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Supporting Information

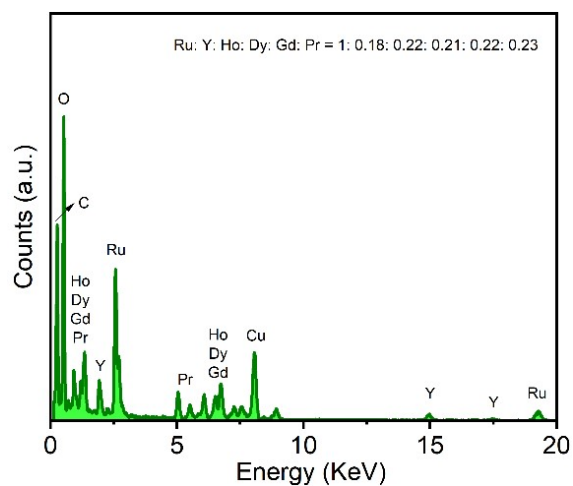
A new-type high-entropy electrocatalyst with a pyrochlore structure for acid-water oxidation

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Phone: +86-551-63607924.



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30 Figure S1. EDS spectrum of HE-YRO. The signals of Cu and C in the spectrum were
31 originated from the copper grid coated by carbon membrane.

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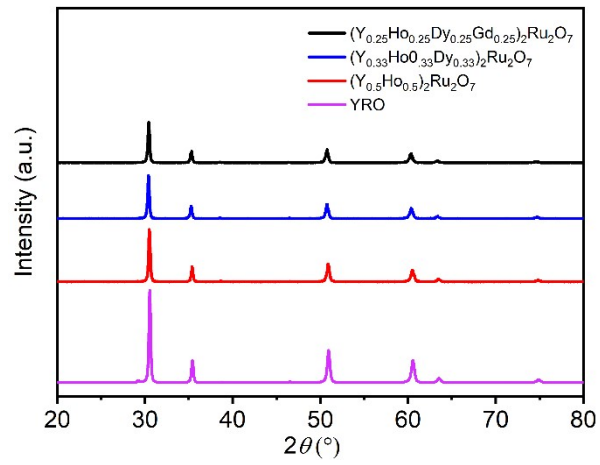
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41 Figure S2. XRD patterns of YRO, $(Y_{0.5}Ho_{0.5})_2Ru_2O_7$, $(Y_{0.33}Ho_{0.33}Dy_{0.33})_2Ru_2O_7$, and
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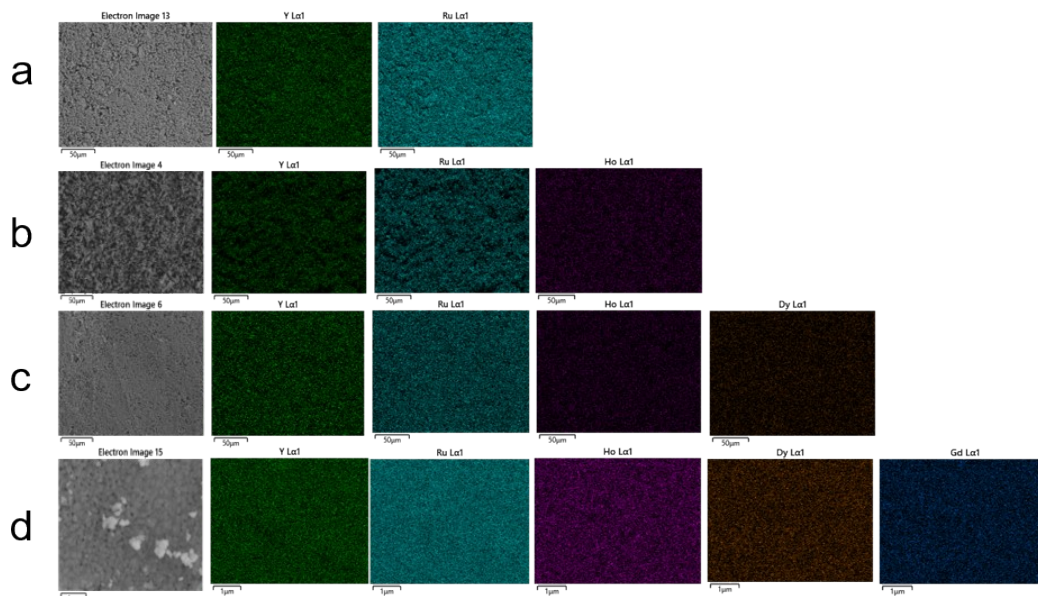
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55 Figure S3. SEM-EDS mappings of (a) YRO, (b) $(Y_{0.5}Ho_{0.5})_2Ru_2O_7$, (c) $(Y_{0.33}Ho_{0.33}Dy_{0.33})_2Ru_2O_7$,
 56 and (d) $(Y_{0.25}Ho_{0.25}Dy_{0.25}Gd_{0.25})_2Ru_2O_7$.

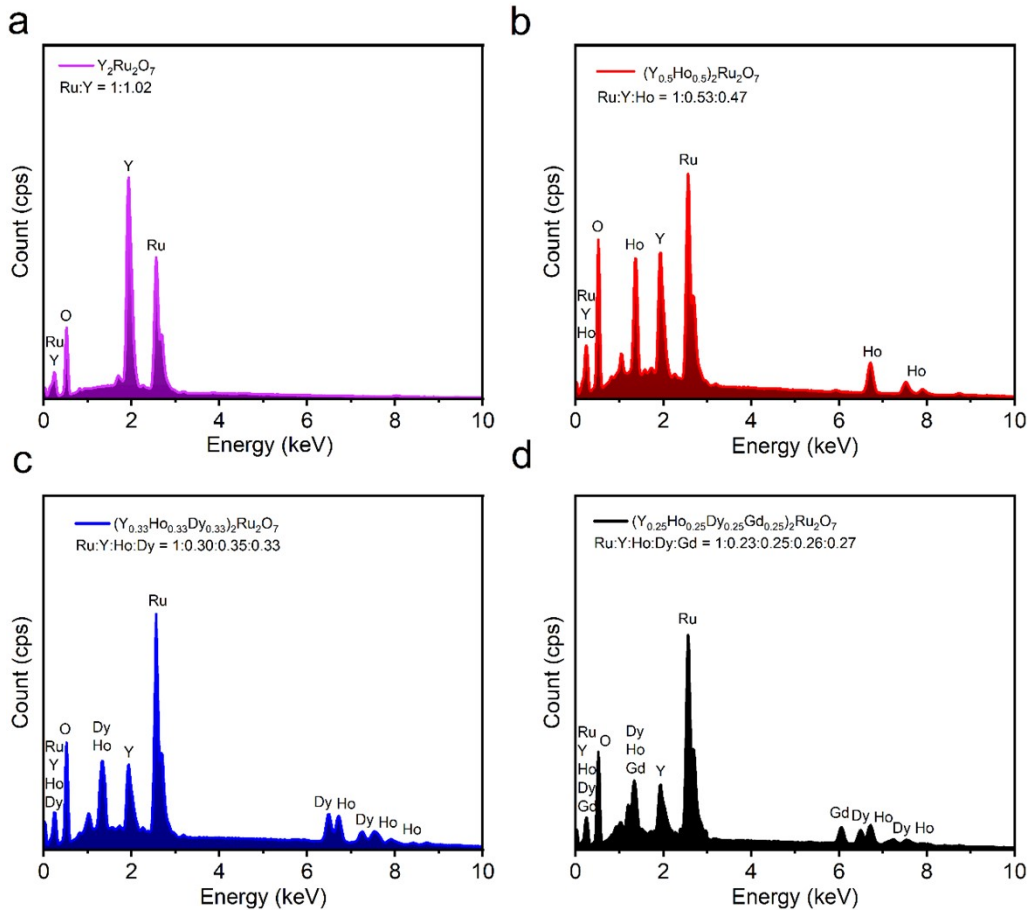
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63 Figure S4. SEM-EDS spectra of (a) YRO, (b) $(Y_{0.5}Ho_{0.5})_2Ru_2O_7$, (c) $(Y_{0.33}Ho_{0.33}Dy_{0.33})_2Ru_2O_7$,

64 and (d) $(Y_{0.25}Ho_{0.25}Dy_{0.25}Gd_{0.25})_2Ru_2O_7$.

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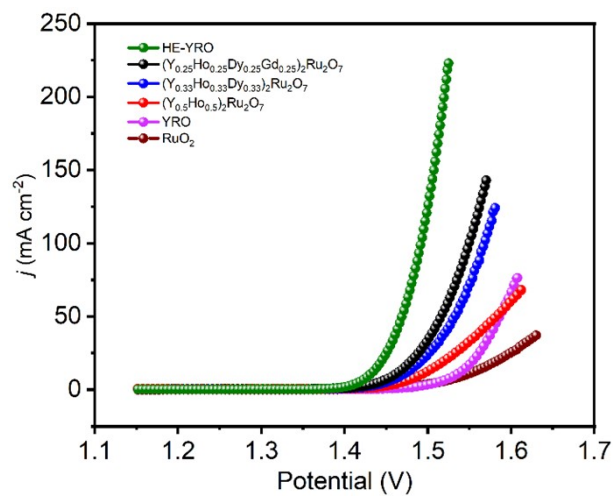
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72 Figure S5. Polarization curves of as-prepared Ru-based catalysts and commercial RuO₂.

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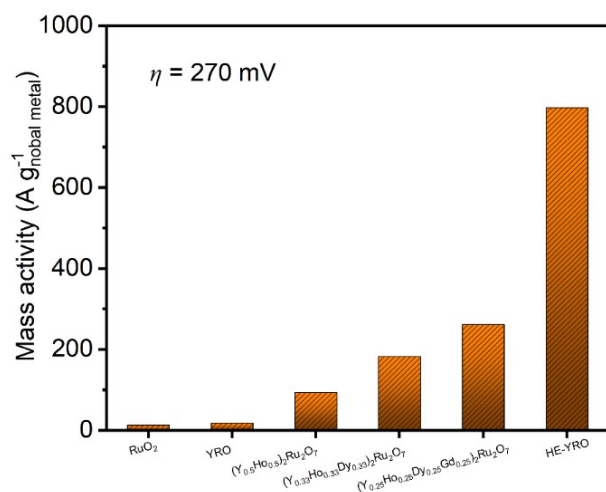
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87 Figure S6. Comparison of mass activity at $\eta = 270$ mV.

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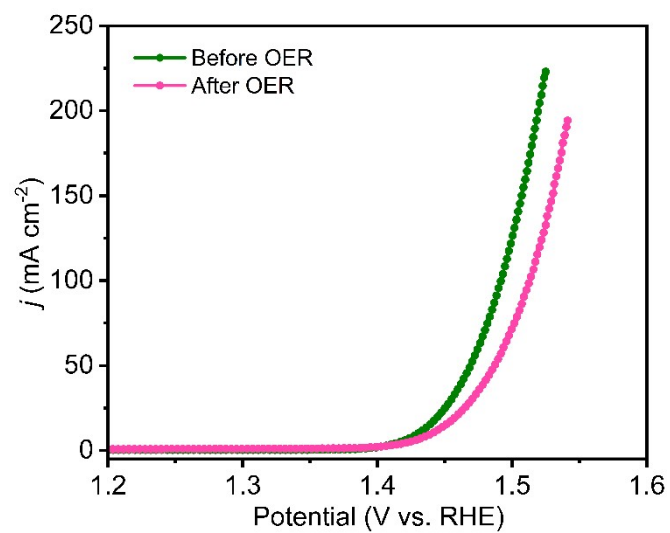
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110 Figure S7. Polarization curves for HE-YRO before and after OER test.

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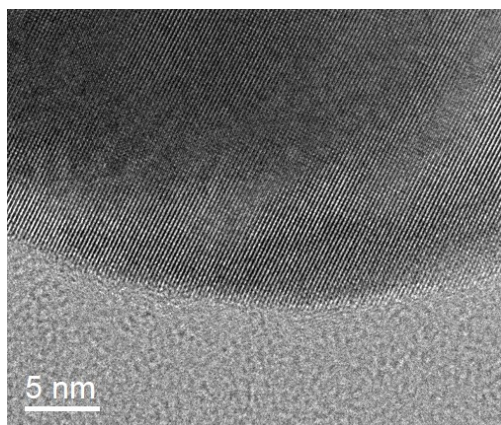
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126 Figure S8. HRTEM image of the HE-YRO electrocatalyst after the stability test.

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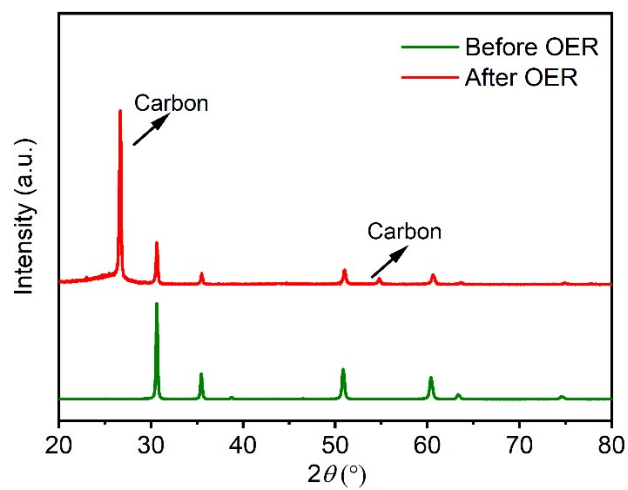
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143 Figure S9. XRD patterns of HE-YRO before and after the OER testing.

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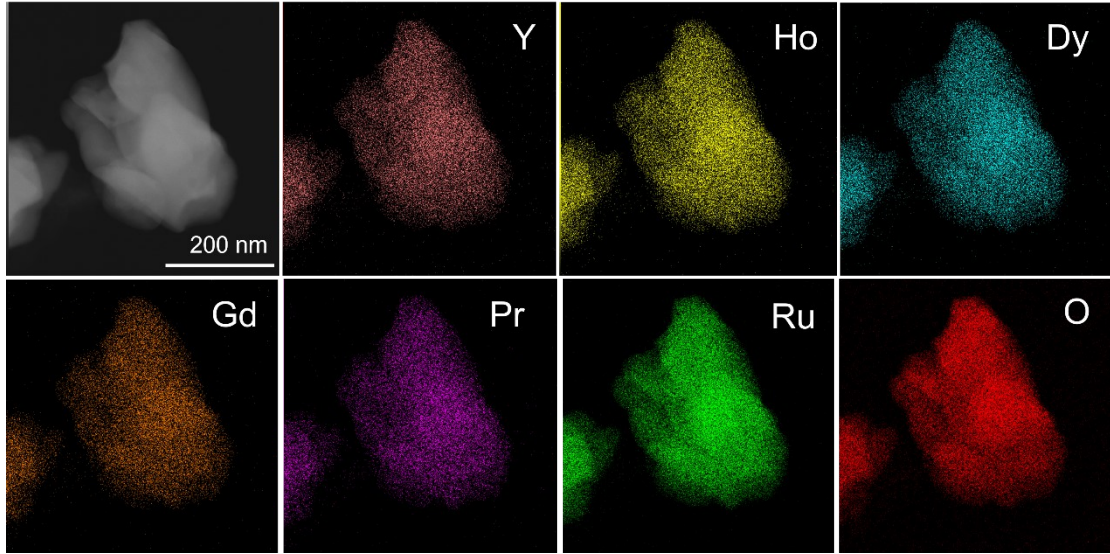
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158 Figure S10. HRTEM-EDS elemental mappings of the HE-YRO sample after OER.

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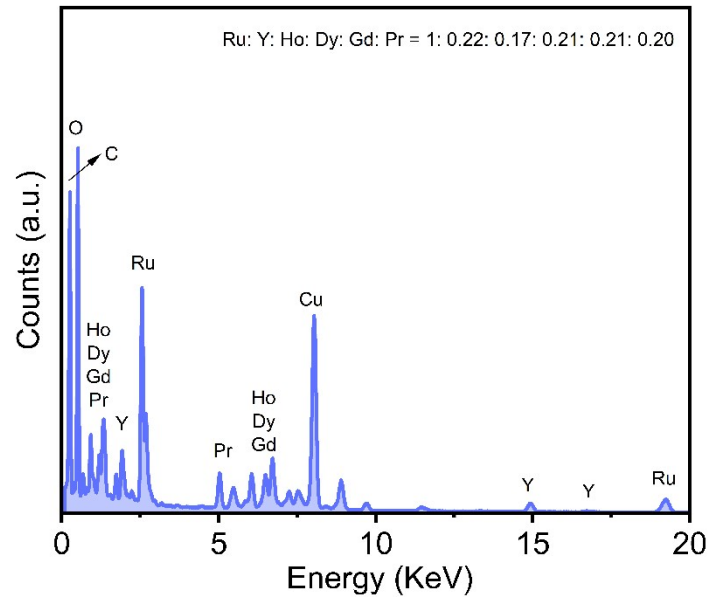
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172 Figure S11. EDS spectrum of HE-YRO after OER. The signals of Cu and C in the
173 spectrum were originated from the copper grid coated by carbon membrane.

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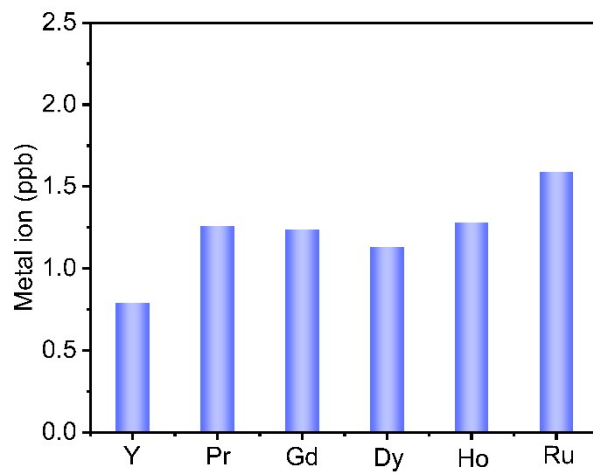
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186 Figure S12. ICP analysis for HE-YRO after OER test.

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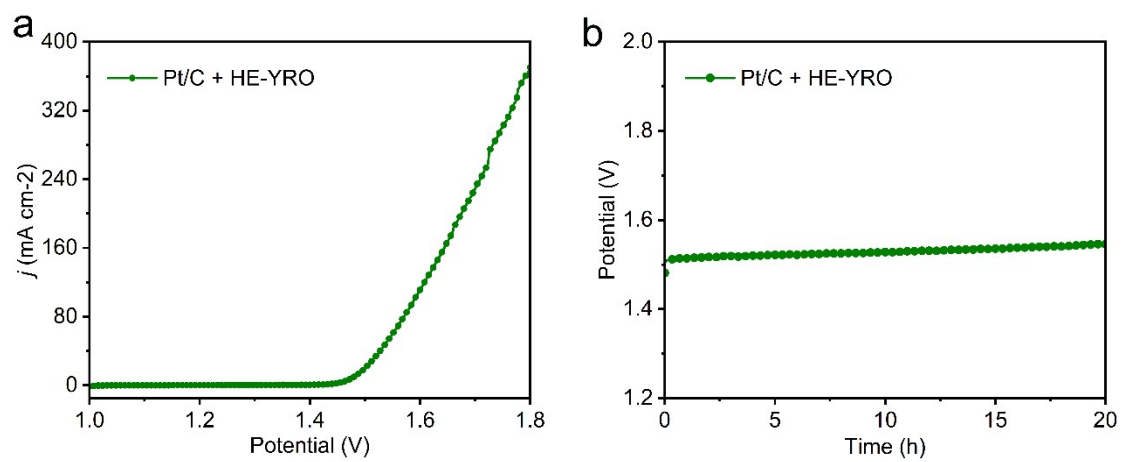
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200 Figure S13. (a) Polarization curve of HE-YRO || Pt/C for overall water splitting. (b)
201 Chronopotentiometric curve water electrolysis for the HE-YRO || Pt/C in a two-electrode
202 setup with a current density of 10 mA cm⁻².

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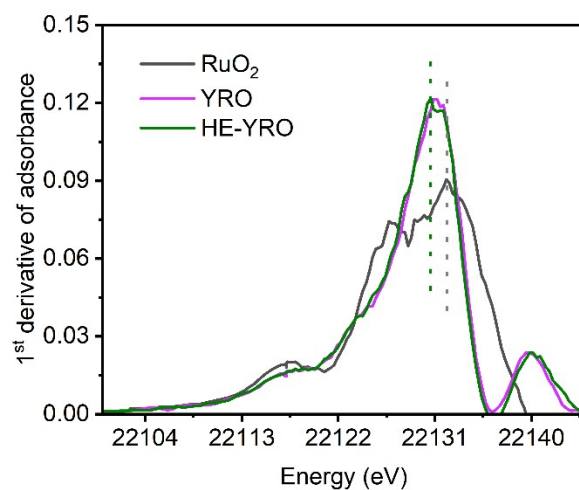
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211 Figure S14. 1st derivative curves of Ru *K*-edge XANES spectra for HE-YRO, YRO, and RuO₂.

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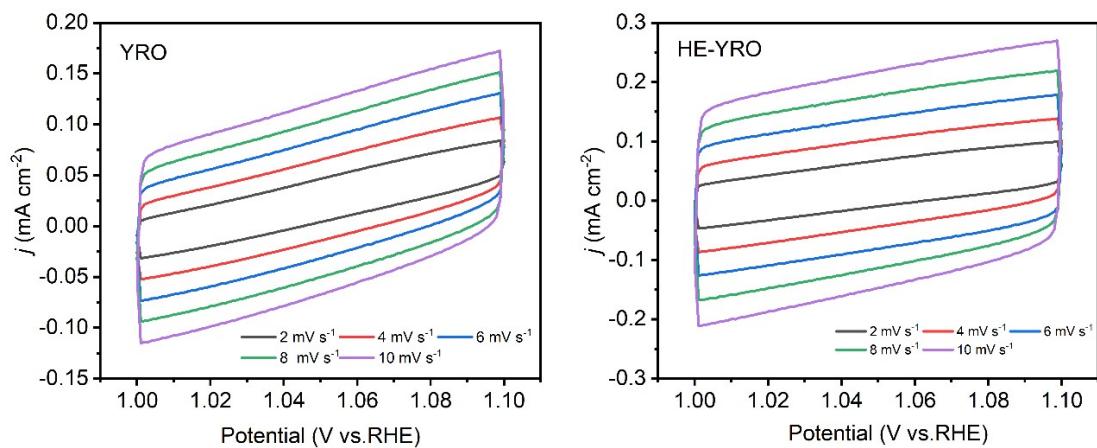
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233 Figure S15. Cyclic voltammetry curves in O²-saturated 0.5M H₂SO₄. The sweep rates are in the

234 range of 2-10 mV s⁻¹.

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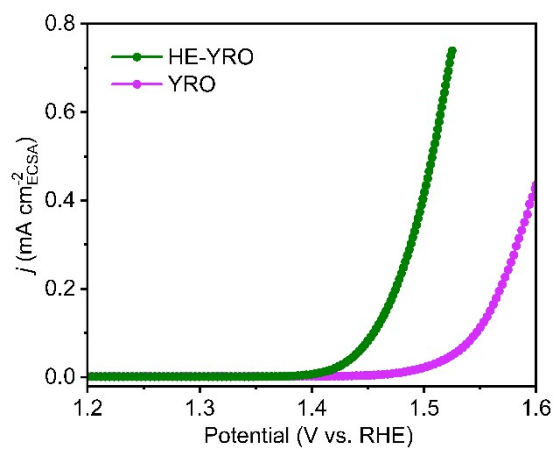
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256 Figure S16. Polarization curves for HE-YRO and YRO normalized by ECSA.

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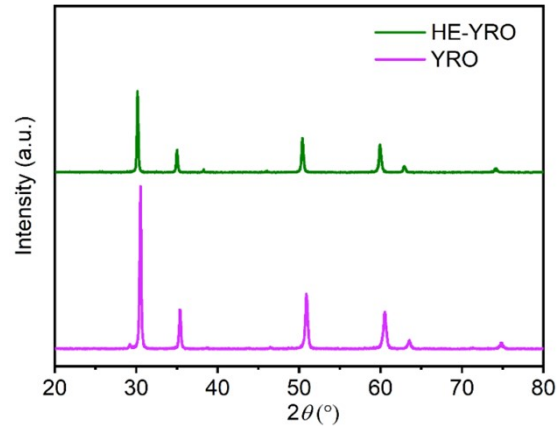
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273 Figure S17. XRD patterns of HE-YRO and YRO.

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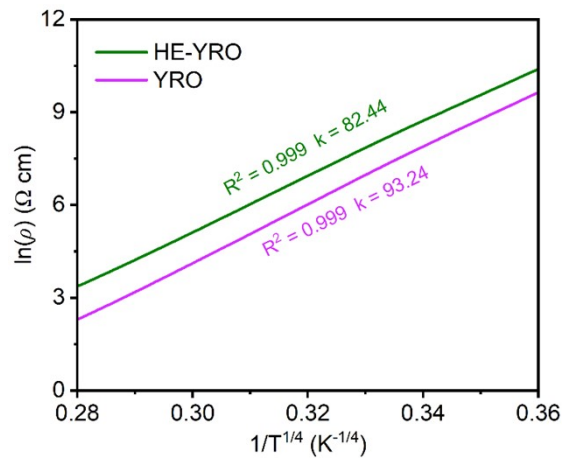
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291 Figure S18. The $\rho(T)$ curves fitted with the three-dimensional VRH model. The hopping energy W

292 ($W=0.25k_B T_0^{1/4} T^{3/4}$, where k_B is the Boltzmann constant) is proportional to $T_0^{1/4}$ ($k = T_0^{1/4}$).

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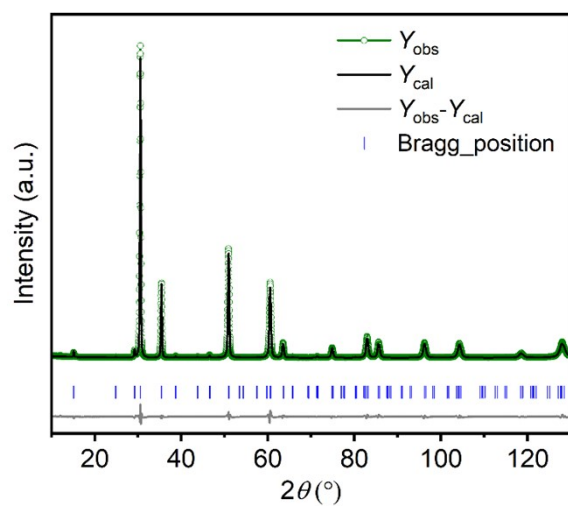
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314 Figure S19. XRD pattern for YRO together with the Rietveld refined results.

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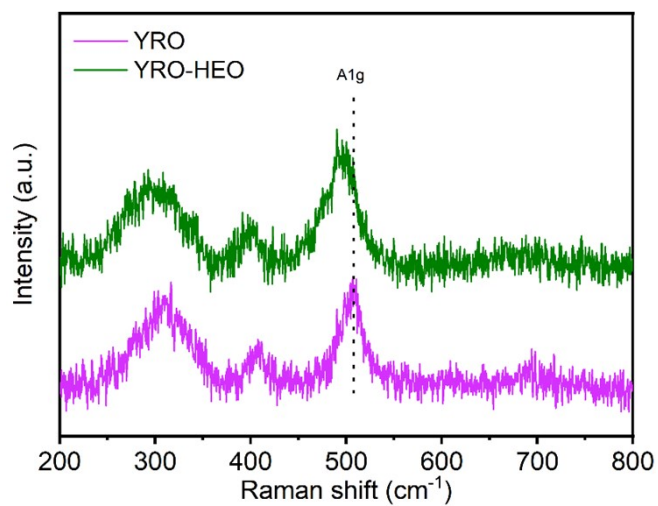
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345 Figure S20. Raman spectra of HE-YRO and YRO.

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377 Table S1. Refined structural parameters of the room temperature XRD patterns for HE-YRO and
378 YRO samples.

Sample	HE-YRO	YRO
Space group	<i>Fd-3m</i>	
<i>a</i> (Å)	10.2121(4)	10.1449(1)
Cell volume (Å ³)	1064.9891(4)	1044.1030(2)
Ru-O bond (Å)	1.9856(6)	1.9964(8)
Ru-O-Ru bond angle (°)	130.78	127.86
<i>R_p</i>	5.93	6.41
<i>R_{wp}</i>	8.21	9.04

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393 Table S2. Element ratios of the HE-YRO before OER test determined using ICP-AES.

Sample	Ru (at. %)	Y (at. %)	Ho (at. %)	Dy (at. %)	Gd (at. %)	Pr (at. %)
HE-YRO	50.3	9.7	10.1	9.6	9.9	10.4

394 The calculated molar ratio of Ru/Y/Ho/Dy/Gd/Pr is 1.00: 0.19: 0.20: 0.19: 0.20: 0.21

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410 Table S3. Comparison of OER activities for the Ru-based catalysts in acid media.

Catalysts	η_{10} (mV)	Mass activity@ η (A g ⁻¹ @ mV)	Electrolytes	References
HE-YRO	200	797.1@270	0.5M H ₂ SO ₄	This work
YRO	304	18.3@270	0.5M H ₂ SO ₄	This work
Y_{1.75}Ca_{0.25}Ru₂O_{7-δ}	275	597.85@270	0.5M H ₂ SO ₄	S1
Y_{1.85}Ba_{0.15}Ru₂O_{7-δ}	278	681.6@270	0.5M H ₂ SO ₄	S2
MoO_x@Y₂Ru₂O_{7-δ}	240	~300@270	0.1M HClO ₄	S3
Y₂RuMnO₇	260	~100@270	0.1M HClO ₄	S4
Y_{1.7}Sr_{0.3}Ru₂O₇	264	418@270	0.5M H ₂ SO ₄	S5
S_{II}-RuCoO NRs	231	783@270	0.5M H ₂ SO ₄	S6
RuIr-HEO	220	666.5@270	0.5M H ₂ SO ₄	S7
Ru₅W₁O_x	235	750@300	0.5M H ₂ SO ₄	S8
RuNi₂@G-250	227	120@300	0.5M H ₂ SO ₄	S9
Y_{1.75}Co_{0.25}Ru₂O_{7-δ}	275	~110@270	0.5M H ₂ SO ₄	S10
Ho₂Ru₂O₇	280	~590@320	0.1M HClO ₄	S11
Ru@IrO_x	282	644.8@330	0.5M H ₂ SO ₄	S12
Y₂[Ru_{1.6}Y_{0.4}] O_{7-δ}	~240	~480@250	0.5M H ₂ SO ₄	S13
Y_{1.85}Zn_{0.15}Ru₂O_{7-δ}	291	412.7@320	0.5M H ₂ SO ₄	S14
Au-Ru nanoparticles	220	-	0.5M H ₂ SO ₄	S15
RuTe₂ nanorods	245	-	0.5M H ₂ SO ₄	S16
RuRh@(RuRh)O₂	245	119.8@250	0.1M HClO ₄	S17

411 (η_{10} : The overpotential of catalysts at 10 mA cm⁻²)

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420 Table S4. XPS analysis of Ru 3p_{3/2} region of HE-YRO and YRO.

Species	HE-YRO		YRO	
	Peak position (eV)	Area (%)	Peak position (eV)	Area (%)
Ru ⁴⁺	463.25	76.6	463.25	85.9
Ru ³⁺	465.22	23.4	465.22	14.1
Ru ³⁺ /Ru ⁴⁺	0.31		0.16	

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436 Table S5. XPS analysis of O 1s region of HE-YRO and YRO.

Species	HE-YRO		YRO	
	Peak position (eV)	Area (%)	Peak position (eV)	Area (%)
O _L	529.1	27.55	529.1	28.00
O _{OH}	531.4	67.24	531.4	59.95
O _W	533.2	5.21	533.2	12.05
O _{OH} /O _L	2.44		2.14	

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