Supporting Information

Layered Double (Ni, Fe) Hydroxide Loaded with Platinum Nanoparticles as an Efficient Catalyst for the Hydrogen Evolution Reaction

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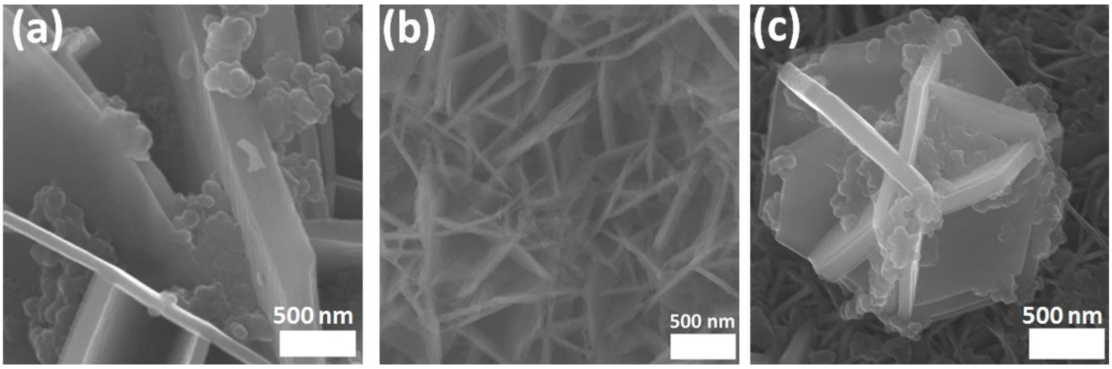

Figure S1. (a) SEM images of NiFe-LDH@NCP+CB (PVP0.3), (b) NiFe-LDH@NCP+CB (PVP0.3)-150-2.5h and (c) Pt@NiFe-LDH@NCP+CB (PVP0.3).

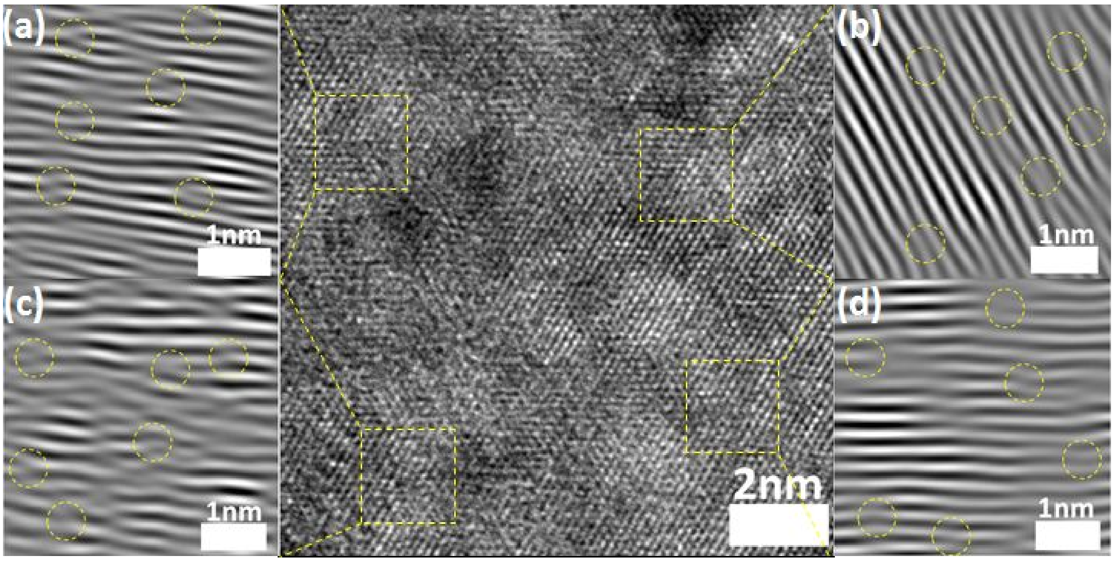

Figure S2. TEM images of defects and dislocations of Pt@NiFe-LDH@NCP+CB(PVP 0.3)-150-2.5h, and after Fourier transform.

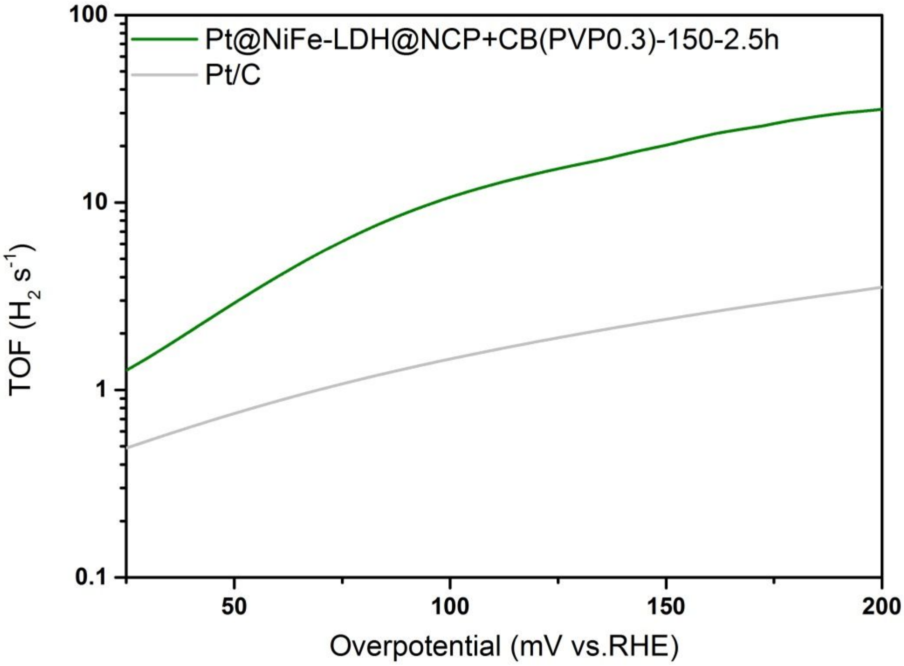

Figure S3. The turnover frequency (TOF) curve of Pt@NiFe-LDH@NCP+CB (PVP 0.3)-150-2.5h, in comparison with commercial Pt/C catalyst.

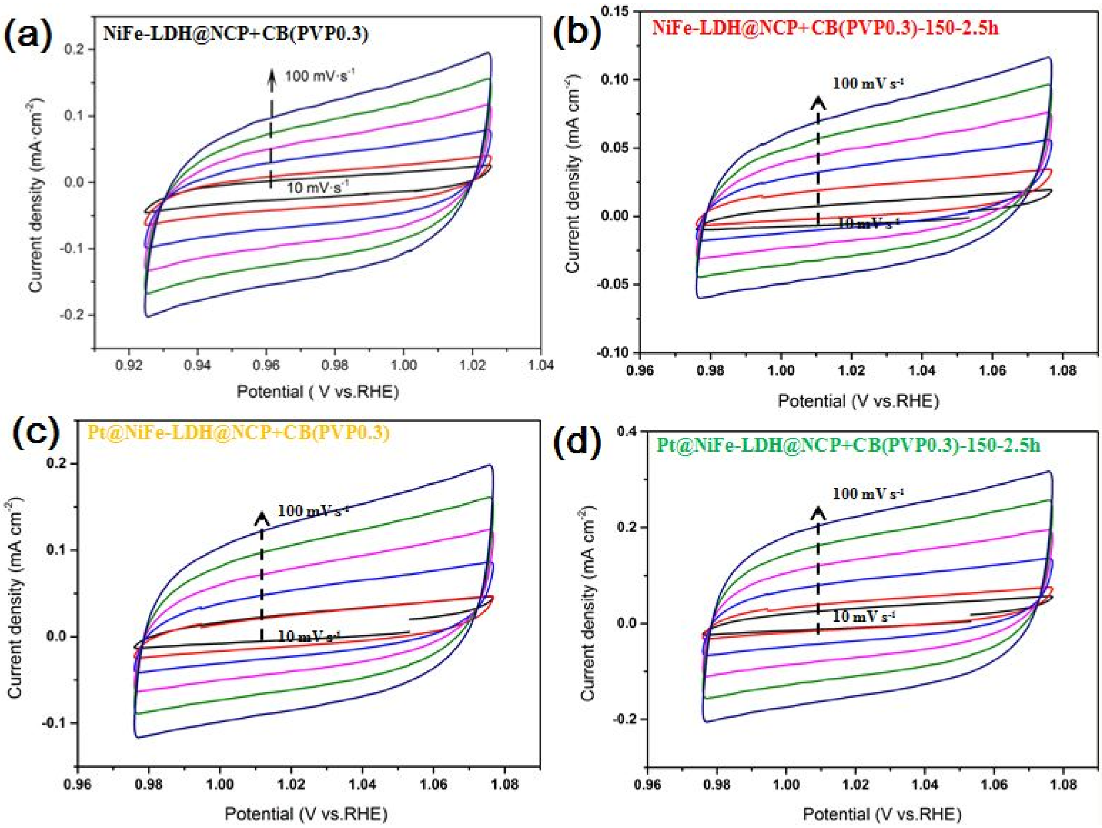

Figure S4. (a) CVs of the NiFe-LDH@NCP+CB (PVP 0.3), (b) NiFe-LDH@NCP+CB(PVP 0.3)-150-2.5h, (c) Pt@NiFe-LDH@NCP+CB(PVP 0.3), and (d) Pt@NiFe-LDH@NCP+CB(PVP 0.3)-150-2.5h at different scan rates of 10, 20, 40, 60, 80 and 100 mV s^{-1}.

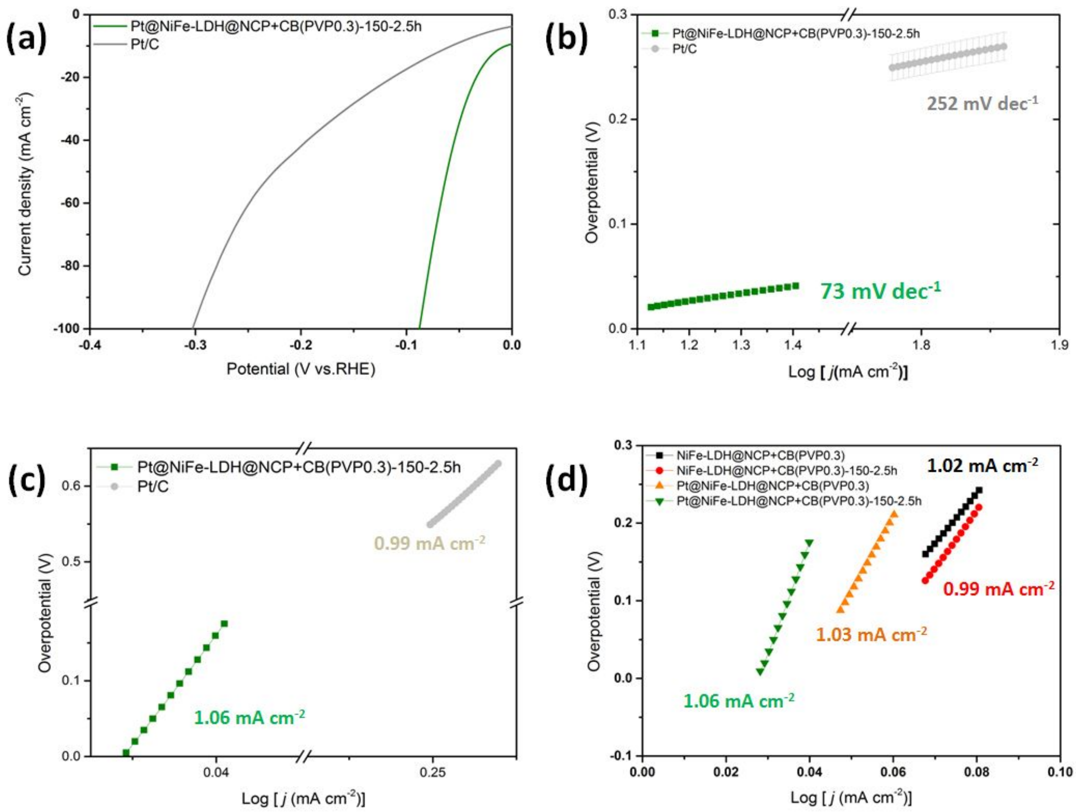

Figure S5. (a) HER LSV curve of the Pt@NiFe-LDH@NCP+CB(PVP 0.3)-150-2.5h and Pt/C，(b) Tafel plots , and (c,d) Exchange current density.

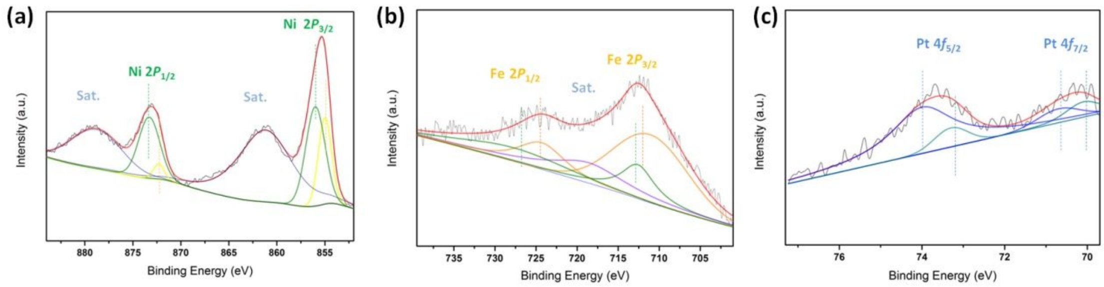

Figure S6. XPS spectra of Ni 2*p* (a), Fe 2*p* (b) and Pt 4*f* (c) of H-Pt@NiFe-LDH@NCP+CB (PVP 0.3)-150-2.5h.

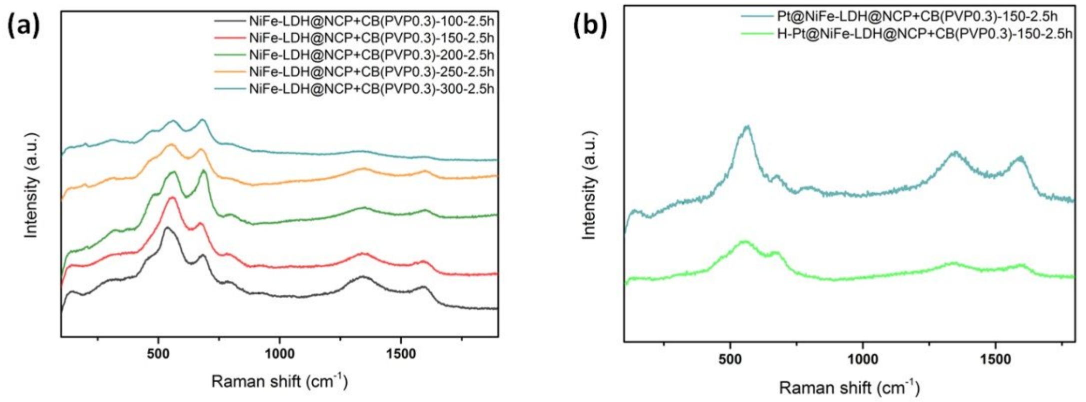

Figure S7. Raman spectra of the samples prepated at different temperatures: (a) NiFe-LDH@NCP+CB(PVP 0.3)-150-2.5h, (b) Pt@NiFe-LDH@NCP+CB(PVP 0.3)-150-2.5h.

Table S1. Comparison of the catalysts with those of the recently reported advanced NiFe oxides or hydroxides electrocatalysts for HER.

Catalyst	**Electrolyte**	**Current density (j, mA cm^{-2})**	**Overpotential (mV vs. RHE)**	**References**
Pt@NiFe-LDH@NCP +CB (PVP 0.3)-150-2.5h	**1 M KOH**	**50**	**63**	**This work**
$Ni_{0.75}Fe_{0.125}V_{0.125}$-LDHs	1 M KOH	10	125	Small 2017, 1703257
NiFe LDH/NF	1 M KOH	10	210	Science 2014, 345,1593-1596
NiFe(II,III)-LDH	1 M KOH	10	113	Small 2019, *15*, 1902551
Cu@ NiFe LDH	1M KOH	10	116	Energy Environ. Sci. 2017, *10*, 1820-1827.
NiFe LDH@NiCoP/NF	1 M KOH	10	120	Adv. Funct. Mater. 2018, 28,1706847
NiFe LDH-NS@DG10	1 M KOH	20	115	Adv. Mater. 2017, *29*, 1700017.
Pt-NiFe-LDH-0.5-12	1 M KOH	50	86	Nano Energy 2020, *72*, 104669
CoP/rGO-400	1 M KOH	50	191	Chem. Sci. 2016, 7, 1690-1695
$Ni_{0.7}Fe_{0.3}S_2$	1 M KOH	50	273	J. Mater. Chem. A 2017, 5, 15838-15844
NiFeOx/CFP	1 M KOH	50	201	Nat. Commun. 2015, *6*, 7261.
Porous MoO_2	1 M KOH	50	97	Adv. Mater. 2016, *28*, 3785-3790
NiFeV-LDHs/NF	1 M KOH	50	214	Small 2018, *14*, 1703257
IPNTs	1 M KOH	50	140	Chemistry 2015, 21, 18062

Table S2 The valence states of Ni in samples in different states analyzed by XPS.

Sample name	Orbit	Binding energy/eV	Area	Ratio of Ni^{3+}/Ni^{2+}
NiFe-LDH@NCP+CB (PVP 0.3)	Ni^{3+}-$2p_{3/2}$	855.5	19850	4.333
	Ni^{2+}-$2p_{3/2}$	854.2	3197	
	Ni^{3+}-$2p_{1/2}$	873.4	12926	
	Ni^{2+}-$2p_{1/2}$	872.7	4367	
Pt@NiFe-LDH@NCP+CB (PVP 0.3)-150-2.5h	Ni^{3+}-$2p_{3/2}$	856.1	136102	∞
	Ni^{2+}-$2p_{3/2}$	-	0	
	Ni^{3+}-$2p_{1/2}$	873.6	59295	
	Ni^{2+}-$2p_{1/2}$	-	0	
H-Pt@NiFe-LDH@NCP+CB (PVP 0.3)-150-2.5h	Ni^{3+}-$2p_{3/2}$	855.8	75805	2.45
	Ni^{2+}-$2p_{3/2}$	855.0	45547	
	Ni^{3+}-$2p_{1/2}$	873.3	51389	
	Ni^{2+}-$2p_{1/2}$	872.5	6470	

Table S3 The valence states of Fe in samples in different states analyzed by XPS.

Sample name	Orbit	Binding energy/eV	Area	Ratio of Fe^{3+}/Fe^{2+}
NiFe-LDH@NCP+CB (PVP 0.3)	Fe^{3+}-2$p_{3/2}$	713.0	6302	0.673
	Fe^{2+}-2$p_{3/2}$	711.1	13299	
	Fe^{3+}-2$p_{1/2}$	724.5	3373	
	Fe^{2+}-2$p_{1/2}$	718.6	1078	
Pt@NiFe-LDH@NCP+CB (PVP 0.3)-150-2.5h	Fe^{3+}-2$p_{3/2}$	713.3	18065	0.398
	Fe^{2+}-2$p_{3/2}$	711.1	72873	
	Fe^{3+}-2$p_{1/2}$	724.5	16665	
	Fe^{2+}-2$p_{1/2}$	718.6	14358	
H-Pt@NiFe-LDH@NCP+CB (PVP 0.3)-150-2.5h	Fe^{3+}-2$p_{3/2}$	713.2	17256	0.461
	Fe^{2+}-2$p_{3/2}$	711.1	55289	
	Fe^{3+}-2$p_{1/2}$	724.3	15621	
	Fe^{2+}-2$p_{1/2}$	718.6	16091	

Table S4 The valence states of Pt in samples in different states analyzed by XPS.

Sample name	Orbit	Binding energy/eV	Area	Ratio of Pt^{2+}/Pt^{0}
NiFe-LDH@NCP+CB (PVP 0.3)	Pt^{2+}-$4f_{7/2}$	-	-	-
	Pt^{0} -$4f_{7/2}$	-	-	
	Pt^{2+}-$4f_{5/2}$	-	-	
	Pt^{0}-$4f_{5/2}$	-	-	
Pt@NiFe-LDH@NCP+CB (PVP 0.3)-150-2.5h	Pt^{2+}-$4f_{7/2}$	73.1	4613	1.838
	Pt^{0}-$4f_{7/2}$	72.3	1840	
	Pt^{2+}-$4f_{5/2}$	75.6	4501	
	Pt^{0}-$4f_{5/2}$	75.1	3119	
H-Pt@NiFe-LDH@NCP+CB (PVP 0.3)-150-2.5h	Pt^{2+}-$4f_{7/2}$	70.7	589	3.857
	Pt^{0}-$4f_{7/2}$	70.1	498	
	Pt^{2+}-$4f_{5/2}$	74.0	3885	
	Pt^{0}-$4f_{5/2}$	73.3	662	