Electronic Supplementary Material (ESI) for Nanoscale Advances.

Supporting Information

First-principles Design of Hetero CoM (M = 3d, 4d, 5d Block Metals) Double Atom Catalysts for Oxygen Evolution Reaction in alkaline condition

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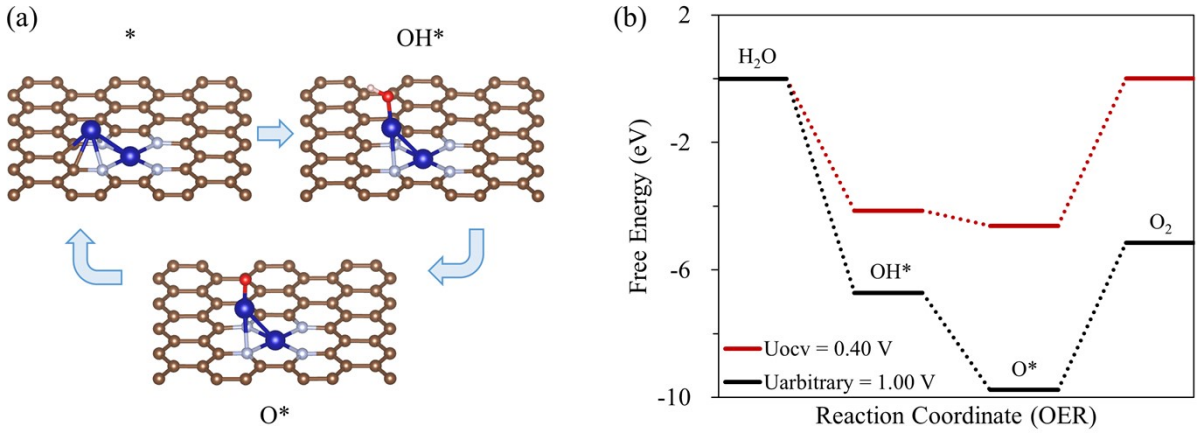

Figure S1. (a) dissociative reaction mechanism of OER on Co_2/N_4G without hydrogen superoxide(OOH*) formation, (b) free energy diagram for dissociative pathway on Co_2/N_4G at OCV condition (U_{OCV} = 0.401 V) and arbitrary potential ($U_{arbitrary}$ = 1.00 V).

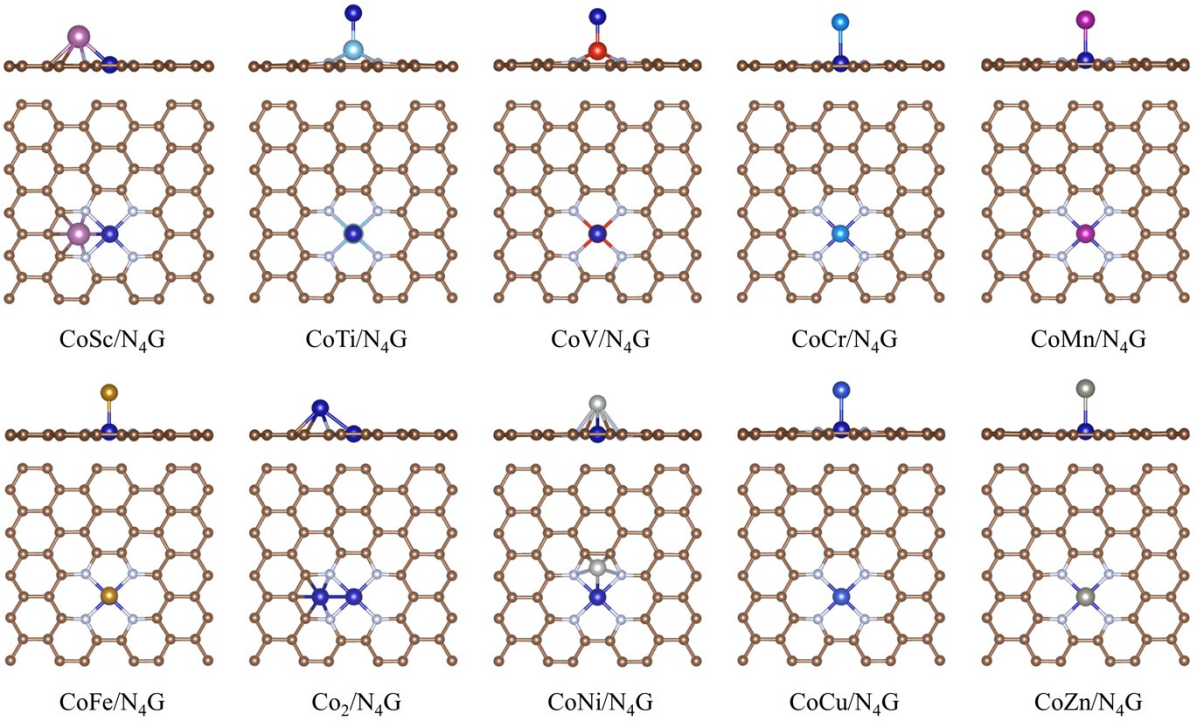

Figure S2. The most stable structures of CoM/N_4G catalysts determined by the formation energy.

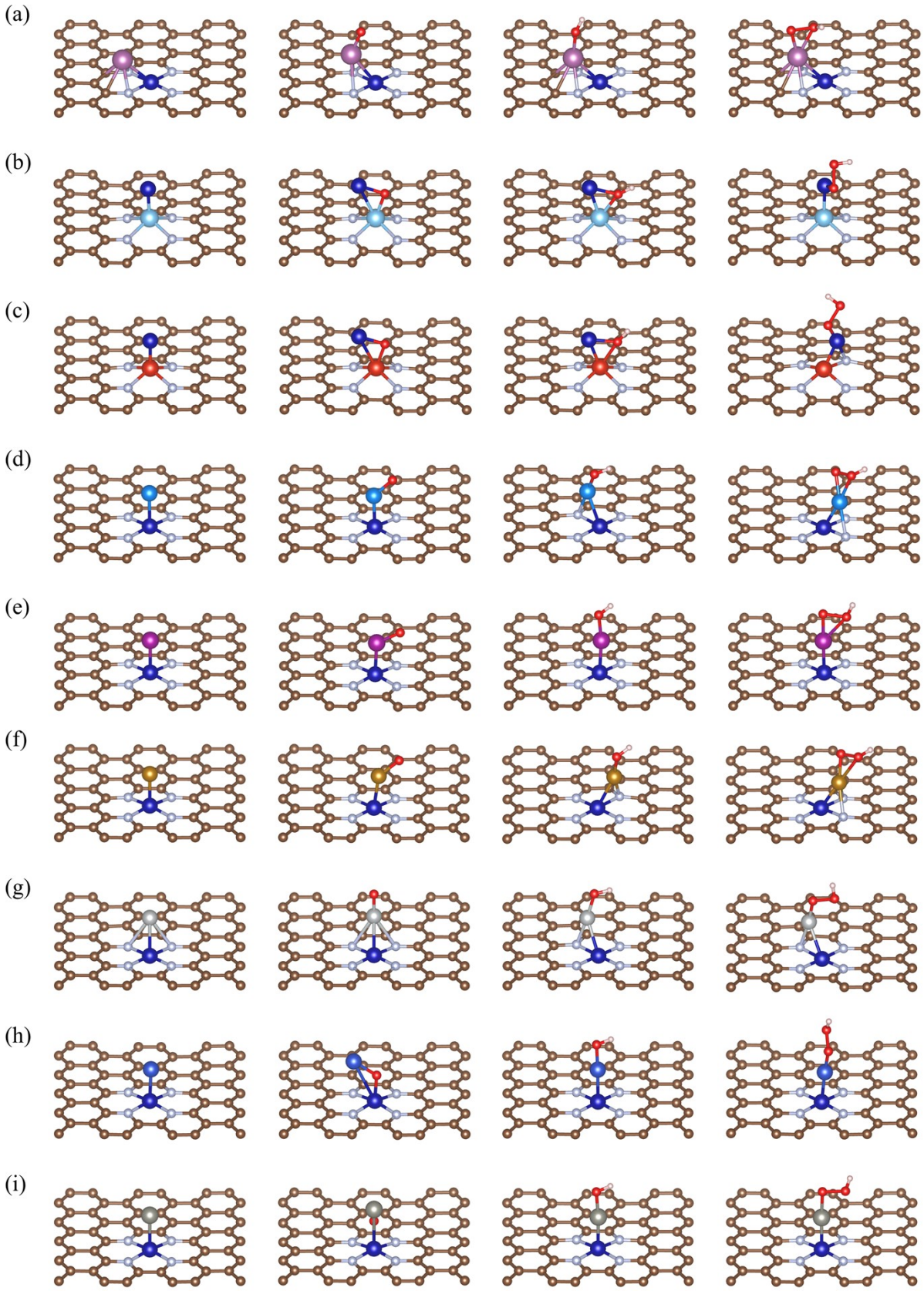

Figure S3. The most favorable adsorption configurations of intermediates for (a) CoSc/N_4G, (b) CoTi/N_4G, (c) CoV/N_4G, (d) CoCr/N_4G, (e) CoMn/N_4G, (f) CoFe/N_4G, (g) CoNi/N_4G, (h) CoCu/N_4G and (i) CoZn/N_4G.

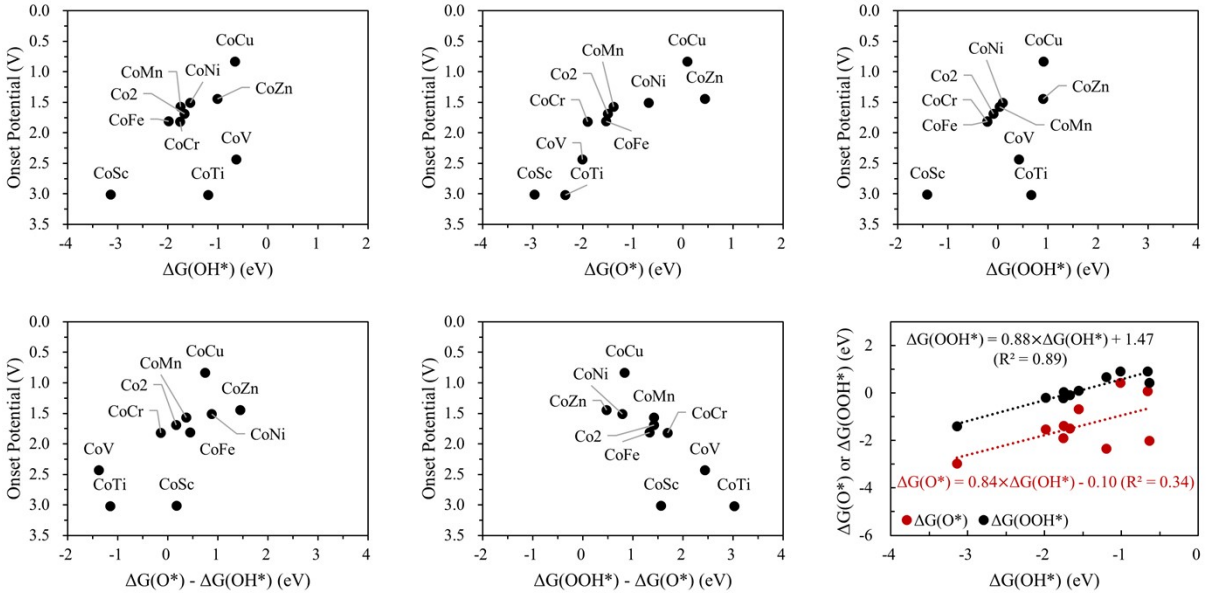

Figure S4. The correlation between OER activity (onset potential) and ΔG(adsorbate*).

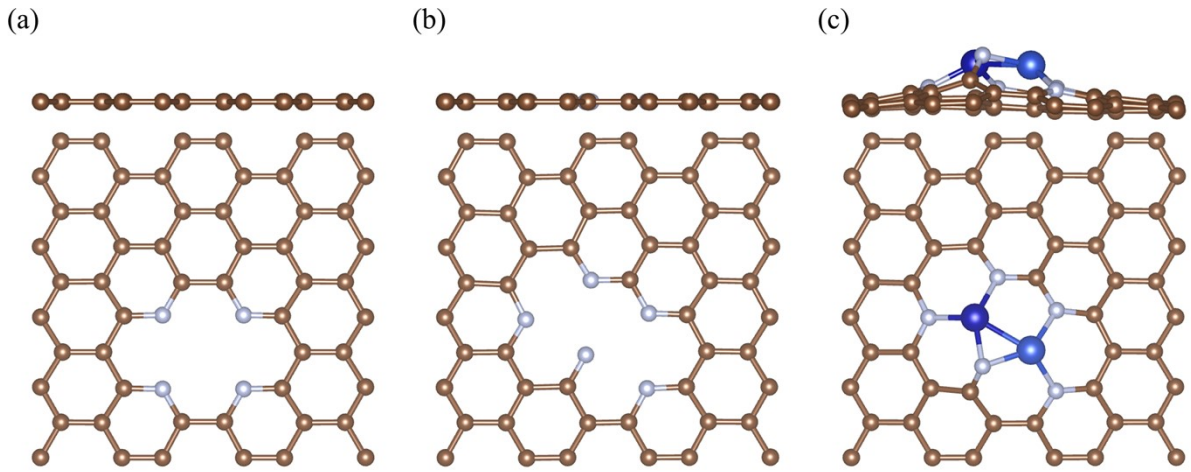

Figure S5. The stable structure of (a) N_4G support (di-vacancy model), (b) N_5G support (two mono-vacancy model) and (c) $CoCu/N_5G$ catalyst.

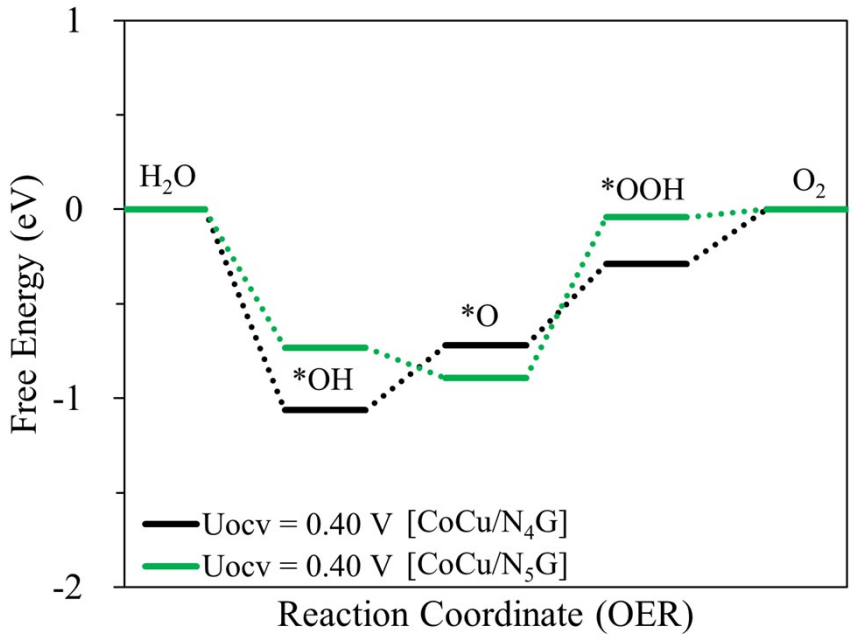

Figure S6. The free energy variations at OCV condition for $CoCu/N_4G$ and $CoCu/N_5G$ catalysts.

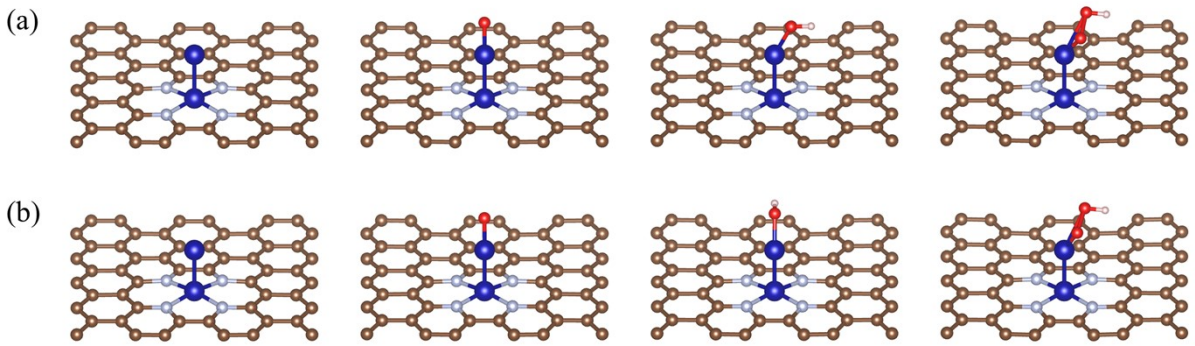

Figure S7. The most favorable adsorption configurations of intermediates for (a) Co_2/N_4G_{config} model and (b) Co_2/N_4G_{strain}.

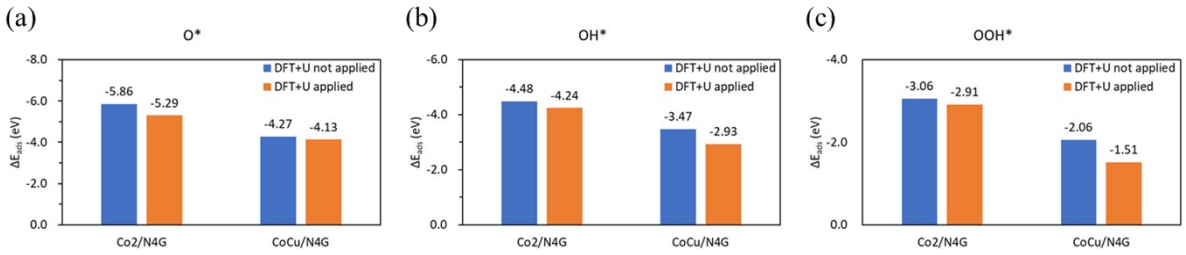

Figure S8. Comparison of adsorption energy on Co_2/N_4G and $CoCu/N_4G$ catalysts. Blue bar and orange bar correspond to the adsorption energy when DFT+U [U=4 (Co) and U=8.5 (Cu)] is not applied and applied, respectively.

Table S1. The dissolution potential of CoM/N_4G catalysts (M = Co or Cu). we calculated dissolution potential of those catalysts, where the one solute atom (single metal atom, Co or Cu) is dissolved into the electrolyte, on the basis of precedent research (Electrochimica Acta, 52 (2007) 5829–5836). The dissolution potential (U_{diss}) can be calculated by following equation: $U_{diss} = U_{diss}^{o} - \frac{E_{bind}}{ne}$ and E_{bind} is defined as $E_{bind} = E_{CoM/N_4G} - E_{Co/N_4G} - E_M$,where U_{diss}^{o}, E_{CoM/N_4G}, E_{Co/N_4G}, E_M, and n are the standard reduction potential of solute M, total energy of CoM/N_4G (M = Co or Cu), Co/N_4G, isolated M atom and the number of electrons involved in dissolution, respectively.

Catalyst	Solute M	U_{diss}
Co_2/N_4G	Co	0.53 V
$CoCu/N_4G$	Co	1.78 V
	Cu	1.04 V

Table S2. The standard reduction potential of Co and Cu bulk.

Reaction	U_{diss}^{o}
$Co^{2+} + 2e^- \rightarrow Co$	−0.28 V
$Cu^{2+} + 2e^- \rightarrow Cu$	0.34 V

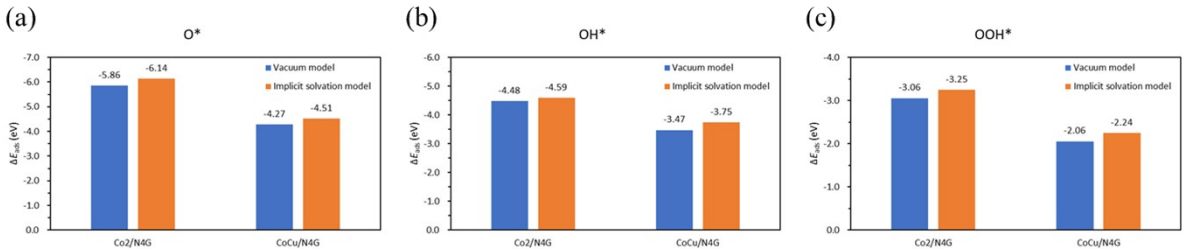

Figure S9. Comparison of adsorption energy on Co_2/N_4G and $CoCu/N_4G$ catalysts. Blue bar and orange bar correspond to the adsorption energy in vacuum model and implicit solvation model, respectively. Here, we took solvent effect into account by using the implicit solvation model implemented in VASPsol, which is simple and cost-effective method in computational study.