

Supporting Information

The Effect of Hydrogen Distribution of Ni-Fe Hydroxide on the Oxygen Evolution Reaction

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Figure S1(a-h). The electronic structure of nickel hydroxide during the OER process at 1-Voh site

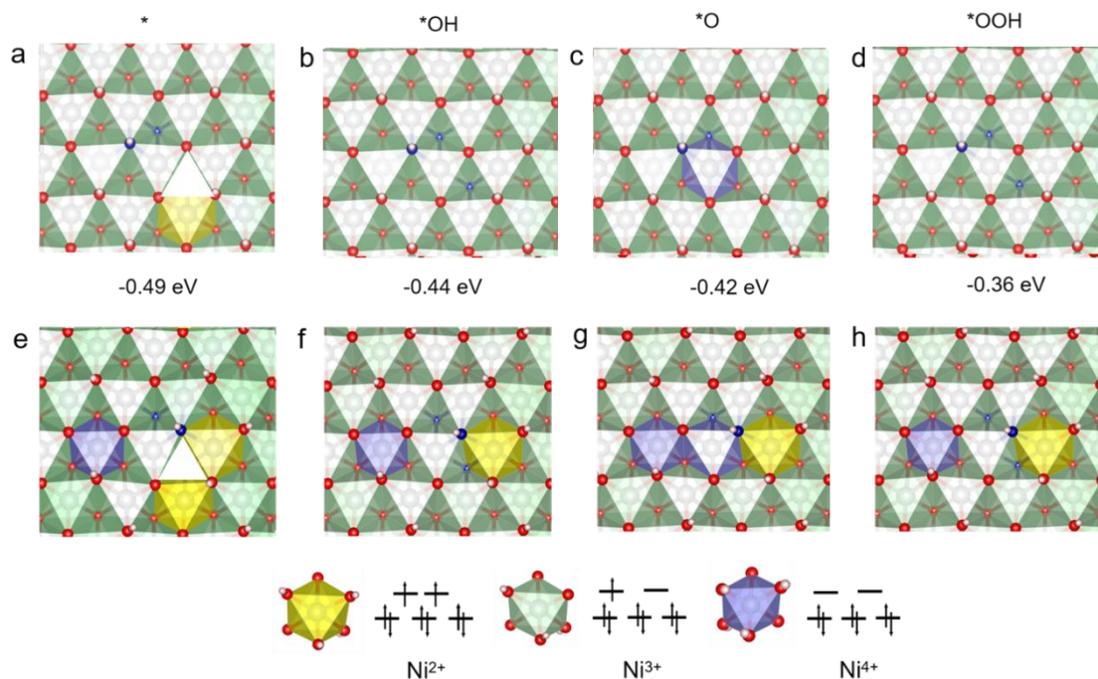


Figure S1 The electronic states of intermediates in the oxygen evolution reaction and the changes in the Gibbs free energy of each intermediate under varying hydrogen conditions at the 1-Voh site for hydroxide. (a-d) represents the electronic state of intermediate species at the site after hydrogen exchange. (e-h) represents the electronic state of intermediate species at the site before hydrogen exchange. The white triangle represents the reaction site. The blue represents the oxygen atoms that still have hydrogen atoms attached to them on the active atom. The numbers represent the ΔG_H of the intermediate ($*$, $*OH$, $*O$, $*OOH$) during hydrogen configuration changes.

Figure S2(a-h). The electronic structure of nickel hydroxide during the OER process at 2-Voh site

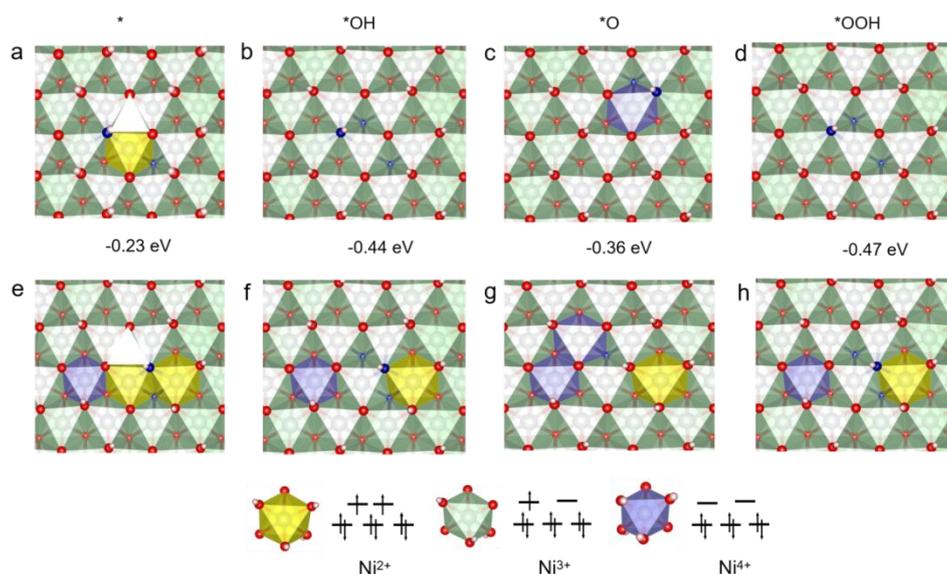


Figure S2 The electronic states of intermediates in the oxygen evolution reaction and the changes in the Gibbs free energy of each intermediate under varying hydrogen conditions at the 2-Voh site for hydroxide. (a-d) represents the electronic state of intermediate species at the site after hydrogen exchange. (e-h) represents the electronic state of intermediate species at the site before hydrogen exchange.

Figure S3(a-h). The electronic structure of nickel hydroxide during the OER process at 1-Voh-s site

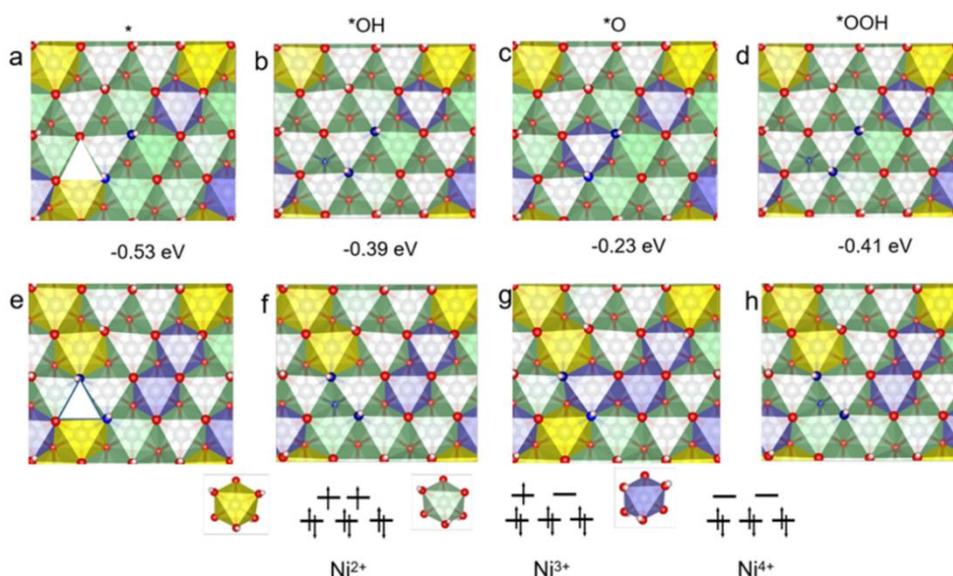


Figure S3 The electronic states of intermediates in the oxygen evolution reaction and the changes in the Gibbs free energy of each intermediate under varying hydrogen conditions at the 1-Voh-s site for hydroxide. (a-d) represents the electronic state of intermediate species at the site after hydrogen exchange. (e-h) represents the electronic state of intermediate species at the site before hydrogen exchange.

Figure S4(a-h). The electronic structure of Fe-doped hydroxide during the OER process at 1-Voh site

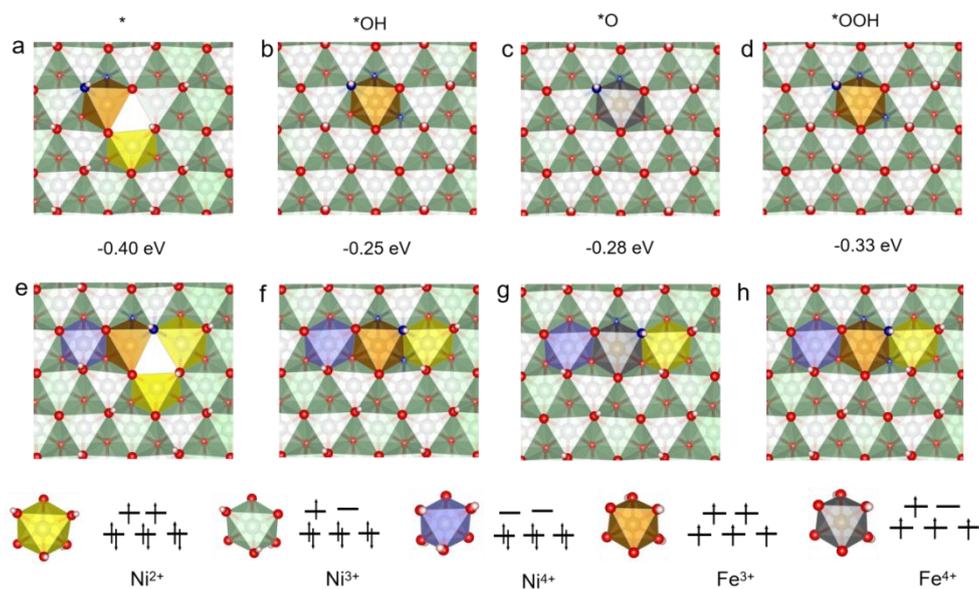


Figure S4 The electronic states of intermediates in the oxygen evolution reaction and the changes in the Gibbs free energy of each intermediate under varying hydrogen conditions at the 1-Voh site for Fe-doped hydroxide. (a-d) represents the electronic state of intermediate species at the site after hydrogen exchange. (e-h) represents the electronic state of intermediate species at the site before hydrogen exchange.

Figure S5(a-h). The electronic structure of Fe-doped hydroxide during the OER process at 1-Voh-s site.

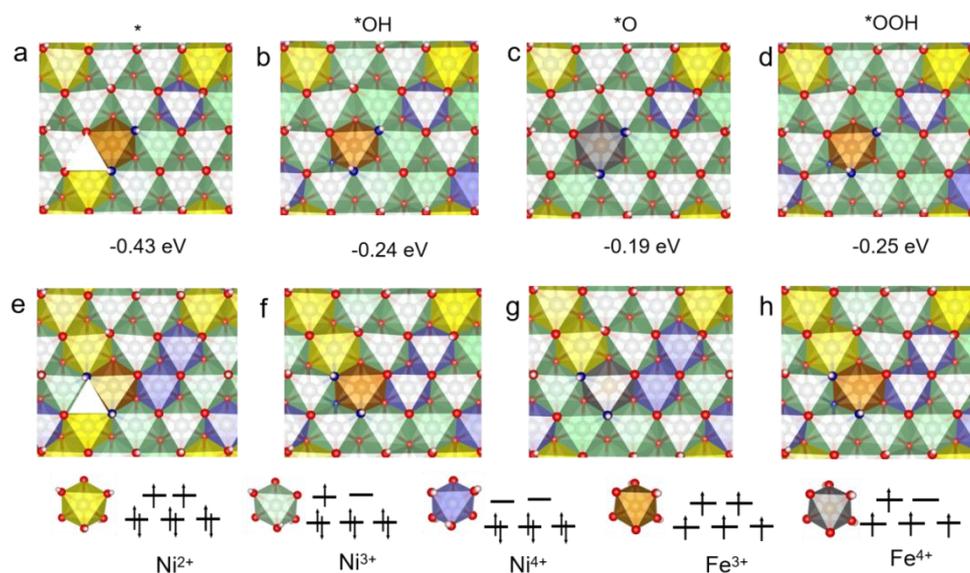


Figure S5 The electronic states of intermediates in the oxygen evolution reaction and the changes in the Gibbs free energy of each intermediate under varying hydrogen conditions at the 1-Voh-s site for Fe-doped hydroxide. (a-d) represents the electronic state of intermediate species at the site after hydrogen exchange. (e-h) represents the electronic state of intermediate species at the site before hydrogen exchange.

Optimized coordinates for β -NiOOH

1.00000000000000

11.8354997634999997	0.0000000000000000	0.0000000000000000
0.0000000000000000	10.0607995986999992	0.0000000000000000
0.0000000000000000	0.0000000000000000	20.1658000945999998

Ni	H	O
16	16	32

Direct

0.2867833665722624	0.7547150701577124	0.1432935765158962
0.9084528240011753	0.5139600815719568	0.1515238664552752
0.0368510272854059	0.7636976064034050	0.1513265093146441
0.1586424163115978	0.5048900050019592	0.1454132303719034
0.7867529061025066	0.7550715540430429	0.1430342394527033
0.5365924112814292	0.7635593175812019	0.1487722910897901
0.6583970363116266	0.5049132918956900	0.1421968770849295
0.2864680170046406	0.2549037400492309	0.1449822277260059
0.9085775641679155	0.0139323287265764	0.1514242502836191
0.0367519089046362	0.2640852424913666	0.1528700318874786
0.1585328308139398	0.0050276941445786	0.1449944058426515
0.7867750376331142	0.2552390259852366	0.1436501940839323
0.4086016818125825	0.0136745396327643	0.1499712910597670
0.5364779513516466	0.2636841155329636	0.1494786332460264
0.6584285212991418	0.0050418671519079	0.1423889077749307
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0.8143825659433390	0.8990201441854936	0.0029162621757000
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0.0612495491574180	0.1196812130683715	0.2935648171789317
0.1318224021849759	0.1489941864367832	0.0047018101166087
0.8147165868687346	0.3983418044444186	0.0031649530218402
0.3856533539874112	0.8691174953956956	0.2908181619581973
0.5624658874206069	0.1193466853834343	0.2902626597677350
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0.5628738636145673	0.6199508110655744	0.2896207053151896
0.6342546507813042	0.6482330904522559	0.0012290400963200
0.3099747610817828	0.3968187087615087	0.0035779187455783
0.9026494673398200	0.3547984610331544	0.2273010927371177
0.2914866739635361	0.9146938651407445	0.0683948056997923
0.1532616908810405	0.6632565894831935	0.0690049808886347

0.0418696495401537	0.6048926109064131	0.2273353679185373
0.2819227986134872	0.5889631988974795	0.2091243610147431
0.9146443473318840	0.6798117918301746	0.0854937912865077
0.0313442108921829	0.9302556200973449	0.0862536930132305
0.1644406415689218	0.3391484493211128	0.2116533416268332
0.4045762024507694	0.3543590836077449	0.2255411798074252
0.7928903310367988	0.9147949079645408	0.0681525462070360
0.6536579937100746	0.6642283569732812	0.0668861781330940
0.5420382754907663	0.6039271984220196	0.2242618418204324
0.7801999573224176	0.5891725518149945	0.2084202691468875
0.4137821871794581	0.6794873128670244	0.0838891310016327
0.5309540274956989	0.9302598726899897	0.0836147007321171
0.6637971817777262	0.3391256065482853	0.2078385564058568
0.9031697609644989	0.8541479932804893	0.2265429615251528
0.2911097071387471	0.4138838904193357	0.0692126834811320
0.1527042549180599	0.1648234373628453	0.0700642353777301
0.0414495611751176	0.1043712077618600	0.2279018412557011
0.2816805399953331	0.0883462091005841	0.2097358798679520
0.9149443180605008	0.1805202979424904	0.0863425639800471
0.0314137892938660	0.4299090231274268	0.0871214604061221
0.1647527146687504	0.8382976763377270	0.2095414685268233
0.4043632459105018	0.8539163383302810	0.2249226798320360
0.7928936897123355	0.4146490272938118	0.0682604149119986
0.6534014956684715	0.1646509149960333	0.0674687259669198
0.5420755065992138	0.1038709339645112	0.2247405955765488
0.7803696217011249	0.0888983038558296	0.2086813149529813
0.4132665696621998	0.1800605371833853	0.0848406385822416
0.5308591705387489	0.4297332432433175	0.0836650115933980
0.6637716467723184	0.8388401714173678	0.2073585912546762

Table S1-S2. The change in Gibbs free energy of the intermediate is caused by the hydrogen configuration.

			ΔG_1 / eV	$\Delta(\Delta G_1)$ / eV	ΔG_2 / eV	$\Delta(\Delta G_2)$ / eV	ΔG_3 / eV	$\Delta(\Delta G_3)$ / eV	ΔG_4 / eV	$\Delta(\Delta G_4)$ / eV
Ni	1-Voh	H vary	-1.01	-0.21	0.46	0.08	0.29	-0.11	0.26	0.24
		primstine	-0.80		0.38		0.40		0.02	
	2-Voh	H vary	-1.01	0.05	0.46	0.02	0.33	0.06	0.22	-0.13
		primstine	-1.06		0.44		0.27		0.35	
	1-Voh-s	H vary	-0.98	0.14	0.41	0.16	0.33	-0.18	0.24	-0.12
		primstine	-1.12		0.25		0.51		0.36	
Fe	1-Voh	H vary	-1.02	0.13	0.35	-0.13	0.53	0.09	0.14	-0.09
		primstine	-1.15		0.48		0.44		0.23	
	2-Voh	H vary	-1.14	0.15	0.41	-0.03	0.40	-0.05	0.32	-0.07
		primstine	-1.29		0.44		0.45		0.39	
	1-Voh-s	H vary	-1.07	0.19	0.25	0.05	0.54	-0.06	0.28	-0.19
		primstine	-1.26		0.20		0.60		0.47	

Table S1 The change in Gibbs free energy of the intermediate is caused by the hydrogen configuration. 1-Voh,2-Voh,1-Voh-s represents different reactive sites.

		$\Delta G_{H1,H2}$ / eV	$\Delta(\Delta G_{H1,2})$ / eV	$\Delta G_{H2,H3}$ / eV	$\Delta(\Delta G_{H2,3})$ / eV	$\Delta G_{H3,H4}$ / eV	$\Delta(\Delta G_{H3,4})$ / eV	$\Delta G_{H4,H1}$ / eV	$\Delta(\Delta G_{H4,1})$ / eV
Ni	1-Voh	-0.23	-0.21	-0.44	0.08	-0.36	-0.11	-0.47	0.24
		-0.44		-0.36		-0.47		-0.23	
	2-Voh	-0.49	0.05	-0.44	0.02	-0.42	0.06	-0.36	-0.13
		-0.44		-0.42		-0.36		-0.49	
	1-Voh-s	-0.53	0.14	-0.39	0.16	-0.23	-0.18	-0.41	-0.12
		-0.39		-0.23		-0.41		-0.53	
Fe	1-Voh	-0.38	0.13	-0.25	-0.13	-0.38	0.09	-0.29	-0.09
		-0.25		-0.38		-0.29		-0.38	
	2-Voh	-0.40	0.15	-0.25	-0.03	-0.28	-0.05	-0.33	-0.07
		-0.25		-0.28		-0.33		-0.40	
	1-Voh-s	-0.43	0.19	-0.24	0.05	-0.19	-0.06	-0.25	-0.19
		-0.24		-0.19		-0.25		-0.43	

Table S2 The ΔG_{Hn} of the intermediate is caused by the hydrogen configuration.

			ΔG_1 / eV	ΔG_2 / eV	ΔG_3 / eV	ΔG_4 / eV
Ni	1-Voh	H vary		PDS		
		primstine			PDS	
	2-Voh	H vary		PDS		
		primstine		PDS		
	1-Voh-s	H vary		PDS		
		primstine			PDS	
Fe	1-Voh	H vary		PDS		
		primstine			PDS	
	2-Voh	H vary			PDS	
		primstine		PDS		
	1-Voh-s	H vary			PDS	
		primstine			PDS	

Table S3 The potential-determining step (PDS) corresponding to different reaction sites under the change of hydrogen