

Supplementary Materials

Reasonable Design of MXene-Supported Dual-Atom Catalysts with High Catalytic Activity for Hydrogen Evolution and Oxygen Evolution Reaction: A First-Principles Investigation

Erpeng Wang, Miaoqi Guo, Jian Zhou * and Zhimei Sun *

School of Materials Science and Engineering, Beihang University, Beijing 100191, China

* Correspondence: jzhou@buaa.edu.cn (J.Z.); zmsun@buaa.edu.cn (Z.S.)

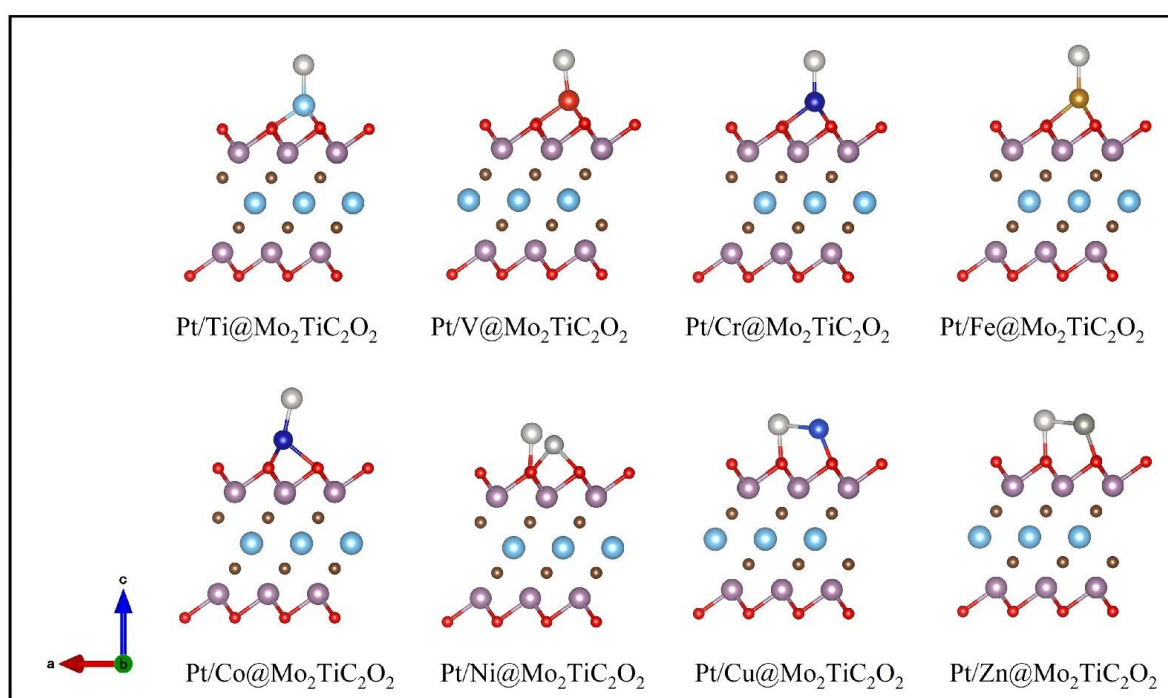
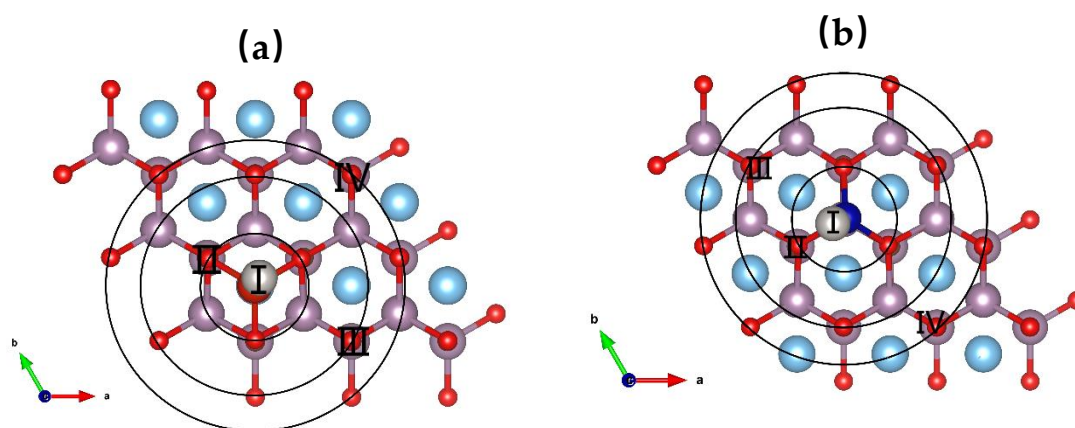


Figure S1. The most stable structures diagram for Pt/TM@Mo₂TiC₂O₂ h-DACs (TM = Ti, V, Cr, Fe, Co, Ni, Cu and Zn).



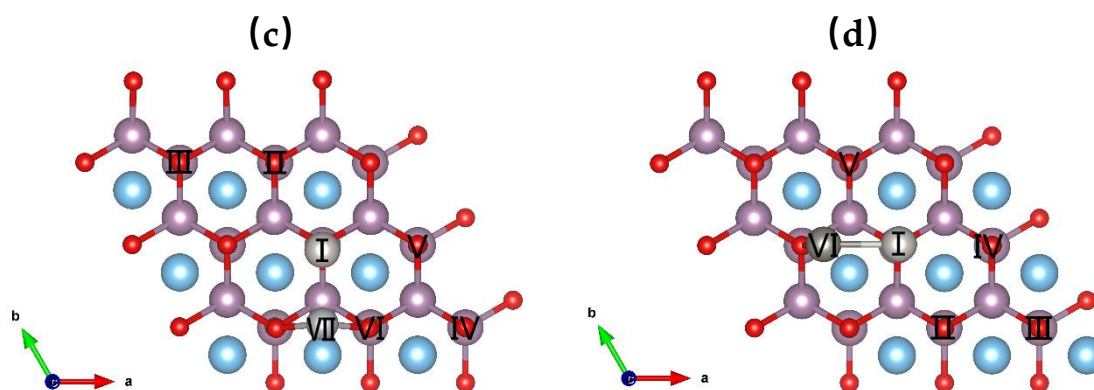


Figure S2. The possible H proton adsorption sites on the (a) Pt/TM@Mo₂TiC₂O₂ h-DACs surface (TM=Ti, V, Cr and Fe); (b) Pt/Co@Mo₂TiC₂O₂ h-DACs surface; (c) Pt/Ni@Mo₂TiC₂O₂ h-DACs surface; (d) Pt/TM@Mo₂TiC₂O₂ h-DACs surface (TM = Cu, Zn).

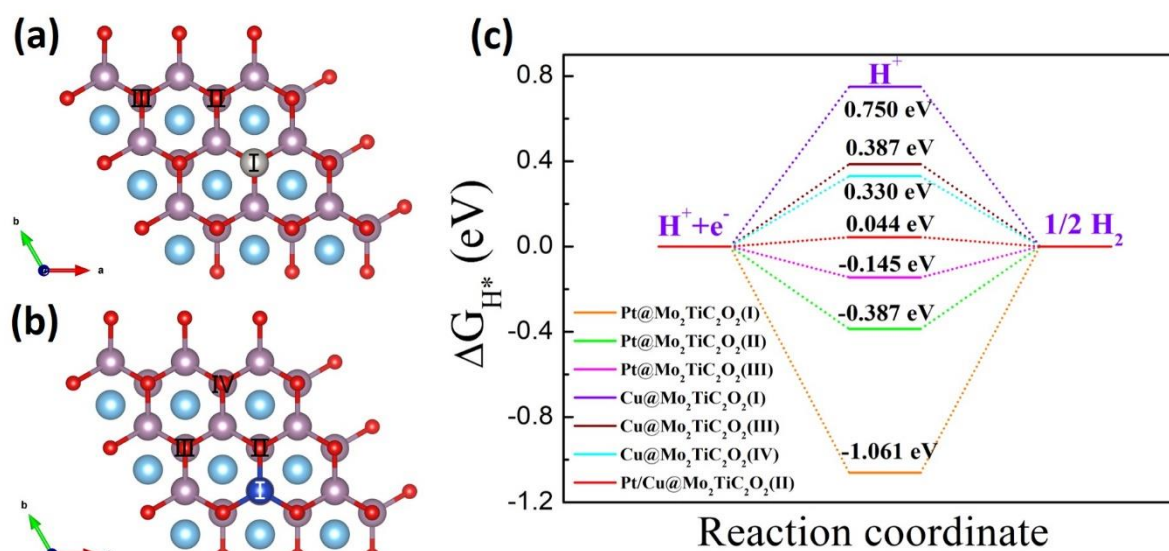


Figure S3. The possible H proton adsorption sites on the (a) Pt@Mo₂TiC₂O₂ SACs surface and (b) Cu@Mo₂TiC₂O₂ SACs. (c) Calculated Gibbs free energy profiles of HER for Pt@Mo₂TiC₂O₂ SACs, Cu@Mo₂TiC₂O₂ SACs and Pt/Cu@Mo₂TiC₂O₂ h-DACs.

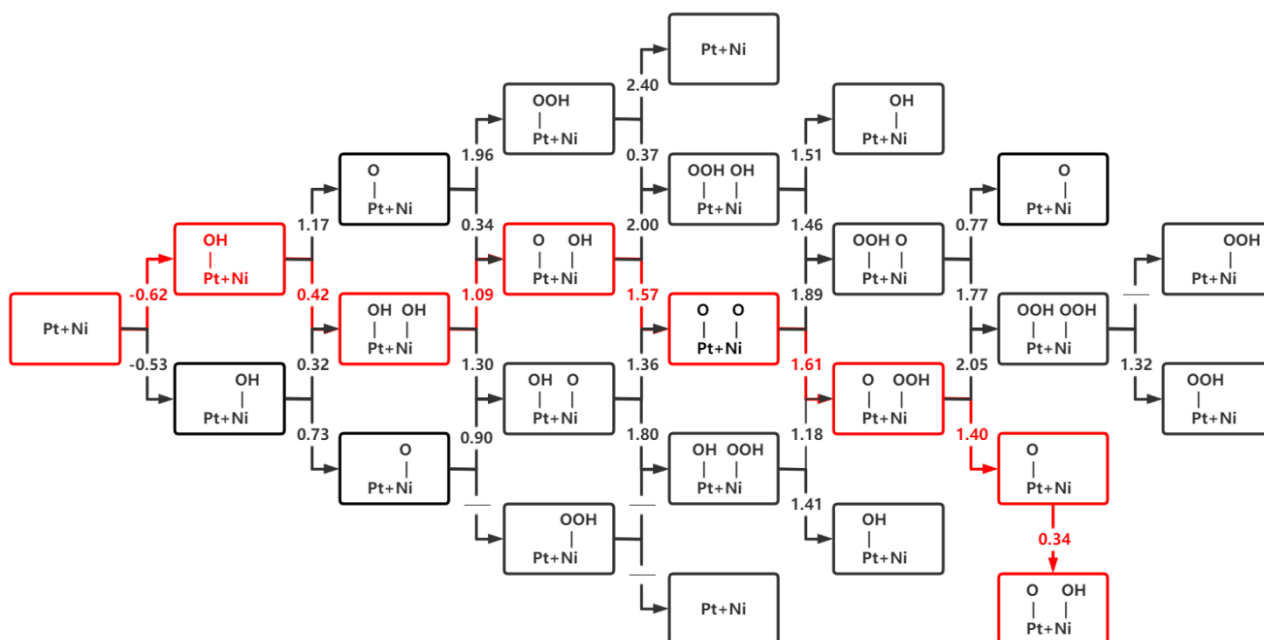


Figure S4. The diagram of all possible ORR pathways and corresponding Gibbs free energy on Pt/Ni@Mo₂TiC₂O₂ h-DACs, red lines represent the energy-favored reaction pathways.

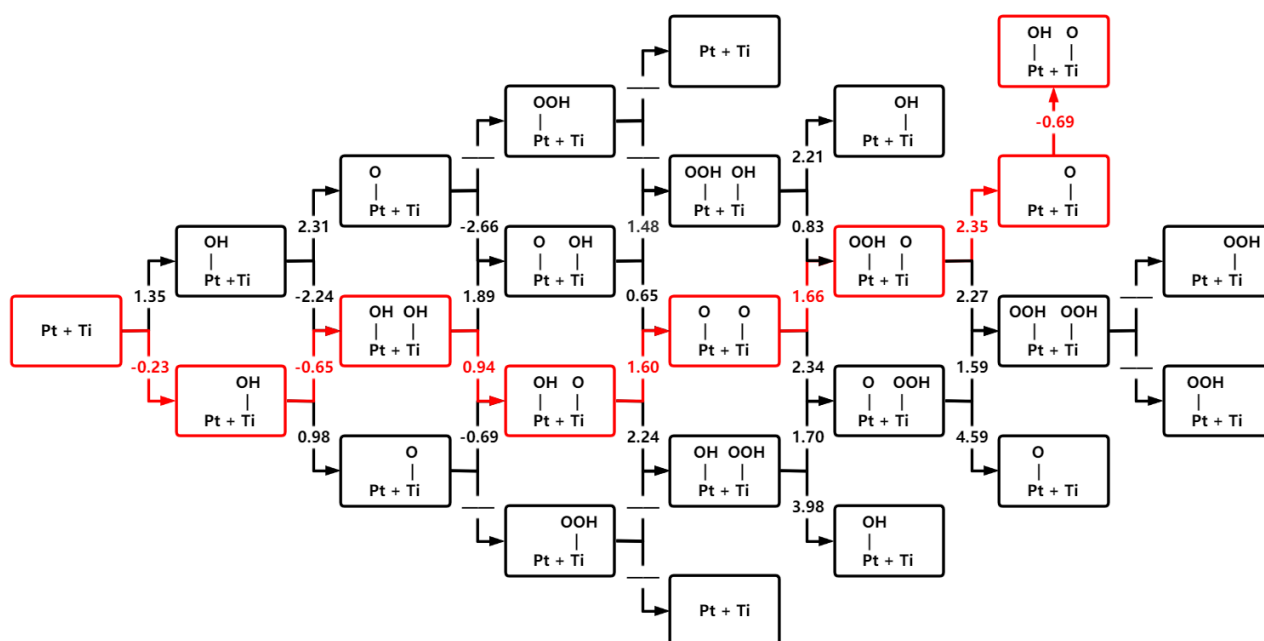


Figure S5. The diagram of all possible ORR pathways and corresponding Gibbs free energy on Pt/Ti@Mo₂TiC₂O₂ h-DACs, red lines represent the energy-favored reaction pathways.

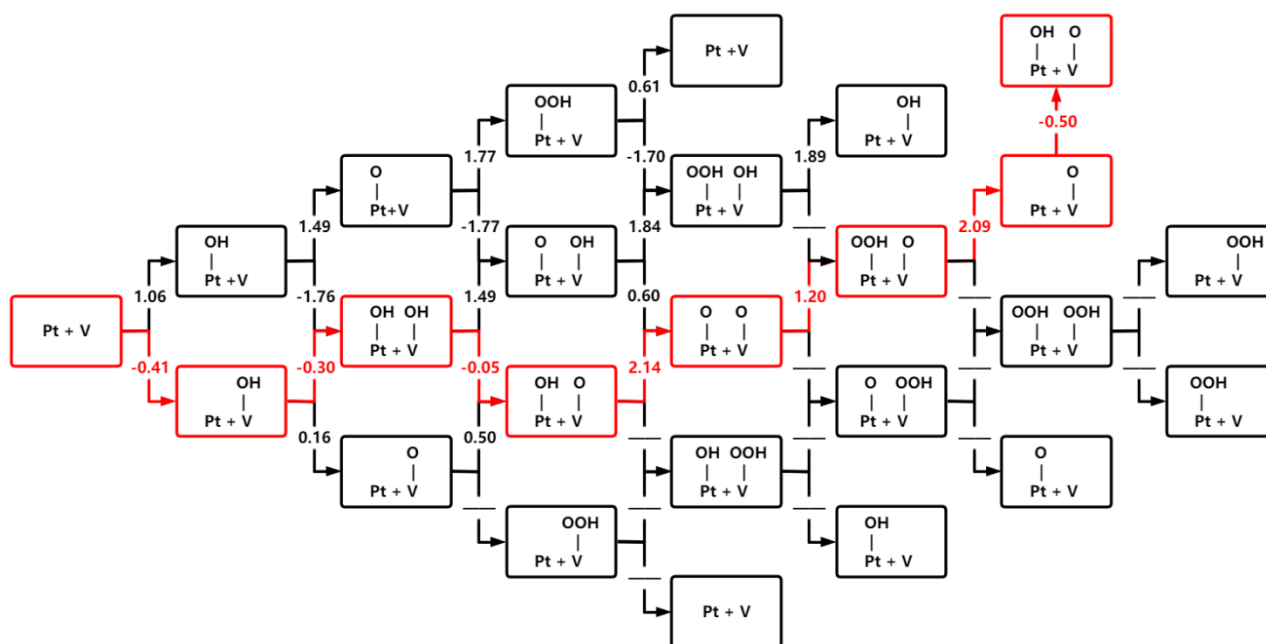


Figure S6. The diagram of all possible ORR pathways and corresponding Gibbs free energy on Pt/V@Mo₂TiC₂O₂h-DACs, red lines represent the energy-favored reaction pathways.

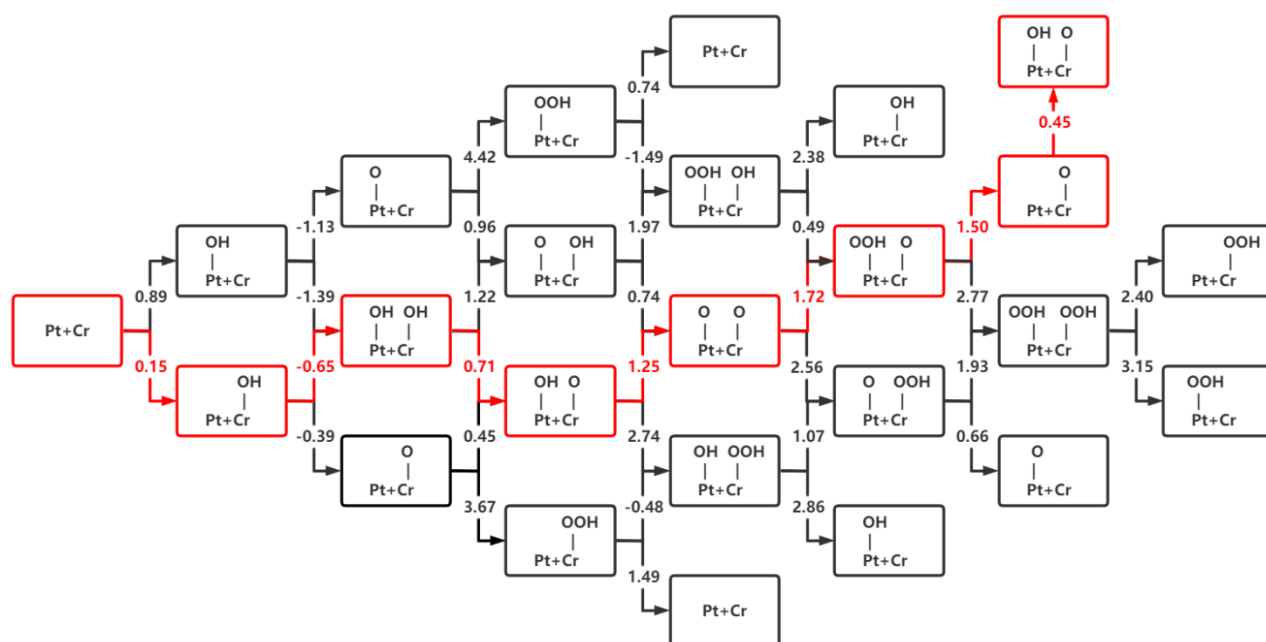


Figure S7. The diagram of all possible ORR pathways and corresponding Gibbs free energy on Pt/Cr@Mo₂TiC₂O₂h-DACs, red lines represent the energy-favored reaction pathways.

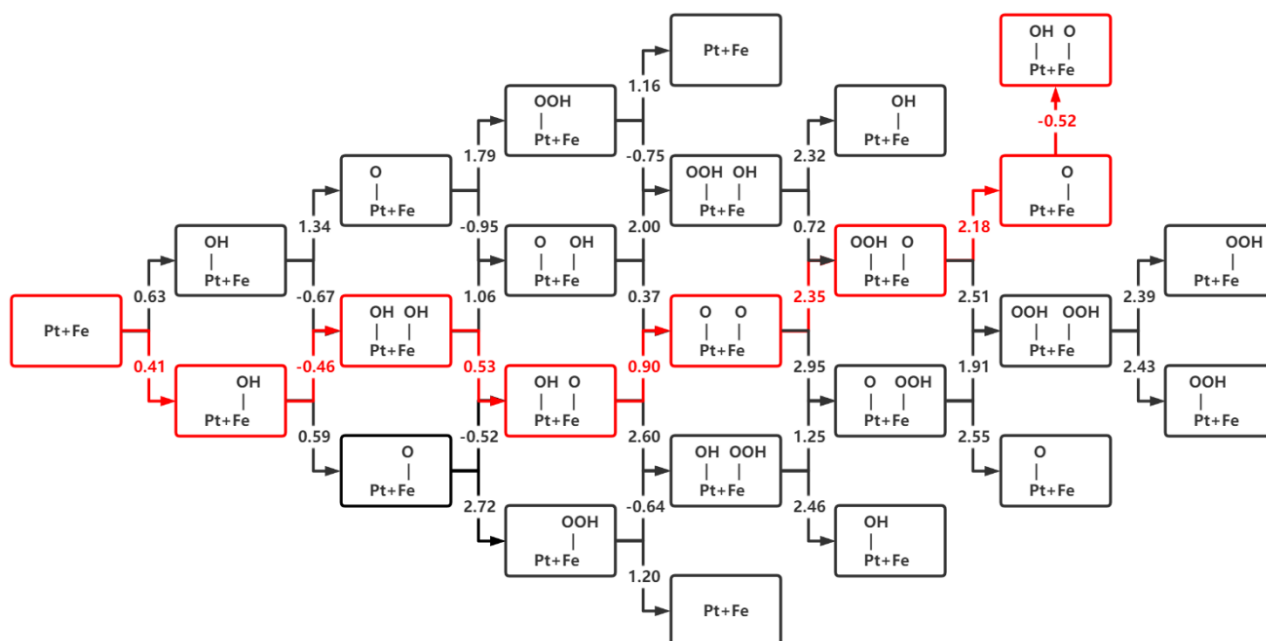


Figure S8. The diagram of all possible ORR pathways and corresponding Gibbs free energy on Pt/Fe@Mo₂TiC₂O₂h-DACs, red lines represent the energy-favored reaction pathways.

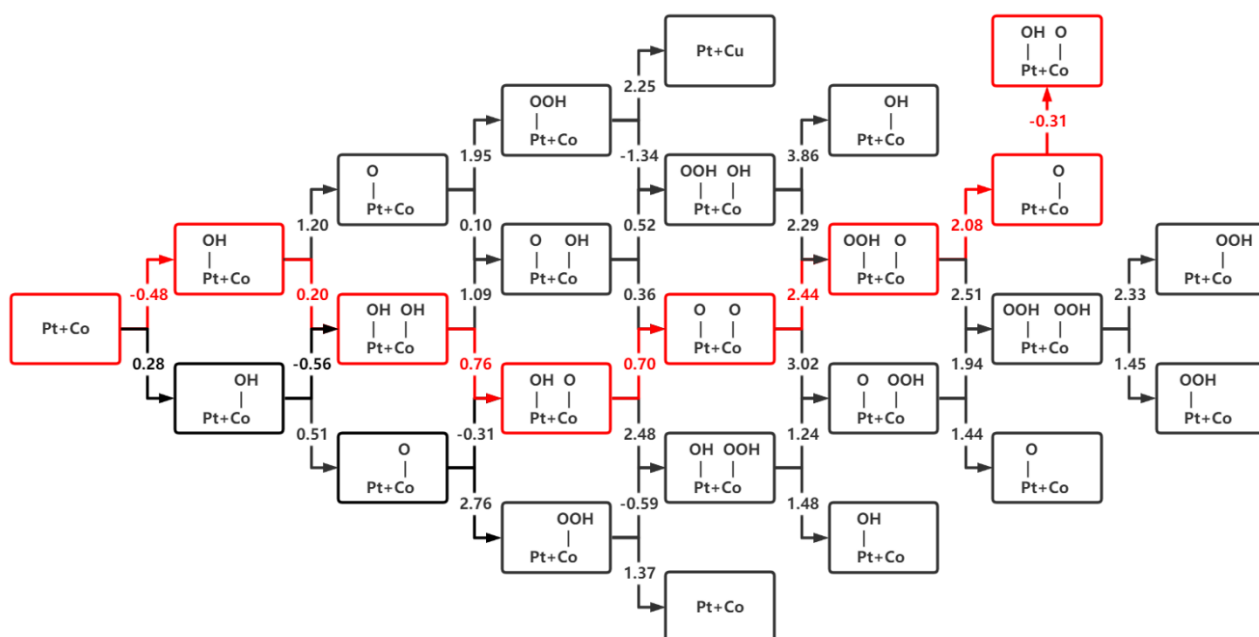


Figure S9. The diagram of all possible ORR pathways and corresponding Gibbs free energy on Pt/Co@Mo₂TiC₂O₂h-DACs, red lines represent the energy-favored reaction pathways.

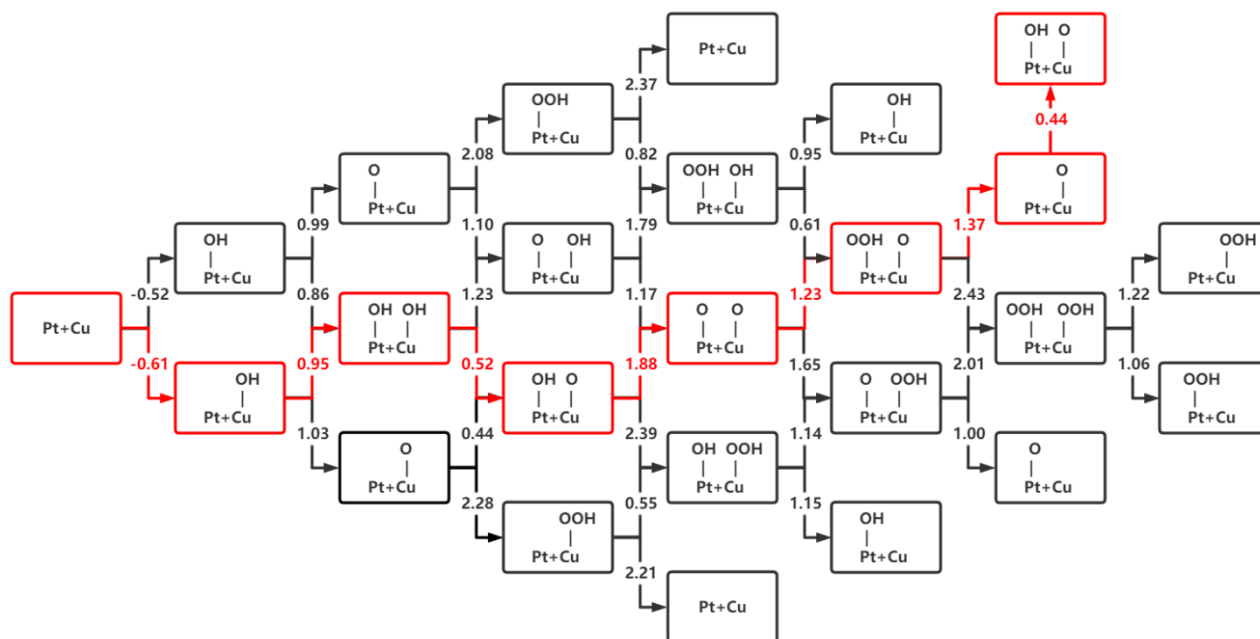


Figure S10. The diagram of all possible ORR pathways and corresponding Gibbs free energy on Pt/Cu@Mo₂TiC₂O₂ h-DACs, red lines represent the energy-favored reaction pathways.

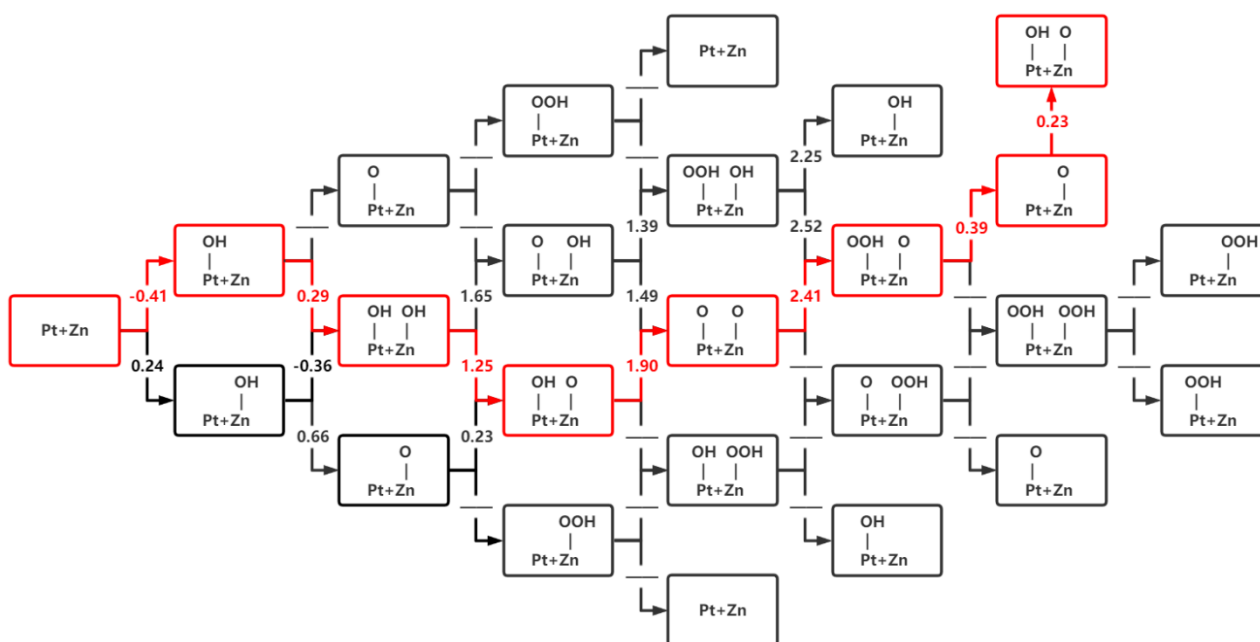


Figure S11. The diagram of all possible ORR pathways and corresponding Gibbs free energy on Pt/Zn@Mo₂TiC₂O₂ h-DACs, red lines represent the energy-favored reaction pathways.

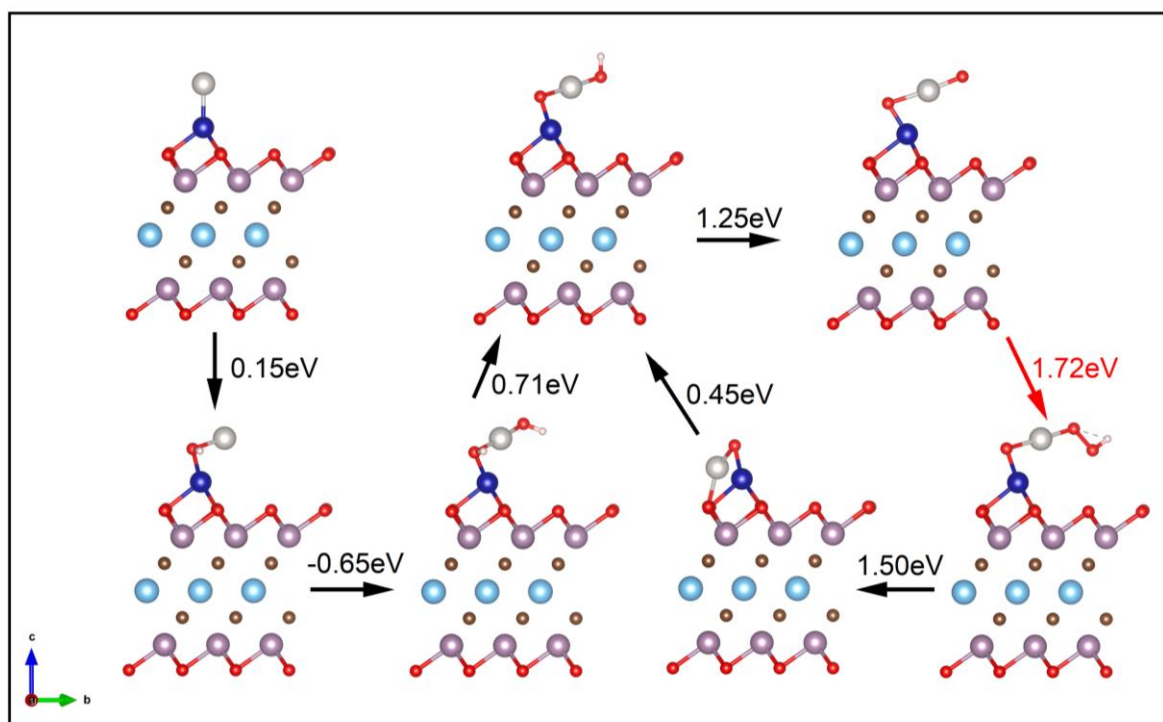


Figure S12. The configurations of oxygenated intermediates in energy-favored reactions pathways of OER on Pt/Cr@Mo₂TiC₂O₂h-DACs. The red line represents the most energy-consuming step.

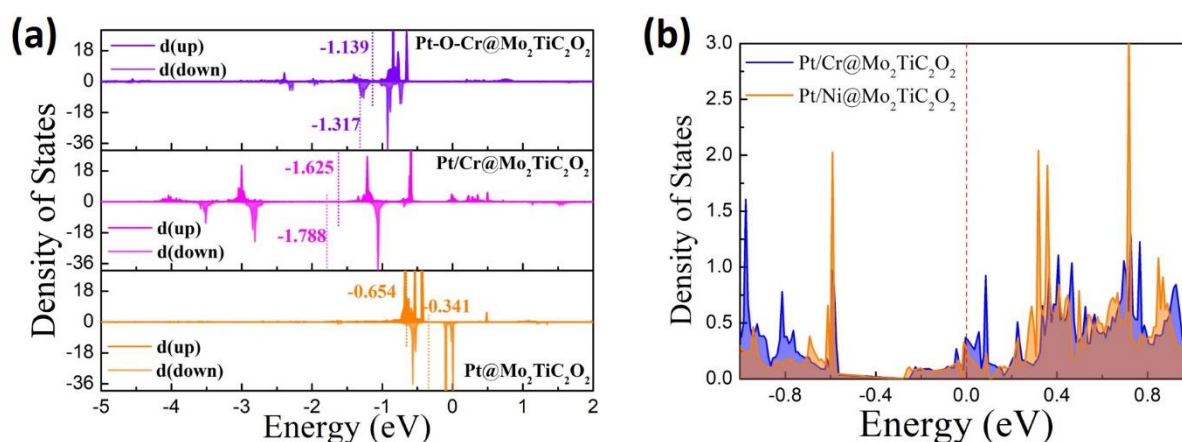


Figure S13. (a) The Projected DOS and the location d-band center of Pt atom for Pt@Mo₂TiC₂O₂, Pt/Cr@Mo₂TiC₂O₂ and Pt-O-Cr@Mo₂TiC₂O₂h-DACs. (b) The electronic DOS (per atom) of Pt/Cr@Mo₂TiC₂O₂h-DACs and Pt/Ni@Mo₂TiC₂O₂h-DACs.

Table S1. ΔG_{H^+} of different H adsorption sites on the Pt/TM@Mo₂TiC₂O₂ DACs surface (TM = Ti, V, Cr, Fe, Co, Ni, Cu and Zn). Wherein, “x” represents structural instability after H proton adsorbed to the site. “-” represents the non-existent sites.

	Site I	Site II	Site III	Site IV	Site V	Site VI	Site VII
Pt/Ti@Mo ₂ TiC ₂ O ₂	0.228	1.026	0.562	0.543	-	-	-
Pt/V@Mo ₂ TiC ₂ O ₂	-0.187	x	0.298	0.301	-	-	-
Pt/Cr@Mo ₂ TiC ₂ O ₂	-0.591	x	x	0.378	-	-	-
Pt/Fe@Mo ₂ TiC ₂ O ₂	-0.724	x	x	0.214	-	-	-
Pt/Co@Mo ₂ TiC ₂ O ₂	-0.813	x	x	0.159	-	-	-
Pt/Ni@Mo ₂ TiC ₂ O ₂	-1.112	0.566	0.493	0.291	x	x	x
Pt/Cu@Mo ₂ TiC ₂ O ₂	-0.706	0.044	0.268	0.354	x	x	-
Pt/Zn@Mo ₂ TiC ₂ O ₂	-1.071	x	x	0.445	x	x	-

Table S2. The number of the charge transfer (Q_e , e) from Single-atom to substrate.

Active center	Charge transfer (Q_e/e)
Ni@Mo ₂ TiC ₂ O ₂	-0.89
Pt@Mo ₂ TiC ₂ O ₂	-0.45
Pt/Ni@Mo ₂ TiC ₂ O ₂	-0.79
O-Pt/Ni@Mo ₂ TiC ₂ O ₂	-0.76
Pt/Cr@Mo ₂ TiC ₂ O ₂	0.38
Pt-O-Cr@Mo ₂ TiC ₂ O ₂	-0.37