Supporting information

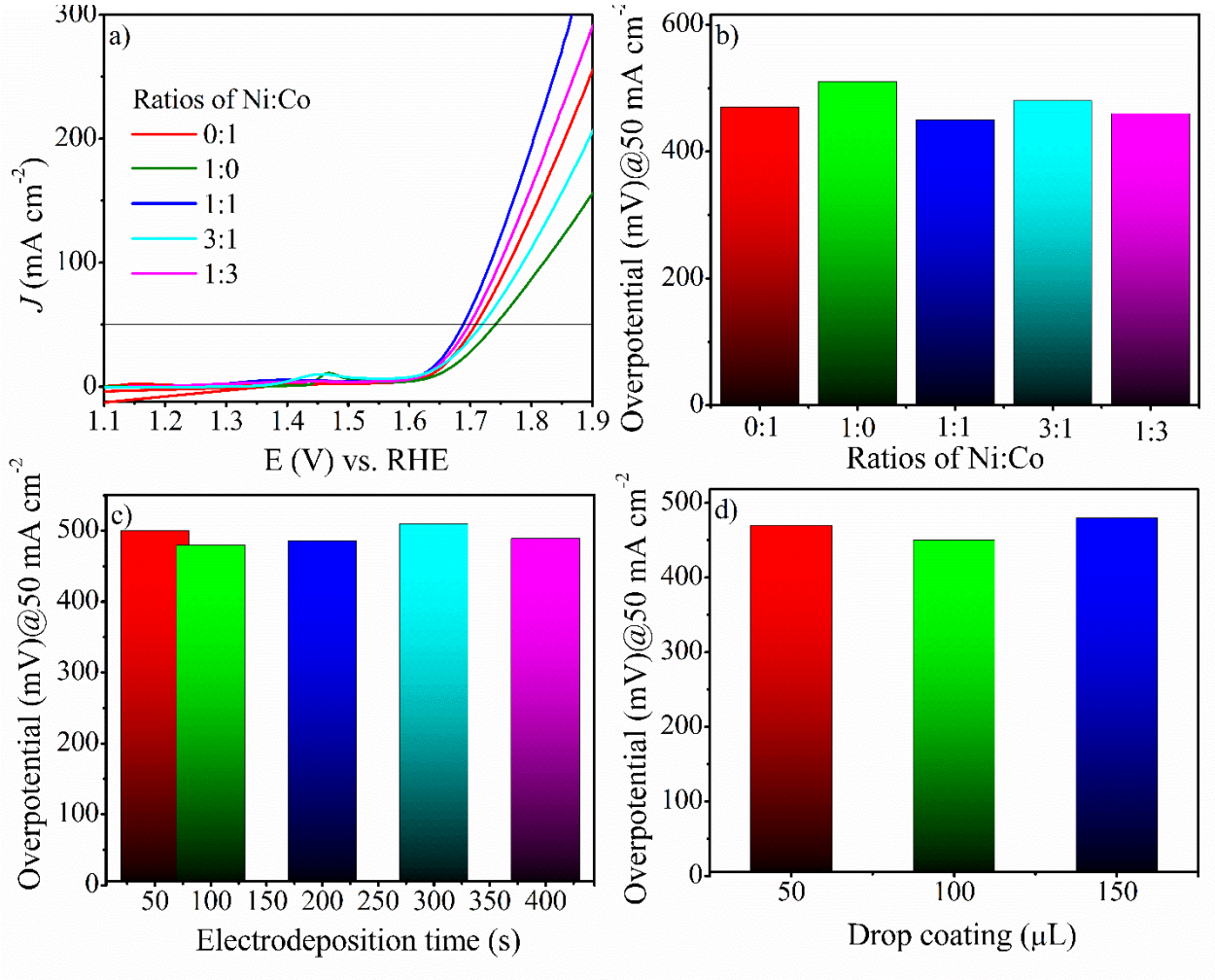

Fig.S1. (a) OER LSVs of NiCoP/NF catalysts in different ratio of Ni:Co in 1 M KOH electrolyte at scan rate of 2 mV s^{-1}.(b) The bar graph of overpotential at 50 mA cm^{-2} of different ratio of Ni:Co. (c) Electrodeposition time of NiCoP on NF. (d) The bar graph of thickness of g-C_3N_4 on NF.

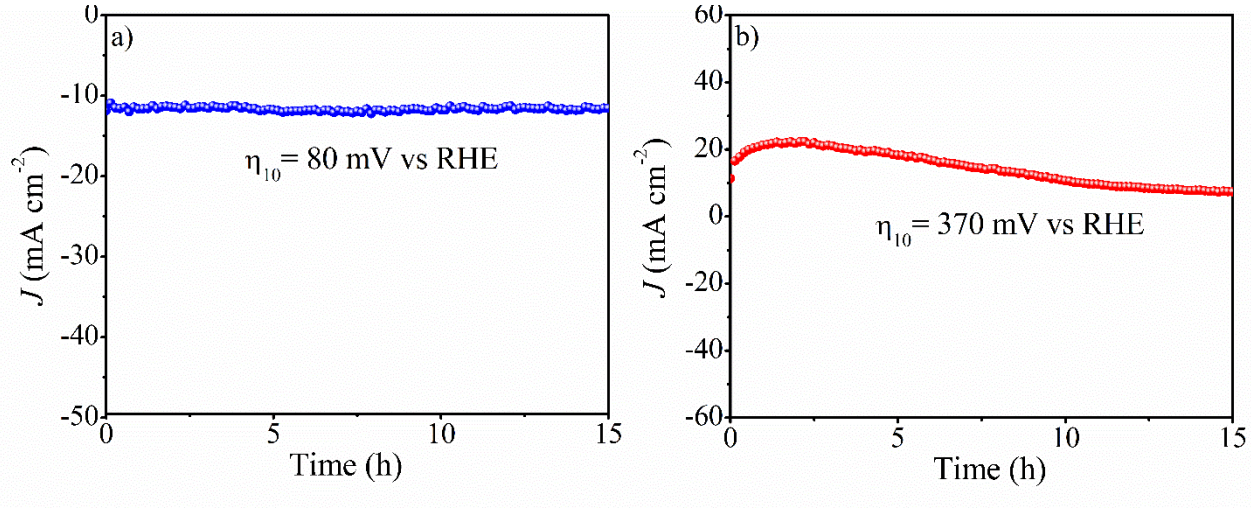

Fig.S2. Chronoamperomograms of g-C_3N_4/NiCoP/NF in 1 M KOH for (a) HER at 80 mV versus RHE, (b) OER at 370 mV vs. RHE.

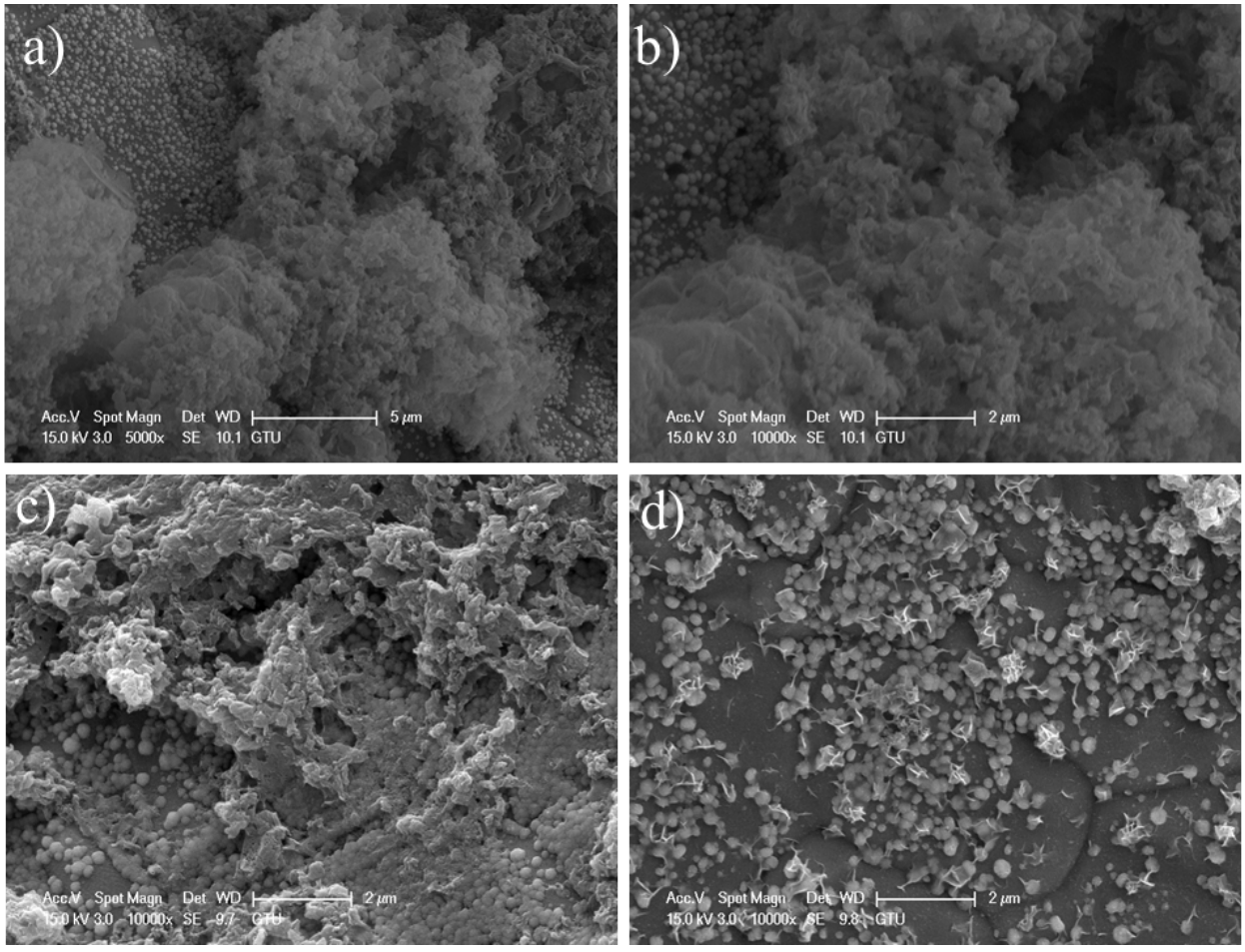

Fig.S3. (a-b) Low- and high-resolution SEM images of g-C_3N_4/NiCoP/NF, (c) Post-HER, (d) Post-OER.

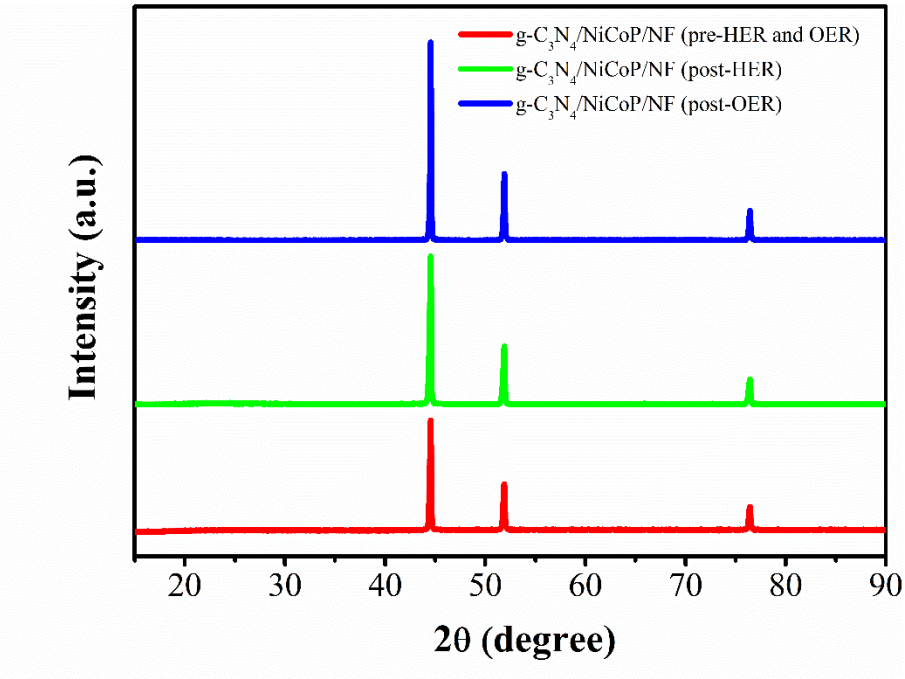

Fig.S4. XRD analysis of g-C_3N_4/NiCoP/NF for Pre-HER and OER, Post-HER and Post-OER.

Table S1. A comparison of electrochemical performances of electrocatalyst containing NiCoP in alkaline electrolyte.

	η_{10} (mV) vs. RHE for HER	Tafel slope (mV dec^{-1})	η_{10}(mV) vs. RHE for OER	Tafel slope (mV dec^{-1})	Cell Voltage (V) for OWS	Ref.
NiCoP/NF	60	51.4	253	49.5	1.55	[1]
$Ni_{0.83}Co_{0.17}P$/NF	-	-	295	45	-	[2]
NiCoP@NC-NCNTs	-	-	227	99	-	[3]
$Ni_{0.51}Co_{0.49}P$/NF	82	43	239	45	1.57	[4]
Ni-Co-P/NF	85	46	-	-	-	[5]
NiCoP NWAs/NF	104	54	~270@η_{20}	116	1.64	[6]
NiCoP@NF-100	98	68	-	-	1.8	[7]
NiCoP	43	77.8	238	96	1.52	[8]
Ti_3C_2@mNiCoP	127	103	237	104	1.57	[9]
NiCoP NR@NS	71	57	268	71	1.57	[10]
NiCoP nanopeapods/CNTs	82	63	250	67	1.558	[11]
NiCoP@PNCNF	98	58	260	63	1.645	[12]
g-C_3N_4/NiCoP/NF	80	89	370	64	1.70	This work

Table S2. The XPS fitting results of NiCoP/NF and g-C_3N_4/NiCoP/NF.

		Peak	Area	BE(eV)	Height	FWHM(eV)
C1s	NiCoP/NF	sp^2C	3880.34	284.90	1539.12	2.37
	g-C_3N_4/NiCoP/NF	sp^2C	5023.28	284.91	2261.12	2.09
			2128.31	287.39	773.70	2.58
O1s	NiCoP/NF	P-O	11517.91	531.44	4632.49	2.34
	g-C_3N_4/NiCoP/NF	P-O	15134.97	531.77	5555.35	2.56
Ni2p	NiCoP/NF	$Ni2p_{3/2}$	3078.76	856.28	1042.40	2.77
		Sat.	2810.87	861.39	484.99	5.44
		$Ni2p_{1/2}$	1364.68	874.03	430.39	2.98
		Sat.	2867.68	879.33	358.97	7.50
	g-C_3N_4/NiCoP/NF	$Ni2p_{3/2}$	3750.61	856.40	1261.06	2.79
		Sat.	2412.28	861.34	471.12	4.81
		$Ni2p_{1/2}$	2160.95	874.20	591.48	3.43
		Sat.	1982.81	879.97	365.04	5.10
Co2p	NiCoP/NF		1688.24	775.54	217.70	7.29
		$Co2p_{3/2}$	1935.98	781.35	624.42	2.91
		Sat.	2960.36	784.76	421.12	6.60
		$Co2p_{1/2}$	1050.21	797.31	324.23	3.04
		Sat.	1327.52	802.48	198.38	6.29
	g-C_3N_4/NiCoP/NF		4182.17	776.19	355.70	11.05
		$Co2p_{3/2}$	3453.05	781.65	1004.56	3.23
		Sat.	2527.72	785.66	460.09	5.16
		$Co2p_{1/2}$	1257.87	797.55	438.32	2.70
		Sat.	1839.66	802.49	263.98	6.55
P2p	NiCoP/NF	P-O	2193.99	133.33	887.43	2.32
	g-C_3N_4/NiCoP/NF	P-O	1681.08	133.15	733.56	2.15

Table S3. The estimated EIS parameters of the catalysts.

	Catalysts	R_s (Ω)	R_{ct}(kΩ)	CPE(S.s^a)
HER	g-C_3N_4	2.09	7.29	428.4x10^{-6}
	NiCoP	2.20	5.16	347.4x10^{-6}
	g-C_3N_4/NiCoP	2.20	4.84	305.3x10^{-6}
OER	g-C_3N_4	1.79	0.27	690.7x10^{-3}
	NiCoP	2.10	14.53	949.7x10^{-6}
	g-C_3N_4/NiCoP	1.81	0.11	35.47x10^{-3}

Table S4. The calculated TOF parameters of the catalysts.

	Catalysts	*I* (mA)	Q (mC)	Γ	TOF (s^{-1})
HER **η= -0.050 mV**	g-C_3N_4	-7.72	-141.9	-8.86x10^{17}	0.027
	NiCoP	-6.42	-118.8	-7.42x10^{17}	0.027
	g-C_3N_4/NiCoP	-9.11	-157.4	-9.82x10^{17}	0.029
OER **η=0.410 mV**	g-C_3N_4	2.36	27.41	1.71x10^{17}	0.022
	NiCoP	7.16	86.72	5.41x10^{17}	0.021
	g-C_3N_4/NiCoP	2.54	28.82	1.80x10^{17}	0.047

References

[1] H.-S. Hu, Y. Li, Y.-R. Shao, K.-X. Li, G. Deng, C.-B. Wang, Y.-Y. Feng, NiCoP nanorod arrays as high-performance bifunctional electrocatalyst for overall water splitting at high current densities, J Power Sources, 484 (2021) 229269.
[2] S. Battiato, L. Bruno, A.L. Pellegrino, A. Terrasi, S. Mirabella, × Optimized electroless deposition of NiCoP electrocalysts for enhanced water splitting, Catalysis Today, (2022).
[3] R. Li, C. Liu, P. Tang, B. Yang, X. Li, G. Yang, L. Xie, NiCoP nanowire arrays embedded in 3D integrated N-doped carbon network for enhanced electrochemical oxygen evolution, Vacuum, 192 (2021) 110395.
[4] J. Yu, Q. Li, Y. Li, C.Y. Xu, L. Zhen, V.P. Dravid, J. Wu, Ternary metal phosphide with triple-layered structure as a low-cost and efficient electrocatalyst for bifunctional water splitting, Adv Funct Mater, 26 (2016) 7644-7651.
[5] C. Yu, F. Xu, L. Luo, H.S. Abbo, S.J. Titinchi, P.K. Shen, P. Tsiakaras, S. Yin, Bimetallic Ni–Co phosphide nanosheets self-supported on nickel foam as high-performance electrocatalyst for hydrogen evolution reaction, Electrochimica Acta, 317 (2019) 191-198.
[6] J. Li, G. Wei, Y. Zhu, Y. Xi, X. Pan, Y. Ji, I.V. Zatovsky, W. Han, Hierarchical NiCoP nanocone arrays supported on Ni foam as an efficient and stable bifunctional electrocatalyst for overall water splitting, J Mater Chem A, 5 (2017) 14828-14837.
[7] L. Chen, Y. Song, Y. Liu, L. Xu, J. Qin, Y. Lei, Y. Tang, NiCoP nanoleaves array for electrocatalytic alkaline H2 evolution and overall water splitting, Journal of Energy Chemistry, 50 (2020) 395-401.
[8] W. Zou, C. Sun, K. Zhao, J. Li, X. Pan, D. Ye, Y. Xie, W. Xu, H. Zhao, L. Zhang, Surface reconstruction of NiCoP pre-catalysts for bifunctional water splitting in alkaline electrolyte, Electrochimica Acta, 345 (2020) 136114.
[9] Q. Yue, J. Sun, S. Chen, Y. Zhou, H. Li, Y. Chen, R. Zhang, G. Wei, Y. Kang, Hierarchical mesoporous MXene–NiCoP electrocatalyst for water-splitting, ACS applied materials interfaces, 12 (2020) 18570-18577.
[10] J.-G. Wang, W. Hua, M. Li, H. Liu, M. Shao, B. Wei, Structurally engineered hyperbranched NiCoP arrays with superior electrocatalytic activities toward highly efficient overall water splitting, ACS applied materials interfaces, 10 (2018) 41237-41245.
[11] J. Bian, Z. Song, Y. Zhang, C. Cheng, NiCoP nanopeapods embedded in carbon nanotube arrays as bifunctional catalysts for efficient overall water splitting, Materials Today Nano, 8 (2019) 100053.
[12] X. Sun, P. Wei, J. Zhang, S. Gu, R. Yang, C. Fang, Q. Li, J. Han, Z. Jiang, J. He, P, N-codoped carbon nanofibers confined ultra-small bimetallic NiCoP for highly efficient overall water splitting, Applied Surface Science, 570 (2021) 151247.