Supporting information

Free-Standing Multi-Hierarchical MoC-based Catalyst for pH-Universal Hydrogen Evolution Reaction at Ultra-High Current Density

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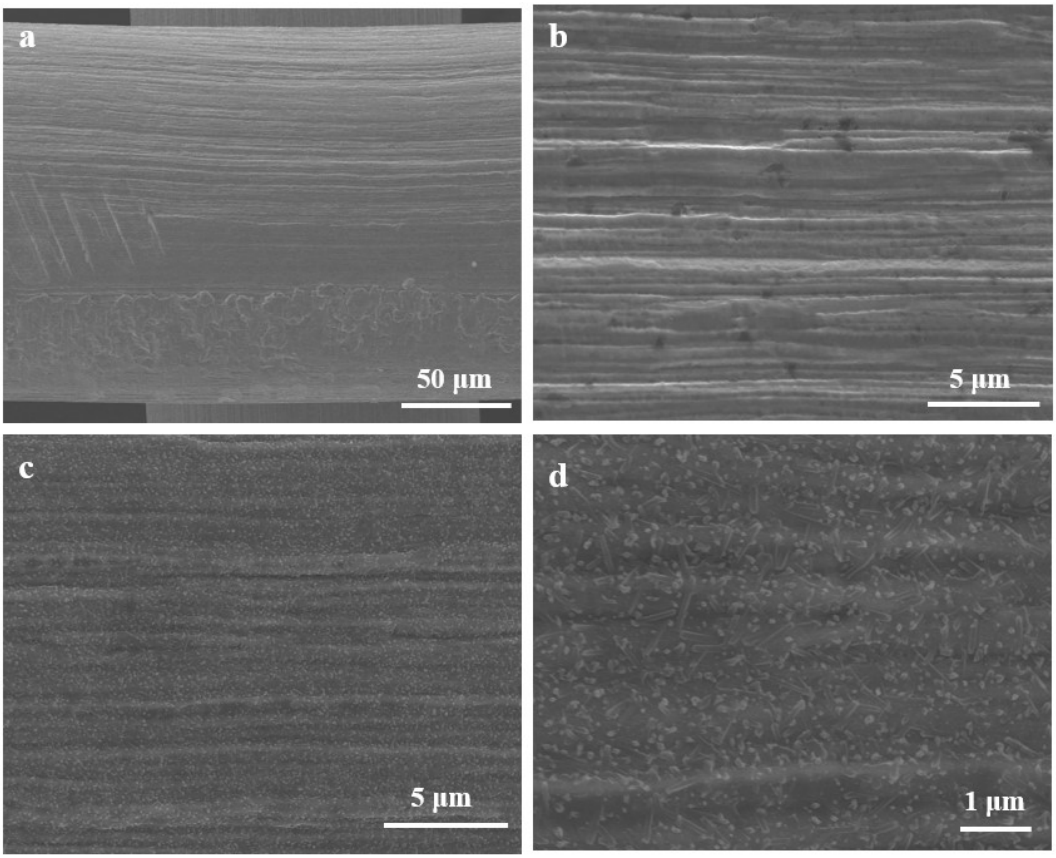

Fig. S1. SEM images of (a-b) Mo mesh and (c-d) MoOx-Mo mesh.

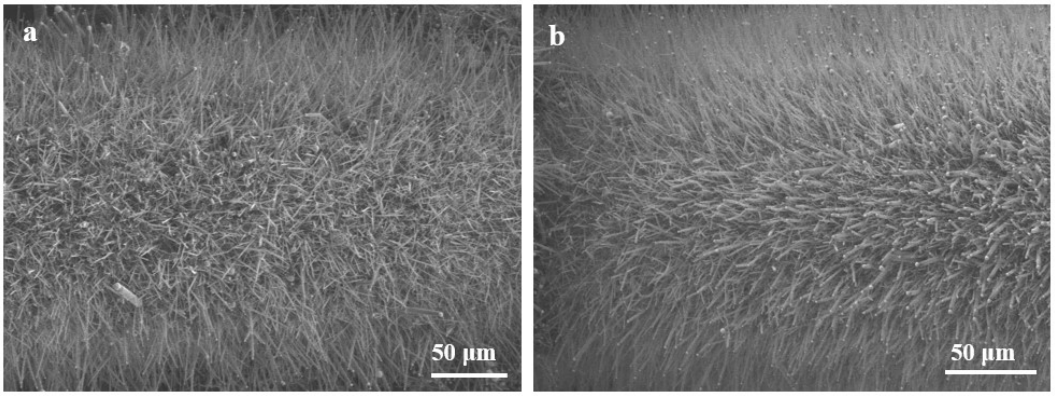

Fig. S2. SEM images of (a) MoC/Ni@NCNTs-MoC@C and (b) P-MoC/Ni@NCNTs-MoC@C.

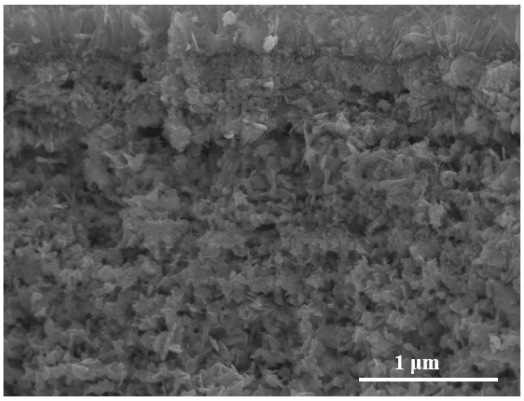

Fig. S3. SEM images of MoC@C shell.

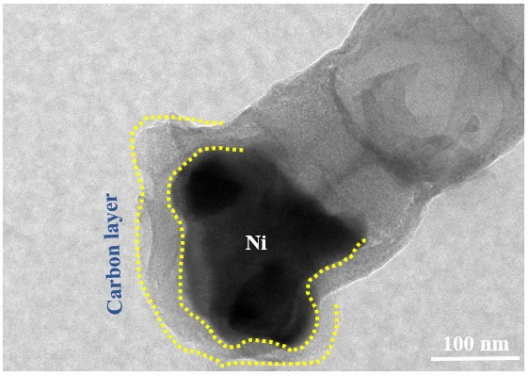

Fig. S4. TEM images of P-MoC/Ni@NCNTs-MoC@C.

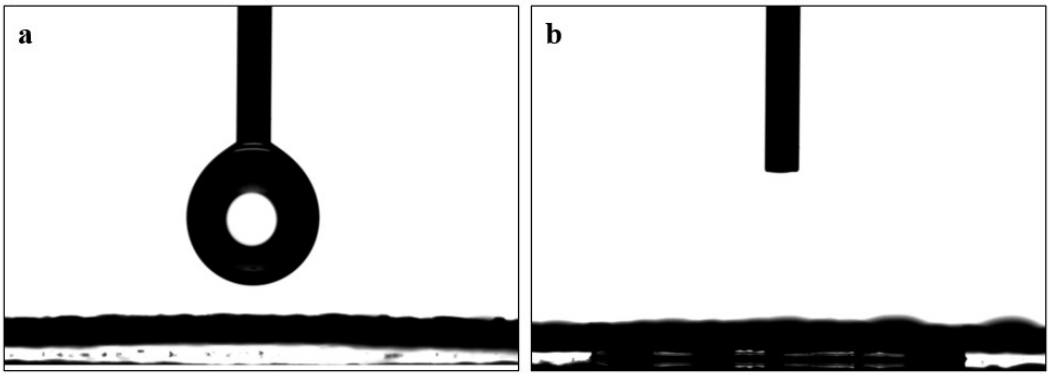

Fig. S5. The connect angles of P-MoC/Ni@NCNTs-MoC@C.

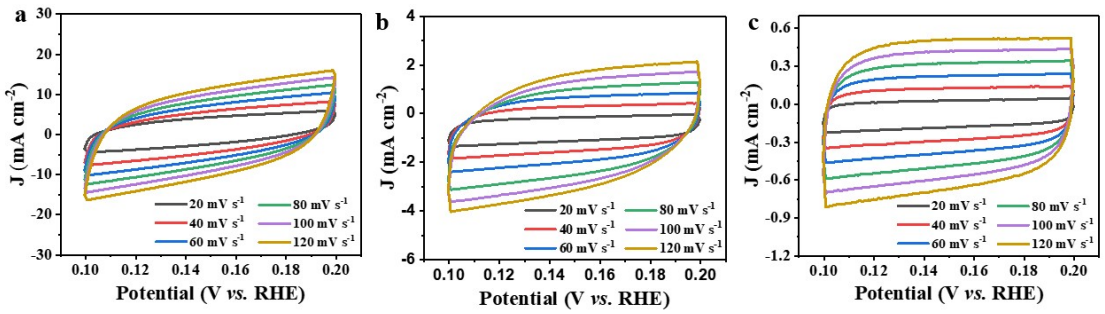

Fig. S6. CVs of (a) MoC/Ni@NCNTs-MoC@C, (b) MoC-Mo mesh, and (c)Mo mesh in 0.5 M H_2SO_4.

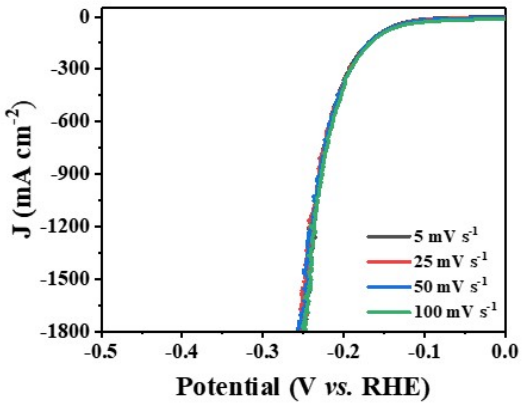

Fig. S7. LSV curves of P-MoC/Ni@NCNTs-MoC@C at different scan Rates in 0.5 M H_2SO_4.

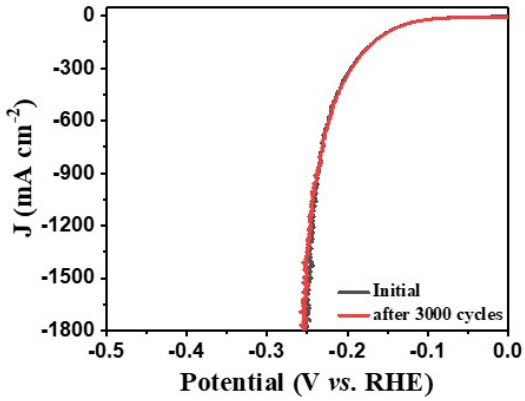

Fig. S8. LSV curves of P-MoC/Ni@NCNTs-MoC@C initial and after 3000 cycles test in 0.5 M H_2SO_4.

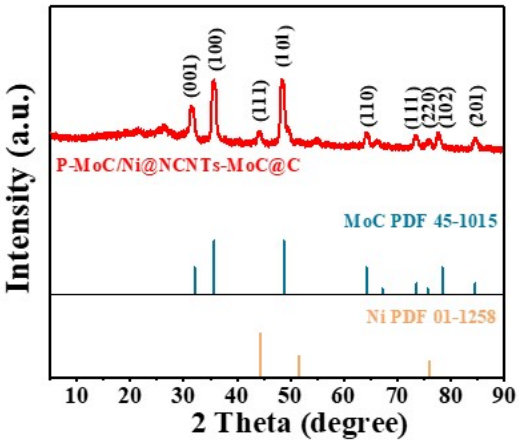

Fig. S9. XRD pattern of P-MoC/Ni@NCNTs-MoC@C after i-t test in 0.5 M H_2SO_4.

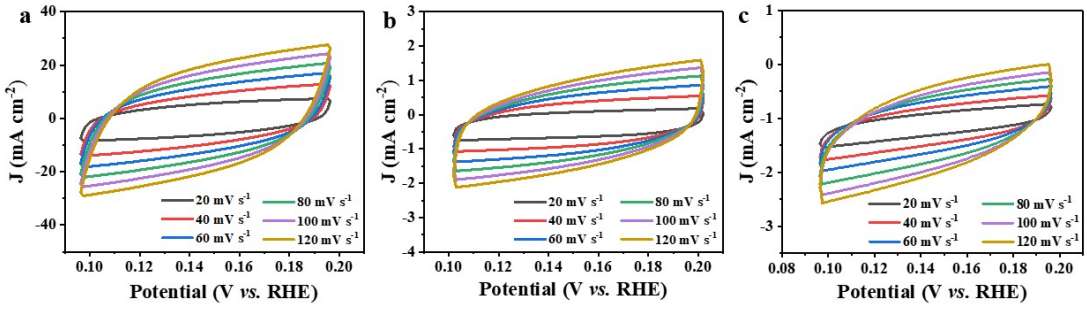

Fig. S10. CVs of (a) MoC/Ni@NCNTs-MoC@C, (b) MoC-Mo mesh, and (c)Mo mesh in 1.0 M KOH.

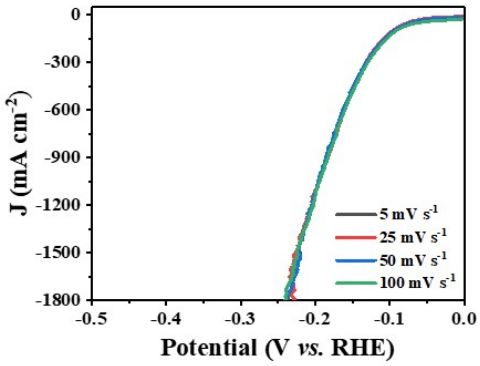

Fig. S11. LSV curves of P-MoC/Ni@NCNTs-MoC@C at different scan rates in 1.0 M KOH.

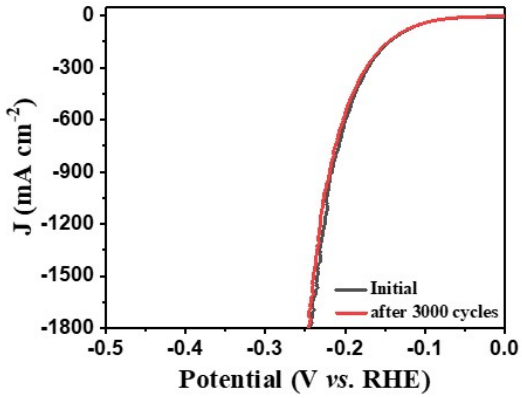

Fig. S12. LSV curves of P-MoC/Ni@NCNTs-MoC@C initial and after 3000 cycles test in 1.0 M KOH.

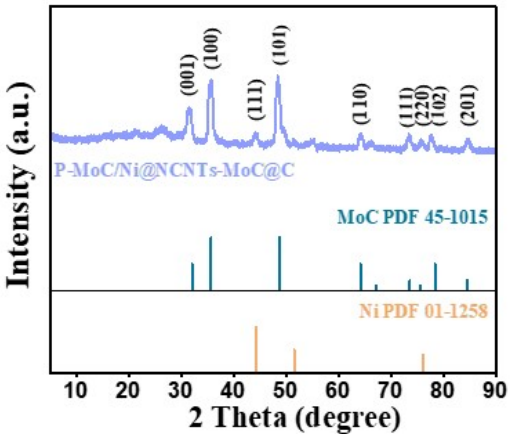

Fig. S13. XRD pattern of P-MoC/Ni@NCNTs-MoC@C after i-t test in 1.0 M KOH.

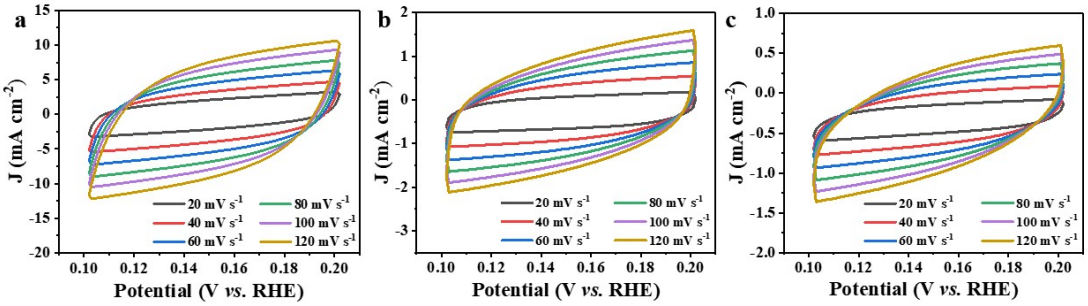

Fig. S14. CVs of (a) MoC/Ni@NCNTs-MoC@C, (b) MoC-Mo mesh, and (c)Mo mesh in 1.0 M PBS.

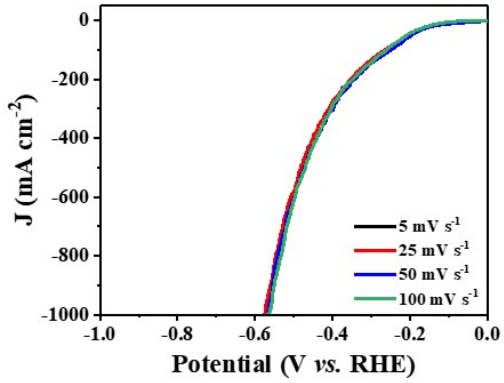

Fig. S15. LSV curves of P-MoC/Ni@NCNTs-MoC@C at different scan Rates in 1.0 M PBS.

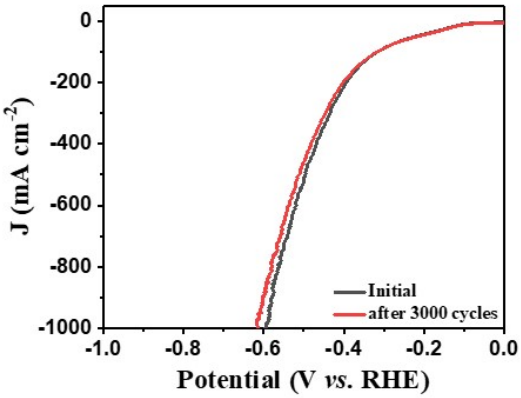

Fig. S16. LSV curves of P-MoC/Ni@NCNTs-MoC@C initial and after 3000 cycles test in 1.0 M PBS.

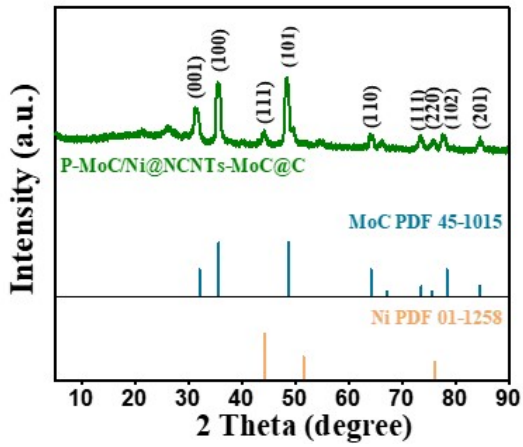

Fig. S17. XRD pattern of P-MoC/Ni@NCNTs-MoC@C after i-t test in 1.0 M PBS.

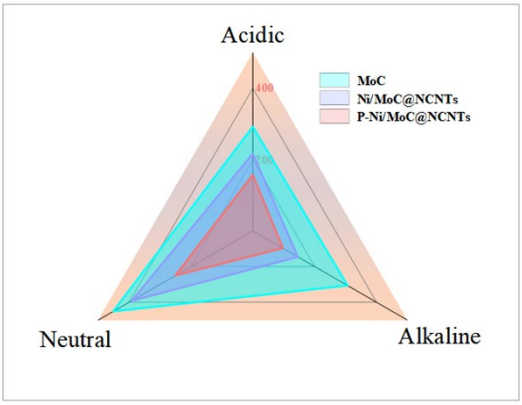

Fig. S18. Comparison of as-prepared samples HER performance in the full pH range.

Table S1. Summary of representative non-noble metal-based composite catalysts in 0.5 M H_2SO_4.

Type of molybdenum carbide electrocatalyst	η_{10} (mV)	η_{100} (mV)	Tafel slope (mV dec^{-1})
This work	91.2	157.3	67.4
CM/Mo_xC@NC	149	248.5	86
Ni-Mo_2C-0.67	165	214	54.1
$Ni_3Mo_3C/Mo_2C/Ti_3C_2T_x$/NC	99	253	58
MoS_2 /Mo_2C	207	308.9	60
Ni-MoS_2/NCNTs-3	158	225.7	69.3
WS_2/Ni_5P_4-Ni_2P	94	211	74
Ni_2P-Co_2P	172	296	67
Co-NCNT/CC	78	155	74
Mo_2C@SNC	146	237	83
PS/MoS_2 1:2	154	250	71
Ni_2P/NPPC	159	261	74
WC/WO_{3-x}-900	34.8	212.6	59.3

Table S2. Summary of representative non-noble metal-based composite catalysts in 1.0 M KOH.

Type of molybdenum carbide electrocatalyst	η_{10} (mV)	η_{100} (mV)	Tafel slope (mV dec^{-1})
This work	12.1	97.2	71.1
MoC@NC	98	465.2	55
Ni-Mo_2C-0.67	151	194	50.9
H-Mo_2C/NG	63	163	48
MS-Mo_2C@NCNS	98	199.1	99
MoOPC@NC-200	69	154	60.4
CoO/Mo_2C	107	285	80
Ni-MoC@NCNT/CC	70	197.6	68.9
$Ni_3Mo_3C/Mo_2C/Ti_3C_2T_x$/NC	98	242.8	69

CoN/MoC/NMCNFs/CC	92.5	266.2	107.9
Ni_3Mo_3N/Mo_2C@C	43	273.2	110
N:Mo_2C@NC	150	294	97
MoS_2/Ni_3S_2-NF	187	274	90
NF-Ni_3S_2-NF	135	225	75

Table S3. Summary of representative non-noble metal-based composite catalysts in 1.0 M PBS.

Type of molybdenum carbide electrocatalyst	**η_{10} (mV)**	**η (mV)**	**Tafel slope (mv dec^{-1})**
This work	85.4	250.5(100)	130.3
SiO_2/PPy NTs-CFs	183	274(40)	100.2
Co–$Mo_{0.4}$-S	213	379(40)	94
WP NAs/CC	200	402(80)	125
Mo_5N_6	249		330
Ni_2P@NPCNFs	185.3	377.2(100)	230.3
3D MoS_2/N-GAs	261	402.4(40)	230
Co_9S_8@C	290		280
$FePSe_3$/NC	140.1	457(100)	167
Co-Fe-P	138	329(100)	138
$Mo_2N_xC_{1-x}$/rGO	332		137