Supporting Information for

Co-doped Ni_3S_2@CNTs Array Anchored on Graphite Foam with Hierarchical Conductive Network for High-Performance Supercapacitor and Hydrogen Evolution Electrode

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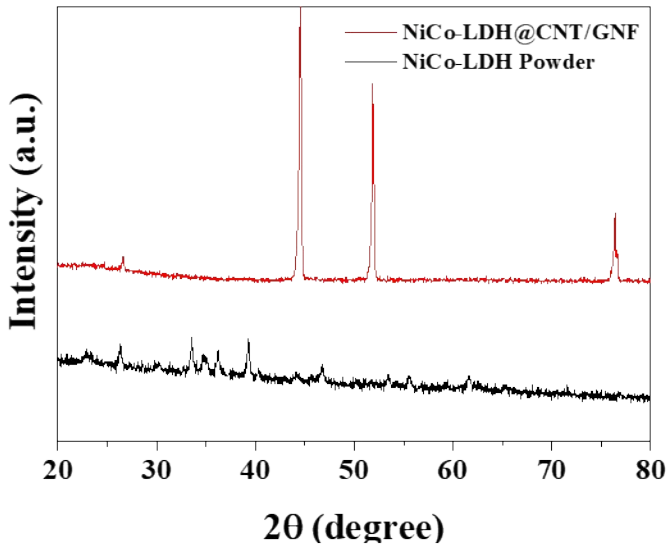

Figure S1 XRD patterns of NiCo-LDH@CNTs/GNF and the NiCo-LDH powder, respectively.

As shown in the figure, the peaks of collected NiCo-LDH powder show obvious specific peaks compared with that of the NiCo-LDH@CNTs/GNF, which may be due to the relatively low loading mass of the NiCo-LDH on the CNTs/GNF struts. Moreover, the peaks located at 22.7°, 33.4°, 34.7° can be correlated with (003), (021) and (101) planes of nickel hydroxide (JCPDS 38-0715), while at 36.3°, 55.9°, 65.2° corresponding to (100), (104) and (110) planes of cobalt hydroxide (JCPDS 26-1107). All of the above results verify the formation of NiCo-LDH composites.

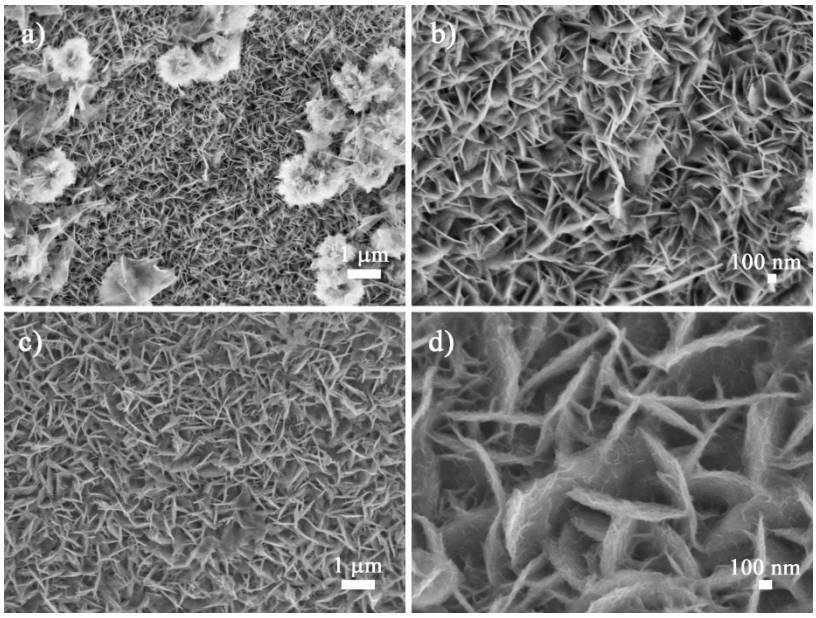

Figure S2. SEM images of a), b) the NiCo-LDH /GNF composites and c), d) Co-Ni_3S_2/GNF composites.

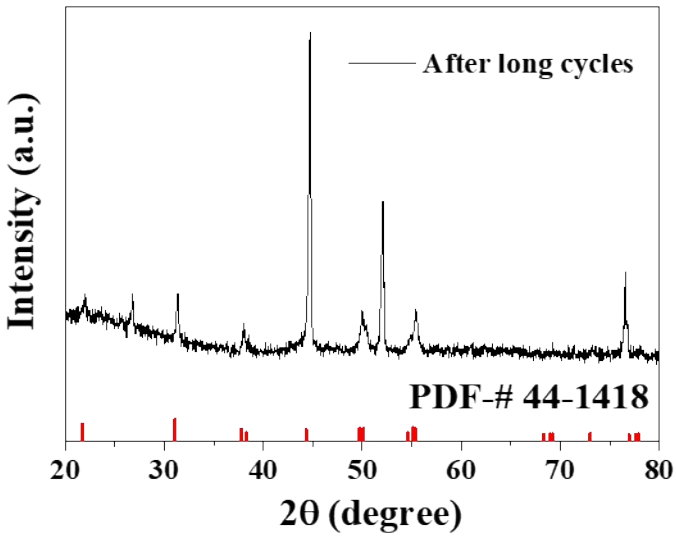

Figure S3 The XRD patterns of the Co-Ni_3S_2@CNTs/GNF sample after cycling test.

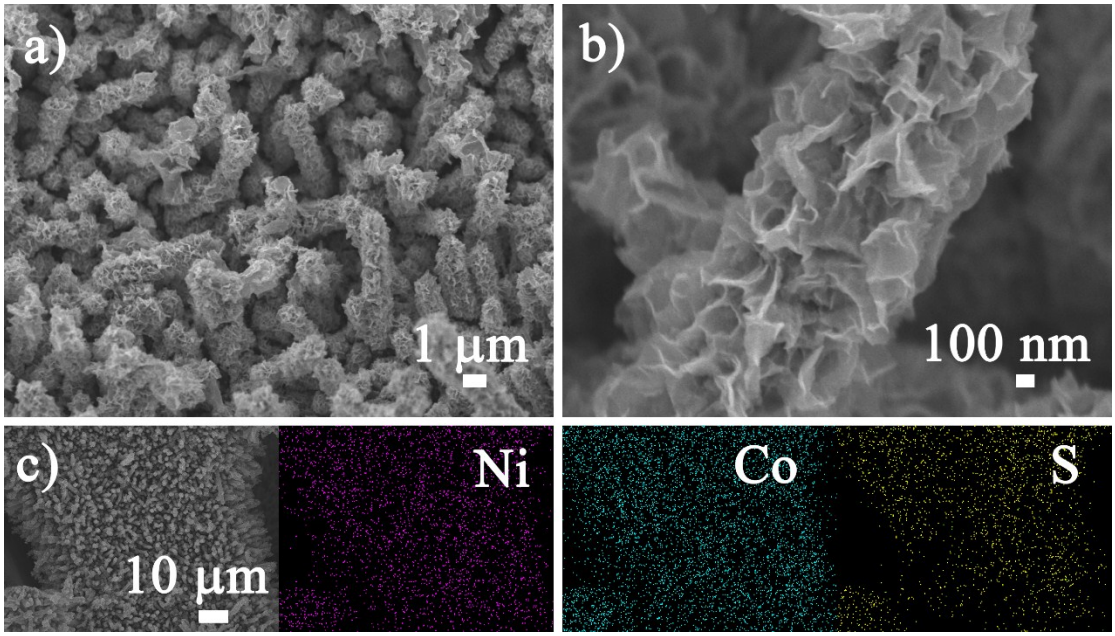

Figure S4 SEM images of a), b) and c) EDS images of the Co-Ni_3S_2@CNT/GNF composites after long cycling test.

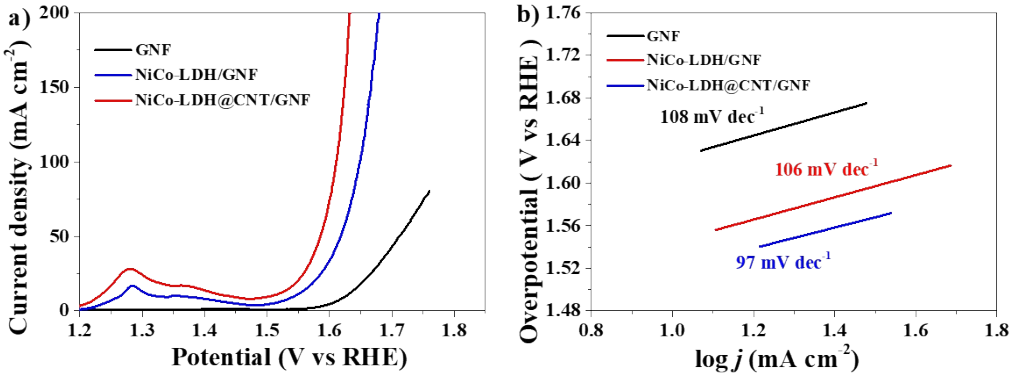

Figure S5 a) LSV curves and b) Tafel curves of NiCo-LDH@CNTs/GNF, NiCo-LDH/GNF and GNF scaffold.

The OER activity of the NiCo-LDH@CNTs/GNF and NiCo-LDH/GNF are also tested, respectively, to further investigate the role of the conductive CNTs array structure (**Figure S5**). As shown in the figures, the LSV curve and the corresponding Tafel curve of the NiCo-LDH@CNTs/GNF composite can also achieve better electrocatalytic activity than that of the NiCo-LDH/GNF and GNF scaffold, which may be due to the increased electron transfer rate.

Table S1 The peak area and the area ratio of the Co/Ni ions with different valence states calculated by the deconvolution of the Co 2p and Ni 2p peaks of the Co-Ni_3S_2@CNTs/GNF electrodes.

		Co(3+)	Co(2+)	Co(3+)/Co(2+)	Ni(3+)	Ni(2+)	Ni(3+)/Ni(2+)
Co-Ni_3S_2 Peak Area	**2p3/2**	**4481.11**	**15975.27**	**0.28**	**42889.90**	**22012.94**	**1.95**
	2p1/2	**2601.53**	**6084.67**	**0.43**	**18706.90**	**9332.31**	**2.00**