Energy Materials

Supplementary Materials

One-step hydrothermal synthesis of Fe single atom doped 1T-MoS_2 nanosheets for high-performance seawater hydrogen production

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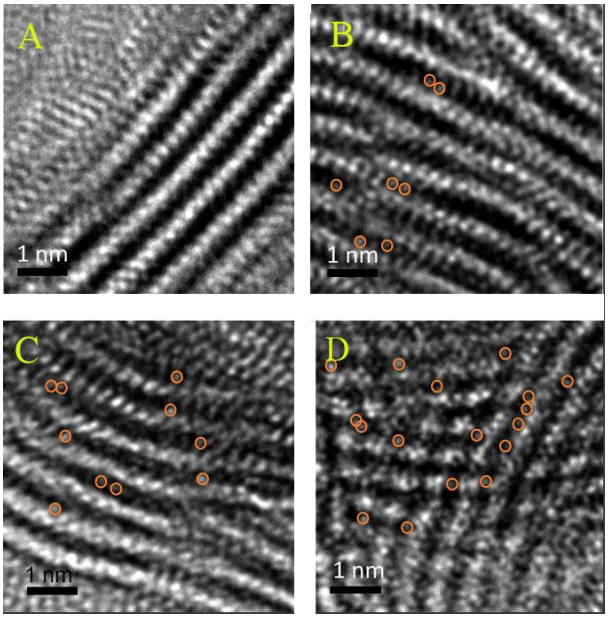

Figure 1. TEM images of (A) sample S0; (B) S2 and (C) S5 and (D) S20 respectively.

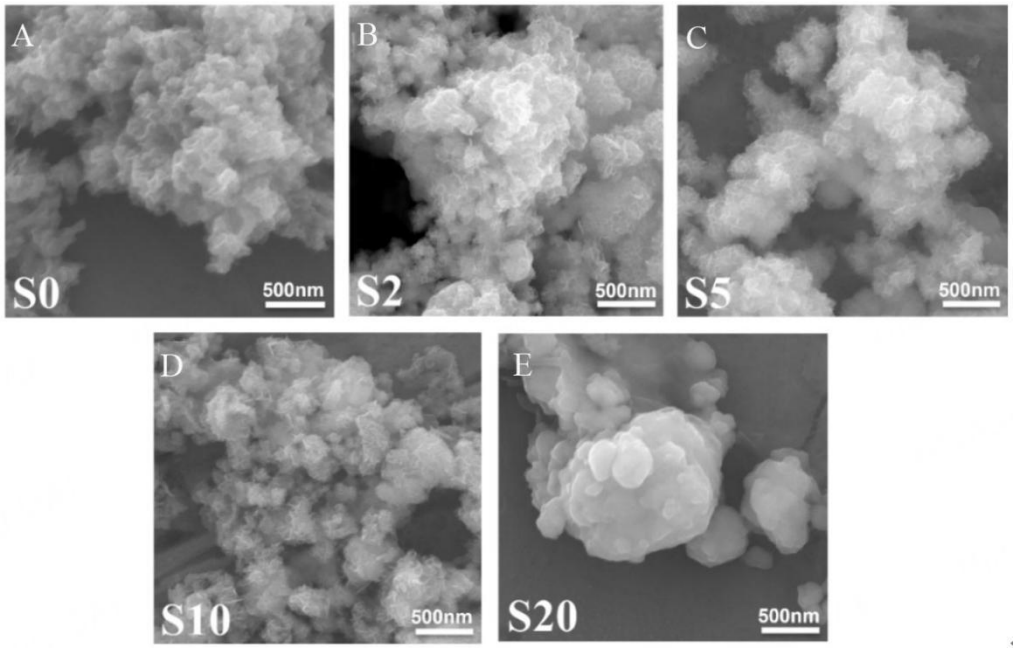

Figure 2. SEM images of sFe-1T/MoS_2 nanosheets for sample (A) S0; (B) S2; (C) S5; (D) S10 and (E) S20, respectively.

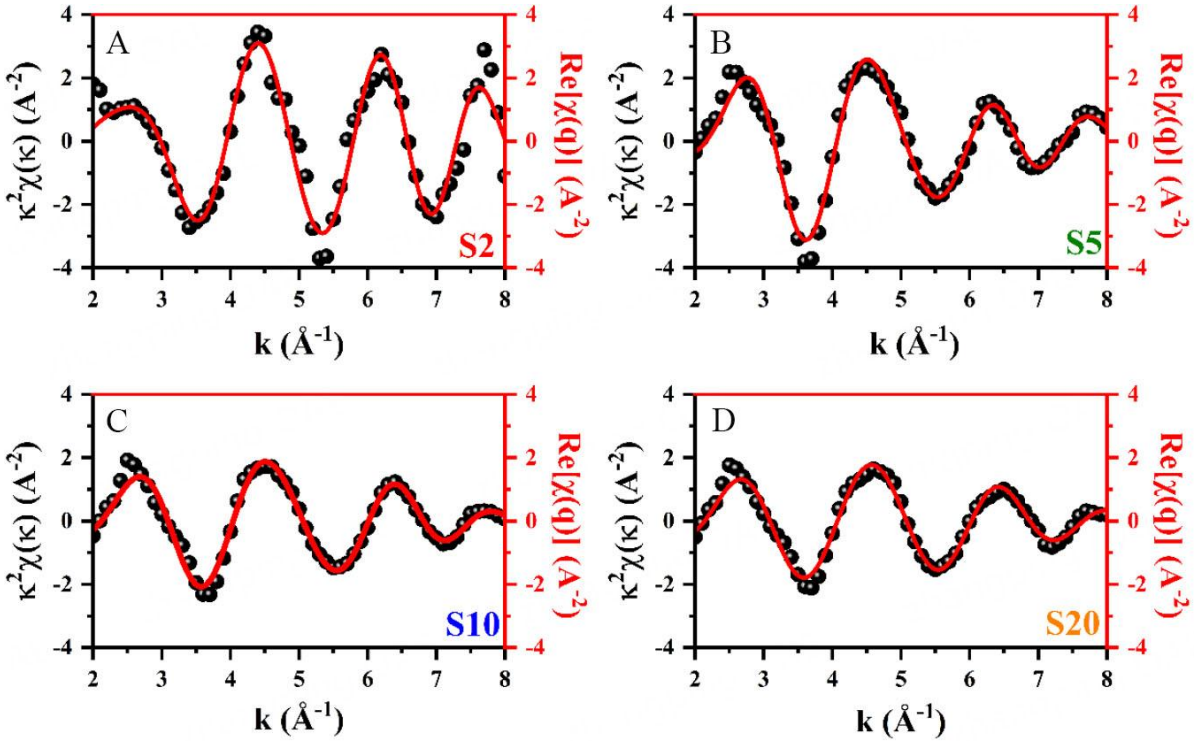

Figure 3. The forward-backward Fourier transformation (FT) transform for (A) S2; (B) S5; (C) S10 and (D) S20.

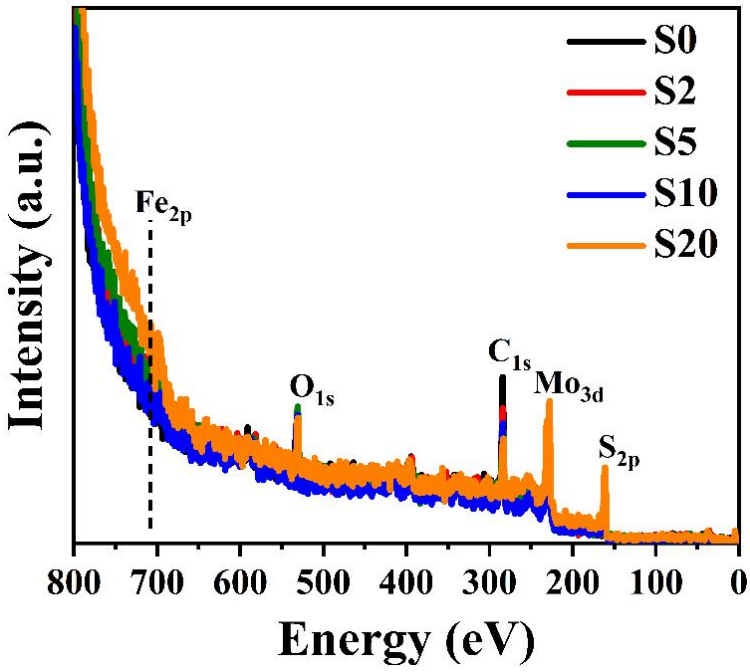

Figure 4. Survey curves of XPS for sFe-1T/MoS_2.

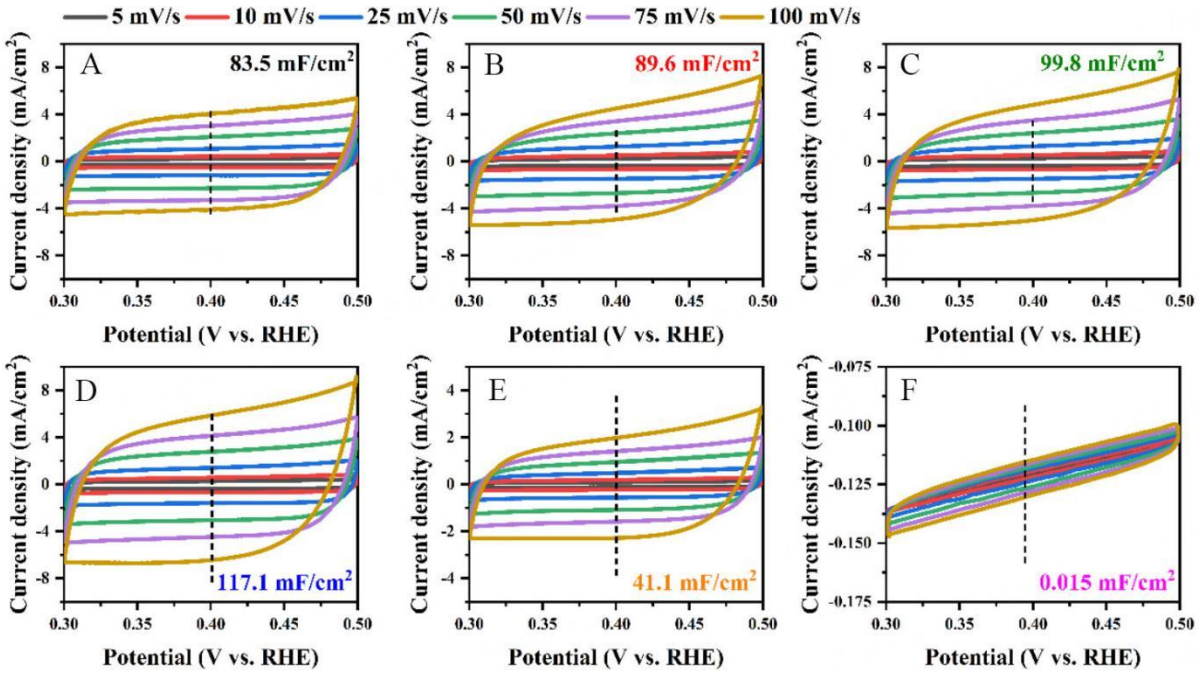

Figure 5. Non-Faradaic region Cyclic voltammetry (CV) curves at different scan rates (5, 10, 25, 50, 75 and 100 mV/s) for (A) S0; (B) S2; (C) S5; (D) S10; (E) S20 and (F) carbon electrode, respectively.

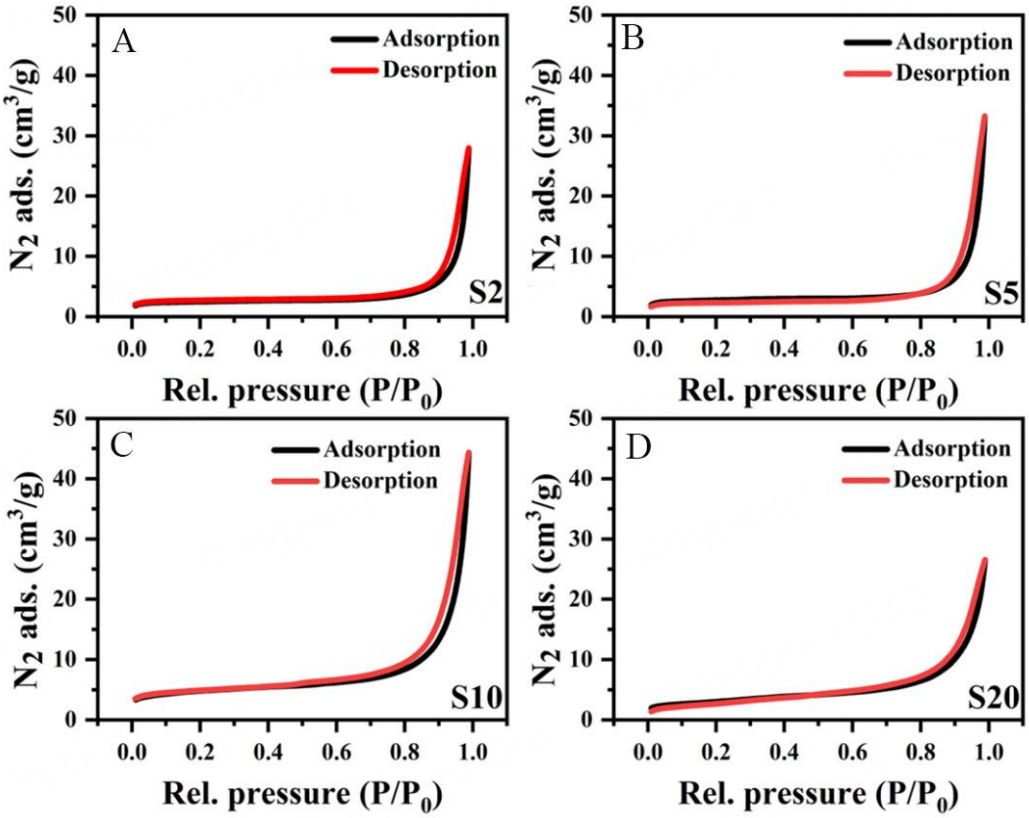

Figure 6. The N_2 absorption–desorption isotherms for sample (A) S2; (B) S5; (C) S10; (D) S20, respectively.

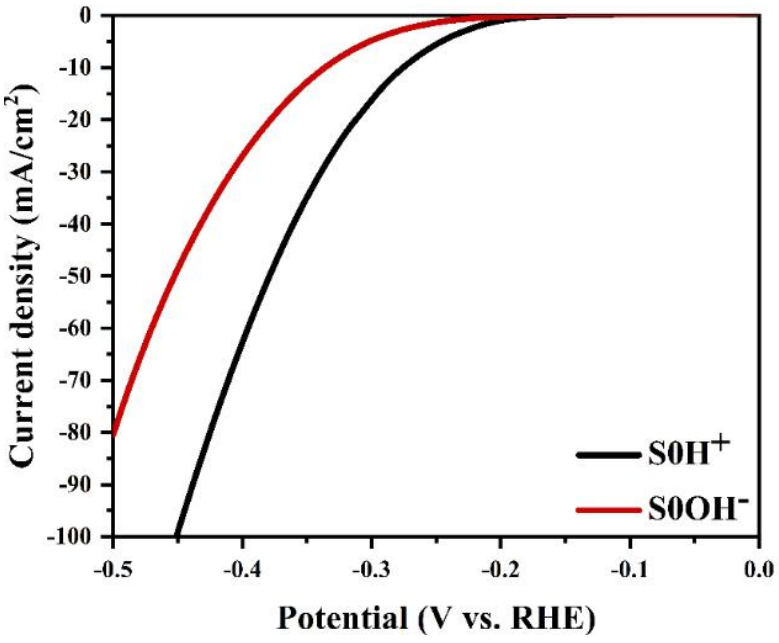

Figure 7. LSV curves of non-doped 1T MoS_2 measured in 0.5M H_2SO_4 and 1M KOH solution.

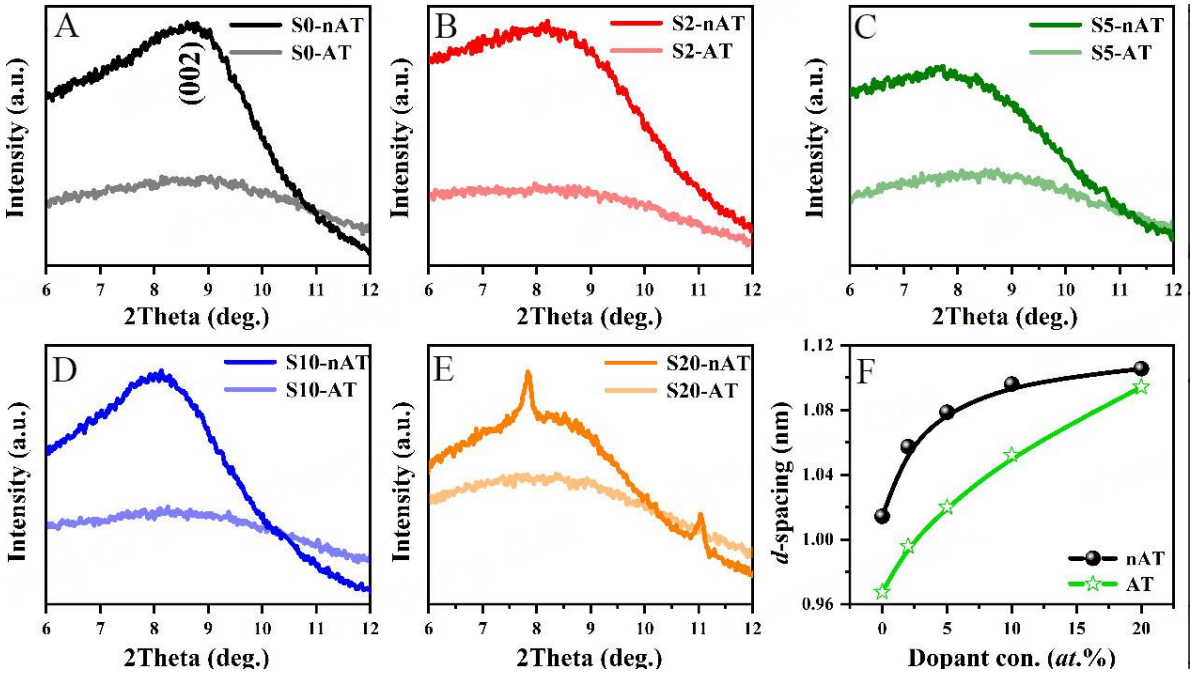

Figure 8. The zoomed-in XRD peak variation with acidic treatment (AT) and without acidic treatment (nAT) for (A) S0; (B) S2; (C) S5; (D) S10 and (E) S20; (F) The summary of *d*-spacing changes by acidic treatment.

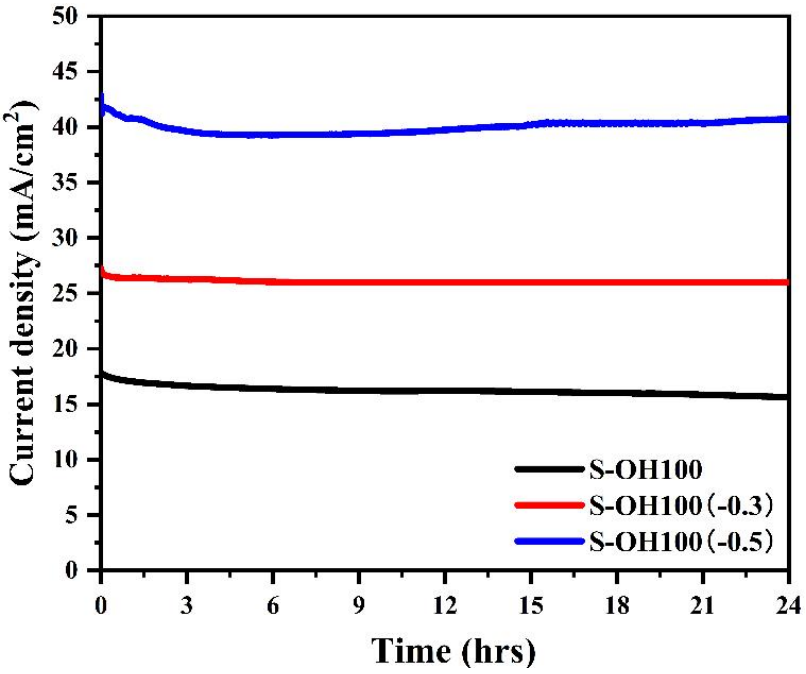

Figure 9. 24hrs stability measurement at a constant RHE potential of - 0.3 V and -0.5 V in alkaline (1M KOH) solutions, respectively.

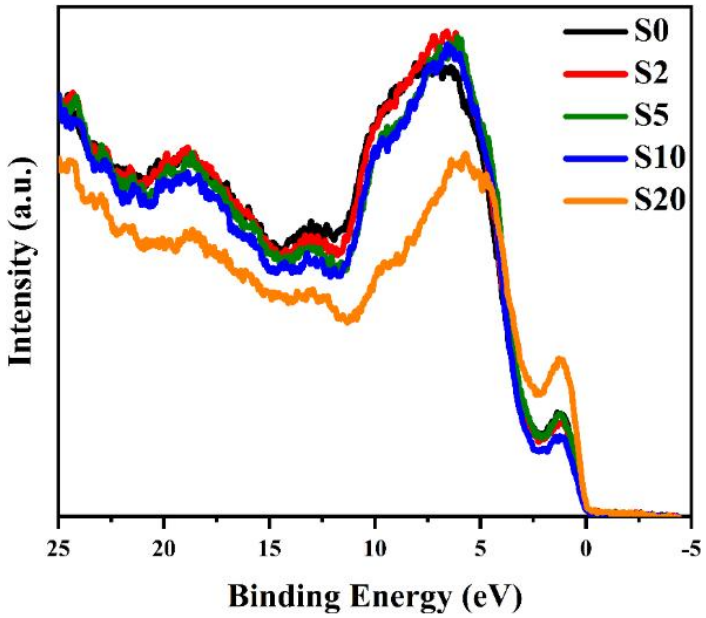

Figure. S10 Overall UPS spectra for sFe-1T/MoS_2.

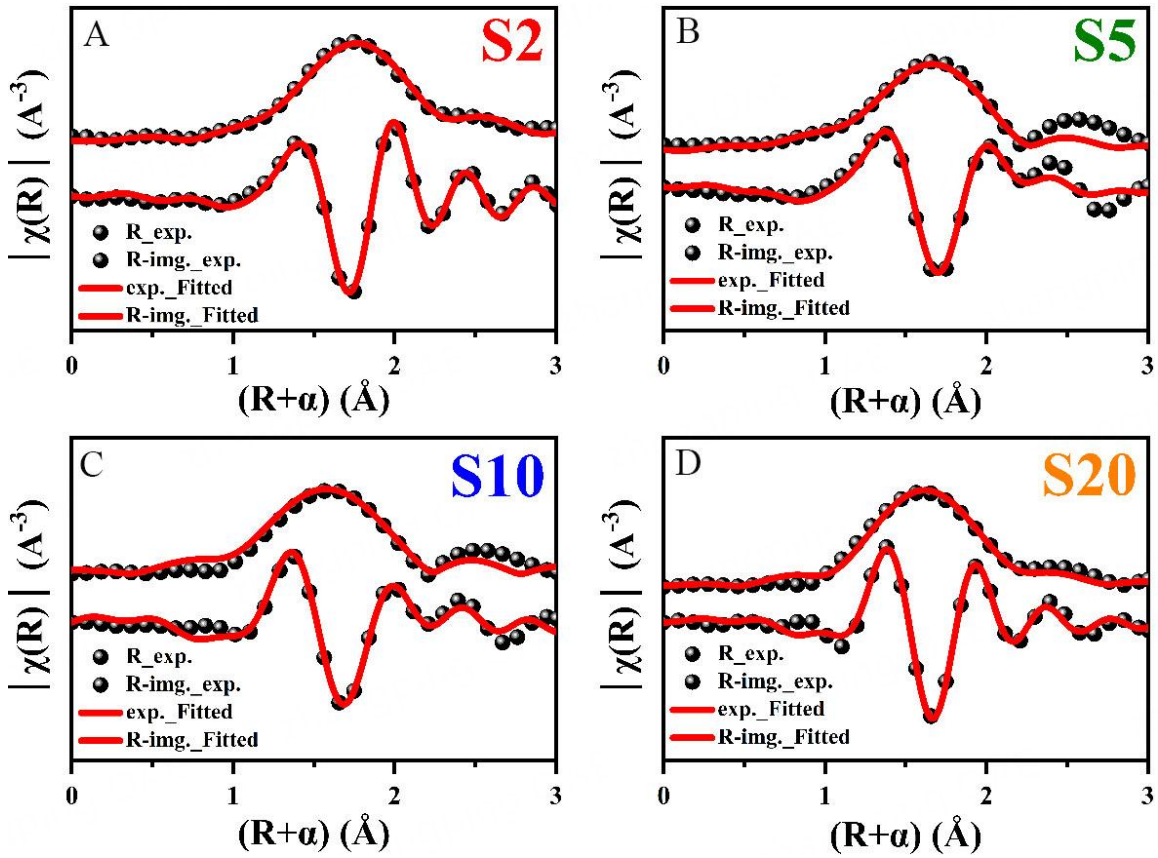

Figure 11. EXAFS Fe *K*-edge fitting curves for (A) S0; (B) S5; (C) S10 and (D) S20 for the real (R_exp.) and imaging (R-img.exp.) part of fitting formular, respectively.

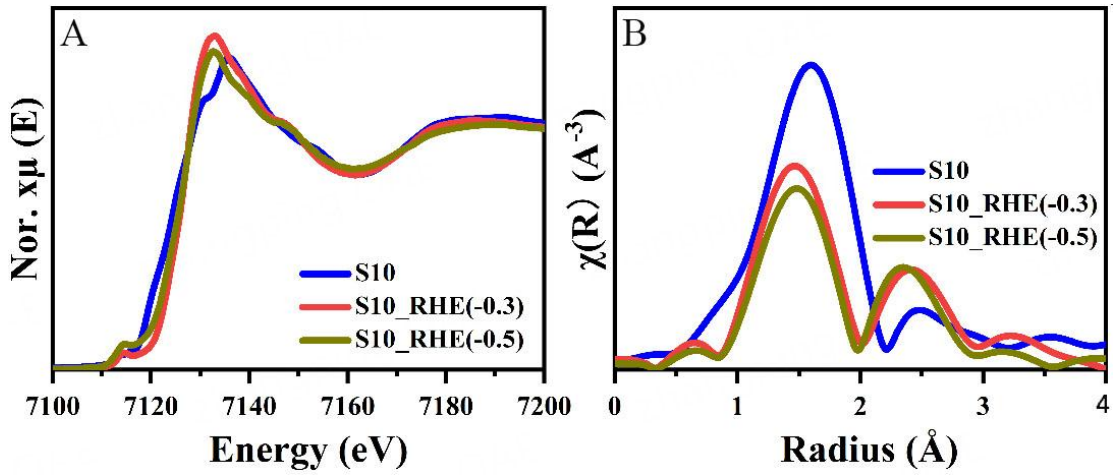

Figure 12. (A) NEXAFS curves of Fe K-edge before and after long-term catalysis at RHE potential of -0.3 V and RHE potential of -0.5 V in alkaline solution for sample S10; (B) Fourier-transform (FT) k^2-weighted NEXAFS spectra of sample S10 before and after long-term catalysis.

Table 1. Recent milestones on single element doping of 1T/MoS_2 on hydrogen evolution reaction (HER).

Dopants	Tafel slope (mV/dec)	Loading mass (mg/cm^2)	Electrolyte	Ref.
V	54	0.26		[1]
P	67	2.8		[2]
W	55	0.28		[3]
N	93	-	0.5M H_2SO_4	[4]
FeS_2	82	8.9		[5]
Pd	62	0.22		[6]
Pt	57	-		[7]
Ni	50	-	1M KOH	[8]
Fe	55	0.04	0.5M H_2SO_4	**This work**

Table 2. Summary of electrochemical measurements for Fe-1T/MoS_2 in acid solution with 40 μg/cm^2 loading mass

Name	Composition	Overpotential (mV)	Tafel slope (mV/dec)	Onset potential (mV)	C_{dl} (mF/cm^2)	EASA (cm^2)
S0	Fe_0-1T/MoS_2	274	70	245	83.3	4165

S2	$Fe_{0.02}$-1T/$Mo_{0.98}S_2$	254	57	223	89.6	4389
S5	$Fe_{0.05}$-1T/$Mo_{0.95}S_2$	234	56	210	99.8	4988
S10	$Fe_{0.1}$-1T/$Mo_{0.9}S_2$	215	55	190	117.1	5853
S20	$Fe_{0.2}$-1T/$Mo_{0.8}S_2$	260	75	230	41.4	2070

*Overpotential is obtained directly from LSV measurement at a current density of 10 mA/cm^2; *Onset potential is directly obtained in Tafel plots fitting at the current density of 1 mA/cm^2.

Table 3. Parameters of the NEXAFS Curve fitting for the Fe *K*-edge of sFe-1T/MoS_2.

Sample	***Path***	***Reff* (Å)**	***N***	***R* (Å)**	**σ^2 (Å)**
S2			6.5	2.327	0.01 ± 0.002
S5			6.4	2.310	0.009 ± 0.002
S10	Fe-S	2.239	6.1	2.293	0.006 ± 0.003
S20			5	2.287	0.002 ± 0.002

Reff: the standard radius distance obtained from the standard structure of FeS; ***N*:** coordination number, also fixed by the standard structure of FeS; *R*: radius distance for real path; σ^2: Debye-Waller factors are fitted based on global fit parameters. S_0^2 was fixed by 0.66, corresponding to previous reports; ΔE_0 was refined as a global fit parameter, returning a value of (-8 ± 1); Data ranges were set as from 2 to 8 for *k* and 1 to 3 for *R*, respectively; The number of variable parameters is 4. The R-factor for this fitting is 1.3%.

Table 4. Summary of seawater splitting performance of 10 *at.*% single-atom-doped Fe-1T/MoS_2 (S10) compared to recent Mo-based seawater electrocatalysts

Composition	**Overpotential (mV) at 10mAcm^{-2}**	***Additional***	**Ref.**
S-H10	318	10% v/v 0.5M	This

		H_2SO_4	work
S-H30	262	30% v/v 0.5M H_2SO_4	
S-OH10	325	10% v/v 1M KOH	
S-OH30	242	30% v/v 1M KOH	
CoNi-MoS$_2$-Pd$_{SA}$Ru$_{SA}$	1450	1M KOH	[9]
CC-MoC/MoS$_2$-H	136	-	[10]
CoMoP-C	450	-	[11]
Mo$_5$N$_6$	257	pH 8.4	[12]
h-MoN-BN-CNT	190	-	[13]
MoS$_2$-QDs/aerogel-100	400	-	[14]
Co$_x$Mo$_{2-x}$C/MXene/NC	312	-	[15]
Fe-MoS$_2$ array	119	-	[16]
MoS$_2$@W-G	115 86	1M KOH 0.5M H_2SO_4	[17]

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