Simultaneously Tackling Discrete Island Effects and Interfacial Resistance in PEM Electrolyzers via a Scalable Bilayer Catalyst Design

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Keywords: Proton exchange membrane electrolyzer, Discrete island effects , Interfacial resistance, Uncoated PTL, Bilayer anode.

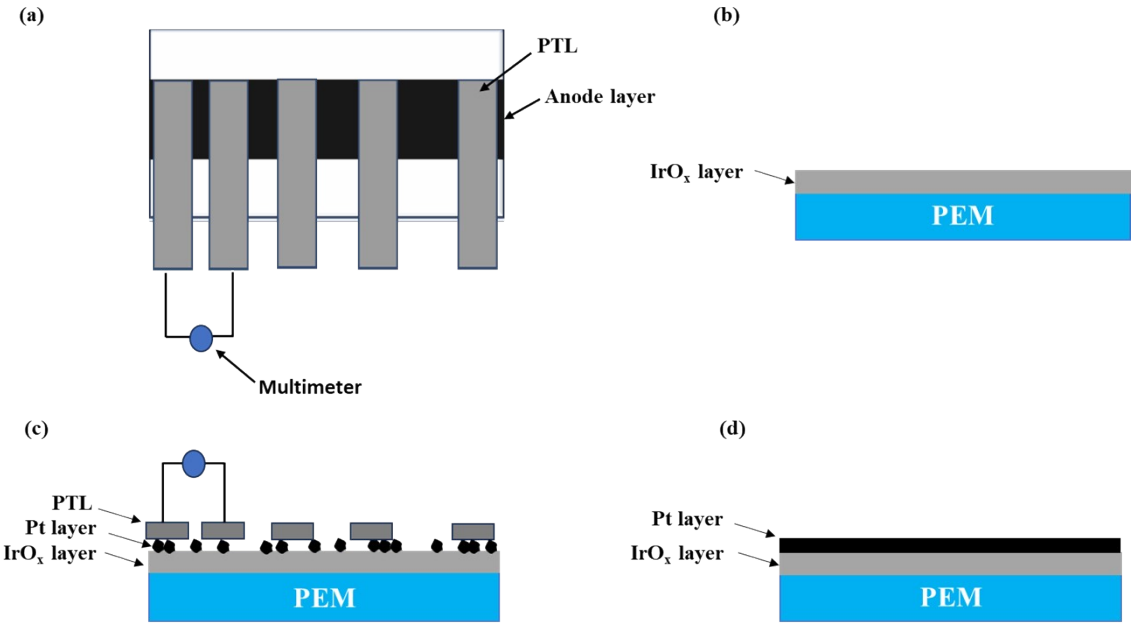

Fig. S1. (a) The schematic of in-plane resistivity measurement method. The anode cross section morphology of (b) Ir1mg; (c) Ir1mgPtYmgB (Y< 0.3); (d) Ir1mgPtYmgB (Y≥0.3).

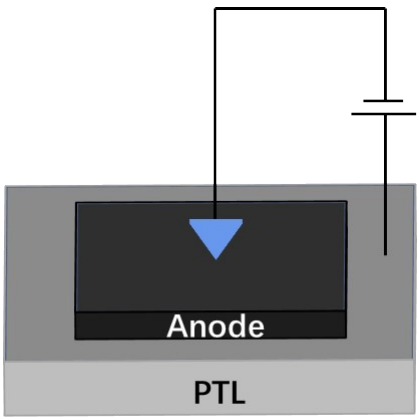

Fig. S2. Testing schematic of the voltammetric curve for the anode /PTL assembly.

Supplementary Note 1:

The contribution of the voltage loss is shown in Fig. S4. The voltage loss consists of kinetics, Ohmic, and mass transfer (MT) sections described in the following equation (S1):

$$E_{cell} = E_{rev} + \eta_{kinetic} + \eta_{Ohmic} + \eta_{MT} \qquad \text{(S1)}$$

Where E_{rev} is the reversible cell voltage, and $\eta_{kinetic}$, η_{Ohmic} and η_{MT} is kinetics, Ohmic, and MT overpotential.

E_{rev} is calculated by the Nernst equation (S2):

$$E_{rev} = E_0 + \frac{RT}{2F}\left[\frac{a(H_2)\cdot\sqrt{a(O_2)}}{a(H_2O)}\right] \qquad \text{(S2)}$$

Where E_0 is standard reversible potential, R is the gas constant, T is temperature, F is the Faraday constant. At 80°C, ambient pressure (1 bar), the saturation pressure of H_2O is 0.47 bar. So $a(H_2O)$ is value to 0.47. While $a(H_2)$ and $a(O_2)$ is equal to $\frac{1\ bar - 0.47\ bar}{1\ bar}$.

$\eta_{kinetic}$ is calculated by the Butler-Volmer equation (S3):

$$\eta_{kinetic} = \frac{RT}{\alpha F}\cdot ln\frac{i}{i_0} \qquad \text{(S3)}$$

Where alpha is the transfer coefficient, i_0 is exchange current density.

η_{Ohmic} is calculated by the following equation (S4):

$$\eta_{Ohmic} = i \cdot R_{Ohmic} \qquad \text{(S4)}$$

Where R_{Ohmic} is equal to the high-frequency resistance (HFR, the high frequency intercept of the Nyquist plot with the real axis) tested by electrochemical impedance spectroscopy (EIS).

The remaining overpotential is treated as η_{MT}.

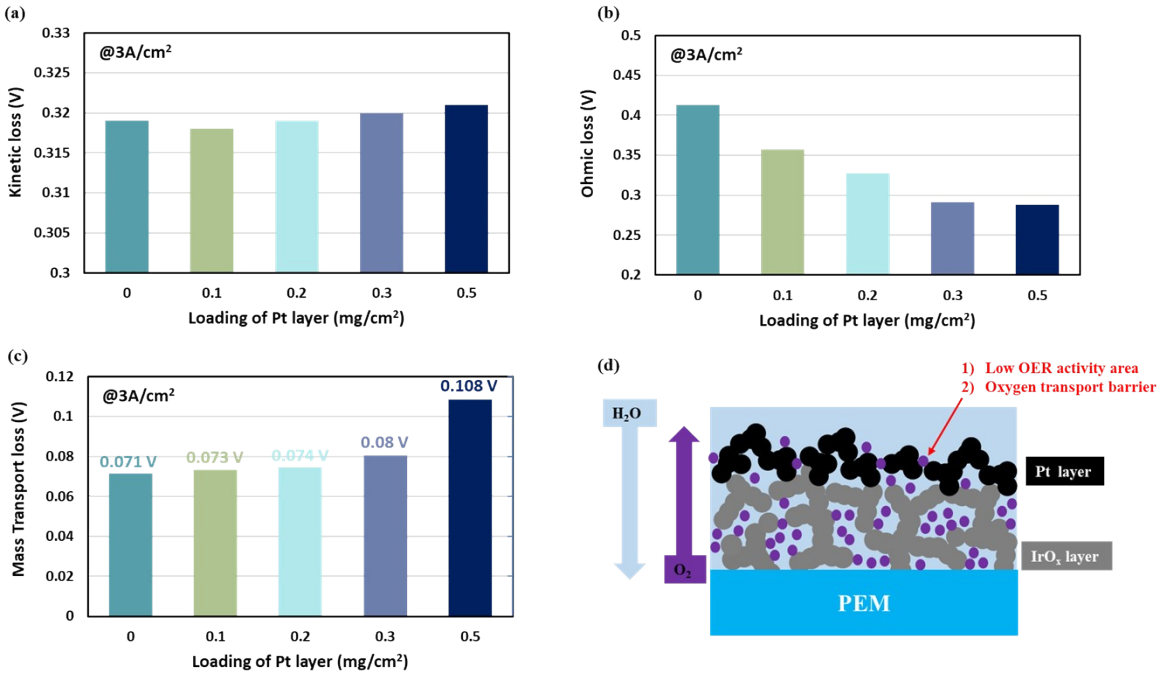

Fig. S3. (a) Kinetic loss; (b) Ohmic loss and (c) Mass Transport loss @ 3 A/cm^2 in Ir1mgPtYmg/UC-PTL (Y= 0 ,0.1 ,0.2, 0.3 and 0.5). (d) Schematic of anode water-gas mass transfer in Pt-IrO_x bilayer.

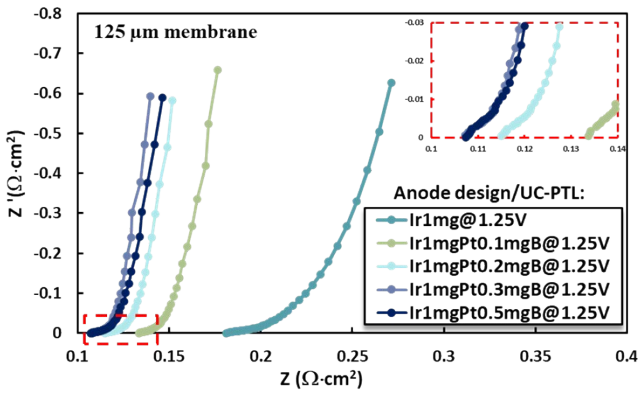

Fig. S4. EIS plot for Ir1mgPtYmg/UC-PTL (Y= 0, 0.1, 0.2, 0.3, and 0.5) at 1.25 V.

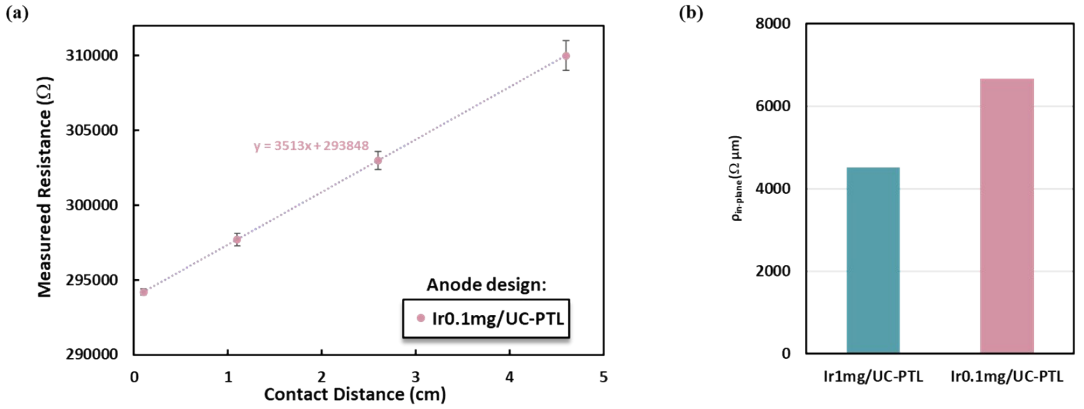

Fig. S5. (a) Measured resistance for Ir0.1mg/UC-PTL. (b) in-plane resistivity for Ir1mg/UC-PTL and Ir0.1mg/UC-PTL.

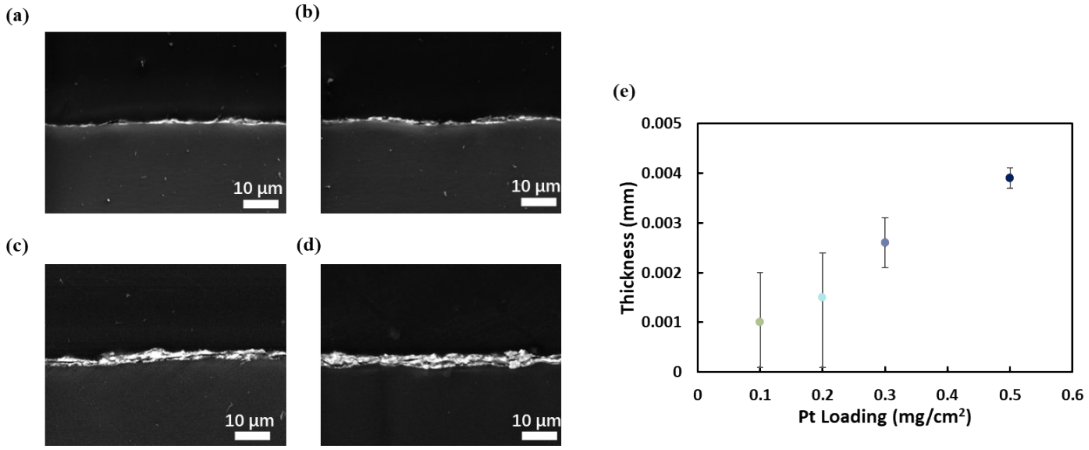

Fig. S6. SEM cross section image for (a) Pt0.1mg; (b) Pt0.2mg; (c) Pt0.3mg and (d) Pt0.5mg. The anode thickness of PtYmg (Y=0.1, 0.2, 0.3 and 0.5).

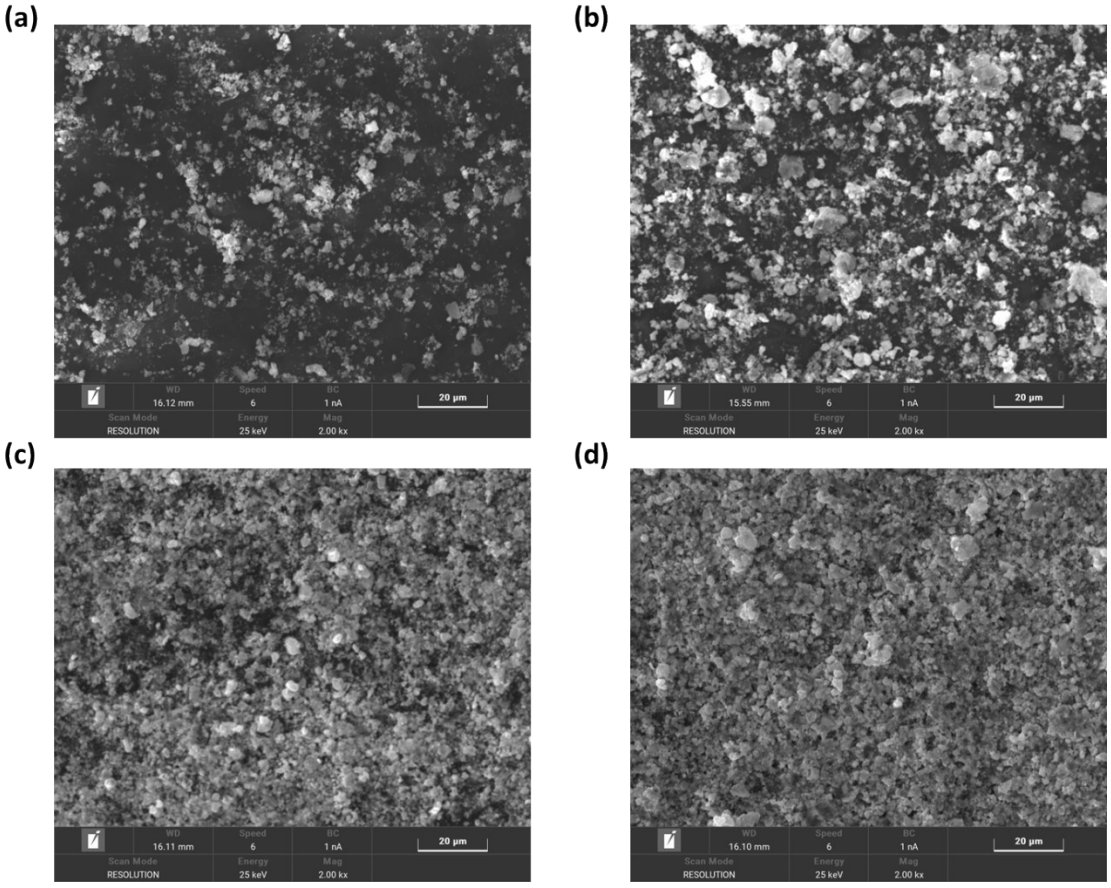

Fig. S7. SEM top-view image for (a) Pt0.1mg; (b) Pt0.2mg; (c) Pt0.3mg and (d) Pt0.5mg

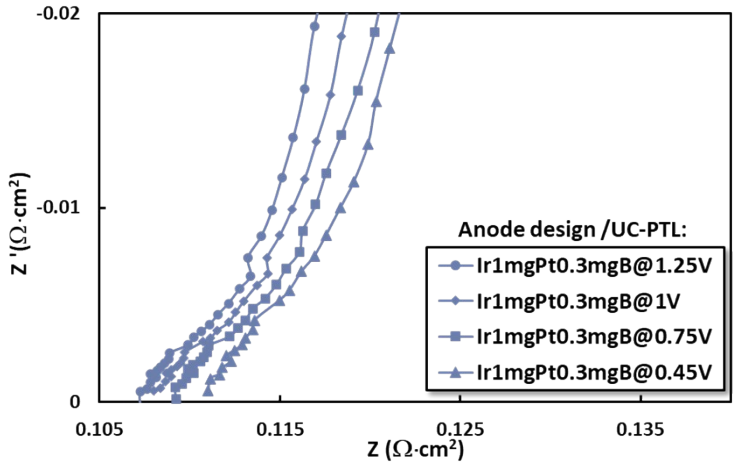

Fig. S8. EIS plot for Ir1mgPt0.3mgB at different voltages.

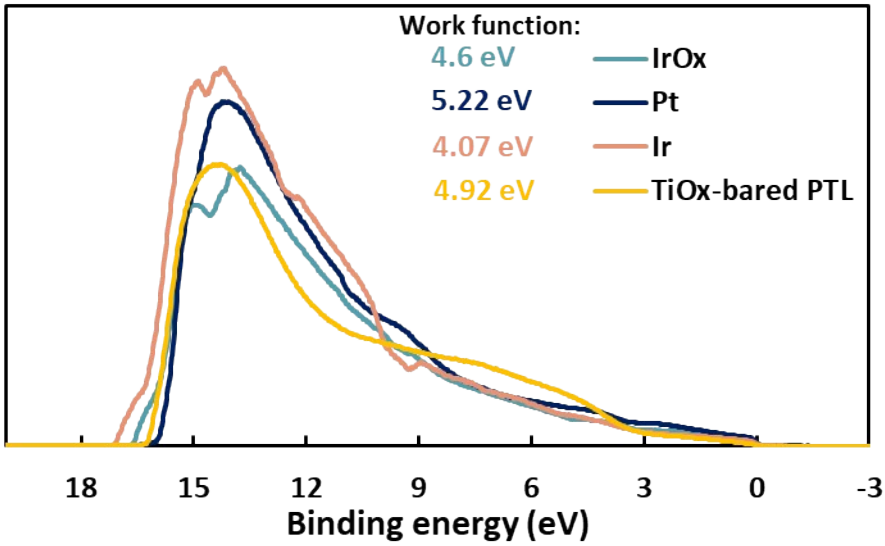

Fig. S9. UPS analysis for IrO_x, Pt, Ir and TiO_x-bared PTL.

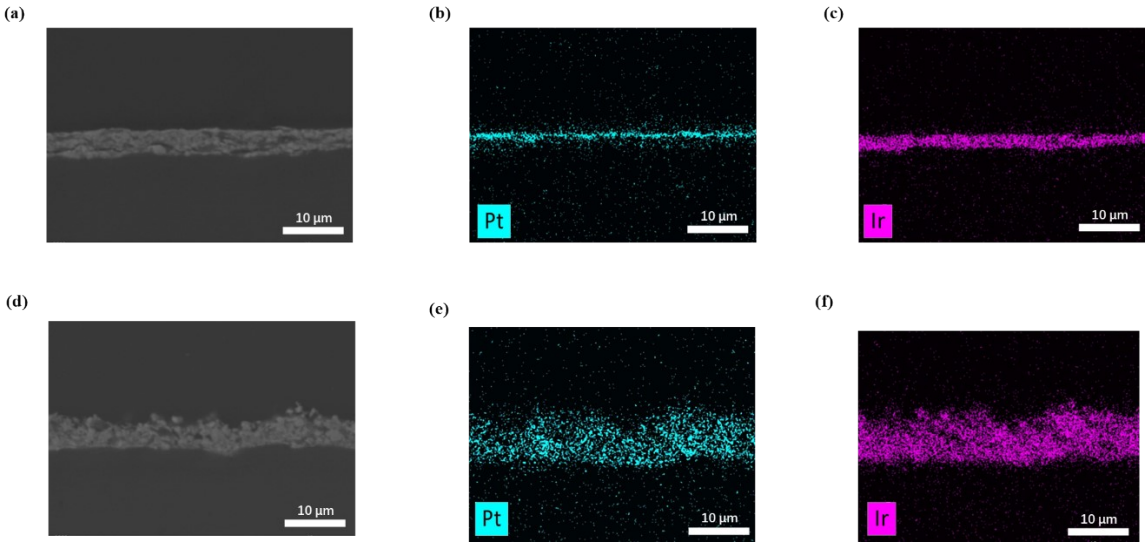

Fig. S10. SEM cross section image for (a) Ir1mgPt0.3mgB and (d) Ir1mgPt0.3mgM. EDS elemental mapping showing the distribution of Pt for (b) Ir1mgPt0.3mgB and (e) Ir1mgPt0.3mgM. EDS elemental mapping showing the distribution of Ir for (c) Ir1mgPt0.3mgB and (f) Ir1mgPt0.3mgM.

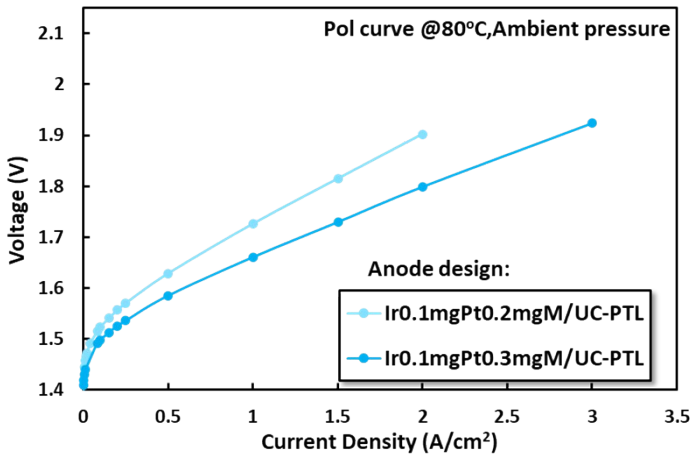

Fig. S11. Polarization curves for Ir0.1mgPt0.2mgM/UC-PTL and Ir0.1mgPt0.3mgM/UC-PTL.

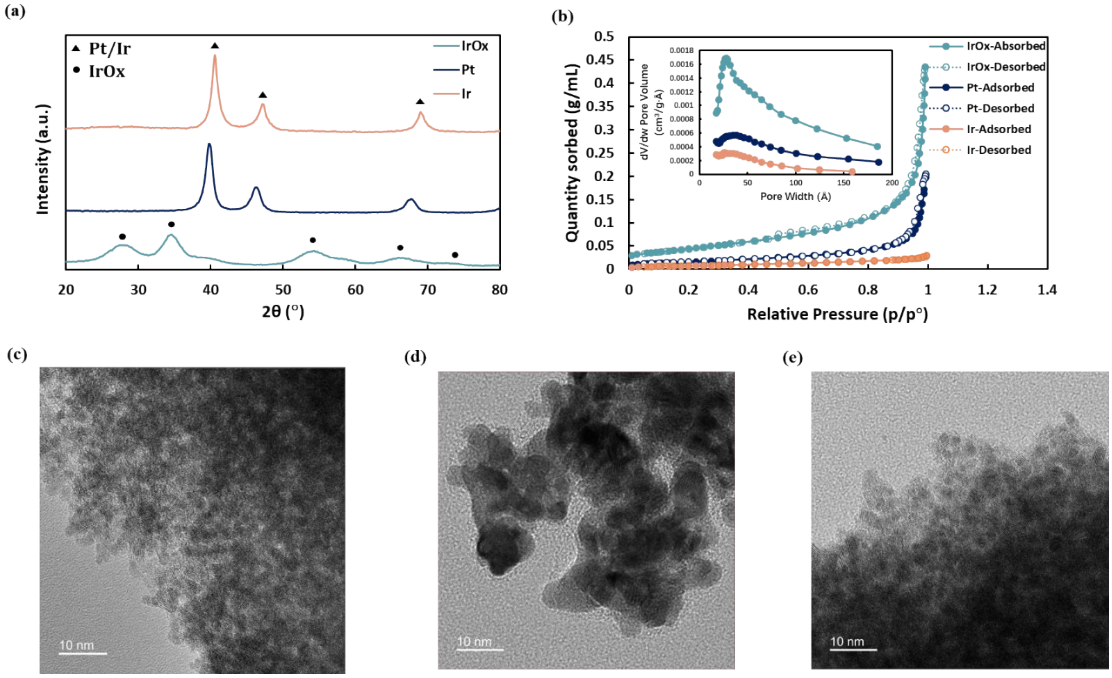

Fig. S12. (a) XRD patterns and (b) Nitrogen adsorption and desorption isotherm (inset shows the BJH pore size distribution (PSD)) for IrO_x, Pt and Ir catalyst. TEM images for (c) IrO_x; (d) Pt and (e) Ir catalyst.

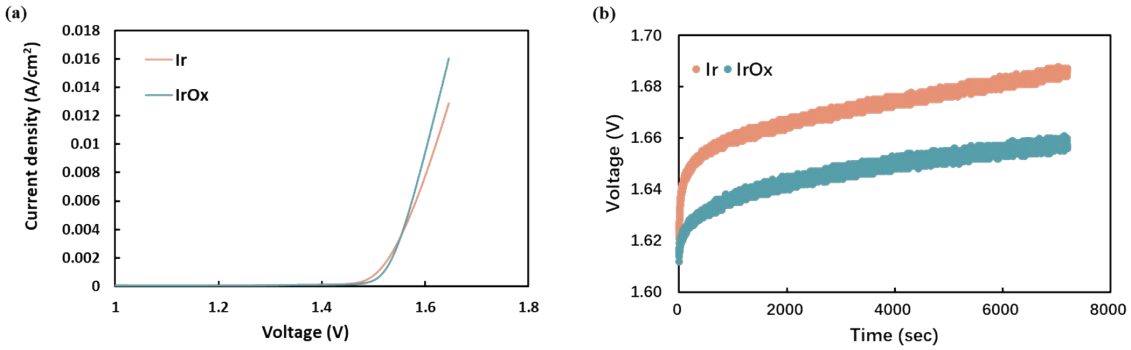

Fig. S13. Electrocatalytic OER performance for Ir and IrO_x catalyst in three-electrode system(a) Linear sweep voltammetry curves; (b) chronopotentiometry test.

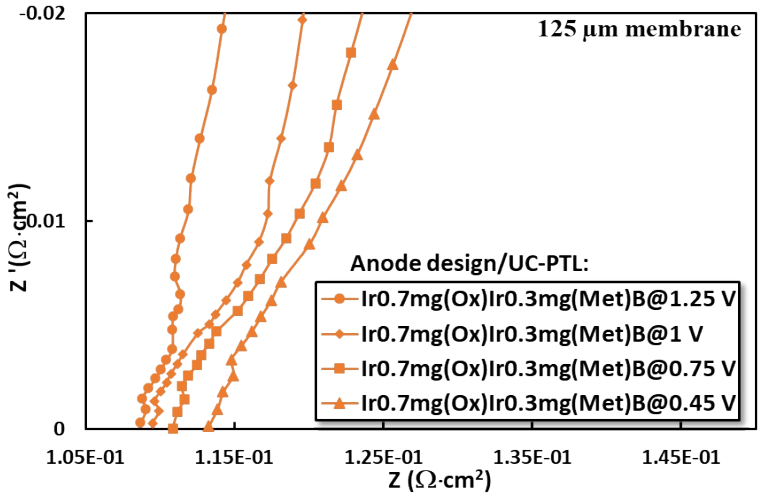

Fig. S14. EIS plot for Ir0.7mg(Ox)Ir0.3mg(Met)B at different voltages.

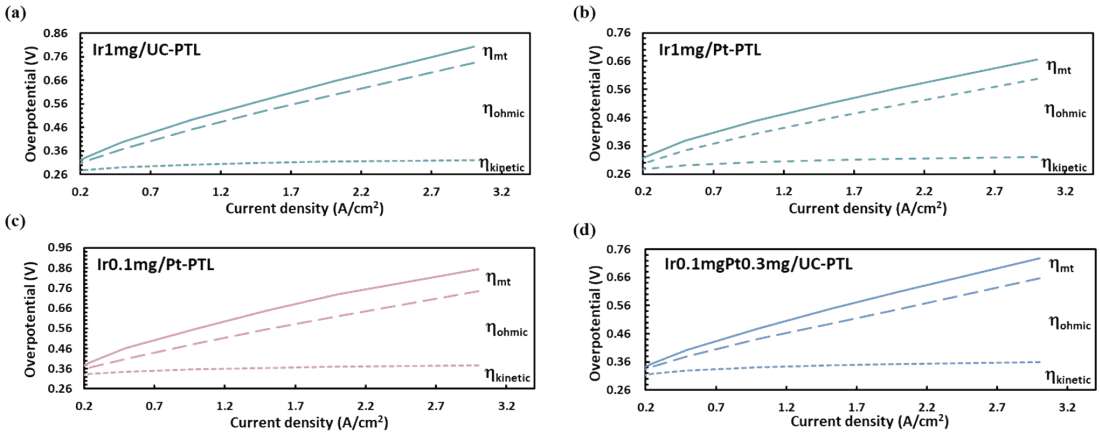

Fig. S15. Voltage loss analysis for (a) Ir1mg/UC-PTL; (b) Ir1mg/Pt-PTL; (c) Ir0.1mg/Pt-PTL and (d) Ir0.1mPt0.3mg/UC-PTL on 125μm membrane.

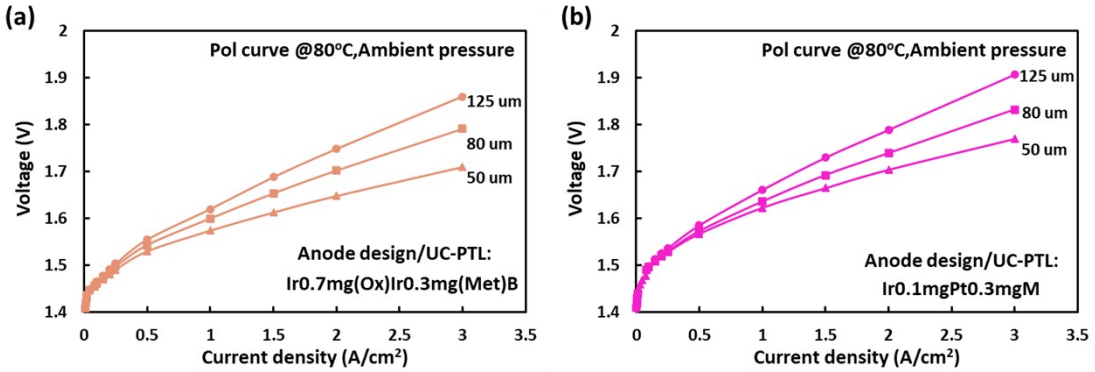

Fig. S16. Polarization curves for (a) Ir0.7mg(Ox)Ir0.3mg(Met)B/UC-PTL and (b) Ir0.1mgPt0.3mgM/UC-PTL on various membrane.

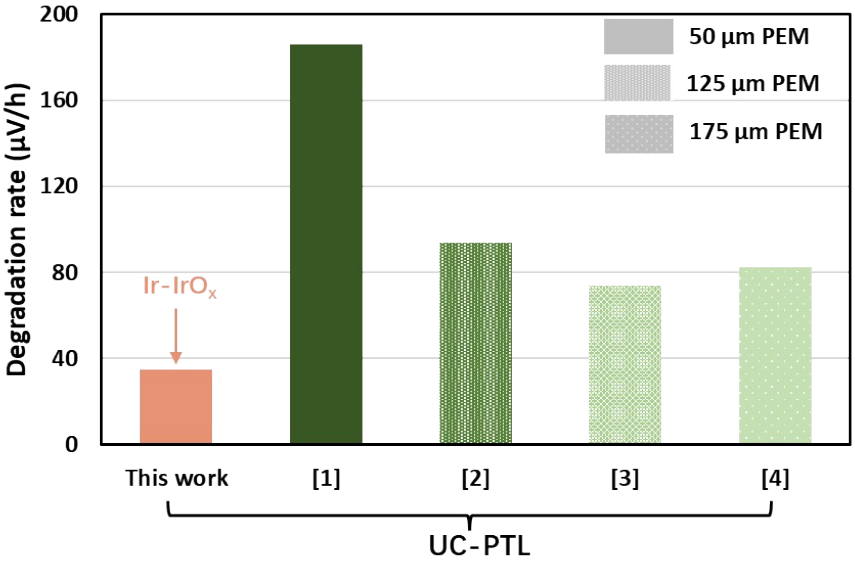

Fig. S17. Comparisons of the PEMWE durability with recent reported works[1-4].

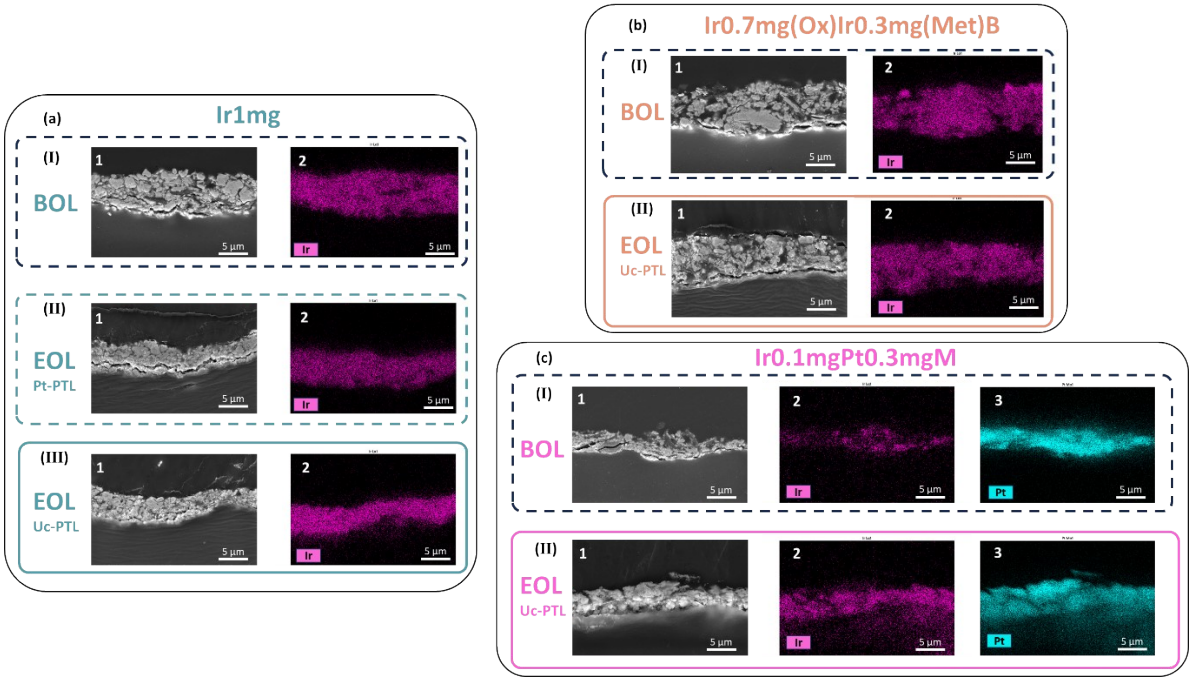

Fig. S18. SEM and EDS elemental (Ir and Pt) mapping analysis for various ACLs.

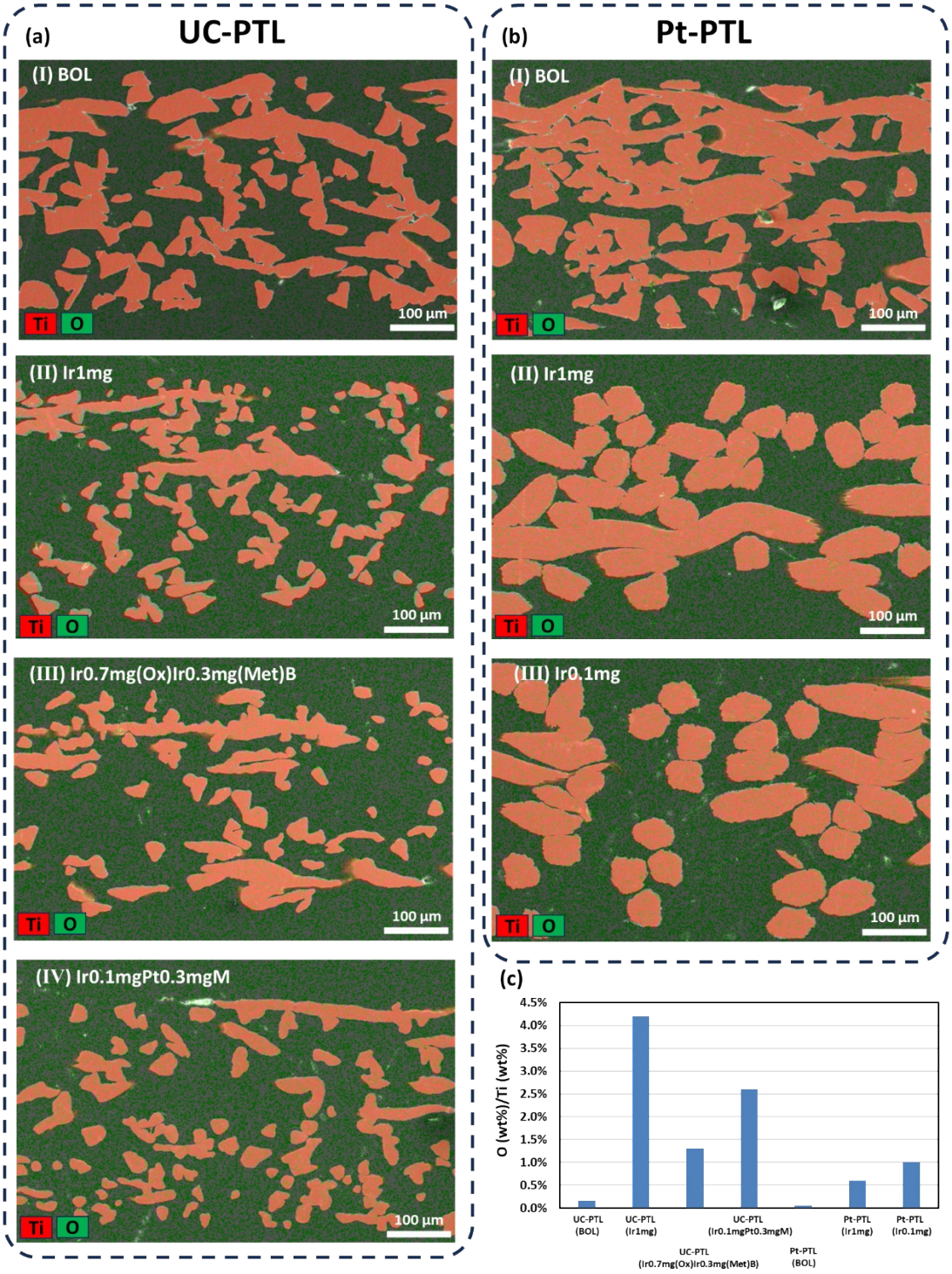

Fig. S19. EDS elemental mapping showing the distribution of Ti and O for (a) UC-PTL and (b) Pt-PTL paired with different ACLs. (c) O(wt%)/T(wt%) in various PTL

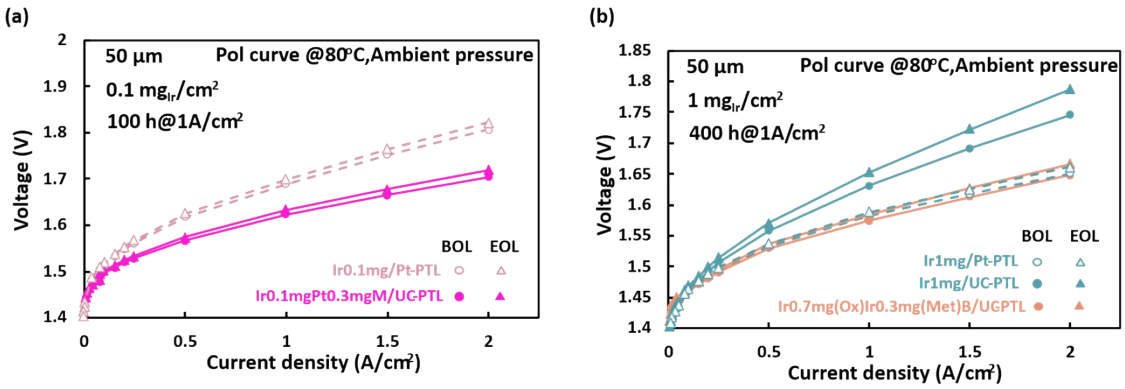

Fig. S20. Polarization curves in BOL and EOL stage for (a) Ir0.1mg/Pt-PTL and Ir0.1mgPt0.3mgM/UC-PTL and (b) Ir1mg/Pt-PTL, Ir1mg/UC-PTL and Ir0.7mg(Ox)Ir0.3mg(Met)B/UC-PTL.

Table S1. Physical properties, RDE mass activities and degradation rate for the catalysts.

Catalyst	BET area (m^2/g)	Crystallite size (nm)	Density (g/cm^3)	Mass activity (A/mg_{metal}@1.6 V)	Degradation rate (mV/h)
IrO_x	148	3	13.5	0.11	20
Ir	33.5	7.7	23.2	0.0086	35
Pt	37.5	7	22.8	—	—

Table S2. The degradation rate for CCM operating at 1 A/cm^2.

Anode	Design			Degradation rate (μV/h)
	Ir	Pt	PTL	
Ir1mg	1	0	Pt-PTL	10 ± 5
Ir0.1	0.1	0	Pt-PTL	100 ± 14
Ir1mg	1	0	UC-PTL	51±10
Ir1mgPt0.3mgB	1	0.3	UC-PTL	74 ± 11
Ir0.1mgPt0.3mgB	0.1	0.3	UC-PTL	104 ± 15
Ir0.1mgPt0.3mgM	0.1	0.3	UC-PTL	101 ± 15
Ir0.7mg(Ox)Ir0.3mg(Met)B	1	0	UC-PTL	35 ± 8

Supplementary Note 2:

For the cost models, a simplified formula (S5) for the levelized cost of hydrogen (LCoH) is used, following the approach adopted from our previous work[5].

$$LCoH = \frac{\text{CapE}x + \text{Discounted Cost of Electricity}}{\text{Discounted amount of } H_2 \text{ produced}} = \frac{C_X + c_E JT \sum_{y=1}^{Y} V_y \frac{1}{(1+q)^y}}{JT\mu \sum_{y=1}^{Y} \frac{1}{(1+q)^y}} \quad \text{(S5)}$$

Table S3. The description of the elements in equation S5

Element	Unit	Description
$LCoH$	\$/kg	Levelized Cost of Hydrogen
C_X	\$/cm^2	CapEx: cost of catalyst and PTL materials/cm^2
J	A/cm^2	Current density. Fixed value of 1 A/cm^2 in this study
V_y	V	Average cell voltage of the year
T	h/y	Annual operating time. Fixed value of 4,000 h/y in this study
c_E	\$/kWh	Cost of input electricity
μ	kg H_2/Ah	H_2 conversion factor: $\mu = 3.7311 \cdot 10^{-5}$ kg/Ah
Y	-	Total number of years calculated from the lifetime of the cell
q	%	Financial discount factor. Set to 6% in this study
Ir cost	\$/g	163
Pt cost	\$/g	31.5

Pt-PTL cost	$/cm^2	0.26
UC-PTL cost	$/cm^2	0.09

Table S4. LCoH for the CCMs operating at 1 A/cm^2.

Anode	CapEx ($/kg H_2)	OpEx ($/kg H_2)	Lifetime (h)	Total ($/kg H_2)
Ir1mg/UC-PTL	1.26	2.2	10,000	3.46
Ir1mg/UC-PTL	0.74	2.68	10,000	3.42
Ir0.1/Pt-PTL	7.85	2.35	1,000	10.17
Ir0.1mgPt0.3mgB/UC-PTL	3.29	2.33	1,000	5.62
Ir0.1mgPt0.3mgM/UC-PTL	3.29	2.32	1,000	5.61
Ir1mgPt0.3mgB/UC-PTL	0.78	2.79	10,000	3.57
Ir0.7mg(Ox)Ir0.3mg(Met)B/UC-PTL	0.75	2.46	10,000	3.21

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