Supplementary Information (SI) for Dalton Transactions.
This journal is © The Royal Society of Chemistry 2025

Supporting Information

Striking the Perfect $Ni(OH)_2$ Balance for Boosted Electrocatalytic Water-Dissociation and H_2 Delivery in Proton-Deficit Environments

Roshini Arulraj[a], Tumpa Sadhukhan[a]*and Anantharaj Sengeni[b]*

[a]Theoretical and Computational Materials Chemistry Laboratory, Department of Chemistry, SRM Institute of Science and Technology, Kattankulathur 603203, Tamil Nadu, India

[b]Laboratory for Electrocatalysis and Energy, Department of Chemistry, Indian Institute of Technology, Kanpur 208 016, Uttar Pradesh, India

* Correspondence should be addressed to: tumpas@srmist.edu.in, and ananths@iitk.ac.in

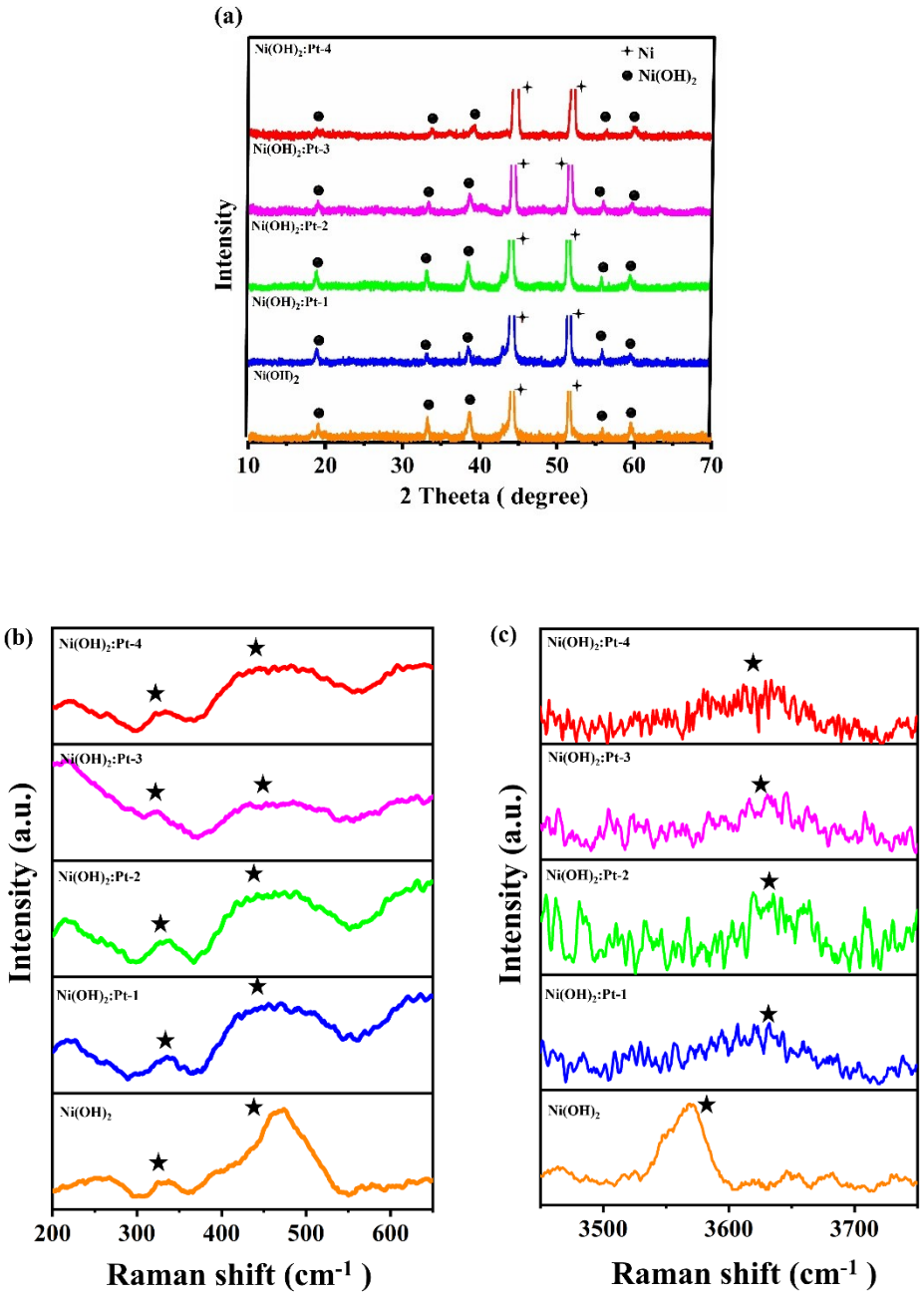


Fig. S1: (a) XRD patterns of $Ni(OH)_2$:Pt , $Ni(OH)_2$:Pt -1, $Ni(OH)_2$:Pt -2, $Ni(OH)_2$:Pt -3 and $Ni(OH)_2$:Pt -4. (b and c) corresponding Raman spectra.

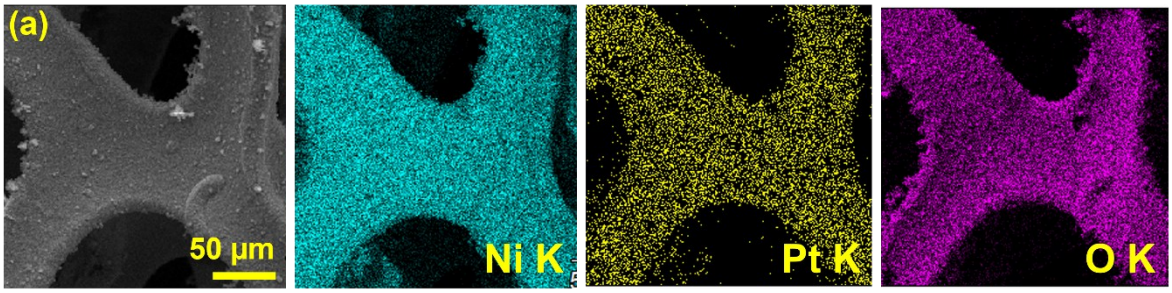
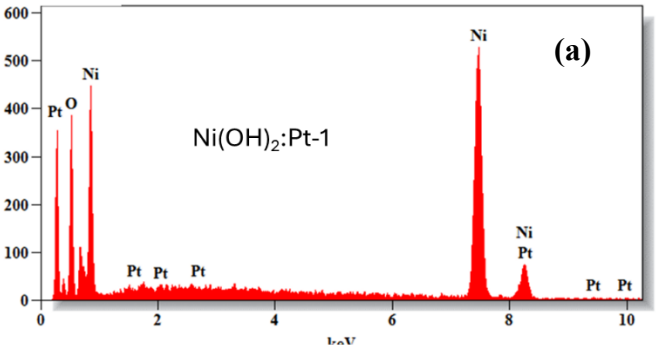
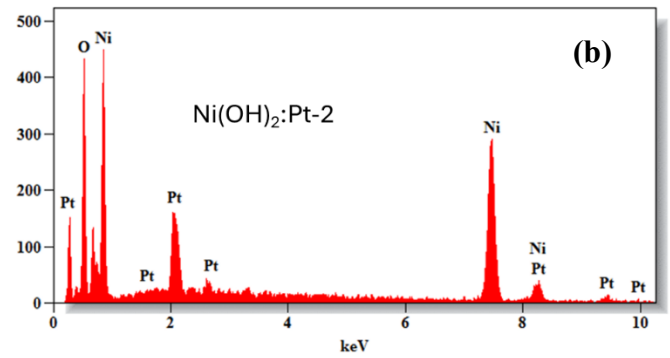


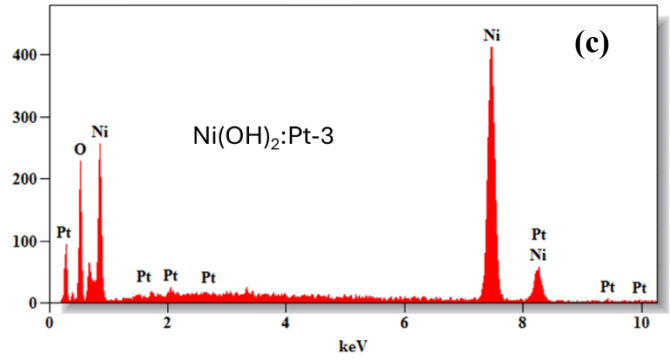
Fig. S2: Elemental mapping of $Ni(OH)_2$:Pt-3 the respective nickel, platinum and oxygen atom.



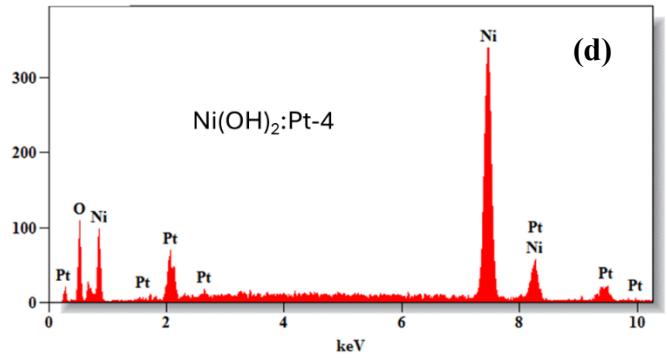
Element	Net Counts	Weight %	Atom %
O	1382	11.77	33.03
Ni	8998	87.29	66.75
Pt	26	0.94	0.22
Total		100.00	100.00



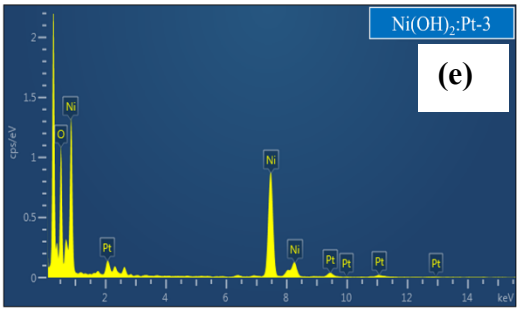
Element	Net Counts	Weight %	Atom %
O	1726	22.91	56.09
Ni	4612	60.98	40.68
Pt	56	2.69	0.66
Total		100.00	100.00



Element	Net Counts	Weight %	Atom %
O	814	9.44	28.09
Ni	6892	87.77	71.26
Pt	328	16.10	3.23
Total		100.00	100.00



Element	Net Counts	Weight %	Atom %
O	450	6.44	24.34
Ni	5685	64.86	66.77
Pt	681	28.70	8.89
Total		100.00	100.00



Element	Line Type	K Factor	Absorption Correction	Wt%	Wt%	Atomic %
O	K series	2.020	1.00	42.90	1.11	75.16
Ni	K series	1.164	1.00	49.86	1.10	23.80
Pt	L series	2.266	1.00	7.24	1.05	1.04
Total:				100.00		100.00

Fig. S3: SEM-EDS spectra of $Ni(OH)_2$:Pt-R (R=1-4) with corresponding elemental composition tables (a-d) and TEM-EDS spectra of $Ni(OH)_2$:Pt-3 with corresponding elemental composition table (e).

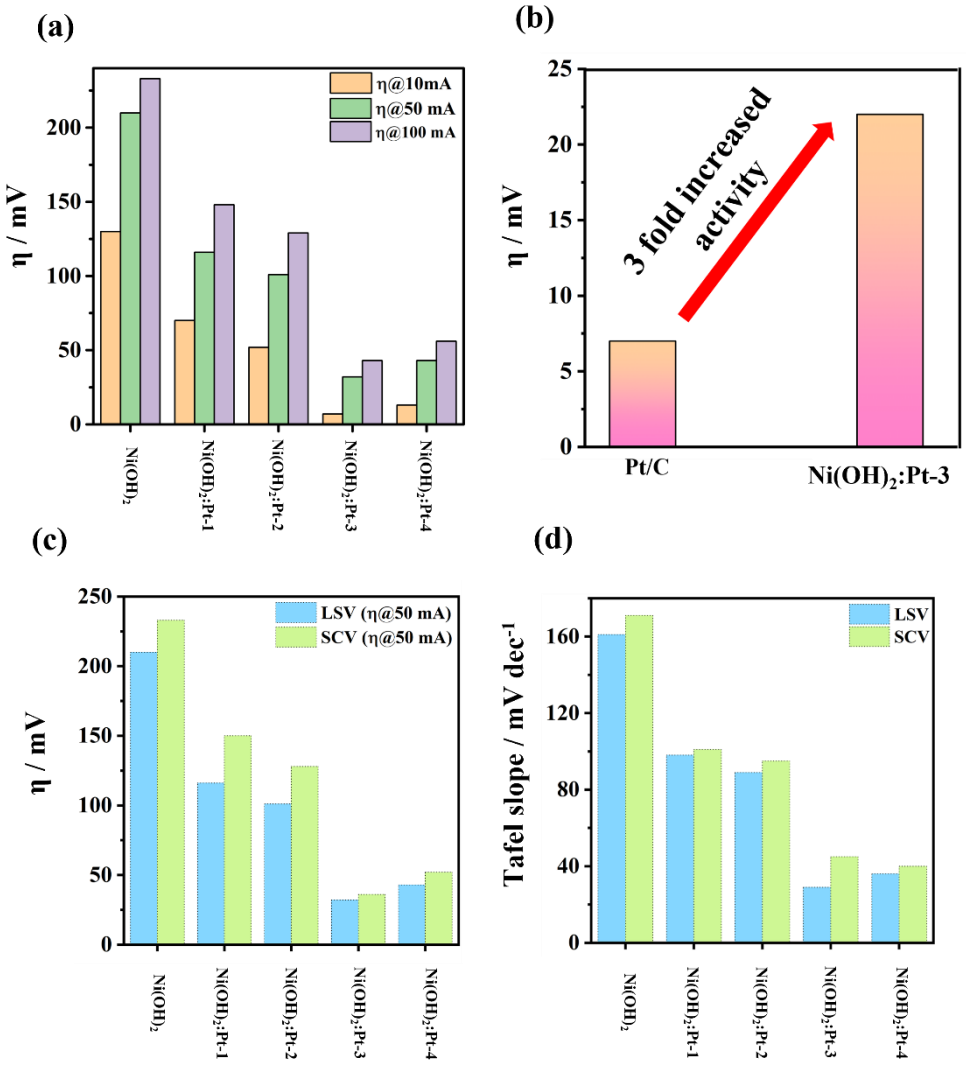


Fig. S4: (a) Comparative bar chart showing overpotentials at current densities of 10, 50, and 100 for the respective electrocatalysts from LSV (b) Comparative plot between Pt/C and Ni(OH)2:Pt-3, and (c-d) Comparison plots of LSV and SCV for overpotential and Tafel slope values.

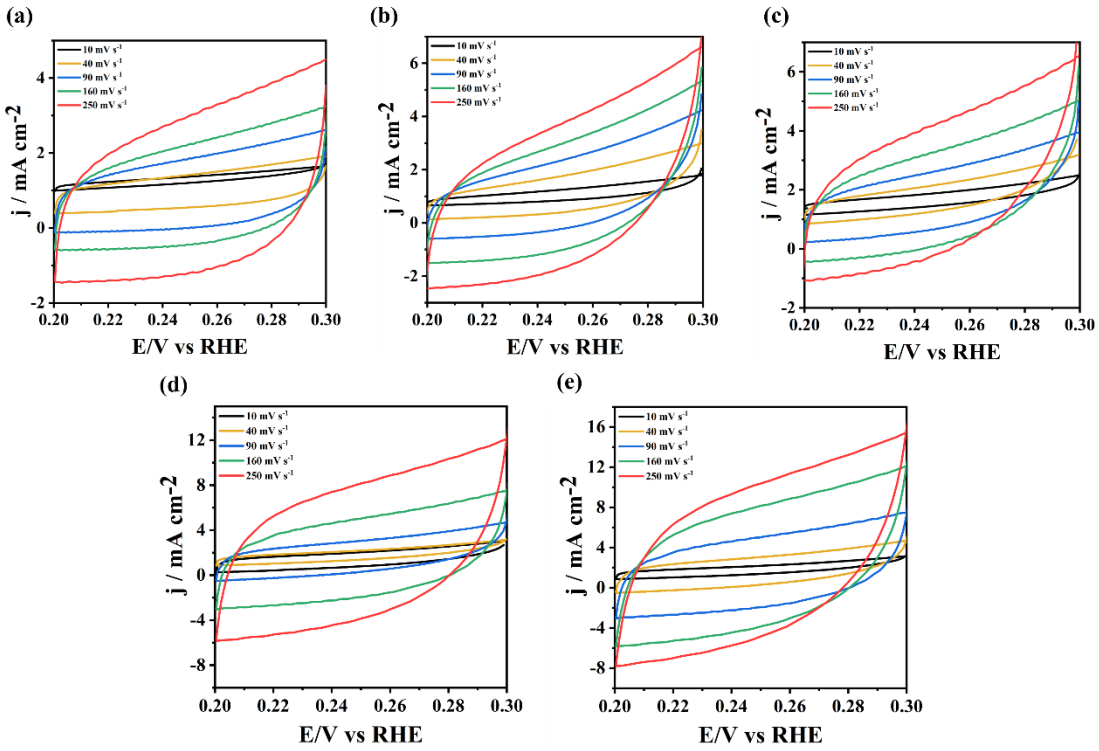


Fig. S5: Cyclic voltammetry (CV) responses in the non-Faradaic region for the determination of double-layer capacitance (C_{dl}) values for (a) $Ni(OH)_2$, (b) $Ni(OH)_2$:Pt-1, (c) $Ni(OH)_2$:Pt-2, (d) $Ni(OH)_2$:Pt-3 and (e) $Ni(OH)_2$:Pt-4 respectively.

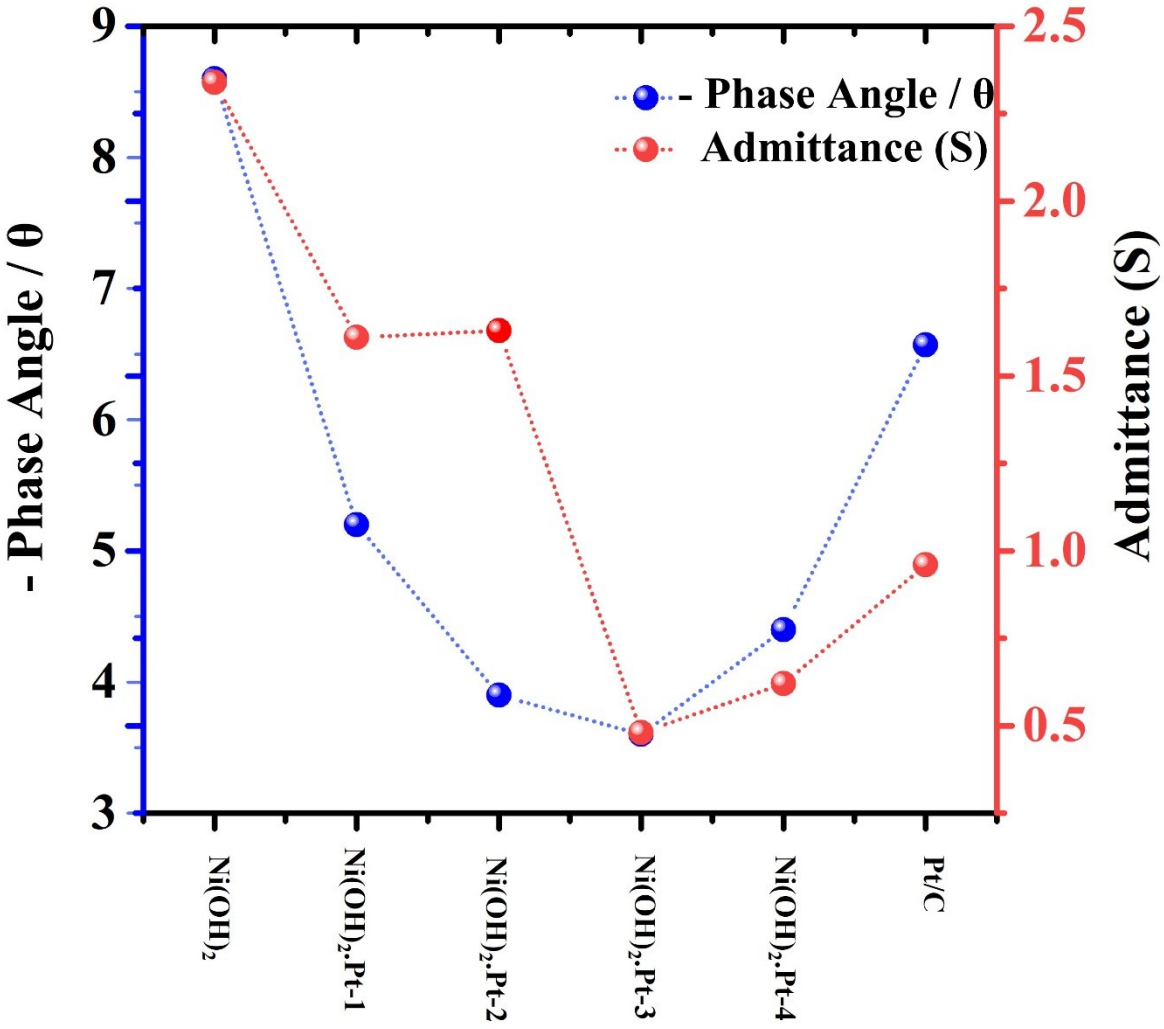


Fig. S6: Admittance and Phase angle values from EIS of all catalysts.

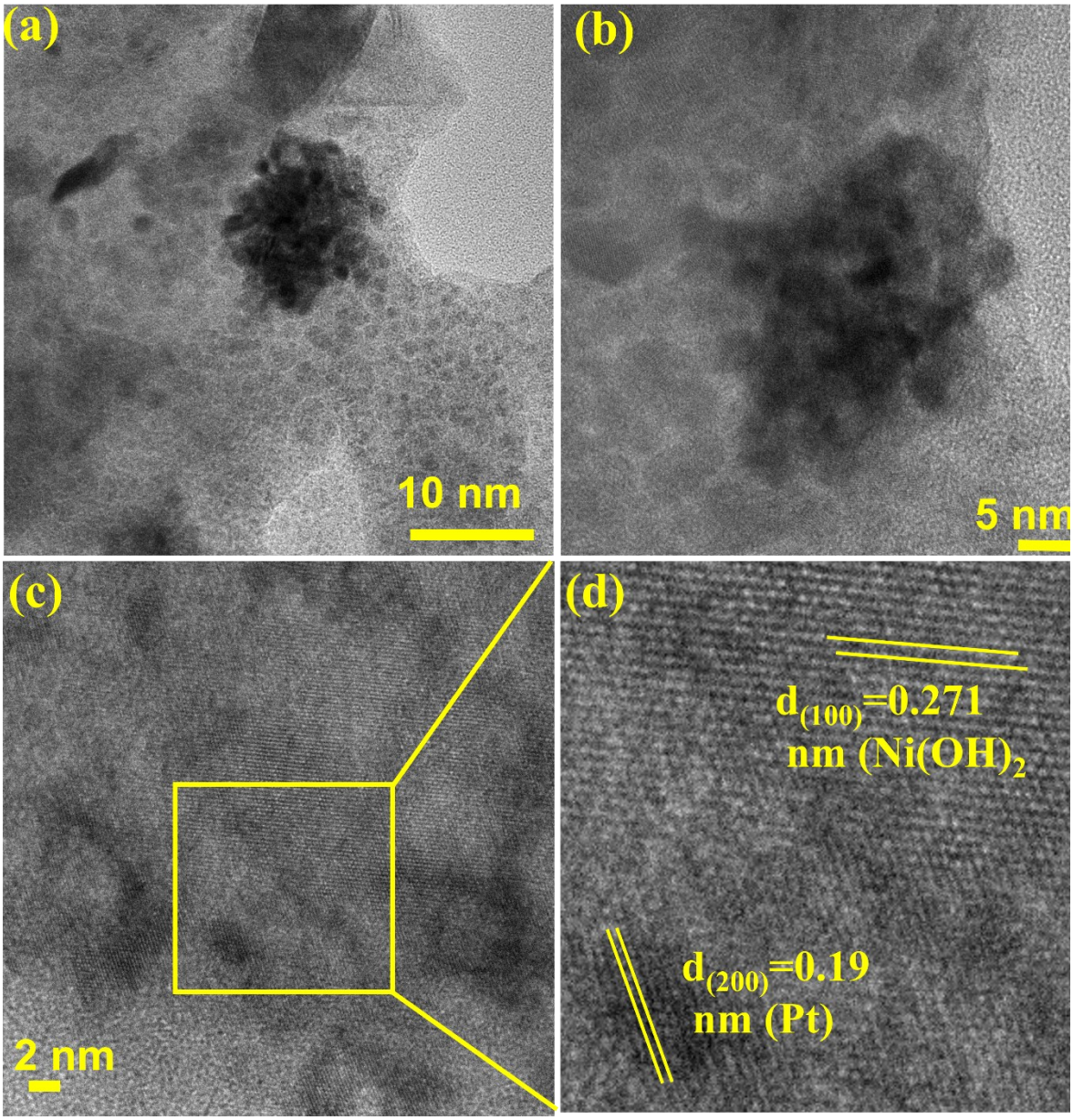


Fig. S7: (a-b) After stability TEM images of $Ni(OH)_2$:Pt-3 at progressively higher magnifications and (c-d) TEM images of the same sample at even higher magnifications, revealing the distinct lattice fringes of $Ni(OH)_2$:Pt-3.

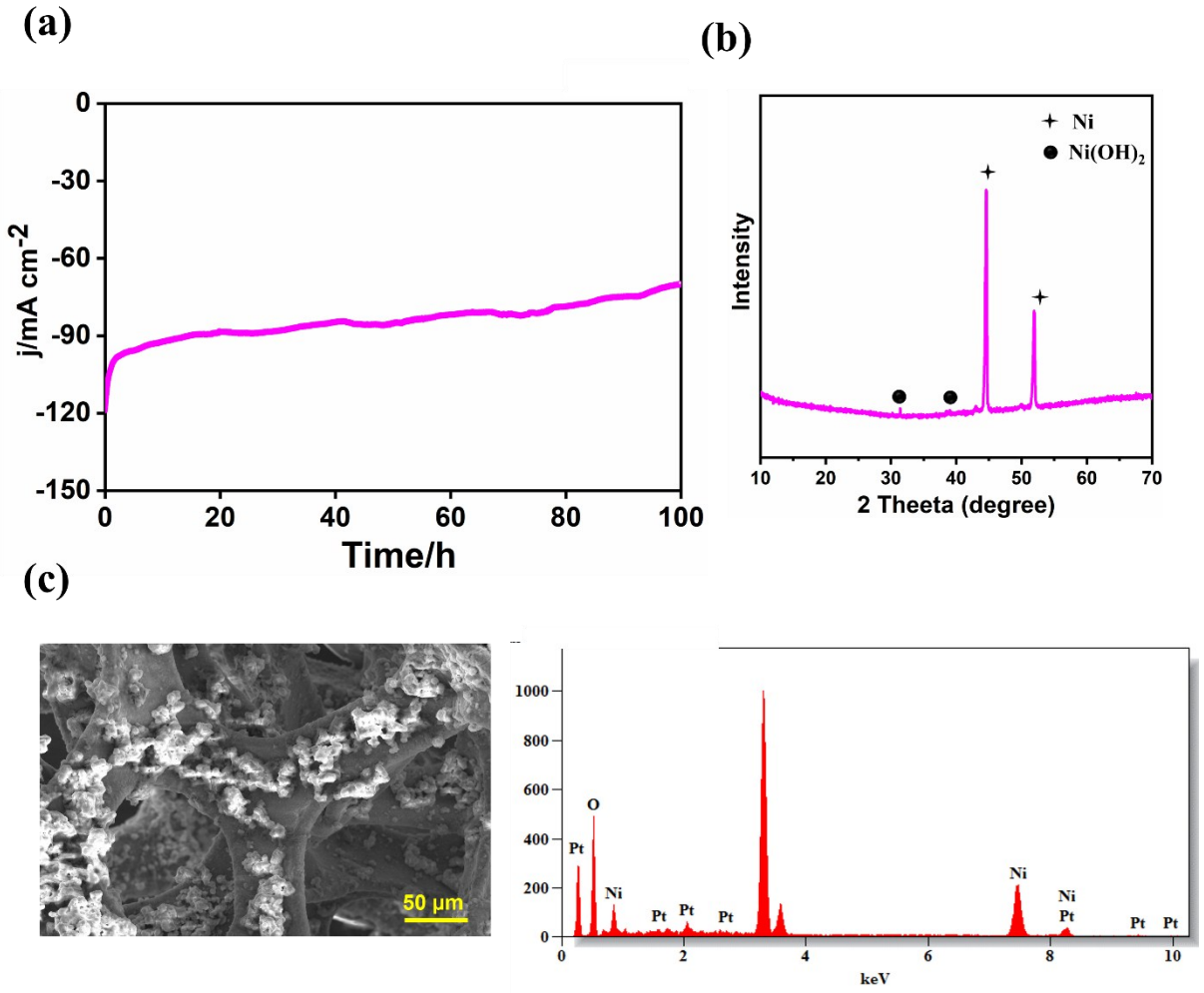


Fig. S8: (a) Stability of $Ni(OH)_2$:Pt-3 after 100 hours. (b) XRD spectrum of $Ni(OH)_2$:Pt-3 after 100 hours. (c) SEM and EDX images of $Ni(OH)_2$:Pt-3 after 100 hours.

Electrocatalyst	Electrolyte	η_{10} (mV)	η_{100} (mV)	Tafel slope (mV dec-1)	Reference
$PtNi_5$	1.0 M KOH	26.8	90.0	19.2	1
Pt–$Co(OH)_2$/CC	1.0 M KOH	32.0	110.0	70.0	2
Pt–CoFe(II) LDHs	1.0 M KOH	15.0	70.0	45.3	3
PtNiP NWs	1.0 M KOH	9.0	44.0	30.5	4
Pt_2Ni_3-P NWs	1.0 M KOH	44.0	—	66.0	5
β-$Ni(OH)_2$/Pt	0.1 M KOH	92.0	—	42.0	6
Pt/TiO_2/Ni(OH)2/NF	1.0 M KOH	26.0	80.0	38.9	7
Pt–Ni_2Fe_1	1.0 M KOH	80.0	350	67.0	8
D-PtNi/C	1.0 M KOH	39.7	—	55.0	9
PtSA–NiO/Ni	1.0 M KOH	26.0	85.0	27.1	10
$Ni(OH)_2$:Pt	1.0 M KOH	9.0	50	40.0	**Our work**

Table. S1: Comparison of electrocatalysts for alkaline hydrogen evolution reaction based on electrochemical activity.

Catalyst	Overpotential (10 mA cm^{-2})	electrolytes	Content of Pt	References
$Ni(OH)_2$-PtO_2 NS/Ti	> 45	1M KOH	5.10 wt%	11
Pt NWs/SL-$Ni(OH)_2$	> 85	1M KOH	38.00 wt%	12
Pt/Nb-$Co(OH)_2$	112 mV	1 M KOH	3.34 wt%	13
Pt@DG	37 mV	1 M KOH	1.57 wt%	14
Pt-$Ni(OH)_x$	58 mV	1 M KOH	0.17 wt%	15
Pt-SA/a-$Ni(OH)_2$	64 mV	1 M KOH	0.07 wt%	16
Pt_5/HMCS	46.2 mV	1 M KOH	5.08 wt%.	17
$Ni(OH)_2$:Pt	9 mV	1 M KOH	16.10 wt%	Our work

Table. S2: Evaluation of the electrocatalytic performance of $Ni(OH)_2$:Pt for HER in comparison to catalysts from Previous studies.

References:

1. C. Zhang, X. Liang, R. Xu, C. Dai, B. Wu, G. Yu, B. Chen, X. Wang, N. Liu, Adv. Funct. Mater., 2021, 31, 2008298.
2. Z. Xing, C. Han, D. Wang, Q. Li, X. Yang, ACS Catal., 2017, 7, 7131–7135.
3. J. Wu, Z. Nie, R. Xie, X. Hu, Y. Yu, N. Yang, J. Power Sources, 2022, 532, 231353.
4. Q. Chen, B. Wei, Y. Wei, P. Zhai, W. Liu, X. Gu, Z. Yang, J. Zou, R. Zhang, Y. Gong, Appl. Catal., B, 2022, 301, 120754.
5. P. Wang, Q. Shao, J. Guo, L. Bu, X. Huang, Chem. Mater., 2020, 32, 3144–3149.
6. X. Yu, J. Zhao, L. R. Zheng, Y. Tong, M. Zhang, G. Xu, C. Li, J. Ma, G. Shi, ACS Energy Lett., 2017, 3, 237–244.
7. A. Kong, M. Peng, M. Liu, Y. Lv, H. Zhang, Y. Gao, J. Liu, Y. Fu, W. Li, J. Zhang, Appl. Catal., B, 2022, 316, 121654.
8. L. Wang, L. Zhang, W. Ma, H. Wan, X. Zhang, X. Zhang, S. Jiang, J. Y. Zheng, Z. Zhou, Adv. Funct. Mater., 2022, 32, 2203342.
9. H. Chen, G. Wang, T. Gao, Y. Chen, H. Liao, X. Guo, H. Li, R. Liu, M. Dou, S. Nan, Q. He, J. Phys. Chem. C, 2020, 124, 5036–5045.
10. K. L. Zhou, Z. Wang, C. B. Han, X. Ke, C. Wang, Y. Jin, Q. Zhang, J. Liu, H. Wang, H. Yan, Nat. Commun., 2021, 12, 3783.
11. L. Xie, X. Ren, Q. Liu, G. Cui, R. Ge, A. M. Asiri, X. Sun, Q. Zhang, L. Chen, J. Mater. Chem. A, 2018, 6, 1967-1970.
12. M. Xie, S. Duan, Y. Shen, K. Fang, Y. Wang, M. Lin, X. Guo, ACS Energy Lett., 2016, 1, 814-819.
13. Y. Tian, M. Wen, A. Huang, Q. Wu, Z. Wang, Q. Zhu, T. Zhou, Y. Fu, *Small*, 2023, 19,

2207569.

14. Q. Yang, H. Liu, P. Yuan, Y. Jia, L. Zhuang, H. Zhang, X. Yan, G. Liu, Y. Zhao, J. Liu, S. Wei, J. Am. Chem. Soc., 2022, 144, 2171-2178.
15. Q. Li, Q. Zhang, W. Xu, R. Zhao, M. Jiang, Y. Gao, W. Zhong, K. Chen, Y. Chen, X. Li, N. Yang, Adv. Energy Mater., 2023, 13, 2203955.Li, Q., Zhang, Q., Xu, W., Zhao, R.,
16. Y. Liu, G. Liu, X. Chen, C. Xue, M. Sun, Y. Liu, J. Kang, X. Sun, L. Guo, Nano-Micro Lett., 2024, 16, 202.
17. X. K. Wan, H. B. Wu, B. Y. Guan, D. Luan, X. W. Lou, *Adv. Mater.*, 2020, 32, 1901349.