

### Supplementary Information

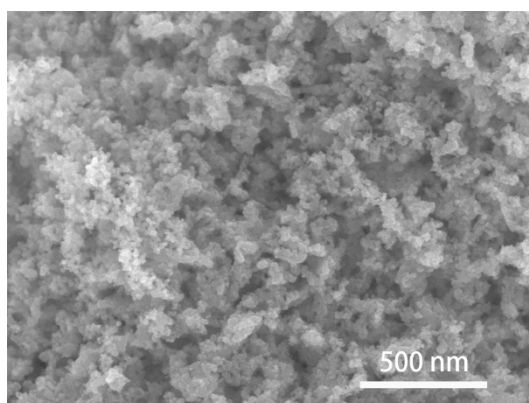


Figure S1. SEM image of Ru<sub>0.9</sub>Cr<sub>0.1</sub>O<sub>2</sub>.

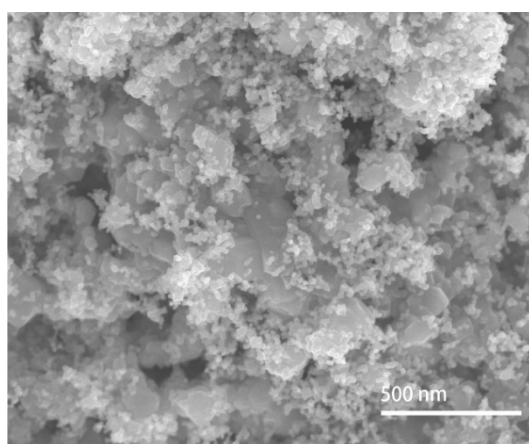


Figure S2. SEM image of RuO<sub>2</sub>.

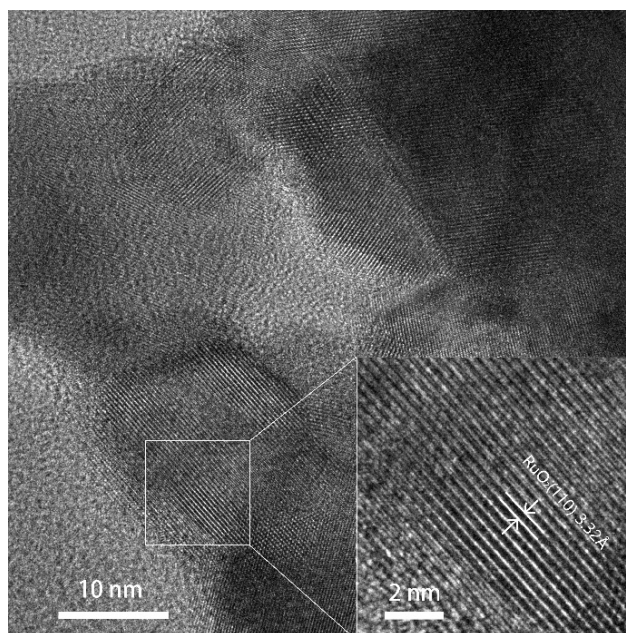


Figure S3. HRTEM images of RuO<sub>2</sub>.

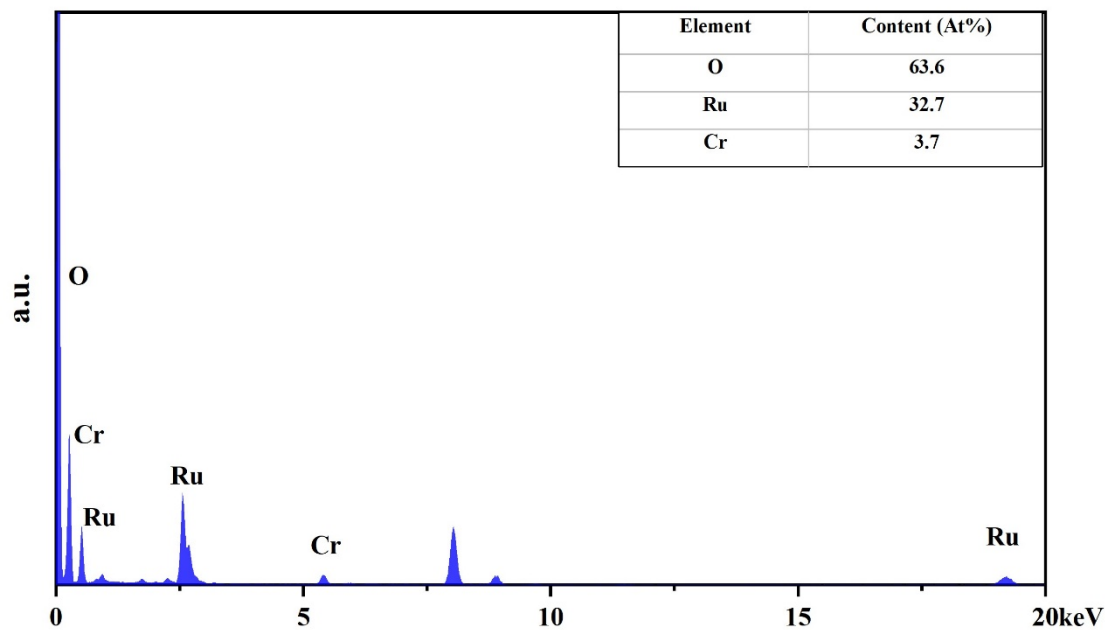


Figure S4. EDS elemental spectra of  $\text{Ru}_{0.9}\text{Cr}_{0.1}\text{O}_2$ .

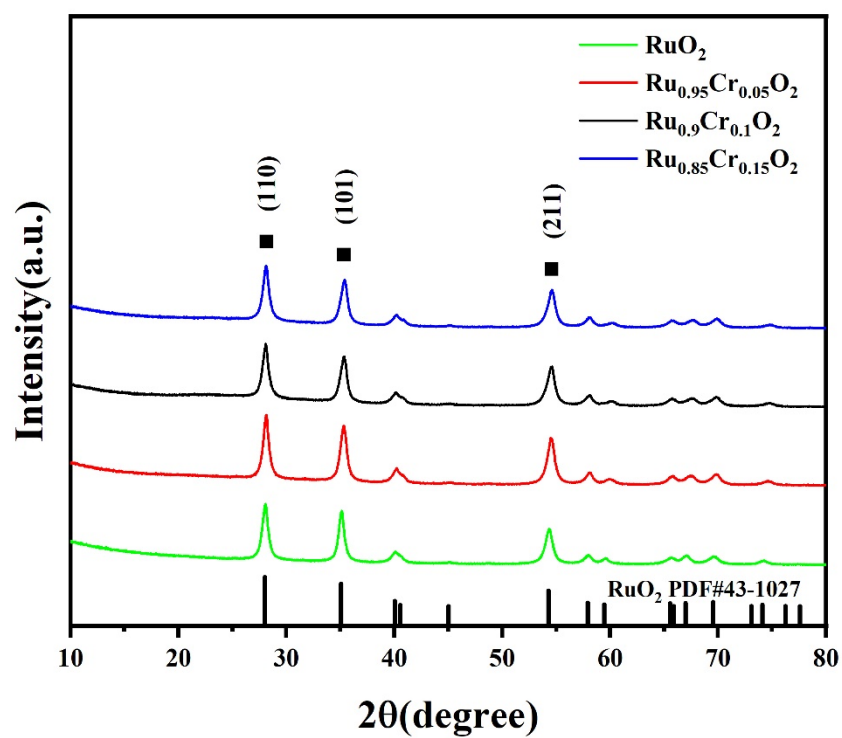


Figure S5. XRD of unmodified and different Cr-doped  $\text{RuO}_2$ .

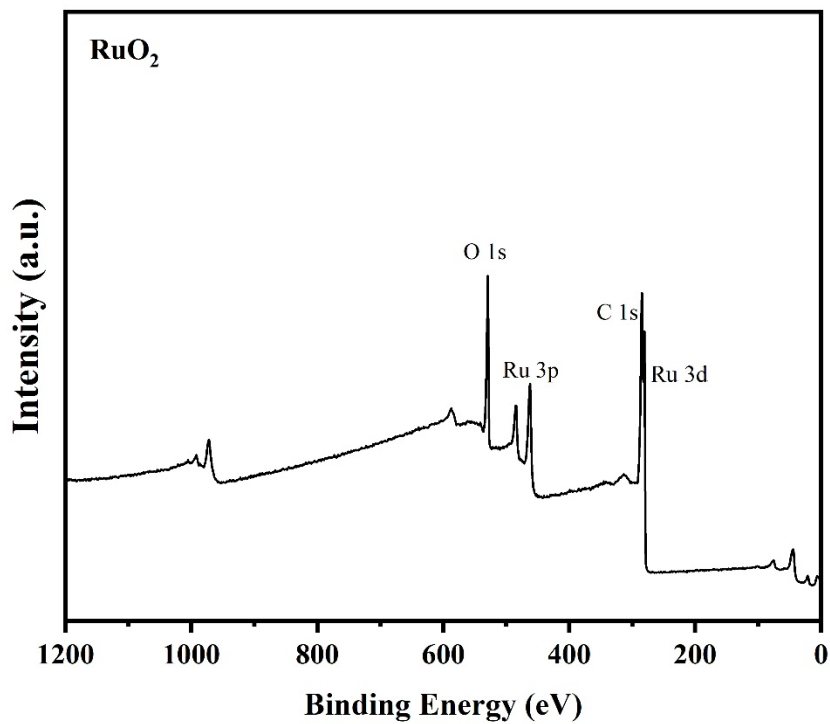


Figure S6. XPS survey pattern of RuO<sub>2</sub>.

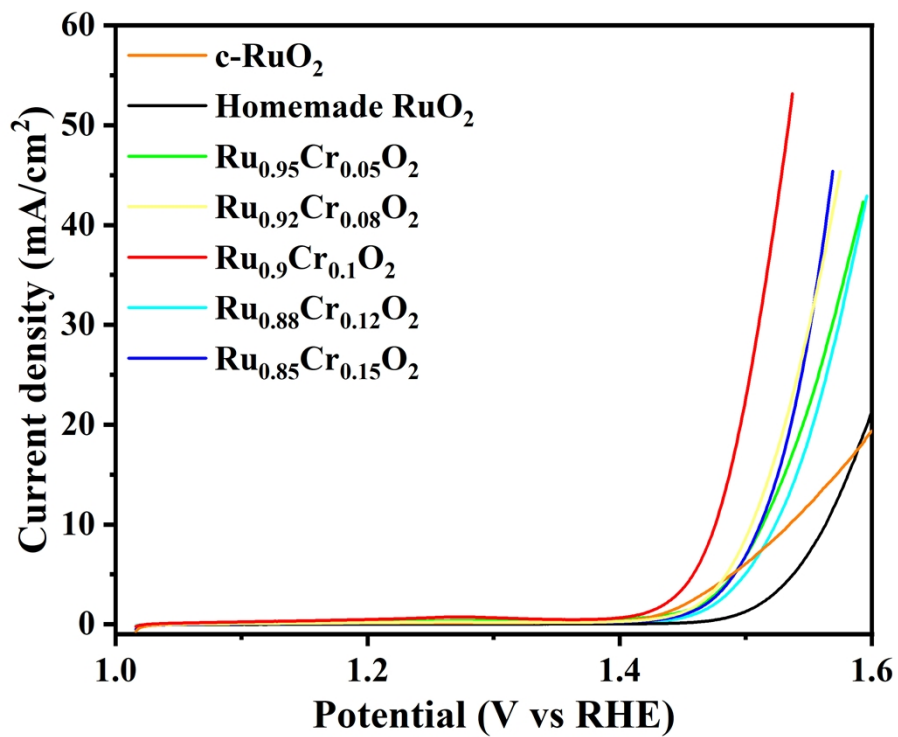


Figure S7. LSV curves of RuO<sub>2</sub> with different Cr-doped.

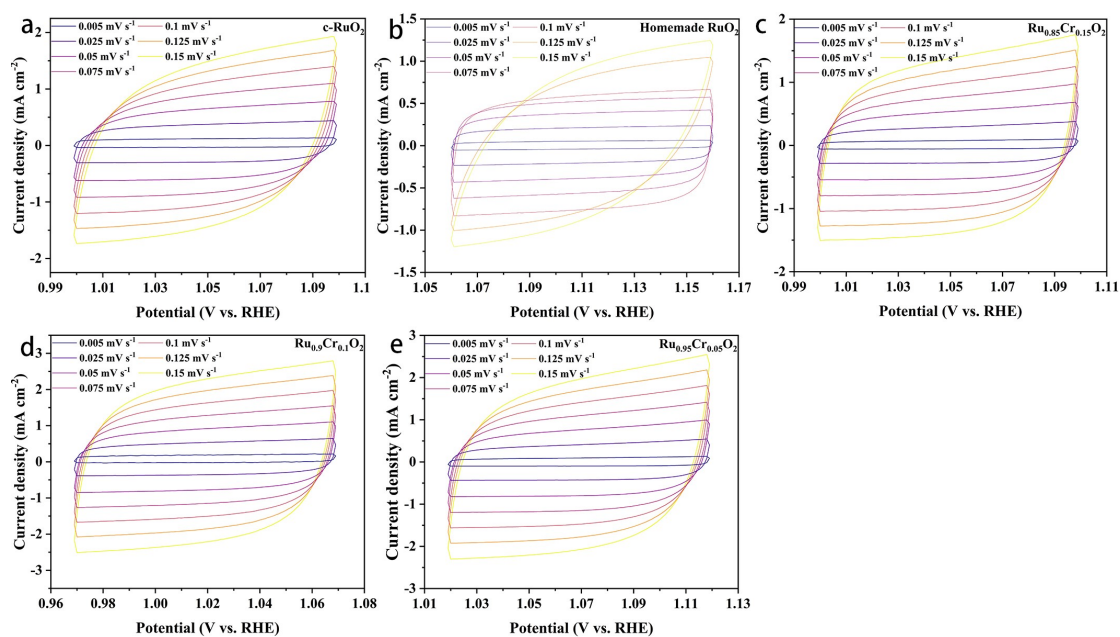


Figure S8. CV curves of (a) c-RuO<sub>2</sub>, (b) homemade RuO<sub>2</sub>, (c) Ru<sub>0.9</sub>Cr<sub>0.1</sub>O<sub>2</sub>, (d) Ru<sub>0.9</sub>Cr<sub>0.1</sub>O<sub>2</sub> and (e) Ru<sub>0.9</sub>Cr<sub>0.1</sub>O<sub>2</sub> in the non-Faradaic region under different scan rates in pH=0 H<sub>2</sub>SO<sub>4</sub>.

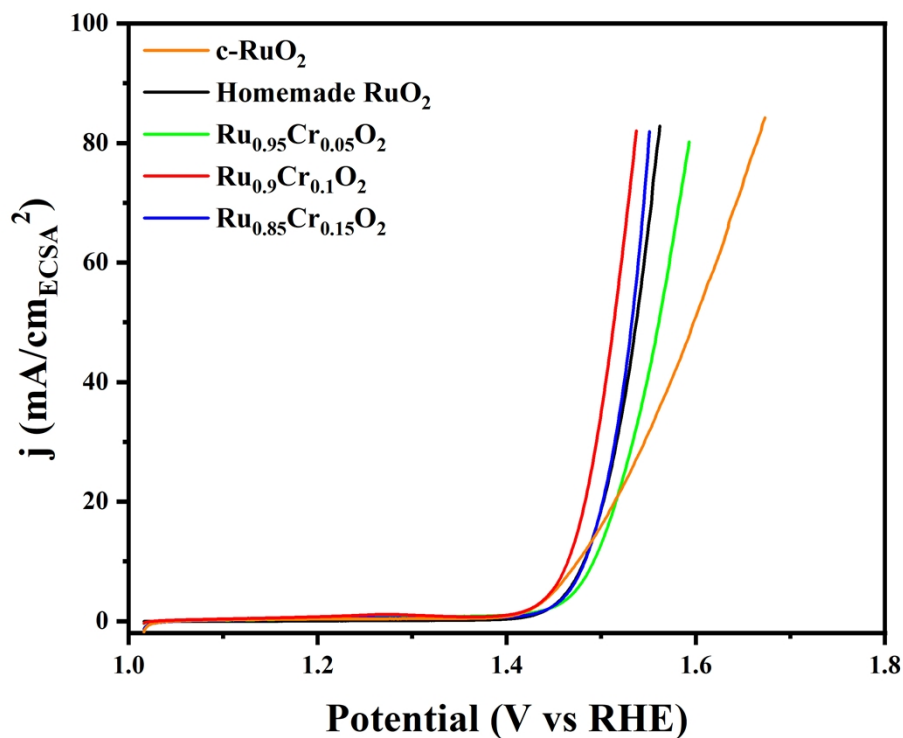


Figure S9. LSV curves after ECSA normalization.

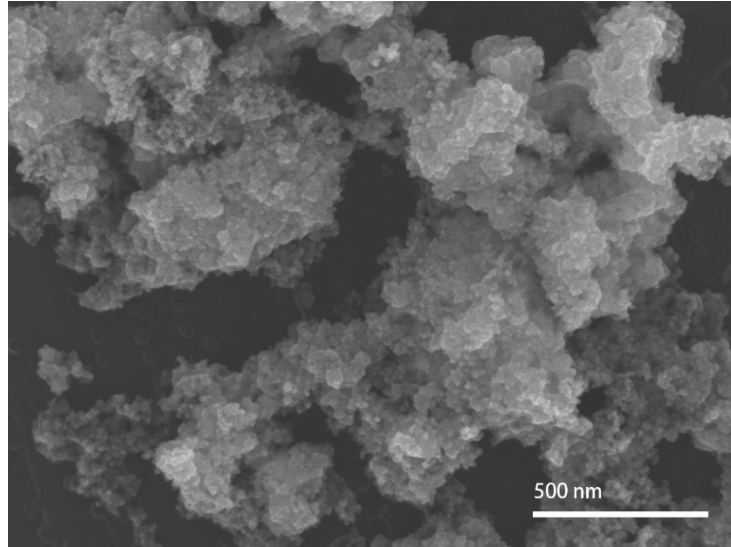


Figure S10. SEM image of Ru<sub>0.9</sub>Cr<sub>0.1</sub>O<sub>2</sub> after 8 h CP test.

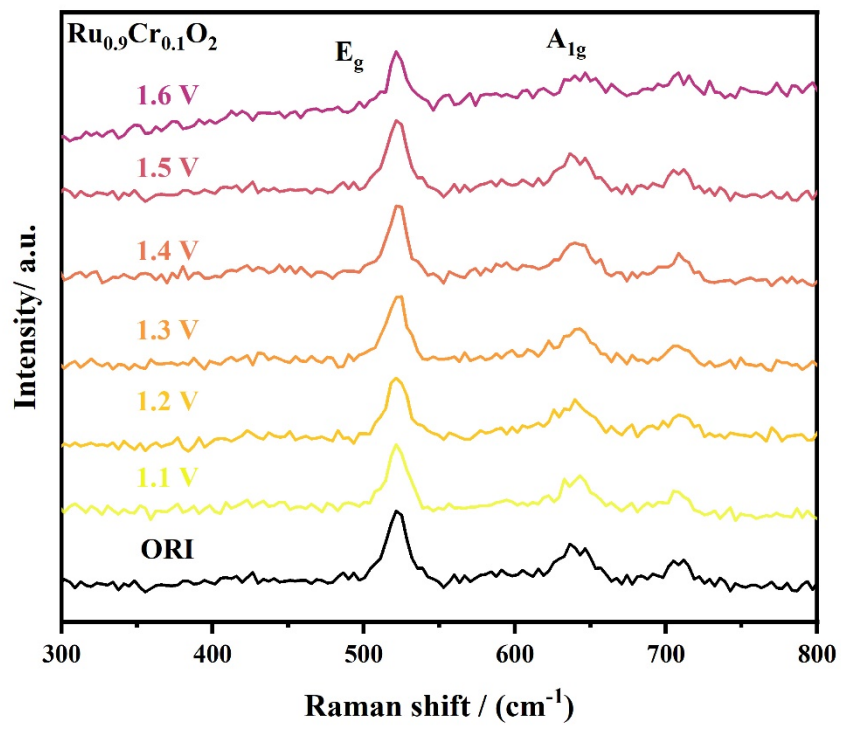


Figure S11. Operando Raman spectra obtained under various applied potential on Ru<sub>0.9</sub>Cr<sub>0.1</sub>O<sub>2</sub>.

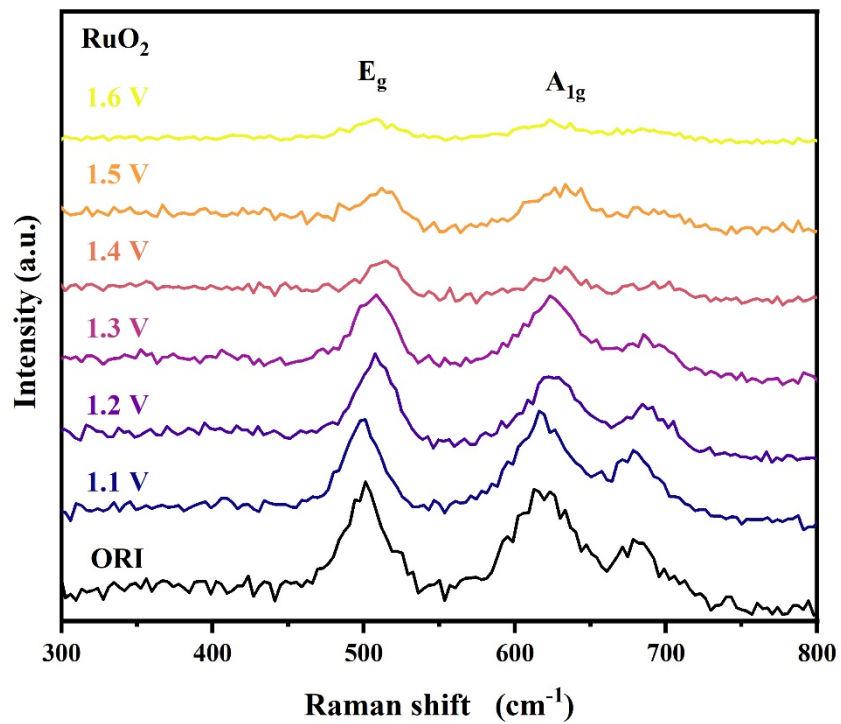


Figure S12. Operando Raman spectra obtained under various applied potential on RuO<sub>2</sub>.

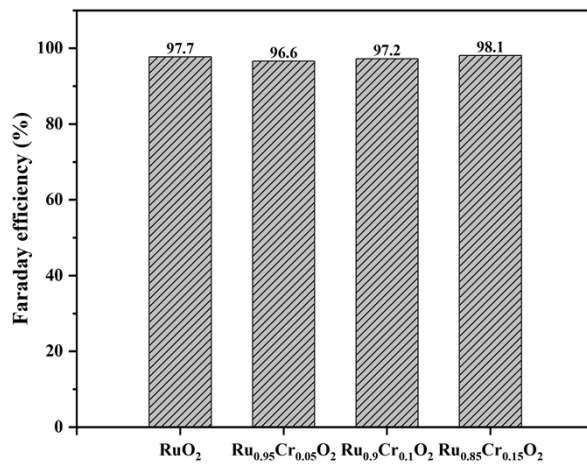


Figure S13. O<sub>2</sub> Faraday efficiencies of different catalysts at 10 mA cm<sup>-2</sup>.

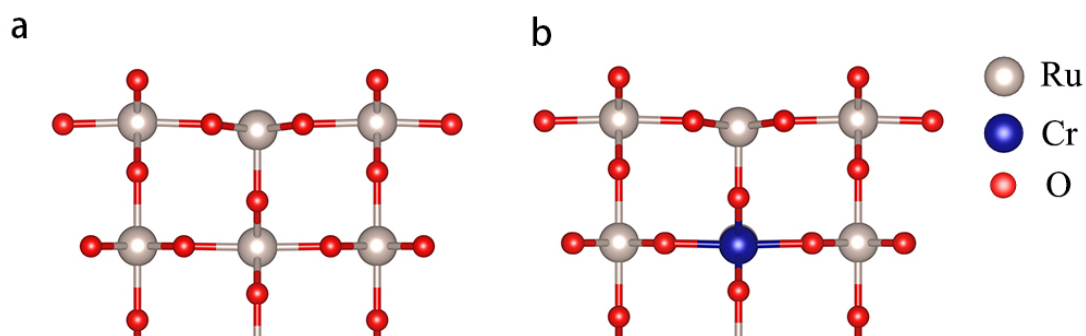


Figure S14. Model for DFT calculations of (a)RuO<sub>2</sub> and (b)Ru<sub>0.9</sub>Cr<sub>0.1</sub>O<sub>2</sub>, where the grey spheres are Ru, the red ones are O and the blue ones are Cr.

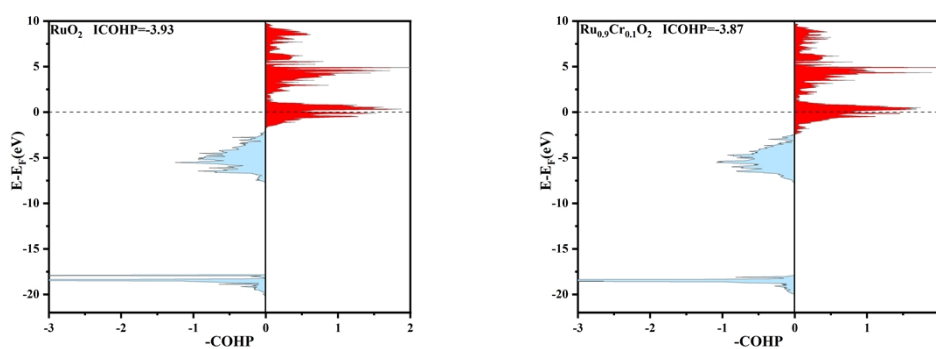


Figure S15. Description of bonding-anti-bonding roles of Ru-O bond in RuO<sub>2</sub> and Ru<sub>0.9</sub>Cr<sub>0.1</sub>O<sub>2</sub> by COHP.

Table S1. ICP-MS results of the electrolytes of RuO<sub>2</sub> and Ru<sub>0.9</sub>Cr<sub>0.1</sub>O<sub>2</sub> after a half-hour CP test.

Sample number	element	elemental concentration C (μg/L)
RuO <sub>2</sub>	Ru	193.963
Ru <sub>0.9</sub> Cr <sub>0.1</sub> O <sub>2</sub>	Cr	127.047
	Ru	15.683

Table S2. Formation energy of surface Ru defects in RuO<sub>2</sub> and Ru<sub>0.9</sub>Cr<sub>0.1</sub>O<sub>2</sub>.

	formation energy of surface Ru defects
RuO <sub>2</sub>	2.40 eV
Ru <sub>0.9</sub> Cr <sub>0.1</sub> O <sub>2</sub>	2.63 eV

Table S3. d-band center of Ru PDOS and p-band center of O PDOS of RuO<sub>2</sub> and Ru<sub>0.9</sub>Cr<sub>0.1</sub>O<sub>2</sub>.

	d-band center Ru PDOS	p-band center O PDOS
RuO <sub>2</sub>	-1.085 eV	-1.237 eV
Ru <sub>0.9</sub> Cr <sub>0.1</sub> O <sub>2</sub>	-2.726 eV	-2.975 eV