Electronic Supplementary Information for

Upcycling red brick into a superior monolithic hydrogen evolution electrocatalyst

Zhengguo Zhang,[*a,b] Yiming Li,[c] Jiao Zhang,[c] Yaoyao Zhao,[a,b] Mengjuan Xu,[a,b] Fang Wang,[a,b] and Shixiong Min[*a,b,c]

[a] *School of Chemistry and Chemical Engineering, North Minzu University, Yinchuan, 750021, P. R. China.*

[b] *Ningxia Key Laboratory of Solar Chemical Conversion Technology, North Minzu University, Yinchuan 750021, P. R. China.*

[c] *Analysis and Testing Center of Ningxia Hui Autonomous Region, North Minzu University, Yinchuan, 750021, P. R. China.*

**Corresponding authors: zhangzhengguo1119@126.com; sxmin@nun.edu.cn.*

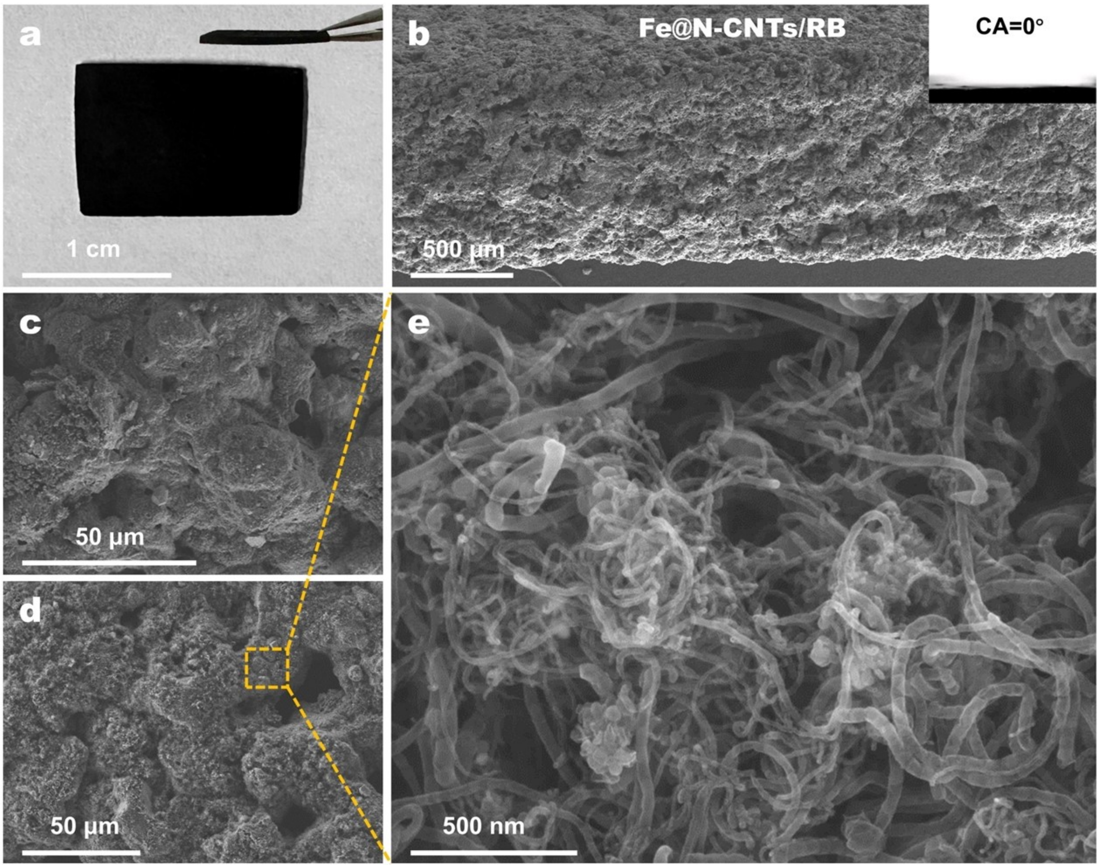

Fig. S1 (a) Photographs of Fe@N-CNTs/RB electrode. (b) SEM image showing the overall view of Fe@N-CNTs/RB electrode, where the inset shows the electrolyte contact angel on Fe@N-CNTs/RB electrode. (c) side-view, (d) Top-view, and (e) high-resolution SEM images of Fe@N-CNTs/RB.

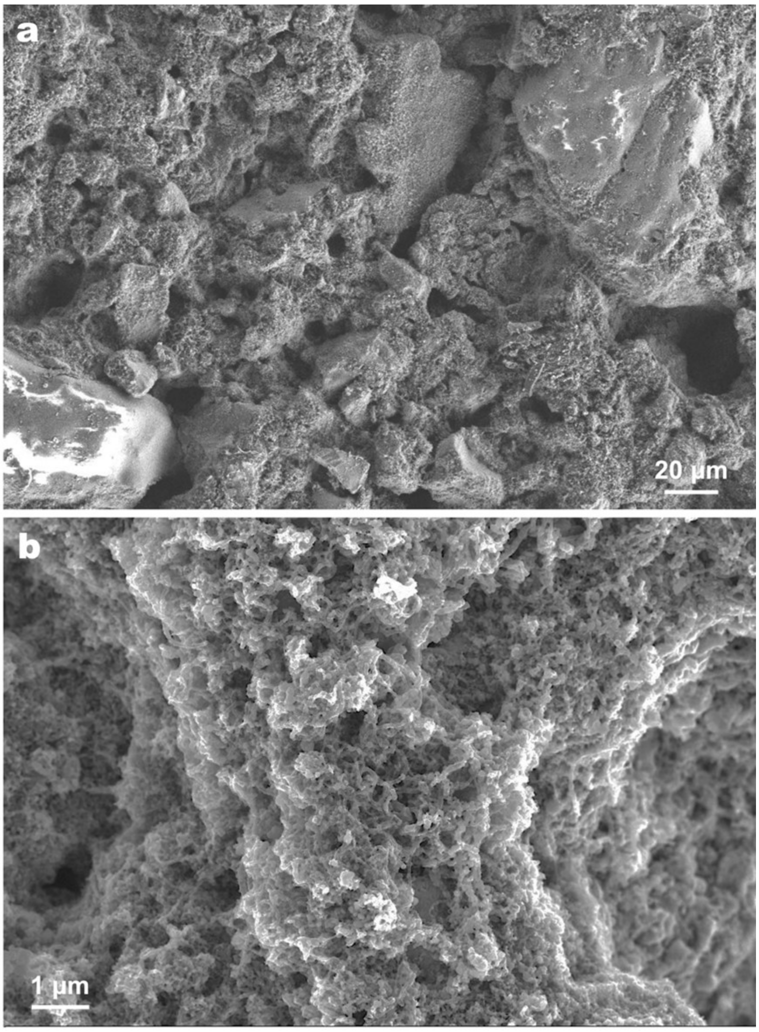

Fig. S2 Medium-magnification SEM images of Fe@N, P-CNTs/RB electrode.

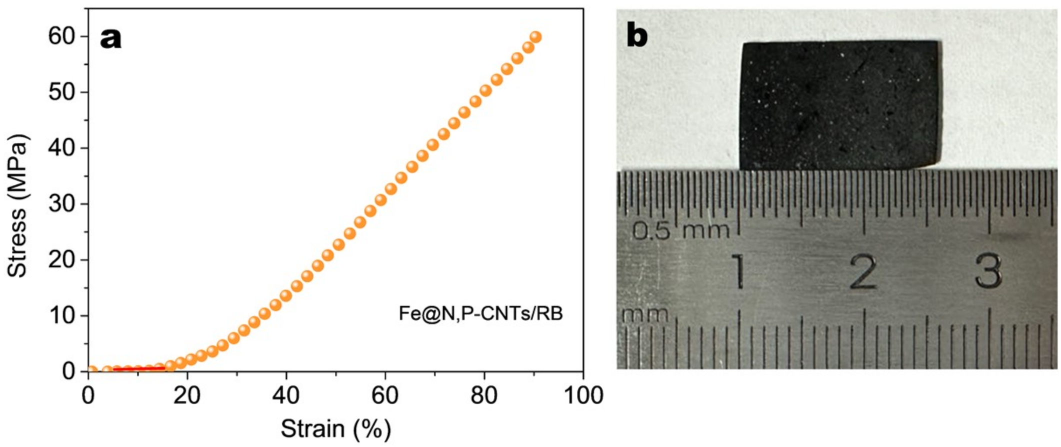

Fig. S3 (a) Compression-strain curve and (b) the photograph of Fe@N, P-CNTs/RB electrode after mechanical strength test.

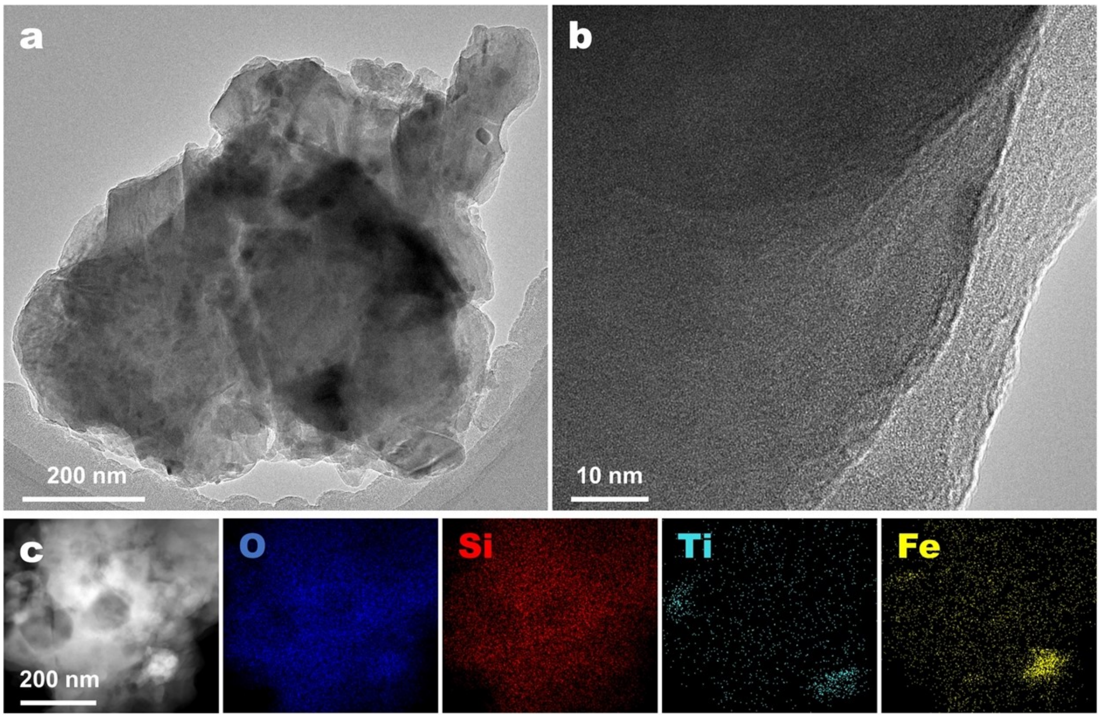

Fig. S4 (a) TEM and (b) HRTEM images of RB. (c) HAADF-STEM image of RB and the corresponding EDX mappings.

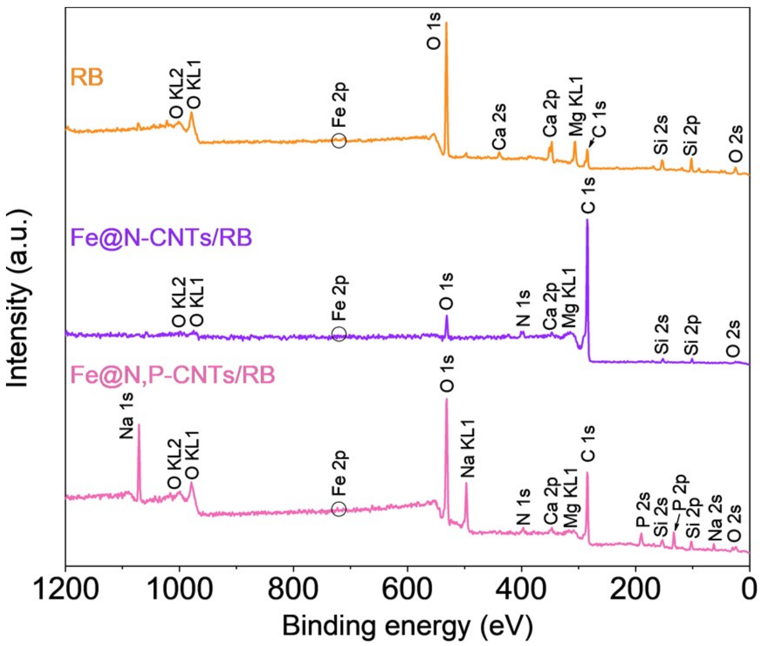

Fig. S5 XPS survey spectra of RB, Fe@N-CNTs/RB, and Fe@N, P-CNTs/RB electrodes.

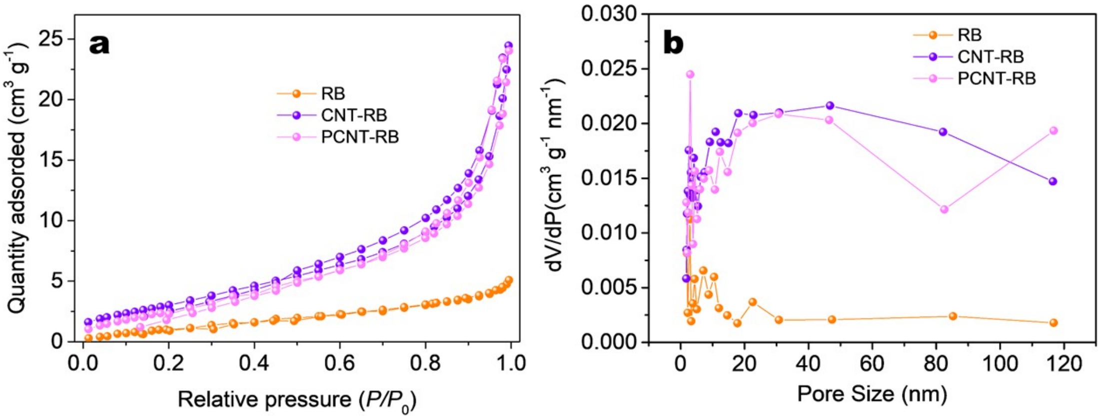

Fig. S6 (a) N_2 adsorption-desorption isotherms and (b) pore size distributions of RB, Fe@N-CNTs/RB, and Fe@N, P-CNTs/RB electrodes.

Table S1 Comparison of electrocatalytic HER performance of Fe@N, P-CNTs/RB with previously reported Fe-based electrocatalysts in acidic solution.

Catalyst	η_{10} (mV)	η_{100} (mV)	Reference
Ru-Fe-O_x/CC	32	90	[1]
Fe-(NiS_2/MoS_2)/CNT	98	194	[2]
Fe_7S_8/C	90.6	-	[3]
Mo-Fe(1/1)-Se-CP	86.9	158.5	[4]
CoFe@NDC@MoS_2	64	-	[5]
CoS_2/FeS_2/CN	76.5	218.5	[6]
FeP/Fe@NC	49	130	[7]
Pt_3Fe/NMCS-A	13	-	[8]
Ir-SA@Fe@NCNT	26	-	[9]
Fe-Ni_3S_2@$FeNi_3$-8	48	-	[10]
$Fe_{0.5}Co_{0.5}P$/CC	37	-	[11]
FeP/C-450	51.1	100.5	[12]
3% $Ni_{0.5}Fe_{0.5}Se_2$/MWCNT	200	-	[13]
Fe/P/$C_{0.5}$-800	256	-	[14]
CW-CNT@N-C-NiFe	179	275	[15]
$FePSe_3$/NC	70	150	[16]
FeP NPs@NPC	130	234	[17]
FeP/CC	34	75	[18]
Fe&FeP@gl-C-15	92	-	[19]
FeP/CC	-	108	[20]
FeP-GS	123	-	[21]
Fe@N, P-CNTs/RB	223.6	285.6	**This work**

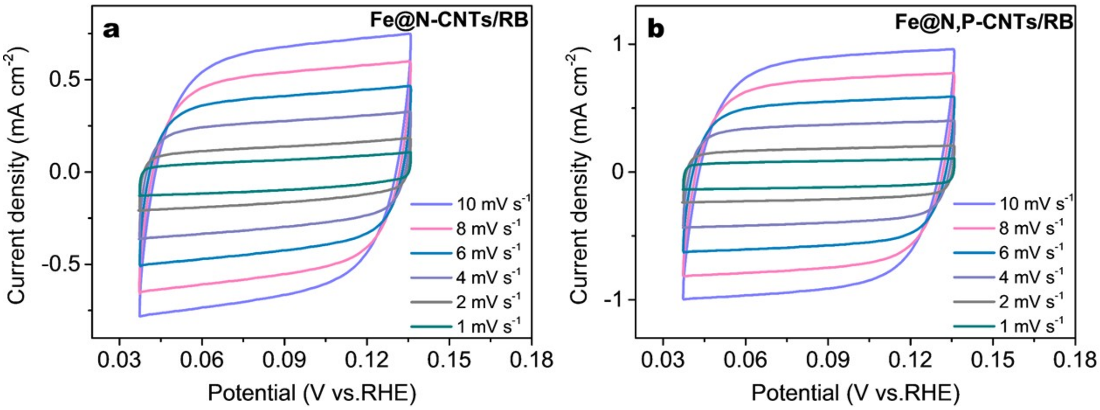

Fig. S7 CV curves of (a) Fe@N-CNTs/RB and (b) Fe@N, P-CNTs/RB monolithic electrodes at different scan rates in 0.5 M H_2SO_4 solution.

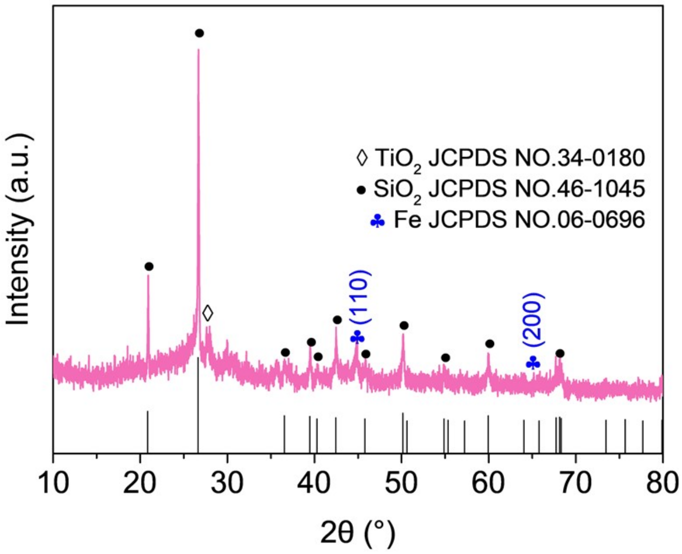

Fig. S8 XRD pattern of the Fe@N, P-CNTs/RB monolithic electrode after long-term HER stability test.

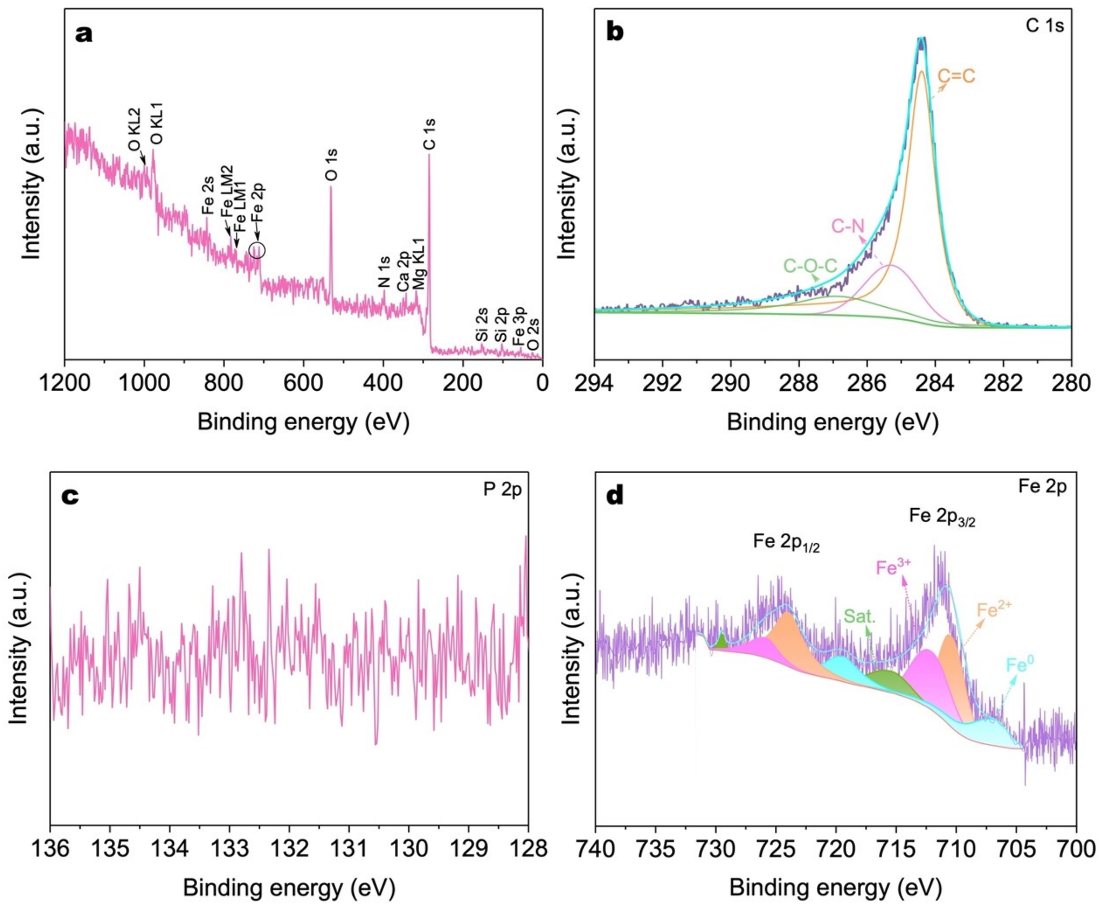

Fig. S9 (a) XPS survey, (b) C 1s, (c) P 2p, and (d) Fe 2p spectra of Fe@N, P-CNTs/RB monolithic electrode after long-term HER stability test.

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