Supplementary Information

High-Throughput Screening of Bifunctional Catalysts for Oxygen Evolution/Reduction Reaction at the Subnanometer Regime

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1. Density Functional Theory (DFT) Details

Text S1:

All the plane-wave spin-polarized density functional calculations were conducted using the Vienna *ab initio* simulation package (VASP) with the Perdew-Burke-Ernzerhof (PBE) functional within the generalized gradient approximation (GGA).[1,2] Ion core and valence electron interactions were described using the projector augmented wave (PAW) method.[3] For relaxation calculations, kinetic energy cut-offs of 500 eV with energy convergence criteria of 10^{-6} eV were employed, and geometric relaxation continued until forces on atoms were smaller than 0.02 eV/Å.[4] Gaussian smearing with a sigma value of 0.2 eV was applied and the Γ-centered (1×1×1) k-point grids were considered for the sampling of the Brillouin zone in congruence with the previous reports.[5,6] However, a higher (3 × 3 × 3) k-mesh was used to calculate the density of states (DOS). A sufficiently large box with dimensions 20 × 20 × 20 $Å^3$ was utilized to optimize the distinct geometries to avoid the possibility of spurious interaction between the adjacent images along each axis.

2. Scaling Relationship Investigation

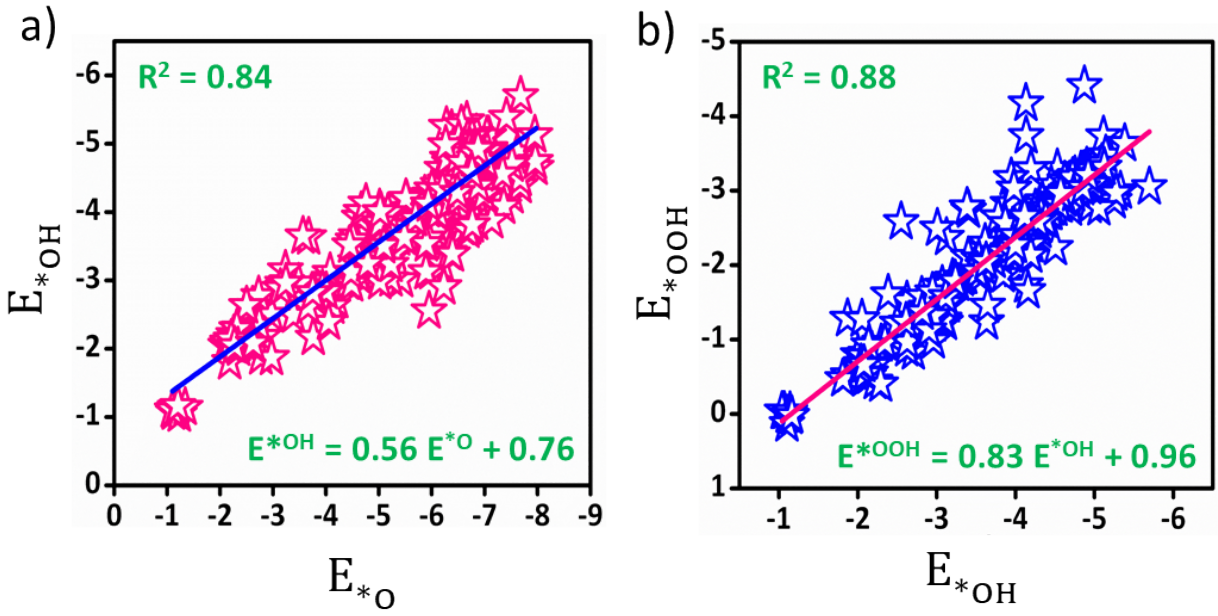

Figure S1. Scaling relationship between **(a)** E_{*O} vs E_{*OH} and **(b)** E_{*OH} vs E_{*OOH} for different-sized TM_n subnano clusters.

3. Reaction Energy Diagram of OER and ORR activity

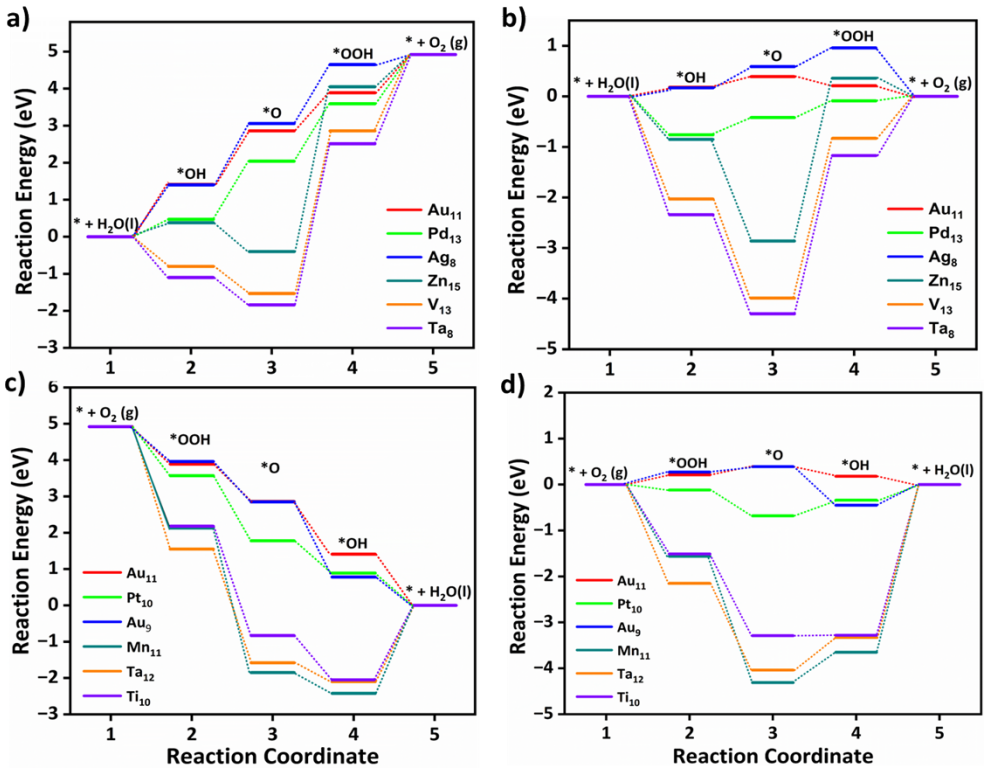

Figure S2. Free-energy diagrams of OER elementary steps at **(a)** 0 V and **(b)** 1.23 V, and ORR elementary steps at **(c)** 0 V and **(d)** 1.23 V.

4. Distribution analysis of I_{df} with E_{*O}, E_{*OH}, and E_{*OOH}.

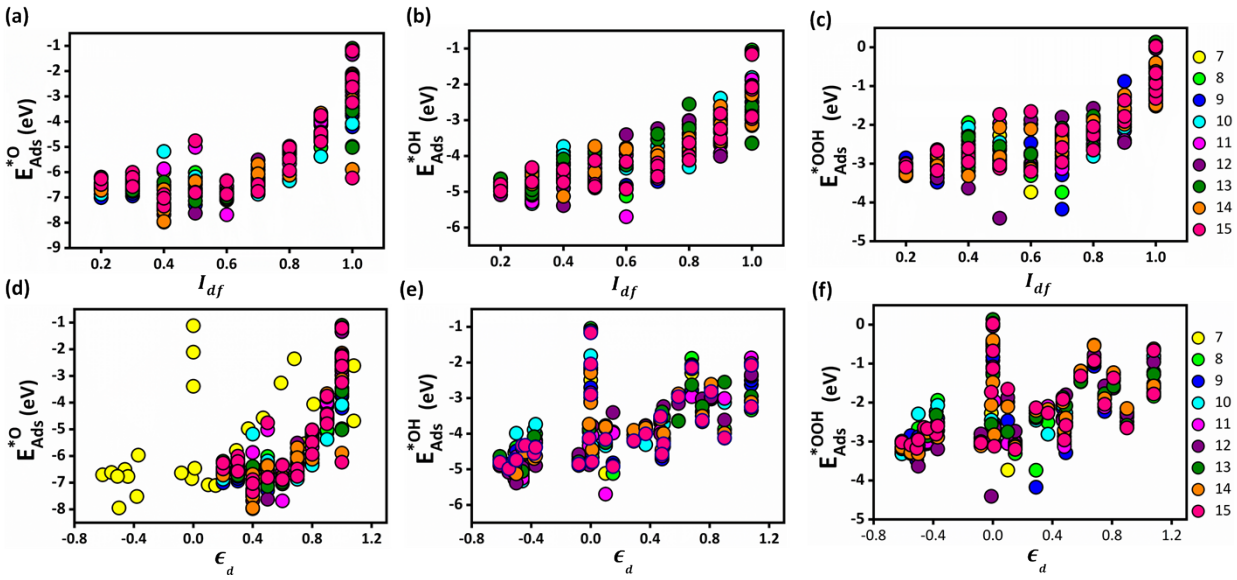

Figure S3. Distribution analysis between **(a-c)** $E_{*O}, E_{*OH}, and\ E_{*OOH}$ and I_{df}, and **(d-f)** $E_{*O}, E_{*OH}, and\ E_{*OOH}$ and ϵ_d.

5. References

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