Supporting Information

Tunable d-band center of NiFeMo alloy with enlarged lattice strain enhancing intrinsic catalytic activity for overall water-splitting

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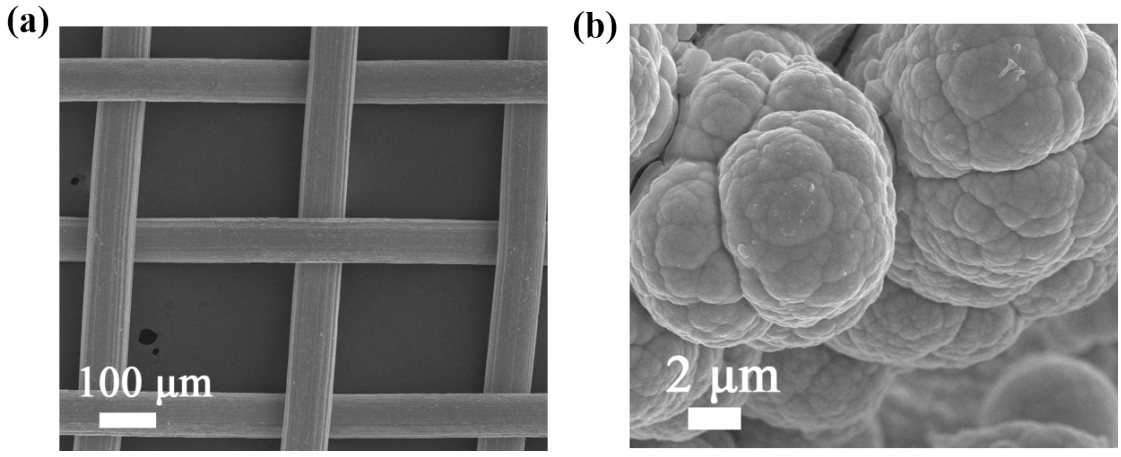

Fig. S1. (a) Scanning electron microscopy (SEM) images of (a) Blank SSM (b) NiFeMo/SSM at further magnification.

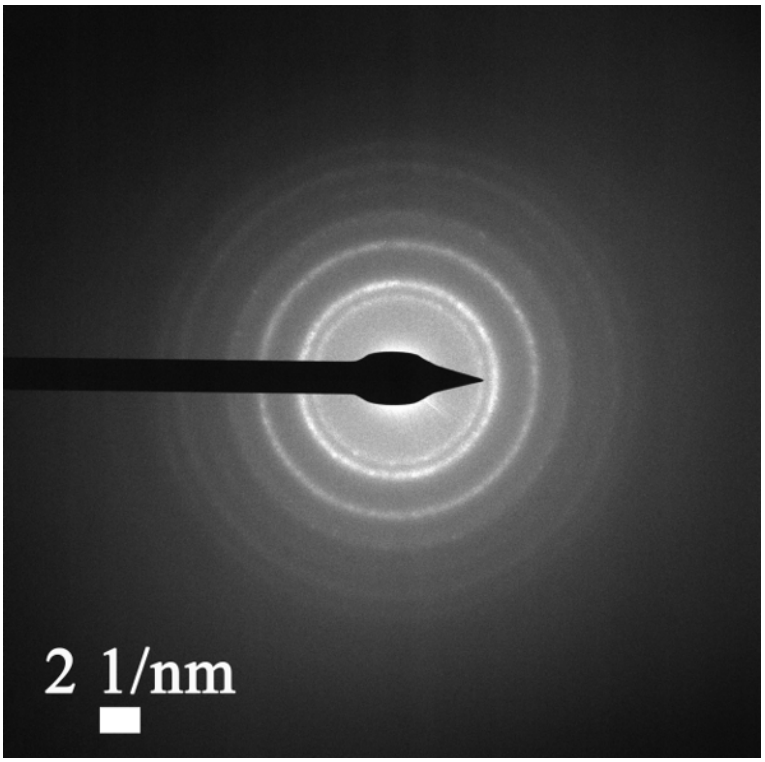

Fig. S2. SAED pattern of NiFeMo/SSM.

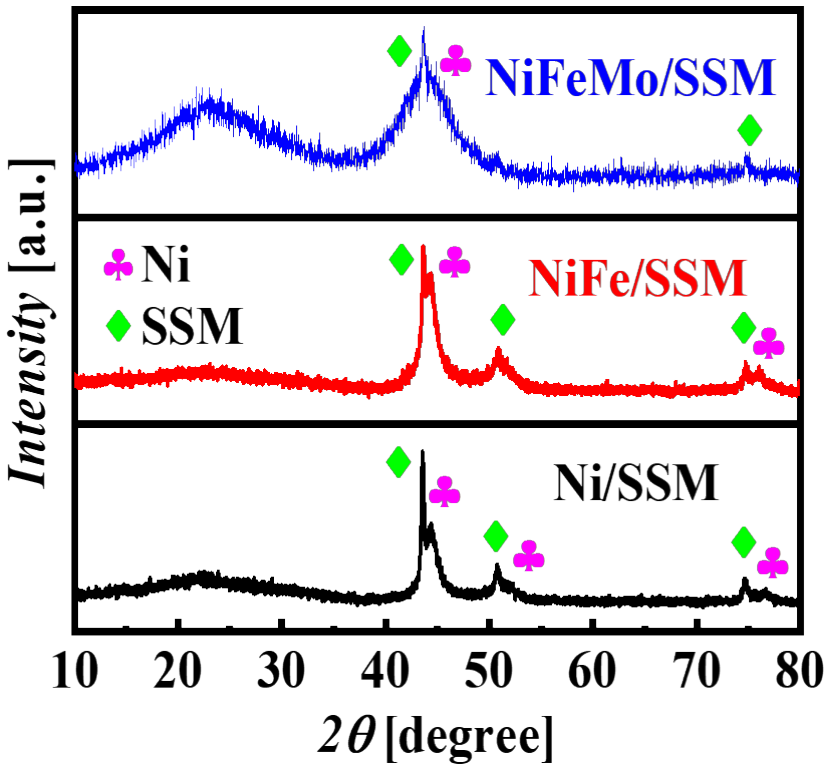

Fig. S3. XRD patterns of Ni/SSM, NiFe/SSM, NiFeMo/SSM.

Table S1. The content of each metal in the catalyst.

Sample	Elements	Atomic Percentage (At%)
NiFe/SSM	Ni	92.98
	Fe	6.61
NiMo/SSM	Ni	94.89
	Mo	4.62
NiFeMo/SSM	Ni	91.99
	Fe	4.27
	Mo	3.02

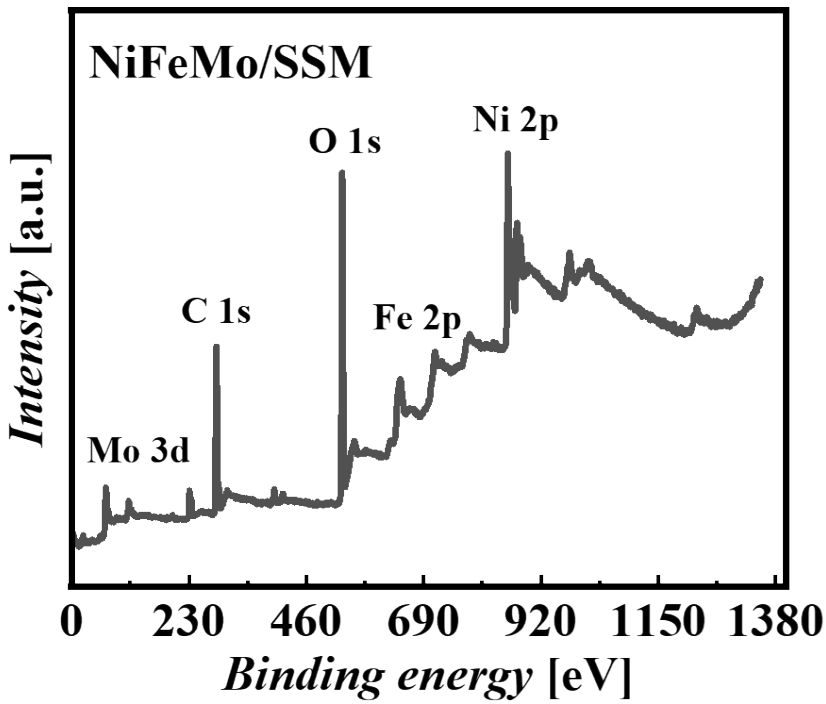

Fig. S4. XPS survey spectra of NiFeMo/SSM.

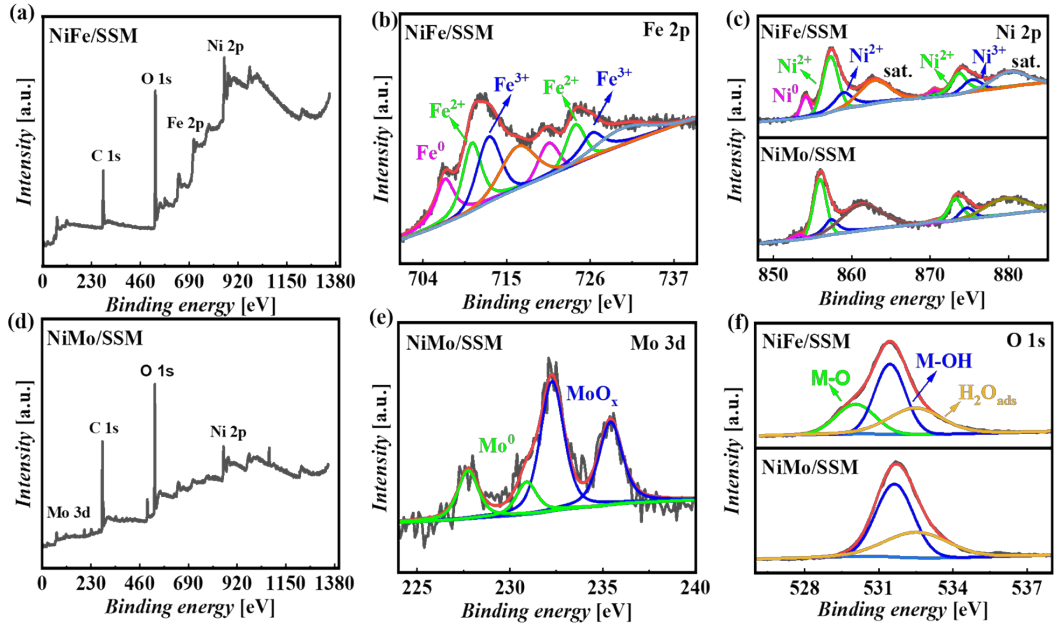

Fig. S5. (a) XPS survey spectra of NiFe/SSM, (b) Fe 2p of NiFe/SSM, (c) Ni 2p of NiFe/SSM and NiMo/SSM, (d) XPS survey spectra of NiMo/SSM, (e) Mo 3d of NiMo/SSM, (f) O 1s of NiFe/SSM and NiMo/SSM.

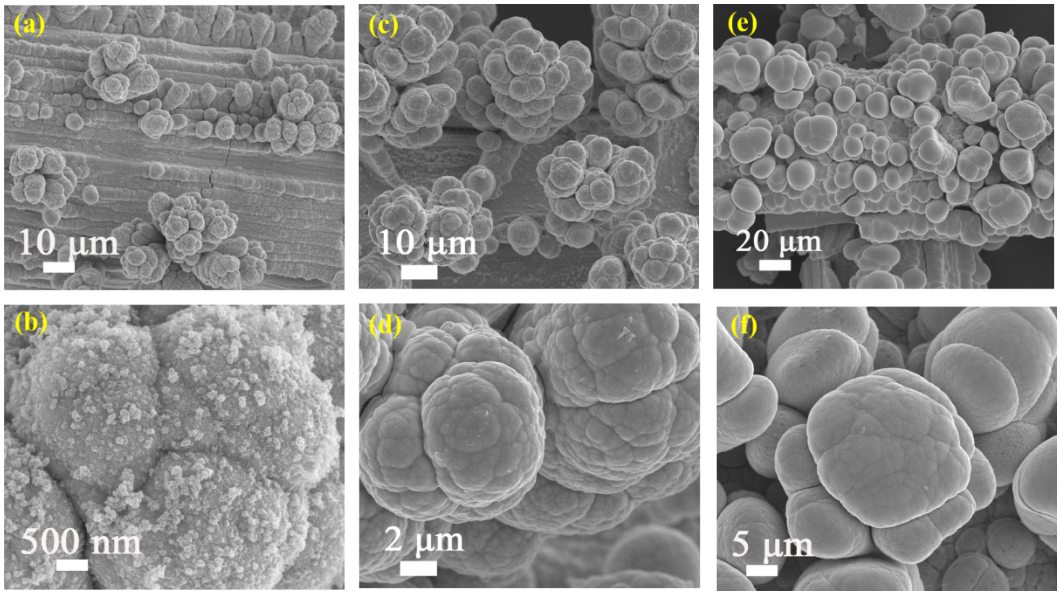

Fig. S6. SEM images with the different magnification of (a) and (b) NiFeMo-(10 mM Mo), (c) and (d) NiFeMo-(20 mM Mo), (e) and (f) NiFeMo-(40 mM Mo).

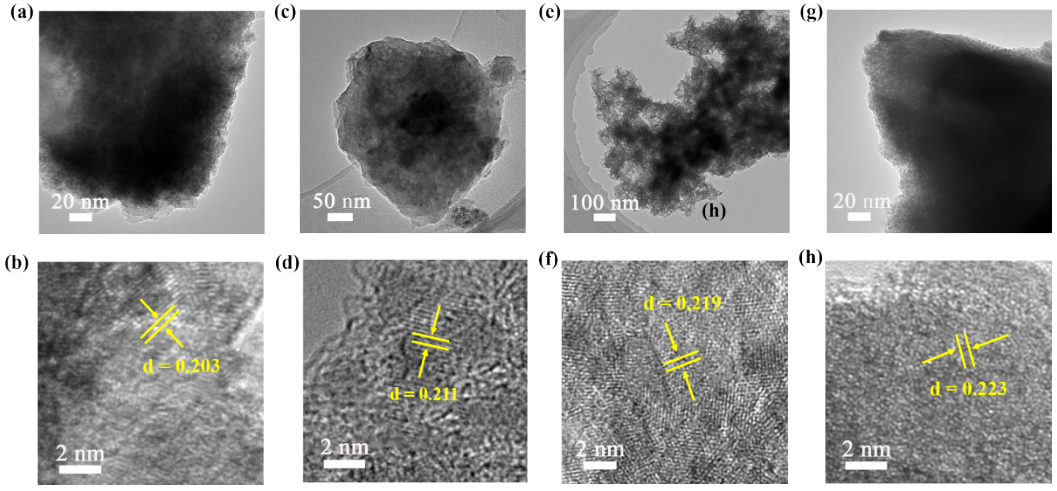

Fig. S7. TEM and HRTEM images of (a-b) Ni, (c-d) NiFeMo-(10 mM Mo), (e-f) NiFeMo-(20 mM Mo), (g-h) NiFeMo-(40 mM Mo).

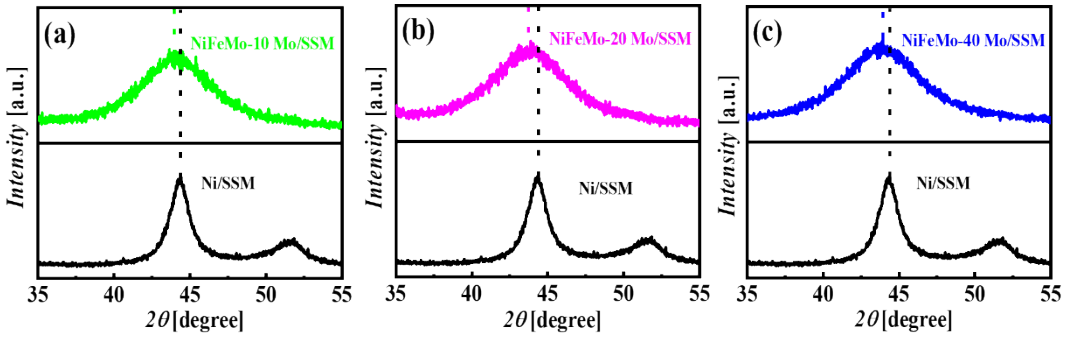

Fig. S8. XRD patterns of (a) Ni and NiFeMo-(10 mM Mo), (b) Ni and NiFeMo-(20 mM Mo), (c) Ni and NiFeMo-(40 mM Mo).

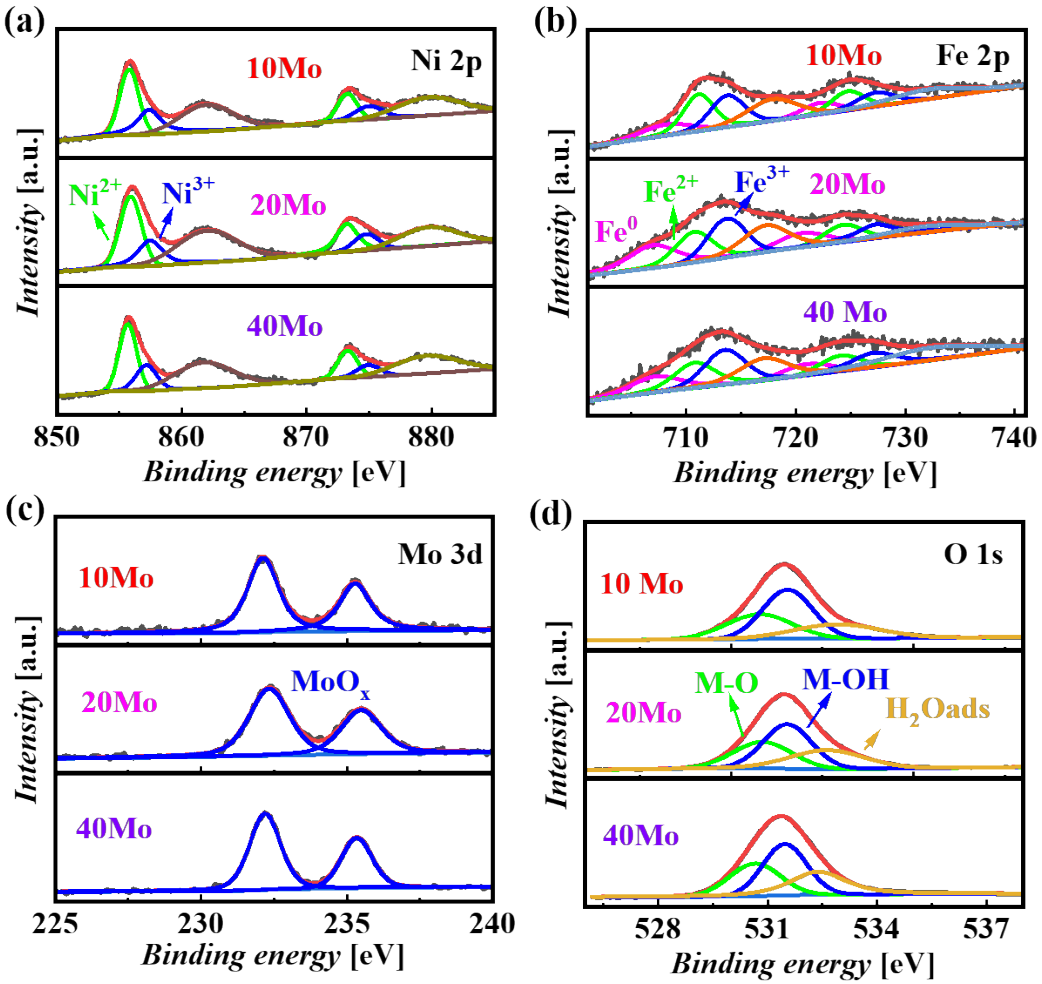

Fig. S9. Comparative chemical structures of NiFeMo-(10 mM Mo), NiFeMo-(20 mM Mo), NiFeMo-(40 mM Mo). a) Ni 2p, b) Fe 2p, c) Mo 3d and d) O1s.

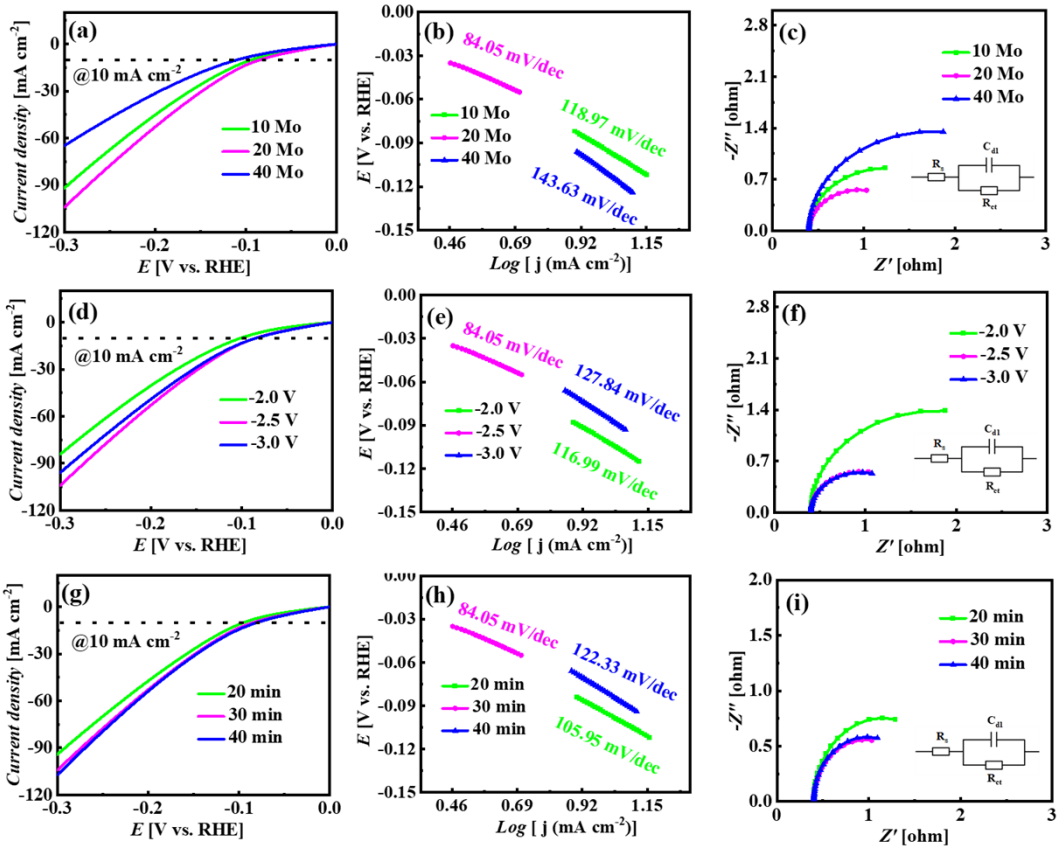

Fig. S10. LSV curve, Tafel plots, and Nyquist plot for different deposition conditions for HER.

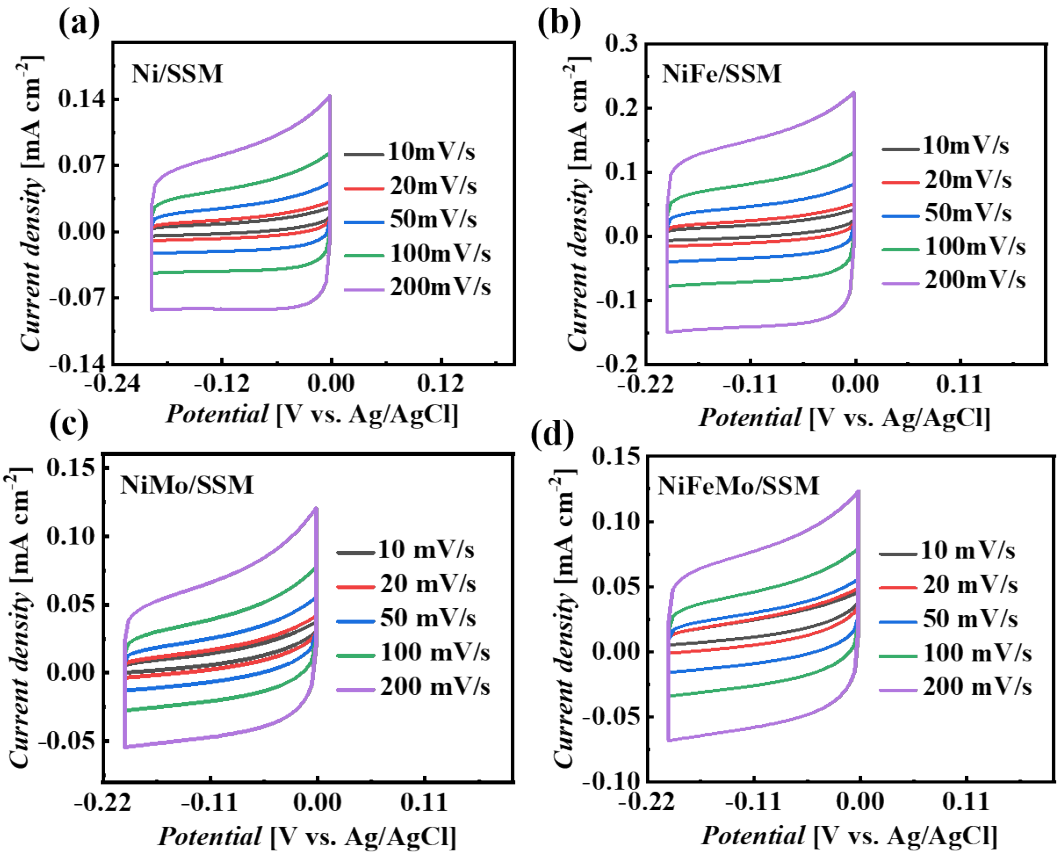

Fig. S11. Cyclic voltammograms within the potential range where no faradaic reaction occurred on (a) Ni/SSM, (b) NiFe/SSM, (c) NiMo/SSM, (d) NiFeMo/SSM.

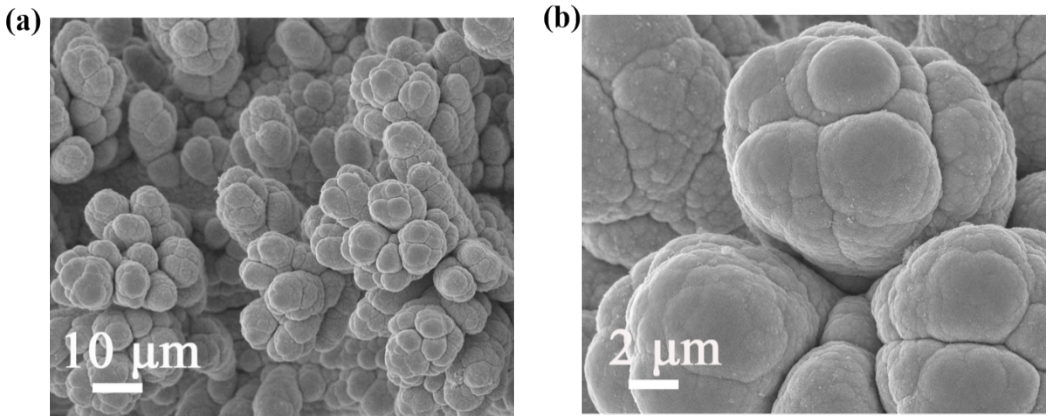

Fig. S12. SEM images of NiFeMo/SSM after HER test.

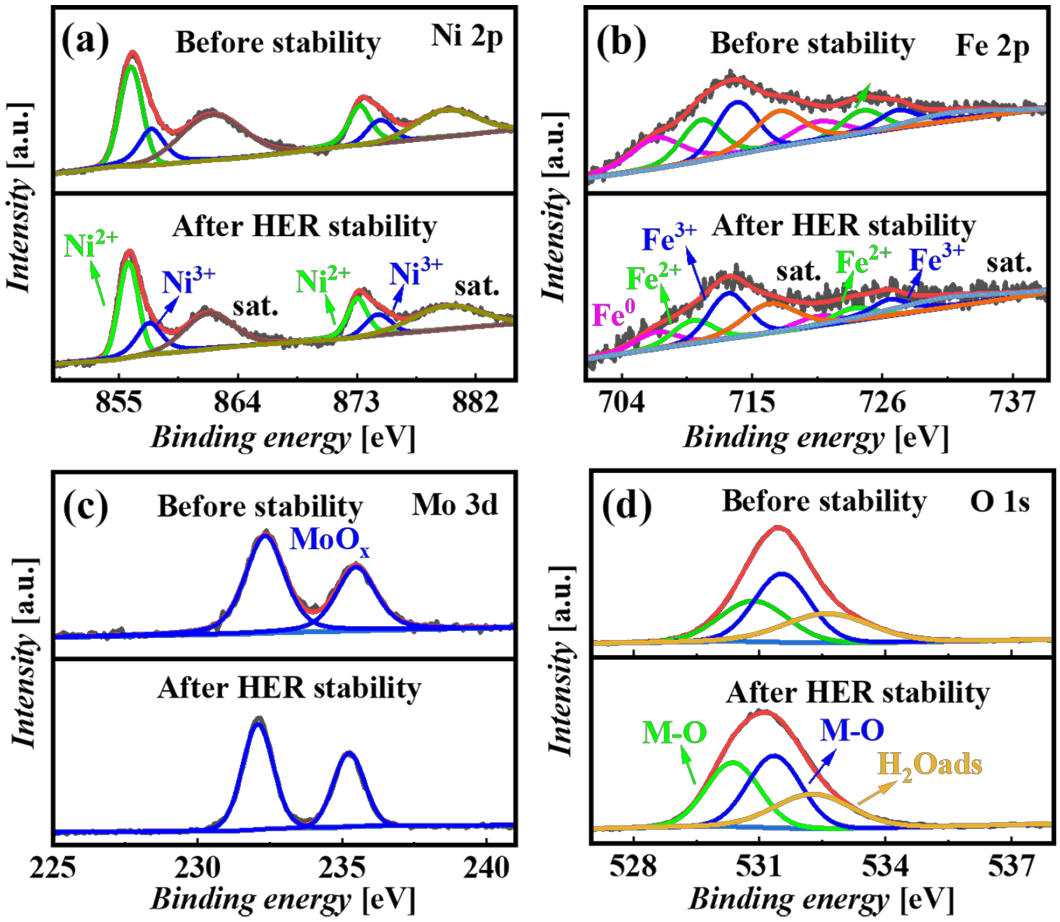

Fig. S13. (a) XPS spectra of Ni 2p of NiFeMo/SSM before and after HER stability test. (b) XPS spectra of Fe 2p of NiFeMo/SSM before and after HER test. (c) XPS spectra of Mo 3d of NiFeMo/SSM before and after HER test. (d) XPS spectra of O 1s of NiFeMo/SSM before and after HER test. (top panel for before test, bottom panel for after HER).

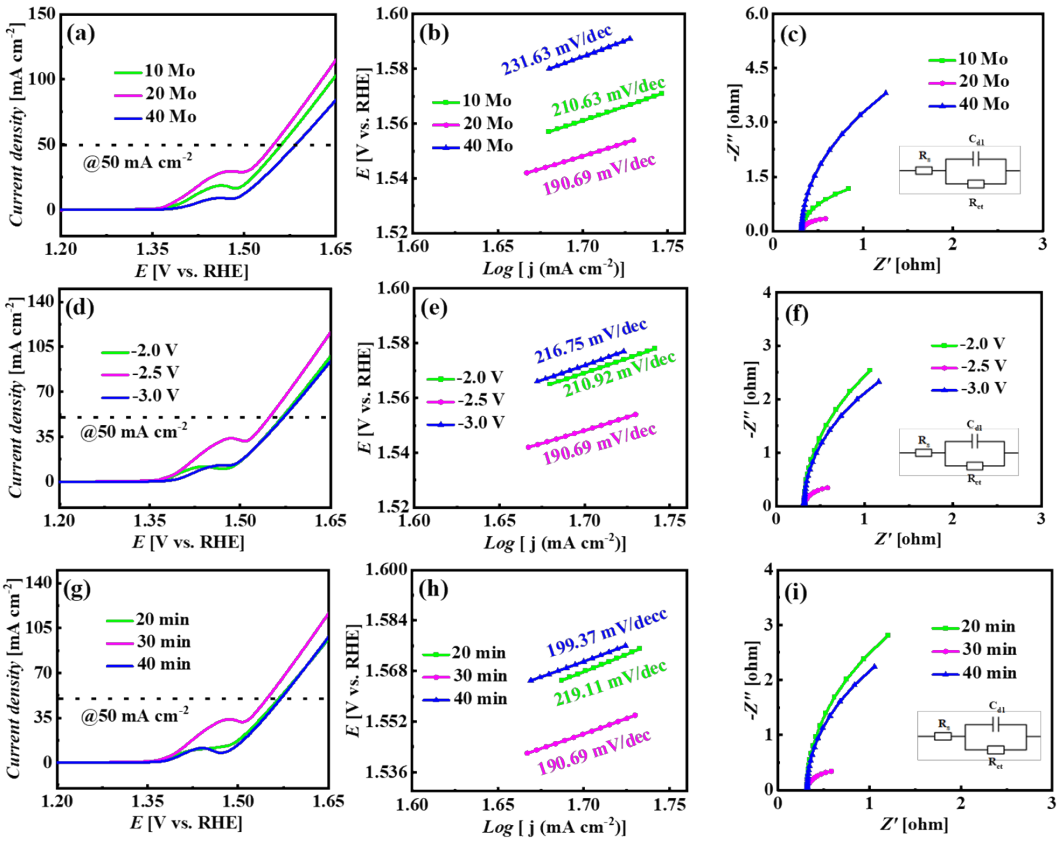

Fig. S14. LSV curve, Tafel plots, and Nyquist plot for different deposition conditions for OER.

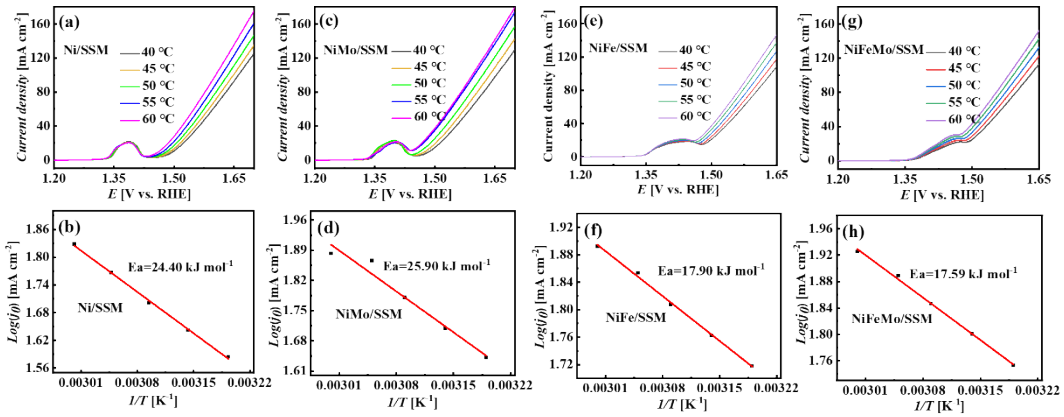

Fig. S15. Polarization curve with different temperatures and corresponding Arrhenius plots of Ni/SSM (a and b), NiMo/SSM (c and d), NiFe/SSM (e and f), NiFeMo/SSM (g and h).

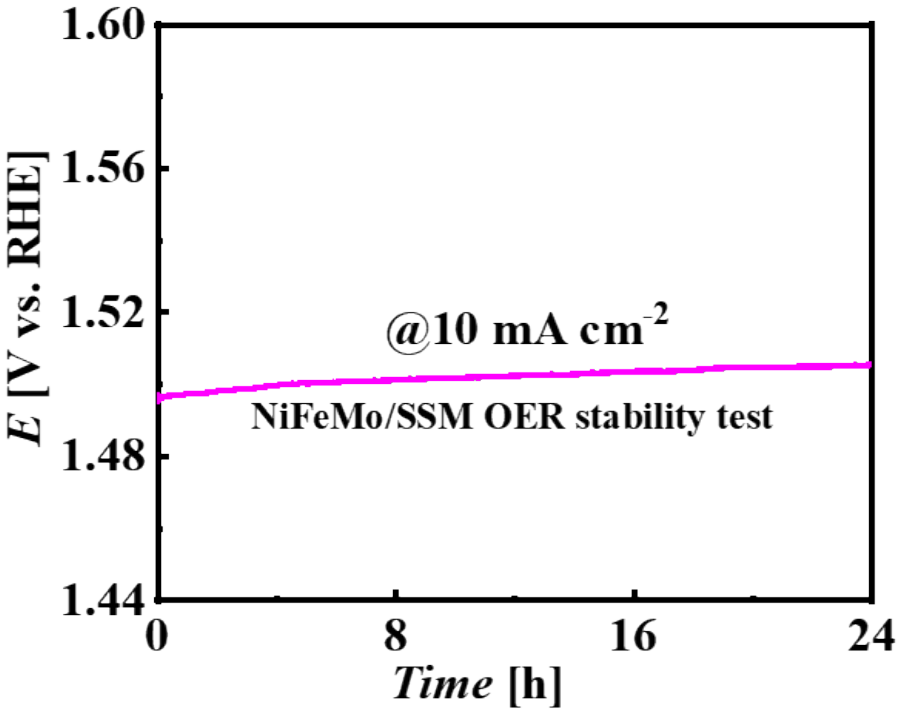

Fig. S16. Long stability test curve of NiFeMo/SSM at the current density of 10 mA cm^{-2} for 24 h.

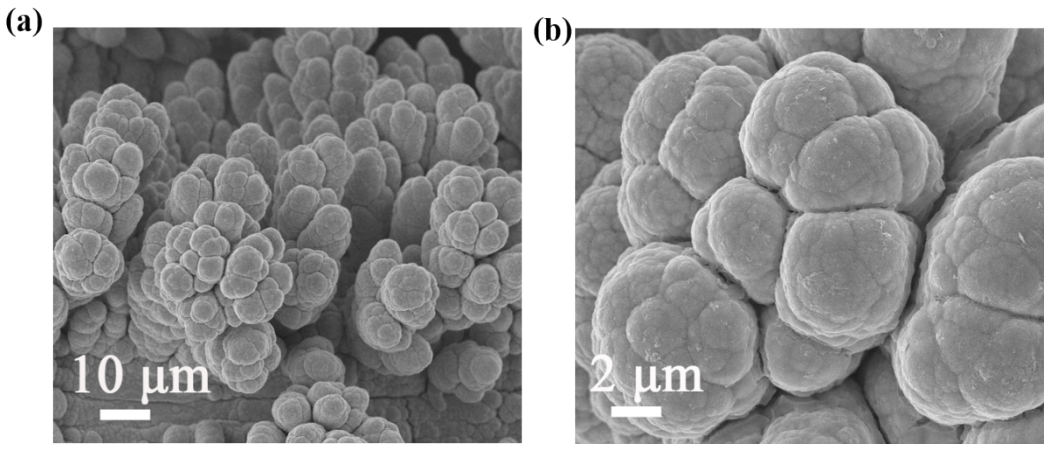

Fig. S17. SEM images of NiFeMo/SSM after OER test.

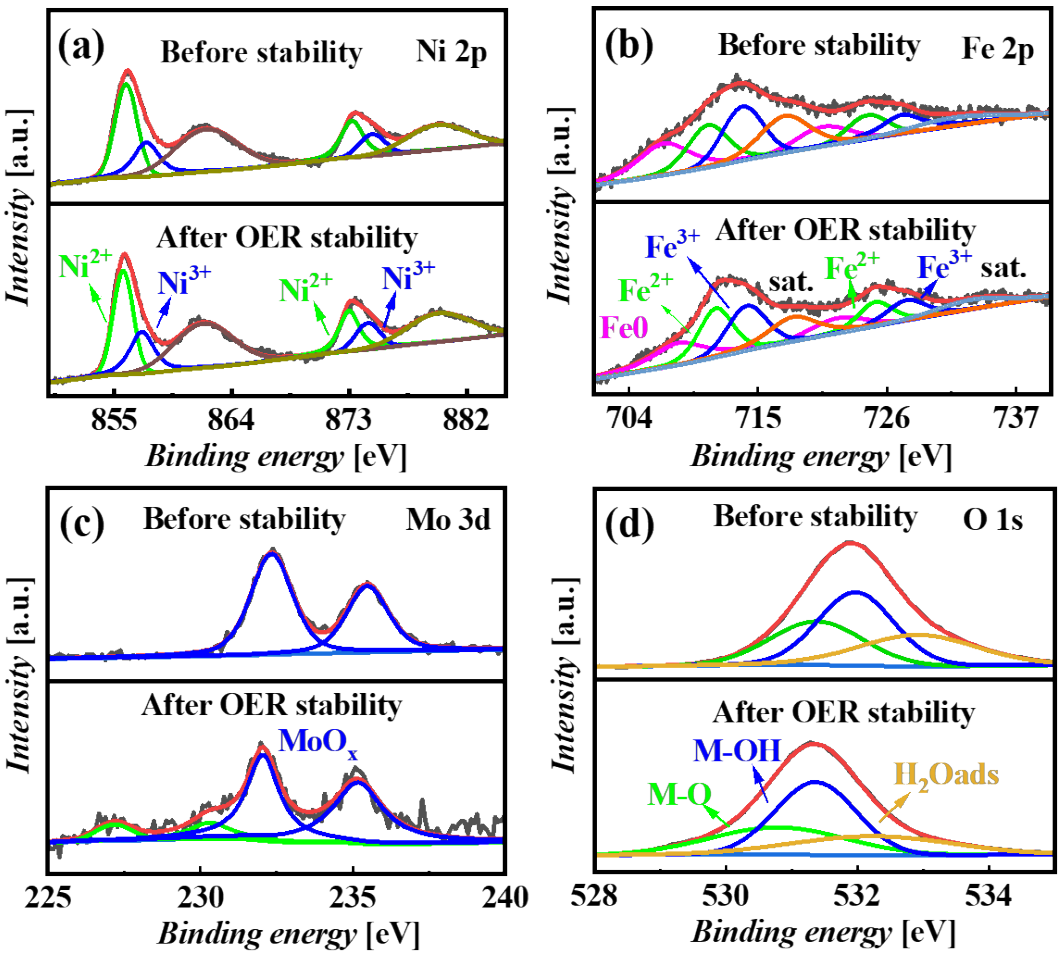

Fig. S18. (a) XPS spectra of Ni 2p of NiFeMo/SSM before and after OER stability test. (b) XPS spectra of Fe 2p of NiFeMo/SSM before and after the OER test. (c) XPS spectra of Mo 3d of NiFeMo/SSM before and after the OER test. (d) XPS spectra of O 1s of NiFeMo/SSM before and after the OER test. (top panel for before test, bottom panel for after OER).

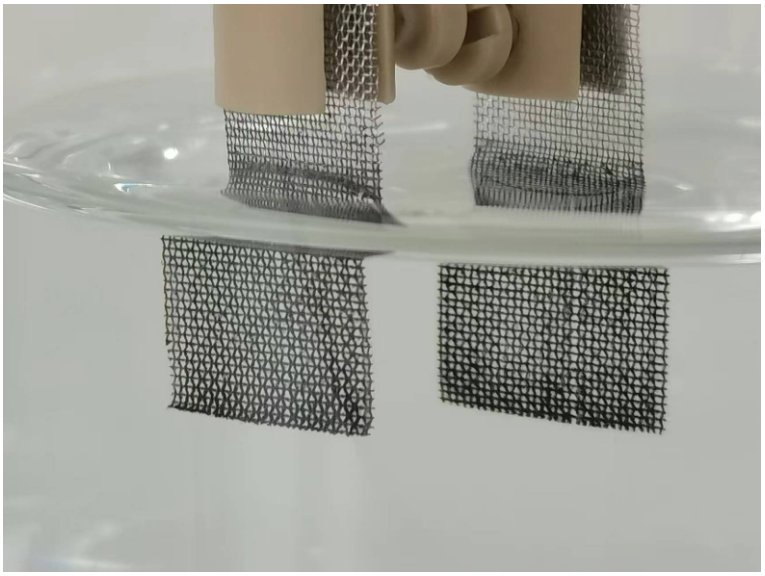

Fig. S19. Overall water splitting for a device with NiFeMo/SSM and 1.5V battery.

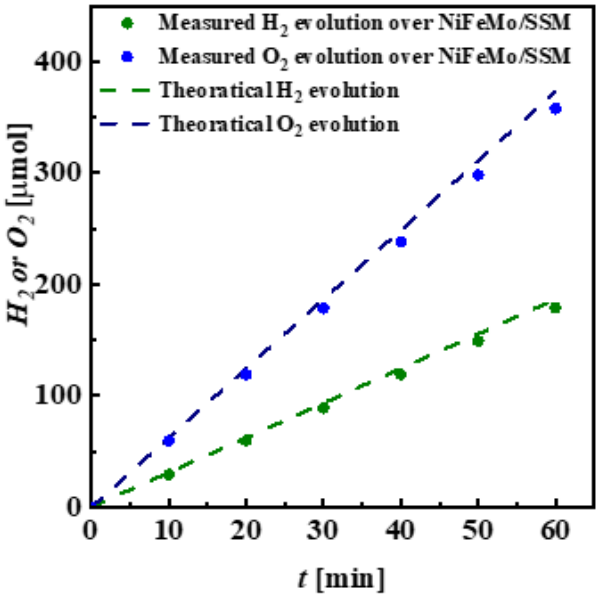

Fig. S20. Experimental and theoretical amounts of H_2 and O_2 by the NiFeMo/SSM electrode at a fixed current density of 10 mA cm^{-2}.

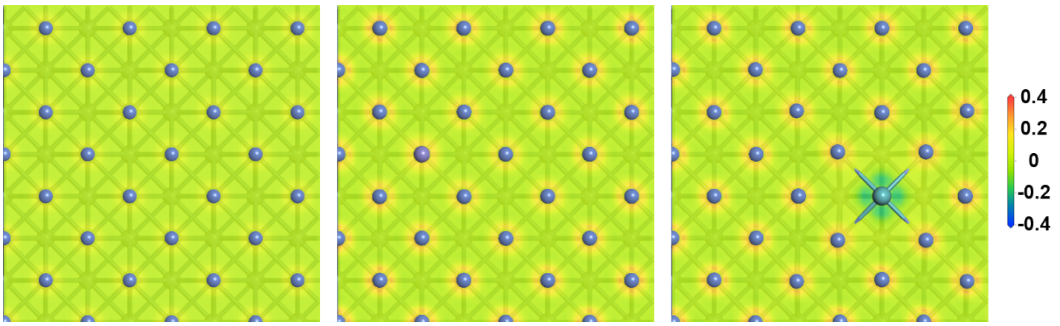

Fig. S21. Charge density difference for Ni、NiFe、NiMo model.

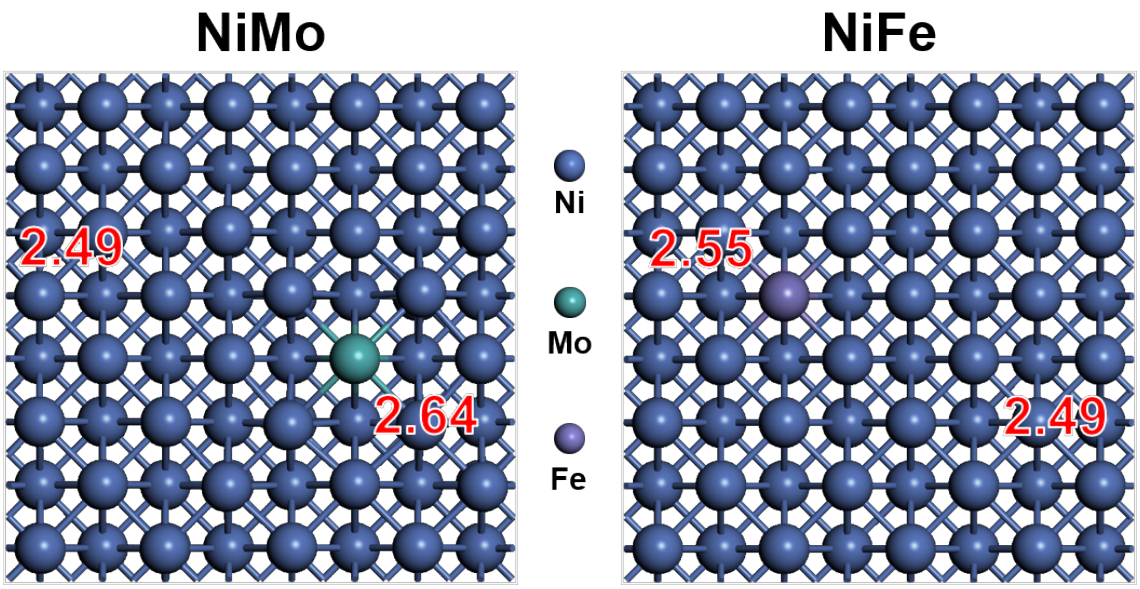

Fig. S22. Constructed computational model of NiMo and NiFe.

Table S2

Comparison of NiFeMo/SSM with recently reported electrocatalyst literature for HER in alkaline solution.

Materials	Electrolyte	Overpotential (mV)	J (mA cm^{-2})	Ref.
NiFeMo/SSM	**1.0 M KOH**	**86**	**10**	**This work**
NiMo	1.0 M KOH	58	10	[1]
FeMo	1.0 M KOH	66	10	[2]
$NiMoO_4$	1.0 M KOH	71	10	[3]
Ni-Mo-O/Ni_4Mo	1.0 M KOH	61	10	[4]
Ni_4Mo	1.0 M KOH	86	100	[5]
Ni/Mo-Ni	1.0 M KOH	24	10	[6]
		89	50	
		159	100	
NiMo	1.0 M KOH	33	10	[7]
		267	1000	
NiFeMo suboxides	1.0 M KOH	22	10	[8]
		117	100	
NiFeMo	1.0 M KOH	33	10	[9]
		249	500	
NiFeMo	1.0 M KOH	84.8	10	[10]
Mo-$NiFe_x$	1.0 M KOH	109.9	10	[11]

Table S3

Comparison of NiFeMo/SSM with recently reported electrocatalyst literature for OER in alkaline solution.

Materials	Electrolyte	Overpotential (mV)	J (mA cm^{-2})	Ref.
NiFeMo/SSM	**1.0 M KOH**	**318**	**50**	**This work**
MoFe-$Ni(OH)_2$/NiOOH	1.0 M KOH	280	100	[12]
NiFeMo	1.0 M KOH	230	20	[13]
NiFeMo suboxides	1.0 M KOH	255 289	10 100	[8]
NiFeMo	1.0 M KOH	230	10	[10]
Mo-NiFe-LDH	1.0 M KOH	317	20	[14]
NiFeMo film	1.0 M KOH	306	10	[15]
NiFeMo	1.0 M KOH	198 293	10 500	[9]
Mo-$NiFe_x$	1.0 M KOH	240	100	[11]
Mo-NiFe-LDH	1.0 M KOH	317	20	[14]

Reference

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