

Supplementary information

**Rational Design of RuO<sub>2</sub>/0.5CeO<sub>2</sub> Heterostructure as an Efficient and Stable Electrocatalyst for Acidic Water Splitting**

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## Supplementary Figures

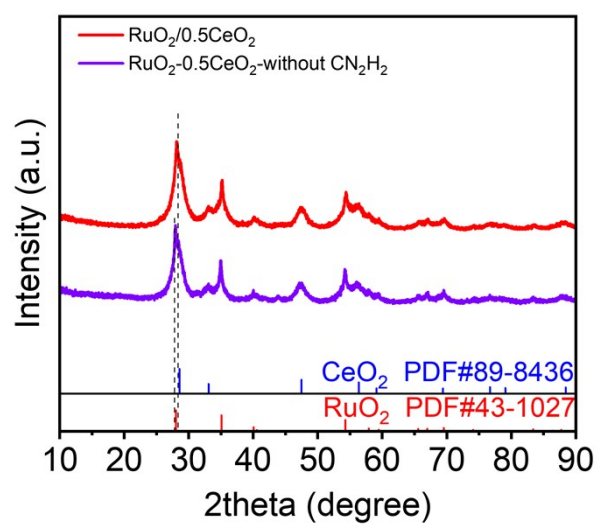


Fig. S1. XRD patterns of RuO<sub>2</sub>/0.5CeO<sub>2</sub> and RuO<sub>2</sub>-0.5CeO<sub>2</sub> (without CN<sub>2</sub>H<sub>2</sub> as precursor).

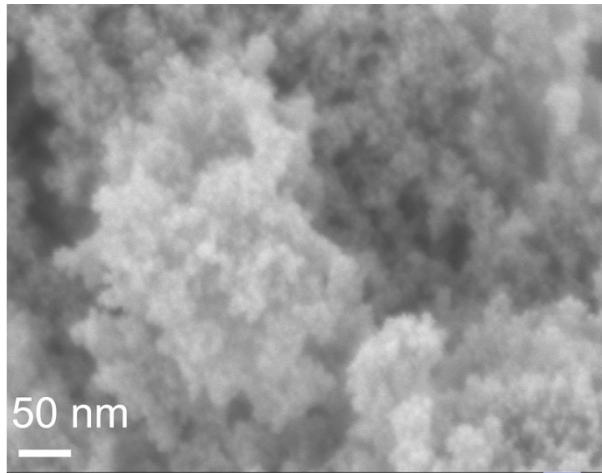


Fig. S2. SEM image of RuO<sub>2</sub>/0.5CeO<sub>2</sub>.

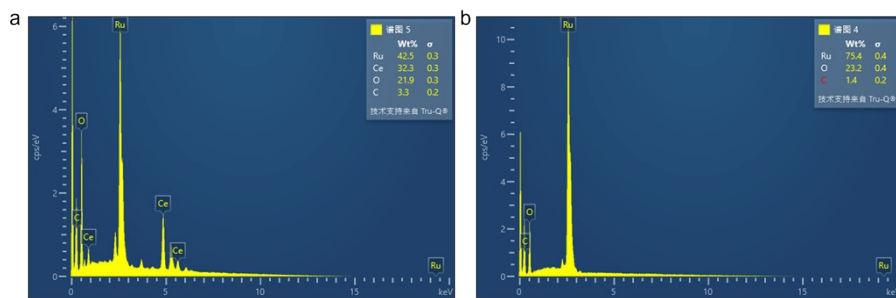


Fig. S3. SEM-EDS analysis of RuO<sub>2</sub>/0.5CeO<sub>2</sub> and commercial RuO<sub>2</sub>.

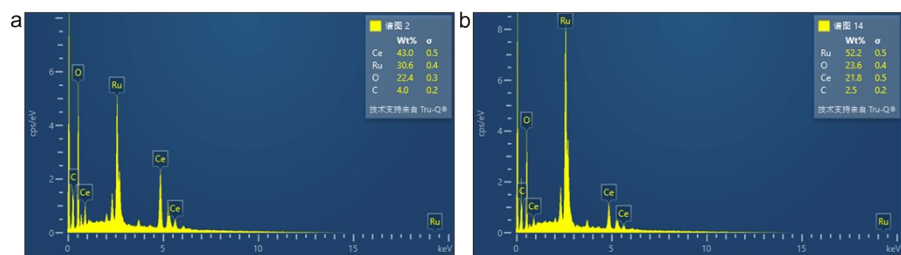


Fig. S4. SEM-EDS analysis of  $\text{RuO}_2/\text{CeO}_2$  and  $\text{RuO}_2/0.25\text{CeO}_2$ .

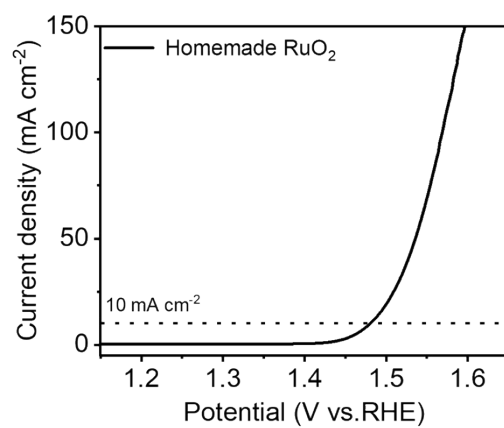


Fig. S5. LSV polarization curves of homemade RuO<sub>2</sub> with iR compensation.

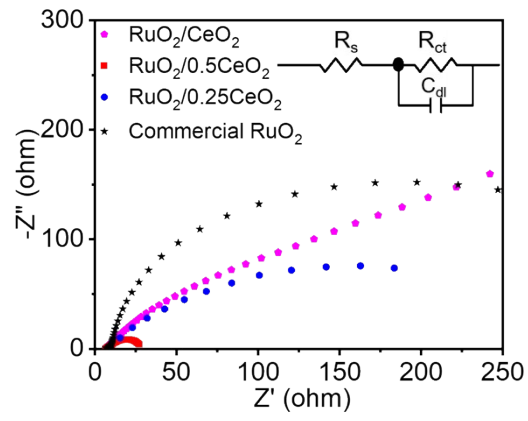


Fig. S6. EIS plot at a constant potential of 1.5 V vs. RHE.

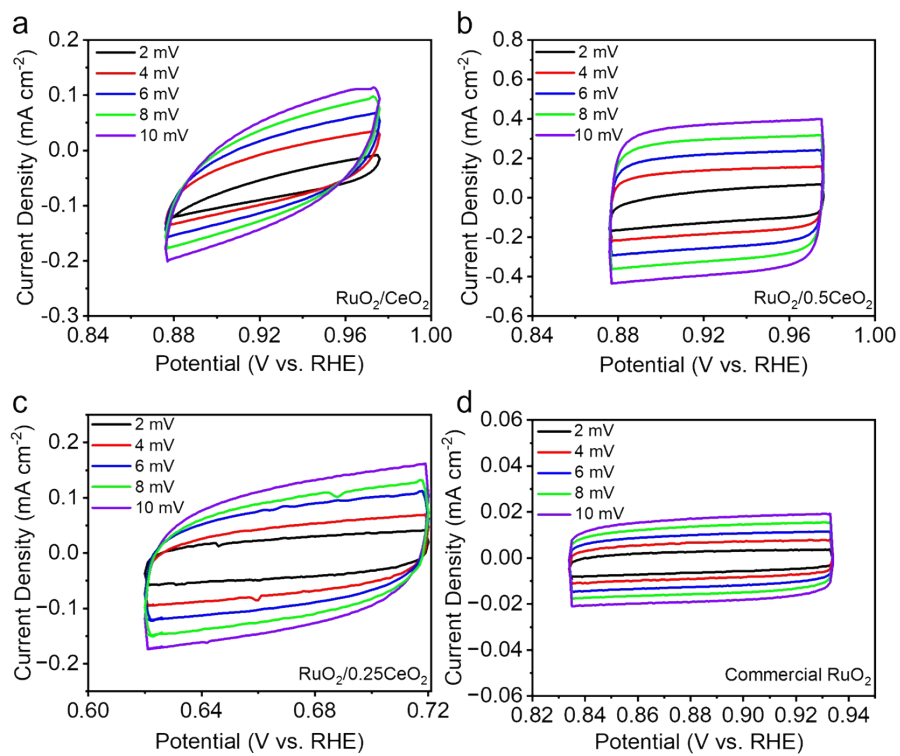


Fig. S7. Cyclic voltammograms curves of (a) RuO<sub>2</sub>/CeO<sub>2</sub>, (b) RuO<sub>2</sub>/0.5CeO<sub>2</sub>, (c) RuO<sub>2</sub>/0.25CeO<sub>2</sub>, and (d) commercial RuO<sub>2</sub> at scan rates from 2 to 10 mV s<sup>-1</sup> within the non-Faradaic potential range.

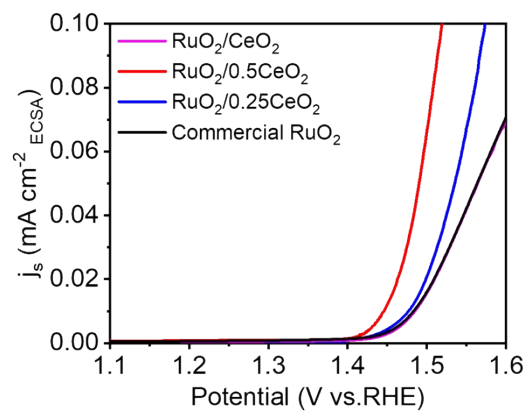


Fig. S8. OER polarization curves for RuO<sub>2</sub>/CeO<sub>2</sub>, RuO<sub>2</sub>/0.5CeO<sub>2</sub>, RuO<sub>2</sub>/0.25CeO<sub>2</sub>, commercial RuO<sub>2</sub> in acid media. The activities of catalysts were normalized to ECSA.

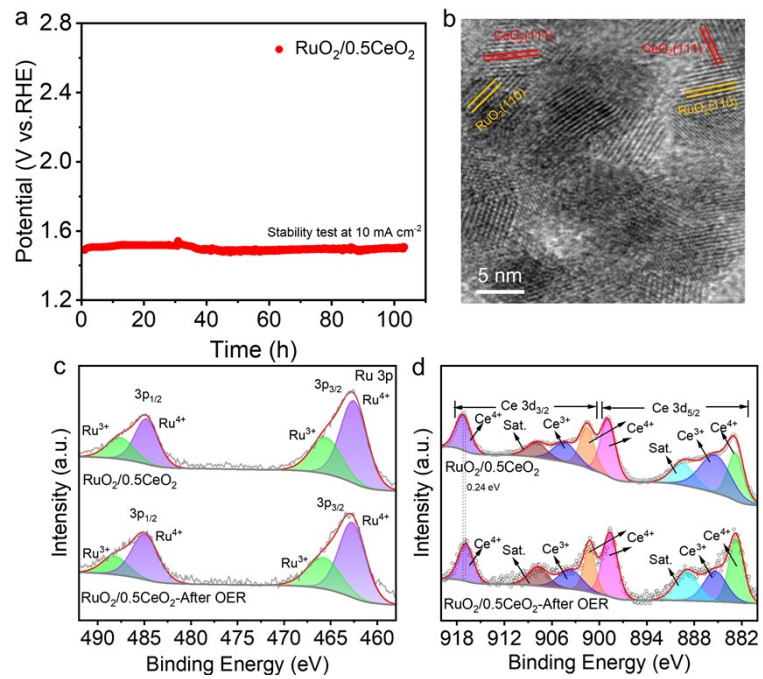


Fig. S9. (a) 100-hour durability test of RuO<sub>2</sub>/0.5CeO<sub>2</sub>. (b) High-magnification TEM of RuO<sub>2</sub>/0.5CeO<sub>2</sub> after 100-hour durability test. High-resolution XPS spectra of (c) Ru 3p and (d) Ce 3d for RuO<sub>2</sub>/0.5CeO<sub>2</sub> before and after 100-hour durability test.

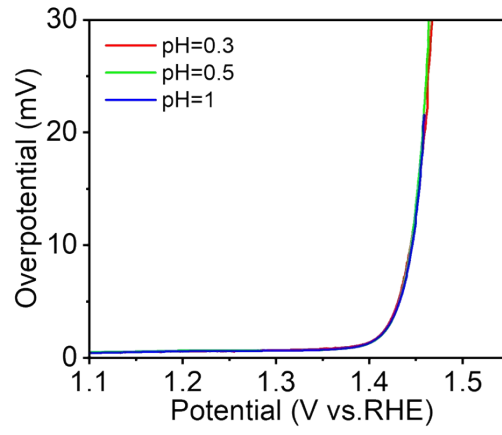


Fig. S10. OER polarization curves for RuO<sub>2</sub>/0.5CeO<sub>2</sub> in electrolytes of different pH.

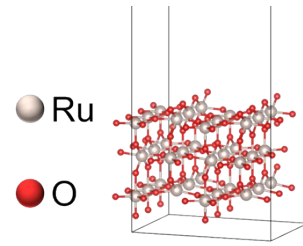


Fig. S11. Crystal structure model of RuO<sub>2</sub>.

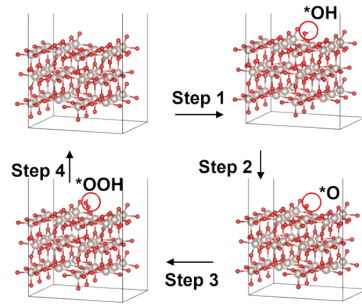


Fig. S12. Four-step AEM of OER for RuO<sub>2</sub>.

## Supplementary Tables

Table S1. The mass percent of Ru, Ce, O and C for RuO<sub>2</sub>/0.5CeO<sub>2</sub> and Commercial RuO<sub>2</sub> measured by SEM-EDS and ICP-OES.

Sample	SEM-EDS				ICP-OES
	Ru (wt%)	Ce (wt%)	O (wt%)	C (wt%)	Weight ratio of Ru/Ce
RuO <sub>2</sub> /CeO <sub>2</sub>	30.6	43.0	22.4	4.0	0.71
RuO <sub>2</sub> /0.5CeO <sub>2</sub>	42.4	31.3	21.93	3.2	1.43
RuO <sub>2</sub> /0.25CeO <sub>2</sub>	52.2	21.8	23.6	2.5	2.46
Commercial RuO <sub>2</sub>	75.39	\	23.2	1.4	\

Table S2. Binding energy positions and relative concentrations of O 1s for RuO<sub>2</sub>/0.5CeO<sub>2</sub> and RuO<sub>2</sub> from high-resolution XPS analysis.

Sample	Binding energy positions (eV)				Relative concentrations (%)			
	O <sub>L</sub>	-OH	H <sub>2</sub> O	C=O/C-O	O <sub>L</sub>	-OH	H <sub>2</sub> O	C=O/C-O
RuO <sub>2</sub> /0.5CeO <sub>2</sub>	529.45	530.82	532.02	533.19	28.15	40.69	22.48	8.68
RuO <sub>2</sub>	529.31	530.26	531.57	533.16	41.67	32.26	20.25	5.82

Table S3. Summary of OER performance of state-of-the-art Ru-based catalysts.

Catalyst	$\eta_{10}$ (mV)	Stability (h@10 mA cm <sup>-2</sup> )	PEMWE (V@1 A cm <sup>-2</sup> )	PEMWE	Reference
				Durability (h@mA cm <sup>-2</sup> ) <sup>2)</sup>	
<b>RuO<sub>2</sub>/0.5CeO<sub>2</sub></b>	<b>214</b>	<b>550</b>	<b>1.59</b>	<b>300 h@500 mA cm<sup>-2</sup></b>	<b>This Work</b>
h-RuO <sub>2-6</sub>	247	600	1.63	130 h@200 mA cm <sup>-2</sup>	<sup>1</sup>

MD-RuZrCoCrCeO <sub>2</sub>	179	800	1.66	600 h@200 mA cm <sup>-2</sup>	2
Ru-Co <sub>3</sub> O <sub>4-x</sub>	198	230	1.85	200 h@500 mA cm <sup>-2</sup>	3
RuO <sub>2</sub> @NCC	215	327	1.65	400 h@250 mA cm <sup>-2</sup>	4
Ru <sub>clusters</sub> /ZnCo <sub>2</sub> O <sub>4</sub>	200	725	1.697	275 h@200 mA cm <sup>-2</sup>	5
RuO <sub>x</sub> -GDY	193	300	1.73	200 h@100 mA cm <sup>-2</sup>	6
a-Ge-RuO <sub>2</sub>	205	500	1.62	400 h@200 mA cm <sup>-2</sup>	7
RuIr-Co <sub>3</sub> O <sub>4</sub>	220	610	1.67@0.5A cm <sup>-2</sup>	500 h@100 mA cm <sup>-2</sup>	8
Tm-RuO <sub>2</sub>	201	300	1.84	N.A.	9
Mn-RuO <sub>2</sub>	209	180	1.9	40 h@100 mA cm <sup>-2</sup>	10
H-Mn <sub>0.1</sub> Ru <sub>0.9</sub> O <sub>2</sub>	169	500	1.65	400 h@500 mA cm <sup>-2</sup>	11
Y-RuO <sub>2</sub>	227	40	1.59	100 h@200 mA cm <sup>-2</sup>	12
RuBa <sub>x</sub> Co <sub>3-x</sub> O <sub>4</sub>	219	N.A.	N.A.	50 h@50 mA cm <sup>-2</sup>	13
W <sub>0.2</sub> Er <sub>0.1</sub> Ru <sub>0.7</sub> O <sub>2-δ</sub>	168	250	N.A.	N.A.	14
Mn <sub>0.73</sub> Ru <sub>0.27</sub> O <sub>2-δ</sub>	208	10	N.A.	N.A.	15
SSPt-RuO <sub>2</sub> HNSs	228	100	N.A.	N.A.	16

Table S4. Concentration of Ru and Ce in the electrolyte and their mass loss percentages after chronopotentiometry stability tests.

Sample	Sample amount	Concentrations of Ru/Ce (μg mL <sup>-1</sup> )	Loss mass percent of Ru/Ce
RuO <sub>2</sub> /0.5CeO <sub>2</sub>	1mg (0.42mg Ru)	0.013/1.43	0.115%/16.4%
Commercial RuO <sub>2</sub>	1mg (0.75mg Ru)	0.037	0.45%

Table S5. Comparison of representative previously reported Ru-CeO<sub>2</sub> heterostructures and Ce-modified Ru catalysts for acidic OER.

Catalyst	Synthesis method	Support type	$\eta_{10}$ (mV)	Stability (h@10 mA cm <sup>-2</sup> )	PEMWE performance	PEMWE Durability	Reference
<b>RuO<sub>2</sub>/0.5CeO<sub>2</sub></b>	<b>impregnation-pyrolysis</b>	\	<b>214</b>	<b>550</b>	<b>1.59 V@1 A cm<sup>-2</sup></b>	<b>300 h@500 mA cm<sup>-2</sup></b>	<b>This Work</b>
RuO <sub>2</sub> -CeO <sub>2</sub> -CC	electrodeposit ion-calcination	Carbon cloth	180	1000	N.A.	N.A.	17
RuO <sub>2</sub> /CeO <sub>2</sub> @C	hydrothermal	Carbon sphere	170	100	1.54 V@0.01 A cm <sup>-2</sup>	100 h@10 mA cm <sup>-2</sup>	18
Ce-RuO <sub>2</sub>	glucose-assisted pyrolysis	\	191	25	N.A.	N.A.	19
Ce-RuO <sub>2</sub>	PVP-mediated pyrolysis	\	226	20	N.A.	N.A.	20
Ce@RuO <sub>2</sub> /CoNC	hydrothermal	CoNC	150	1000	1.56 V@0.2 A cm <sup>-2</sup>	1000 h@200 mA cm <sup>-2</sup>	21
Ru <sub>3</sub> MoCeO <sub>x</sub>	glucose-assisted pyrolysis	\	160	100	N.A.	N.A.	22
Ru <sub>0.85</sub> Ce <sub>0.15</sub> O <sub>x</sub>	glucose-blowing method	\	146	200	1.71 V@1 A cm <sup>-2</sup>	100 h@200 mA cm <sup>-2</sup>	23

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