$ adsorb at the inner Helmholtz plane, displacing $\text{OH}^-$ and lowering its interfacial concentration, disrupting equilibrium and promoting $\text{OH}^-$ release, accelerating surface reconstruction.	S45
$\text{NO}_3^-$	$\text{III}_d + \text{I}_s$	Surface-adsorbed $\text{NO}_3^-$ on $\text{NiFe LDH}$ builds negative charge, repelling chloride and lowering capacitance, while restructuring interfacial water to hinder ion diffusion, enhance hydroxide transport, and stabilize active sites, improving oxygen evolution performance.	S46
$\text{NO}_3^-/\text{CO}_3^{2-}$	III	$\text{NO}_3^-$ and $\text{CO}_3^{2-}$ in $\text{NiFe-LDH}$ interlayers leach out and are replaced by hydroxide, decreasing layer thickness, increasing spacing and distortion, and promoting $\text{Fe}^{2+}$ -rich substitution.	S47
$\text{MoO}_4^{2-}/\text{SO}_4^{2-}$	$\text{III}_d + \text{I}_s$	During OER on the $(\text{Ni,Fe})\text{S}_2/\text{MoS}_2$ heterostructure, $\text{MoO}_4^{2-}$ and $\text{SO}_4^{2-}$ guide precatalyst reconstruction, modulating active sites and promoting asynchronous Ni and Fe oxidation.	S48
$-\text{SO}_3^-$	III	$-\text{SO}_3^-$ groups are covalently attached to the $\text{Ir/IrOx}$ surface, creating a hydrogen-bond network that enhances proton transport, promotes PCET, facilitates $\text{OOH}$ formation, and improves stability in acidic OER conditions.	S49
$\text{SO}_4^{2-}$	III	Externally added components and barium-fixed sulfate use electrostatic repulsion, steric hindrance, and competitive adsorption to form a protective layer that suppresses chloride corrosion and enables dynamic self-healing.	S50
$\text{CO}_3^{2-}$	III	Nickel oxalate converts in situ to $\text{CO}_3^{2-}$ on $\text{NiOOH}$ , forming a weakly bound interface that depletes hydroxide, suppresses OER over a wide range.	S51
$\text{CO}_3^{2-}$	$\text{III}_d + \text{I}_s$	$\text{CO}_3^{2-}$ formed during $\text{NiFe}$ oxalate transformation is charged, repels chloride, suppresses side reactions, enhances $\text{NiFeOOH}$ activity, and stabilizes a protective layer for sustained seawater oxidation.	S52
$\text{MoO}_4^{2-}$	III	$\text{MoO}_4^{2-}$ formed in situ from $\text{RuMoNi}$ creates a stable negatively charged layer via reversible $\text{NiMoO}_4$ , repelling chloride, limiting corrosion and side reactions, and sustaining high-current oxygen evolution.	S53

$\text{PO}_4^{3-}/\text{SO}_4^{2-}/\text{ClO}_4^-$ $/\text{NO}_3^-$	III	Oxyanions tune Helmholtz layer thickness and interfacial fields, affecting Ir-site solvation, adsorption, proton transfer, and stability, with adsorption strength ranking phosphate > sulfate >> nitrate $\approx$ perchlorate.	S54
$\text{B}(\text{OH})_4^-/\text{BO}_3^{3-}$	III	$\text{B}(\text{OH})_4^-/\text{BO}_3^{3-}$ briefly adsorbs on NiFe hydroxide, stabilizing charge during potential fluctuations, buffering interfacial pH, regulating the double layer, and limiting reconstruction, dissolution, and activity loss.	S55

**Table S3 (continued)**

Oxyanions	Classification Type (I, II, III)	Key mechanism	Ref.
$\text{PO}_4^{3-}$	III	Post phosphorylation introduces phosphate species that promote proton transfer, accelerate PCET, improve hydration, stabilize intermediates, enhance kinetics, and reinforce oxide stability for efficient, durable, noble-metal-free OER catalysis.	S56
$\text{BO}_3^{3-}$ / $\text{B(OH)}_4^-$	III	Exogenous borate in the electrolyte buffers interfacial protons, stabilizes pH, accelerates proton coupled electron transfer, optimizes hydration, improves mass transport, and enhances stability and corrosion resistance.	S57
$\text{WO}_4^{2-}$	III	In situ formed $\text{WO}_4^{2-}$ serves as a surface Bronsted base, increasing local hydroxide levels, facilitating water splitting, and improving OH adsorption and activation on NiCoOOH sites.	S58
$\text{RuO}_5^{2-}$	III	The honeycomb ATO nanocavities restrict outward diffusion of soluble $\text{RuO}_5^{2-}$ , increasing its surface concentration and shifting equilibrium to suppress $\text{RuO}_2$ over-oxidation and degradation.	S59
$\text{PO}_4^{3-}$ + $\text{CO}_3^{2-}$	III	During OER, the phytic-acid shell oxidizes into phosphate and carbonate, forming an oxyanion layer on NiFeOOH that blocks halides, suppresses dissolution, enhances mass transport, and tunes structure.	S60
$\text{SiO}_3^{2-}$	III	$\text{SiO}_3^{2-}$ weakly adsorb on $\text{Ni(OH)}_2$ , restructuring hydrogen bond networks, boosting water dissociation, repelling chloride, enhancing hydrophilicity, promoting Ni oxidation, and improving seawater OER activity and stability.	S61

I, II, and III denote Type I (electronic-structure regulators), Type II (charge-buffering/electron-relay species), and Type III (microenvironment regulators), respectively. Subscripts d, w, and s indicate dominant, weak, and synergistic roles.

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