

Supporting Information

Entropy-Driven Durability Enhancement of PtM₃
(M=Transition Metal) Type Alloy Catalysts for
Oxygen Reduction Reaction

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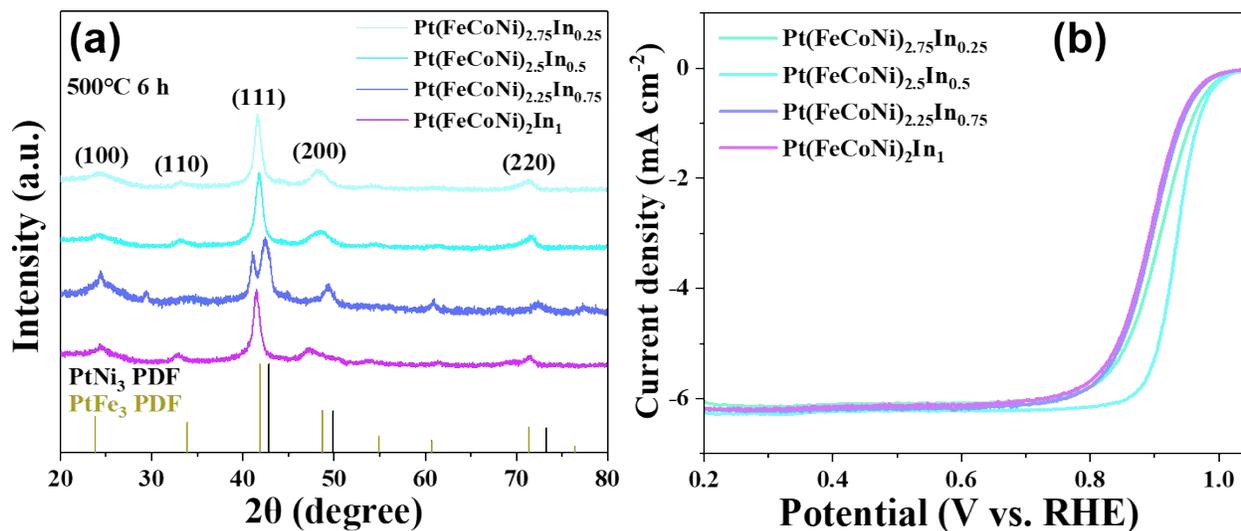


Figure S1. (a) XRD patterns of $\text{Pt}(\text{FeCoNi})_{3-x}\text{In}_x$ ($x=0.25, 0.5, 0.75, 1$) annealed at 500°C for 6 h. (b) LSV curves of as-prepared $\text{Pt}(\text{FeCoNi})_{3-x}\text{In}_x$ ($x=0.25, 0.5, 0.75, 1$) tested in O_2 -saturated $0.1 \text{ mol L}^{-1} \text{ HClO}_4$ at a rotation speed of 1,600 rpm.

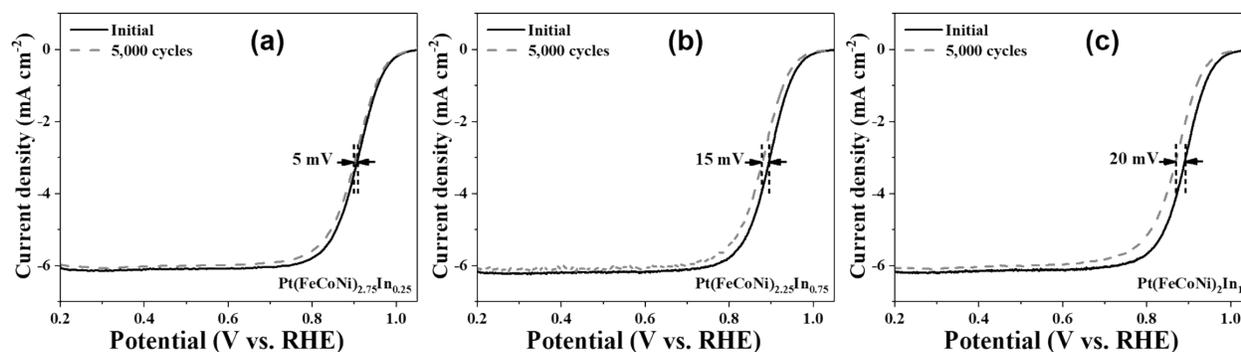


Figure S2. LSV curves of $\text{Pt}(\text{FeCoNi})_{3-x}\text{In}_x$ ($x=0.25, 0.75, 1$) at the beginning and after 5,000 ADT cycles, tested in O_2 -saturated $0.1 \text{ mol L}^{-1} \text{ HClO}_4$ at a rotation speed of 1,600 rpm: (a) $x=0.25$; (b) $x=0.75$; (c) $x=1$;

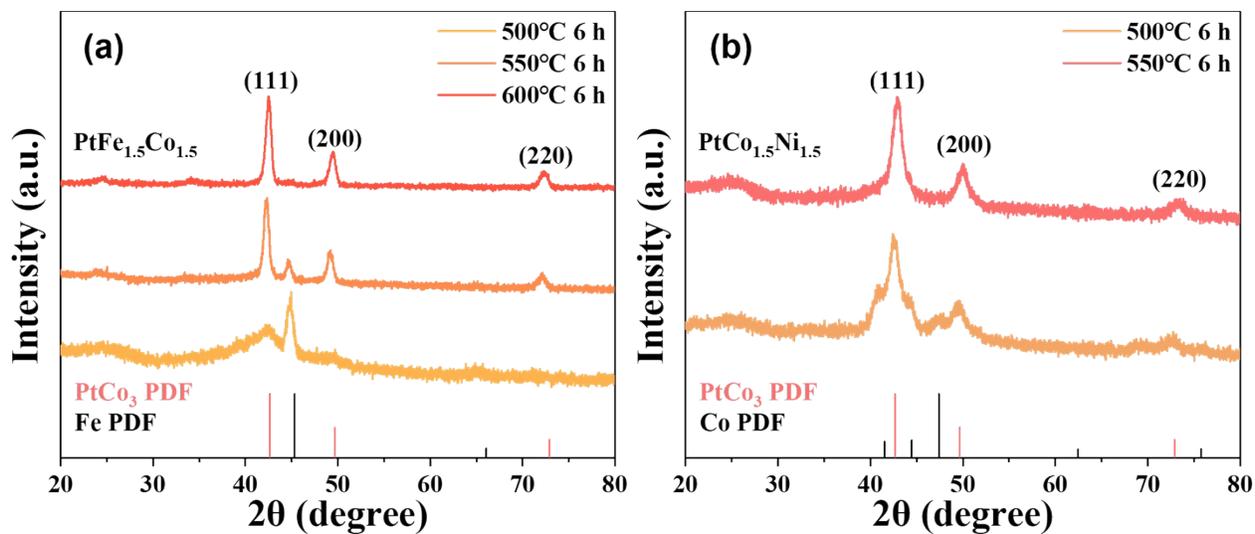


Figure S3. XRD patterns of (a) PtFe_{1.5}Co_{1.5} and (b) PtCo_{1.5}Ni_{1.5} annealed at different temperatures. PDF cards: PtCo₃ (PDF #96-152-4154), Co (PDF #04-007-7985), Fe (PDF #04-016-2968)

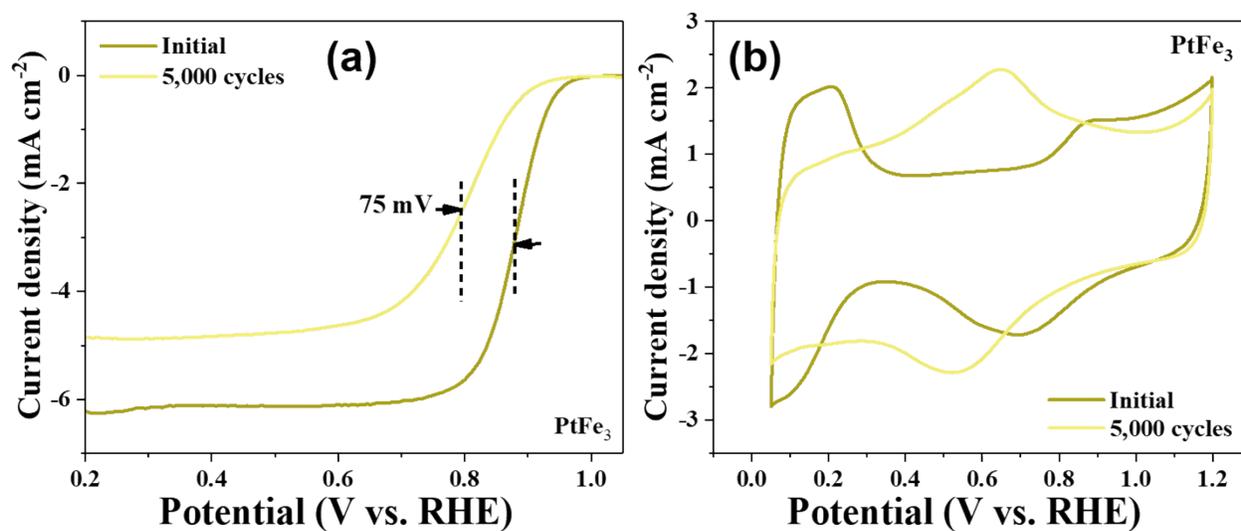


Figure S4. RDE test results of PtFe₃ in 0.1 mol L⁻¹ HClO₄ at the beginning and after different cycles: (a) LSV curves; (b) CV curves.

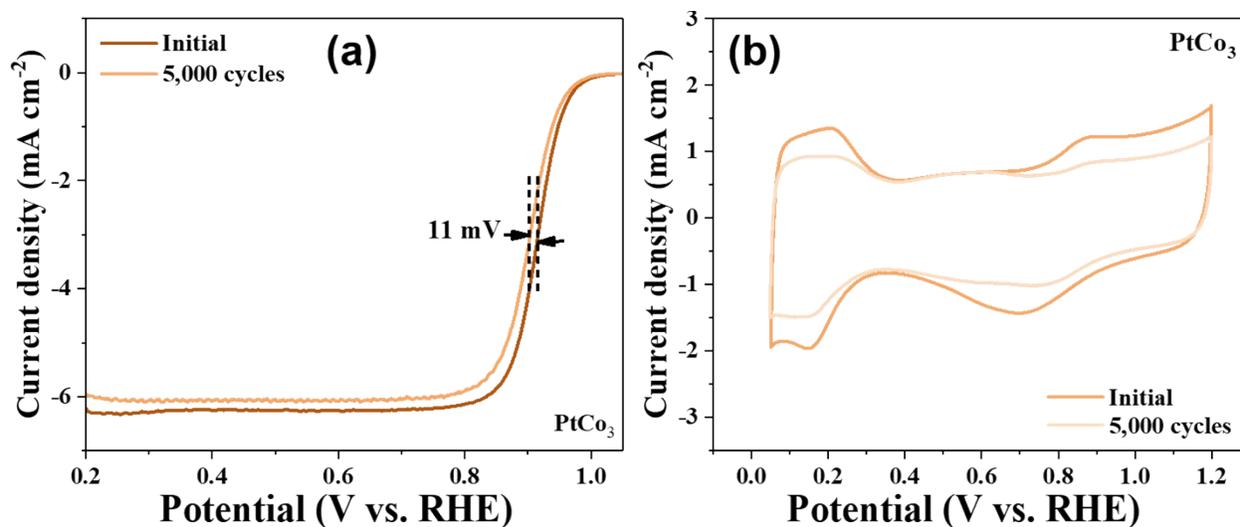


Figure S5. RDE test results of PtCo₃ in 0.1 mol L⁻¹ HClO₄ at the beginning and after different cycles: (a) LSV curves; (b) CV curves.

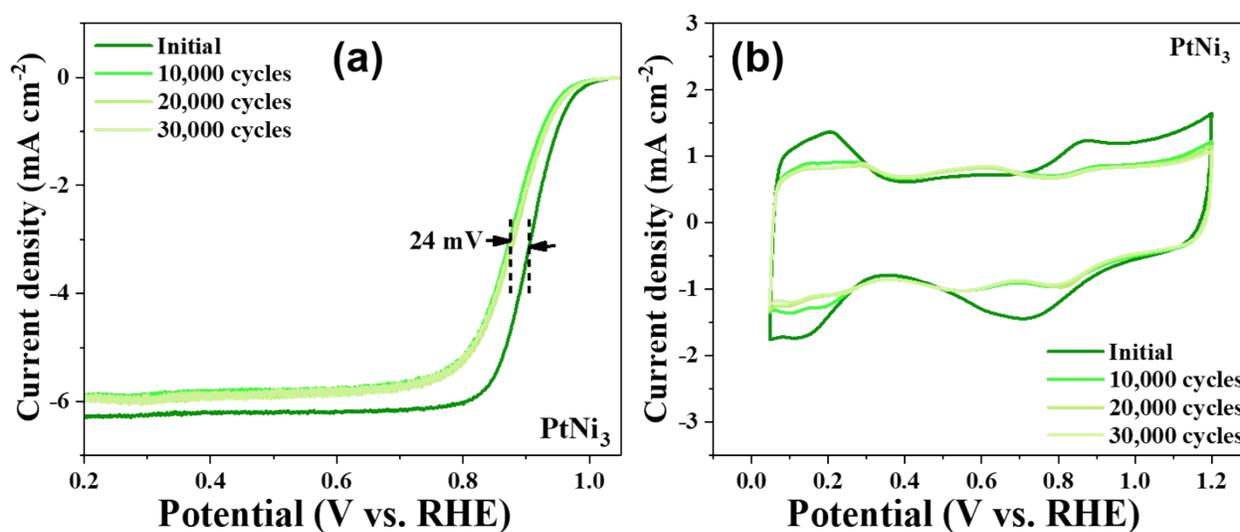


Figure S6. RDE test results of PtNi₃ in 0.1 mol L⁻¹ HClO₄ at the beginning and after different cycles: (a) LSV curves; (b) CV curves.

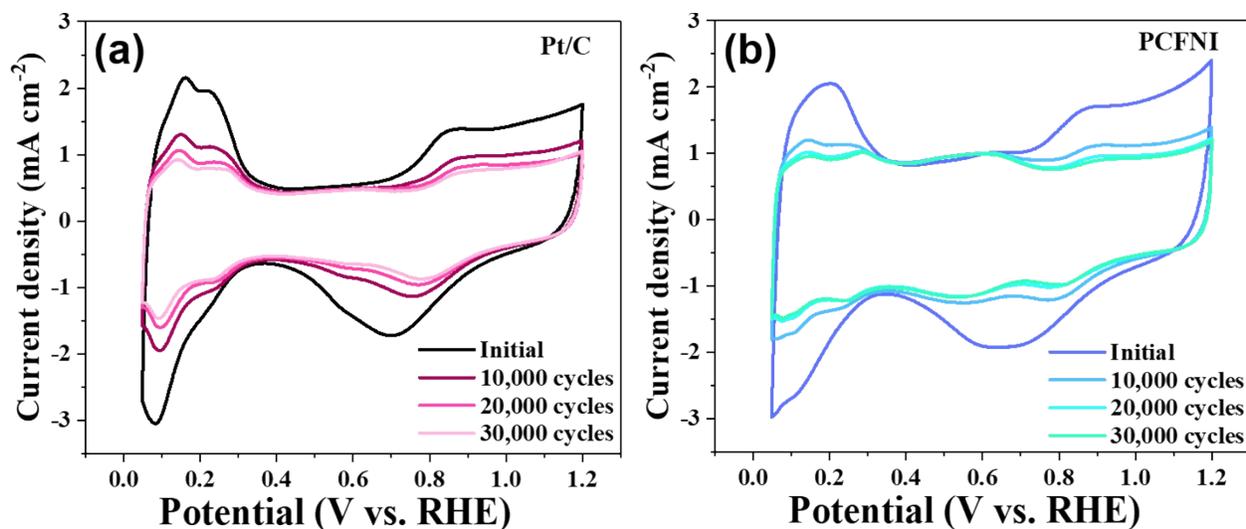


Figure S7. CV curves of (a) commercial Pt/C and (b) PCFNI tested in N_2 -saturated $0.1 \text{ mol L}^{-1} \text{ HClO}_4$ at the beginning and after different cycles.

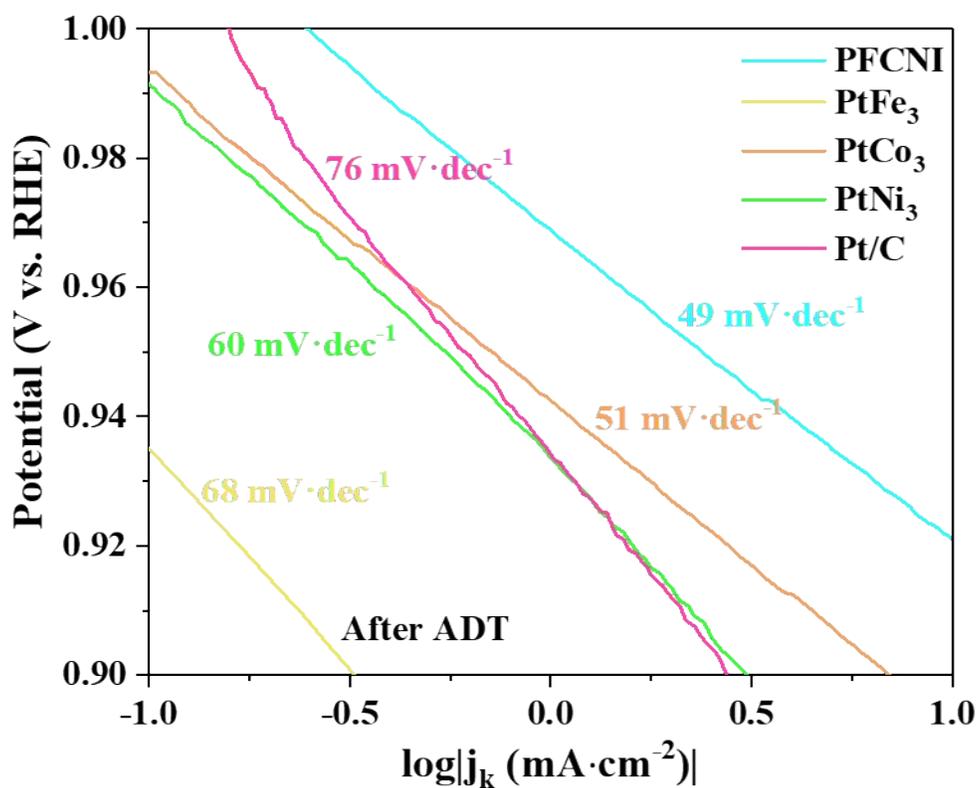


Figure S8. Tafel plots of PFCNI, PtFe_3 , PtCo_3 , PtNi_3 and commercial Pt/C tested in $0.1 \text{ mol L}^{-1} \text{ HClO}_4$ after 30,000 ADT cycles.

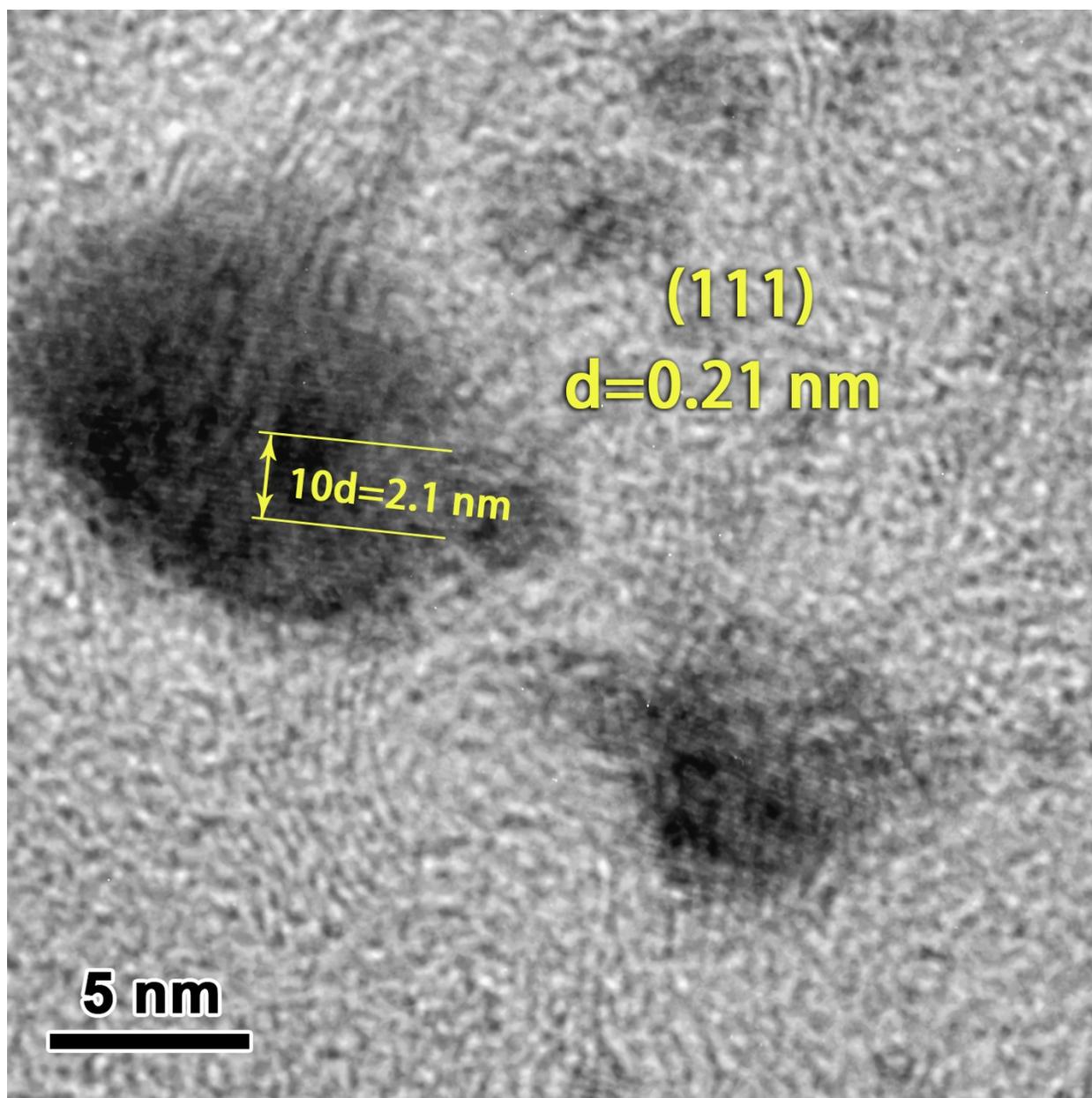


Figure S9. High-resolution TEM image of as-prepared PtNi₃

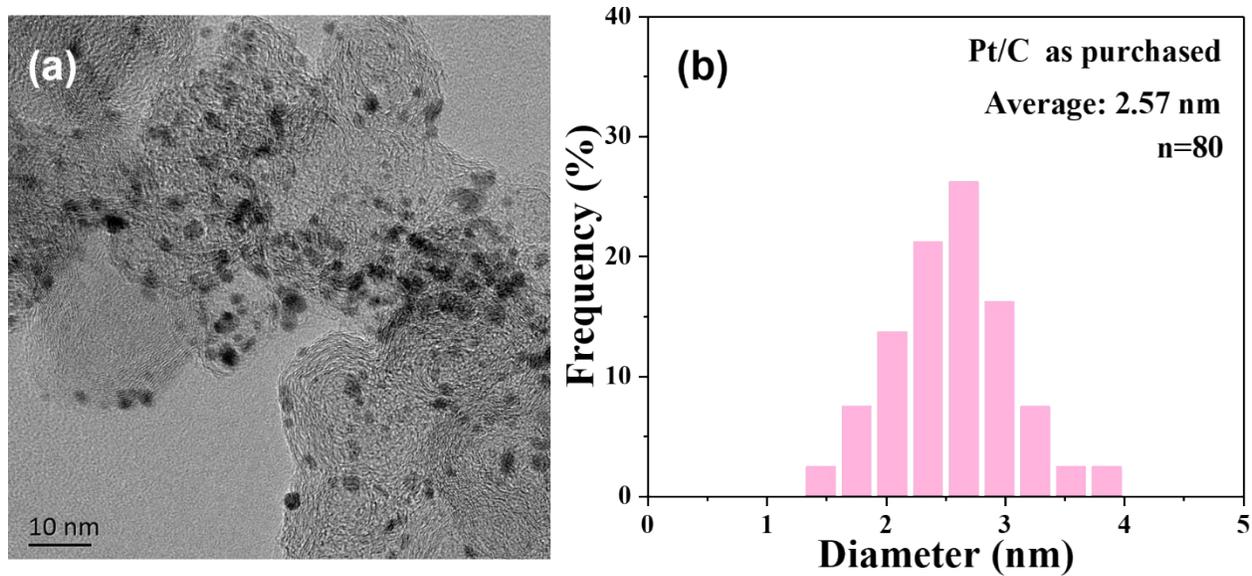


Figure S10. TEM image and particle size distribution of commercial Pt/C (as purchased)

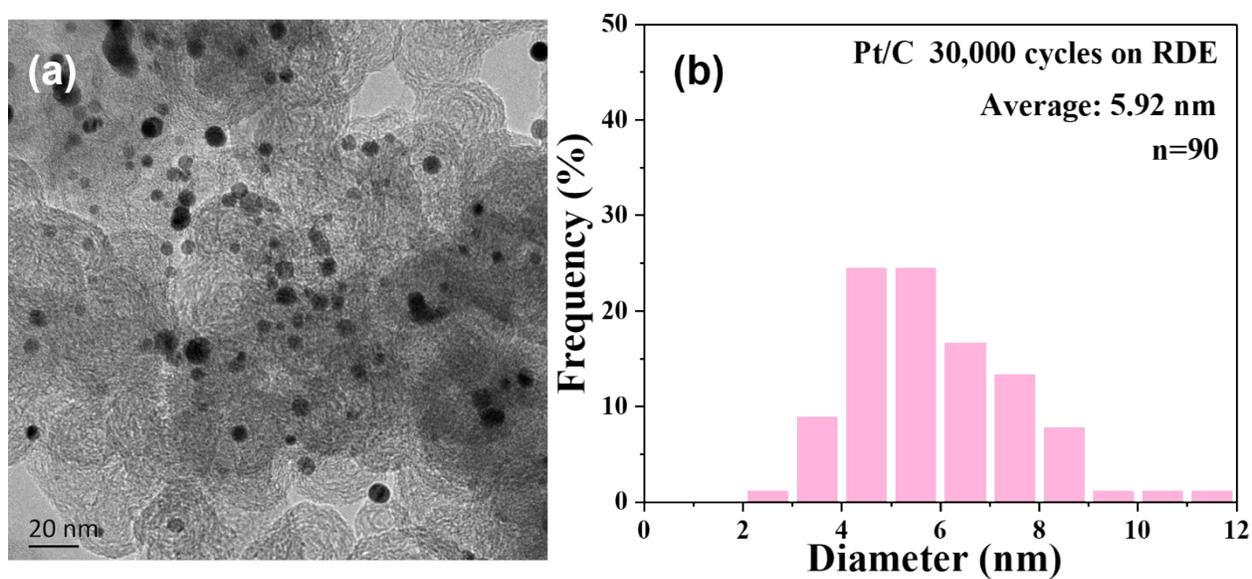


Figure S11. TEM image and particle size distribution of commercial Pt/C after 30000 cycles on RDE

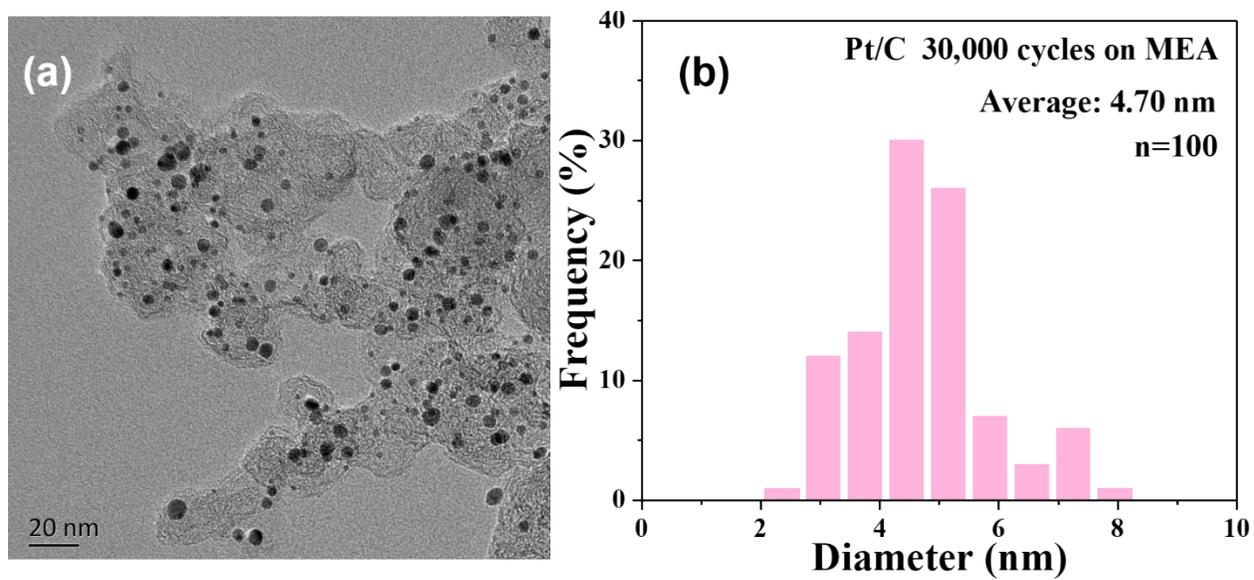


Figure S12. TEM image and particle size distribution of commercial Pt/C after 30000 cycles on MEA

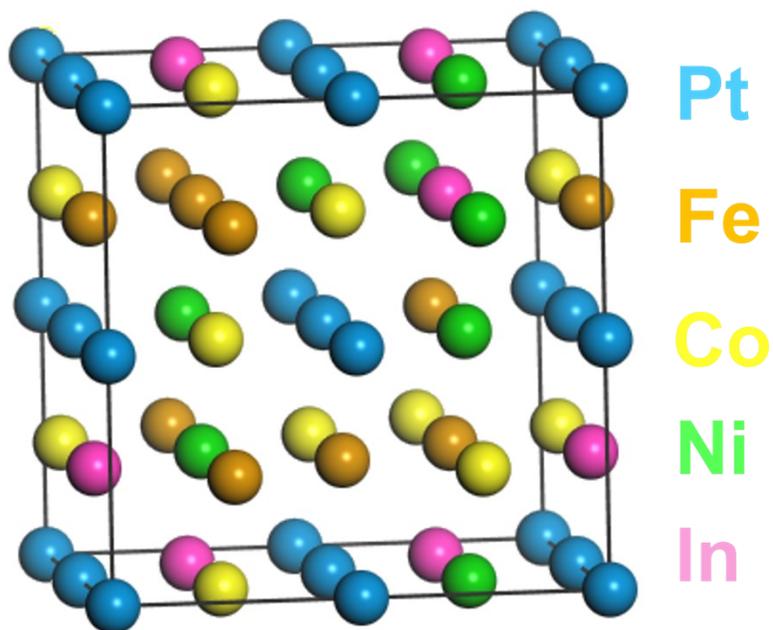


Figure S13. The 2x2x2 supercell model of PFCNI

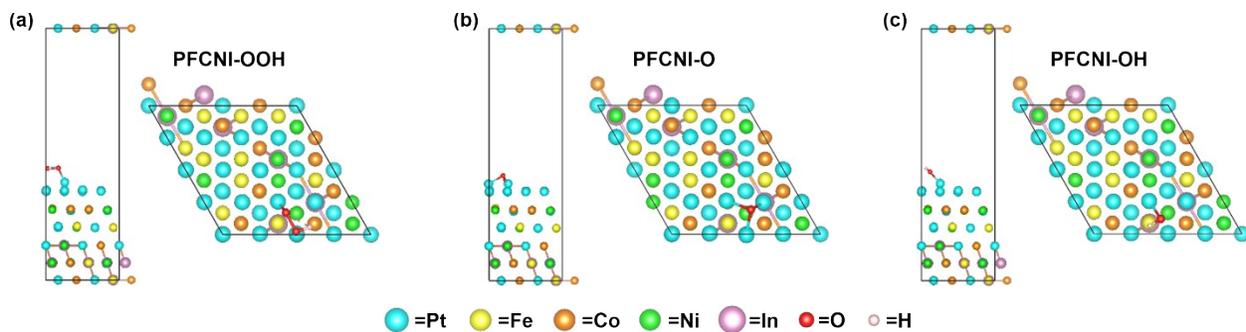


Figure S14. DFT models for surface adsorption calculation of PFCNI from the view of X and Z axis: (a) OOH*;
(b) O*;
(c) OH*

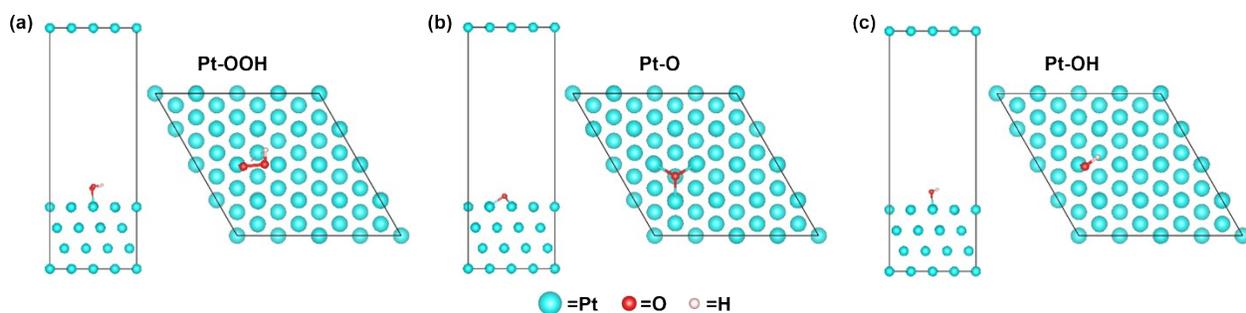


Figure S15. DFT models for surface adsorption calculation of Pt (111) from the view of X and Z axis: (a) OOH*;
(b) O*;
(c) OH*

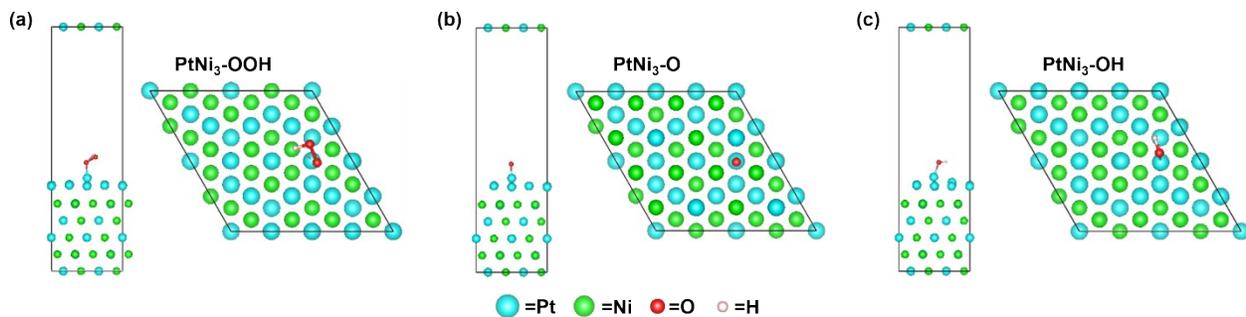


Figure S16. DFT models for surface adsorption calculation of PtNi₃ from the view of X and Z axis: (a) OOH*;
(b) O*;
(c) OH*

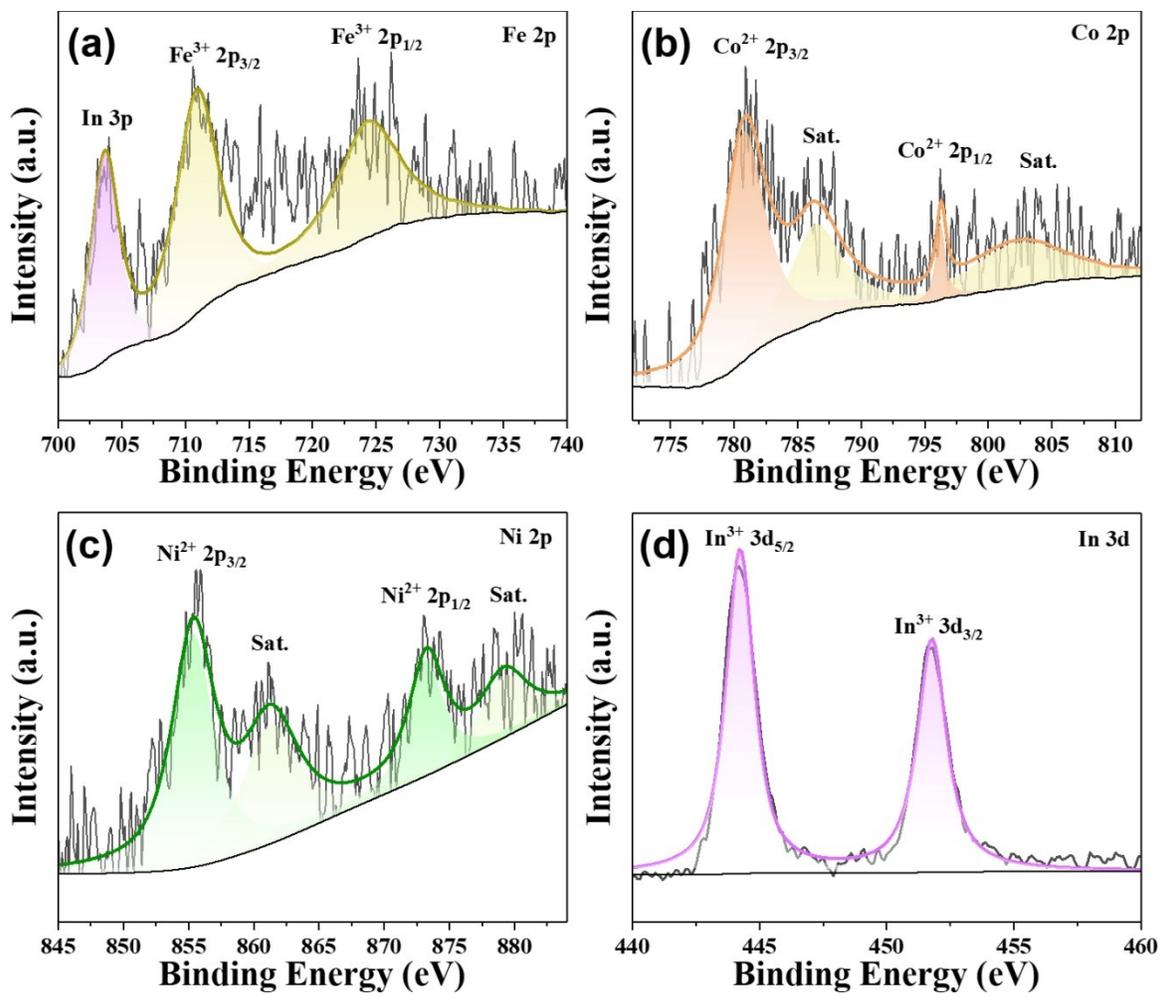


Figure S17. XPS spectra of as-prepared PFCNI measured at: (a) Fe 2p; (b) Co 2p; (c) Ni 2p; (d) In 3d

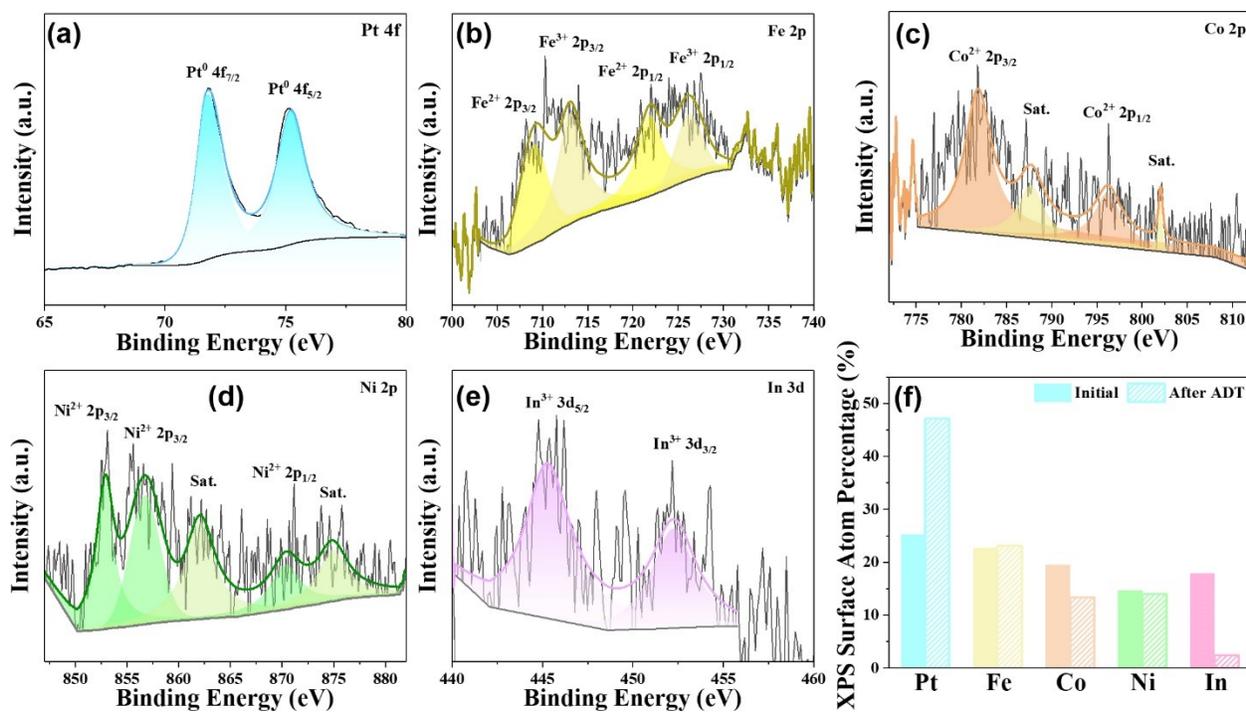


Figure S18. XPS spectra of after-cycling PFCNI measured at: (a) Pt 4f; (b) Fe 2p; (c) Co 2p; (d) Ni 2p; (e) In 3d; and (f) the comparison of XPS surface atom percentage (%) before and after cycling.

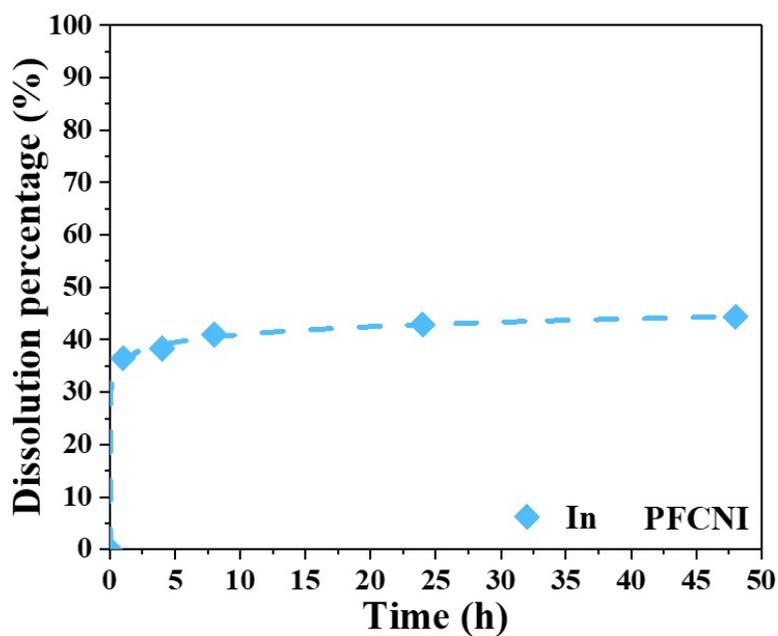


Figure S19. The corresponding In dissolution percentage measured by ICP-AES, with the as-prepared PFCNI soaked in 1 mol L⁻¹ HNO₃ and sampled at 1 h, 4 h, 8 h, 24 h, 48 h

Table S1. Comparison of PtM₃ type ORR catalysts' electrochemical stability on RDE in 0.1 mol L⁻¹ HClO₄ solution.

Catalyst (Ref.)	Initial mass activity (A mg _{Pt} ⁻¹)	ADT cycles	Mass activity preservation
PtNi ₃ ¹	About 0.5~0.9	10,000	About 50%~70%
PtNi ₃ ²	About 0.7~0.85	10,000	About 60~70%
PtCo ₃ ²	About 0.65~0.75	10,000	About 65~80%
PtFe ₃ ²	About 0.35~0.7	10,000	About 60~85%
O-PtCo ₃ @HNCS ³	0.54	30,000	90.1%
Pt(Fe, Co, Ni) ₃ /C-700 ⁴	0.32	10,000	83%
HIFs Pt@PtCu ₃ ⁵	0.36	15,000	83%
O-PtCo ₃ /C-600 ⁶	0.545	10,000	96.5%
O-PtCo ₃ /C-700 ⁶	0.447	10,000	97.5%
D-PtCo ₃ /C ⁶	0.413	10,000	81.4%
PtRhNiFeCu/C ⁷	1.23	10,000	75.8%
PtPdFeCoNi ⁸	1.23	30,000	82.1%
PtCoNiGaZn@Pt/C ⁹	0.65	20,000	73.0%

Pt(FeCoNiCuZn) ₃ /C ¹⁰	0.70	30,000	97.1%
PtNi ₃ (This work)	0.232	30,000	41.4%
PFCNI (This work)	1.04	30,000	85.7%

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