Supporting Information for

N-doping induced lattice-strained porous PdIr bimetallene for pH-universal hydrogen evolution electrocatalysis

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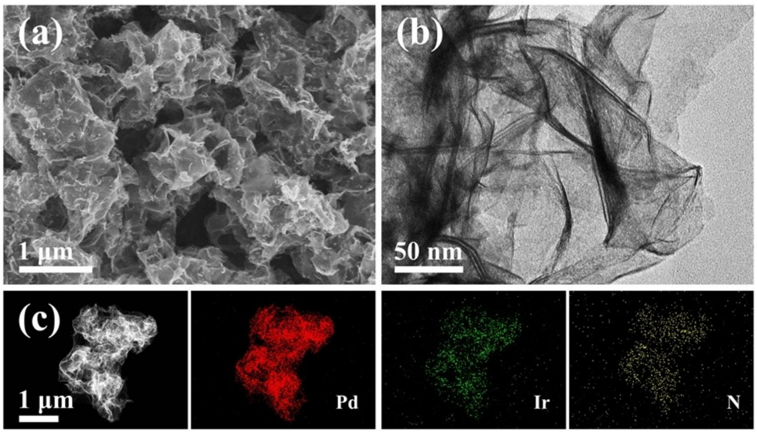


Fig. S1 (a) SEM and (b)TEM images of the PdIr bimetallene. (c) HAADF-STEM image and associated element mapping images.

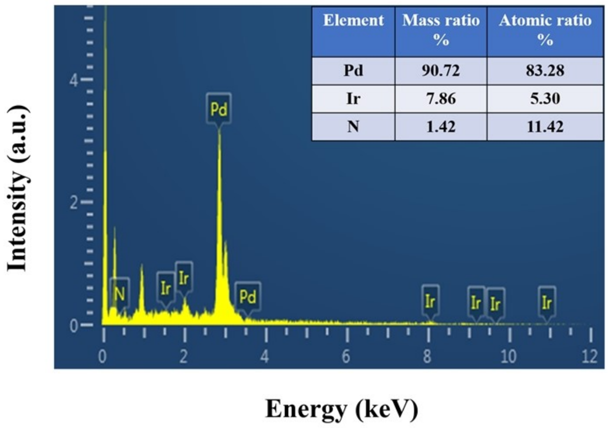


Fig. S2 EDX spectrum of the PdIr bimetallene.

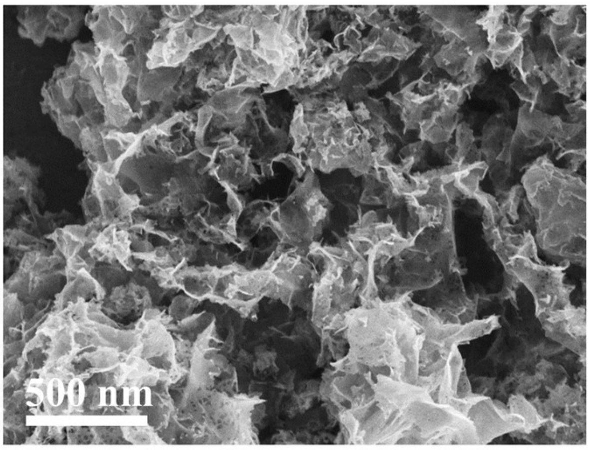


Fig. S3 Large-scale SEM image of the N-PdIr bimetallene.

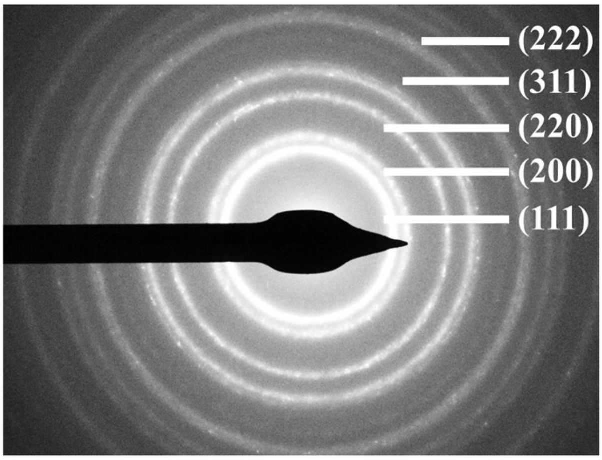
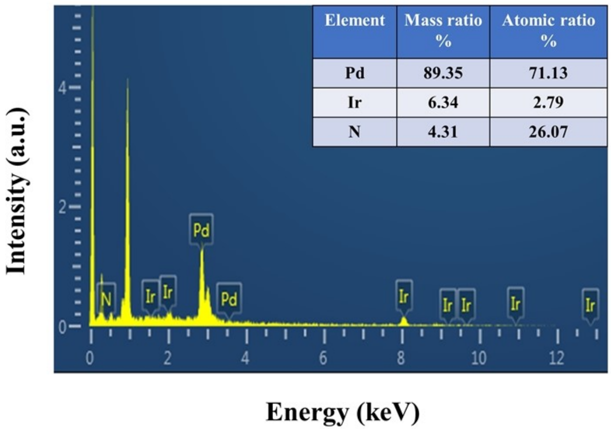


Fig. S4 The SAED pattern of the N-PdIr bimetallene.



Element	Mass ratio %	Atomic ratio %
Pd	89.35	71.13
Ir	6.34	2.79
N	4.31	26.07

Fig. S5 EDX spectrum of the N-PdIr bimetallene.

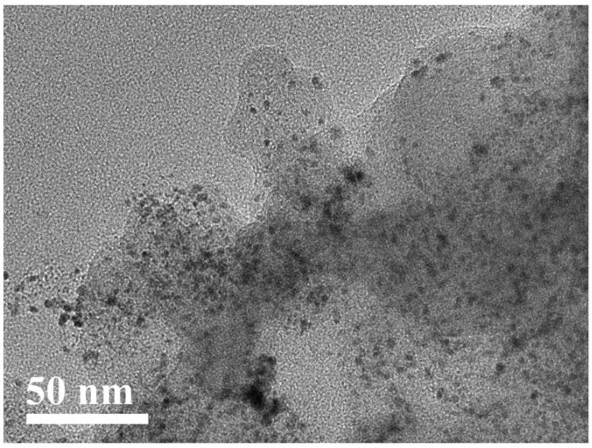


Fig. S6 The TEM image of the Pt/C.

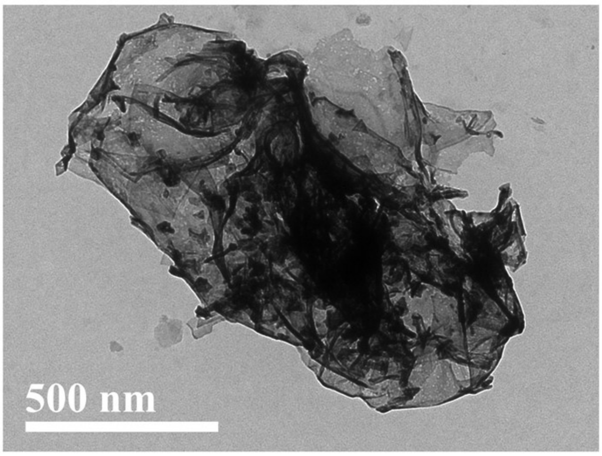


Fig. S7 The TEM image of the N-Pd metallene.

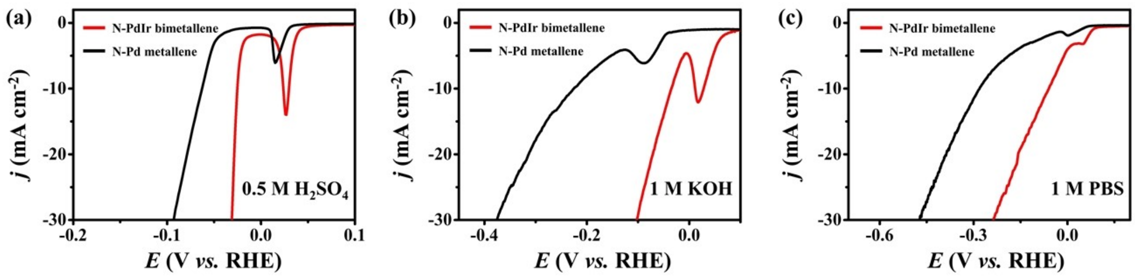


Fig. S8 (a, b, and c) HER polarization curves of the N-PdIr bimetallene and N-Pd metallene in acidic, alkaline, and neutral electrolytes.

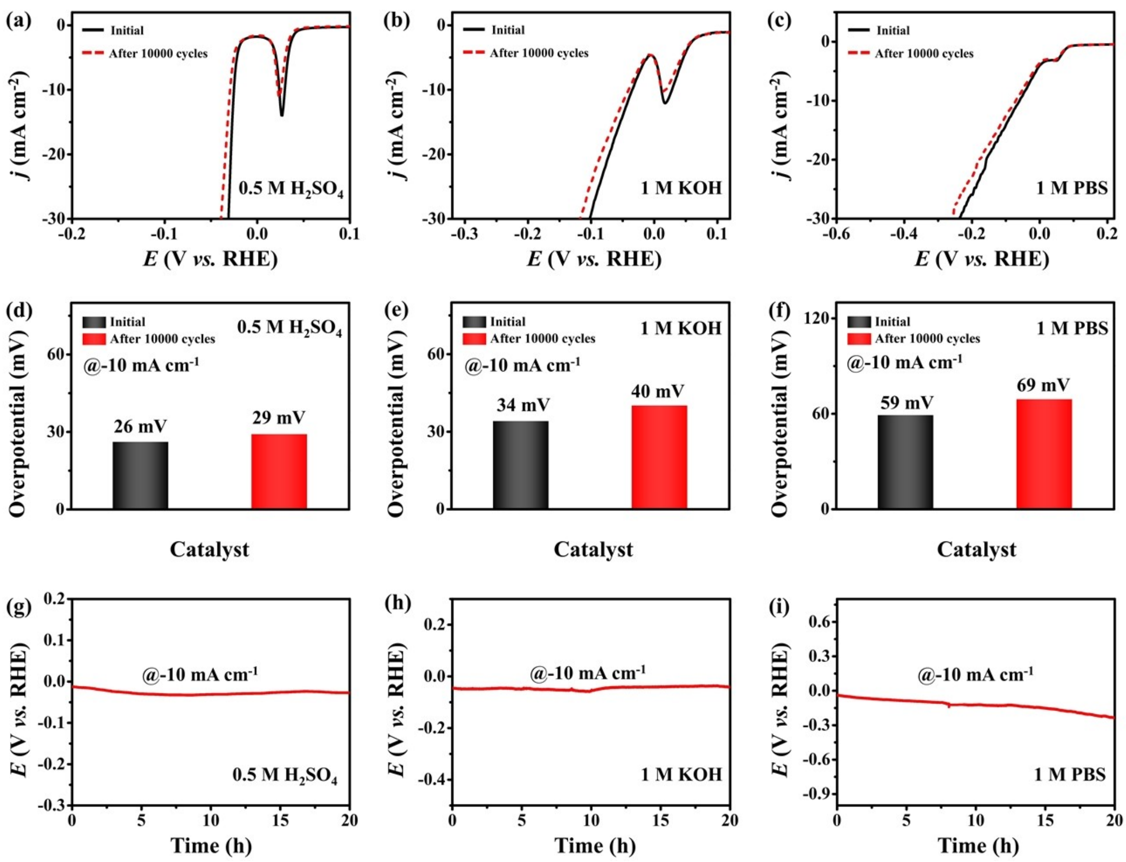


Fig. S9 (a, b, and c) HER polarization curves of the N-PdIr bimetallene before and after 10000 cycles in acidic, alkaline, and neutral electrolytes, respectively. (d, e, and f) Comparison of the required overpotentials of the N-PdIr bimetallene before and after 10000 cycles at current density of -10 mA cm^{-2} in acidic, alkaline, and neutral electrolytes, respectively. (g, h, and i) Chronopotentiometry curves of the N-PdIr bimetallene under the cathodic current density of 10 mA cm^{-2} in acidic, alkaline, and neutral electrolytes, respectively.

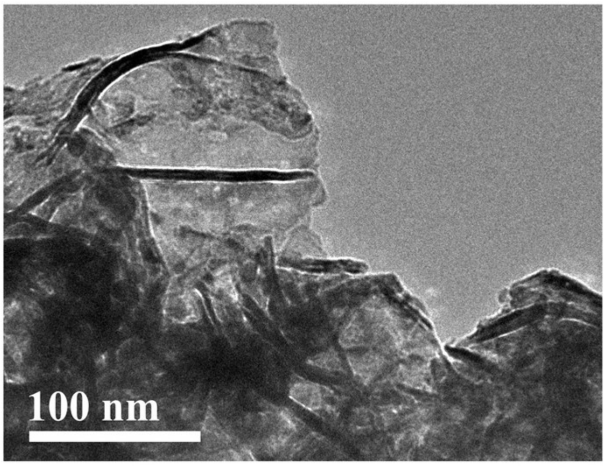


Fig. S10 TEM image of the N-PdIr bimetallene after catalytic stability testing.

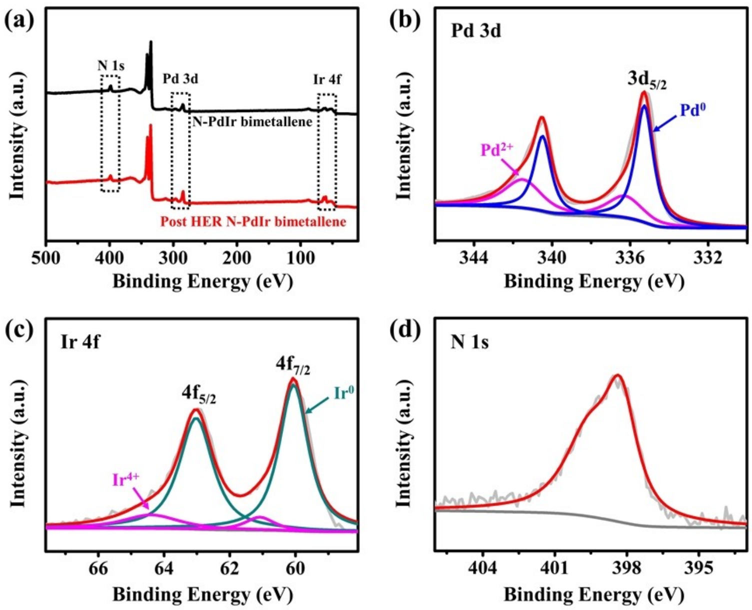


Fig. S11 (a) XPS survery spectra of inital and post-HER N-PdIr bimetallene catalysts. (b) Pd 3d, (c) Ir 4f and (d) N 1s XPS spectra for post-HER N-PdIr bimetallene after stability testing.

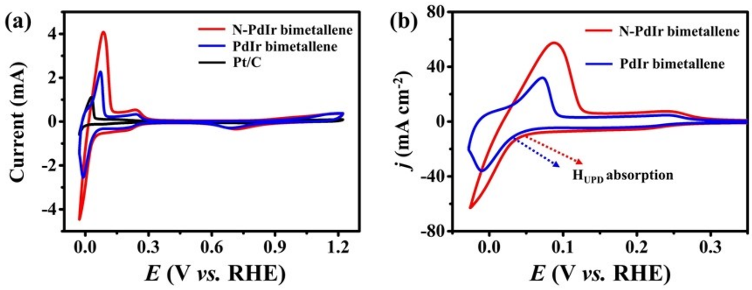


Fig. S12 (a) CV curves for various catalysts recorded in 0.5 M H_2SO_4 with a scan rate of 50 mV s^{-1}. (b) The regions between -0.05 and 0.35 V from (a).

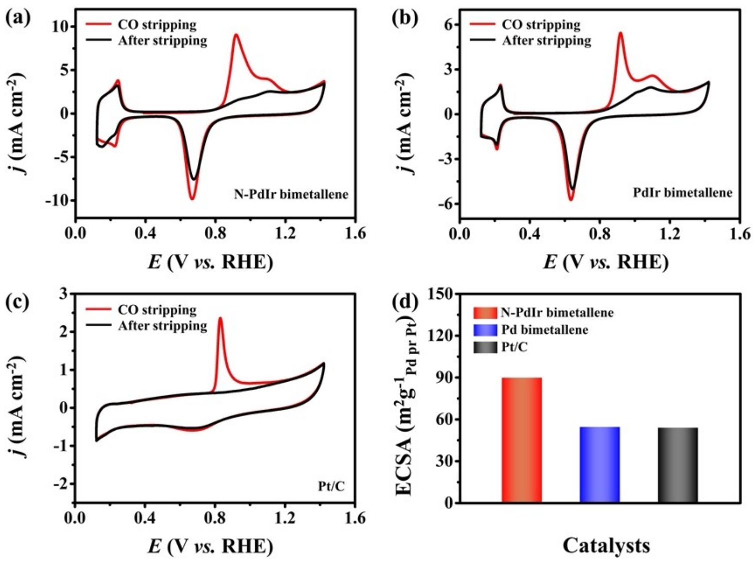


Fig. S13 CO stripping measurements of (a) N-PdIr bimetallene, (b) PdIr bimetallene and (c) Pt/C in 0.5 M H_2SO_4 at a scan rate of 20 mV s^{-1}. (d) the corresponding ECSA.

Table S1. Comparison of HER activities in a 0.5 M H_2SO_4 for N-PdIr bimetallene with other reported electrocatalysts.

Catalysts	Overpotential at 10 mA cm^{-2} (mV)	Electrolyte	Ref.
N-PdIr bimetallene	**26**	**0.5 M H_2SO_4**	**This work**
Pd-CN_X	55	0.5 M H_2SO_4	[1]
Pd-CoS_2-MoS_2/C-600	144	0.5 M H_2SO_4	[2]
hollow RhCo nanoparticles	28	0.5 M H_2SO_4	[3]
$Rh_3Pb_2S_2$ nanocage	87	0.5 M H_2SO_4	[4]
PdCu@Pd NCs	68	0.5 M H_2SO_4	[5]
Rh/F-graphene-2 catalysts	46	0.5 M H_2SO_4	[6]
$Pd_{83.5}Ir_{16.5}$ nanocatalyst	73	0.5 M H_2SO_4	[7]
Ru_2P nanoparticles	55	0.5 M H_2SO_4	[8]
$Mo_2TiC_2T_X$–PtSA	30	0.5 M H_2SO_4	[9]
Li-PPS ND catalyst	91	0.5 M H2SO4	[10]

Table S2. Comparison of HER activities in a 1 M KOH for N-PdIr bimetallene with other reported electrocatalysts.

Catalysts	Overpotential at 10 mA cm^{-2} (mV)	Electrolyte	Ref.
N-PdIr bimetallene	**34**	**1 M KOH**	**This work**
Ru_2P nanoparticles	50	1 M KOH	[8]
PtNi/CNFs	82	1 M KOH	[11]
Pd-Pt-S	71	1 M KOH	[12]
Mn-Co-P/Ti	76	1 M KOH	[13]
$NiCo_2S_4$/Pd	57	1 M KOH	[14]
Pt_2Ni_3-P NWs	44	1 M KOH	[15]
Ni_3N/Pt	50	1 M KOH	[16]
PtPd@NLS	46	1 M KOH	[17]
Rh NSs	43	1 M KOH	[18]
PtNi-O nanostructure	40	1 M KOH	[19]

Table S3. Comparison of HER activities in a 1 M PBS for N-PdIr bimetallene with other reported electrocatalysts.

Catalysts	Overpotential at 10 mA cm^{-2} (mV)	Electrolyte	Ref.
N-PdIr bimetallene	**59**	**1 M PBS**	**This work**
RhCoB aerogel	113	1 M PBS	[20]
OsP_2@NPC	54	1 M PBS	[21]
PdP_2/CB	85	1 M PBS	[22]
Pt@NOMC-A	65	1 M PBS	[23]
Ru@Co-SAs/N-C	55	1 M PBS	[24]
Pd-TiO_2	155	1 M PBS	[25]

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