Supplementary information

Ultrafast flash joule heating synthesis of the Pt/MoO_x heterostructure for enhancing the electrocatalytic hydrogen evolution reaction

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Supplementary Figures and Tables

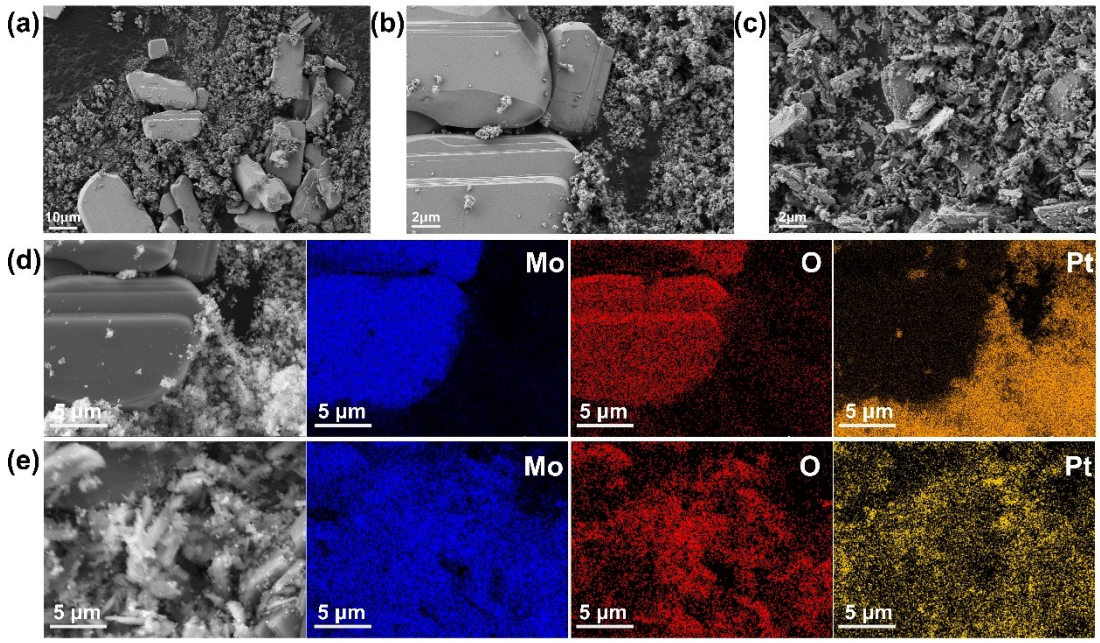

Fig. S1 SEM images of ungrounded Pt-MoO_3 mixture at 10 μm scale (a) and at 2 μm scale (b). SEM image of grounded Pt-MoO_3 mixture at 2 μm scale (c). The elemental mapping images of ungrounded (d) and grounded (e) Pt-MoO_3 mixture.

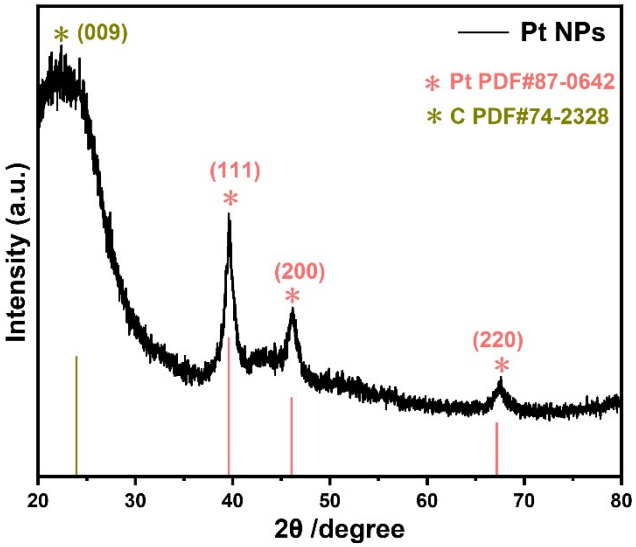

Fig. S2 The XRD pattern of the Pt NPs.

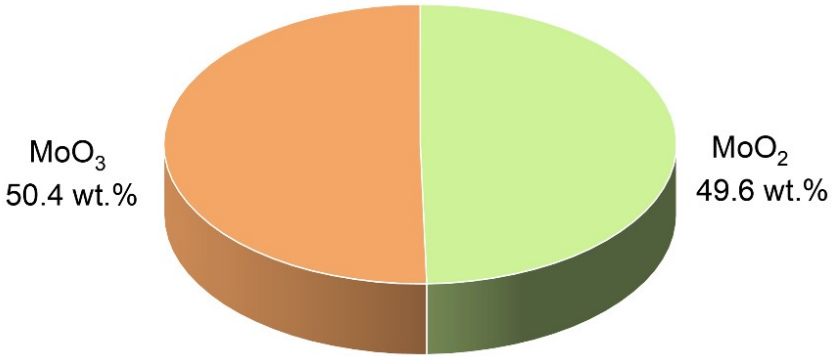

Fig. S3 The mass ratio of MoO_2 and MoO_3 from the quantitative analysis of the XRD of Pt/MoO_x-F.

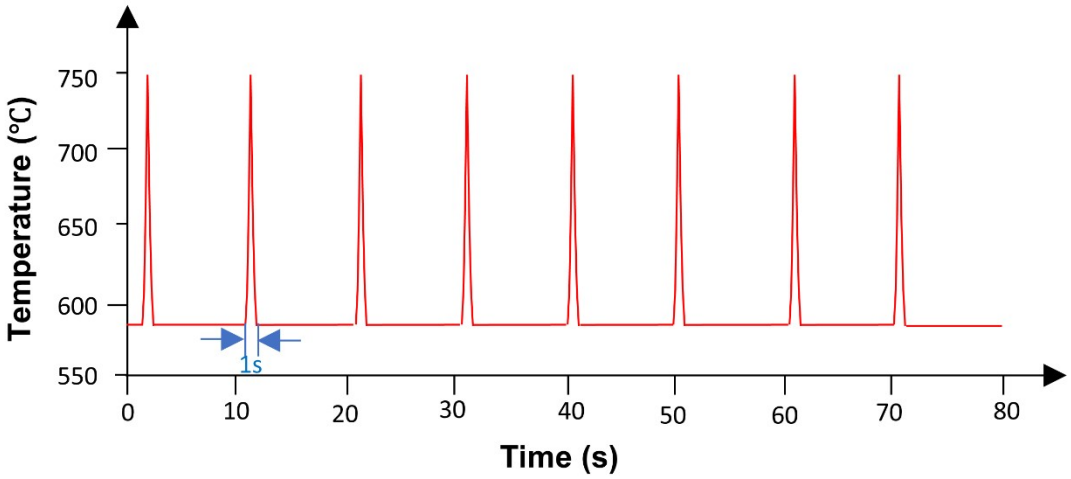

Fig. S4 Schematic diagram of the temperature-time evolution of the ultrafast joule heating eating method.

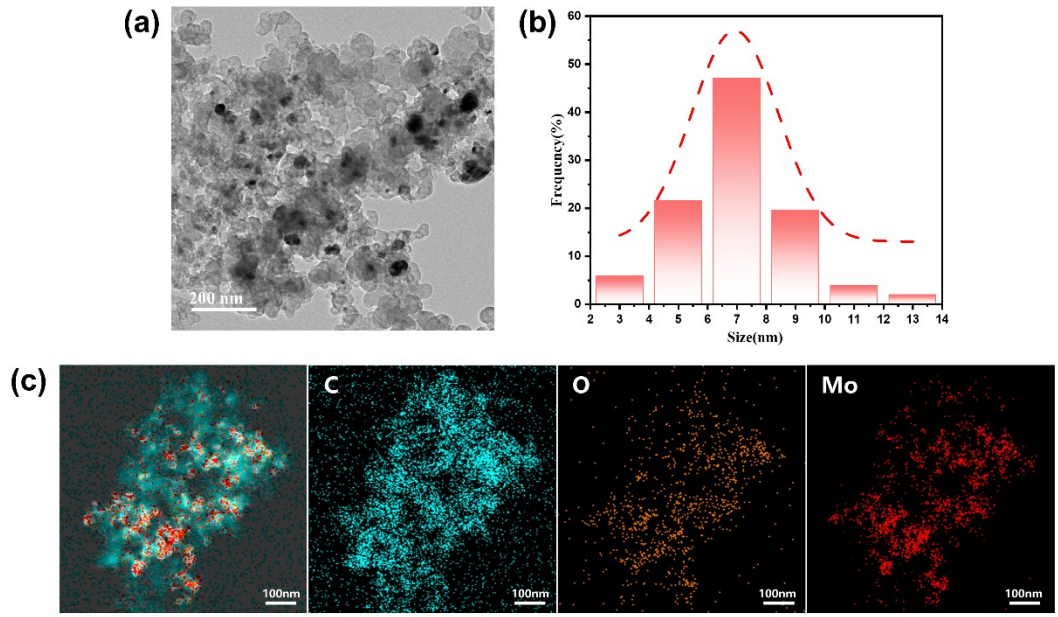

Fig. S5 (a) The TEM image of MoO_x. (b) The size distribution diagram of the metal nanoparticles on the Pt/MoO_x-F. (c) The elemental mapping images of MoO_x.

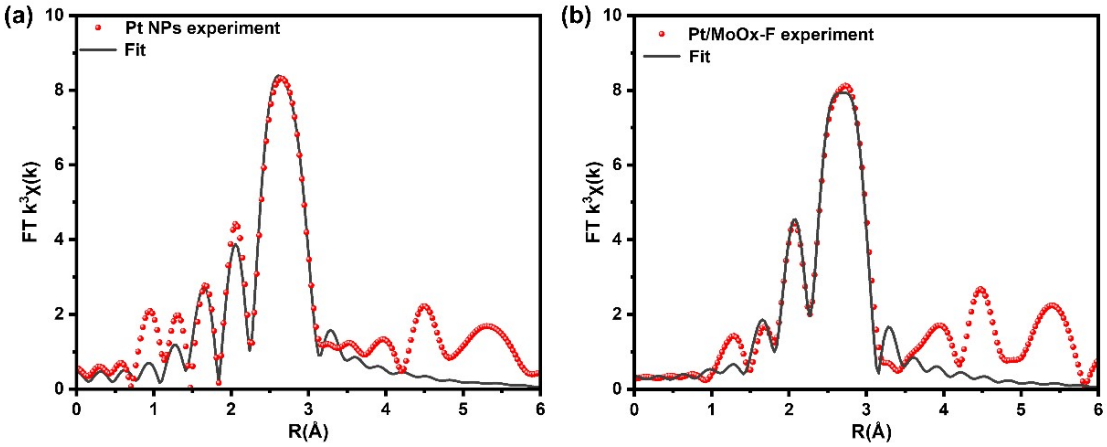

Fig. S6 Experimental and fitted EXAFS spectra of (a) Pt NPs and (b) Pt/MoO_x-F.

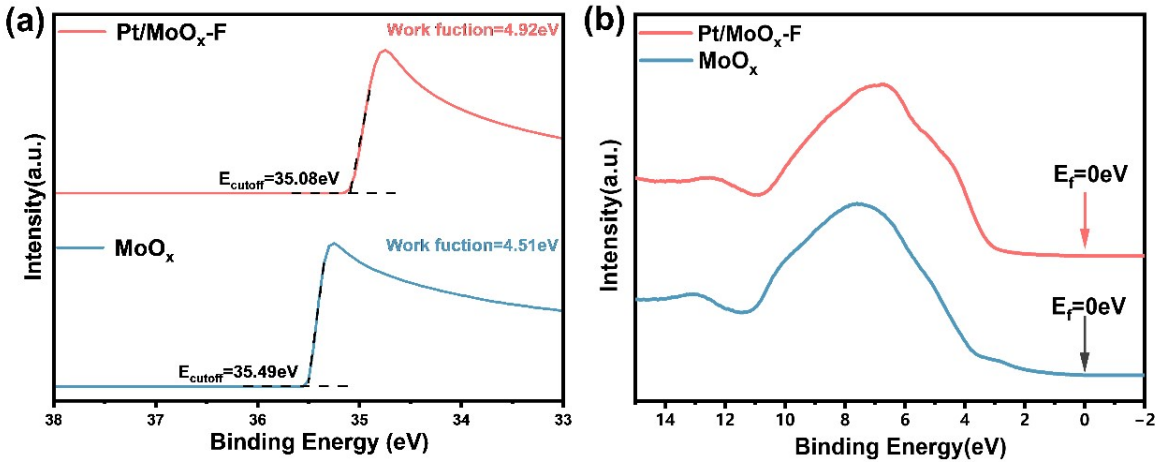

Fig. S7 UPS spectra of (a) SEC and (b) VB of MoO_x and Pt/MoO_x-F samples.

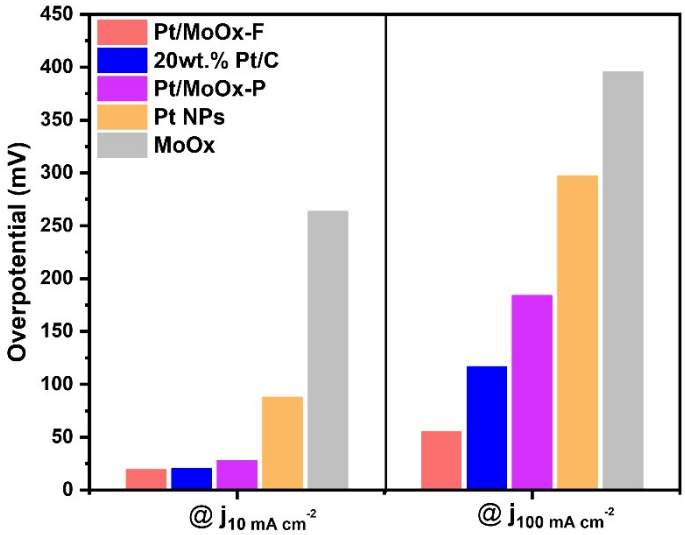

Fig. S8 The overpotential comparison of Pt/MoO_x-F and other reference samples at the current density of 10 mA cm^{-2} and 100 mA cm^{-2}.

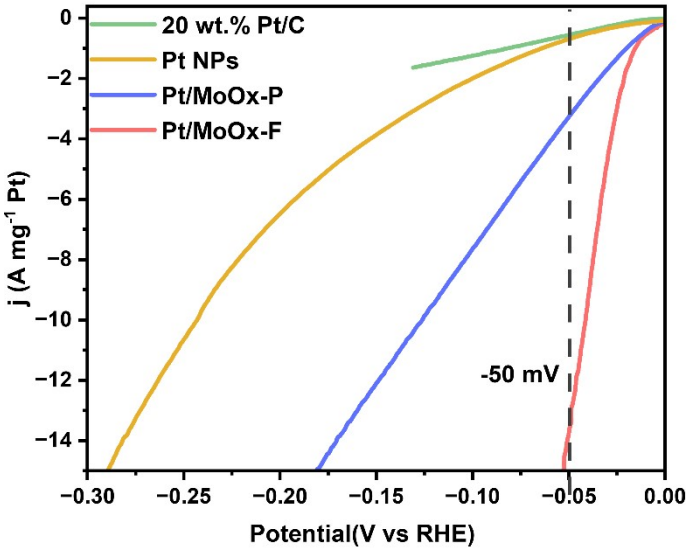

Fig. S9 LSV curves of as-prepared samples normalized by mass loading of Pt species.

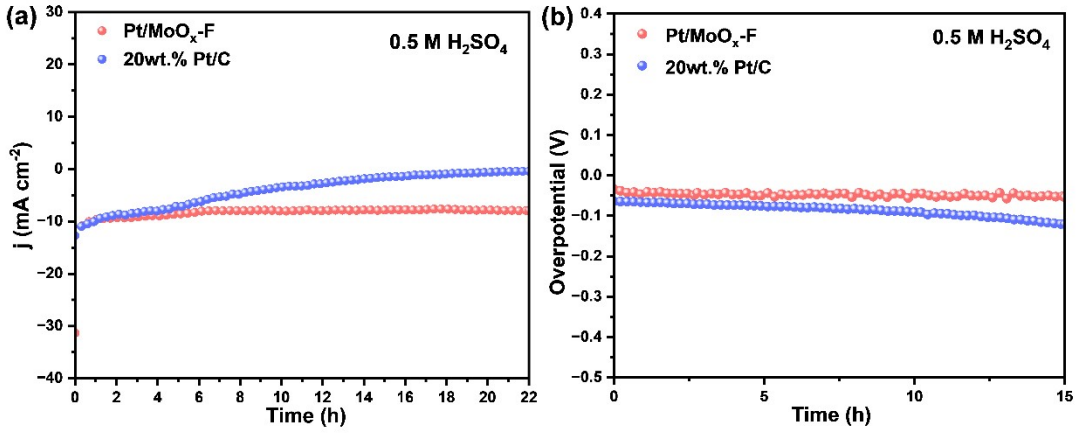

Fig. S10 (a) The plot of current density versus time for Pt/MoO_x-F under a constant voltage test in 0.5M H_2SO_4 electrolyte. (b) The plot of overpotential versus time for Pt/MoO_x-F under a constant current test.

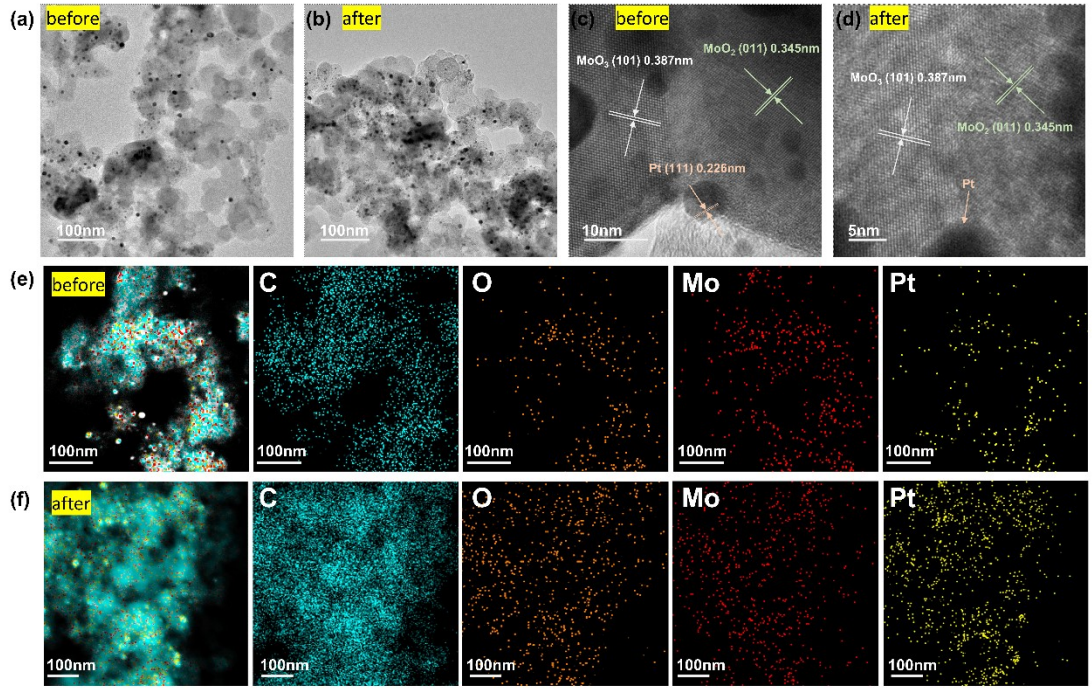

Fig. S11 Structural changes in the Pt/MoO_x-F before and after the HER. TEM images of Pt/MoO_x-F before (a) and after (b) the electrochemical stability tests. HR-TEM images of Pt/MoO_x-F before (c) and after (d) the electrochemical stability tests. The element distribution images of Pt/MoO_x-F before (e) and after (f) the electrochemical stability tests.

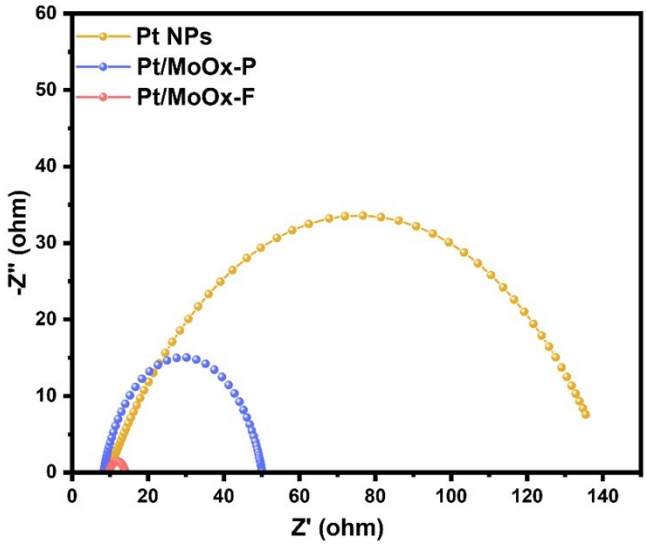

Fig. S12 Nyquist plots of Pt-MoO_x-F and reference samples.

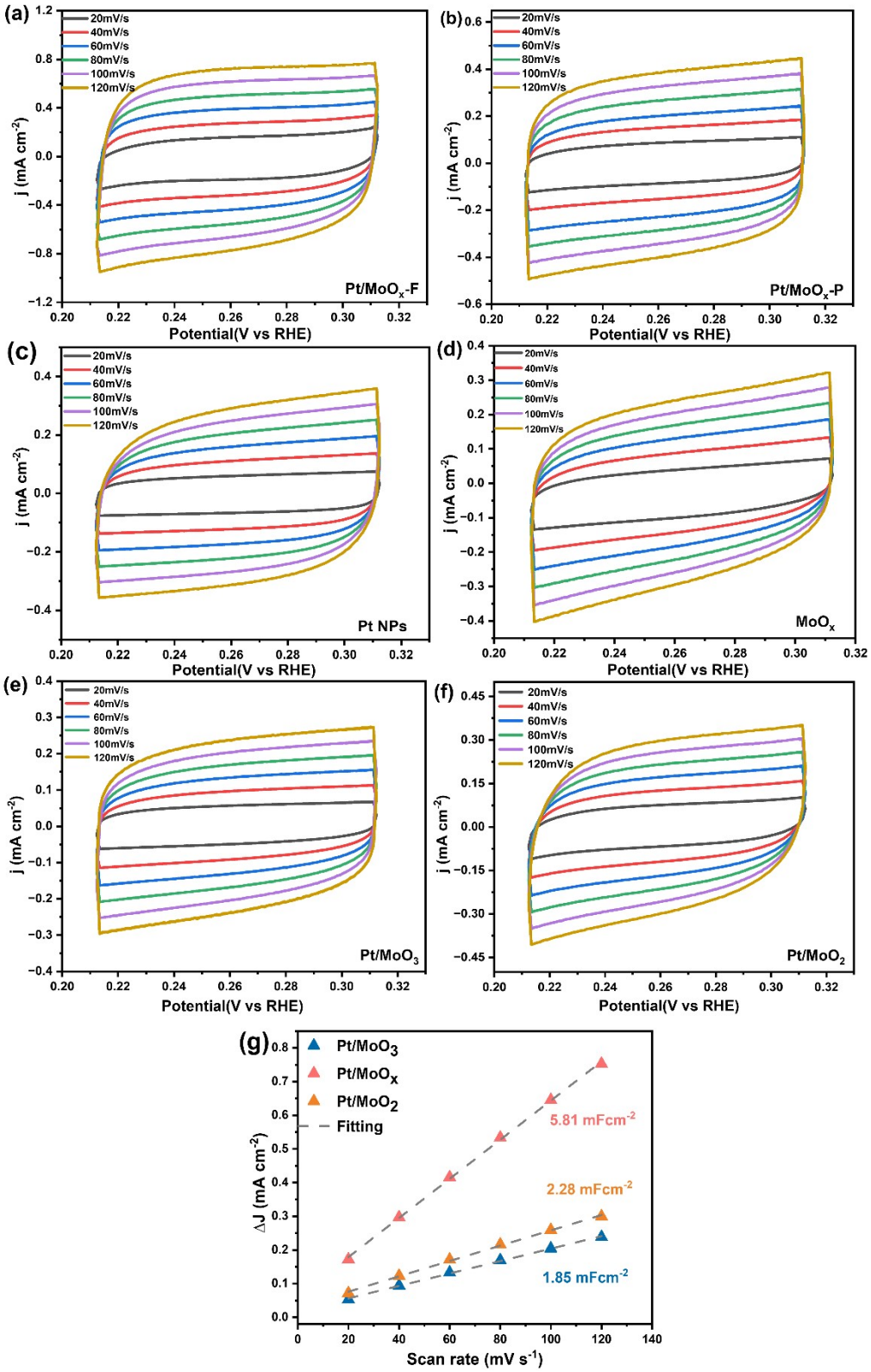

Fig. S13 Current difference at different scan rates for the estimation of the double-layered capacitance of (a) Pt/MoO_x-F, (b) Pt/MoO_x-P, (c) Pt NPs, (d) MoO_x, (e) Pt/MoO_3 and (f) Pt/MoO_2 in 0.5 M H_2SO_4. (g) Capacitive currents as a function of the scan rate of Pt/MoO_2 and Pt/MoO_3 samples.

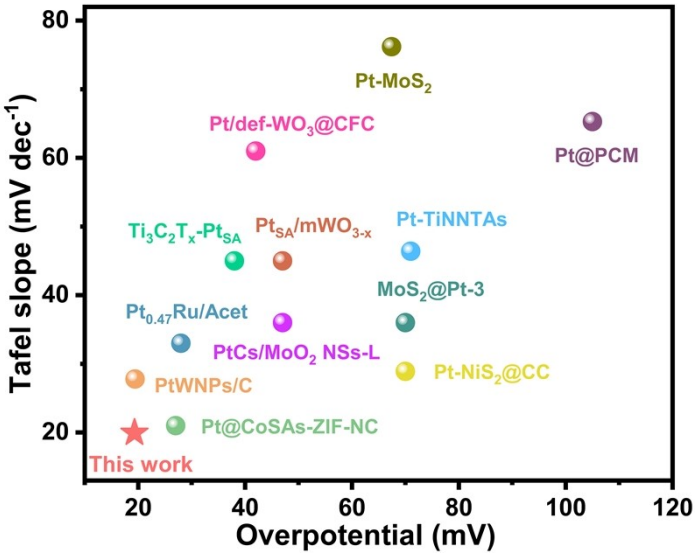

Fig. S14 In 0.5 M H_2SO_4 solution, comparison of the overpotentials and Tafel slopes at 10mA cm^{-2} with the references.

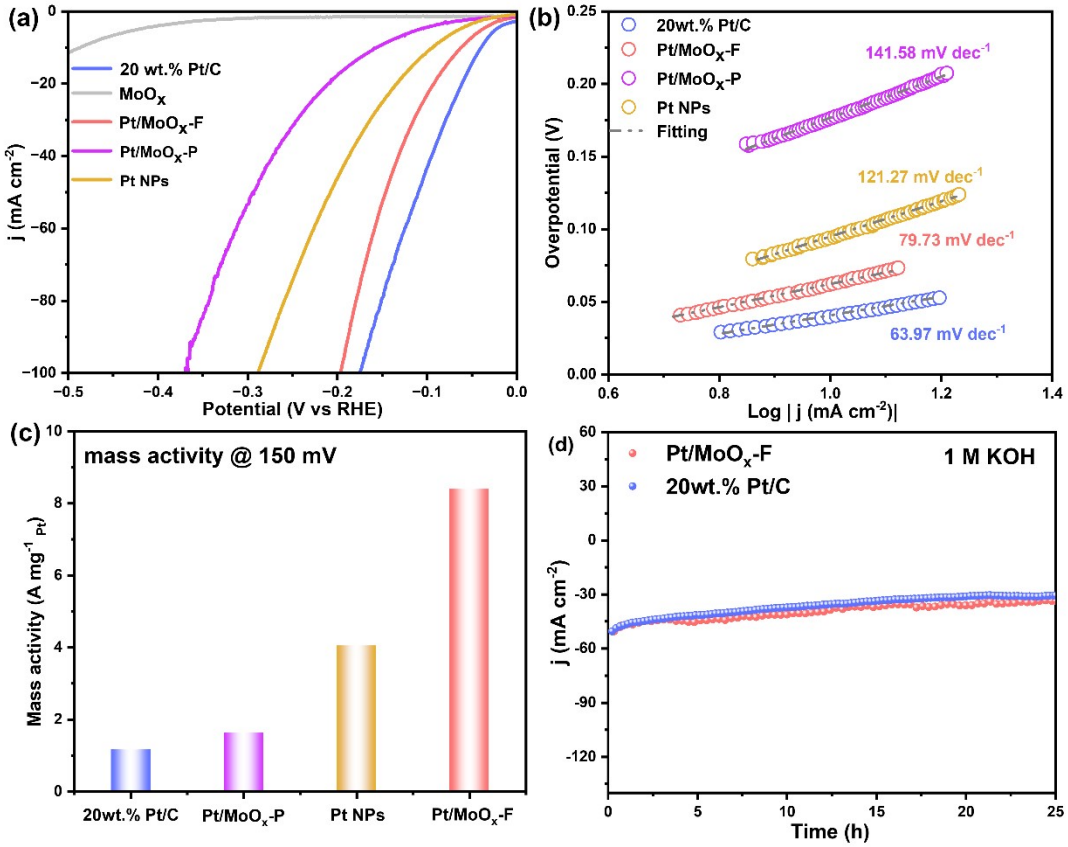

Fig. S15 Electrochemical HER performance under alkaline electrolyte. (a) Polarization curves of Pt/MoO_x-F in comparison with 20 wt.% Pt/C and other reference samples in KOH. (b) Tafel plots for Pt/MoO_x-F and other reference samples. (c) Mass activity at η =150 mV of the as-prepared samples. (d) The plot of current density versus time for Pt/MoO_x-F under a constant voltage test.

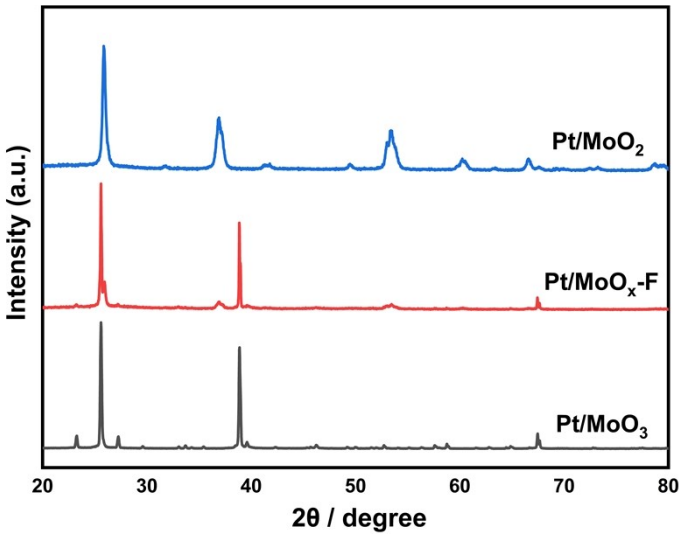

Fig. S16 XRD patterns of Pt/MoO_x-F, Pt/MoO_2 and Pt/MoO_3 samples.

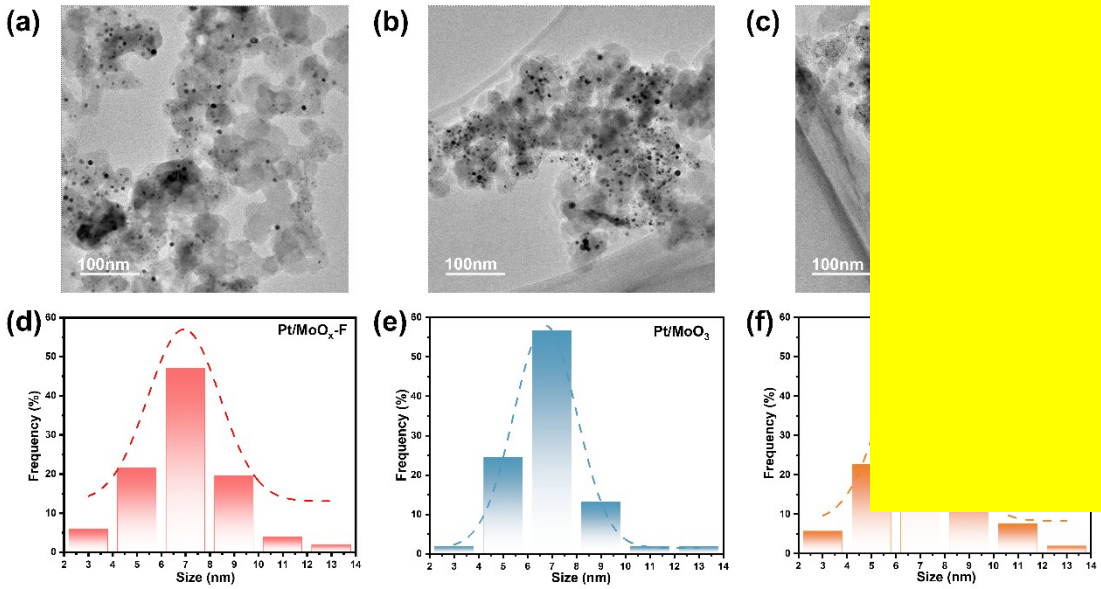

Fig. S17 (a) TEM image of Pt/MoO_x-F. (b) TEM image of Pt/MoO_3. (b) TEM image of Pt/MoO_2. The size distribution diagram of the metal nanoparticles on the Pt/MoO_x-F (d), Pt/MoO_3 (e), and Pt/MoO_2 (f).

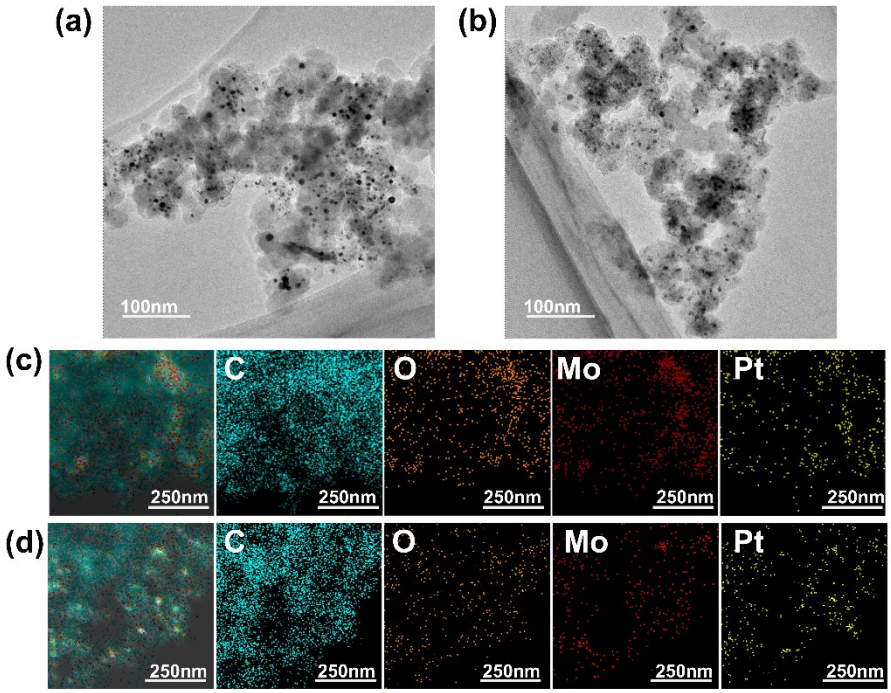

Fig. S18 (a) TEM image of Pt/MoO_3. (b) TEM image of Pt/MoO_2. (c) The elemental mapping images of Pt/MoO_3. (d) The elemental mapping images of Pt/MoO_2.

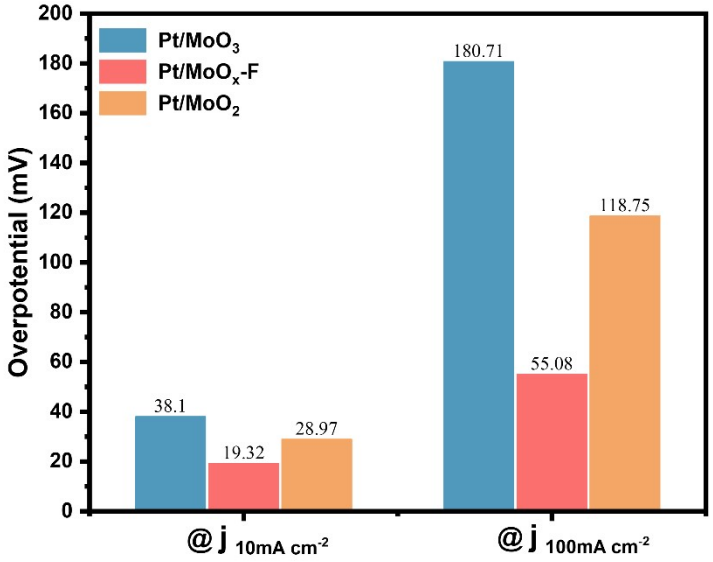

Fig. S19 The overpotential comparison of Pt/MoO_x-F and other reference samples at the current density of 10 mA cm^{-2} and 100 mA cm^{-2}.

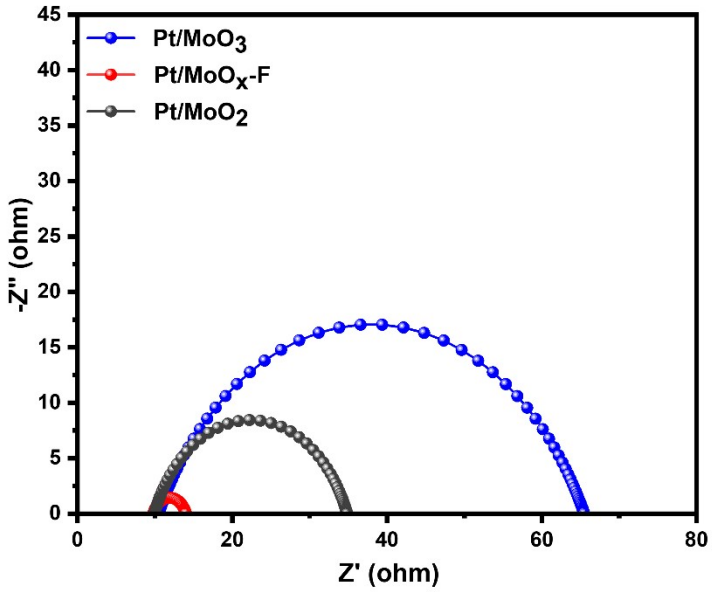

Fig. S20 Nyquist plots of Pt-MoO_x-F and reference samples.

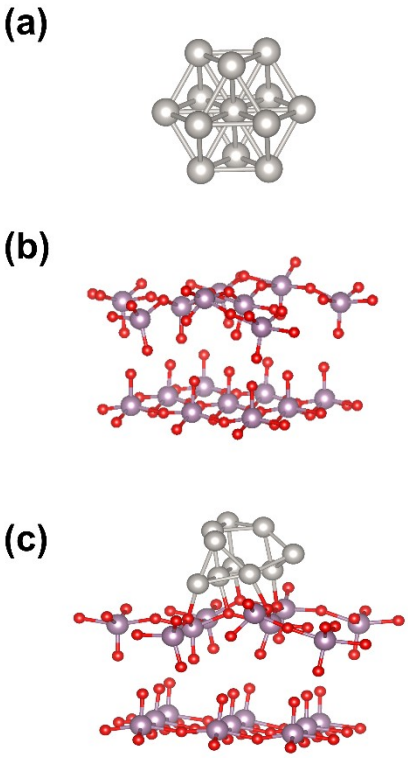

Fig. S21 The DFT computational models of (a) Pt, (b) MoO_x-F and (c) Pt/MoO_x-F.

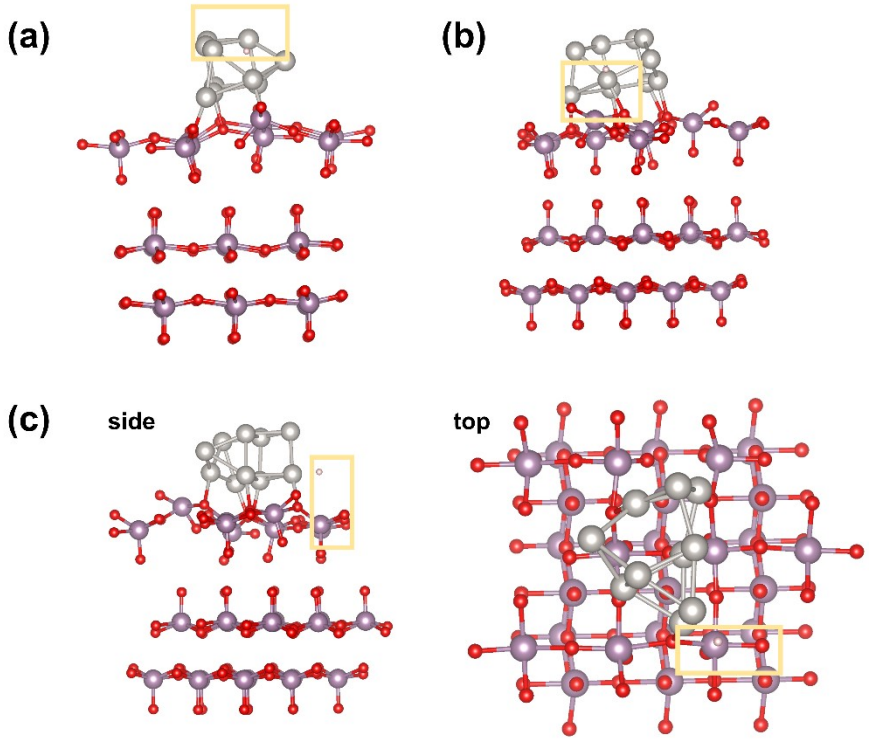

Fig. S22 The DFT model diagram of three different adsorption hydrogen sites on the Pt/MoO_x-F.

Table S1 The spectrogram of the total number of distribution maps of Pt/MoO_x-F.

Spectrogram of the total number of distribution maps	
Element	Atom %
O	3.76
Mo	1.57

Table S2 The percentage of Pt loading of as-prepared samples was measured by ICP-AES.

Sample	Pt Content (wt.%)
Pt/MoO_x-F	1.853
Pt/MoO_x-P	1.854
Pt NPs	1.857
Pt/MoO_3	1.853
Pt/MoO_2	1.856

Table S3 The EXAFS fitting results of Pt/MoO_x-F and reference samples.

sample	path	CN	Bond length(Å)	Debye-Walker coefficient ($Å^2$)	$\triangle E_0$(eV)	R factor
Pt foil	Pt-Pt	12	2.76	0.004	7.97	0.001
PtO_2	Pt-O	6	2.00	0.003	9.37	0.009
Pt NPs	Pt-Pt	6.18	2.76	0.005	8.23	0.008
	Pt-O	1.12	2.00	0.007		
Pt/MoO_x-F	Pt-Pt	8.67	2.76	0.006	7.74	0.016
	Pt-O	0.51	2.01	0.009		
	Pt-Mo	0.94	2.75	0.008		

Binding Energies (eV)								
Samples	Mo^{4+}		Mo^{5+}		Mo^{6+}		Pt	
	$Mo3d_{5/2}$	$Mo3d_{3/2}$	$Mo3d_{5/2}$	$Mo3d_{3/2}$	$Mo3d_{5/2}$	$Mo3d_{3/2}$	$Pt4f_{7/2}$	$Pt4f_{5/2}$
MoO_x	229.25	232.38	231.41	234.54	232.71	235.84	-	-
Pt/MoO_x-F	-	-	231.63	234.76	232.65	235.78	71.65	74.95
Pt NPs	-	-	-	-	-	-	72.23	75.53

Table S4 The XPS spectra-specific parameters.

Table S5. Comparison of HER performance between Pt/MoO_x-F and the recently reported Pt-based electrocatalysts in 0.5 M H_2SO_4.

Sample	η_{10} (mV)	η_{100} (mV)	Mass activity ($A\ mg^{-1}_{Pt}$)	Tafel slope ($mV\ dec^{-1}$)	Reference
Pt/MoO_x-F	19.32	55.08	13.80@50mV	19.94	This work
Pt-SAs/WS_2	32	170	130.2@100mV	28	[1]
$Mo_2TiC_2T_xPt_{SA}$	30	77	8.3@77 mV	30	[2]
Pt_{SA}/mWO_{3-x}	47	-	12.8@50 mV	45	[3]
Pt-TiO_{2-x} NSs	36	180	~0.85@150mV	32.1	[4]
$Pt_{0.47}Ru$/Acet	28	~80	2.63@100mV	33.3	[5]
Pt/Ni-DA	18	-	2.13@50mV	34	[6]
Pt–WO_x/WS_2	42	~300	0.68@112 mV	26	[7]
Pt-MoS_2	67.4	-	-	76.2	[8]
Pt_{SA}/NT/NF	30	88	0.93@100mV	-	[9]
$Pt/V_2O_3/V_8C_7$	45	78	0.64@100mV	30.2	[10]
Pd7@Pt3	33	90	-	23.1	[11]
0.8% Pt-Naf-CV	34	143	-	33	[12]
PtW NPs/C	19.4	-	0.566@20mV	27.8	[13]
Pt_{SA}/α-MoC_{1-x}@C-0.75	12	120	31.56@100mV	27	[14]

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