Electronic Supplementary Material (ESI) for Sustainable Energy & Fuels.

Supplymentary Information

Triple synergistic effect from pitaya-like $MoNi_x$-MoC_x hybrids encapsulated in N-doped C nanospheres for efficient hydrogen evolution

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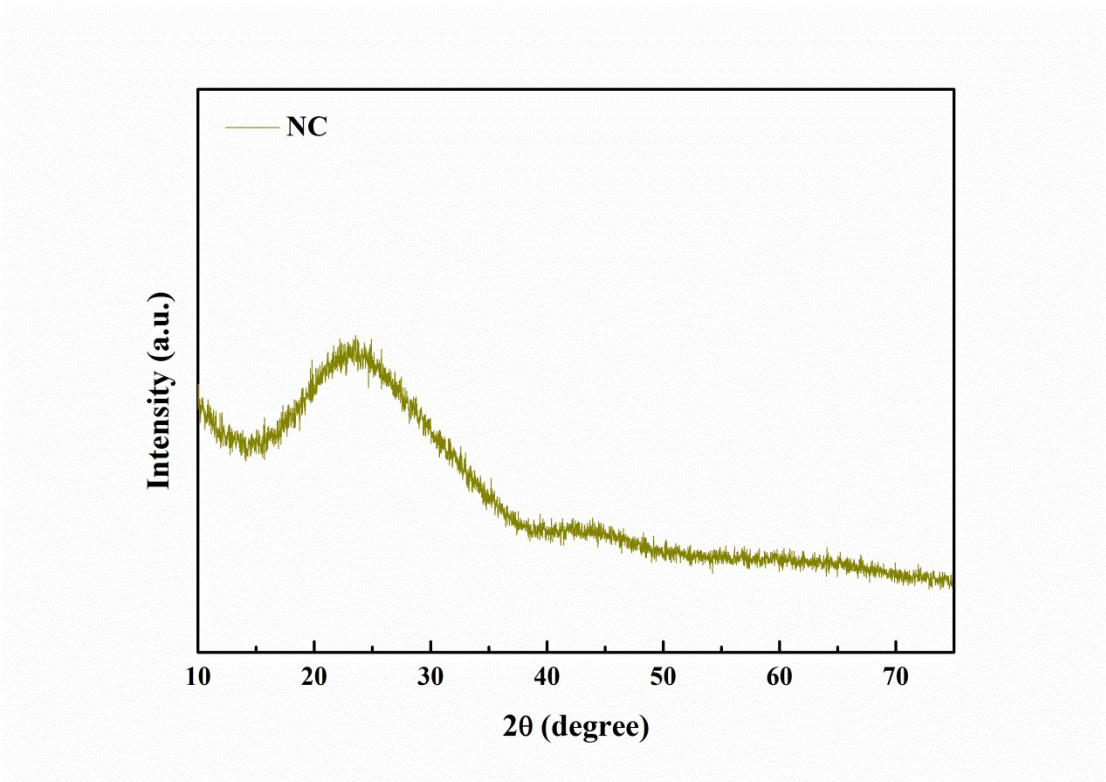


Fig. S1 XRD patterns of NC.

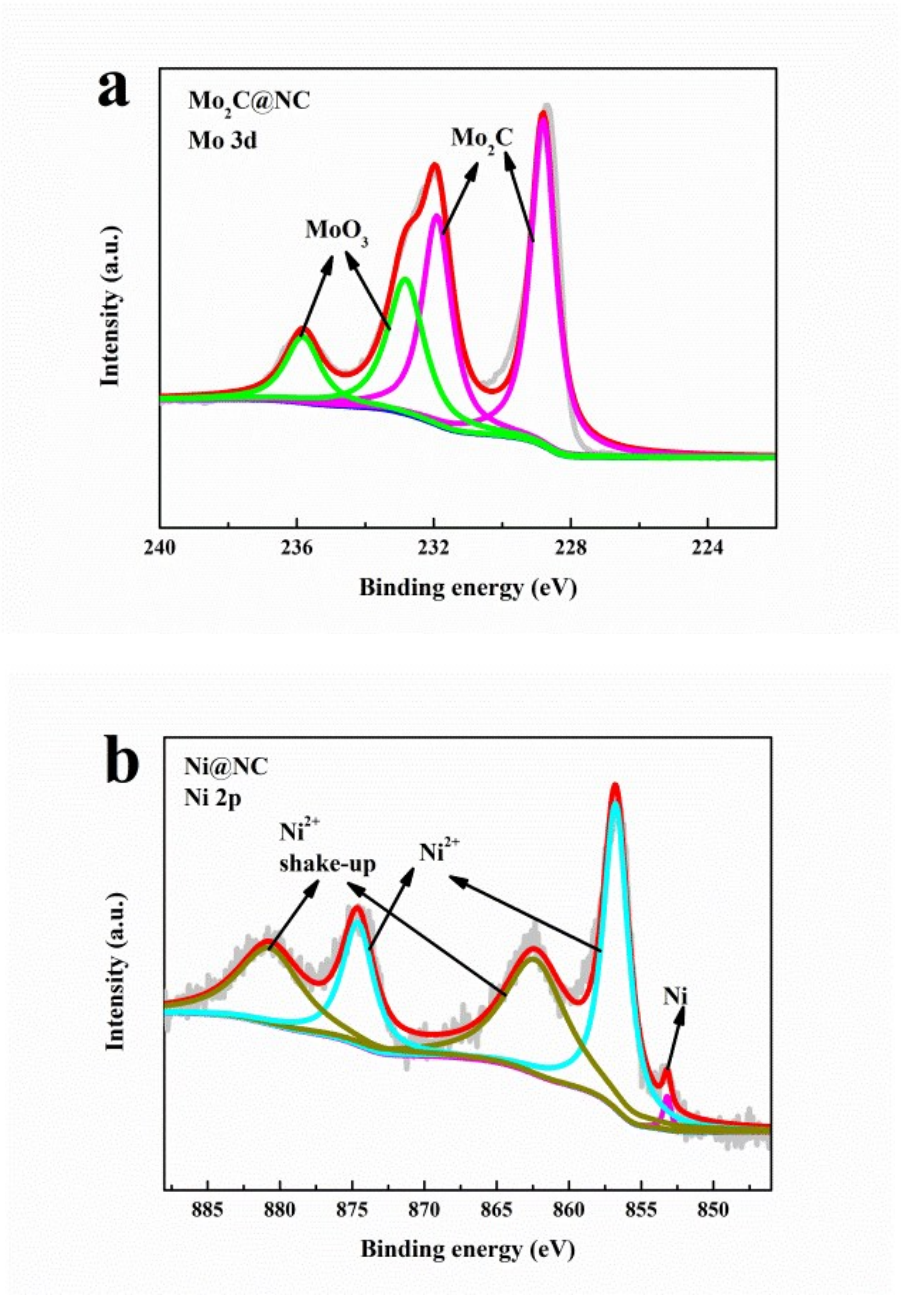


Fig. S2 (a) XPS high-resolution scans for Mo_2C@NC in Mo 3d and (b) XPS high-resolution scans for Ni@NC in Ni 2p.

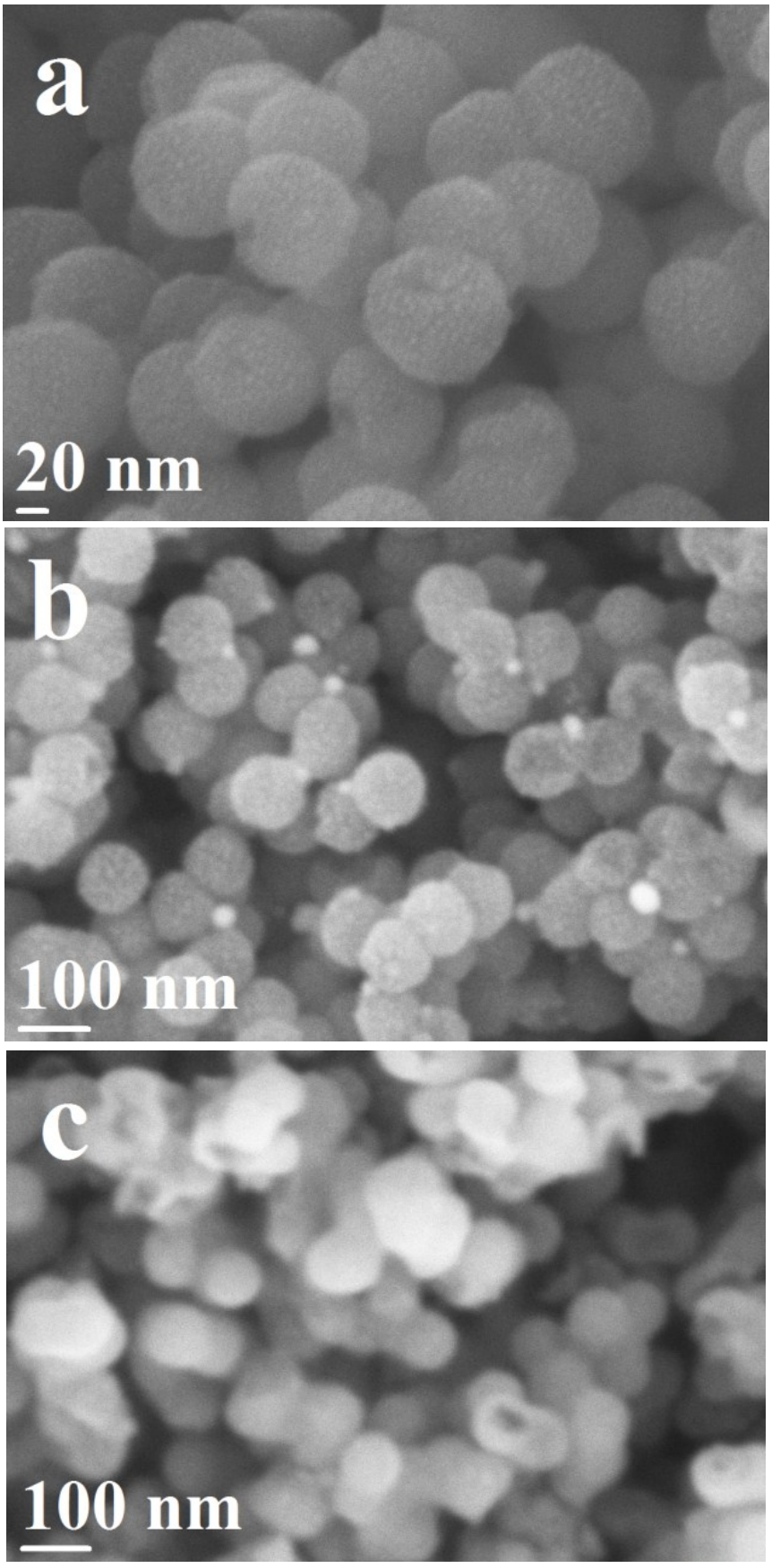


Fig. S3 SEM images of NC, Ni@NC and MoC_x@NC.

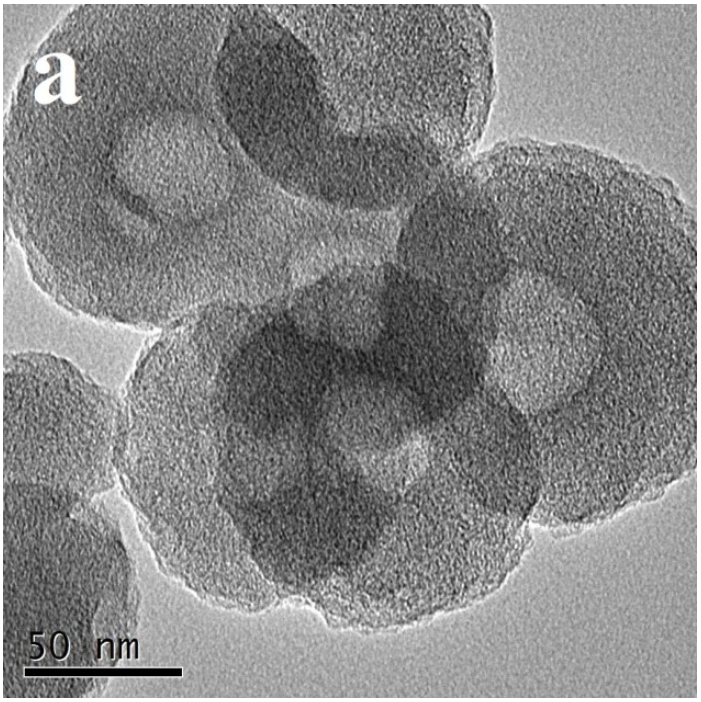


Fig. S4 TEM image of NC.

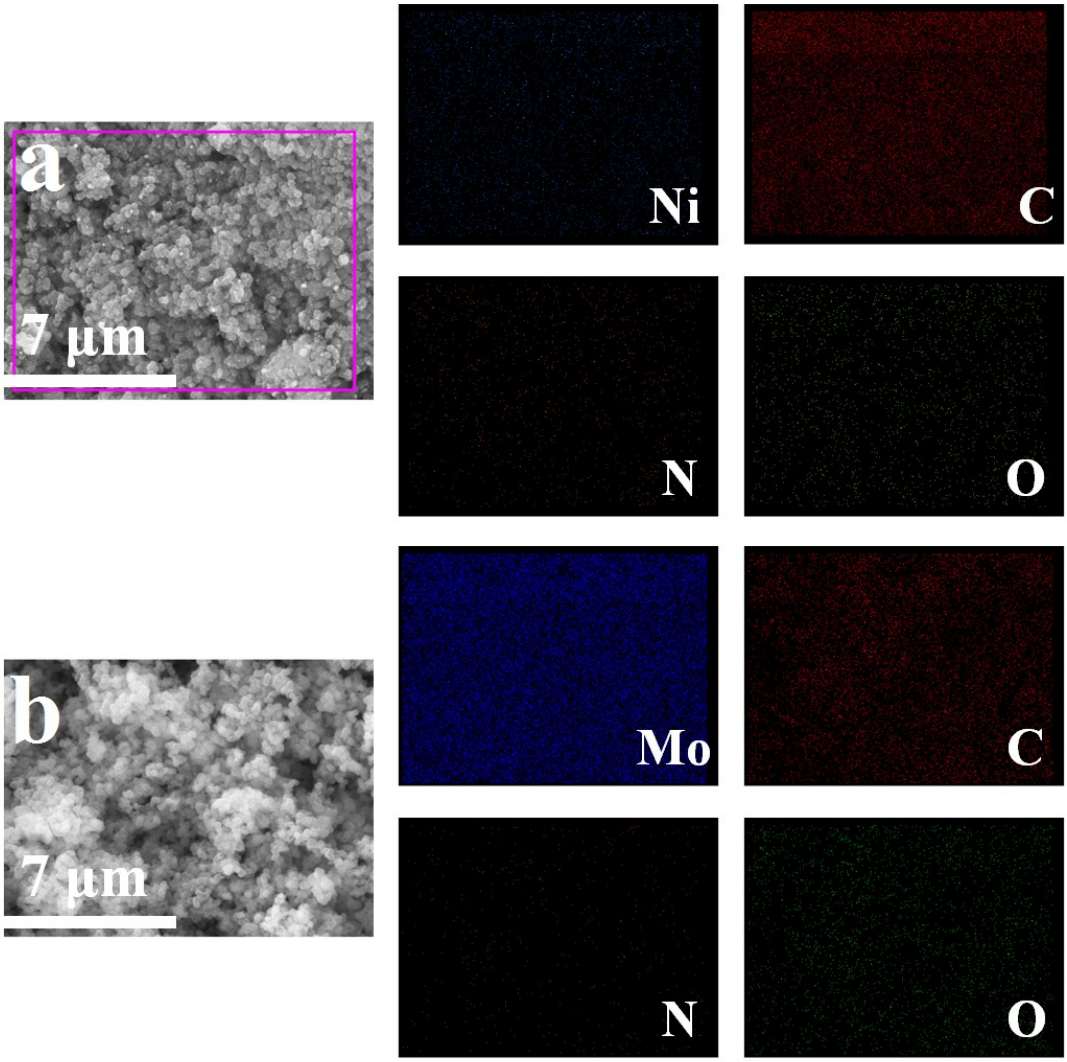
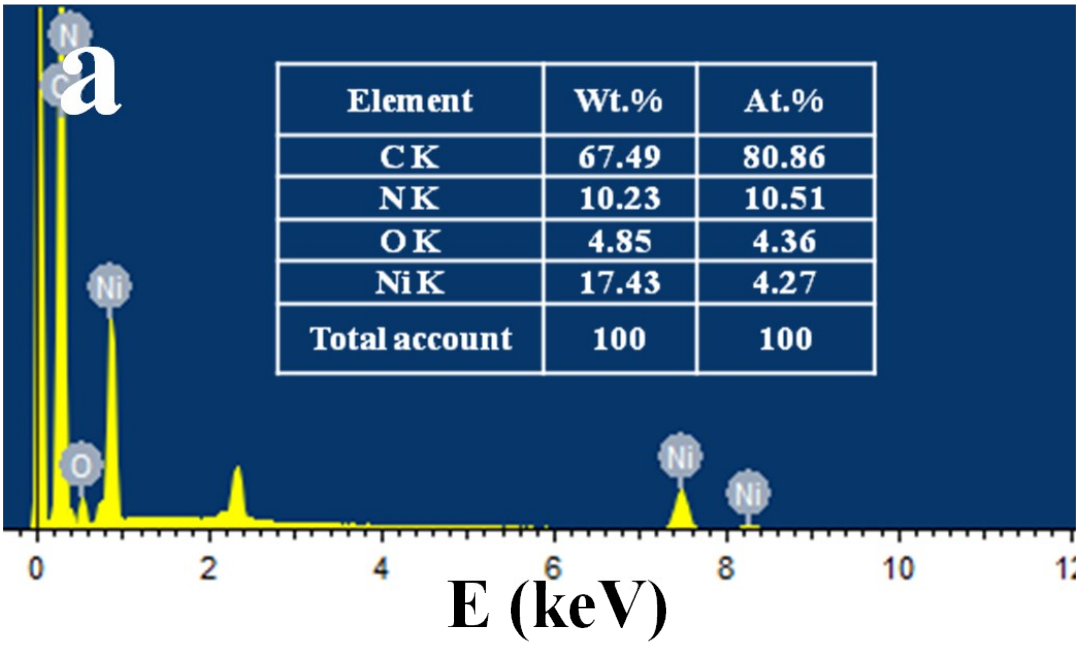
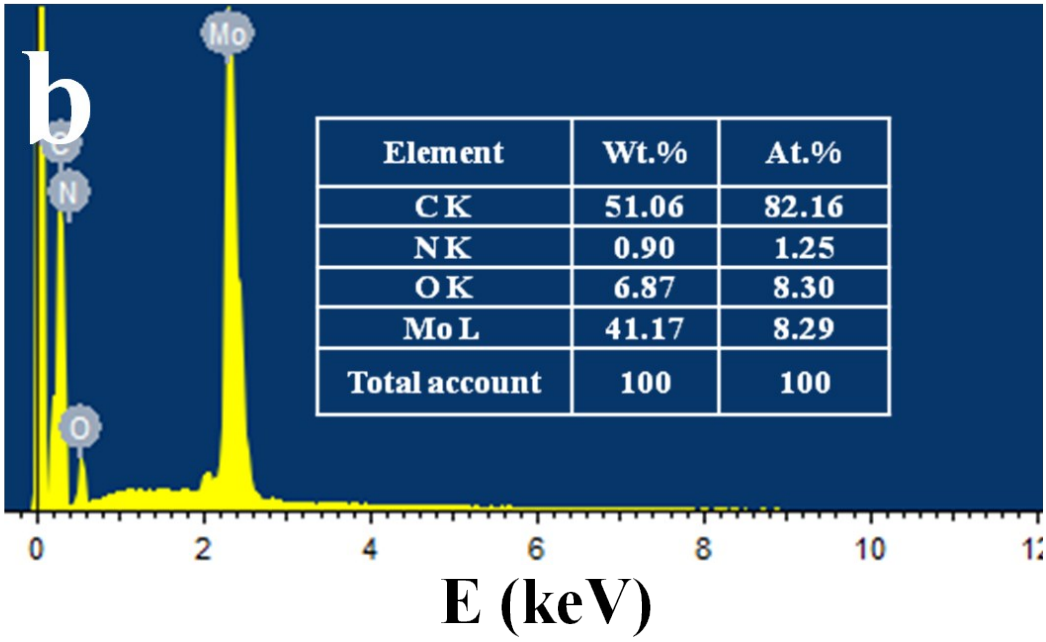


Fig. S5 SEM mapping of (a) Ni@NC and (b) MoC_x@NC.



Element	Wt.%	At.%
C K	67.49	80.86
N K	10.23	10.51
O K	4.85	4.36
Ni K	17.43	4.27
Total account	100	100



Element	Wt.%	At.%
C K	51.06	82.16
N K	0.90	1.25
O K	6.87	8.30
Mo L	41.17	8.29
Total account	100	100

Fig. S6 EDX of (a) Ni@NC and (b) MoC_x@NC.

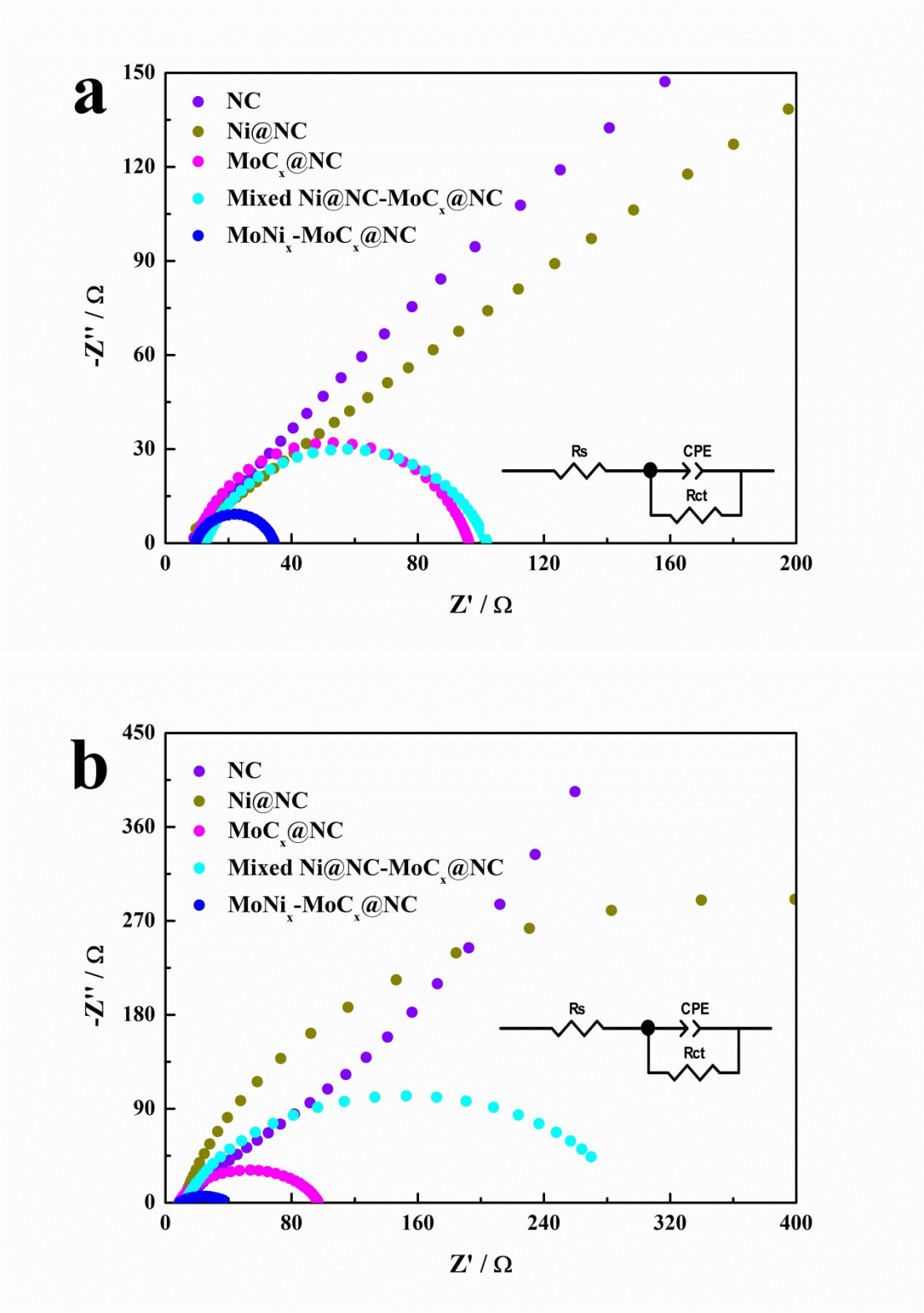


Fig. S7 Nyquist plots of NC, Ni@NC, MoC_x@NC, Mixed Ni@NC-MoC_x@NC and $MoNi_x$-MoC_x@NC in (a) 0.5 M H_2SO_4 and (b) 1 M KOH.

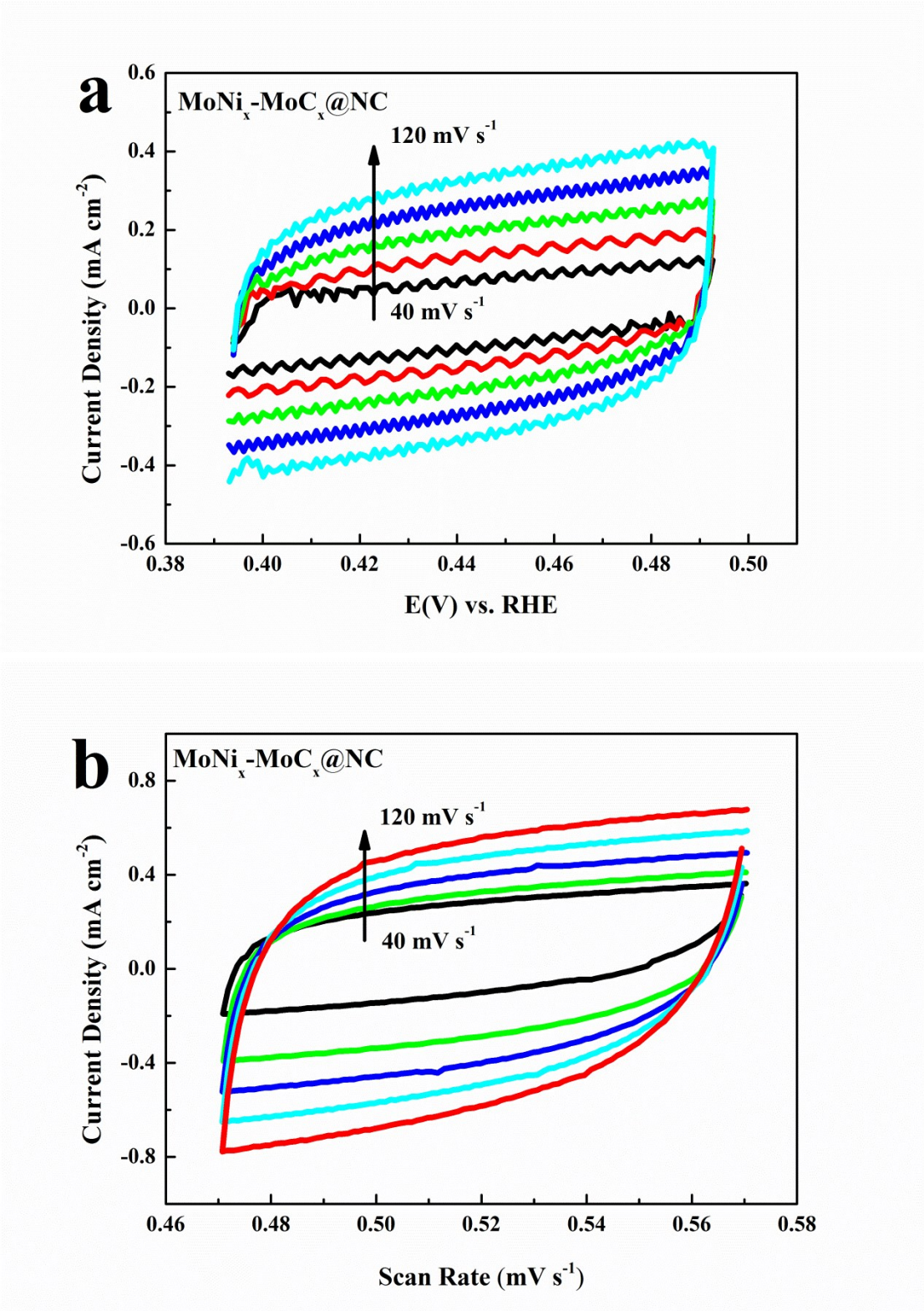


Fig. S8 Double-layer capacitance measurements for determining ECSA of the $MoNi_x$-MoC_x@NC in (a) 0.5 M H_2SO_4 and (b) 1 M KOH.

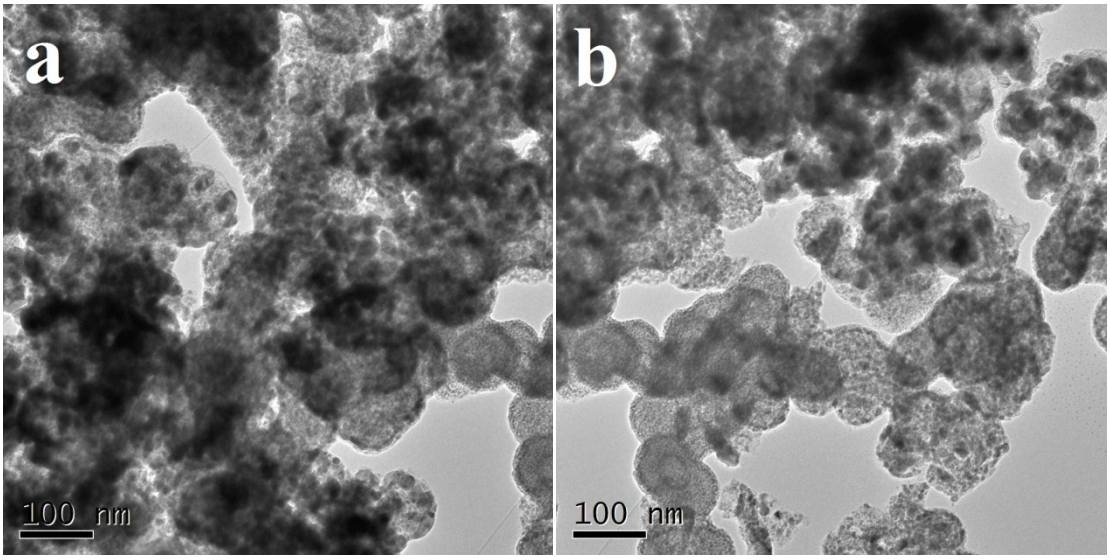


Fig. S9 TEM images of $MoNi_x$-MoC_x@NC after 1000 CV test in (a) 0.5 M H_2SO_4 and (b) 1 M KOH.

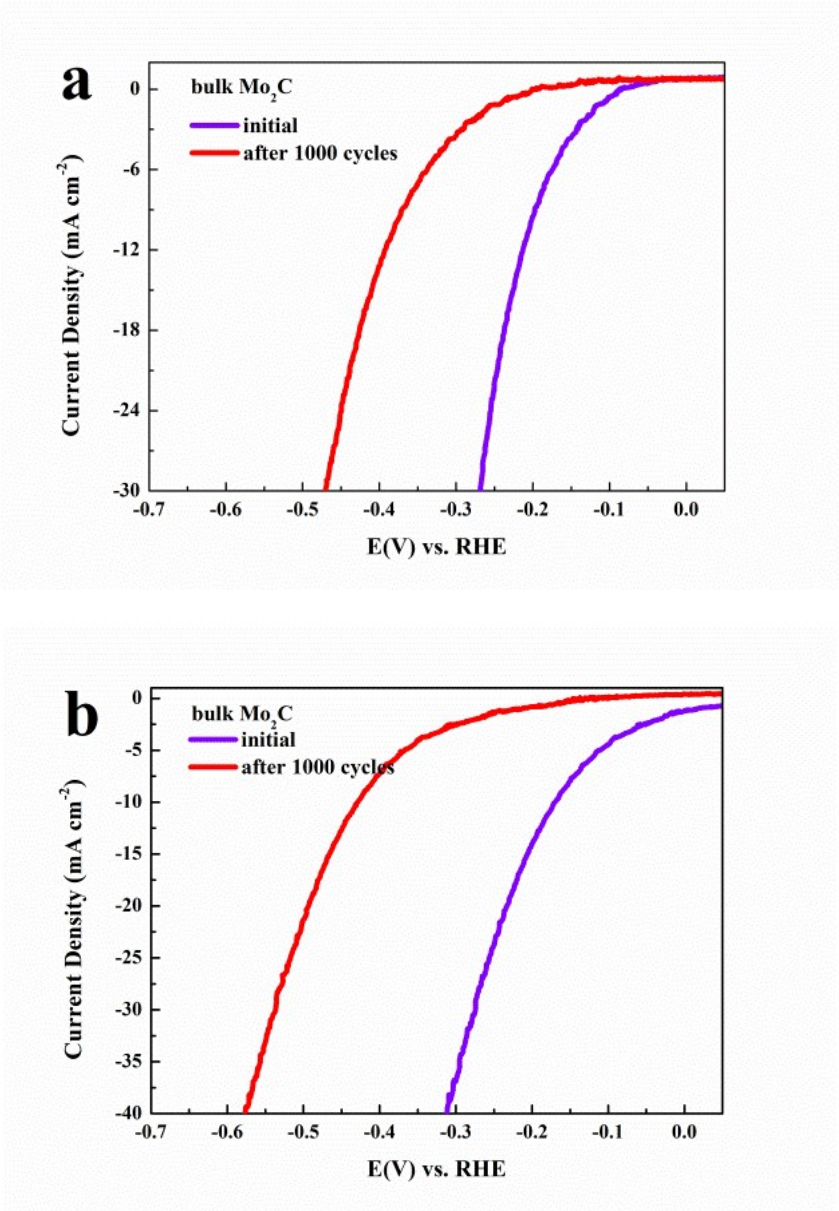


Fig. S10 HER polarization curves of bulk Mo_2C before and after 1000 CV cycles in (a) 0.5 M H_2SO_4 and (b) 1.0 M KOH.

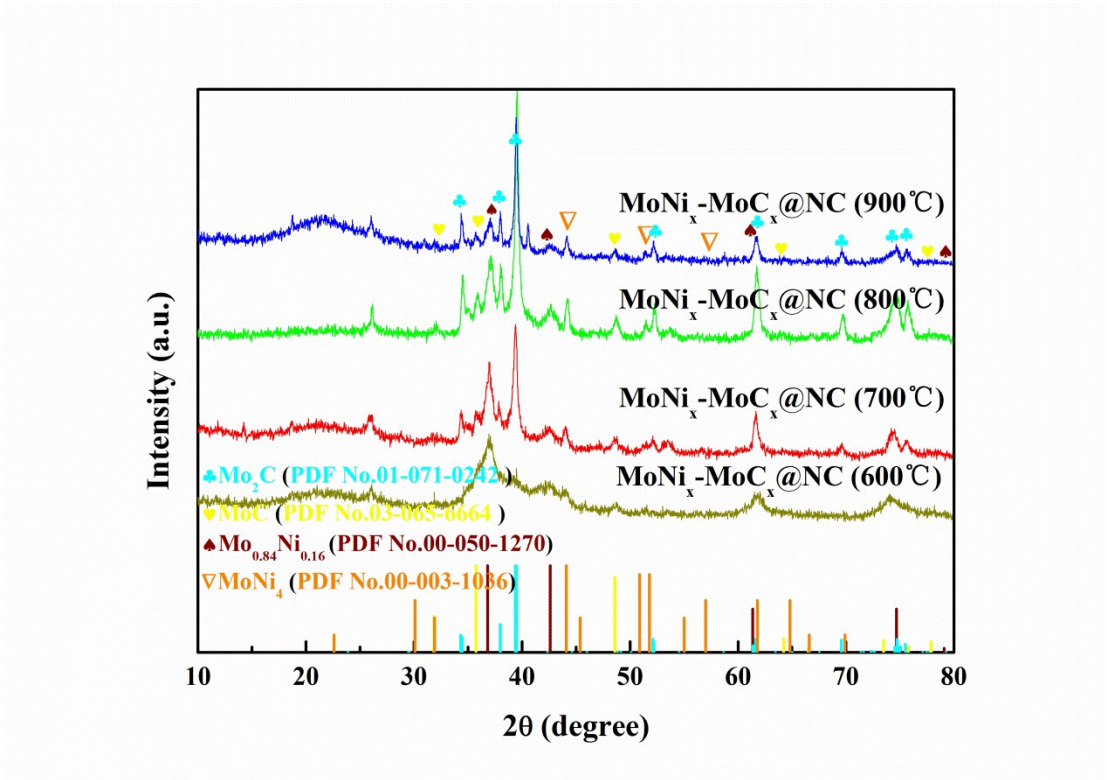


Fig. S11 XRD patterns of $MoNi_x$-MoC_x@NC obtained at 600 °C, 700 °C, 800 °C, and 900 °C.

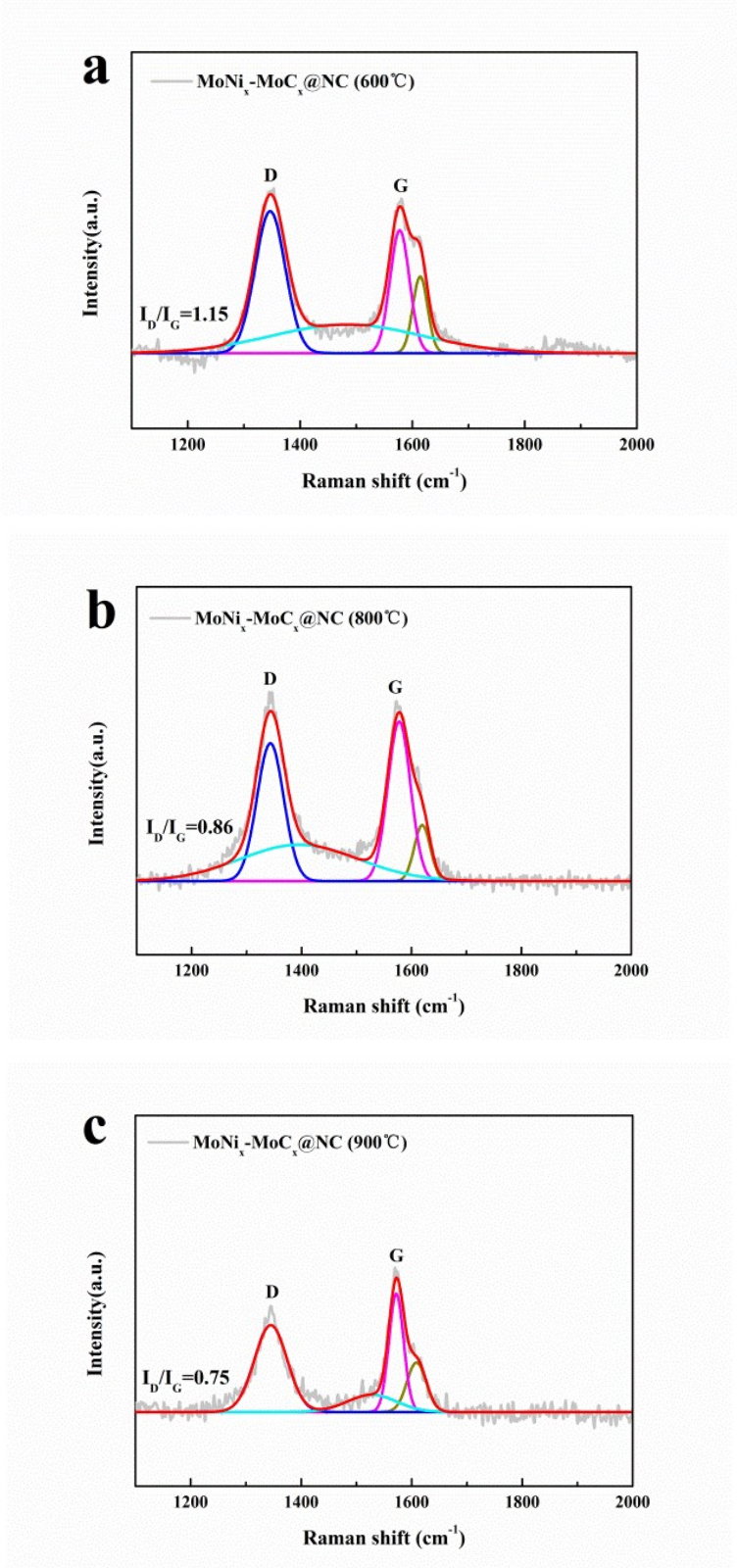
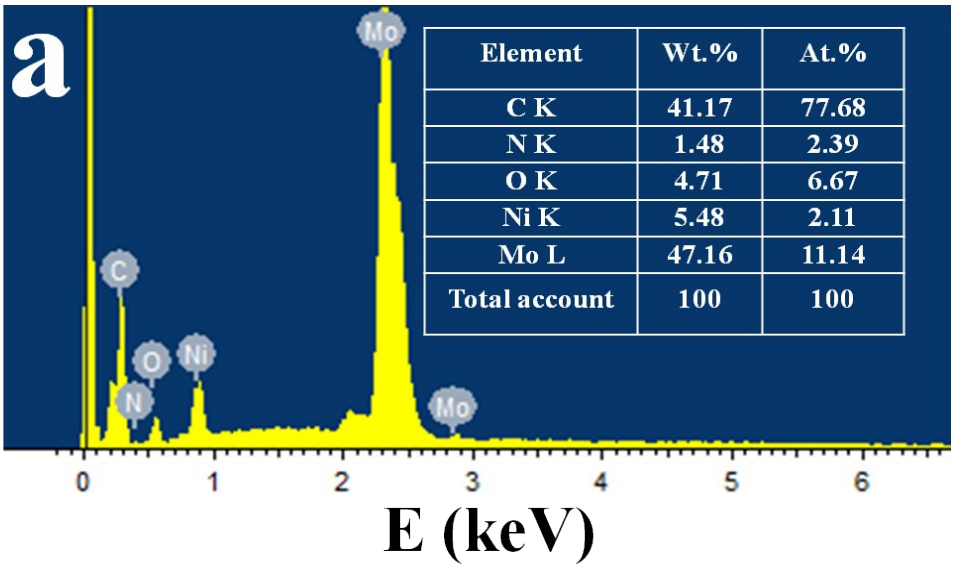
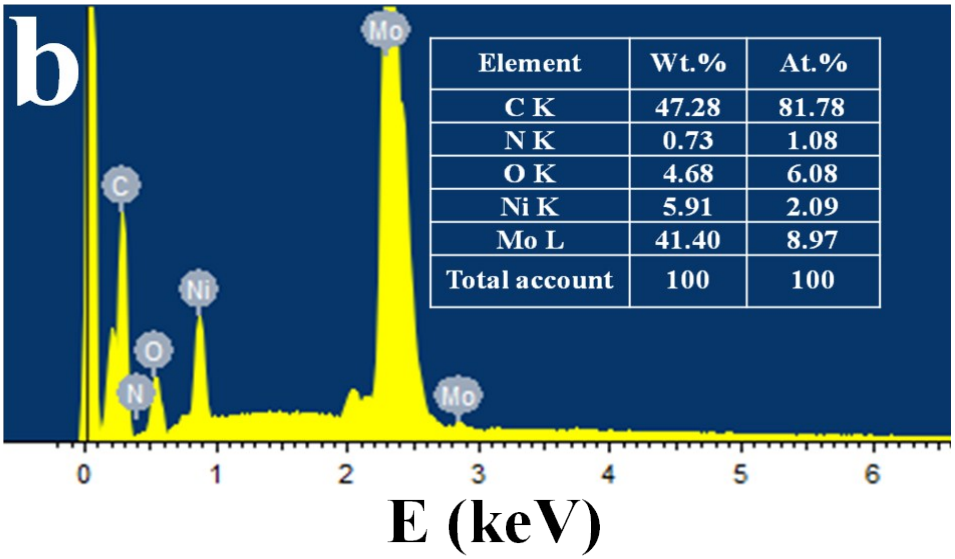


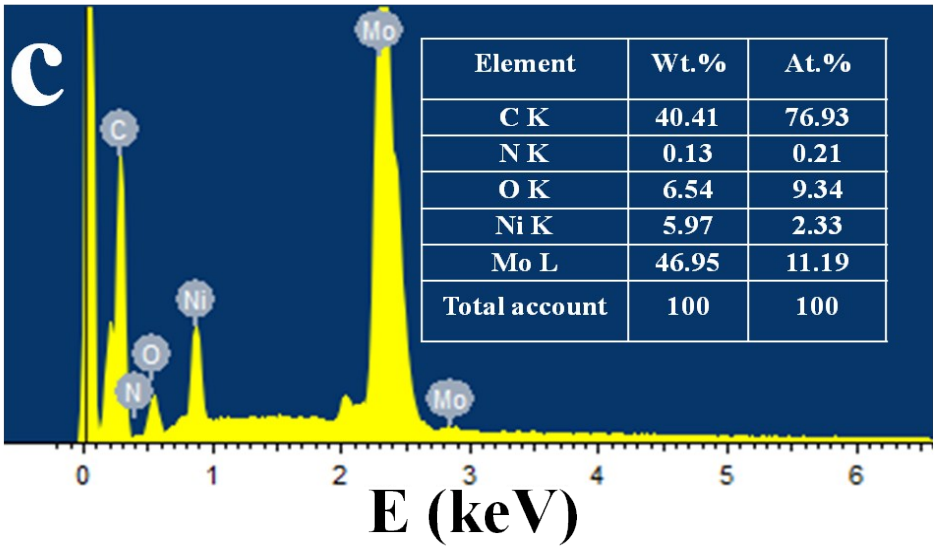
Fig. S12 Raman spectrum of $MoNi_x$-MoC_x@NC obtained at (a) 600 °C, (b) 800 °C, and (c) 900 °C.



Element	Wt.%	At.%
C K	41.17	77.68
N K	1.48	2.39
O K	4.71	6.67
Ni K	5.48	2.11
Mo L	47.16	11.14
Total account	100	100



Element	Wt.%	At.%
C K	47.28	81.78
N K	0.73	1.08
O K	4.68	6.08
Ni K	5.91	2.09
Mo L	41.40	8.97
Total account	100	100



Element	Wt.%	At.%
C K	40.41	76.93
N K	0.13	0.21
O K	6.54	9.34
Ni K	5.97	2.33
Mo L	46.95	11.19
Total account	100	100

Fig. S13 EDX of $MoNi_x$-MoC_x@NC obtained at 600 °C, 800 °C, and 900 °C.

Table S1. Comparison of the HER activity of $MoNi_x$-MoC_x@NC with the other reported TMPs HER electrocatalysts in acidic and alkaline media.

Electrocatalyst	j ($mA\ cm^{-2}$)	η (mV)	b ($mV\ dec^{-1}$)	Electrolyte solution	Ref.
$MoNi_x$-MoC_x@NC	10	172	73	0.5 M H_2SO_4	This work
	10	168	71	1.0 M KOH	This work
Mo_2C-NCNTs	10	147	71	0.5 M H_2SO_4	1
	10	257	/	1.0 M KOH	
Mo_2C nanoparticles	10	198	56	0.5 M H_2SO_4	2
	10	176	58	1.0 M KOH	
Mo_2C	10	210	/	0.5 M H_2SO_4	3
	10	190	/	1.0 M KOH	
Mo_2C nanowires	10	200	55.8	0.5 M H_2SO_4	4
Mo_2C/GCSs	10	210	62.6	0.5 M H_2SO_4	5
MoS_2/Mo_2C-NCNTs	25	210	69	0.5 M H_2SO_4	6
$Mo_{0.06}W_{1.94}C$/CB	10	220	/	0.5 M H_2SO_4	7
Mo_2C-carbon	5	260	/	0.5 M H_2SO_4	8
Co_4Mo_2@NC	10	218	73.5	1.0 M KOH	9
Mo_2C-GNR	10	217	64	1.0 M NaOH	10
$MoC_{0.654}$@CNS	10	220	/	1.0 M KOH	11
Mo_2C-PC	10	238	140	1.0 M KOH	12
Ni/Mo_2C-PC	10	179	59	1.0 M KOH	12

Table S2 Elemental values of fitted equivalent circuit for NC, Ni@NC, MoC_x@NC, Mixed Ni@NC-MoC_x@NC and $MoNi_x$-MoC_x@NC in acidic and alkaline media.

Samples	0.5 M H_2SO_4		1.0 M KOH	
	R_s/Ω	R_{ct}/Ω	R_s/Ω	R_{ct}/Ω
NC	11.66	28514	14.16	10661
Ni@NC	12.04	2612	11.01	716.4
MoC_x@NC	9.44	86.89	9.28	99.23
Mixed Ni@NC-MoC_x@NC	13.27	91.37	12.79	285.2
$MoNi_x$-MoC_x@NC	12.46	47.38	9.45	19.23

Table S3. Elemental values of fitted equivalent circuit for $MoNi_x$-MoC_x@NC obtained at 600 °C, 700 °C, 800 °C and 900 °C in acidic and alkaline media.

Samples	0.5 M H_2SO_4		1.0 M KOH	
	R_s/Ω	R_{ct}/Ω	R_s/Ω	R_{ct}/Ω
600 °C	9.25	92.29	13.78	40.04
700 °C	12.46	47.38	9.45	19.23
800 °C	11.57	82.53	9.96	36.23
900 °C	12.12	85.33	9.43	30.48

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