Supplementary material

Interface engineering towards overall water electrolysis upon $NiCo_2O_4$/NiMo hybrid catalysts

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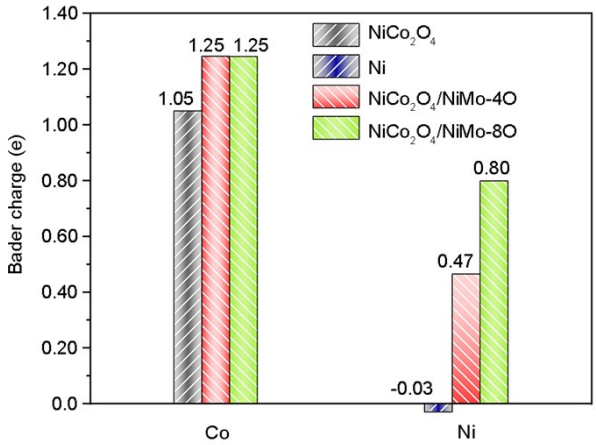

Fig. S1 The Bader charge of Co and Ni sites at the $NiCo_2O_4$/NiMo-xO (x=4, 8) interface.

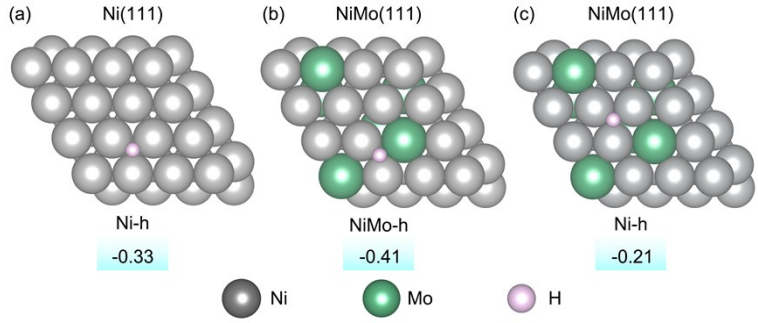

Fig. S2 The configurations for H adsorbed on Ni(111) and NiMo(111) with the adsorption site of Ni-h and NiMo-h. The corresponding Gibbs free energies of hydrogen adsorption (ΔG_{*H}) are labeled below the picture, in the unit of eV.

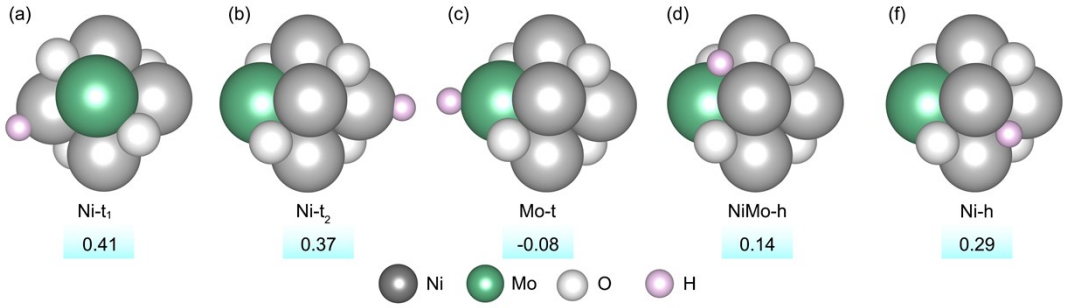

Fig. S3 The configurations for H adsorbed on Ni_5Mo-4O cluster with adsorption sites of Ni-t, Mo-t, NiMo-h, and Ni-h. The subscript 1 in Ni-t_1 means this Ni atom is closer to the Mo atom than that of Ni-t_2. The corresponding ΔG_{*H} is labeled below the picture in the unit of eV.

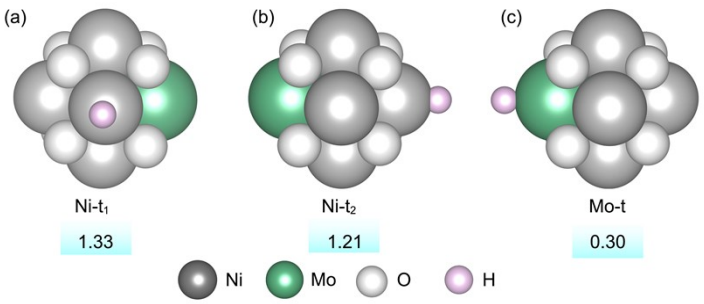

Fig. S4 The configurations for H adsorbed on Ni_5Mo-8O cluster with adsorption sites of Ni-t and Mo-t. The subscript 1 in Ni-t_1 means this Ni atom is closer to the Mo atom than that of Ni-t_2. The corresponding ΔG_{*H} is labeled below the picture in the unit of eV.

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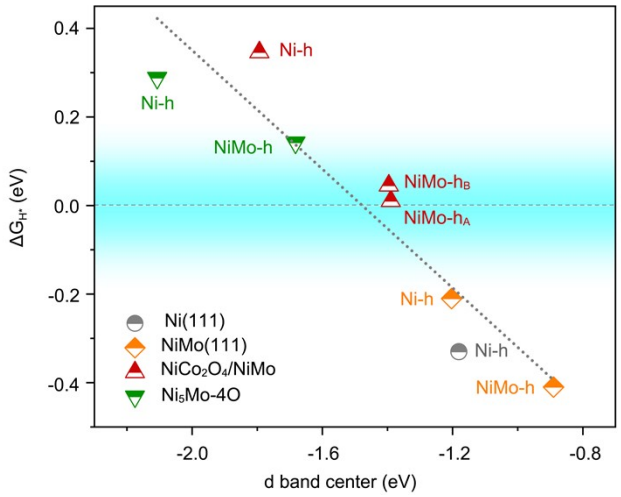

Fig. S5 Correlation between Gibbs free energies of hydrogen adsorption (ΔG_{*H}) and *d* band center (ε_d) of the adsorption sites.

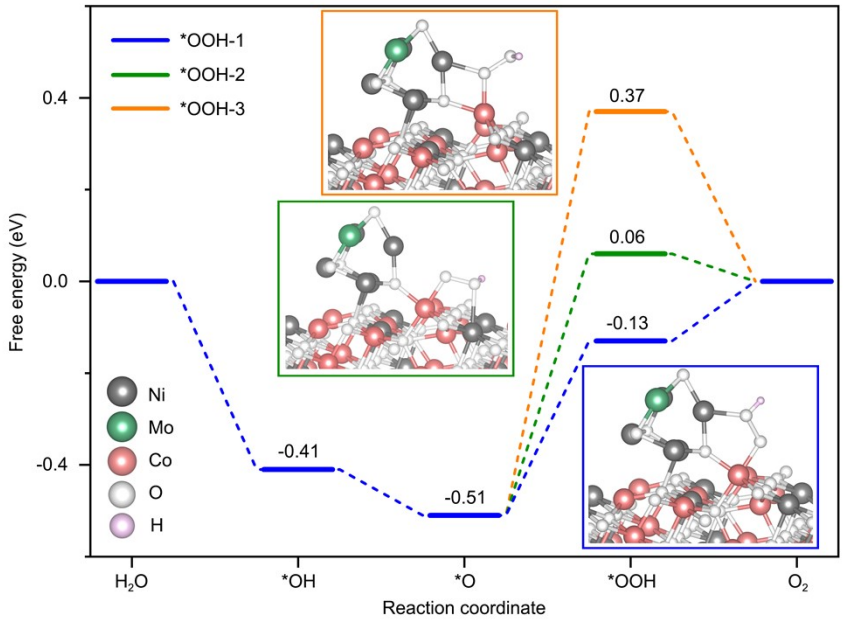

Fig. S6 Free energy diagram of OER process on $NiCo_2O_4$/NiMo hybrid catalyst with three different adsorption configurations for *OOH.

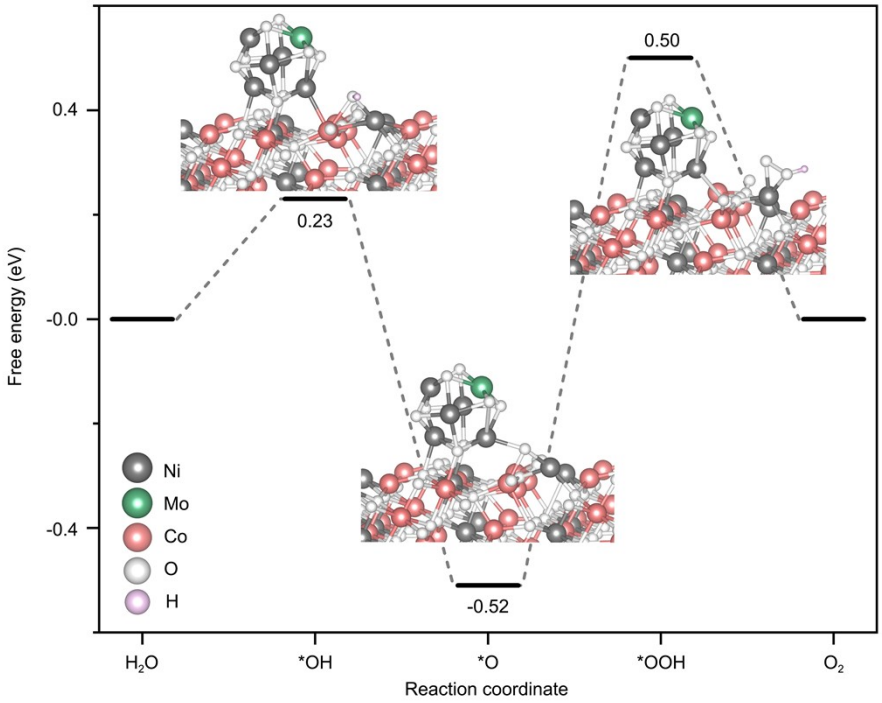

Fig. S7 Free energy diagram of OER process on $NiCo_2O_4$/Ni_5Mo-8O. The active center Co of $NiCo_2O_4$ forms a Co-Ni bond with the Ni of NiMo cluster at the interface. The OER overpotential is 1.02 V.

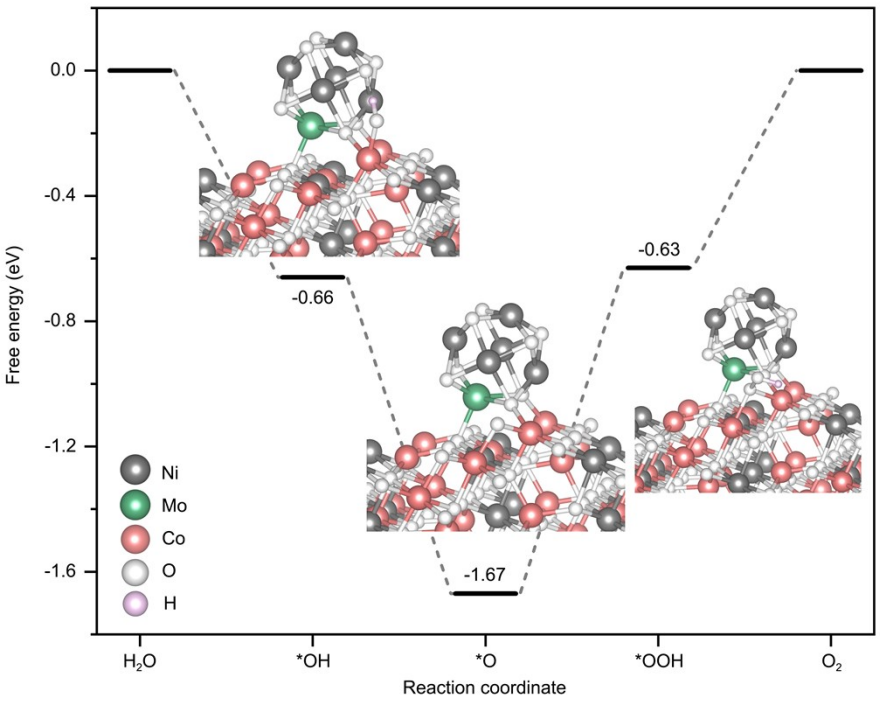

Fig. S8 Free energy diagram of OER process on $NiCo_2O_4/Ni_5Mo$-8O. The active center Co of $NiCo_2O_4$ forms a Co-O bond with the O of Ni_5Mo-8O cluster at the interface. The OER overpotential is 1.04 V.

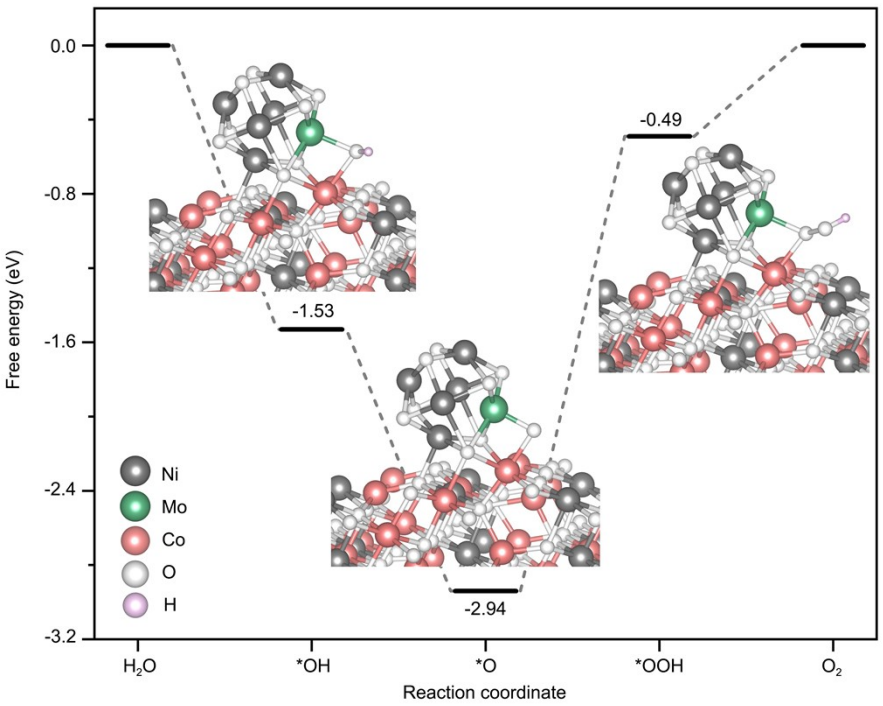

Fig. S9 Free energy diagram of OER process on $NiCo_2O_4/Ni_5Mo$-8O. The active center Co of $NiCo_2O_4$ forms two Co-O bonds with the O of Ni_5Mo-8O cluster at the interface. The Mo atom of Ni_5Mo-8O at the interface strongly adsorbs intermediates $^*O$ and $^*OH$. The OER overpotential increases to 2.45 V.