



Supplementary Information

Breaking the activity-stability trade-off in acidic oxygen evolution reaction via steering charge-compensation mechanisms in RuO₂

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[†] Electronic supplementary information (ESI) available.

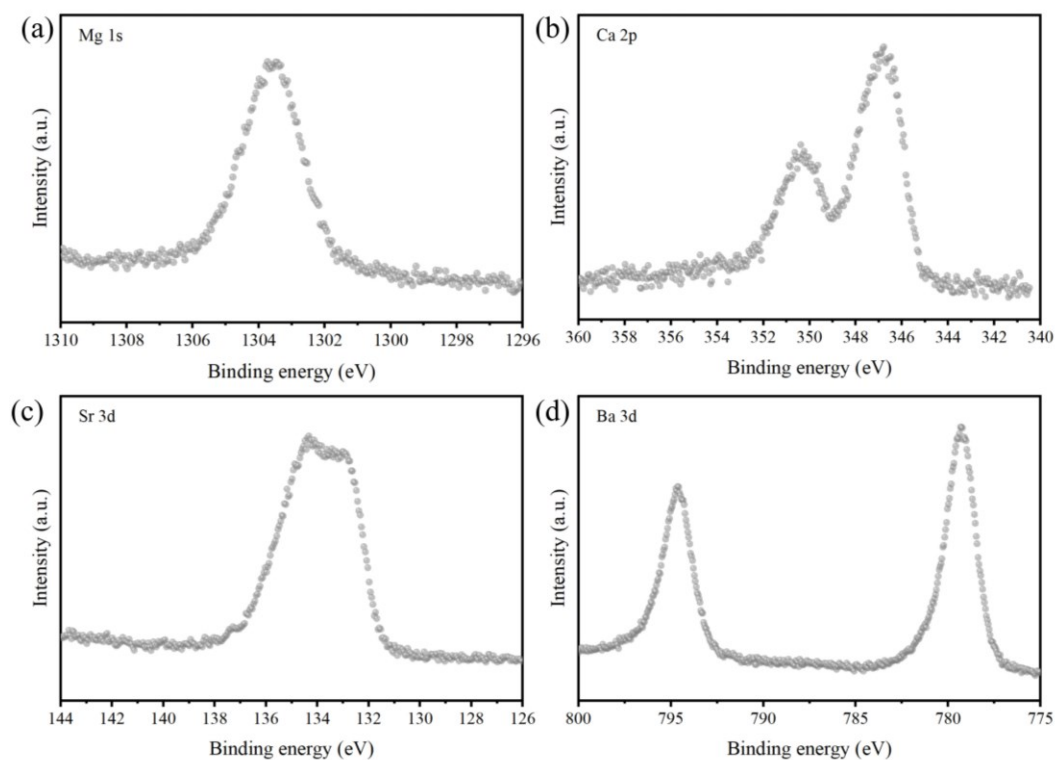


Fig. S1. XPS spectra of Mg 1s (a), Ca 2p (b), Sr 3d (c) and Ba 3d (d).

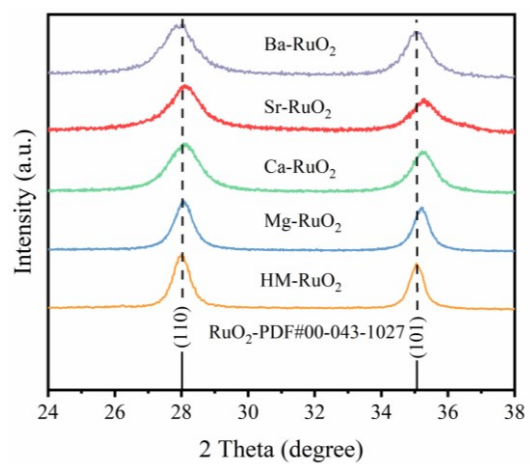


Fig. S2 Magnified XRD patterns of catalysts.

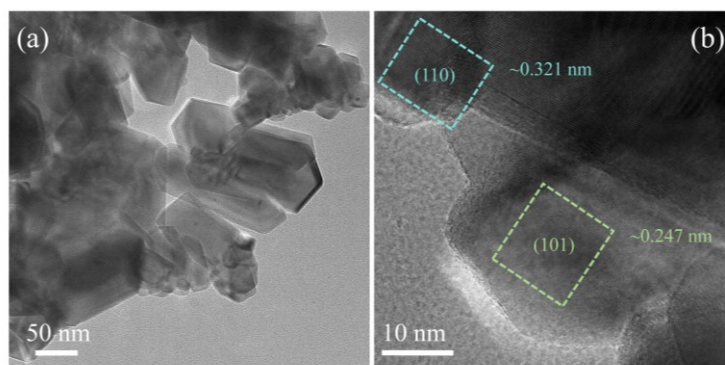


Fig. S3 TEM (a) and HRTEM (b) images of HM-RuO₂.

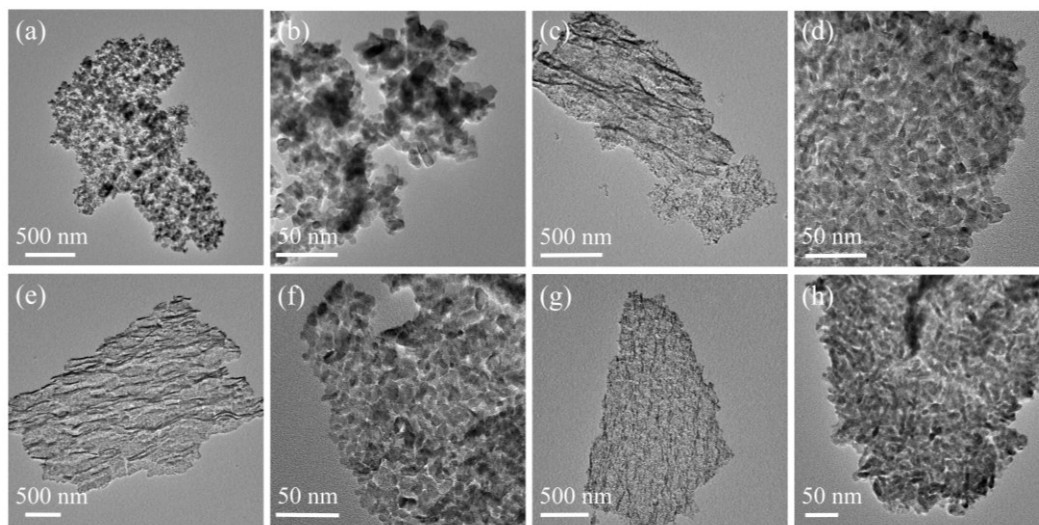


Fig. S4 TEM images of Mg-RuO₂ (a, b), Ca-RuO₂ (c, d), Sr-RuO₂ (e, f) and Ba-RuO₂ (g, h).

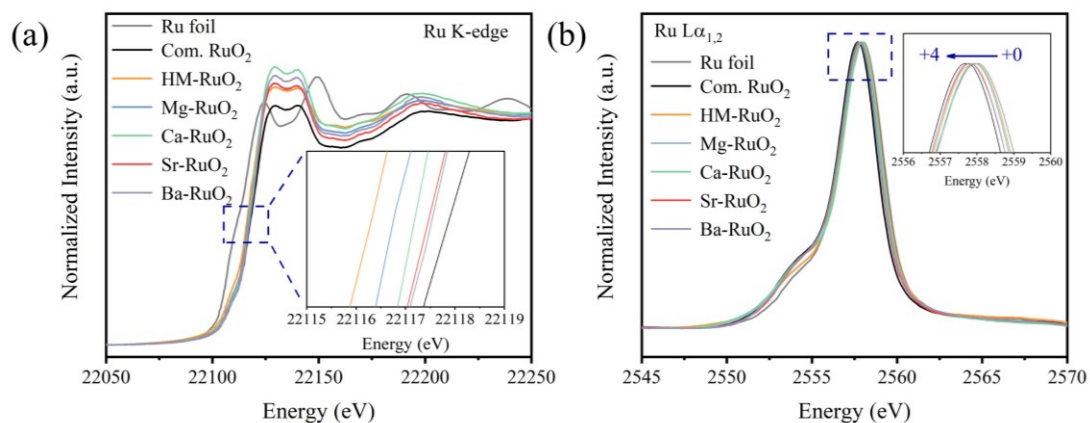


Fig. S5 (a) Normalized Ru K-edge XANES spectra of samples. (b) Normalized XES spectra of Ru L $\alpha_{1,2}$ of samples.

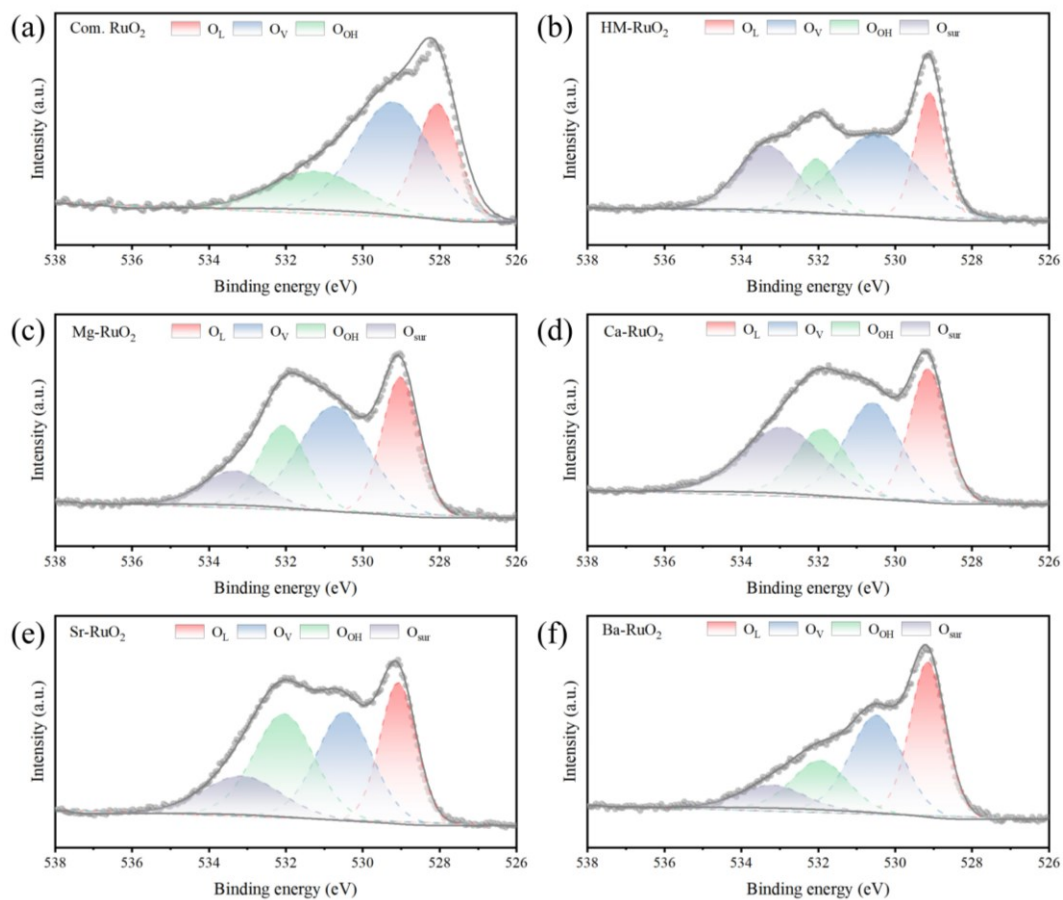


Fig. S6 O 1s spectra of Com. RuO_2 (a), HM- RuO_2 (b), Mg- RuO_2 (c), Ca- RuO_2 (d), Sr- RuO_2 (e) and Ba- RuO_2 (f).

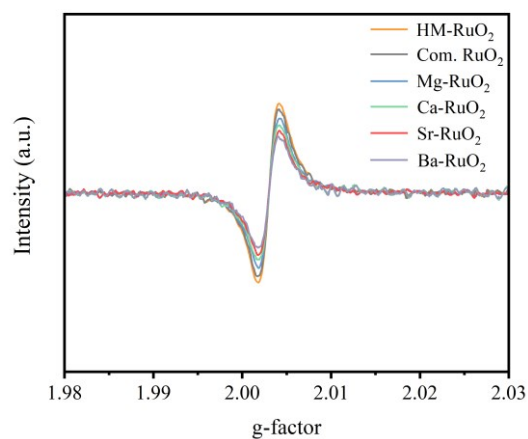


Fig. S7 EPR spectra of catalysts with different O_v densities.

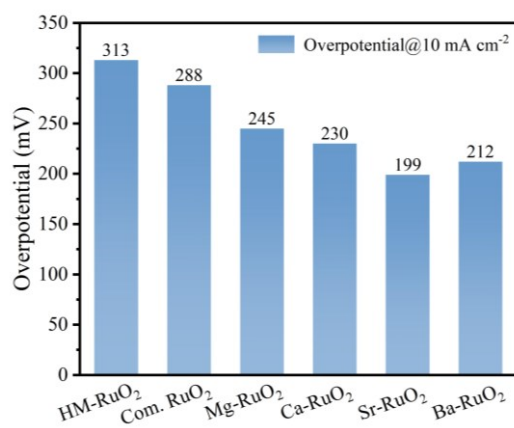


Fig. S8 Overpotential of catalysts at 10 mA cm⁻².

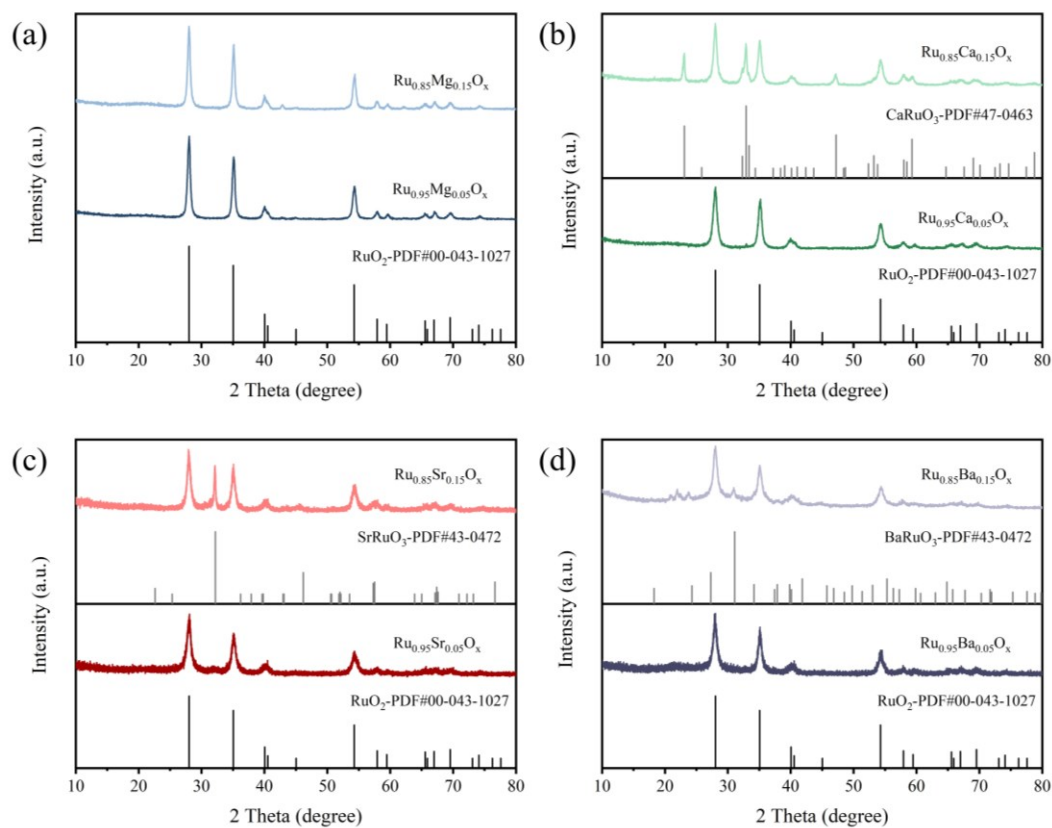


Fig. S9 (a) XRD patterns of RuO₂ catalysts with varying Mg²⁺ doping concentrations. (b) XRD patterns of RuO₂ catalysts with varying Ca²⁺ doping concentrations. (c) XRD patterns of RuO₂ catalysts with varying Sr²⁺ doping concentrations. (d) XRD patterns of RuO₂ catalysts with varying Ba²⁺ doping concentrations.

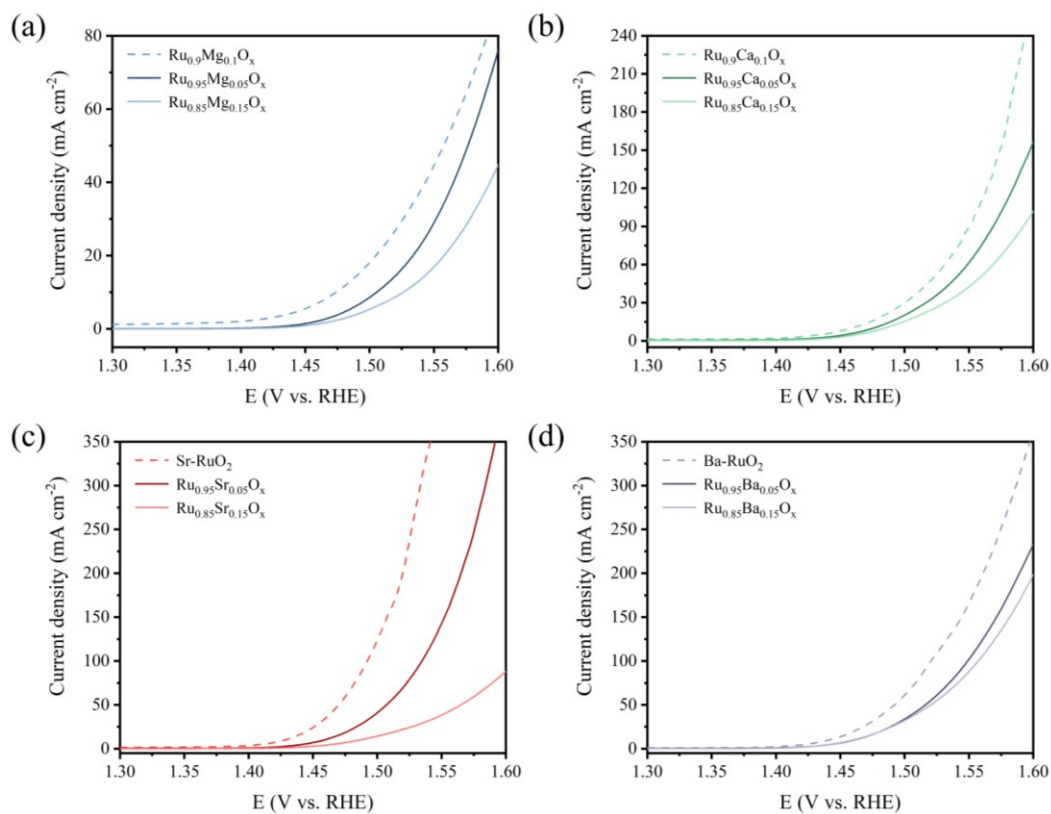


Fig. S10 (a) LSV curves of RuO₂ catalysts with varying Mg²⁺ doping concentrations. (b) LSV curves of RuO₂ catalysts with varying Ca²⁺ doping concentrations. (c) LSV curves of RuO₂ catalysts with varying Sr²⁺ doping concentrations. (d) LSV curves of RuO₂ catalysts with varying Ba²⁺ doping concentrations.

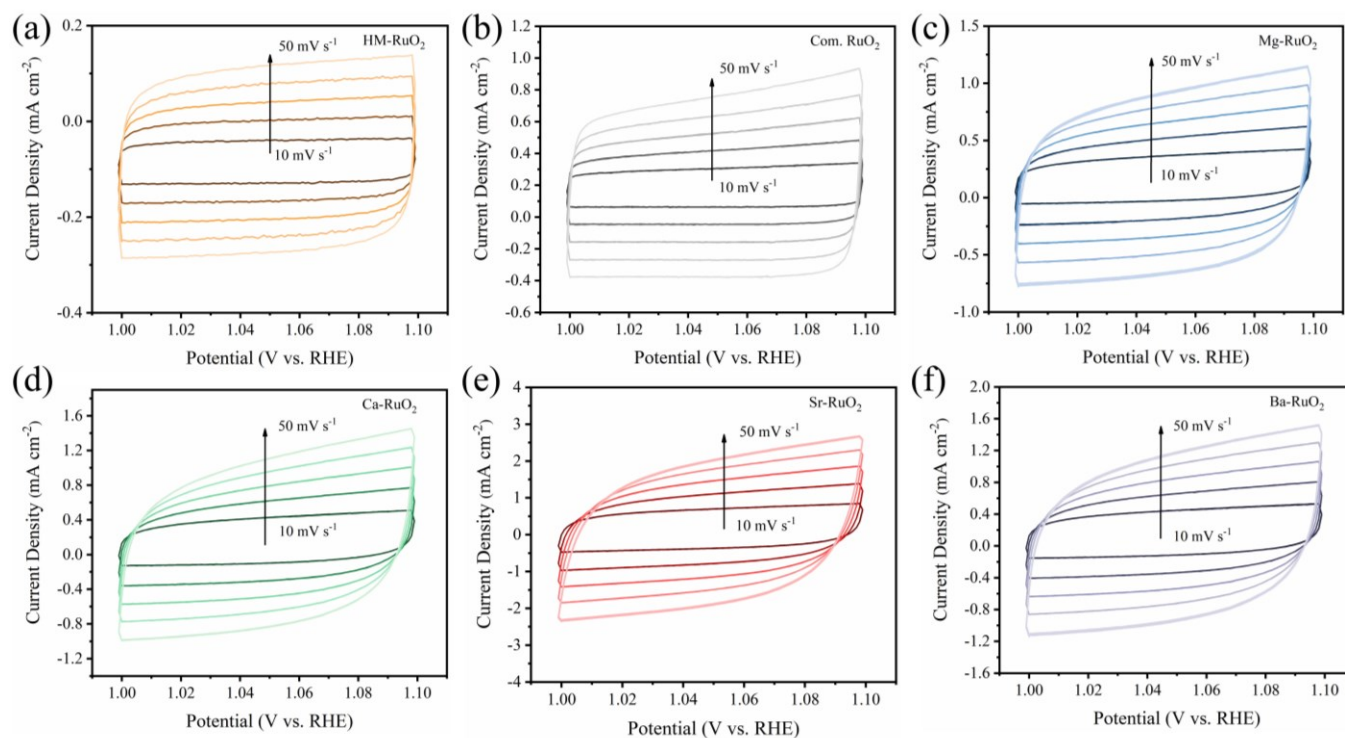


Fig. S11 CV curves from the non-reactive region for (a) HM-RuO₂, (b) Com. RuO₂, (c) Mg-RuO₂, (d) Ca-RuO₂, (e) Sr-RuO₂ and (f) Ba-RuO₂.

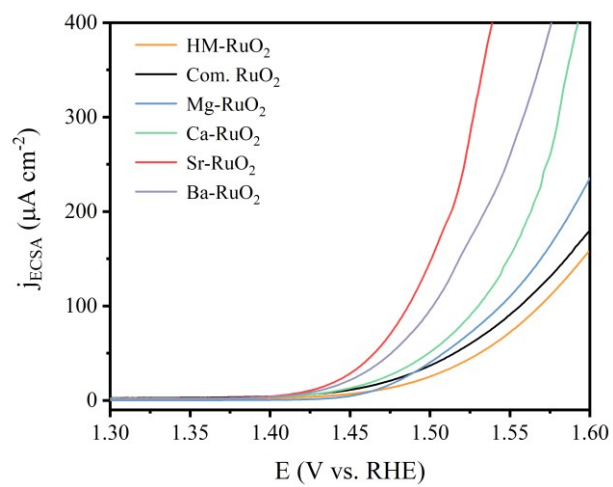


Fig. S12 OER polarization LSV curves normalized by ECSA.

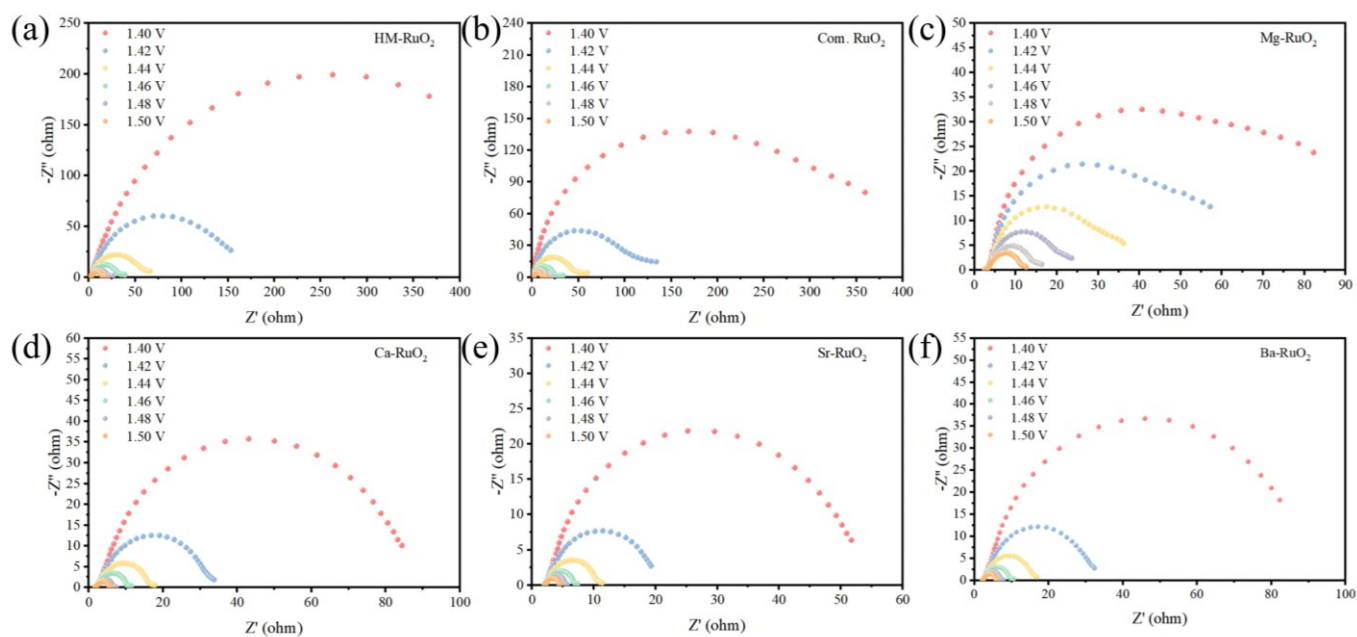


Fig. S13 Nyquist plots of catalysts from 1.40 to 1.50 V vs. RHE.

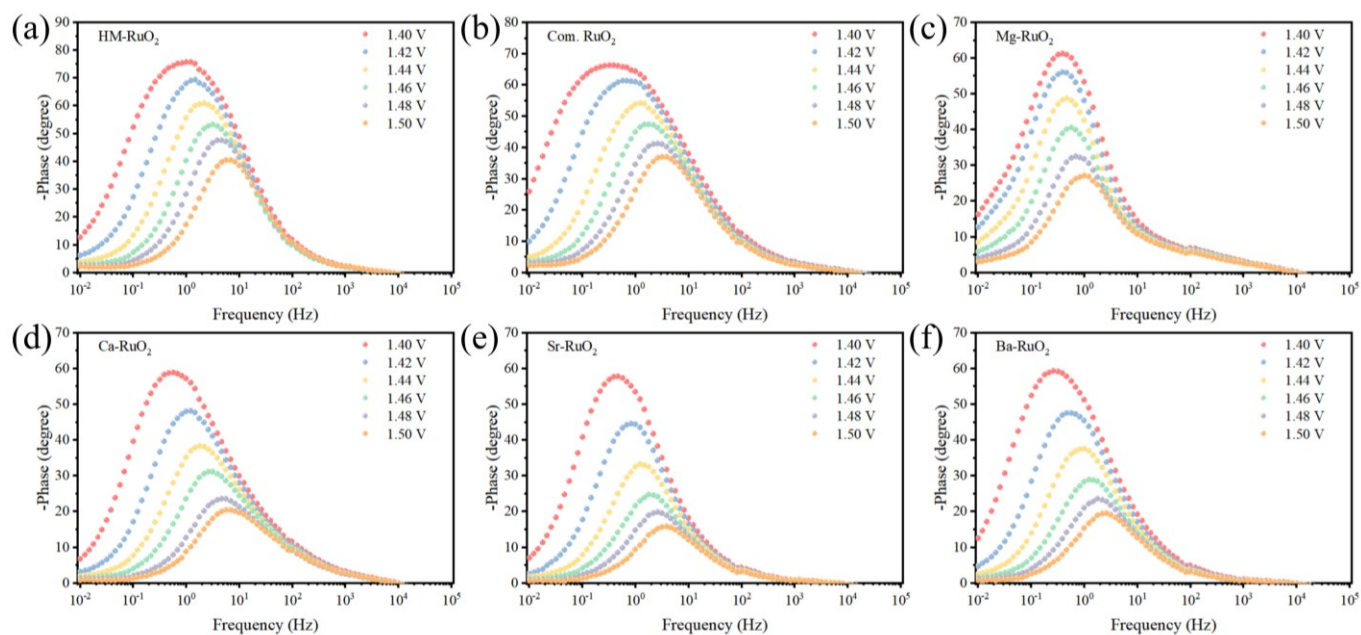


Fig. S14 Operando Bode phase plots of catalysts from 1.40 to 1.50 V vs. RHE.

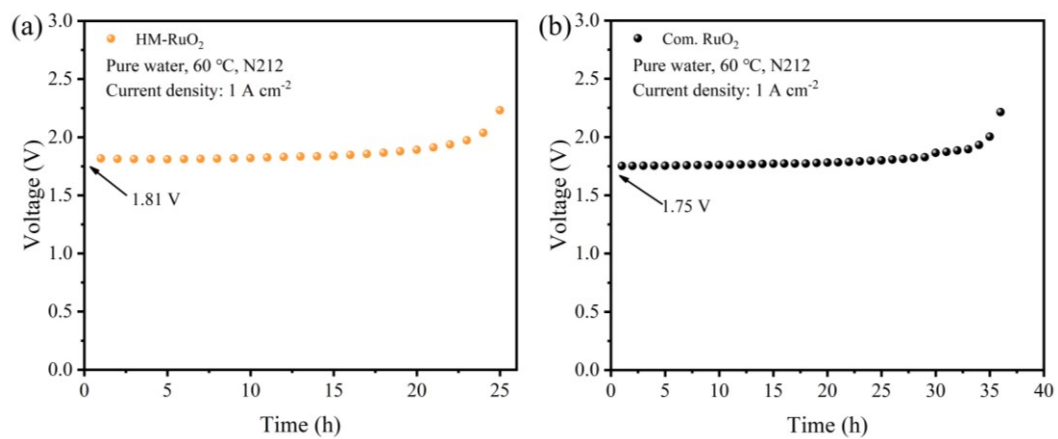


Fig. S15 Long-term durability tests at current density of 1 A cm^{-2} of HM-RuO₂ (a) and Com. RuO₂.

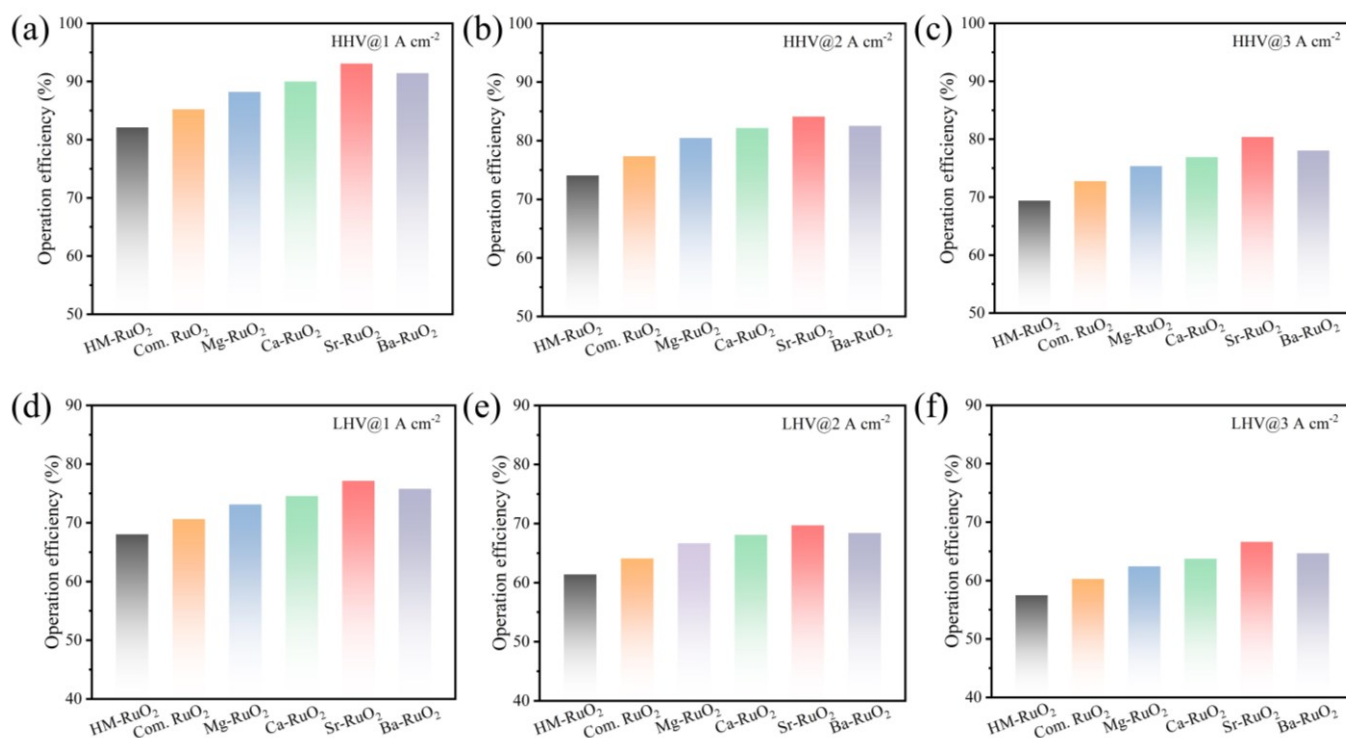


Fig. S16 The operation efficiency (calculating using higher heating value of H₂, HHV=1.43×10⁵ J g⁻¹) of PEMWE utilizing HM-RuO₂, Com. RuO₂, Mg-RuO₂, Ca-RuO₂, Sr-RuO₂ and Ba-RuO₂ as the anodes at 1 A cm⁻² (a), 2 A cm⁻² (b) and 3 A cm⁻² (c). The operation efficiency (calculating using lower heating value of H₂, LHV=1.185×10⁵ J g⁻¹) of PEMWE utilizing HM-RuO₂, Com. RuO₂, Mg-RuO₂, Ca-RuO₂, Sr-RuO₂ and Ba-RuO₂ as the anodes at 1 A cm⁻² (d), 2 A cm⁻² (e) and 3 A cm⁻² (f) ¹.

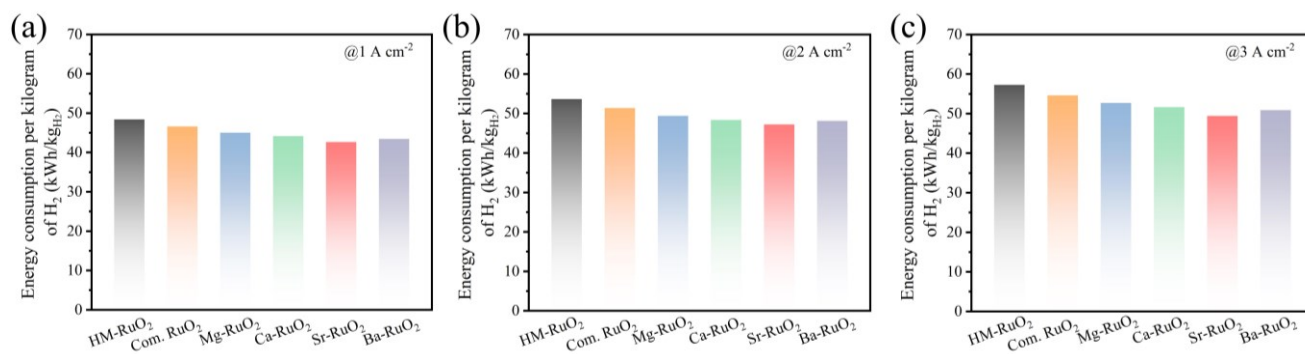


Fig. S17 The energy consumption per kilogram of H_2 for PEMWE utilizing HM-RuO₂, Com. RuO₂, Mg-RuO₂, Ca-RuO₂, Sr-RuO₂ and Ba-RuO₂ as the anodes at 1 A cm⁻² (a), 2 A cm⁻² (b) and 3 A cm⁻².

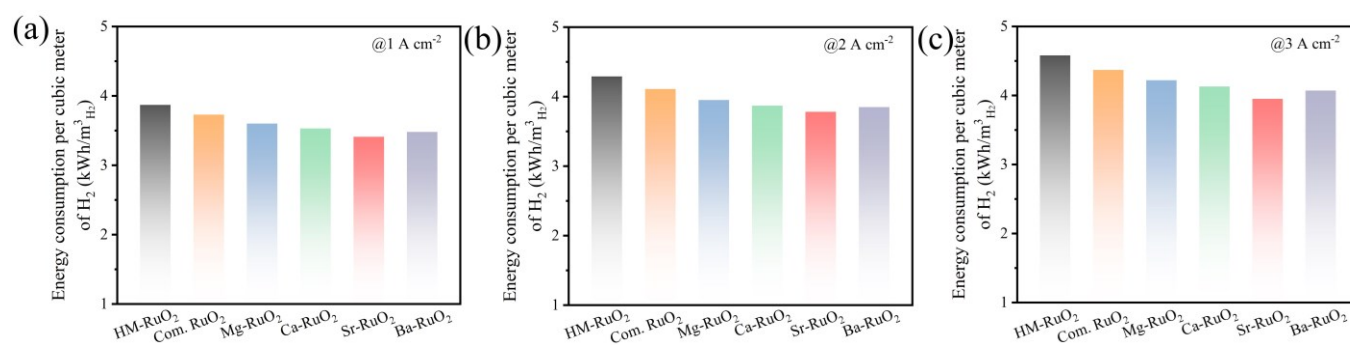


Fig. S18 The energy consumption per cubic meter of H₂ for PEMWE utilizing HM-RuO₂, Com. RuO₂, Mg-RuO₂, Ca-RuO₂, Sr-RuO₂ and Ba-RuO₂ as the anodes at 1 A cm⁻² (a), 2 A cm⁻² (b) and 3 A cm⁻².

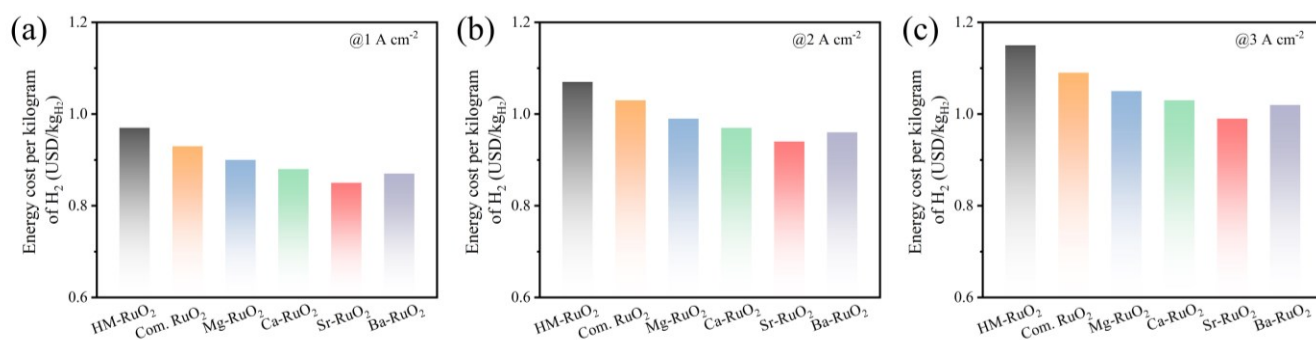


Fig. S19 The energy cost per kilogram of H₂ for PEMWE utilizing HM-RuO₂, Com. RuO₂, Mg-RuO₂, Ca-RuO₂, Sr-RuO₂ and Ba-RuO₂ as the anodes at 1 A cm⁻² (a), 2 A cm⁻² (b) and 3 A cm⁻².

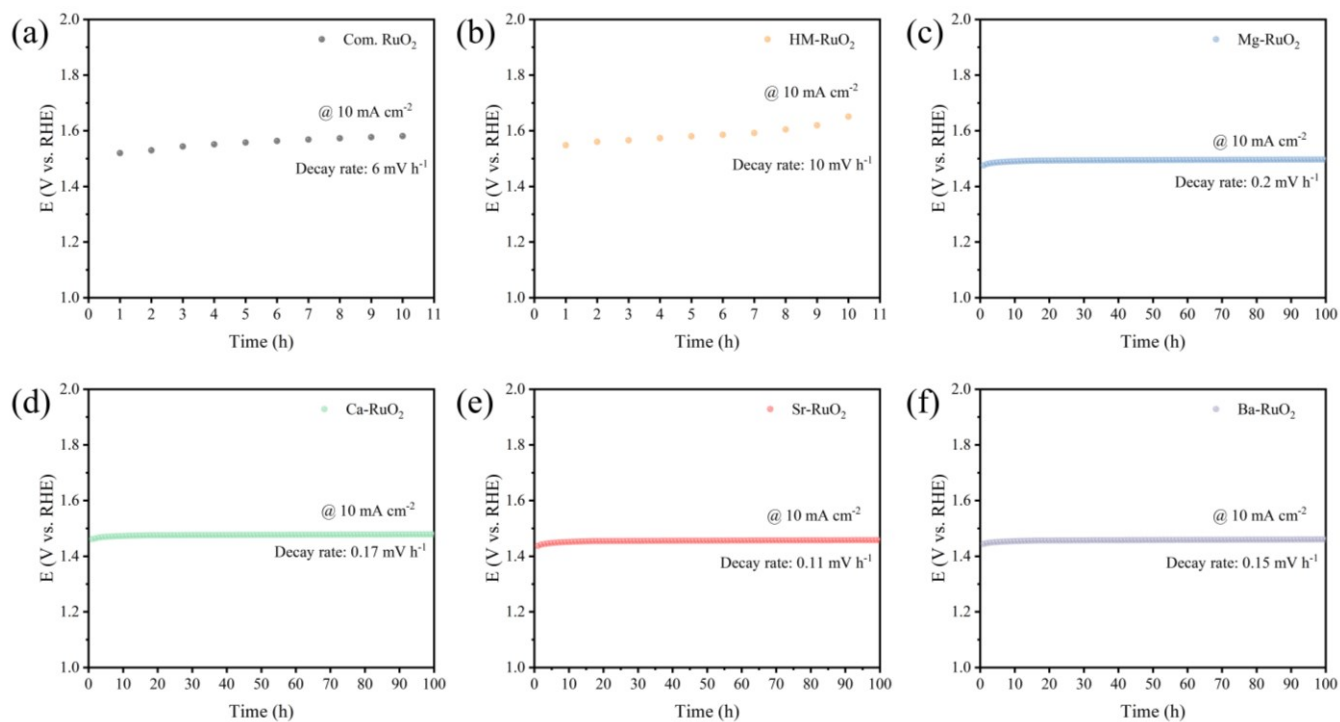


Fig. S20 Chronopotentiometric (CP) durability test at 10 mA cm⁻².

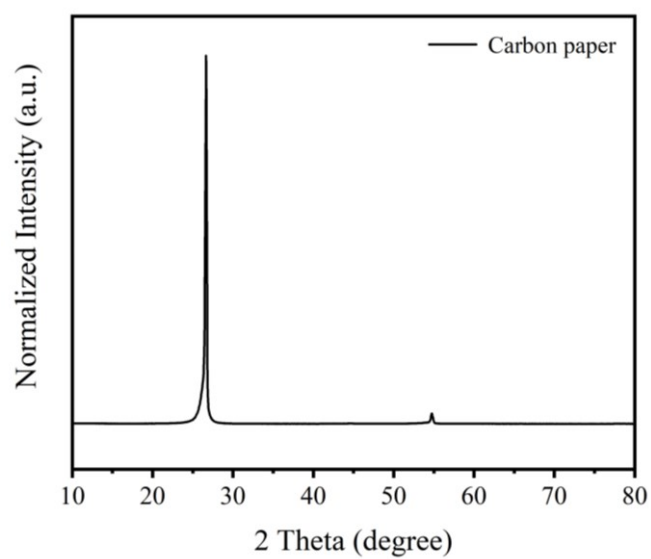


Fig. S21 XRD patterns of carbon paper.

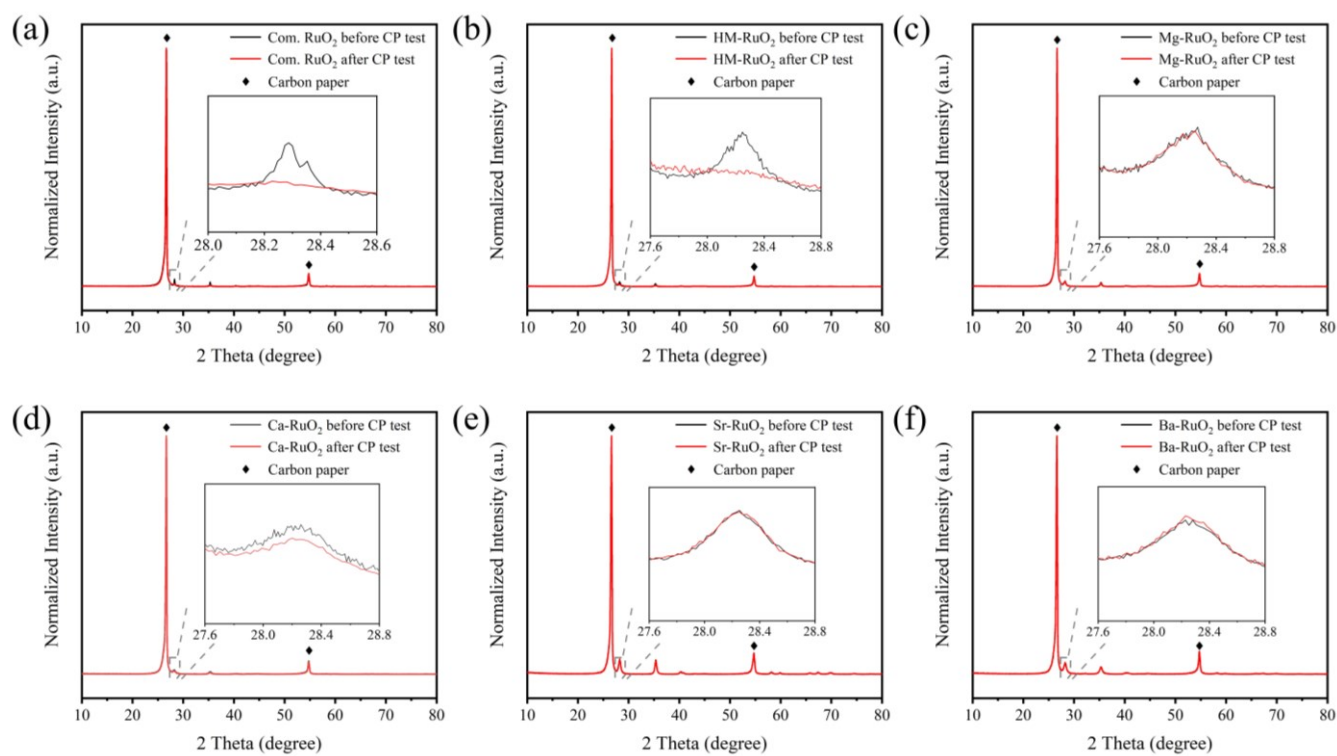


Fig. S22 XRD patterns of catalysts before and after CP tests.

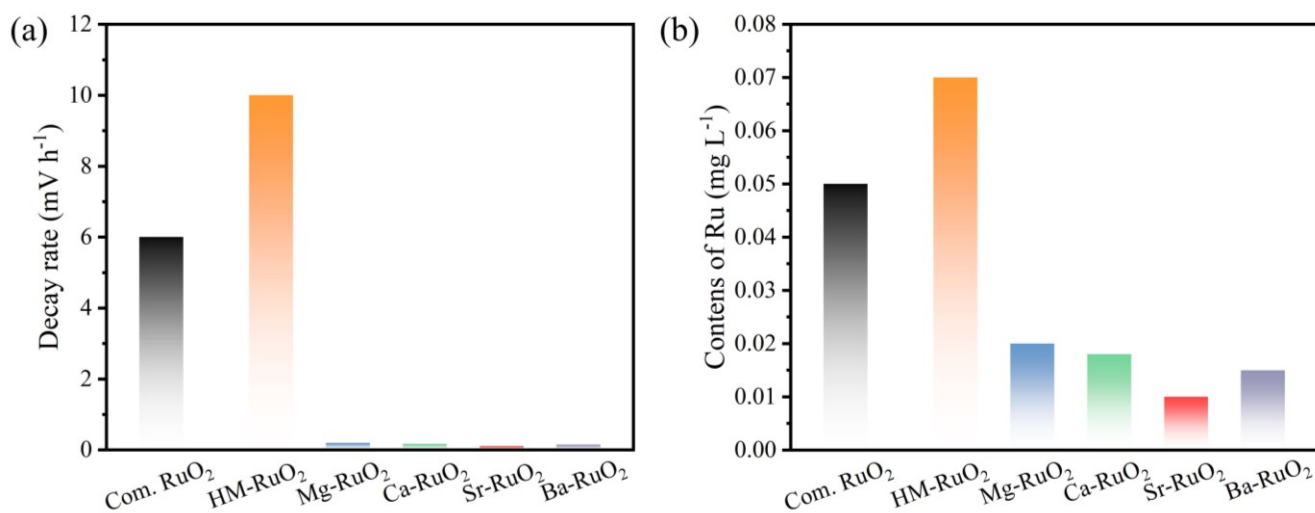


Fig. S23 (a) Decay rate of catalysts at 10 mA cm⁻². (b) Dependence of Ru dissolution in the electrolyte on the OER reaction time at 10 mA cm⁻².

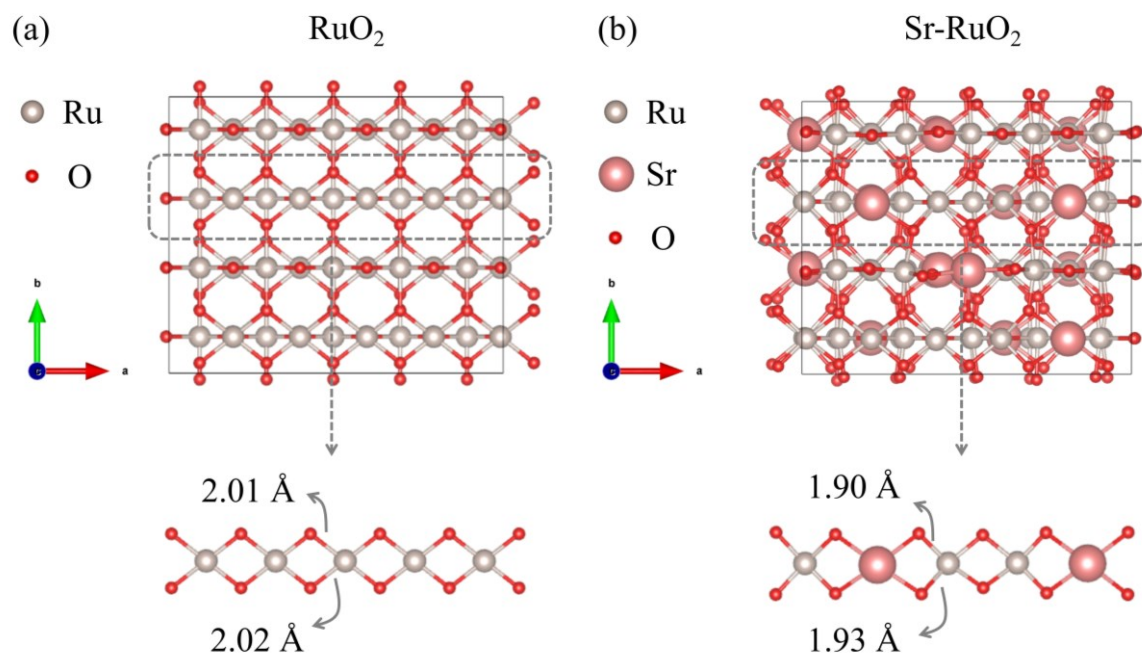


Fig. S24 Crystal structure simulation of the RuO₂ (a) and Sr-RuO₂ (b) catalysts, respectively.

Table S1. Chemical composition of the catalysts determined by ICP-OES.

Catalysts	Ru (wt%)	Ae (wt%)	Ru/Ae at. ratio
HM-RuO ₂	76.32%	/	/
Mg-RuO ₂	71.45%	1.92%	8.95
Ca-RuO ₂	71.80%	3.15%	9.04
Sr-RuO ₂	69.10%	6.61%	9.06
Ba-RuO ₂	66.33%	10.04%	8.97

Table S2. Ionic radii in rutile phase.

Ion	CN	$R_i/\text{\AA}$
Ru^{3+}	6	0.68
Ru^{4+}	6	0.62
Ru^{5+}	6	0.57
Mg^{2+}	6	0.72
Ca^{2+}	6	1.0
Sr^{2+}	6	1.18
Ba^{2+}	6	1.35

Table S3. The interplanar spacing of the catalyst.

Catalysts	(110)	(101)
HM-RuO ₂	0.321	0.247
Mg-RuO ₂	0.315	0.226
Ca-RuO ₂	0.317	0.229
Sr-RuO ₂	0.319	0.231
Ba-RuO ₂	0.324	0.241

Table S4. Fitting results of O 1s XPS spectra for catalysts.

	O _L	O _V	O _{OH}	O _{sur}	O _V /O _L
Com. RuO ₂	28.68%	50.82%	20.5%	/	1.77
HM-RuO ₂	22.47%	41.78%	9.96%	25.79%	1.86
Mg-RuO ₂	27.67%	39.83%	21.05%	11.45%	1.44
Ca-RuO ₂	25.25%	29.65%	30.27%	14.82%	1.17
Sr-RuO ₂	25.93%	27.48%	18.08%	28.51%	1.06
Ba-RuO ₂	37.38%	34.31%	18.33%	9.98%	0.92

Table S5. PEMWE activity based on Ae^{2+} - RuO_2 , Com. RuO_2 , HM- RuO_2 and those previously reported catalysts.

Catalysts	Loading amount	Membrane	Cell temperature ($^{\circ}\text{C}$)	Cell voltage (V) at 1 A cm^{-2}	Stability (h)	Reference
Mg- RuO_2	2 mg cm^{-2}	Nafion 212	$60\text{ }^{\circ}\text{C}$	1.68	$80@1\text{ A cm}^{-2}$	This work
Ca- RuO_2	2 mg cm^{-2}	Nafion 212	$60\text{ }^{\circ}\text{C}$	1.65	$110@1\text{ A cm}^{-2}$	This work
Sr- RuO_2	2 mg cm^{-2}	Nafion 212	$60\text{ }^{\circ}\text{C}$	1.59	$200@1\text{ A cm}^{-2}$	This work
Ba- RuO_2	2 mg cm^{-2}	Nafion 212	$60\text{ }^{\circ}\text{C}$	1.62	$150@1\text{ A cm}^{-2}$	This work
Com. RuO_2	2 mg cm^{-2}	Nafion 212	$60\text{ }^{\circ}\text{C}$	1.74	$35@1\text{ A cm}^{-2}$	This work
HM- RuO_2	2 mg cm^{-2}	Nafion 212	$60\text{ }^{\circ}\text{C}$	1.81	$25@1\text{ A cm}^{-2}$	This work
LD-B/ RuO_2	2 mg cm^{-2}	Nafion 117	$80\text{ }^{\circ}\text{C}$	1.63	$300@0.25\text{ A cm}^{-2}$	Ref. 2
Ir- RuO_2	3 mg cm^{-2}	Nafion 115	$80\text{ }^{\circ}\text{C}$	1.60	$300@1\text{ A cm}^{-2}$	Ref. 3
MD- RuO_2 -BN	1 mg cm^{-2}	Nafion 115	$80\text{ }^{\circ}\text{C}$	1.64	$50@1\text{ A cm}^{-2}$	Ref. 4
$\text{Cr}_{0.2}\text{Ru}_{0.8}\text{O}_{2-x}$	3 mg cm^{-2}	Nafion 115	$60\text{ }^{\circ}\text{C}$	1.77	$200@1\text{ A cm}^{-2}$	Ref. 5
RuTiO_x	1 mg cm^{-2}	Nafion 117	$80\text{ }^{\circ}\text{C}$	1.60	$100@0.5\text{ A cm}^{-2}$	Ref. 6
GB- RuO_2	1 mg cm^{-2}	Nafion 117	$80\text{ }^{\circ}\text{C}$	1.54	$100@0.1\text{ A cm}^{-2}$	Ref. 7
Er- RuO_x	3 mg cm^{-2}	Nafion 117	$80\text{ }^{\circ}\text{C}$	1.59	$100@1\text{ A cm}^{-2}$	Ref. 8
Pb- RuO_2	1 mg cm^{-2}	Nafion 212	$80\text{ }^{\circ}\text{C}$	1.68	$250@0.5\text{ A cm}^{-2}$	Ref. 9
4f-Nd- RuO_2	2 mg cm^{-2}	Nafion 115	$60\text{ }^{\circ}\text{C}$	2.38	$200@0.1\text{ A cm}^{-2}$	Ref. 10
$\text{Ba}_{0.3}(\text{SO}_4)_{\delta}\text{W}_{0.2}\text{Ru}_{0.5}\text{O}_{2-\delta}$	3 mg cm^{-2}	Nafion 115	$80\text{ }^{\circ}\text{C}$	1.68	$300@0.5\text{ A cm}^{-2}$	Ref. 11

Table S6. The power and efficiency of the PEMWE by the Ae^{2+} - RuO_2 , Com. RuO_2 and HM- RuO_2 as anodic catalyst at different current densities.

Anodic catalyst	Current density (A cm^{-2})	Efficiency (HHV)	Efficiency (LHV)	Energy consumption (kWh/kg H_2)	Energy consumption ($\text{kWh/m}^3 \text{H}_2$)	Energy cost per kilogram of H_2 (USD/kg H_2)
Mg- RuO_2	1	88.22%	73.11%	45.02	3.6	0.9
	2	80.46%	66.68%	49.37	3.95	0.99
	3	75.35%	62.44%	52.72	4.22	1.05
Ca- RuO_2	1	89.99%	74.57%	44.14	3.53	0.88
	2	82.16%	68.08%	48.34	3.87	0.97
	3	76.91%	63.74%	51.64	4.13	1.03
Sr- RuO_2	1	93.09%	77.14%	42.67	3.41	0.85
	2	84.12%	69.71%	47.22	3.78	0.94
	3	80.39%	66.62%	49.41	3.95	0.99
Ba- RuO_2	1	91.43%	75.77%	43.44	3.48	0.87
	2	82.54%	68.4%	48.12	3.85	0.96
	3	78.04%	64.67%	50.9	4.07	1.02
Com. RuO_2	1	85.23%	70.63%	46.61	3.73	0.93
	2	77.35%	64.1%	51.35	4.11	1.03
	3	72.75%	60.28%	54.6	4.37	1.09
HM- RuO_2	1	82.11%	68.04%	48.38	3.87	0.97
	2	74.07%	61.38%	53.63	4.29	1.07
	3	69.37%	57.48%	57.26	4.58	1.15

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