Supporting Information

Tuning Metal Single-Atoms Embedded in N_xC_y Moiety Toward High-Performance Electrocatalysis

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Supplementary Note:

Charateristic in Co K-edge XANES of Co-N_nC_m-C_N

The Co K-edge XANES of Co–N_nC_m–G_N (see **Figure 4** for clear image) show edge shifts to the higher energy than Co bulk, while giving a rising pre-edge. Co atoms on single vacancy sites have +1 oxidation state so that the Co K-edge XANES of Co–$N_{n=0-3}C_{3-n}$–G_N have higher edge energy than their bulk in XANES. Co–N_3–G_N shows a distinctive pattern different from other structures. This comes from weak metal-ligand interaction between Co and N atoms. The p-d hybridization occurs readily in tetrahedral symmetry structures. The weak metal-ligand interaction leads to weak splitting and occupies p orbitals of hybridized p-d orbitals. As a result, 1s electron excitations to 3d orbitals in Co–N_3–G_N increase, giving intense pre-edge in the XANES spectra. In the systems including C atoms, the metal-ligand interaction becomes stronger. Therefore, the p orbitals in hybridized d-p orbitals are in higher energy levels and so they are not occupied. Then 1s electron excites mainly to 4p orbitals which give sharp rising peaks. Meanwhile, all Co–$N_{n=0-4}C_{4-n}$–G_N show similar XANES patterns to each other with the increased edge energy due to 2+ oxidation state. The differences between coordination environments appear in the intensity of the pre-edge peak of XANES in the Co case. The p-d hybridization is not high in square-planar structure, as in Co–N_4–G_N XANES. However, the intensity of the pre-edge peak increases with the increment of C atoms in the structure because C atoms interact with p orbitals of Co atoms with increased p-d hybridization. It is also notable that Co–N_2C_2–G_N and Co–$N_2C_2{}^c$–G_N have distinguishable pre-edge structures from each other. These differences could be found in all other TMs in M–N_nC_m–G_N systems shown in the above figure. The pre-edge peaks are usually broader and less intense in 4d (TM)SAs, and additional effects like symmetry and distortion of a structure may affect XANES, which helps in characterizing the coordination environments for each (TM)SA.

Descriptors for classification between good and bad catalysts

In this section we report a supervised machine learning approach to classification of double vacancy configurations into good and bad catalysts.

We searched for a descriptor taking properties of atomic elements constituting the configuration. Namely, we considered valence electrons in the occupied d orbital of the metal element (θ_d), unpaired valence electrons in the occupied d orbital of the metal element (θ_d^u), Pauling electronegativity (χ), the number of carbon (n_C) and nitrogen (n_N) atoms closest to the metallic center, atomic numbers (Z), atomic radius, atomic volume, group, period, dipole polarizability, Mendeleev number, Pettifor number, and first ionization energies. We took these values from the mendeleev python package[3]. We also considered the products of these properties such as $\chi\theta_d$, $\chi\theta_d^u$, χ^2, χn_C, χn_N, etc. In addition, we took the Coulomb matrix elements[4] between the metal site and neighboring carbon or nitrogen atoms as well as between metal and adsorbent atoms (hydrogen or oxygen), and between the adsorbents and the closest non-metal surface atom (carbon or nitrogen). The Coulomb matrix element between two atoms with the atomic numbers Z_1 and Z_2 located at a distance r_{12} from each other is given by the formula

$$CM = \frac{Z_1 Z_2}{r_{12}} \quad (1)$$

We considered the following machine learning algorithms: Gaussian naïve Bayes, logistic regression, k-nearest neighbors, radius neighbors classifier, support vector classifier, neural network, and decision trees as well as the following ensemble methods: random forest, extremely randomized trees, and gradient boosting. Before feeding into the learning algorithms, the feature columns were normalized to have zero mean and unit variance. The hyper-parameters search and the model performance estimation was done using the Monte-Carlo cross validation[5]. The data samples were randomly split 100 times into different training (85%) and test (15%) sets using stratified splitting for which we employed the StratifiedShuffleSplit

object from the scikit-learn library[6] in python. This library was used for all data analysis computations in this section. The best hyper-parameters were found using an exhaustive search implemented in GridSearchCV. The Area Under the Receiver Operating Characteristic Curve (ROC AUC)[7] was chosen as the optimization metrics. The mean and standard deviation of the ROC AUC among 100 splits were calculated with the cross_val_score object.

We found that descriptors that give the best ROC AUC are composed of Coulomb matrix elements and the first ionization energy, I_1 of the transition metal. In order to get the actual distances r_{12} between atoms for the Coulomb matrix elements, DFT calculations must be done for geometry optimization. However, desirable descriptors for high-throughput exploration of catalytic configurations are those that do not require DFT calculations. We therefore replaced the actual distances r_{12} by sums of the covalent radii of the corresponding atoms taken from the mendeleev python package. We also compared the results for these descriptors with the descriptors containing the actual DFT-optimized distances. The details are given in the following subsections.

(i) Descriptors for HER

The configurations are classified as good catalysts are those with $|\Delta G|$ close to zero, and bad catalysts are those with $|\Delta G|$ far from zero. We choose the threshold to be 0.2. Therefore, we classify all configurations into two classes, class 1: $|\Delta G| \leq 0.2$, and class 0: $|\Delta G| > 0.2$. Our data set consists of 438 double-vacancy configurations. A configuration is formed by one of thirty different transition metals from Sc through Hg. For each metallic center sitting in the middle of a double vacancy, there are six possible arrangements of its four closest neighbors, where each neighbor can be either carbon or nitrogen. Besides that, configurations differ based on to which atom the hydrogen binds.

Our descriptor is a 9-dimensional vector given by the following components:

$$d = (CM_{UL}, CM_{UR}, CM_{LR}, CM_{LL}, CM_{HC}, CM_{HN}, CM_{ULLR}, CM_{URLL}, I_1) \quad (2)$$

where $CM_{UL}, CM_{UR}, CM_{LR}, CM_{LL}$ are the Coulomb matrix elements between the metallic center and the upper-left, upper-right, lower-right, lower-left atoms, closest to the metal (**Figure S19**). The distances r_{12} in these elements are approximated by the sums of the covalent radii of the corresponding atoms. The other Coulomb matrix elements are as follows:

$$CM_{HC} = \frac{Z_H Z_C}{\sqrt{(r_C+r_M)^2+(r_H+r_M)^2}} \quad (3)$$

$$CM_{HN} = \frac{Z_H Z_N}{\sqrt{(r_N+r_M)^2+(r_H+r_M)^2}} \quad (4)$$

$$CM_{ULLR} = \frac{Z_{UL} Z_{LR}}{r_{UL}+2r_M+r_{LR}} \quad (5)$$

$$CM_{URLL} = \frac{Z_{UR} Z_{LL}}{r_{UR}+2r_M+r_{LL}} \quad (6)$$

where Z_i is the atomic number of the atom i; the subscript Mrefers to the metallic center, and the subscripts UL, LR, UR, LL refer to the upper-left, upper-right, lower-right, lower-left atoms, closest to the metal. This means that CM_{HC} approximates the Coulomb matrix element between the hydrogen and carbon atoms, CM_{HN} – between hydrogen and nitrogen atoms, CM_{ULLR} and CM_{URLL} between atoms situated diagonally around the metallic center. The descriptor based on the DFT-optimized distances is a 7-dimensional vector given by

$$d = (CM_1, CM_2, CM_3, CM_4, CM_{HM}, CM_H, I_1) \quad (7)$$

where $CM_1, \dots, CM_4$ are the Coulomb matrix elements between the transition metal and the neighboring carbon and nitrogen atoms sorted according to the distance in an ascending order; CM_{HM} is the Coulomb matrix element between the transition metal and hydrogen; CM_H is the Coulomb matrix element between hydrogen and the closest non-metal (carbon or nitrogen); I_1 is the first ionization energy of the transition metal. The values of the ROC AUC for different machine learning algorithms for both descriptors are reported in **Figure S20a** and **Table S32**. The best algorithm working with the descriptor based on elemental properties is random forest for which we got ROC AUC = 0.79. In the case of the DFT-optimized descriptor, the best

algorithm is found to be the k-nearest neighbors for which we got ROC AUC = 0.91. The Receiver Operating Characteristic curve for the best classifiers (random forest in the case of elemental properties, and k-nearest neighbors in the case of DFT-optimized geometry) are shown in **Figure S20a**. We performed principal component analysis (PCA) on both descriptors. The cumulative explained variance ratio as a function of the number of principal components is shown in **Figure S21a**. We see from the Figure that almost all the variance in the data is explained by 5 principal components for both descriptors.

(ii) Descriptors for OER, and ORR

The configurations are classified as good catalysts are those with the over-potential η between 0 and 1. Therefore, we classify all configurations into two classes, class 1: $\eta \leq 1$, and class 0: $\eta > 1$. Our data set consists of 210 double-vacancy configurations. A configuration is formed by one of thirty different transition metals from Sc through Hg. For each metallic center sitting in the middle of a double vacancy, we have its closest neighbors either four carbon atoms, four nitrogen atoms, or two carbons and two nitrogens in a trans configuration.

Our descriptor is a 9-dimensional vector given by the following components:

$$d = (CM_{UL}, CM_{UR}, CM_{LR}, CM_{LL}, CM_{OC}, CM_{ON}, CM_{ULLR}, CM_{URLL}, I_1) \quad (8)$$

where $CM_{UL}, CM_{UR}, CM_{LR}, CM_{LL}$ are the Coulomb matrix elements between the metallic center and the upper-left, upper-right, lower-right, lower-left atoms, closest to the metal (**Figure S19**). The distances r_{12} in these elements are approximated by the sums of the covalent radii of the corresponding atoms. The other Coulomb matrix elements are as follows:

$$CM_{OC} = \frac{Z_O Z_C}{\sqrt{(r_C + r_M)^2 + (r_O + r_M)^2}} \quad (9)$$

$$CM_{ON} = \frac{Z_O Z_N}{\sqrt{(r_N + r_M)^2 + (r_O + r_M)^2}} \quad (10)$$

CM_{OC} approximates the Coulomb matrix element between the oxygen and carbon atoms, CM_{ON} – between oxygen and nitrogen atoms. The elements CM_{ULLR} and CM_{URLL} between atoms situated diagonally around the metallic center were described in the previous subsection.

Here we consider three descriptors based on the DFT-optimized geometries. When only one oxygen atom adsorbed, the descriptor consists of the first ionization energy of the transition metal, I_1, and two Coulomb matrix elements – between the oxygen atom and the metallic center, and between oxygen, and the closest non-metal (carbon or nitrogen). The second descriptor is built from the first one with the addition of the Coulomb matrix elements for the system where one oxygen and one hydrogen atoms are adsorbed. It consists of eight components: three from the previous descriptor, and five related to the system with O and H adsorbed, namely, the Coulomb matrix elements between O and the metal, between H and the metal, between O/H and the closest non-metal, and between O and H. The third descriptor is built from the previous one with the addition of the Coulomb matrix elements for the two oxygen and one hydrogen atoms are adsorbed. It consists of 17 components: Coulomb matrix elements between each atom in the OOH configuration, between each O, O, H, and the metallic center, and between each O, O, H, and the closest non-metal. The values of the ROC AUC for different machine learning algorithms for all descriptors are reported in **Figure S20b** & **Table S33** for the OER and **Figure S20c** & **Table S34** for the ORR.

The best algorithm working with the descriptor based on elemental properties for both OER and ORR is random forest for which we got ROC AUC = 0.87 in the case of OER, and 0.81 in the case of ORR. For the three DFT-optimized descriptors, the best algorithm is also random forest for both OER and ORR. The Receiver Operating Characteristic curve for the best classifier (random forest) are shown in the **Figure S20b** for OER, and **Figure S20c** for ORR. The cumulative explained variance ratio as a function of the number of principal components is shown in the **Figure S21b**. We see from the Figure that, as in the case or HER, almost all

the variance in the data is explained by 5 principal components for the descriptors based on elemental properties.

Supplementary Figures

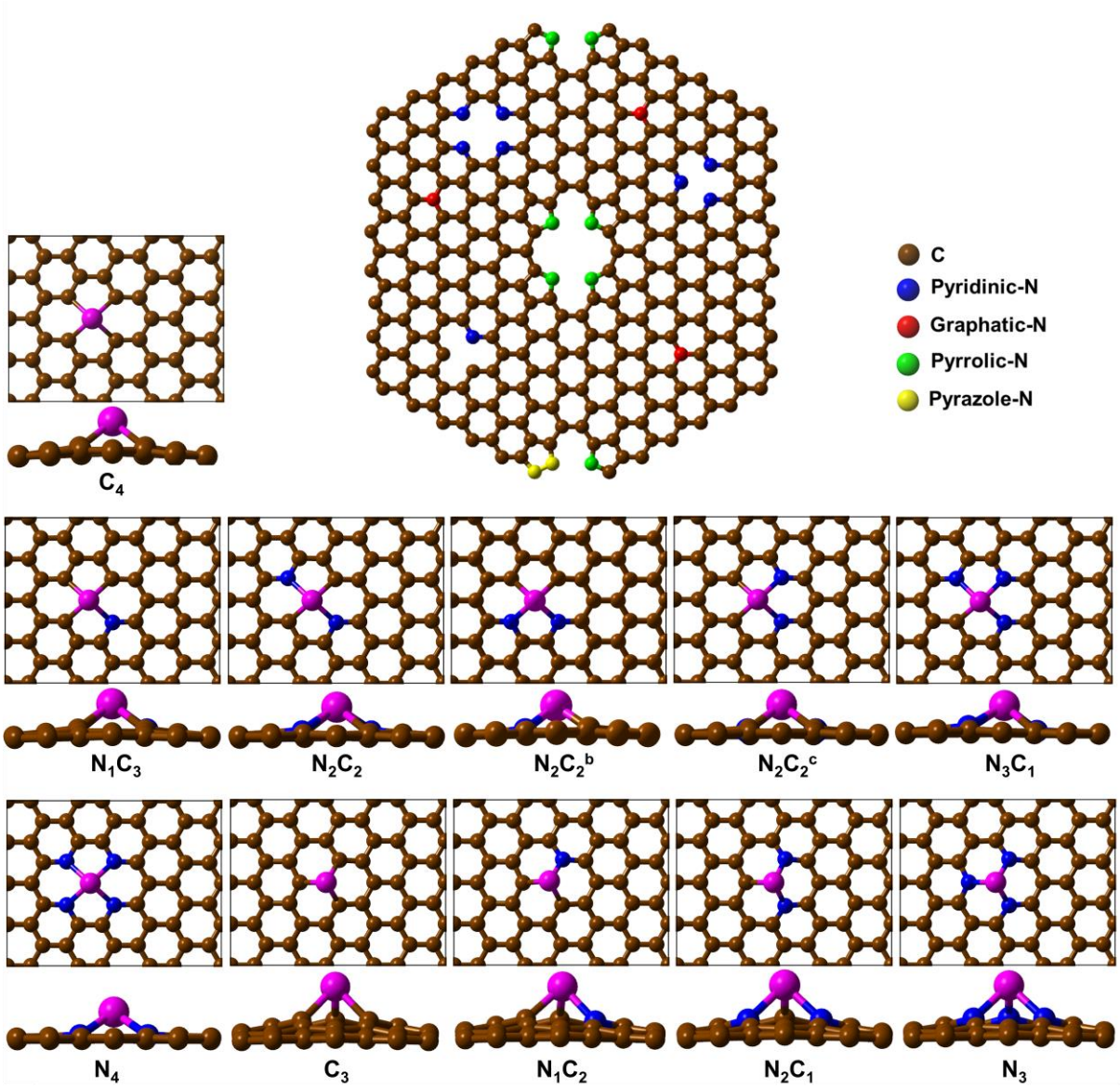
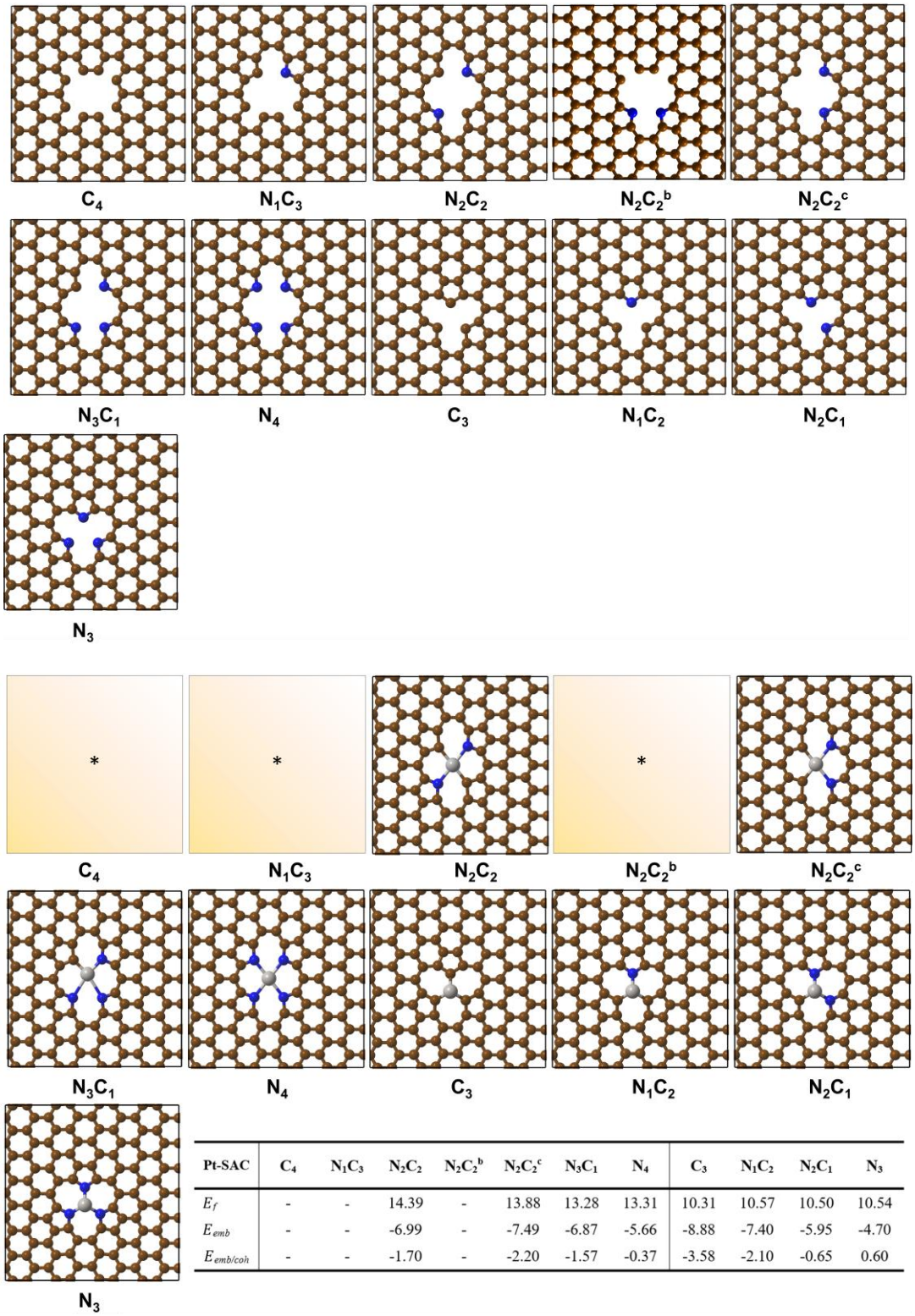


Figure S1. Possible vacant sites and eleven pyridinic/graphitic configurations of Zr-($N_{n=0-4}C_{4-n}$, $N_{n=0-3}C_{3-n}$)-G_N. The Zr atom (purple) is located somewhat above the graphene sheet due to its large atomic radius.



Pt-SAC	C_4	N_1C_3	N_2C_2	$N_2C_2^b$	$N_2C_2^c$	N_3C_1	N_4	C_3	N_1C_2	N_2C_1	N_3
E_f	-	-	14.39	-	13.88	13.28	13.31	10.31	10.57	10.50	10.54
E_{emb}	-	-	-6.99	-	-7.49	-6.87	-5.66	-8.88	-7.40	-5.95	-4.70
$E_{emb/coh}$	-	-	-1.70	-	-2.20	-1.57	-0.37	-3.58	-2.10	-0.65	0.60

Figure S2. Configurations of N-doped ($N_{n=0\text{-}4}C_{4\text{-}n}$, $N_{n=0\text{-}3}C_{3\text{-}n}$)-$G_N$ pyrrolic graphene systems and their Pt-embedding srtructures. Pt-embeddings in C_4 and N_1C_3 pyrrolic defect sites are not stable. The defect formation energy (E_f), Pt-SA embedding energy (E_{emb}), Pt-SA stability over metal cohesion ($E_{emb/coh}$) in pyrrolic systems are given as a table below the figure (energies in eV). These pyrrolic data against the pyridinic data (**Tables S5-S7**) indicate that the pyridinic defect sites are preferred over the pyrrolic ones in G_N/Gr.

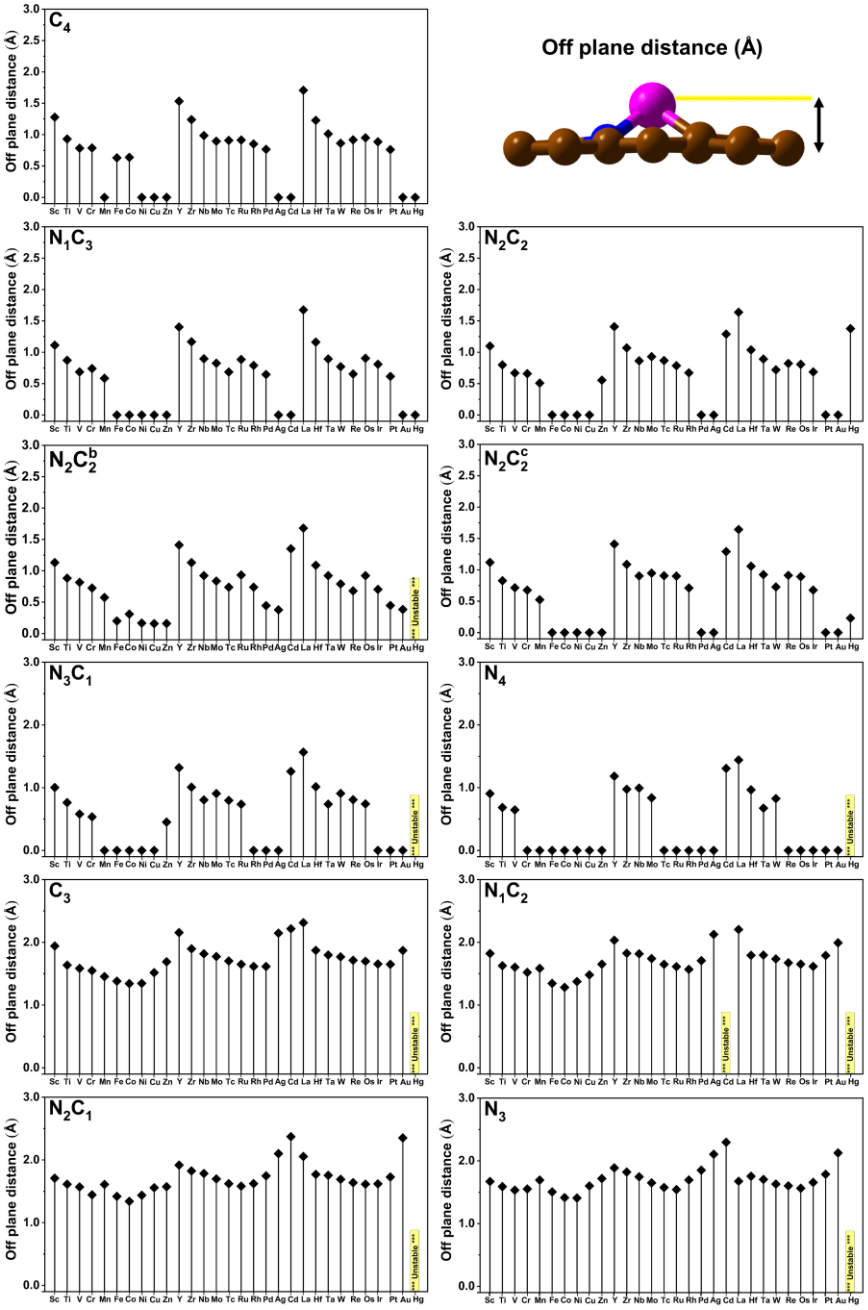


Figure S3. Off-plane distances (Å) of various (TM)SAs for M-($N_{n=0-4}C_{4-n}$, $N_{n=0-3}C_{3-n}$)-G_N. Some unstabilities (in yellow bar) are shown for a few cases of Hg (and Cd). See **Table S8** for details. Perfect planar structures before adsorption are found for divacant moieties M-$N_{n=0-4}C_{4-n}$-G_N [(Mn, Fe, Co), Ni, Cu, (Zn); (Rh, Pd), Ag, (Cd); (Pt), Au, (Hg)]. Metals (Mn), {Fe}, Ni, Cu, (Zn, Ag, Cd), Au, and Hg are in plane of M-C_4-G_N; (Fe), Co, Ni, Cu, Pd, Ag, (Pt) and Au are in plane of M–N_2C_2–G_N; (Cr), Mo, Fe, Co, Ni, (Cu, Zn, Tc, Ru), Rh, Pd, (Re, Os, Ir), Pt, and Au are in plane of M–N_4–G_N. Here, (TM)SAs are in plane regardless of H-adsorption, while "()" denotes the non-planar case upon H-adsorption. Among –N_nC_m moieties, the –N_4 moiety tends to have more planar structures for various elements. However, many of the metal elements M in –N_nC_m–G_N are not in a plane but are located slightly off the plane. Upon adsorbing H, the planarity tends to be further weakened. For all M-N_mC_n-G_N systems, d^1s^2-series{La($5d^16s^2$), Y($4d^15s^2$), Sc($3d^14s^2$)} and d^2s^2-series {Hf($5d^26s^2$), Zr($4d^25s^2$), Ti($3d^24s^2$)} tend to very strongly favor SA-embedding. The strength of SA-embedding in M-N_2C_2 systems is in the following order: {Pt($4d^96s^1$), Pd($4d^{10}$), Mn($3d^24s^2$)}: very strong, {Au($5d^{10}6s^1$), Ir($5d^76s^1$), Rh($4d^85s^1$), Zn($3d^{10}4s^2$), Ni($3d^84s^2$), Cr($3d^54s^1$), V($3d^34s^2$)}: strong, {Ta($5d^36s^2$), Tc($4d^55s^2$), Nb($4d^35s^2$), Cu($3d^{10}4s^1$), Co($3d^74s^2$), Fe($3d^64s^2$)}: moderate, {Cd($4d^{10}5s^2$), Ag($4d^{10}5s^1$), Ru($4d^75s^1$), Mo($4d^55s^1$)}: weak (which indicates significant clustering into NPs). In the case of N_2C_2, the (TM)SA stability over the TM cohesion ($E_{emb/coh}$ in eV) shows the trend in the order Mn, Sc, Y (-5.2~-4.3: very strong stability) > Ti, Zr, Hf (~-3.8: strong) > Ni, Pt, Pd, Zn, Cu (-3.4~-3.0: somewhat strong) > V, Co, Fe, Cr, Rh, Ir, Ta, Nb (-2.8~-2.2: moderate) > Ru, Cd, Ag, Mo (-1.7~-1.2: somewhat weak) > Os, W, Re, Tc (-0.9~-0.4: weak stability). In some systems, Cd and Hg are not embedded because of their low stabilities.

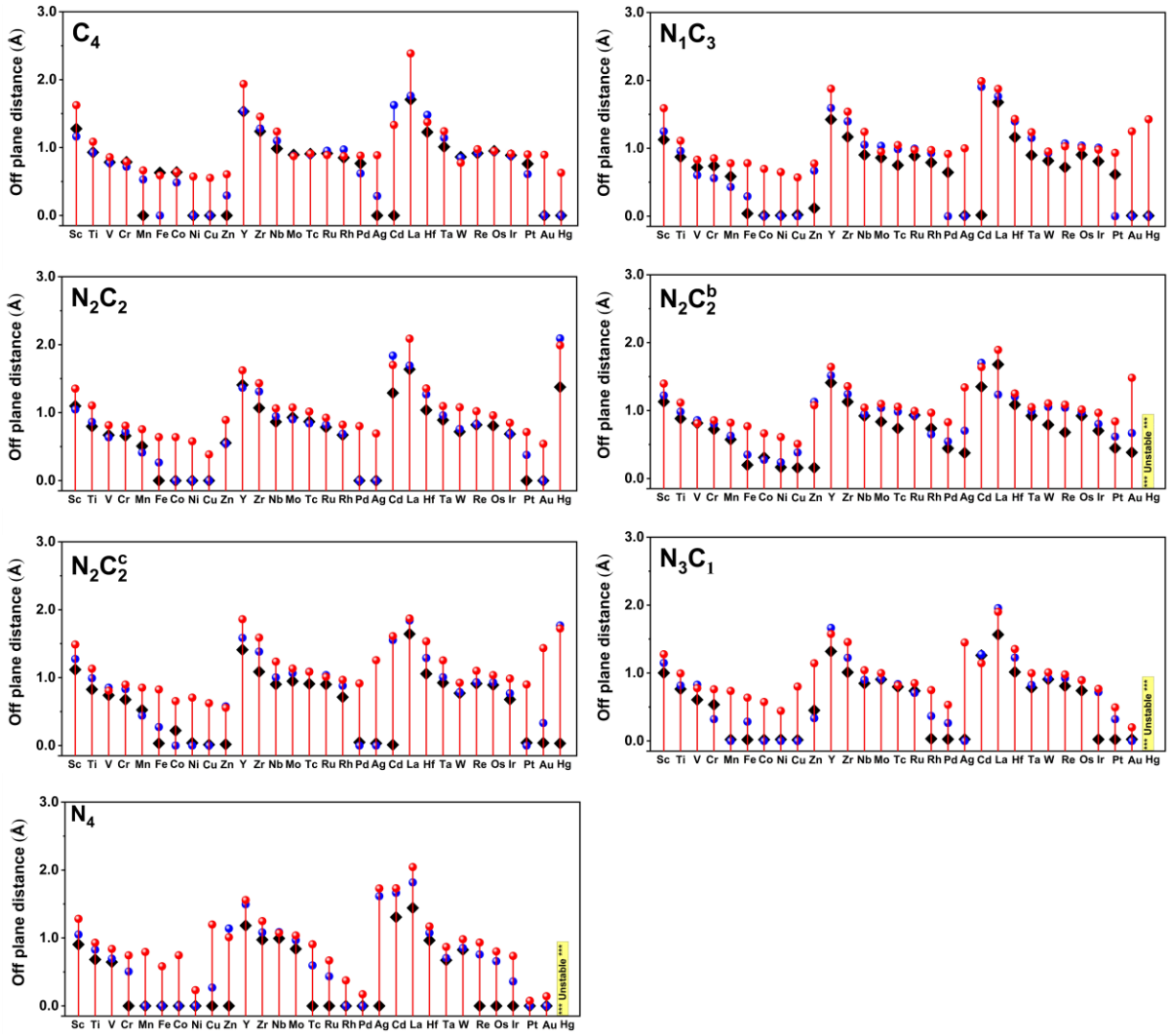


Figure S4. Off-plane distances (Å) of various (TM)SAs for the pristine (black), H-adsorbed (blue) and O-adsorbed (red) M-($N_{n=0-4}C_{4-n}$, $N_{n=0-3}C_{3-n}$)-G_N. Some unstabilities (in yellow bar) are shown for a few cases of Hg (and Cd). See **Tables S8 & S10** for details. Fe-C_4 is an exceptional case that is non-planar without H but is pseudo-planar with a distorted structure upon H-adsorption.

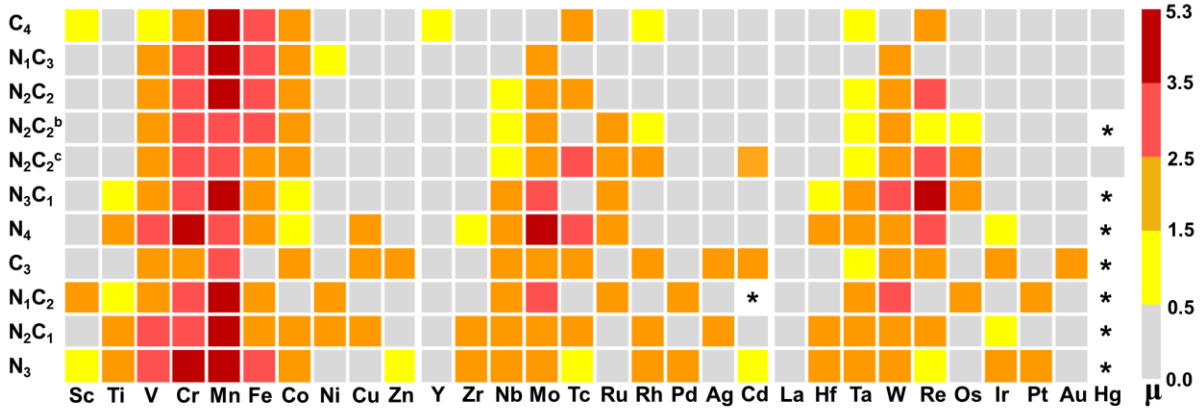


Figure S5. Magnetization color map of (TM)SAs in M–N_nC_m–G_N. Most TM elements show no magnetization, while Cr, Mn, Mo, Re and these neigboring elements show strong magnetization. The metal elements in di-vacancy sites tend to have less magnetization than those in mono-vacancy sites, and more N-doped systems tend to have rather stronger magnetization. See the TM(SA)s magnetization before/after H–adsorption (**Table S15/S16**) and after *O/*OH/*OOH-adsorption (**Table S17**). "*" in the figure denotes an unstable state.

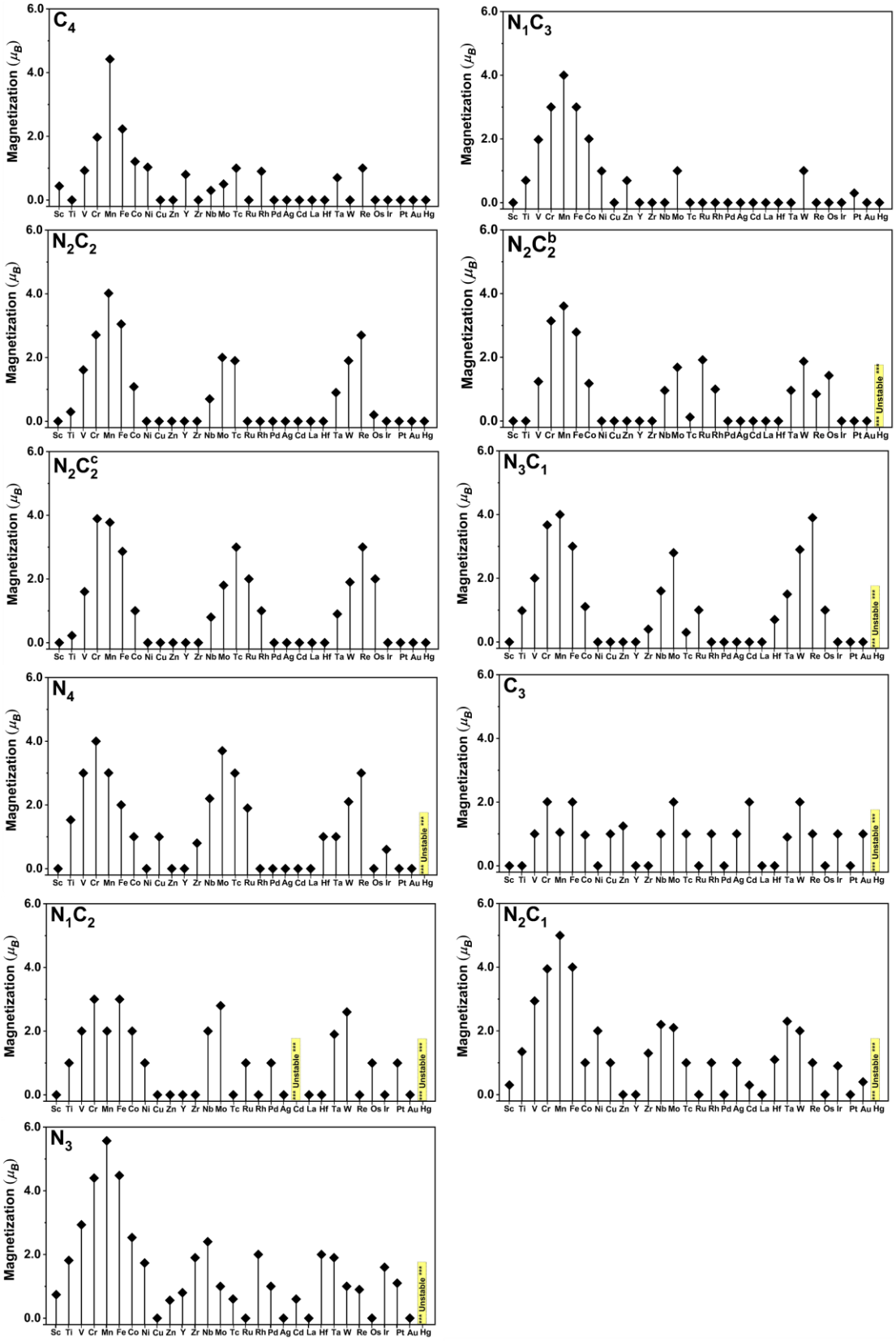


Figure S6. Magnetization of (TM)SAs in M–N_nC_m–G_N. Representation in drop-lines. The magnetization of (TM)SAs changes according to the coordination environment and reflects their unfilled orbital state (see the examples in **Fig. 4** and **Figure S8**).

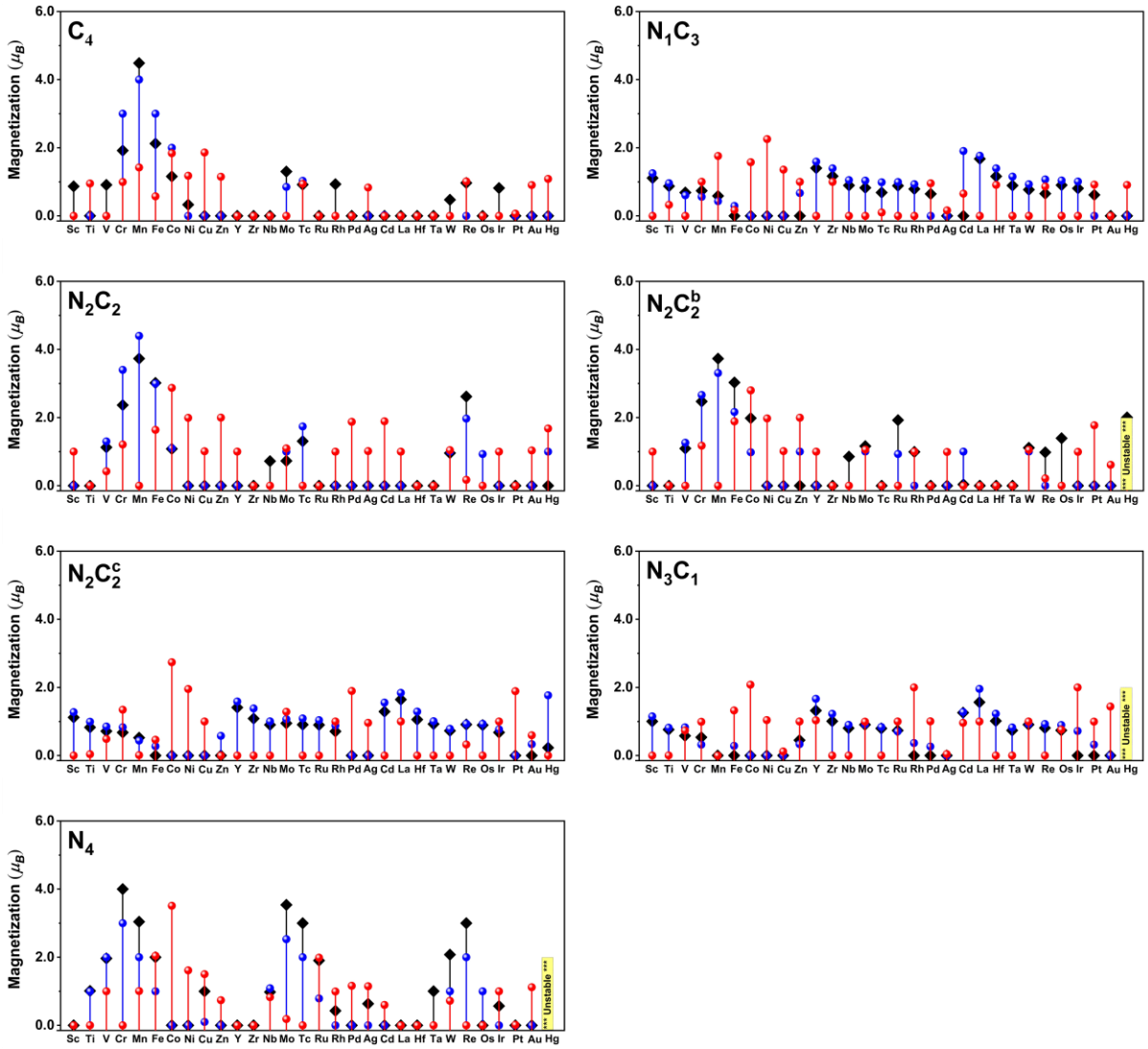


Figure S7. Magnetization of (TM)SAs before (black) and after H/O-adsorption (blue/red) on M–N_nC_m–G_N. See the TM(SA)s magnetization before/after H–adsorption (**Table S15/16**) and after *O/*OH/*OOH-adsorption (**Table S17**). The significant changes in magnetization before/after intermediate adsorption are often noted in 3d transition metals.

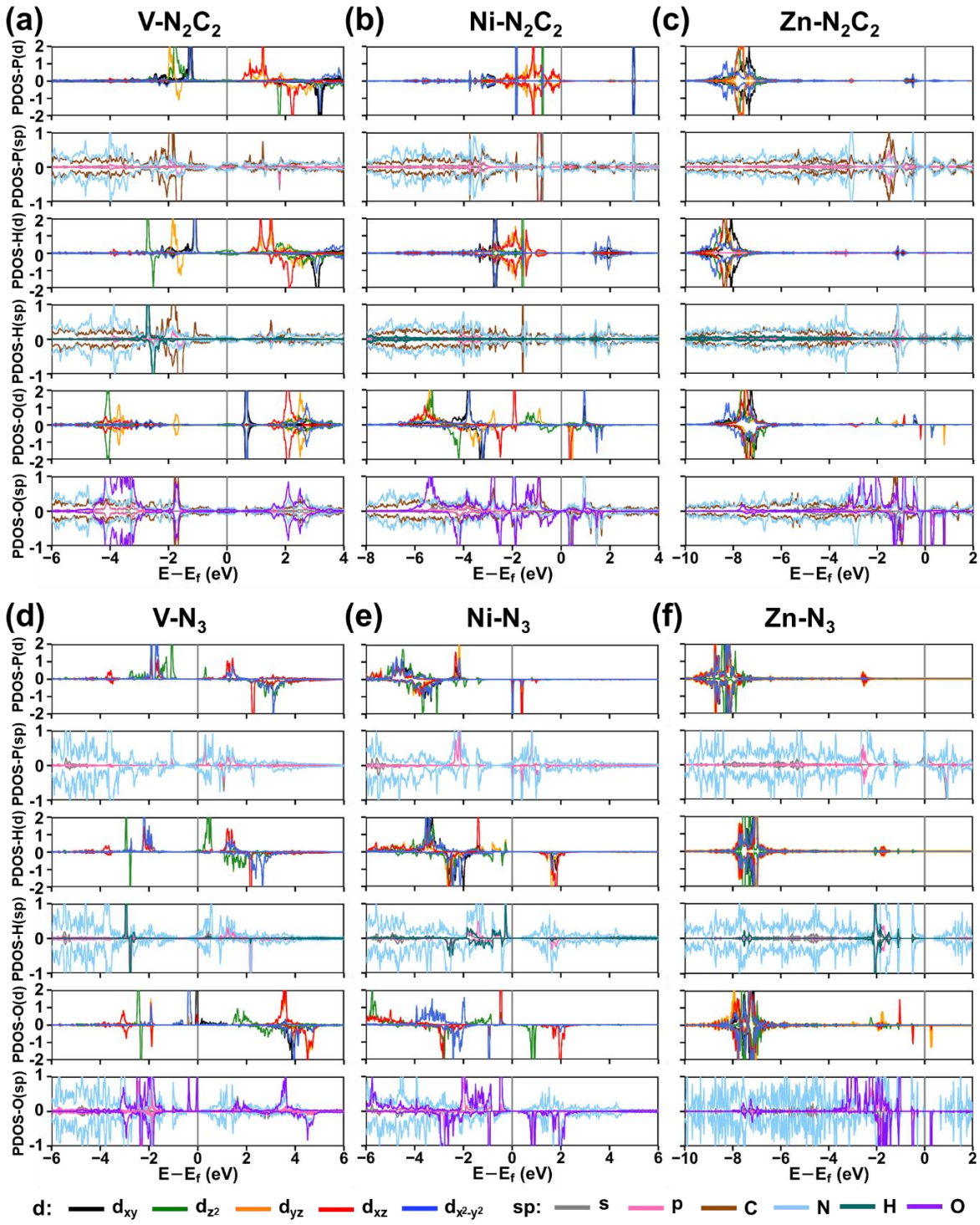


Figure S8. Projected density of states (PDOS) of pristine (PDOS-P), H-adsorbed (PDOS-H), and O-adsorbed (PDOS-O) M–N_2C_2–G_N (a-c) and M–N_3–G_N systems (d-f). a,d, Case for M=V [early transition metal d^3], b,e, Case for M=Ni [square planar d^8; 16 electron rule]. c,f, Case for M=Zn [group 12; fully occupied d orbital d^{10}]. The major-spin (positive) and minor-spin (negative) PDOS of M(d orbitals) are plotted in each upper box, and those of M(s, p orbitals) and C, N and H are plotted in each lower box. (Ni, Zn)-N_2C_2-G_N have planar structures, while V-N_2C_2-G_N has a non-planar structure. See the text for the planar d^4+dsp^2 electron configuration favoring the 16-electron rule and the octahedral sp^3d^2 electron configuration favoring the 18-electron rule in the Ni case.

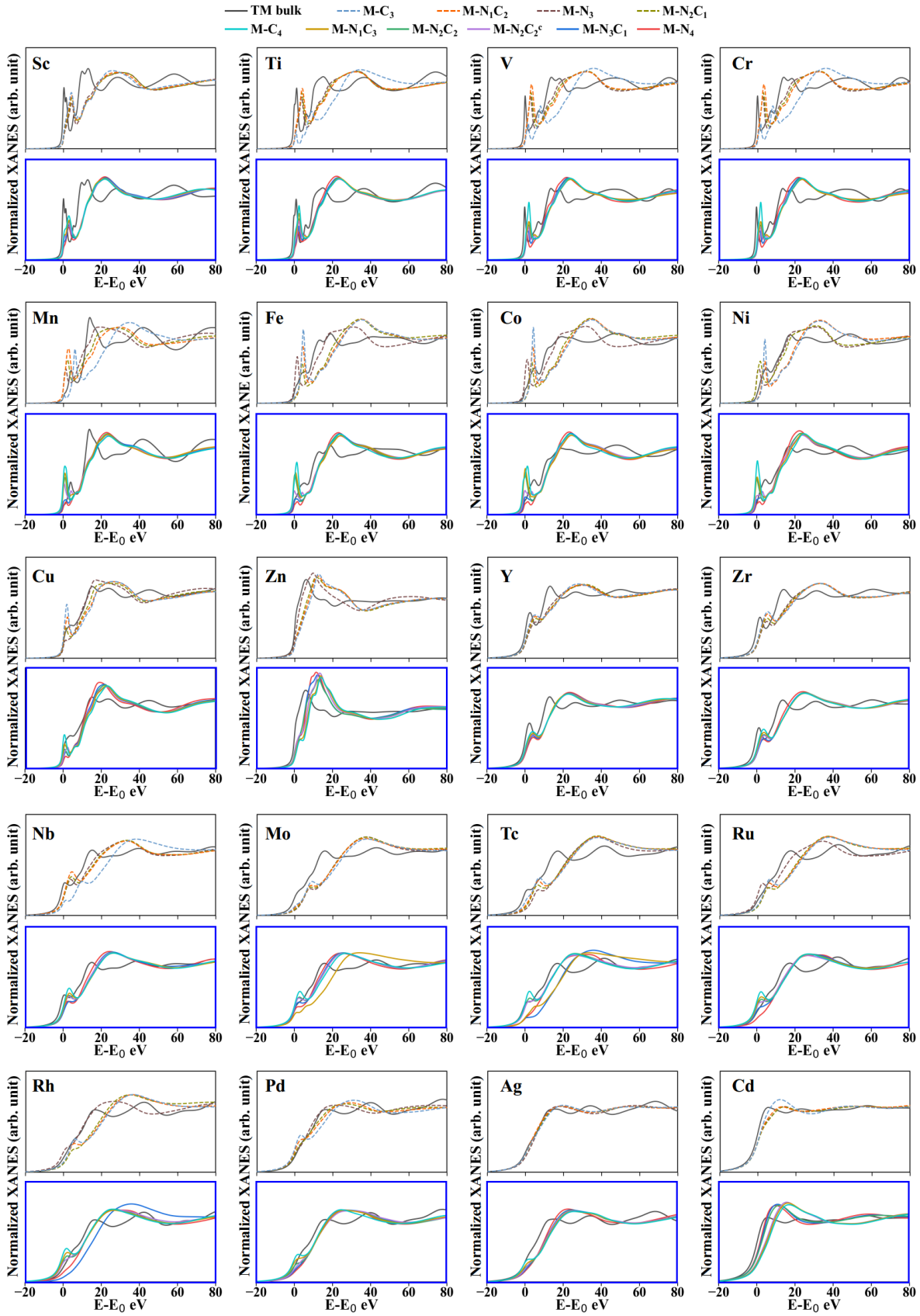


Figure S9. Calculated transition metal K-edge X-ray adsorption near edge structure (XANES) of M-N_nC_m-G_N. The XANES for TM in mono-vacancy site (n+m=3) are in each upper thin black box; those in di-vacancy sites (n+m=4) are in each lower thick blue box. The major difference between coordination environments can be noted at the pre-edge peak.

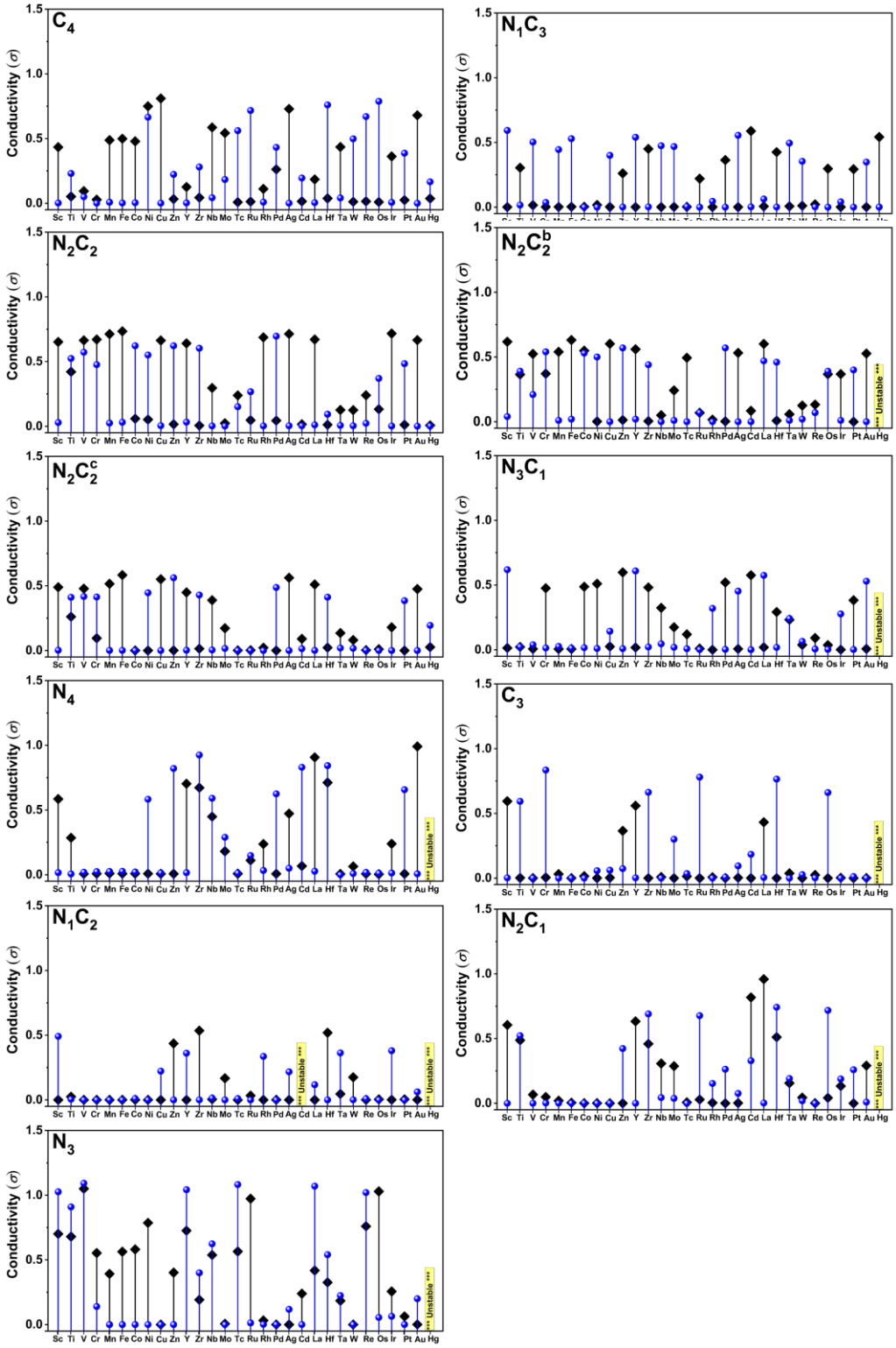


Figure S10. Electrical conductivity (σ) of M–N_nC_m–G_N before (black) and after (blue) H-adsorption with respect to the σ of 2.1% N-doped Gr (σ^o; $\sigma^o/\sigma(Gr) = \sim 7\times10^2$). The σ's are calculated from the band structures. Assuming the scattering rates (τ) of M–N_nC_m–G_N being similar to that of G_N (the experimental value of ~100 fs)[1], the σ(M–N_nC_m–G_N)/σ^o values are reported in **Tables S18-S19**. Overall, among the divalent N_nC_{4-n} moities, the N_2C_2 moiety tends to show high conductivity for most of TM(SA)s embedded in G_N.

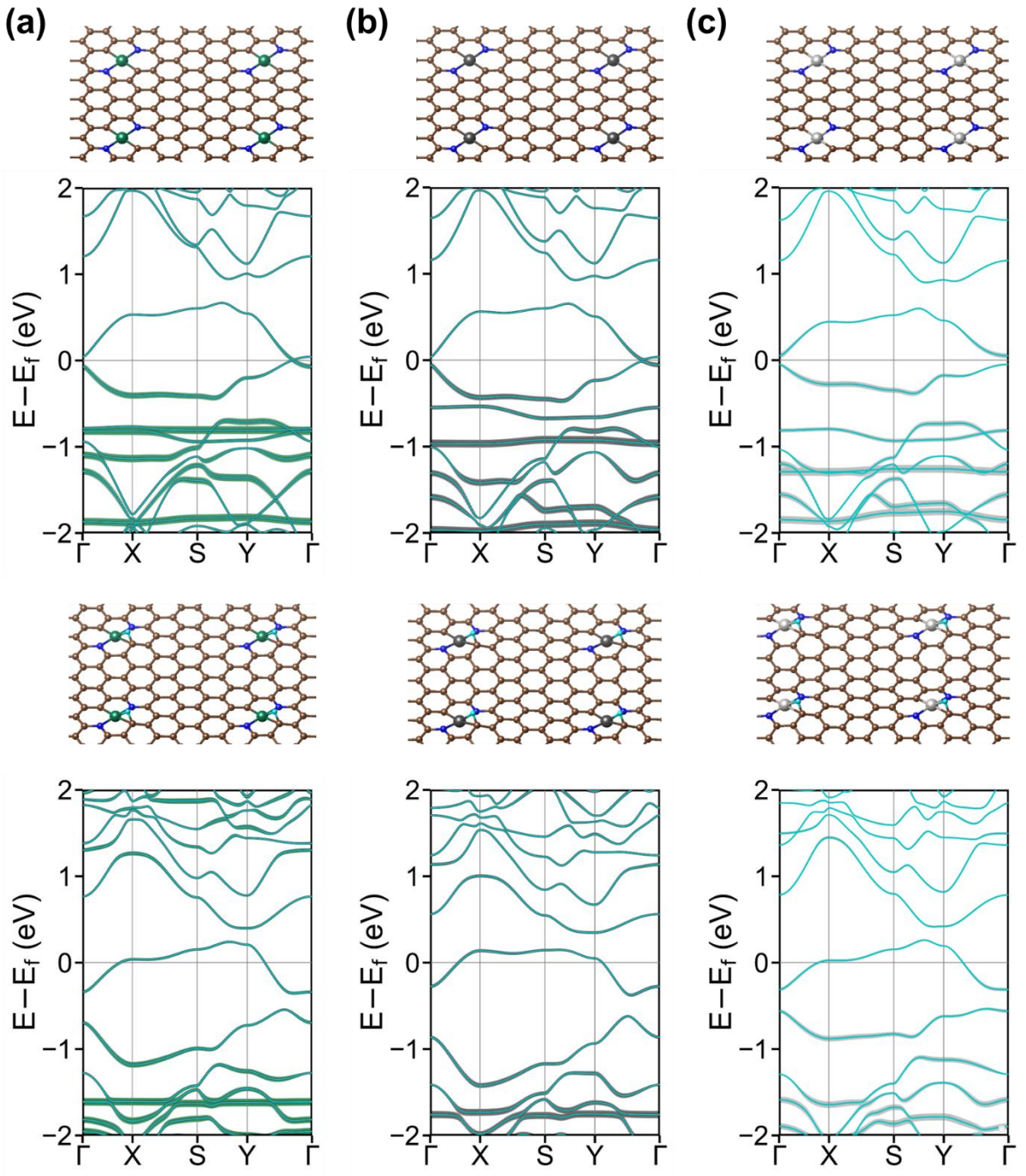


Figure S11. Geometries and band structures before/after H-adsorption. a, Ni–N_2C_2–G_N, b, Pd–N_2C_2–G_N, and c, Pt–N_2C_2–G_N. Ni (dark green), Pd (dark grey), Pt (silver), and G_N (C: brown, N: blue). Black and cyan lines of band structures are major and minor spin components, respectively. The electrical conductivities are calculated from the band structures. These systems are not conductive in their pristine structure due to (very) small band gaps near E_F but become conductive after H-adsorption with one conducting channel at the Fermi level.

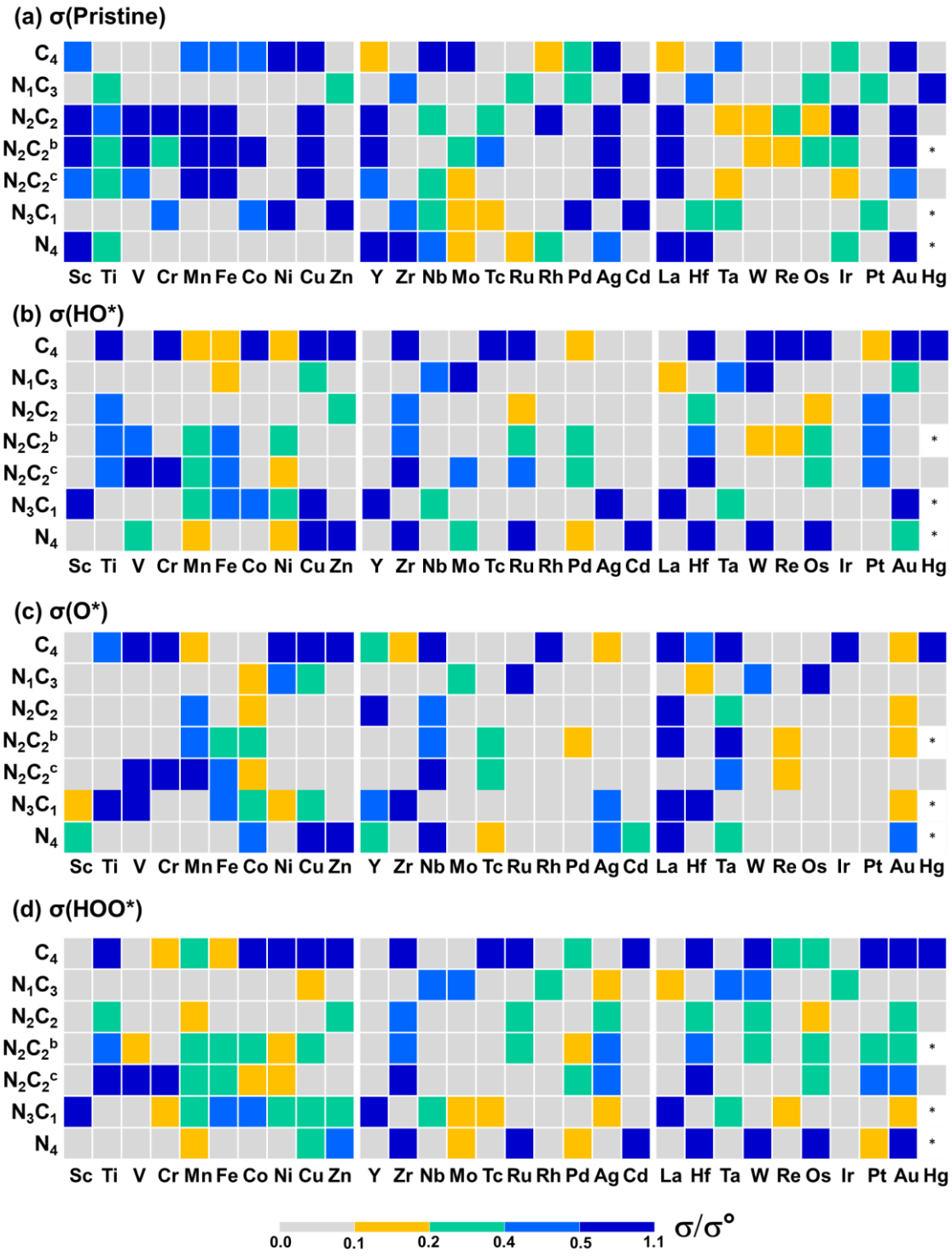


Figure S12. Electrical conductivity (σ) of M–N_nC_m–G_N after HO/O/HOO-adsorption with respect to 2.1 % N-doped Gr (σ^o where $\sigma^o/\sigma(Gr) = \sim 7\times10^2$) "*" denotes an unstable state. The conductivity is reduced after the HO/O/HOO-adsorption in most cases. The $\sigma(M–N_nC_m–G_N)/\sigma^o$ values are reported in **Tables S19** (See **Fig. 5** for the upon H-adsorption).

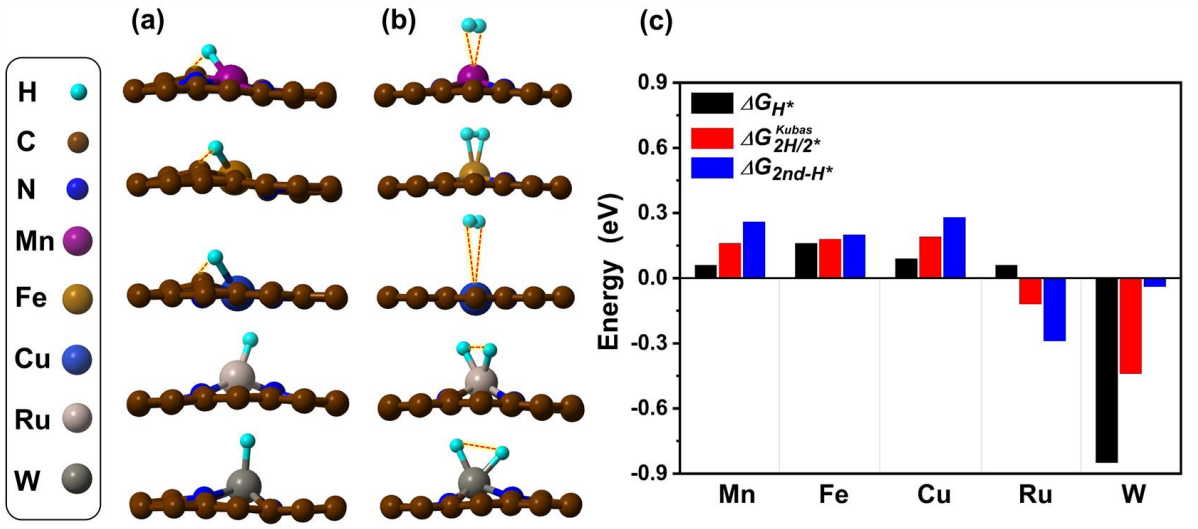


Figure S13. Kubas type HER. a, Volmer-type and b, 2nd H adsorbed conformations of the selected M–N_2C_2–G_N systems and (c) adsorption free energies (eV) per H of 1st H (ΔG_{H^*}), Kubas-type H_2 ($\Delta G^{Kubas}_{2H^*/2}$), and 2nd H ($\Delta G_{2nd-H^*} = 2 \times \Delta G^{Kubas}_{2H^*/2} - \Delta G_{H^*}$) (see **Table S23** for details). Free energies are referenced to gaseous ½ H_2(g) in equilibrium with a solvated proton at the standard condition. When 2nd H adsorbs as Kubas type, the free energy tends to increase because it is generally more difficult to let 2nd H adsorbed on an active metal site as compared with 1st H. However, a few cases show decreased free energies. For example, Ru shows that $\Delta G^{Kubas}_{2H*/2}$ is lower than ΔG_{H*}, which is exceptional. Ru ($4d^7 5s^1$) in Ru–N_2C_2–G_N along with 4 pairs of two electrons (d^2sp hybrid orbitals) from –(N_2C_2), fills 16 electrons in total; it has 17 electrons upon H-adsorption, and then fully occupied 18 electrons upon 2nd H-adsorption. Thus, the 2nd H-adsorption stabilizes the Ru–N_2C_2–G_N system significantly, explaining its particular stability over 1st H-adsorption. Similarly, Tc also shows a similar trend. However, both cases are not effective Kubas systems, because two H atoms attached on Ru is somewhat too far away (>1.6 Å) from each other to form a H_2 molecule easily. The effective Kubas type 2nd H, which is located very close to 1st H, would help in higher turnover frequency. In the case of (Fe,Co)–N_2C_2–G_N, the Kubas reaction helps in accelerating H_2-desorption with $\Delta G^{Kubas}_{2H*/2}$ =0.18 eV per H atom.

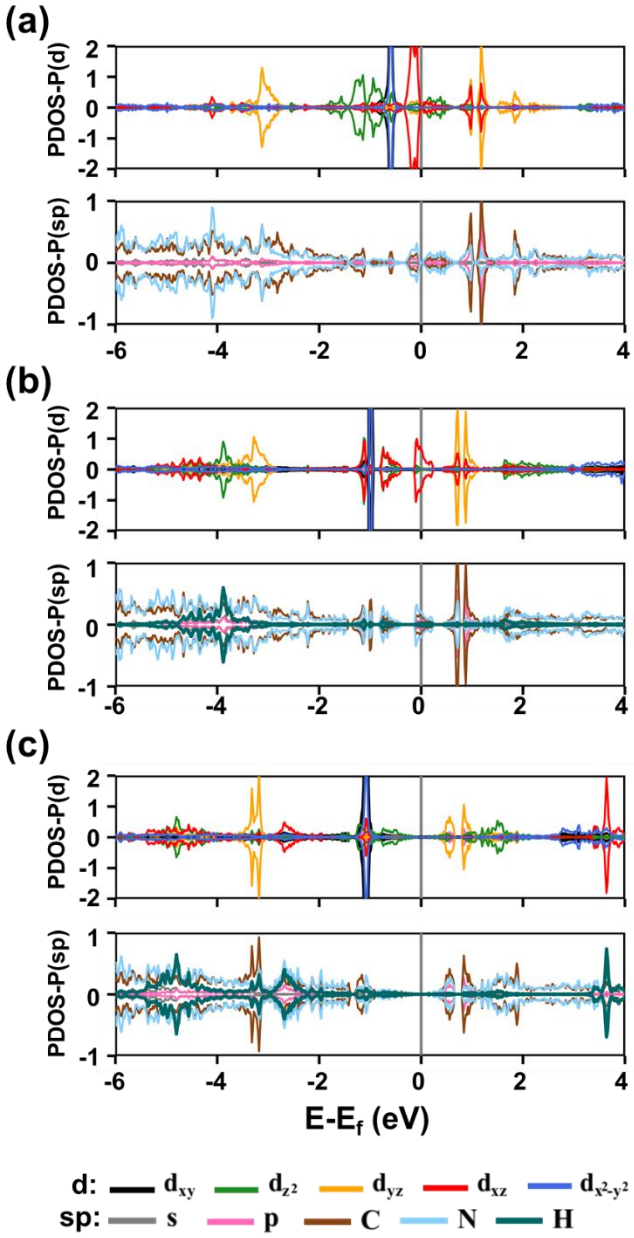


Figure S14. PDOS for a, pristine, b, 1H-adsorbed and **c,** 2H-adsorbed Ru-N_2C_2-G_N. The half-filled antibonding state in 1H adsorbed Ru-N_2C_2-G_N makes ΔG_{H*} higher than $\Delta G_{2H*/2}^{Kubas}$. In Ru-N_2C_2-G_N, the unfilled d_{yz} orbital and antibonding d_{xz} orbital are hybridized. When the first H adsorbs on Ru-N_2C_2-G_N, H interacts with the antibonding d_{xz} state and fills this state by half. After that, the second H interacts with the unfilled d_{yz} state and it breaks the hybridization of unfilled d_{yz} orbital and antibonding d_{xz} orbital. Then, the d orbitals in bonding state are filled with 18 electrons in 2H-adsorbed Ru-N_2C_2-G_N.

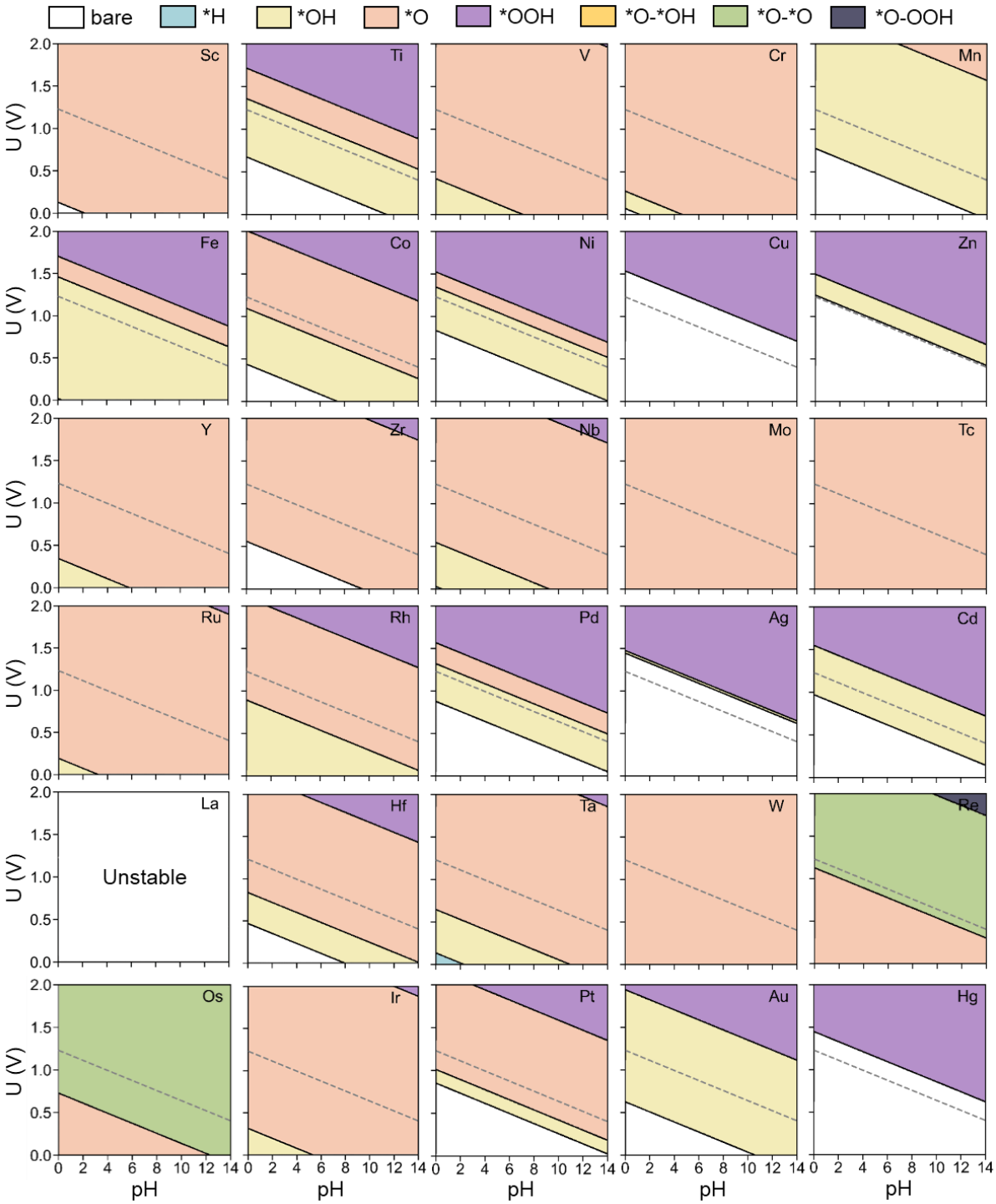


Figure S15. Pourbaix diagram of M-C_4. The pourbaix diagrams are obtained using the Gibbs free energies of each intermediates on metal site at given U and pH. The grey dashed line represent the equilibrium potential for OER/ORR. The intermediates (*O-*OH, *O-*O, *O-*OOH) in path-II are also considered for Re and Os-C_4 (See **Method** for details). Intermdediates are not stable in La-C_4. The *O is expected to form on the active site in most of M-C_4 systems in a 0.7–1.4 V range.

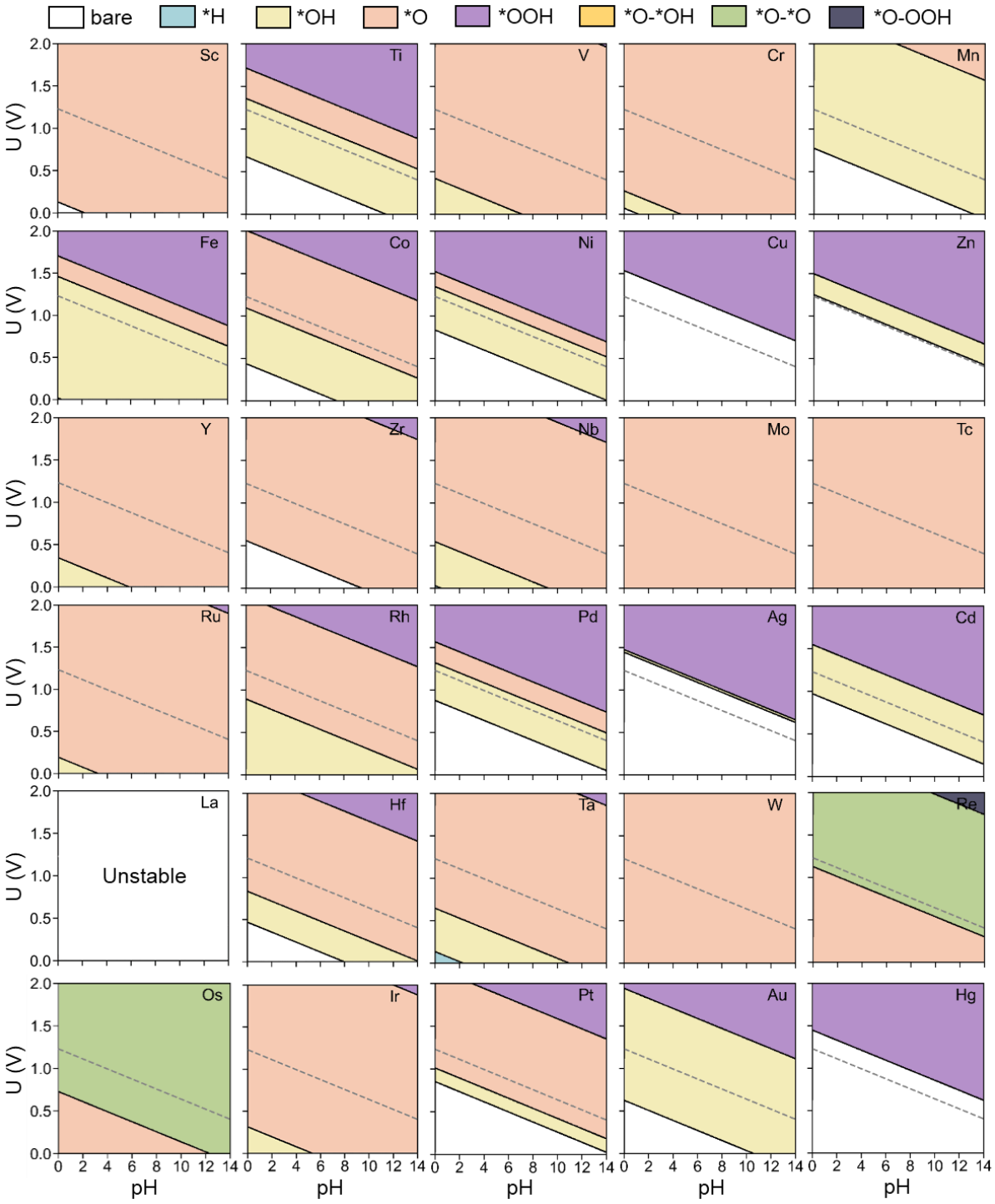


Figure S16. Pourbaix diagram of M-N_2C_2. The pourbaix diagrams are obtained using the Gibbs free energies of each intermediates on metal site at given U and pH. The grey dashed line represent the equilibrium potential for OER/ORR. The intermediates (*O-*OH, *O-*O, *O-*OOH) in path-II are also considered for Fe, Mo, Tc, Ru, Ta, W, Re, and Os-N_2C_2 (See **Methods** for details). Intermdediates are not stable in La-N_2C_2. The *O is expected to form on the active site in most of M-N_2C_2 systems in a 0.7–1.4 V range.

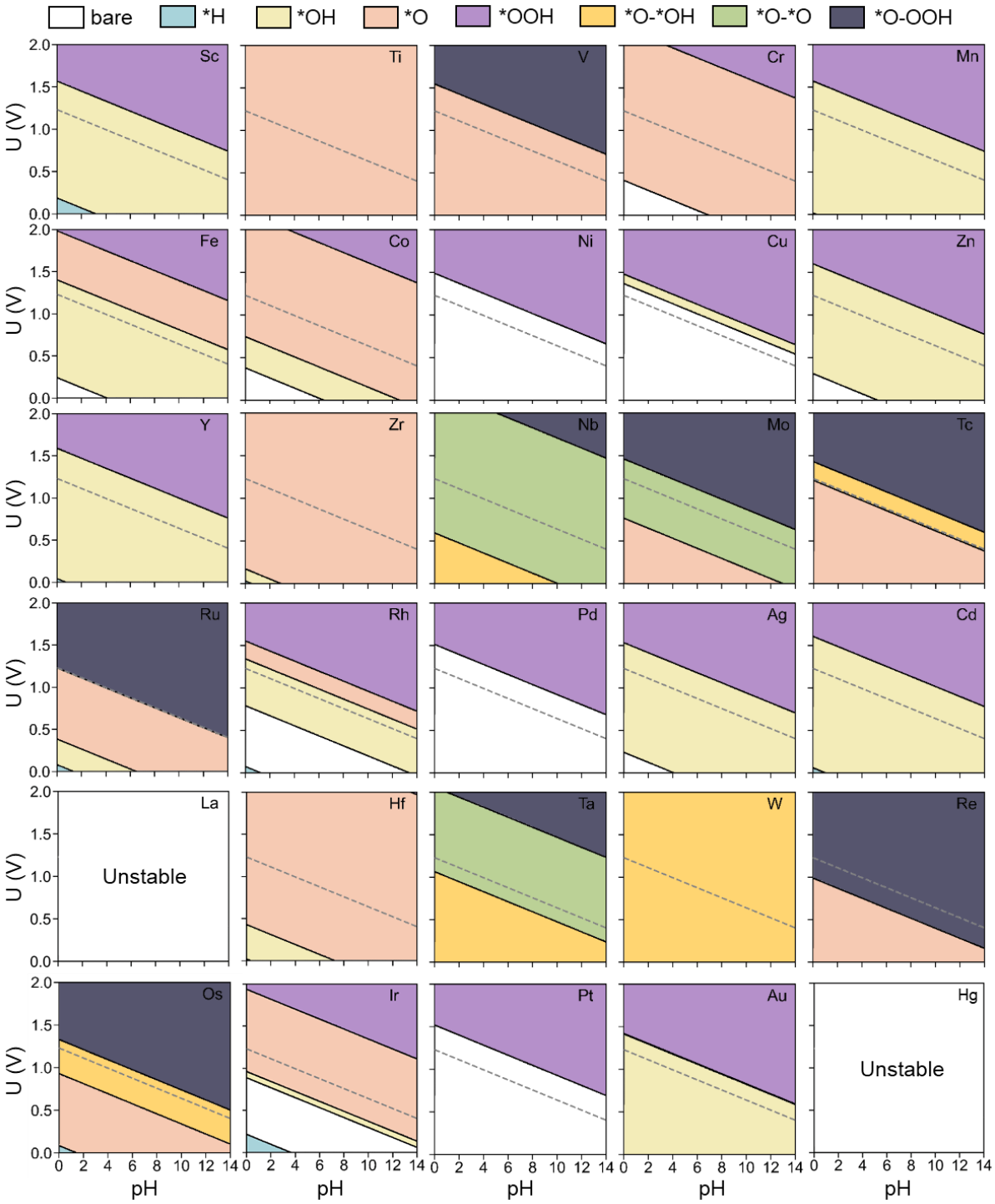


Figure S17. Pourbaix diagram of M-N_4. The pourbaix diagrams are obtained using the Gibbs free energies of each intermediate on a metal site at given U and pH. The grey dashed line represents the equilibrium potential for OER/ORR. The intermediates (*O-*OH, *O-*O, *O-*OOH) in path-II are also considered for V, Nb, Mo, Tc, Ru, Ta, Re, and Os-N_4 (See **Methods** for details). Intermdediates are not stable in La-N_4 and Hg-N_4 is not stable. The *O is expected to form on the active site in most of M-N_4 systems in a 0.7–1.4 V range. For the M-N_4 systems following path-II, The *O formation at the *O-M site is expected in the same range.

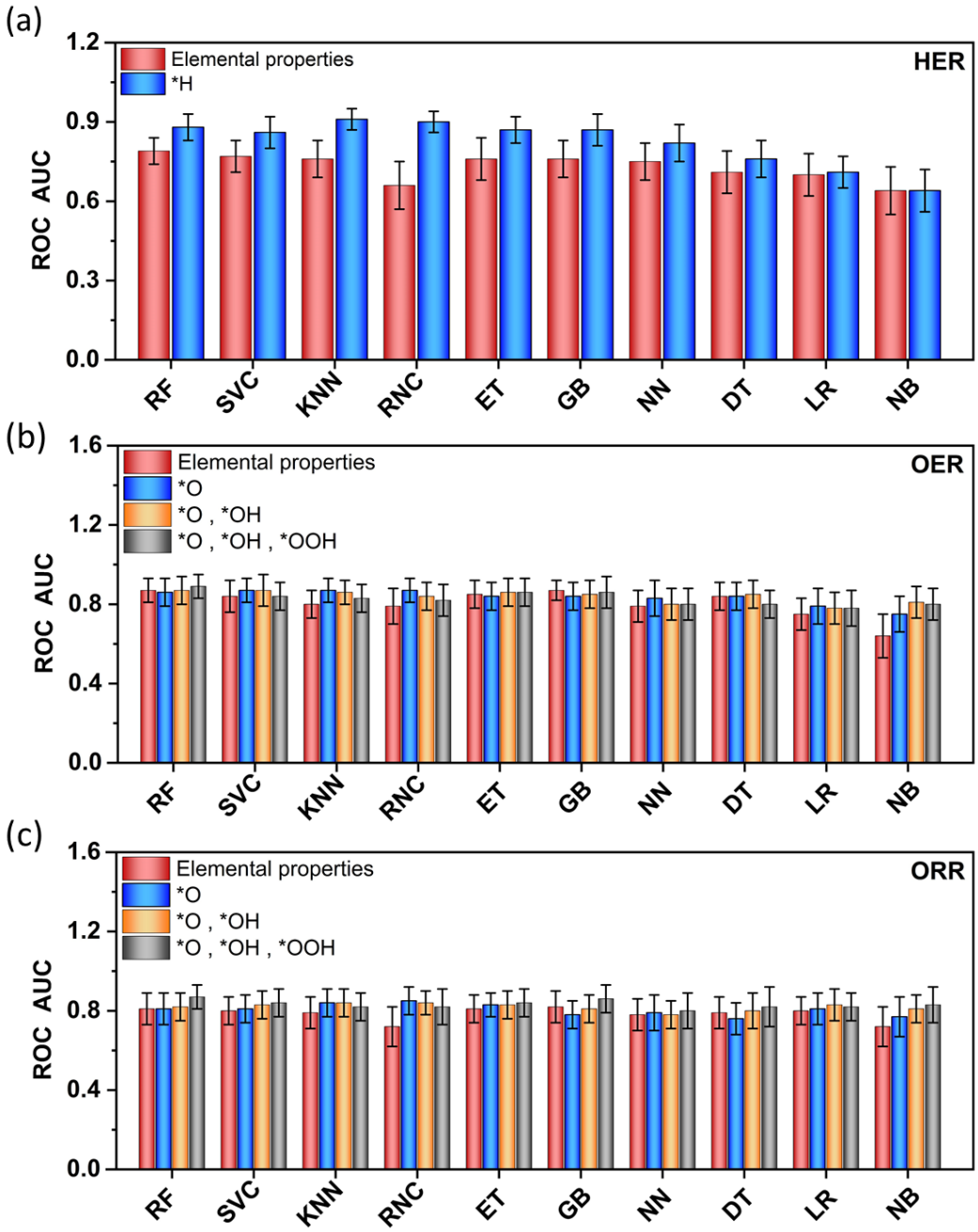


Figure S18. Comparison of predictive accuracy of various machine learning models trained with different descriptors for predicting the overpotentials of HER, OER and ORR. Mean and standard deviation values of the Area Under the Receiver Operating Characteristic Curve (ROC AUC) for different machine learning methods.

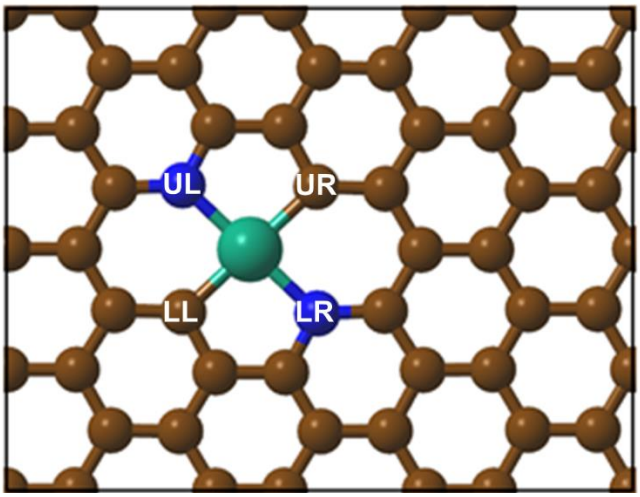


Figure S19. Atom positions used for the Coulomb marix. The atomic positions of upper-left (UL), upper-right (UR), lower-right (LR), and lower-left (LL).

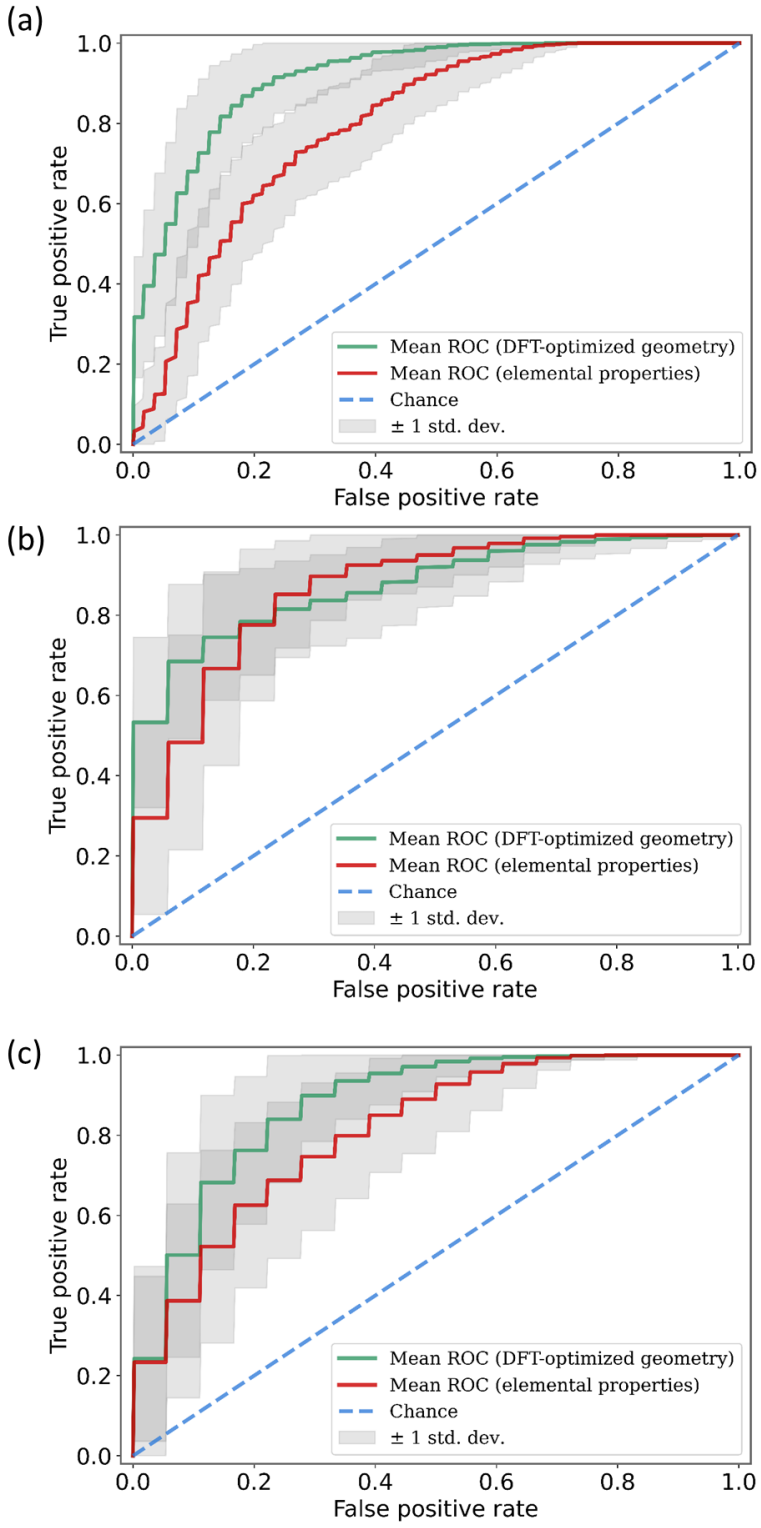


Figure S20. Receiver operating characteristic (ROC) curve for different classifiers. The mean ROC are calculated among curves for 100 random splits into the training and validation sets for predicting the overpotentials of (a) HER, (b) OER and (c) ORR.

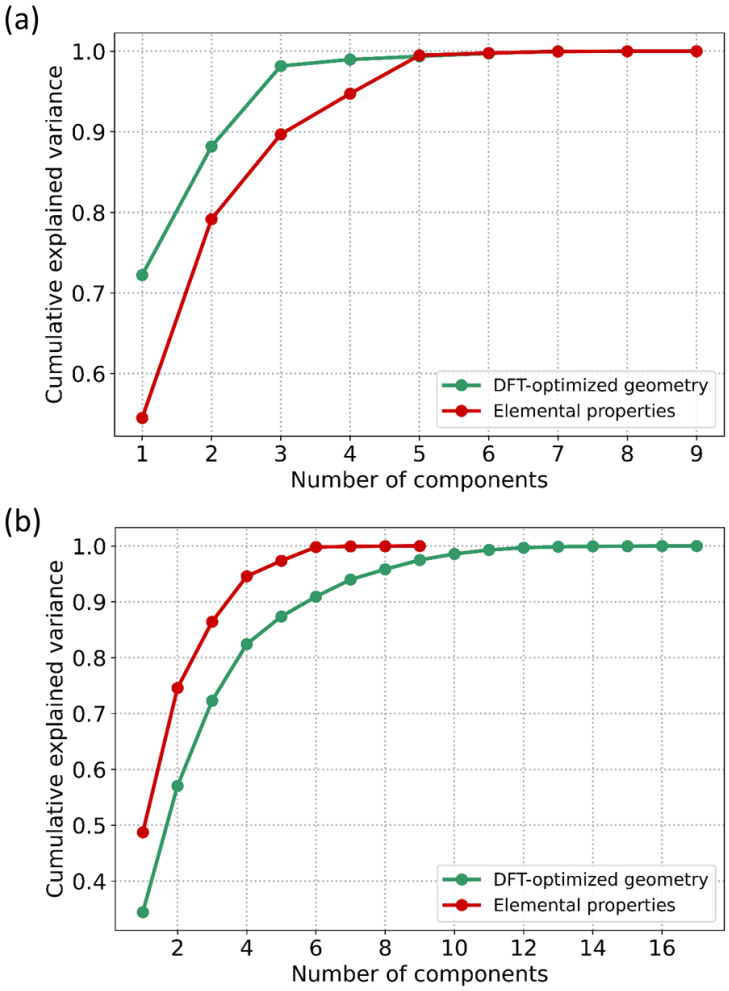


Figure S21. Cumulative explained variance ratios as a function of the number of principal components. a, HER: best classifiers (random forest in the case of elemental properties, and k-nearest neighbors in the case of DFT-optimized geometry). b, OER and ORR: best classifier (random forest).

Supplementary Tables

Table S1. Experimental (E_{coh}^{exp}) and calculated(E_{coh}^{DFT}) cohesive energy (eV) of bulk transition metals. The difference of the calculated cohesive energy from the experimental value ($E_{coh}^{DFT} - E_{coh}^{exp}$) is given in brackets.

Transition Metal	E_{-coh}^{exp}	E_{-coh}^{DFT}					
		PBE	PBE+TS	RPBE	RPBE+TS	RPBE(U)+TS	AVG[b]
Sc HCP	3.90	4.14 [0.24]	5.07 [1.17]	3.76 [-0.14]	4.64 [0.74]	4.01 [0.11]	4.33 [0.43]
Ti HCP	4.84	5.42 [0.58]	6.53 [1.69]	4.93 [0.09]	6.01 [1.17]	4.46 [-0.38]	5.23 [0.39]
V BCC	5.30	5.38 [0.08]	6.55 [1.25]	4.86 [-0.44]	6.02 [0.72]	3.93 [-1.37]	4.98 [-0.32]
Cr BCC	4.09	4.05 [-0.04]	5.42 [1.33]	3.47 [-0.62]	4.81 [0.72]	2.26 [-1.83]	3.54 [-0.55]
Fe BCC	4.28	4.86 [0.58]	5.77 [1.49]	4.25 [-0.03]	5.13 [0.85]	3.35 [-0.93]	4.24 [-0.04]
Co HCP	4.43	5.15 [0.72]	5.97 [1.54]	4.57 [0.14]	5.38 [0.95]	3.54 [-0.89]	4.46 [0.03]
Ni FCC	4.44	4.79 [0.35]	5.78 [1.34]	4.21 [-0.23]	5.17 [0.73]	3.91 [-0.53]	4.54 [0.10]
Cu FCC	3.48	3.49 [0.01]	4.10 [0.62]	3.01 [-0.47]	3.59 [0.11]	3.18 [-0.30]	3.39 [-0.09]
Zn HCP	1.35	1.09 [-0.26]	1.51 [0.16]	0.72 [-0.63]	1.12 [-0.23]	1.11 [-0.24]	1.11 [-0.24]
Y HCP	4.39	4.17 [-0.22]	4.95 [0.56]	3.78 [-0.61]	4.53 [0.14]	4.53 [0.14]	4.53 [0.14]
Zr HCP	6.29	6.20 [-0.09]	7.36 [1.07]	5.74 [-0.55]	6.87 [0.58]	6.87 [0.58]	6.87 [0.58]
Nb BCC	7.44	7.00 [-0.44]	8.39 [0.95]	6.40 [-1.04]	7.76 [0.32]	7.76 [0.32]	7.76 [0.32]
Mo BCC	6.80	6.26 [-0.54]	7.71 [0.91]	5.61 [-1.19]	7.04 [0.24]	7.04 [0.24]	7.04 [0.24]
Tc HCP	7.13	6.92 [-0.21]	9.15 [2.02]	6.22 [-0.91]	8.41 [1.28]	8.41 [1.28]	8.41 [1.28]
Ru HCP	6.74	6.75 [0.01]	7.80 [1.06]	6.00 [-0.74]	7.02 [0.28]	7.02 [0.28]	7.02 [0.28]
Rh FCC	5.72	5.75 [0.03]	6.54 [0.82]	5.03 [-0.69]	5.80 [0.08]	5.80 [0.08]	5.80 [0.08]
Pd FCC	3.90	3.73 [-0.17]	4.00 [0.10]	3.11 [-0.79]	3.37 [-0.53]	3.37 [-0.53]	3.37 [-0.53]
Ag FCC	2.94	2.52 [-0.42]	2.97 [0.03]	2.01 [-0.93]	2.43 [-0.51]	2.43 [-0.51]	2.43 [-0.51]
Cd HCP	1.16	0.72 [-0.44]	1.11 [-0.05]	0.33 [-0.83]	0.70 [-0.46]	0.70 [-0.46]	0.70 [-0.46]
Hf HCP	6.42	6.43 [0.01]	7.50 [1.08]	5.92 [-0.50]	6.96 [0.54]	6.96 [0.54]	6.96 [0.54]
Ta BCC	8.09	8.12 [0.03]	9.25 [1.16]	7.49 [-0.60]	8.60 [0.51]	8.60 [0.51]	8.60 [0.51]
W BCC	8.79	8.84 [0.05]	9.99 [1.20]	8.10 [-0.69]	9.23 [0.44]	9.23 [0.44]	9.23 [0.44]
Re HCP	8.02	7.76 [-0.26]	8.82 [0.80]	7.00 [-1.02]	8.05 [0.03]	8.05 [0.03]	8.05 [0.03]
Os HCP	8.17	8.34 [0.17]	9.38 [1.21]	7.58 [-0.59]	8.62 [0.45]	8.62 [0.45]	8.62 [0.45]
Ir FCC	6.92	7.23 [0.31]	7.79 [0.87]	6.55 [-0.37]	7.10 [0.18]	7.10 [0.18]	7.10 [0.18]
Pt FCC	5.85	5.49 [-0.36]	6.01 [0.16]	4.80 [-1.05]	5.30 [-0.55]	5.30 [-0.55]	5.30 [-0.55]
Au FCC	3.81	3.04 [-0.77]	3.42 [-0.39]	2.45 [-1.36]	2.81 [-1.00]	2.81 [-1.00]	2.81 [-1.00]
ME		-0.04	0.89	-0.62	0.29	-0.16	0.06
MAE		0.27	0.93	0.64	0.53	0.54	0.38
MSE		0.12	1.13	0.52	0.39	0.47	0.23

ME: mean error, **MAE:** mean absolute error, **MSE:** mean square error. HCP**:** hexagonal closed packed, BCC**:** body centered cubic, FCC**:** face centered cubic. Since Mn and La have quite different crysral structures from other transition metals and Hg is liquid, their direct comparison between experimental and calaculation cohesive energies are not made here. Energy cutoff for Y is used with 600eV due to its low energy maximum of pseudopotential. For covenience's sake (because the cohesive energy is oppositely signed with respect to the formation energy), we define $E_{-coh} \equiv -E_{coh}$ for our further discussion. For the 3d metals in the order of Sc through Zn, the Hubbard U_{eff} are used with 2.11, 2.58, 2.72, 2.79, 3.06, 3.29, 3.42, 3.40, 3.87, and 4.12 eV, according to the literature based on HER study[2]. [b]Average value of RPBE+TS and RPBE(U)+TS.

Table S2. Experimental (d^{exp}) and calculated (d^{DFT}) shortest interatomic distances (Å) of bulk transition metals. The difference from the calculated distance to the experimental value (d^{DFT}–d^{exp}) is given in parentheses.

Transition Metal	d^{exp}	d^{DFT}					
		PBE	PBE+TS	RPBE	RPBE+TS	RPBE(U)+TS	AVG
Sc HCP	3.254	3.244 (-0.010)	3.087 (-0.167)	3.286 (0.032)	3.132 (-0.122)	3.211 (-0.043)	3.171 (-0.083)
Ti HCP	2.897	2.880 (-0.017)	2.789 (-0.108)	2.906 (0.009)	2.813 (-0.084)	2.860 (-0.037)	2.836 (-0.061)
V BCC	2.613	2.595 (-0.018)	2.543 (-0.07)	2.615 (0.002)	2.557 (-0.056)	2.590 (-0.023)	2.574 (-0.039)
Cr BCC	2.498	2.463 (-0.035)	2.406 (-0.092)	2.482 (-0.016)	2.421 (-0.077)	2.437 (-0.061)	2.429 (-0.069)
Fe BCC	2.460	2.455 (-0.005)	2.412 (-0.048)	2.479 (0.019)	2.431 (-0.029)	2.520 (0.060)	2.476 (0.016)
Co HCP	2.497	2.476 (-0.021)	2.443 (-0.054)	2.499 (0.002)	2.462 (-0.035)	2.464 (-0.033)	2.463 (-0.034)
Ni FCC	2.493	2.485 (-0.008)	2.418 (-0.075)	2.511 (0.018)	2.438 (-0.055)	2.406 (-0.087)	2.422 (-0.071)
Cu FCC	2.556	2.568 (0.012)	2.505 (-0.051)	2.599 (0.043)	2.533 (-0.023)	2.521 (-0.035)	2.527 (-0.029)
Zn HCP	2.665	2.666 (0.001)	2.603 (-0.062)	2.704 (0.039)	2.639 (-0.026)	2.624 (-0.041)	2.632 (-0.033)
Y HCP	3.556	3.549 (-0.007)	3.403 (-0.153)	3.591 (0.035)	3.453 (-0.103)	3.453 (-0.103)	3.453 (-0.103)
Zr HCP	3.179	3.184 (0.005)	3.067 (-0.112)	3.207 (0.028)	3.092 (-0.087)	3.092 (-0.087)	3.092 (-0.087)
Nb BCC	2.859	2.871 (0.012)	2.776 (-0.083)	2.887 (0.028)	2.789 (-0.070)	2.789 (-0.070)	2.789 (-0.070)
Mo BCC	2.725	2.740 (0.015)	2.672 (-0.053)	2.753 (0.028)	2.683 (-0.042)	2.683 (-0.042)	2.683 (-0.042)
Tc HCP	2.710	2.718 (0.008)	2.628 (-0.082)	2.730 (0.020)	2.638 (-0.072)	2.638 (-0.072)	2.638 (-0.072)
Ru HCP	2.650	2.664 (0.014)	2.620 (-0.030)	2.676 (0.026)	2.632 (-0.018)	2.632 (-0.018)	2.632 (-0.018)
Rh FCC	2.539	2.708 (0.169)	2.669 (0.130)	2.727 (0.188)	2.685 (0.146)	2.685 (0.146)	2.685 (0.146)
Pd FCC	2.753	2.786 (0.033)	2.765 (0.012)	2.812 (0.059)	2.790 (0.037)	2.790 (0.037)	2.790 (0.037)
Ag FCC	2.889	2.928 (0.039)	2.874 (-0.015)	2.977 (0.088)	2.911 (0.022)	2.911 (0.022)	2.911 (0.022)
Cd HCP	2.979	3.040 (0.061)	2.961 (-0.018)	3.097 (0.118)	3.012 (0.033)	3.012 (0.033)	3.012 (0.033)
Hf HCP	3.131	3.134 (0.003)	3.043 (-0.088)	3.162 (0.031)	3.067 (-0.064)	3.067 (-0.064)	3.067 (-0.064)
Ta BCC	2.860	2.870 (0.010)	2.803 (-0.057)	2.889 (0.029)	2.817 (-0.043)	2.817 (-0.043)	2.817 (-0.043)
W BCC	2.741	2.760 (0.019)	2.713 (-0.028)	2.770 (0.029)	2.722 (-0.019)	2.722 (-0.019)	2.722 (-0.019)
Re HCP	2.567	2.752 (0.185)	2.715 (0.148)	2.761 (0.194)	2.725 (0.158)	2.718 (0.151)	2.718 (0.151)
Os HCP	2.675	2.693 (0.018)	2.661 (-0.014)	2.702 (0.027)	2.670 (-0.005)	2.670 (-0.005)	2.670 (-0.005)
Ir FCC	2.715	2.741 (0.026)	2.719 (0.004)	2.750 (0.035)	2.729 (0.014)	2.729 (0.014)	2.729 (0.014)
Pt FCC	2.772	2.804 (0.032)	2.777 (0.005)	2.819 (0.047)	2.794 (0.022)	2.794 (0.022)	2.794 (0.022)
Au FCC	2.879	2.938 (0.059)	2.906 (0.027)	2.968 (0.089)	2.934 (0.055)	2.934 (0.055)	2.934 (0.055)
ME		0.0222	-0.0420	0.0461	-0.0201	-0.0125	-0.0163
MAE		0.0312	0.0662	0.0473	0.0561	0.0529	0.0535
MSE		0.0029	0.0065	0.0046	0.0047	0.0041	0.0042

[a] See the legend of Table S1.

Table S3. Vacancy formation energies (E_v in eV) of N_nC_m-G_N by the presence of h H atoms [$E_v = E_{H_hN_nC_mGr} + n\mu_C - (E_{C_{m+n}Gr} + n\mu_N + h\mu_H)$].

Number of H (h)	C_4	N_1C_3	N_2C_2	N_2C_2[b]	N_2C_2[c]	N_3C_1	N_4	C_3	N_1C_2	N_2C_1	N_3
0H	8.44	7.96	7.35	7.48	6.44	5.96	4.62	7.54	5.64	5.30	4.09
1H	7.68	5.87	5.14	5.25	5.10	3.70	3.00	5.66	4.78	3.32	1.79
2H	6.67	4.81	3.01	3.14	3.08	2.43	1.69	4.92	2.90	1.97	1.99
3H	4.41	2.52	2.60	2.61	2.25	2.11	2.36	3.15	2.47	2.76	3.08
4H	2.86	2.93	2.41	2.43	3.17	2.95	3.17	-	-	-	-

In the absence of metal SAs, in general, highly N-doped moieties can be more easily formed. However, the vacancy formation is often prcessed through harsh conditions such as high temperature, acid treatment, plasma treatment, laser beam, ultrasonics, etc. Then, even for large energy differences, various possibilities occur, and so diverse structures can be generated. Nevertheless, the H adsorption pheneomena are governed almost by steady state thermodyanamics. Most moieties for N_nC_m-G_N favor to form 2H or 3H adsorbed structures, while C_4 and N_2C_2 favor 4H and N_3 favors 1H.

Table S4. Vacancy formation energies (E_v in eV) of pyrrolic defect sites in N-doped graphene by the presence of h H atoms [$E_v = E_{H_hN_nC_mGr} + n\mu_C - (E_{C_{m+n}Gr} + n\mu_N + h\mu_H)$].

Number of H (h)	C_4	N_1C_3	N_2C_2	$N_2C_2^b$	$N_2C_2^c$	N_3C_1	N_4	C_3	N_1C_2	N_2C_1	N_3
0H	-	-	16.09	-	16.08	14.85	13.68	13.89	12.67	11.14	9.94
1H	-	-	13.43	-	13.42	12.18	11.27	11.83	10.03	8.45	7.23
2H	-	-	10.79	-	10.78	9.89	8.92	9.49	7.71	6.73	5.82
3H	-	-	9.61	-	9.59	8.69	8.17	7.37	6.52	5.97	5.45
4H	-	-	8.51	-	8.57	8.07	7.67	-	-	-	-

In comparison of the formation energies of pyrrolic and pyridinic defect sites, it is easier to form pyridinic defect sites than pyrrolic defect sites. The C_4, N_1C_3 and $N_2C_2^b$ types of pyrrolic defect sites change to pyridinic defect sites by geometry optimization.

Table S5. (TM)SA+vacancy formation energy [$E_f = E_{MN_nC_mGr} + n\mu_C - (E_{C_{m+n}Gr} + n\mu_N + \mu_M)$ in eV] for porphyrin type configurations of TM embedded SAC in M-N_nC_m-G_N (n+m= 4: di-vacancy sites, n+m=3: mono-vacancy sites). (RPBE(U)+TS results are in parentheses).

Transition Metal	C_4	N_1C_3	N_2C_2	N_2C_2[b]	N_2C_2[c]	N_3C_1	N_4	C_3	N_1C_2	N_2C_1	N_3
Sc	7.07 (7.15)	5.13 (5.25)	3.66 (3.80)	3.72 (3.85)	3.70 (3.84)	2.23 (2.38)	1.22 (1.39)	6.00 (6.21)	4.35 (4.55)	3.52 (3.70)	2.82 (2.94)
Ti	6.67 (6.55)	5.40 (5.33)	4.16 (4.09)	4.27 (4.18)	4.20 (4.09)	3.36 (3.02)	2.52 (2.18)	5.79 (5.74)	5.22 (5.07)	4.61 (4.44)	3.96 (3.65)
V	7.29 (6.65)	6.28 (5.54)	5.21 (4.30)	5.33 (4.39)	5.18 (4.24)	4.23 (3.19)	3.21 (1.95)	6.56 (6.00)	5.71 (4.94)	5.09 (4.11)	4.45 (3.20)
Cr	7.69 (6.58)	6.66 (5.26)	5.46 (4.04)	5.73 (4.23)	5.54 (4.05)	4.36 (2.70)	3.03 (1.21)	6.56 (5.52)	5.88 (4.55)	5.64 (4.08)	5.40 (3.48)
Mn	7.81 (5.64)	6.01 (3.82)	5.02 (2.87)	4.93 (2.74)	4.73 (2.57)	3.89 (1.70)	2.92 (0.97)	6.28 (5.62)	6.08 (4.82)	5.62 (3.11)	4.82 (1.99)
Fe	7.43 (6.95)	6.25 (5.48)	5.36 (4.61)	5.20 (4.45)	4.90 (4.19)	4.14 (3.55)	3.05 (2.36)	6.09 (6.34)	5.68 (5.65)	5.56 (4.91)	5.10 (3.98)
Co	7.31 (6.82)	6.33 (5.39)	5.22 (4.54)	5.07 (4.39)	4.72 (4.09)	3.86 (3.14)	2.82 (2.04)	5.79 (5.65)	5.54 (5.32)	5.31 (4.64)	4.92 (3.96)
Ni	7.23 (7.24)	5.75 (5.78)	4.66 (4.42)	4.49 (4.29)	4.06 (3.87)	3.42 (3.10)	2.62 (2.19)	6.50 (6.19)	6.05 (5.54)	5.70 (4.95)	5.17 (4.39)
Cu	6.84 (6.97)	5.50 (5.58)	5.04 (5.11)	4.78 (4.84)	4.43 (4.47)	4.24 (4.24)	3.46 (3.21)	7.92 (7.89)	6.64 (6.49)	5.83 (5.63)	4.83 (4.51)
Zn	6.77 (6.84)	6.16 (6.13)	5.03 (4.86)	5.32 (5.16)	4.98 (4.79)	4.05 (3.73)	2.71 (2.26)	7.88 (7.72)	6.32 (6.09)	4.75 (4.45)	3.86 (3.62)
Y	7.06	5.22	3.72	3.77	3.77	2.25	1.20	6.07	4.43	3.56	2.75
Zr	6.73	5.46	4.18	4.29	4.26	3.48	2.85	6.14	5.69	5.25	4.76
Nb	7.45	6.41	5.84	5.85	5.87	5.17	4.71	7.38	6.85	6.58	6.23
Mo	8.18	7.23	6.78	6.80	6.81	6.29	5.79	7.61	7.46	7.31	7.03
Tc	8.61	7.97	7.60	7.64	7.68	7.23	6.51	8.03	7.74	7.59	7.69
Ru	7.43	7.08	6.33	6.80	6.49	6.02	5.24	6.24	6.27	6.30	6.54
Rh	7.13	6.24	5.57	5.83	5.50	4.77	3.74	5.60	5.59	5.83	6.10
Pd	7.14	5.98	5.00	4.74	4.37	3.69	2.88	6.17	5.97	5.44	5.32
Ag	8.36	6.92	6.40	6.09	5.82	5.56	5.58	8.46	6.92	6.20	4.87
Cd	8.53	7.91	6.35	6.91	6.79	5.36	4.09	8.46	*	5.71	4.46
La	4.80	3.14	1.55	1.64	1.58	0.1	-1.03	3.65	2.03	1.11	0.29
Hf	6.82	5.57	4.32	4.40	4.45	3.47	2.73	6.21	5.78	5.32	4.83
Ta	7.52	6.53	5.79	5.88	5.88	5.28	4.66	7.56	7.25	6.98	6.71
W	8.41	7.66	7.16	7.15	7.14	6.74	6.31	8.23	8.19	8.13	7.95
Re	8.16	7.65	7.32	7.34	7.40	7.08	6.61	7.95	7.90	7.85	8.22
Os	7.91	7.66	7.11	7.47	7.22	6.81	6.15	7.24	7.41	7.78	8.24
Ir	7.03	6.22	5.67	5.78	5.65	4.95	4.05	5.96	6.23	6.89	7.54
Pt	6.55	5.55	4.84	4.49	4.20	3.68	2.94	6.21	6.41	6.48	6.86
Au	7.02	5.80	5.47	5.08	4.92	4.72	4.99	8.38	7.11	6.17	5.87
Hg	8.12	7.77	7.05	*	7.41	*	*	*	*	*	*

"*" denotes unstable systems.

Table S6. Embedding energy [$E_{emb} = E_{M@G^d} - (E_{G^d} + E_M)$ in eV] for configurations of (TM)SA embedded in M-N_nC_m-G_N (G^d denotes graphene with defect). (RPBE(U)+TS results are in parentheses).

Transition Metal	C_4	N_1C_3	N_2C_2	N_2C_2[b]	N_2C_2[c]	N_3C_1	N_4	C_3	N_1C_2	N_2C_1	N_3
Sc	-5.97	-7.78	-9.01	-8.32	-7.30	-8.65	-7.92	-6.52	-5.88	-6.40	-5.82
	(-5.26)	(-7.03)	(-8.23)	(-7.56)	(-6.53)	(-7.86)	(-7.12)	(-5.67)	(-5.04)	(-5.59)	(-5.07)
Ti	-7.73	-8.87	-9.87	-9.13	-8.16	-8.87	-7.98	-8.09	-6.37	-6.68	-6.04
	(-6.30)	(-7.40)	(-8.39)	(-7.68)	(-6.73)	(-7.66)	(-6.77)	(-6.59)	(-4.97)	(-5.30)	(-4.80)
V	-7.13	-8.01	-8.84	-8.09	-7.20	-8.02	-7.31	-7.34	-5.89	-6.21	-5.56
	(-5.67)	(-6.65)	(-7.65)	(-6.93)	(-6.04)	(-6.97)	(-6.48)	(-5.80)	(-4.57)	(-5.09)	(-4.72)
Cr	-5.52	-6.42	-7.37	-6.48	-5.63	-6.68	-6.28	-6.13	-4.51	-4.45	-3.41
	(-4.08)	(-5.27)	(-6.25)	(-5.43)	(-4.57)	(-5.79)	(-5.55)	(-4.62)	(-3.30)	(-3.46)	(-2.78)
Mn	-5.03	-6.70	-7.45	-6.92	-6.07	-6.79	-6.03	-6.04	-3.95	-4.11	-3.62
	(-3.88)	(-5.57)	(-6.28)	(-5.78)	(-4.91)	(-5.65)	(-4.65)	(-3.38)	(-1.88)	(-3.30)	(-3.13)
Fe	-6.10	-7.15	-7.79	-7.33	-6.58	-7.22	-6.57	-6.92	-5.03	-4.85	-4.03
	(-4.79)	(-6.13)	(-6.76)	(-6.29)	(-5.51)	(-6.03)	(-5.48)	(-4.88)	(-3.28)	(-3.71)	(-3.36)
Co	-6.46	-7.32	-8.18	-7.71	-7.01	-7.74	-7.06	-7.46	-5.42	-5.34	-4.45
	(-5.11)	(-6.42)	(-7.02)	(-6.54)	(-5.80)	(-6.62)	(-5.99)	(-5.77)	(-3.80)	(-4.18)	(-3.58)
Ni	-6.33	-7.69	-8.54	-8.08	-7.46	-7.98	-7.05	-6.55	-4.70	-4.75	-4.00
	(-5.06)	(-6.40)	(-7.51)	(-7.02)	(-6.40)	(-7.04)	(-6.22)	(-5.60)	(-3.95)	(-4.24)	(-3.52)
Cu	-5.14	-6.36	-6.57	-6.21	-5.52	-5.58	-4.62	-3.55	-2.53	-3.03	-2.75
	(-4.60)	(-5.87)	(-6.09)	(-5.74)	(-5.06)	(-5.17)	(-4.46)	(-3.17)	(-2.27)	(-2.83)	(-2.67)
Zn	-2.74	-3.22	-4.11	-3.19	-2.49	-3.29	-2.90	-1.11	-0.38	-1.64	-1.25
	(-2.66)	(-3.25)	(-4.27)	(-3.34)	(-2.67)	(-3.61)	(-3.34)	(-1.26)	(-0.60)	(-1.93)	(-1.48)
Y	-5.85	-7.57	-8.83	-8.15	-7.11	-8.50	-7.82	-6.33	-5.68	-6.24	-5.77
Zr	-8.53	-9.68	-10.70	-9.98	-8.96	-9.61	-8.51	-8.60	-6.76	-6.89	-6.11
Nb	-8.71	-9.62	-9.94	-9.31	-8.25	-8.83	-7.55	-8.26	-6.49	-6.47	-5.53
Mo	-7.24	-8.07	-8.28	-7.64	-6.58	-6.98	-5.74	-7.30	-5.16	-5.01	-4.00
Tc	-8.20	-8.71	-8.84	-8.17	-7.09	-7.42	-6.40	-8.26	-6.25	-6.10	-4.72
Ru	-7.98	-8.21	-8.72	-7.62	-6.89	-7.23	-6.28	-8.66	-6.33	-5.99	-4.48
Rh	-7.06	-7.82	-8.24	-7.36	-6.65	-7.25	-6.56	-8.07	-5.79	-5.25	-3.69
Pd	-4.62	-5.65	-6.39	-6.02	-5.35	-5.91	-4.98	-5.07	-2.98	-3.21	-2.04
Ag	-2.46	-3.78	-4.05	-3.74	-2.96	-3.10	-1.34	-1.85	-1.09	-1.51	-1.55
Cd	-0.56	-1.06	-2.37	-1.18	-0.26	-1.57	-1.10	-0.11	*	-0.26	-0.24
La	-7.78	-9.31	-10.65	-9.94	-8.95	-10.31	-9.70	-8.40	-7.73	-8.35	-7.88
Hf	-8.53	-9.65	-10.66	-9.96	-8.87	-9.72	-8.73	-8.63	-6.76	-6.92	-6.12
Ta	-9.47	-10.34	-10.83	-10.11	-9.07	-9.55	-8.43	-8.92	-6.93	-6.90	-5.89
W	-9.21	-9.84	-10.09	-9.47	-8.44	-8.72	-7.41	-8.88	-6.62	-6.38	-5.27
Re	-8.28	-8.66	-8.75	-8.10	-7.00	-7.20	-5.93	-7.97	-5.73	-5.48	-3.83
Os	-9.10	-9.23	-9.53	-8.54	-7.76	-8.04	-6.96	-9.26	-6.79	-6.12	-4.38
Ir	-8.47	-9.15	-9.46	-8.71	-7.81	-8.38	-7.54	-9.02	-6.45	-5.48	-3.56
Pt	-7.14	-8.02	-8.48	-8.21	-7.45	-7.85	-6.85	-6.97	-4.46	-4.10	-2.44
Au	-4.18	-5.27	-5.37	-5.13	-4.24	-4.33	-2.32	-2.31	-1.28	-1.92	-0.94
Hg	-0.48	-0.70	-1.18	*	0.85	*	*	*	*	*	*

"*" denotes unstable systems. It should be noted that among all possible M-N_nC_m-G_N configurations, the cases of M-N_2C_2-G_N are particularly stable with the largest negative embedding energies; thus, the N_2C_2 moiety can be most easily made.

Table S7. Stability of (TM)SA-embedding over metal-cohesion [$E_{emb/coh} = E_{M@G^d} - (E_{G^d} + \mu_M) = E_{emb} - E_{-coh}$ in eV] for M-N_nC_m-G_N {see Figure 3}. (RPBE(U)+TS results are in parentheses).

Transition Metal	C_4	N_1C_3	N_2C_2	N_2C_2[b]	N_2C_2[c]	N_3C_1	N_4	C_3	N_1C_2	N_2C_1	N_3
Sc	-1.32 (-1.24)	-3.14 (-3.02)	-4.36 (-4.22)	-3.67 (-3.55)	-2.65 (-2.52)	-4.00 (-3.85)	-3.27 (-3.11)	-1.87 (-1.66)	-1.24 (-1.03)	-1.76 (-1.58)	-1.18 (-1.06)
Ti	-1.72 (-1.84)	-2.87 (-2.94)	-3.86 (-3.93)	-3.13 (-3.22)	-2.16 (-2.27)	-2.87 (-3.20)	-1.97 (-2.31)	-2.08 (-2.13)	-0.36 (-0.51)	-0.67 (-0.84)	-0.03 (-0.34)
V	-1.11 (-1.74)	-1.99 (-2.72)	-2.81 (-3.72)	-2.07 (-3.01)	-1.18 (-2.11)	-2.00 (-3.04)	-1.29 (-2.55)	-1.32 (-1.87)	0.13 (-0.64)	-0.19 (-1.16)	0.46 (-0.79)
Cr	-0.70 (-1.81)	-1.61 (-3.00)	-2.56 (-3.98)	-1.67 (-3.16)	-0.81 (-2.31)	-1.87 (-3.52)	-1.47 (-3.28)	-1.32 (-2.35)	0.30 (-1.03)	0.36 (-1.20)	1.40 (-0.52)
Mn	-0.58 (-2.75)	-2.25 (-4.44)	-3.00 (-5.15)	-2.47 (-4.65)	-1.63 (-3.79)	-2.34 (-4.52)	-1.58 (-3.52)	-1.59 (-2.26)	0.50 (-0.76)	0.34 (-2.17)	0.82 (-2.01)
Fe	-0.97 (-1.44)	-2.01 (-2.78)	-2.66 (-3.41)	-2.20 (-2.94)	-1.45 (-2.16)	-2.09 (-2.68)	-1.44 (-2.14)	-1.78 (-1.53)	0.10 (0.07)	0.28 (-0.36)	1.10 (-0.01)
Co	-1.08 (-1.57)	-1.94 (-2.88)	-2.80 (-3.48)	-2.33 (-3.00)	-1.63 (-2.26)	-2.36 (-3.08)	-1.68 (-2.45)	-2.08 (-2.23)	-0.04 (-0.26)	0.04 (-0.64)	0.93 (-0.04)
Ni	-1.16 (-1.15)	-2.51 (-2.48)	-3.37 (-3.60)	-2.91 (-3.10)	-2.29 (-2.49)	-2.80 (-3.12)	-1.87 (-2.31)	-1.37 (-1.69)	0.47 (-0.04)	0.42 (-0.33)	1.17 (0.39)
Cu	-1.55 (-1.42)	-2.77 (-2.69)	-2.98 (-2.91)	-2.62 (-2.56)	-1.93 (-1.88)	-1.99 (-1.99)	-1.03 (-1.28)	0.04 (0.01)	1.06 (0.91)	0.56 (0.35)	0.84 (0.51)
Zn	-1.62 (-1.55)	-2.10 (-2.14)	-2.99 (-3.16)	-2.07 (-2.24)	-1.37 (-1.56)	-2.18 (-2.50)	-1.78 (-2.24)	0.01 (-0.16)	0.74 (0.51)	-0.52 (-0.83)	-0.14 (-0.37)
Y	-1.33	-3.05	-4.30	-3.63	-2.59	-3.97	-3.30	-1.80	-1.15	-1.71	-1.25
Zr	-1.66	-2.81	-3.84	-3.11	-2.09	-2.75	-1.64	-1.74	0.11	-0.02	0.76
Nb	-0.94	-1.86	-2.18	-1.54	-0.48	-1.06	0.21	-0.50	1.27	1.30	2.23
Mo	-0.21	-1.04	-1.24	-0.60	0.46	0.06	1.29	-0.27	1.88	2.03	3.03
Tc	0.22	-0.30	-0.42	0.24	1.33	1.00	2.02	0.15	2.16	2.31	3.69
Ru	-0.96	-1.19	-1.69	-0.60	0.13	-0.21	0.74	-1.63	0.69	1.03	2.54
Rh	-1.26	-2.02	-2.45	-1.56	-0.85	-1.45	-0.76	-2.28	0.01	0.55	2.10
Pd	-1.26	-2.29	-3.02	-2.66	-1.98	-2.54	-1.62	-1.71	0.39	0.16	1.33
Ag	-0.03	-1.35	-1.62	-1.31	-0.53	-0.67	1.08	0.58	1.34	0.92	0.88
Cd	0.14	-0.36	-1.67	-0.49	0.43	-0.87	-0.40	0.58	*	0.44	0.46
La	-3.60	-5.13	-6.47	-5.76	-4.77	-6.13	-5.52	-4.22	-3.55	-4.17	-3.70
Hf	-1.57	-2.69	-3.71	-3.00	-1.91	-2.76	-1.77	-1.67	0.20	0.04	0.84
Ta	-0.87	-1.73	-2.23	-1.51	-0.47	-0.95	0.17	-0.32	1.67	1.70	2.71
W	0.02	-0.61	-0.86	-0.24	0.79	0.51	1.81	0.35	2.61	2.85	3.96
Re	-0.23	-0.61	-0.70	-0.06	1.04	0.85	2.12	0.08	2.31	2.57	4.22
Os	-0.48	-0.61	-0.91	0.08	0.86	0.58	1.66	-0.64	1.83	2.50	4.24
Ir	-1.37	-2.05	-2.35	-1.61	-0.71	-1.28	-0.44	-1.92	0.65	1.62	3.54
Pt	-1.85	-2.72	-3.19	-2.91	-2.15	-2.55	-1.55	-1.67	0.83	1.20	2.86
Au	-1.37	-2.46	-2.56	-2.31	-1.43	-1.51	0.50	0.51	1.53	0.89	1.88
Hg	-0.28	-0.49	-0.97	*	1.06	*	*	*	*	*	*

For covenience's sake (because the cohesive energy is oppositely signed with respect to the formation energy), we define $E_{-coh} \equiv -E_{coh}$ for our further discussion. TM-SAs on di-vacancy sites ($N_{n=0\text{-}4}C_{4\text{-}n}$-$G_N$), in particular, in $N_2C_2G_N$ have strongly negative $E_{emb/coh}$, indicating easy formation of SAs over NPs, whereas those on mono-vacancy sites ($N_{n=1\text{-}3}C_{3\text{-}n}$-$G_N$) show instability of SAs over NPs with mostly positive $E_{emb/coh}$, indicating easy formation of NPs over SAs, and those on C_3-G_N show weakly negative $E_{emb/coh}$, indicating some mixtures of SAs and NPs with more SAs than NPs. "*" denotes unstable systems. The red-marked positive values indicate that the SA formation is not favored, but even adsorbed SAs can be readily desorbed which results in NPs aggregation.

Table S8. Off-plane distances (Å) (before slash) of a (TM)SA embedded M-N_nC_m-G_N graphene systems before/after H adsorption for the global minima configurations.

Transition Metal	C_4	N_1C_3	N_2C_2	N_2C_2[b]	N_2C_2[c]	N_3C_1	N_4	C_3	N_1C_2	N_2C_1	N_3
Sc	1.28/1.16	1.11/1.25	1.10/1.05	1.13/1.22	1.12/1.28	1.00/1.15	0.90/1.05	1.94/2.11	1.82/2.17	1.71/1.90	1.67/1.85
Ti	0.93/0.93	0.87/0.96	0.80/0.86	0.88/0.98	0.83/0.99	0.76/0.82	0.68/0.83	1.64/1.98	1.63/1.85	1.61/1.66	1.59/1.71
V	0.78/0.77	0.69/0.61	0.67/0.64	0.82/0.86	0.71/0.85	0.58/0.83	0.65/0.69	1.58/1.72	1.60/1.61	1.57/1.61	1.53/1.63
Cr	0.79/0.72	0.74/0.56	0.66/0.72	0.72/0.80	0.68/0.83	0.54/0.32	0/0.51	1.55/1.72	1.52/1.61	1.44/1.51	1.55/1.54
Mn	0/0.53	0.59/0.43	0.51/0.41	0.57/0.63	0.52/0.44	0/0	0/0	1.45/1.62	1.58/1.42	1.61/1.67	1.69/1.92
Fe	0.63/0	0/0.29	0/0.27	0.20/0.35	0/0.27	0/0.28	0/0	1.38/1.54	1.34/1.37	1.42/1.65	1.51/1.68
Co	0.64/0.49	0/0	0/0	0.31/0.28	0/0	0/0	0/0	1.34/1.48	1.28/1.41	1.34/1.53	1.41/1.63
Ni	0/0	0/0	0/0	0.17/0.24	0/0	0/0	0/0	1.35/1.56	1.37/1.42	1.44/1.59	1.41/1.61
Cu	0/0	0/0	0/0	0.16/0.38	0/0	0/0	0/0.27	1.52/1.80	1.48/1.67	1.56/1.54	1.60/1.81
Zn	0/0.30	0/0.67	0.55/0.55	0.16/1.13	0/0.58	0.45/0.33	0/1.14	1.69/1.99	1.65/1.76	1.57/1.87	1.72/1.82
Y	1.53/1.54	1.40/1.59	1.41/1.37	1.41/1.52	1.41/1.59	1.32/1.67	1.18/1.50	2.15/2.29	2.03/2.22	1.92/2.11	1.89/2.13
Zr	1.24/1.28	1.17/1.40	1.07/1.31	1.13/1.24	1.09/1.38	1.01/1.23	0.97/1.08	1.90/2.26	1.83/2.06	1.82/1.99	1.82/1.88
Nb	0.99/1.10	0.90/1.05	0.86/0.95	0.92/0.95	0.90/1.00	0.80/0.90	0.99/1.09	1.82/2.00	1.82/1.83	1.78/1.97	1.75/1.77
Mo	0.89/0.88	0.82/1.04	0.93/0.90	0.84/1.04	0.95/1.07	0.91/0.92	0.84/0.97	1.77/1.98	1.74/1.81	1.70/1.85	1.65/1.78
Tc	0.91/0.90	0.69/0.99	0.87/0.84	0.74/0.98	0.91/1.09	0.80/0.84	0/0.60	1.70/1.87	1.65/1.75	1.62/1.72	1.58/1.68
Ru	0.91/0.95	0.89/0.99	0.79/0.82	0.93/0.93	0.90/1.04	0.74/0.71	0/0.44	1.65/1.87	1.61/1.65	1.58/1.61	1.54/1.78
Rh	0.85/0.97	0.79/0.93	0.67/0.69	0.74/0.65	0.71/0.88	0/0.37	0/0	1.62/1.74	1.57/1.73	1.62/1.74	1.70/1.87
Pd	0.77/0.62	0.65/0	0/0	0.44/0.55	0/0	0/0.26	0/0	1.61/1.88	1.71/1.74	1.75/1.92	1.85/1.92
Ag	0/0.29	0/0	0/0	0.38/0.70	0/0	0/0	0/1.62	2.15/2.02	2.13/2.05	2.10/2.03	2.10/2.18
Cd	0/1.63	0/1.90	1.29/1.84	1.35/1.70	1.29/1.55	1.26/1.28	1.31/1.67	2.22/2.32	*	2.37/2.34	2.30/2.13
La	1.71/1.76	1.67/1.77	1.64/1.69	1.68/1.24	1.64/1.84	1.57/1.96	1.44/1.82	2.31/2.53	2.20/2.42	2.05/2.29	1.67/2.28
Hf	1.23/1.48	1.16/1.40	1.04/1.27	1.09/1.21	1.06/1.29	1.02/1.23	0.97/1.08	1.87/2.16	1.79/1.96	1.77/1.89	1.75/1.82
Ta	1.01/1.14	0.89/1.15	0.89/0.96	0.92/0.99	0.92/1.01	0.74/0.82	0.67/0.71	1.80/1.88	1.80/1.83	1.75/1.86	1.70/1.89
W	0.86/0.85	0.77/0.93	0.72/0.76	0.79/1.06	0.73/0.79	0.91/0.93	0.83/0.85	1.77/1.97	1.74/1.91	1.69/1.80	1.63/1.70
Re	0.92/0.92	0.65/1.07	0.82/0.83	0.68/1.04	0.91/0.93	0.81/0.93	0/0.76	1.71/1.78	1.67/1.74	1.64/1.78	1.60/1.72
Os	0.95/0.94	0.90/1.04	0.81/0.96	0.92/0.97	0.89/0.93	0.74/0.90	0/0.66	1.70/1.76	1.65/1.72	1.61/1.74	1.56/1.71
Ir	0.89/0.88	0.81/1.01	0.69/0.69	0.70/0.80	0.68/0.77	0/0.72	0/0.36	1.65/1.89	1.62/1.78	1.62/1.76	1.66/1.86
Pt	0.76/0.61	0.62/0	0/0.38	0.45/0.62	0/0	0/0.32	0/0	1.65/1.82	1.79/1.79	1.73/1.89	1.79/1.94
Au	0/0	0/0	0/0	0.38/0.67	0/0.33	0/0	0/0	1.87/1.87	1.99/2.07	2.35/2.05	2.13/2.29
Hg	0/0	0/0	1.38/2.09	*	0.23/177	*	*	*	*	*	*

"*" denotes unstable systems.

Table S9. Metal-to-H bond distances (Å) upon the H adsorption (For di-vacancy sites, the M-H bond distances are given here only for the global minima configurations).

Transition Metal	C_4	N_1C_3	N_2C_2	N_2C_2[b]	N_2C_2[c]	N_3C_1	N_4	C_3	N_1C_2	N_2C_1	N_3
Sc	2.05	2.06	2.11	2.03	2.16	1.88	1.87	1.91	1.92	1.90	1.89
Ti	2.03	1.76	1.75	1.75	1.75	1.75	1.75	1.80	1.79	1.78	1.79
V	1.67	1.68	1.69	1.69	1.68	1.70	1.68	1.72	1.72	1.73	1.72
Cr	1.92	2.16	1.63	1.94	1.81	2.09	1.61	1.67	1.67	1.68	1.68
Mn	2.15	2.13	1.85	1.56	2.23	2.22	1.53	1.63	1.61	1.64	1.66
Fe	1.84	2.02	1.97	1.72	2.08	1.72	1.50	1.58	1.57	1.59	1.61
Co	1.74	1.87	1.79	1.67	1.44	1.63	1.45	1.56	1.51	1.54	1.57
Ni	1.82	1.79	1.74	1.72	1.74	1.87	2.20	1.56	1.47	1.48	1.54
Cu	2.03	1.98	1.97	1.88	2.17	2.04	2.18	1.53	1.54	1.49	1.51
Zn	2.19	2.23	2.12	1.92	2.19	2.08	1.56	1.54	1.53	1.54	1.53
Y	2.14	2.14	2.24	2.21	2.33	2.05	2.04	2.06	2.07	2.08	2.07
Zr	1.92	1.91	1.91	1.91	1.91	1.91	1.90	1.97	1.95	1.94	1.94
Nb	1.80	1.82	1.82	1.82	1.82	1.82	1.83	1.87	1.86	1.86	1.85
Mo	1.74	1.76	1.76	1.76	1.76	1.75	1.72	1.80	1.79	1.78	1.77
Tc	1.72	1.68	1.69	1.70	1.66	1.67	1.63	1.75	1.73	1.72	1.71
Ru	1.66	1.62	1.63	1.62	1.61	1.61	1.58	1.70	1.69	1.70	1.67
Rh	1.82	1.82	1.76	1.55	1.54	1.54	1.53	1.69	1.62	1.65	1.65
Pd	2.30	2.12	2.07	1.92	2.20	2.31	2.43	1.70	1.58	1.59	1.57
Ag	2.24	2.24	2.33	1.97	2.38	2.48	1.62	1.66	1.67	1.63	1.62
Cd	1.70	1.69	2.74	2.41	1.70	1.70	1.70	1.69	*	1.69	1.68
La	2.21	2.21	2.42	2.09	2.50	2.21	2.20	2.17	2.16	2.18	2.15
Hf	1.90	1.89	1.89	1.89	1.89	1.89	1.88	1.95	1.94	1.87	1.92
Ta	1.80	1.81	1.82	1.81	1.81	1.81	1.80	1.87	1.80	1.85	1.85
W	1.75	1.76	1.76	1.76	1.75	1.75	1.73	1.81	1.79	1.79	1.78
Re	1.72	1.70	1.70	1.71	1.68	1.69	1.65	1.76	1.74	1.73	1.72
Os	1.70	1.65	1.83	1.64	1.63	1.63	1.61	1.71	1.70	1.70	1.69
Ir	1.64	1.59	1.62	1.59	1.57	1.57	1.56	1.69	1.64	1.65	1.64
Pt	1.82	2.33	1.85	1.79	2.42	2.45	2.47	1.69	1.59	1.59	1.59
Au	2.40	2.44	2.48	1.96	2.48	2.47	2.49	1.64	1.64	1.59	1.57
Hg	2.50	2.45	1.65	*	1.64	*	*	*	*	*	*

"*****" denotes unstable systems.

Table S10. Off-plane distances (Å) of a (TM)SA embedded M-N_nC_m-G_N graphene systems before/after the *O adsorption ("prist/‡") as OER/ORR reaction intermediates and the bond distances (Å) from metal to *O , *OH and *OOH.

TM	C_4				N_1C_3				N_2C_2				N_2C_2[b]			
	prist/‡	*O	*OH	*OOH	prist/‡	*O	*OH	*OOH	prist/‡	*O	*OH	*OOH	prist/‡	*O	*OH	*OOH
Sc	1.28/1.62	1.73	1.90	2.02	1.13/1.59	1.75	1.91	2.03	1.10/1.35	1.86	1.93	2.04	1.13/1.40	1.80	1.91	2.03
Ti	0.93/1.09	1.67	1.85	1.92	0.87/1.11	1.66	1.85	1.92	0.80/1.10	1.66	1.84	1.93	0.88/1.12	1.66	1.84	1.92
V	0.78/0.86	1.61	1.80	1.83	0.72/0.83	1.61	1.80	1.86	0.67/0.81	1.61	1.80	1.83	0.82/0.81	**1.82**	**1.81**	**1.82**
Cr	0.79/0.79	1.58	1.77	1.78	0.74/0.85	1.58	1.78	1.74	0.66/0.81	1.59	1.79	1.74	0.72/0.86	**1.83**	**1.82**	**1.83**
Mn	0/0.66	1.58	1.78	1.74	0.59/0.78	1.58	1.82	1.82	0.51/0.76	1.57	1.83	1.83	0.57/0.82	1.57	1.83	1.84
Fe	0.63/0.59	1.57	1.83	1.83	0.04/0.78	1.59	1.85	1.79	0/0.64	1.61	1.80	1.64	0.20/0.77	1.61	1.84	1.86
Co	0.64/0.64	1.61	1.77	1.73	0.01/0.70	1.64	1.79	1.75	0/0.64	1.65	1.84	1.77	0.31/0.66	1.65	1.83	1.82
Ni	0/0.57	1.69	1.84	1.83	0.01/0.65	1.72	1.85	1.86	0/0.58	1.74	1.90	1.93	0.17/0.61	1.75	1.88	1.88
Cu	0/0.55	1.83	1.91	2.08	0.02/0.57	1.88	1.92	1.90	0/0.39	1.92	2.01	2.13	0.16/0.51	1.92	1.91	1.91
Zn	0/0.61	1.95	1.96	2.12	0.12/0.78	1.90	1.93	1.97	0.55/0.89	1.88	1.92	2.04	0.16/1.08	1.88	1.89	1.94
Y	1.53/1.94	1.86	2.05	2.18	1.42/1.88	1.88	2.06	2.19	1.41/1.62	1.94	2.07	2.20	1.41/1.64	1.94	2.06	2.19
Zr	1.24/1.46	1.81	2.00	2.09	1.17/1.54	1.81	1.99	2.08	1.07/1.43	1.80	1.99	2.10	1.13/1.36	1.80	1.97	2.08
Nb	0.99/1.24	1.74	1.94	1.99	0.90/1.24	1.74	1.94	2.01	0.86/1.06	1.74	1.94	2.01	0.92/1.04	1.74	1.91	1.98
Mo	0.89/0.87	1.70	1.91	1.92	0.86/0.95	1.72	1.91	1.92	0.93/1.08	**1.96**	**2.69**	**2.69**	0.84/1.10	**1.96**	**1.92**	**1.95**
Tc	0.91/0.91	1.69	1.90	1.86	0.75/1.05	**1.92**	**1.91**	**1.97**	0.87/1.02	**1.97**	**2.69**	**2.75**	0.74/1.06	**1.96**	**1.91**	**1.93**
Ru	0.91/0.89	1.69	1.89	1.81	0.89/0.98	1.70	1.90	1.77	0.79/0.93	**2.22**	**2.41**	**2.59**	0.93/0.99	**1.93**	**1.94**	**1.94**
Rh	0.85/0.88	1.75	1.91	1.90	0.79/0.97	1.76	1.97	1.91	0.67/0.82	1.79	1.96	1.94	0.74/0.97	1.78	1.96	1.93
Pd	0.77/0.88	1.82	2.01	2.08	0.65/0.92	1.88	1.98	1.99	0/0.80	1.92	2.08	2.12	0.44/0.83	1.91	1.94	1.95
Ag	0/0.89	1.98	2.07	2.13	0.01/1.00	2.03	2.03	1.98	0/0.69	2.14	2.20	2.59	0.38/1.34	1.98	2.05	1.97
Cd	0/1.33	2.06	2.07	2.29	0.02/1.99	2.04	2.04	2.07	1.29/1.70	2.05	2.05	2.10	1.35/1.64	2.03	2.07	2.12
La	1.71/2.39	1.85	2.17	2.30	1.68/1.88	1.88	2.17	2.31	1.64/2.09	1.90	2.20	2.33	1.68/1.89	1.89	2.18	2.31
Hf	1.23/1.38	1.82	1.95	2.06	1.16/1.43	1.82	1.96	2.07	1.04/1.36	1.81	1.95	2.07	1.09/1.25	1.81	1.94	2.06
Ta	1.01/1.24	1.77	1.92	1.96	0.90/1.24	1.77	1.94	2.01	0.89/1.10	**1.96**	**2.33**	**2.40**	0.92/1.05	**1.95**	**1.91**	**1.93**
W	0.86/0.78	1.74	1.92	1.99	0.82/0.95	1.75	1.91	1.91	0.72/1.08	**1.95**	**2.78**	**2.78**	0.79/1.11	**1.92**	**1.93**	**1.91**
Re	0.92/0.98	**1.96**	**2.67**	**2.69**	0.72/1.03	**1.93**	**1.92**	**1.98**	0.82/1.02	**1.97**	**2.71**	**2.77**	0.68/1.09	**1.96**	**1.93**	**1.95**
Os	0.95/0.95	**2.01**	**2.63**	**2.73**	0.91/1.01	**1.97**	**1.94**	**1.97**	0.81/0.96	**2.19**	**2.46**	**2.56**	0.92/1.02	**1.95**	**1.94**	**1.94**
Ir	0.89/0.91	1.76	1.95	1.88	0.81/0.98	1.77	1.98	1.85	0.69/0.85	1.80	1.99	1.90	0.70/0.97	1.79	1.97	1.84
Pt	0.76/0.90	1.82	2.02	2.02	0.62/0.93	1.86	1.98	2.06	0/0.71	1.87	2.07	2.12	0.45/0.84	1.89	1.94	1.95
Au	0/0.89	1.96	2.87	3.25	0.01/1.25	1.87	1.98	3.13	0/0.54	2.19	2.28	3.14	0.38/1.48	1.91	1.97	3.38
Hg	0/0.63	2.32	2.32	2.69	0.01/1.43	2.02	2.08	2.23	1.38/1.99	2.02	2.05	2.08	*	*	*	*

Table S10. Continued...

TM	N_2C_2[c]				N_3C_1				N_4			
	prist/‡	***O**	***OH**	***OOH**	**prist/‡**	***O**	***OH**	***OOH**	**prist/‡**	***O**	***OH**	***OOH**
Sc	1.12/1.49	1.75	1.92	2.03	1.00/1.28	1.77	1.91	2.03	0.90/1.28	1.76	1.90	2.02
Ti	0.83/1.13	1.66	1.83	1.89	0.76/0.99	1.66	1.83	1.88	0.68/0.93	1.65	1.82	1.86
V	0.74/0.81	**1.83**	**1.83**	**1.81**	0.61/0.78	1.61	1.79	1.78	0.65/0.84	**1.88**	**1.88**	**1.92**
Cr	0.68/0.90	**1.84**	**1.83**	**1.83**	0.54/0.76	1.59	1.81	1.77	0/0.74	1.58	1.86	1.85
Mn	0.52/0.85	1.57	1.86	1.87	0.02/0.74	1.58	1.86	1.89	0/0.79	1.60	1.90	1.93
Fe	0.03/0.83	**1.83**	**1.87**	**1.77**	0.02/0.64	1.63	1.84	1.67	0/0.58	1.66	1.84	1.79
Co	0.22/0.66	1.66	1.83	1.82	0.02/0.57	1.68	1.85	1.83	0/0.75	1.66	1.89	1.92
Ni	0.04/0.71	1.74	1.84	1.85	0.02/0.44	1.80	1.85	1.85	0/0.23	1.95	2.04	2.23
Cu	0.01/0.62	1.89	1.88	1.88	0.02/0.80	1.82	1.89	1.89	0/1.20	1.76	1.97	2.15
Zn	0.02/0.56	1.94	1.89	1.93	0.45/1.14	1.86	1.87	1.95	0/1.01	1.87	1.89	1.94
Y	1.41/1.86	1.86	2.06	2.19	1.32/1.58	1.93	2.06	2.19	1.18/1.56	1.91	2.06	2.18
Zr	1.09/1.59	1.80	1.98	2.08	1.01/1.46	1.80	1.97	2.09	0.97/1.25	1.79	1.95	2.07
Nb	0.90/1.24	**1.93**	**1.95**	**2.03**	0.85/1.04	**1.96**	**1.94**	**1.98**	0.99/1.07	**1.75**	**1.98**	**2.04**
Mo	0.95/1.13	**1.97**	**1.92**	**1.94**	0.91/1.00	**1.96**	**1.92**	**1.96**	0.84/1.04	**2.02**	**2.05**	**2.00**
Tc	0.91/1.09	**1.98**	**1.94**	**1.95**	0.80/0.82	**1.96**	**1.91**	**1.97**	0/0.91	**2.09**	**2.14**	**2.02**
Ru	0.90/1.01	**1.97**	**1.95**	**1.94**	0.74/0.85	**2.02**	**1.96**	**1.92**	0/0.67	**2.06**	**2.06**	**1.94**
Rh	0.71/0.97	1.78	1.96	1.92	0.03/0.75	1.81	1.92	1.91	0/0.38	1.85	2.01	2.03
Pd	0.05/0.92	1.91	1.91	1.91	0.03/0.53	1.92	1.91	1.91	0/0.17	2.24	2.29	3.18
Ag	0.03/1.26	1.98	2.03	1.95	0.02/1.45	1.90	1.95	1.94	0/1.73	1.99	2.06	2.11
Cd	0.01/1.61	2.01	2.07	2.10	1.26/1.14	2.06	2.06	2.11	1.31/1.73	2.06	2.08	2.12
La	1.64/1.87	1.87	2.18	2.30	1.57/1.90	1.90	2.19	2.31	1.44/2.04	1.89	2.18	2.31
Hf	1.06/1.53	1.81	1.95	2.07	1.02/1.35	1.80	1.95	2.07	0.97/1.17	1.80	1.93	2.05
Ta	0.92/1.25	1.77	1.92	1.78	0.78/1.00	**1.94**	**1.94**	**2.04**	0.67/0.87	**1.78**	**1.98**	**2.05**
W	0.77/0.92	**1.93**	**1.93**	**1.99**	0.91/1.01	**0.97**	**1.92**	**1.95**	0.83/0.98	**1.75**	**1.97**	**1.98**
Re	0.91/1.10	**1.99**	**1.94**	**1.94**	0.81/0.98	**1.98**	**1.92**	**1.99**	0/0.93	**2.08**	**2.11**	**1.92**
Os	0.89/1.04	**2.07**	**1.95**	**1.93**	0.74/0.89	**2.08**	**1.96**	**1.90**	0/0.80	**2.08**	**2.06**	**1.95**
Ir	0.68/0.99	**2.04**	**2.00**	**1.99**	0.02/0.77	1.82	1.93	1.93	0/0.74	1.85	2.01	2.02
Pt	0.04/0.90	1.89	1.91	2.27	0.02/0.49	1.92	1.91	2.21	0/0.08	2.00	2.31	3.45
Au	0.04/1.43	1.90	1.94	2.87	0.02/0.20	1.94	1.94	2.96	0/0.14	2.61	2.49	2.94
Hg	0.03/1.72	1.91	2.05	2.08	*	*	*	*	*	*	*	*

"**prist/‡**" represents "the prstine state without O-adsorption / the O*-adsorbed intermediate state" of the embedded (TM)SA (The values in blue follows path-II shown in Figure 6).

Table S11. Clinging energies (E_{2M} in eV) of 2nd (TM)SA attached to 1st (TM)SA in SAC [$E_{2M} = E_{M_2G^d} - (E_{MG^d} + E_M)$] for M-$N_nC_m$-$G_N$. (RPBE(U)+TS results are in parentheses).[a]

Transition Metal	C_4	N_1C_3	N_2C_2	N_2C_2[b]	N_2C_2[c]	N_3C_1	N_4	C_3	N_1C_2	N_2C_1	N_3
Sc	-0.50	-2.57	-0.57	-0.59	-0.64	-0.74	-1.09	-0.52	-0.62	-1.07	-1.07
	(-0.41)	(-1.80)	(-0.55)	(-0.50)	(-0.56)	(-0.66)	(-0.89)	(-0.52)	(-0.59)	(-0.47)	(-1.20)
Ti	-1.02	-3.22	-1.35	-1.39	-3.25	-2.76	-1.95	-0.95	-1.31	-1.71	-1.82
	(-0.72)	(-2.01)	(-0.92)	(-0.99)	(-2.20)	(-1.42)	(-1.00)	(-0.71)	(-1.04)	(-1.24)	(-1.29)
V	-2.91	-2.84	-2.45	-2.91	-2.27	-2.42	-1.63	-1.18	-1.28	-1.88	-1.88
	(-1.73)	(-1.64)	(-0.91)	(-1.49)	(-1.77)	(-1.24)	(-0.04)	(-0.80)	(-0.73)	(-0.64)	(-0.95)
Cr	-1.61	-1.61	-0.63	-1.52	-1.96	-0.63	-1.12	-0.61	-0.80	-0.96	-0.96
	(-0.26)	(-0.10)	(-0.33)	(-1.26)	(-0.12)	(-0.90)	(-0.93)	(-0.02)	(-0.64)	(-0.67)	(-0.83)
Mn	-2.57	-1.11	-1.16	-1.27	-1.38	-1.09	-0.87	-1.00	-1.29	-1.17	-1.17
	(-1.86)	(-1.00)	(-0.46)	(-0.77)	(-0.88)	(-0.56)	(-0.41)	(-1.46)	(-0.81)	(-0.62)	(-0.70)
Fe	-2.57	-2.48	-1.80	-2.42	-2.40	-1.97	-1.36	-1.56	-1.72	-1.78	-1.78
	(-1.88)	(-1.12)	(-0.96)	(-1.03)	(-1.06)	(-0.93)	(-0.29)	(-0.94)	(-0.55)	(-0.92)	(-1.34)
Co	-1.81	-2.85	-2.00	-2.60	-2.48	-2.06	-1.33	-1.81	-1.76	-1.81	-1.81
	(-2.18)	(-1.59)	(-1.44)	(-1.63)	(-1.59)	(-1.32)	(-0.53)	(-1.41)	(-1.38)	(-1.28)	(-1.80)
Ni	-3.12	-2.45	-1.27	-2.17	-2.21	-1.75	-0.54	-1.72	-2.38	-2.58	-2.58
	(-2.57)	(-2.19)	(-1.50)	(-1.64)	(-1.52)	(-1.35)	(-0.44)	(-0.99)	(-1.31)	(-1.46)	(-1.85)
Cu	-1.61	-1.18	-1.37	-1.20	-1.30	-1.29	-0.43	-1.41	-1.35	-1.72	-1.72
	(-1.54)	(-1.05)	(-1.23)	(-1.05)	(-1.15)	(-1.11)	(-0.32)	(-1.13)	(-1.06)	(-1.46)	(-1.08)
Zn	-0.20	-0.19	-0.17	0.00	-0.14	-0.24	-0.23	-0.56	-0.54	-0.56	-0.56
	(-0.21)	(-0.20)	(-0.17)	(-0.03)	(-0.14)	(-0.20)	(-0.17)	(-0.51)	(-0.48)	(-0.49)	(-0.66)
Y	-0.36	-0.39	-0.50	-0.49	-0.55	-0.61	-0.89	-2.54	-0.55	-0.96	-0.96
Zr	-0.88	-3.41	-1.29	-1.27	-3.34	-1.73	-2.14	-2.76	-3.03	-3.76	-3.76
Nb	-1.58	-1.75	-2.20	-2.16	-3.11	-2.38	-2.63	-3.14	-1.78	-3.53	-3.53
Mo	-2.85	-1.94	-1.74	-1.62	-1.80	-2.61	-2.74	-2.05	-2.42	-2.42	-2.42
Tc	-3.13	-3.64	-4.13	-3.63	-4.41	-4.74	-4.53	-2.54	-2.99	-3.45	-3.45
Ru	-2.41	-2.92	-2.82	-3.33	-3.21	-3.13	-2.84	-1.66	-2.41	-2.57	-2.57
Rh	-2.20	-2.76	-1.98	-2.99	-2.54	-2.72	-1.76	-2.06	-2.06	-2.25	-2.25
Pd	-0.97	-1.88	-1.66	-1.65	-1.54	-1.42	-0.58	-1.43	-1.40	-1.13	-1.13
Ag	-1.32	-0.70	-0.64	-0.50	-0.60	-0.49	-1.30	-0.97	-1.12	-1.12	-1.12
Cd	-0.44	-0.37	-0.27	-0.48	-0.50	-0.43	-0.48	-0.44	*	-0.54	-0.54
La	-0.29	-0.29	-0.30	-0.28	0.07	-2.13	-0.56	-0.32	-0.35	-1.89	-1.89
Hf	-0.96	-1.28	-1.37	-1.43	-3.28	-1.81	-2.04	-0.85	-1.62	-3.44	-3.44
Ta	-2.20	-3.80	-2.70	-2.81	-2.82	-3.04	-3.09	-2.16	-2.62	-4.06	-4.06
W	-3.23	-3.70	-3.70	-3.87	-4.30	-4.17	-4.49	-2.69	-3.35	-3.78	-3.78
Re	-2.52	-2.92	-3.55	-3.85	-3.83	-4.24	-4.38	-2.19	-2.59	-3.36	-3.36
Os	-3.67	-4.27	-4.42	-4.76	-4.66	-4.74	-4.22	-2.82	-3.64	-4.51	-4.51
Ir	-3.65	-3.41	-3.23	-3.27	-3.33	-2.82	-2.25	-3.29	-3.84	-4.40	-4.40
Pt	-2.24	-2.86	-1.55	-2.28	-1.38	-2.05	-0.97	-3.46	-3.49	-3.60	-3.60
Au	-0.80	-0.71	-0.91	-0.80	-0.96	-0.66	-0.78	-2.47	-2.55	-2.00	-2.00
Hg	-0.13	-0.13	-0.13	*	-0.13	*	*	*	*	*	*

[a] Zn, Cd, and Hg have small negative E_{2M}, compared with other metal atoms. "*" denotes unstable systems.

Table S12. Stability of clinging energies ($E_{2M/coh}$ in eV) of 2nd (TM)SA attached to 1st SAC TM (E_{2M}) over metal cohesion (E_{coh}) for M-N_nC_m-G_N, which is related to SAs' aggregation into cluster, consistent with 16 vs 18 electron rules. (RPBE(U)+TS results are in parentheses).[a]

Transition Metal	C_4	N_1C_3	N_2C_2	N_2C_2[b]	N_2C_2[c]	N_3C_1	N_4	C_3	N_1C_2	N_2C_1	N_3
Sc	4.15 (3.61)	2.08 (2.22)	4.08 (3.46)	4.06 (3.52)	4.00 (3.45)	3.90 (3.36)	3.55 (3.12)	4.12 (3.50)	4.03 (3.42)	3.57 (3.13)	3.57 (2.82)
Ti	4.99 (3.74)	2.79 (2.45)	4.65 (3.53)	4.62 (3.47)	2.76 (2.25)	3.24 (3.04)	4.06 (3.45)	5.05 (3.75)	4.69 (3.42)	4.30 (3.22)	4.18 (3.17)
V	3.11 (2.20)	3.18 (2.29)	3.57 (3.02)	3.11 (2.44)	3.75 (2.16)	3.60 (2.69)	4.39 (3.89)	4.85 (3.13)	4.75 (3.19)	4.15 (3.29)	4.15 (2.98)
Cr	3.20 (2.01)	3.21 (2.17)	4.18 (1.94)	3.29 (1.00)	2.85 (2.14)	4.18 (1.37)	3.69 (3.20)	4.20 (2.25)	4.02 (1.62)	3.85 (1.60)	3.85 (1.44)
Mn	1.87 (-0.73)	3.34 (0.12)	3.29 (0.66)	3.18 (0.35)	3.07 (0.25)	3.36 (0.57)	3.58 (0.72)	3.45 (-0.34)	3.16 (0.32)	3.28 (0.50)	3.28 (0.42)
Fe	2.56 (1.46)	2.65 (2.22)	3.33 (2.38)	2.71 (2.32)	2.73 (2.29)	3.16 (2.42)	3.78 (3.06)	3.57 (2.40)	3.42 (2.80)	3.35 (2.42)	3.35 (2.01)
Co	3.57 (1.36)	2.53 (1.95)	3.38 (2.10)	2.77 (1.91)	2.90 (1.95)	3.32 (2.22)	4.05 (3.01)	3.57 (2.13)	3.62 (2.16)	3.57 (2.26)	3.57 (1.74)
Ni	2.05 (1.35)	2.72 (1.72)	3.90 (2.41)	3.00 (2.27)	2.96 (2.39)	3.42 (2.56)	4.63 (3.47)	3.46 (2.93)	2.79 (2.61)	2.59 (2.45)	2.59 (2.06)
Cu	1.98 (1.64)	2.41 (2.13)	2.22 (1.95)	2.39 (2.13)	2.29 (2.03)	2.30 (2.07)	3.16 (2.86)	2.18 (2.05)	2.24 (2.12)	1.87 (1.72)	1.87 (2.11)
Zn	0.92 (0.90)	0.93 (0.90)	0.94 (0.94)	1.12 (1.08)	0.98 (0.96)	0.88 (0.90)	0.89 (0.93)	0.56 (0.60)	0.58 (0.63)	0.56 (0.61)	0.56 (0.45)
Y	4.16	4.13	4.02	4.04	3.98	3.92	3.64	1.98	3.97	3.57	3.57
Zr	5.98	3.45	5.58	5.60	3.53	5.13	4.72	4.11	3.84	3.10	3.10
Nb	6.18	6.02	5.56	5.60	4.66	5.38	5.13	4.62	5.99	4.23	4.23
Mo	4.19	5.09	5.30	5.41	5.24	4.43	4.30	4.99	4.61	4.62	4.62
Tc	5.84	5.33	4.83	4.78	4.56	4.22	4.44	6.42	5.98	5.51	5.51
Ru	4.61	4.10	4.20	3.69	3.81	3.90	4.18	5.36	4.61	4.45	4.45
Rh	3.59	3.04	3.82	2.81	3.26	3.07	4.03	3.74	3.73	3.54	3.54
Pd	2.39	1.48	1.70	1.71	1.83	1.94	2.79	1.93	1.96	2.24	2.24
Ag	1.11	1.72	1.79	1.93	1.83	1.94	1.13	1.45	1.31	1.31	1.31
Cd	0.26	0.33	0.43	0.21	0.20	0.27	0.22	0.25	*	0.16	0.16
La	3.89	3.89	3.88	3.90	4.25	2.05	3.62	3.86	3.83	2.29	2.29
Hf	6.00	5.68	5.58	5.53	3.68	5.15	4.92	6.11	5.34	3.52	3.52
Ta	6.40	4.80	5.90	5.79	5.79	5.56	5.51	6.44	5.98	4.54	4.54
W	6.00	5.53	5.53	5.35	4.93	5.06	4.74	6.54	5.87	5.45	5.45
Re	5.52	5.13	4.50	4.20	4.21	3.80	3.67	5.86	5.46	4.68	4.68
Os	4.95	4.35	4.20	3.86	3.96	3.88	4.40	5.80	4.98	4.11	4.11
Ir	3.45	3.69	3.87	3.83	3.77	4.28	4.85	3.81	3.26	2.70	2.70
Pt	3.06	2.44	3.75	3.02	3.92	3.25	4.32	1.84	1.81	1.69	1.69
Au	2.02	2.10	1.91	2.02	1.85	2.16	2.03	0.34	0.27	0.82	0.82
Hg	0.08	0.08	0.08	*	0.08	*	*	*	*	*	*

[a] Cd, Hg, Zn / Ag, Au, Pd have rather very-small / small $E_{2M/coh}$, which help form these NPs easily. Cu and Mn have moderate values, while others have large values (> 3eV). "*" denotes unstable systems.

Table S13. Dissolution potential U_{ds} [= U°_{dis}(bulk metal) − $E_{emb/coh}$ / en_e in eV] for configurations of (TM)SA embedded in M-N_nC_m-G_N (G^d denotes graphene with defect). (RPBE(U)+TS results are in parentheses).

Transition Metal	C_4	N_1C_3	N_2C_2	N_2C_2[b]	N_2C_2[c]	N_3C_1	N_4	C_3	N_1C_2	N_2C_1	N_3
Sc	-1.64 (-1.67)	-1.03 (-1.07)	-0.63 (-0.67)	-0.86 (-0.94)	-1.20 (-1.24)	-0.75 (-0.80)	-0.99 (-1.04)	-1.46 (-1.53)	-1.67 (-1.74)	-1.49 (-1.55)	-1.69 (-1.73)
Ti	-0.77 (-0.71)	-0.20 (-0.16)	0.30 (0.33)	-0.07 (-0.02)	-0.55 (-0.54)	-0.20 (-0.03)	-0.65 (-0.48)	-0.59 (-0.56)	-1.45 (-1.37)	-1.29 (-1.21)	-1.61 (-1.46)
V	-0.63 (-0.31)	-0.19 (0.18)	0.23 (0.68)	-0.15 (0.32)	-0.59 (-0.12)	-0.18 (0.34)	-0.54 (0.09)	-0.52 (-0.24)	-1.25 (-0.86)	-1.09 (-0.60)	-1.41 (-0.78)
Cr	-0.56 (0.00)	-0.11 (-0.59)	0.37 (1.08)	-0.08 (0.67)	-0.50 (0.24)	0.03 (0.85)	-0.18 (0.73)	-0.25 (0.27)	-1.06 (-0.39)	-1.09 (-0.31)	-1.61 (-0.65)
Mn	-0.90 (0.18)	-0.06 (1.03)	0.31 (1.39)	0.04 (1.14)	-0.38 (0.70)	-0.02 (1.07)	-0.40 (0.57)	-0.39 (-0.06)	-1.44 (-0.81)	-1.36 (-0.11)	-1.60 (-0.19)
Fe	0.03 (0.27)	0.56 (0.94)	0.88 (1.25)	0.65 (1.02)	0.27 (0.63)	0.60 (0.89)	0.27 (0.62)	0.44 (0.32)	-0.50 (-0.49)	-0.59 (-0.27)	-1.00 (-0.44)
Co	0.26 (0.51)	0.69 (1.16)	1.12 (1.46)	0.88 (1.22)	0.54 (0.85)	0.90 (1.26)	0.56 (0.95)	0.76 (0.83)	-0.26 (-0.15)	-0.30 (0.04)	-0.74 (-0.26)
Ni	0.32 (0.32)	1.00 (0.98)	1.42 (1.54)	1.19 (1.29)	0.89 (0.98)	1.14 (1.30)	0.68 (0.89)	0.43 (0.58)	-0.50 (-0.24)	-0.47 (-0.09)	-0.85 (-0.46)
Cu	1.11 (1.05)	1.72 (1.68)	1.83 (1.80)	1.65 (1.62)	1.30 (1.28)	1.34 (1.34)	0.86 (0.98)	0.32 (0.34)	-0.19 (-0.12)	0.06 (0.17)	-0.08 (0.08)
Zn	0.05 (0.01)	0.29 (0.31)	0.73 (0.82)	0.28 (0.36)	-0.07 (0.02)	0.33 (0.49)	0.13 (0.36)	-0.76 (-0.68)	-1.13 (-1.01)	-0.50 (-0.35)	-0.69 (-0.57)
Y	-1.93	-1.35	-0.94	-1.16	-1.51	-1.05	-1.27	-1.77	-1.99	-1.80	-1.95
Zr	-1.03	-0.75	-0.49	-0.67	-0.93	-0.76	-1.04	-1.02	-1.48	-1.44	-1.64
Nb	-0.79	-0.48	-0.37	-0.59	-0.94	-0.75	-1.17	-0.93	-1.52	-1.53	-1.84
Mo	-0.13	0.15	0.21	0.00	-0.35	-0.22	-0.63	-0.11	-0.83	-0.88	-1.21
Tc	0.29	0.55	0.61	0.28	-0.26	-0.10	-0.61	0.32	-0.68	-0.75	-1.45
Ru	0.94	1.05	1.31	0.76	0.39	0.57	0.09	1.28	0.12	-0.05	-0.81
Rh	1.23	1.61	1.82	1.38	1.03	1.33	0.98	1.74	0.60	0.32	-0.45
Pd	1.58	2.09	2.46	2.28	1.94	2.22	1.76	1.80	0.76	0.87	0.29
Ag	0.83	2.15	2.42	2.11	1.33	1.47	-0.28	0.22	-0.54	-0.12	-0.08
Cd	-0.47	-0.22	0.43	-0.16	-0.62	0.03	-0.20	-0.70	*	-0.62	-0.63
La	-1.18	-0.67	-0.22	-0.46	-0.79	-0.34	-0.54	-0.97	-1.20	-0.99	-1.15
Hf	-1.16	-0.88	-0.62	-0.80	-1.07	-0.86	-1.11	-1.13	-1.60	-1.56	-1.76
Ta	-0.31	-0.02	0.14	-0.10	-0.44	-0.28	-0.66	-0.49	-1.16	-1.17	-1.50
W	0.09	0.30	0.39	0.18	-0.16	-0.07	-0.50	-0.02	-0.77	-0.85	-1.22
Re	0.38	0.50	0.53	0.32	-0.05	0.02	-0.41	0.27	-0.47	-0.56	-1.11
Os	0.90	0.92	0.95	0.83	0.73	0.77	0.63	0.92	0.61	0.53	0.31
Ir	1.62	1.84	1.94	1.70	1.40	1.59	1.31	1.80	0.94	0.62	-0.02
Pt	2.10	2.54	2.77	2.63	2.26	2.45	1.96	2.01	0.76	0.58	-0.25
Au	1.96	2.32	2.35	2.27	1.98	2.00	1.33	1.33	0.99	1.20	0.87
Hg	1.07	1.29	1.76	*	-0.26	*	*	*	*	*	*

Table S14. Classification of 330 possible stabilities against (TM)SA-aggregation/dissolution for different M-N_nC_m-G_N template types in combination of 30 different (TM)SAs into four different categrories.

(TM)SA Systems	Unaggregatable Indissoluble	Unaggregatable Dissoluble	Aggregatable Indissoluble	Aggregatable Dissoluble
C_4	16	11	2	1
N_1C_3	21	9	-	-
N_2C_2	24	6	-	-
N_2C_2[b]	12	15	3	-
N_2C_2[c]	12	10	2	6
N_3C_1	16	8	2	4
N_4	12	7	3	8
C_3	10	12	5	3
N_1C_2	-	9	7	14
N_2C_1	1	11	7	11
N_3	-	10	4	16
Total	**124**	**108**	**35**	**63**

Table S15. Magnetization (μ in Bohr) before/after hydrogen adsorption for M-N_nC_m-G_N. (RPBE(U)+TS results are given for 3d metals).

Transition Metal	C_4	N_1C_3	N_2C_2	N_2C_2[b]	N_2C_2[c]	N_3C_1	N_4	C_3	N_1C_2	N_2C_1	N_3
Sc	0.8/0 (0.4/0)	0/0 (0/0)	0/0 (0/0)	0/0 (0/0)	0/0 (0/0)	0/0 (0/0)	0/0 (0/0)	0 (0)	1.0 (0)	0 (0.3)	0.5 (0.7)
Ti	0/0 (0/0)	0/0 (0.7/0)	0/0 (0.3/0)	0/0 (0.3/0)	0/0 (0.2/0)	0.8/0 (1.0/0)	1.4/0.6 (1.5/1.0)	0 (0)	0.7 (1.0)	1.6 (1.3)	2.0 (1.8)
V	0.9/0 (0.9/0)	1.3/0 (2.0/0)	1.2/0.7 (1.6/1.3)	1.2/0.7 (1.6/1.3)	1.1/0.6 (1.6/1.5)	2.0/1.0 (2.0/1.0)	3.0/2.0 (3.0/2.0)	1.0 (1.0)	2.0 (2.0)	2.6 (2.9)	2.6 (2.9)
Cr	1.9/3.0 (2.0/3.0)	2.9/3.4 (3.0/4.0)	2.6/3.1 (2.7/3.4)	3.1/1.6 (3.5/2.7)	3.5/3.0 (3.9/3.3)	3.2/3.9 (3.7/4.0)	4.0/3.0 (4.0/3.0)	2.0 (2.0)	3.0 (3.0)	2.5 (3.9)	4.3 (4.4)
Mn	4.4/3.9 (4.4/4.0)	4.0/3.1 (4.0/3.4)	3.7/3.4 (4.0/4.4)	3.6/2.9 (3.9/3.3)	3.5/4.0 (3.8/4.0)	3.8/3.1 (4.0/5.0)	3.0/2.0 (3.0/2.0)	3.0 (1.0)	4.0 (2.0)	4.5 (5.0)	5.3 (5.6)
Fe	3.2/3.0 (2.2/3.0)	3.0/2.0 (3.0/3.8)	2.7/2.6 (3.0/3.0)	2.8/2.0 (3.0/2.2)	2.3/3.0 (2.9/3.0)	2.4/2.0 (3.0/2.0)	2.0/1.0 (2.0/1.0)	0 (2.0)	1.0 (3.0)	2.0 (4.0)	3.2 (4.5)
Co	1.1/1.4 (1.2/2.0)	2.0/1.0 (2.0/1.0)	1.2/0 (1.1/1.1)	1.2/0 (1.8/1.0)	1.0/0 (1.0/2.0)	0.9/0.8 (1.1/1.0)	0.9/0 (1.0/0)	1.0 (1.0)	0 (2.0)	1.0 (1.0)	2.3 (2.5)
Ni	0/0 (1.0/0)	0.7/0 (1.0/0)	0/0 (0/0)	0/0 (0/0)	0/0 (0/0)	0/0 (0/0)	0/0 (0/0)	0 (0)	1.0 (1.0)	2.0 (2.0)	1.6 (1.7)
Cu	0/0 (0/0)	0/0 (0/0)	0/0 (0/0)	0/0 (0/0)	0/0 (0/0)	0.1/0 (0/0)	1.0/0 (1.0/0.1)	1.0 (1.0)	0 (0)	1.0 (1.0)	0 (0)
Zn	0/1.0 (0/0)	0.1/0 (0.7/0)	0/0 (0/0)	0/1.0 (0/1.0)	0/0 (0/0)	0/0 (0/0)	0/0 (0/0)	1.3 (1.2)	0 (0)	0 (0)	0.9 (0.6)
Y	0.8/0	0/0	0/0	0/0	0/0	0/0	0/0	0	0	0	0.8
Zr	0/0	0/0	0/0	0/0	0/0	0.4/0	0.8/0	0	0	1.3	1.9
Nb	0.3/0	0/0	0.7/0	1.0/0	0.8/0	1.6/0.9	2.2/1.1	1.0	2.0	2.2	2.4
Mo	0.5/0.9	1.0/1.1	2.0/0	1.7/1.0	1.8/1.0	2.8/2.0	3.7/2.5	2.0	2.8	2.1	1.0
Tc	1.0/1.0	0/1.0	1.9/1.0	0.1/0	3.0/2.0	0.3/1.0	3.0/2.0	1.0	0	1.0	0.6
Ru	0/0	0/0	0/1.7	1.9/0.9	2.0/1.0	1.0/0	1.9/0.8	0	1.0	0	0
Rh	0.9/0	0/0.9	0/0	1.0/0	1.0/0	0/0	0/0	1.0	0	1.0	2.0
Pd	0/0	0/0	0/0	0/0	0/0	0/0	0/0	0	1.0	0	1.0
Ag	0/0	0/0	0/0	0/0	0/0	0/0	0/0	1.0	0	1.0	0
Cd	0/0	0/0	0/0	0/1.0	1.5/1.0	0/0	0/0	2.0	*	0.3	0.6
La	0/0	0/0.9	0/0	0/0	0/0	0/1.9	0/0	0	0	0	0
Hf	0/0	0/0	0/0	0/0	0/0	0.7/0.7	1.0/0	0	0	1.1	2.0
Ta	0.7/0	0/0	0.9/0	1.0/0	0.9/0	1.5/1.9	1.0/0	0.9	1.9	2.3	1.9
W	0/0	1.0/0.7	1.9/1.0	1.9/1.0	1.9/1.0	2.9/1.0	2.1/1.0	2.0	2.6	2.0	1.0
Re	1.0/0	0/0	2.7/2.0	0.8/0	3.0/2.0	3.9/0	3.0/2.0	1.0	0	1.0	0.9
Os	0/0	0/0	0.2/0.9	1.4/0	2.0/1.0	1.0/0.4	0/1.0	0	1.0	0	0
Ir	0/0	0/1.0	0/0	0/0	0/0	0/0	0.6/0	1.0	0	0.9	1.6
Pt	0/0	0.3/0	0/0	0/0	0/0	0/0	0/0	0	1.0	0	1.1
Au	0/0	0/0	0/0	0/0	0/0	0/0	0/0	1.0	0	0.4	0
Hg	0/0	0/0	0/1.0	*	0/1.0	*	*	*	*	*	*

"*" denotes unstable systems.

Table S16. Magnetization (μ in Bohr) before "prist" and after adsorption of *O, *OH and *OOH for M-N_nC_m-G_N in the absence of solvent. (RPBE(U)+TS results are given for 3d metals).

TM	C_4				N_1C_3				N_2C_2				N_2C_2[b]			
	prist	*O	*OH	*OOH	prist	*O	*OH	*OOH	prist	*O	*OH	*OOH	prist	*O	*OH	*OOH
Sc	0.8	0.9	2.0	0.0	0.0	0.0	1.0	1.0	0.0	1.0	0.0	0.0	0.0	1.0	0.0	0.0
	(0.4)	(0.9)	(2.0)	(2.0)	(0.0)	(0.0)	(1.0)	(1.0)	(0.0)	(1.0)	(0.0)	(0.0)	(0.0)	(1.0)	(0.0)	(0.0)
Ti	0.0	1.3	0.0	0.0	0.0	0.9	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	(0.0)	(1.5)	(0.0)	(0.0)	(0.7)	(1.1)	(0.0)	(0.0)	(0.3)	(0.0)	(0.0)	(0.0)	(0.3)	(0.0)	(0.0)	(0.0)
V	0.9	0.0	0.0	0.0	1.3	0.0	0.6	0.6	1.2	0.0	1.0	1.0	**0.0**	**0.0**	**0.3**	**0.6**
	(0.9)	(0.0)	(0.0)	(0.0)	(2.0)	(0.0)	(1.0)	(1.0)	(1.6)	(0.0)	(1.3)	(1.4)	**(0.0)**	**(1.0)**	**(1.3)**	**(1.8)**
Cr	1.9	0.9	1.1	1.0	2.9	1.0	1.8	1.9	2.6	1.1	1.6	2.2	**1.1**	**1.3**	**2.5**	**2.5**
	(2.0)	(1.0)	(1.6)	(1.1)	(3.0)	(1.0)	(1.9)	(2.0)	(2.7)	(1.2)	(2.5)	(2.5)	**(1.2)**	**(2.0)**	**(2.9)**	**(3.0)**
Mn	4.4	1.0	1.9	1.5	4.0	1.0	2.8	2.8	3.7	0.0	2.1	2.1	3.6	0.0	2.7	2.3
	(4.4)	(1.2)	(2.1)	(2.3)	(4.0)	(0.0)	(2.9)	(2.9)	(4.0)	(0.0)	(3.5)	(3.4)	(3.9)	(0.0)	(3.6)	(3.6)
Fe	3.2	0.0	2.5	2.5	3.0	0.0	3.8	1.7	2.7	1.0	1.1	0.0	2.8	0.5	2.9	2.9
	(2.2)	(0.0)	(2.9)	(2.9)	(3.0)	(1.0)	(3.9)	(1.9)	(3.0)	(1.5)	(3.2)	(2.5)	(3.0)	(1.8)	(3.3)	(2.7)
Co	1.1	0.2	0.0	0.0	2.0	1.8	1.0	1.0	1.2	1.4	2.0	0.0	1.2	1.9	1.9	1.9
	(1.2)	(1.0)	(1.5)	(0.9)	(2.0)	(1.9)	(1.0)	(1.0)	(1.1)	(2.8)	(2.0)	(1.9)	(1.8)	(2.6)	(2.0)	(1.9)
Ni	0.0	1.2	0.0	0.0	0.7	2.4	0.0	0.0	0.0	2.0	1.0	1.0	0.0	2.0	0.5	0.9
	(1.0)	(1.3)	(1.0)	(0.0)	(1.0)	(2.6)	(0.0)	(0.0)	(0.0)	(2.0)	(0.9)	(0.9)	(0.0)	(2.0)	(0.9)	(0.9)
Cu	0.0	1.8	0.5	1.0	0.0	1.8	0.7	0.8	0.0	1.0	0.0	0.0	0.0	1.1	0.0	0.0
	(0.0)	(1.8)	(0.4)	(0.9)	(0.0)	(1.8)	(0.8)	(0.8)	(0.0)	(1.0)	(0.0)	(0.0)	(0.0)	(1.1)	(0.0)	(0.2)
Zn	0.0	1.3	0.3	0.6	0.1	1.0	0.0	0.0	0.0	2.0	1.0	1.0	0.0	2.0	1.0	1.0
	(0.0)	(1.4)	(0.4)	(0.6)	(0.7)	(1.0)	(0.0)	(0.0)	(0.0)	(2.0)	(1.0)	(1.0)	(0.0)	(2.0)	(1.0)	(1.0)
Y	0.0	0.9	2.0	0.0	0.0	0.0	1.0	1.0	0.0	1.0	0.0	0.0	0.0	1.0	0.0	0.0
Zr	0.0	0.1	0.0	0.0	0.0	1.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Nb	0.3	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.7	0.0	0.0	0.0	1.0	0.0	0.0	0.0
Mo	0.5	0.0	1.0	1.0	1.0	0.0	1.2	1.1	**1.1**	**0.0**	**1.0**	**1.0**	**1.1**	**0.0**	**1.0**	**1.0**
Tc	1.0	0.9	1.1	1.1	**0.1**	**0.0**	**1.0**	**1.0**	**0.0**	**0.0**	**0.0**	**0.0**	**0.0**	**0.0**	**0.0**	**0.1**
Ru	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	**0.0**	**0.0**	**0.0**	**1.0**	**0.0**	**0.0**	**0.0**	**0.0**
Rh	0.9	0.0	0.0	0.0	0.0	0.0	1.0	0.0	0.0	1.0	0.0	0.0	1.0	1.0	0.0	0.0
Pd	0.0	0.0	0.9	0.0	0.0	1.0	0.0	0.0	0.0	2.0	0.5	0.5	0.0	1.8	0.5	0.8
Ag	0.0	1.1	0.0	0.0	0.0	2.0	1.0	0.9	0.0	1.1	0.0	0.5	0.0	1.0	0.0	0.7
Cd	0.0	2.0	1.0	0.6	0.0	1.0	0.0	0.0	0.0	2.0	1.0	1.0	0.0	0.0	1.0	1.0
La	0.0	0.0	0.0	0.0	0.0	0.0	0.9	0.9	0.0	1.0	0.0	0.0	0.0	1.7	0.0	0.0
Hf	0.0	0.5	0.0	0.0	0.0	0.8	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Ta	0.7	0.0	0.0	0.0	0.0	0.0	0.0	0.0	**0.0**	**0.0**	**0.0**	**0.0**	**0.0**	**0.0**	**0.0**	**0.0**
W	0.0	0.0	0.0	0.0	1.0	0.0	1.1	1.1	**1.0**	**0.0**	**1.0**	**1.0**	**1.0**	**0.0**	**0.8**	**0.8**
Re	**1.0**	**0.7**	**0.2**	**0.5**	**0.9**	**0.0**	**1.0**	**1.0**	**0.2**	**0.7**	**0.0**	**0.0**	**0.3**	**0.8**	**0.2**	**0.1**
Os	**0.0**	**1.0**	**0.0**	**0.0**	**0.0**	**0.0**	**0.0**	**0.0**	**0.0**	**0.0**	**0.5**	**0.5**	**0.0**	**0.0**	**0.0**	**0.0**
Ir	0.0	0.0	0.0	0.0	0.0	0.0	1.0	0.0	0.0	1.0	0.0	0.0	0.0	1.0	0.0	0.0
Pt	0.0	0.0	0.8	0.0	0.3	1.0	0.0	0.0	0.0	0.0	0.0	0.1	0.0	1.9	0.0	0.0
Au	0.0	1.1	0.0	0.9	0.0	0.0	0.0	0.9	0.0	1.1	0.0	0.7	0.0	0.9	0.0	0.8
Hg	0.0	1.4	0.3	0.7	0.0	1.0	0.0	0.0	0.0	1.9	1.0	1.0	*	*	*	*

Table S16. Continued...

TM	N_2C_2[c]				N_3C_1				N_4			
	prist	***O**	***OH**	***OOH**	**prist**	***O**	***OH**	***OOH**	**prist**	***O**	***OH**	***OOH**
Sc	0.0	0.9	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.2	0.0	0.0
	(0.0)	(0.9)	(0.0)	(0.0)	(0.0)	(0.2)	(0.0)	(0.0)	(0.0)	(0.0)	(0.0)	(0.0)
Ti	0.0	0.1	0.0	0.0	0.8	0.0	0.0	0.0	1.4	0.0	0.0	0.5
	(0.2)	(0..0)	(0.0)	(0.0)	(1.0)	(0.0)	(0.0)	(0.0)	(1.5)	(0.0)	(1.0)	(1.0)
V	**0.0**	**0.3**	**0.0**	**0.1**	2.0	0.0	1.0	1.0	**1.0**	**0.6**	**0.0**	**0.0**
	(0.0)	**(1.0)**	**(2.0)**	**(2.0)**	(2.0)	(1.1)	(1.0)	(1.1)	**(1.0)**	**(1.0)**	**(0.0)**	**(0.0)**
Cr	**1.4**	**1.4**	**2.9**	**2.9**	3.2	1.0	2.0	2.0	4.0	0.2	3.0	3.0
	(1.5)	**(2.0)**	**(3.0)**	**(3.0)**	(3.7)	(1.0)	(2.1)	(2.1)	(4.0)	(0.0)	(3.0)	(3.0)
Mn	3.5	0.0	3.8	3.6	3.8	0.0	3.1	3.1	3.0	0.8	3.5	3.5
	(3.8)	(0.0)	(4.0)	(3.9)	(4.0)	(0.0)	(3.2)	(3.2)	(3.0)	(1.0)	(3.9)	(3.9)
Fe	**0.0**	**4.0**	**3.0**	**1.0**	2.4	1.2	2.1	0.0	2.0	2.0	1.0	1.0
	(2.0)	**(4.0)**	**(5.0)**	**(3.0)**	(3.0)	(1.6)	(2.3)	(2.3)	(2.0)	(2.0)	(2.9)	(2.9)
Co	1.0	2.7	2.0	2.0	0.8	2.1	1.0	1.1	0.9	3.0	0.0	1.6
	(1.0)	(2.9)	(2.0)	(2.0)	(1.1)	(2.2)	(1.3)	(1.4)	(1.0)	(3.3)	(1.9)	(2.0)
Ni	0.0	2.0	1.0	1.0	0.0	1.1	0.0	0.0	0.0	1.9	0.9	1.0
	(0.0)	(2.0)	(1.0)	(1.0)	(0.0)	(1.1)	(0.2)	(0.2)	(0.0)	(1.9)	(0.9)	(1.0)
Cu	0.0	1.0	0.0	0.0	0.1	1.8	0.0	0.7	1.0	1.7	1.2	1.7
	(0.0)	(1.1)	(0.0)	(0.0)	(0.0)	(1.9)	(0.0)	(0.6)	(1.0)	(1.8)	(1.2)	(1.7)
Zn	0.0	0.0	0.8	0.8	0.0	1.0	0.0	0.2	0.0	1.0	0.0	0.0
	(0.0)	(0.0)	(0.9)	(0.8)	(0.0)	(1.0)	(0.0)	(0.2)	(0.0)	(1.0)	(0.0)	(0.0)
Y	0.0	0.0	0.0	0.0	0.0	0.6	0.0	0.0	0.0	0.4	0.0	0.0
Zr	0.0	0.0	0.0	0.0	0.4	0.0	0.0	0.0	0.8	0.0	0.0	0.0
Nb	**0.0**	**0.0**	**0.0**	**0.0**	**0.0**	**0.0**	**0.0**	**0.0**	**0.7**	**0.0**	**0.0**	**0.0**
Mo	**1.3**	**0.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.9**	**1.5**	**0.3**	**0.0**	**1.0**	**0.9**
Tc	**0.0**	**0.9**	**2.0**	**2.0**	**2.0**	**0.0**	**0.9**	**0.8**	**0.0**	**0.6**	**0.2**	**0.0**
Ru	**0.0**	**0.0**	**0.8**	**0.6**	**1.0**	**1.0**	**0.0**	**0.0**	**1.8**	**0.0**	**1.3**	**0.0**
Rh	1.0	1.0	2.0	0.0	0.0	2.0	0.9	0.9	0.0	1.0	0.0	0.0
Pd	0.0	2.0	1.0	1.0	0.0	1.0	0.0	0.0	0.0	2.0	0.9	0.9
Ag	0.0	1.0	0.0	0.4	0.0	0.5	0.0	0.8	0.7	1.6	0.1	0.0
Cd	0.0	0.0	1.0	0.9	0.0	1.0	0.0	0.0	0.0	0.9	0.0	0.0
La	0.0	0.0	0.0	0.0	0.0	1.0	0.0	0.0	0.0	0.0	0.0	0.0
Hf	0.0	0.0	0.0	0.0	0.7	0.0	0.0	0.0	1.0	0.0	0.0	0.0
Ta	0.9	0.0	0.0	0.0	1.5	0.0	0.8	0.0	**0.0**	**0.0**	**0.0**	**0.0**
W	**0.0**	**0.0**	**0.9**	**0.0**	**0.0**	**0.0**	**0.0**	**0.0**	**0.8**	**0.0**	**0.3**	**0.0**
Re	**0.4**	**0.0**	**2.0**	**2.0**	**1.0**	**1.0**	**0.0**	**0.6**	**0.0**	**0.9**	**0.4**	**0.0**
Os	**0.0**	**0.0**	**0.8**	**0.0**	**0.0**	**0.0**	**1.0**	**0.4**	**0.0**	**0.0**	**0.0**	**0.0**
Ir	**1.0**	**0.9**	**0.0**	**0.0**	**0.0**	**0.9**	**0.0**	**0.0**	0.6	1.0	0.0	0.0
Pt	0.0	2.0	0.0	1.0	0.0	0.0	0.0	0.0	0.0	0.0	1.0	0.9
Au	0.0	0.9	0.0	0.5	0.0	1.8	0.0	0.9	0.0	1.5	0.4	0.6
Hg	0.0	0.0	1.0	1.0	*	*	*	*	*	*	*	*

"**prist**" the prstine state without O-adsorption for the embedded (TM)SA (The values in blue follows path-II shown in Figure 6).

Table S17. Magnetization (μ in Bohr) before " prist" and after adsorption of *O, *OH and *OOH for M-N_nC_m-G_N in the presence of solvent (dielectric constant 80). (RPBE(U)+TS results are given for 3d metals)

TM	C_4				N_1C_3				N_2C_2				N_2C_2[b]			
	prist	*O	*OH	*OOH	prist	*O	*OH	*OOH	prist	*O	*OH	*OOH	prist	*O	*OH	*OOH
Sc	0.9	0.0	0.0	1.9	0.0	0.0	0.9	1.0	0.0	1.0	0.0	0.0	0.0	1.0	0.0	0.0
	(0.9)	(0.0)	(0.0)	(1.9)	(0.0)	(0.0)	(0.9)	(1.0)	(0.0)	(1.0)	(0.0)	(0.0)	(0.0)	(1.0)	(0.0)	(0.0)
Ti	0.0	1.0	0.0	0.0	0.0	0.3	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	(0.0)	(1.0)	(0.0)	(0.0)	(0.0)	(0.4)	(0.0)	(0.0)	(0.0)	(0.0)	(0.0)	(0.0)	(0.0)	(0.0)	(0.0)	(0.0)
V	0.7	0.0	0.0	0.0	0.6	0.0	0.6	0.5	1.0	0.0	1.0	1.0	**0.0**	**0.0**	**0.3**	**0.0**
	(0.9)	(0.0)	(0.0)	(0.0)	(1.9)	(0.0)	(1.0)	(1.0)	(1.1)	(0.4)	(1.3)	(1.5)	**(0.0)**	**(1.0)**	**(1.4)**	**(1.9)**
Cr	1.9	1.0	1.1	1.0	1.1	1.0	1.8	1.9	2.1	1.1	1.6	2.2	**1.1**	**1.4**	**2.6**	**2.6**
	(1.9)	(1.0)	(1.6)	(1.1)	(3.0)	(1.0)	(1.9)	(1.9)	(2.4)	(1.2)	(2.5)	(2.6)	**(1.2)**	**(2.0)**	**(2.9)**	**(3.0)**
Mn	2.8	1.0	1.9	1.5	3.9	1.8	2.8	2.8	3.5	0.0	2.1	2.1	3.5	0.0	3.2	2.3
	(4.5)	(1.4)	(2.2)	(2.5)	(4.0)	(0.0)	(2.9)	(2.9)	(3.7)	(0.0)	(3.8)	(3.8)	(3.7)	(0.0)	(3.8)	(3.7)
Fe	3.5	0.0	0.2	2.5	3.0	0.2	3.8	1.6	2.7	1.2	1.1	0.0	2.8	0.9	2.9	2.9
	(2.1)	(0.6)	(2.9)	(2.9)	(3.0)	(1.0)	(3.9)	(1.9)	(3.0)	(1.6)	(3.2)	(2.9)	(3.0)	(1.9)	(3.3)	(2.9)
Co	1.1	0.2	0.0	0.0	2.0	1.6	1.0	1.0	1.1	1.7	2.0	0.0	1.2	2.2	1.9	1.9
	(1.2)	(1.8)	(1.4)	(0.9)	(2.0)	(1.9)	(1.0)	(1.0)	(1.1)	(2.9)	(2.0)	(1.9)	(2.0)	(2.8)	(2.9)	(1.9)
Ni	0.0	1.1	0.0	0.0	0.7	2.3	0.0	0.0	0.0	1.9	0.9	1.0	0.0	1.9	0.0	0.9
	(0.3)	(1.2)	(1.0)	(0.0)	(0.0)	(2.4)	(0.0)	(0.0)	(0.0)	(2.0)	(0.9)	(0.9)	(0.0)	(2.0)	(2.0)	(0.9)
Cu	0.0	1.9	0.6	1.0	0.0	1.4	0.1	0.8	0.0	1.0	0.0	0.0	0.0	1.0	0.0	0.0
	(0.0)	(1.9)	(0.5)	(1.0)	(0.0)	(1.4)	(0.2)	(0.8)	(0.0)	(1.0)	(0.0)	(0.0)	(0.0)	(1.0)	(0.9)	(0.0)
Zn	0.0	1.1	0.1	0.5	0.4	1.0	0.0	0.0	0.0	2.0	1.0	1.0	0.0	2.0	1.0	1.0
	(0.0)	(1.1)	(0.1)	(0.5)	(0.7)	(1.0)	(0.0)	(0.0)	(0.0)	(2.0)	(1.0)	(1.0)	(0.0)	(2.0)	(0.0)	(1.0)
Y	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.5	0.0	1.0	0.0	0.0	0.0	1.0	0.0	0.0
Zr	0.0	0.0	0.0	0.0	0.0	1.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Nb	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.7	0.0	0.0	0.0	0.9	0.0	0.0	0.0
Mo	1.3	0.0	1.0	0.9	1.0	0.0	1.2	1.2	**1.1**	**0.0**	**1.0**	**1.0**	**1.1**	**0.0**	**1.0**	**1.0**
Tc	0.9	0.9	1.1	1.1	**0.1**	**0.0**	**1.0**	**1.0**	**0.0**	**0.0**	**0.0**	**0.0**	**0.0**	**0.0**	**0.0**	**0.0**
Ru	0.0	0.0	0.0	0.0	1.0	0.0	0.0	0.0	**0.0**	**0.0**	**0.7**	**1.0**	**0.0**	**0.0**	**0.0**	**0.0**
Rh	0.9	0.0	0.0	0.0	0.0	0.0	1.0	0.9	0.0	1.0	0.0	0.0	1.0	1.0	0.0	0.0
Pd	0.0	0.0	0.7	0.4	0.0	1.0	0.0	0.0	0.0	1.9	0.7	0.7	0.0	0.0	0.8	0.9
Ag	0.0	0.8	0.0	0.0	0.0	0.2	1.0	0.8	0.0	1.0	0.0	0.0	0.0	1.0	0.0	0.4
Cd	0.0	0.0	1.0	0.5	0.0	0.7	0.0	0.0	0.0	1.9	1.0	1.0	0.0	0.0	1.0	1.0
La	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Hf	0.0	0.0	0.0	0.0	0.0	0.9	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Ta	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	**0.0**	**0.0**	**0.0**	**0.0**	**0.0**	**0.0**	**0.0**	**0.0**
W	0.5	0.0	0.0	0.0	1.0	0.0	1.1	0.0	**1.0**	**0.0**	**1.0**	**1.0**	**1.1**	**0.0**	**0.8**	**0.4**
Re	**1.0**	**0.9**	**0.5**	**0.6**	**0.9**	**0.0**	**1.0**	**1.0**	**0.2**	**0.6**	**0.0**	**0.0**	**0.2**	**0.8**	**0.1**	**0.0**
Os	**0.0**	**0.0**	**0.0**	**0.0**	**0.0**	**0.0**	**0.0**	**0.0**	**0.0**	**0.0**	**0.5**	**0.8**	**0.0**	**0.0**	**0.0**	**0.0**
Ir	0.8	0.0	0.0	0.0	0.0	0.0	1.0	0.4	0.0	1.0	0.0	0.0	0.0	1.0	0.0	0.0
Pt	0.0	0.1	0.0	0.0	0.0	0.9	0.0	0.0	0.0	0.0	0.3	0.3	0.0	1.8	0.0	0.4
Au	0.0	0.9	0.0	0.9	0.0	0.0	0.0	0.9	0.0	1.0	0.0	0.5	0.0	0.6	0.0	0.6
Hg	0.0	1.1	0.0	0.7	0.0	0.9	0.0	0.0	0.0	1.7	1.0	1.0	*	*	*	*

Table S17. Continued...

TM	N_2C_2[c]				N_3C_1				N_4			
	prist	*O	*OH	*OOH	prist	*O	*OH	*OOH	prist	*O	*OH	*OOH
Sc	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	(0.0)	(0.0)	(0.0)	(0.0)	(0.0)	(0.0)	(0.0)	(0.0)	(0.0)	(0.0)	(0.0)	(0.0)
Ti	0.1	0.0	0.0	0.0	0.2	0.0	0.0	0.0	0.8	0.0	0.4	0.5
	(0.0)	(0.0)	(0.0)	(0.0)	(0.0)	(0.0)	(0.0)	(0.1)	(1.0)	(0.0)	(1.0)	(0.9)
V	**0.5**	**0.6**	**0.0**	**0.4**	1.3	0.7	1.0	1.0	**1.0**	**0.9**	**0.0**	**0.0**
	(0.0)	**(1.0)**	**(2.0)**	**(2.0)**	(0.0)	(1.1)	(1.0)	(1.1)	**(1.0)**	**(1.0)**	**(0.6)**	**(0.1)**
Cr	**1.3**	**1.5**	**2.8**	**2.9**	3.0	1.0	2.0	2.0	4.0	0.1	3.0	3.0
	(1.4)	**(2.0)**	**(3.0)**	**(3.0)**	(1.8)	(1.0)	(2.1)	(2.2)	(4.0)	(0.0)	(1.0)	(3.0)
Mn	3.2	0.0	3.8	3.7	3.9	0.0	3.1	3.1	3.0	0.9	3.6	3.6
	(3.6)	(0.0)	(4.0)	(3.9)	(3.3)	(0.0)	(3.3)	(3.3)	(3.0)	(1.0)	(3.9)	(3.9)
Fe	**0.5**	**3.9**	**3.0**	**1.0**	2.4	1.3	2.1	0.0	2.0	2.0	1.0	1.0
	(2.0)	**(4.0)**	**(5.0)**	**(3.0)**	(4.0)	(2.1)	(2.4)	(2.4)	(2.0)	(2.0)	(4.9)	(3.0)
Co	1.0	2.7	2.0	2.0	0.8	2.1	1.0	1.0	0.9	3.2	0.0	0.0
	(0.0)	(2.9)	(2.0)	(2.0)	(3.0)	(2.2)	(1.4)	(1.4)	(0.0)	(3.5)	(2.0)	(2.0)
Ni	0.0	2.0	1.0	1.0	0.0	1.0	0.0	0.0	0.0	3.0	0.8	0.9
	(0.0)	(2.0)	(1.0)	(1.0)	(0.1)	(1.1)	(0.3)	(0.3)	(0.0)	(1.6)	(0.9)	(0.9)
Cu	0.0	1.0	0.0	0.0	0.0	0.1	0.0	0.0	1.0	1.5	1.1	1.4
	(0.0)	(1.0)	(0.0)	(0.0)	(0.0)	(2.0)	(0.0)	(0.1)	(1.0)	(1.5)	(1.1)	(1.5)
Zn	0.0	0.0	1.0	1.0	0.0	1.0	0.0	0.0	0.0	0.7	0.0	0.0
	(0.0)	(0.0)	(1.0)	(1.0)	(0.0)	(1.0)	(0.0)	(0.0)	(0.0)	(0.7)	(0.0)	(0.0)
Y	0.0	0.0	0.0	0.0	0.0	1.0	0.0	0.0	0.0	0.0	0.0	0.0
Zr	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Nb	**0.0**	**0.0**	**0.0**	**0.0**	**0.0**	**0.0**	**0.0**	**0.0**	**0.8**	**0.0**	**0.0**	**0.0**
Mo	**1.3**	**0.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.9**	**1.8**	**0.2**	**0.0**	**1.0**	**1.0**
Tc	**0.0**	**0.9**	**2.0**	**0.0**	**0.0**	**0.0**	**0.9**	**0.1**	**0.0**	**0.0**	**0.0**	**0.0**
Ru	**0.0**	**0.0**	**0.7**	**0.5**	**1.0**	**1.0**	**0.0**	**0.8**	**2.0**	**0.0**	**0.0**	**0.0**
Rh	1.0	1.0	2.0	0.0	0.0	2.0	0.9	0.9	0.4	1.0	0.0	0.0
Pd	0.0	1.9	1.0	1.0	0.0	1.0	0.0	0.0	0.0	1.2	0.7	0.8
Ag	0.0	1.0	0.0	0.0	0.0	0.0	0.0	0.6	0.6	1.1	0.0	0.0
Cd	0.0	0.0	1.0	1.0	0.0	1.0	0.0	0.0	0.0	0.6	0.0	0.0
La	-	-	-	-	-	-	-	-	-	-	-	-
Hf	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Ta	0.0	0.0	0.0	0.0	**0.0**	**0.0**	**0.0**	**0.0**	**0.0**	**0.0**	**0.0**	**0.0**
W	**0.0**	**0.0**	**0.9**	**0.4**	**1.0**	**1.0**	**0.0**	**0.0**	**0.7**	**0.0**	**0.6**	**0.0**
Re	**0.3**	**0.0**	**2.0**	**2.0**	**0.0**	**0.0**	**1.0**	**0.0**	**0.0**	**0.8**	**0.2**	**0.2**
Os	**0.0**	**0.0**	**0.8**	**0.0**	**0.8**	**0.8**	**0.0**	**1.0**	**0.0**	**0.0**	**0.0**	**0.0**
Ir	**1.0**	**0.7**	**0.0**	**0.0**	0.0	2.0	1.0	1.0	0.6	1.0	0.0	0.0
Pt	0.0	1.9	0.0	1.0	0.0	1.0	0.0	0.0	0.0	0.0	0.9	0.7
Au	0.0	0.6	0.0	0.3	0.0	1.4	0.0	0.7	0.0	1.1	0.0	0.4
Hg	0.0	0.0	0.0	1.0	*	*	*	*	*	*	*	*

"**prist**" the prstine state without O-adsorption for the embedded (TM)SA (The values in blue follows path-II shown in Figure 6). The La calculations were not converged when the solvation effect was taken into account.

Table S18. Relative electrical conductivity of M-N_nC_m-G_N with respect to 2.1% N-doped graphene [σ(M-N_nC_m-G_N)/σ^o)] at the prsitine state. (RPBE(U)+TS results are given for 3d metals).

Transition Metal	C_4	N_1C_3	N_2C_2	N_2C_2[b]	N_2C_2[c]	N_3C_1	N_4	C_3	N_1C_2	N_2C_1	N_3
Sc	0.434	0.001	0.652	0.618	0.489	0.014	0.585	0.594	0.000	0.605	0.701
Ti	0.052	0.304	0.421	0.365	0.261	0.022	0.285	0.002	0.025	0.487	0.680
V	0.094	0.015	0.664	0.524	0.478	0.007	0.007	0.000	0.000	0.067	1.050
Cr	0.028	0.002	0.671	0.371	0.095	0.476	0.008	0.005	0.000	0.049	0.553
Mn	0.488	0.002	0.713	0.540	0.515	0.006	0.010	0.031	0.000	0.020	0.392
Fe	0.499	0.003	0.734	0.631	0.583	0.006	0.008	0.000	0.000	0.004	0.563
Co	0.479	0.003	0.059	0.549	0.000	0.487	0.009	0.015	0.001	0.001	0.581
Ni	0.750	0.017	0.052	0.002	0.000	0.510	0.008	0.000	0.000	0.001	0.786
Cu	0.810	0.002	0.663	0.602	0.551	0.025	0.007	0.003	0.000	0.000	0.000
Zn	0.033	0.261	0.016	0.013	0.001	0.598	0.007	0.365	0.436	0.000	0.402
Y	0.126	0.001	0.641	0.560	0.449	0.017	0.703	0.559	0.000	0.633	0.726
Zr	0.044	0.450	0.006	0.005	0.014	0.482	0.672	0.000	0.535	0.459	0.192
Nb	0.586	0.001	0.296	0.050	0.389	0.324	0.449	0.008	0.005	0.307	0.538
Mo	0.543	0.002	0.023	0.242	0.172	0.174	0.181	0.000	0.167	0.287	0.006
Tc	0.009	0.002	0.239	0.494	0.001	0.119	0.008	0.014	0.000	0.007	0.565
Ru	0.013	0.220	0.047	0.069	0.002	0.009	0.111	0.000	0.033	0.029	0.973
Rh	0.111	0.001	0.687	0.016	0.021	0.000	0.237	0.007	0.000	0.004	0.032
Pd	0.262	0.364	0.044	0.003	0.000	0.520	0.007	0.000	0.001	0.000	0.000
Ag	0.729	0.001	0.714	0.532	0.562	0.007	0.472	0.003	0.000	0.002	0.000
Cd	0.015	0.588	0.016	0.084	0.090	0.576	0.067	0.000	*	0.818	0.239
La	0.185	0.006	0.671	0.601	0.510	0.019	0.907	0.432	0.000	0.959	0.418
Hf	0.038	0.425	0.012	0.008	0.022	0.292	0.712	0.000	0.519	0.511	0.326
Ta	0.435	0.007	0.126	0.058	0.135	0.232	0.006	0.037	0.046	0.156	0.184
W	0.012	0.011	0.125	0.125	0.081	0.038	0.064	0.000	0.175	0.044	0.000
Re	0.015	0.021	0.240	0.132	0.003	0.091	0.007	0.024	0.001	0.001	0.760
Os	0.010	0.297	0.132	0.367	0.010	0.036	0.004	0.000	0.004	0.042	1.029
Ir	0.362	0.001	0.717	0.368	0.179	0.001	0.239	0.002	0.001	0.133	0.256
Pt	0.025	0.293	0.012	0.000	0.000	0.383	0.007	0.000	0.004	0.000	0.063
Au	0.680	0.001	0.666	0.527	0.475	0.007	0.991	0.001	0.001	0.291	0.001
Hg	0.037	0.542	0.007	*	0.026	*	*	*	*	*	*

"*" denotes unstable systems.

Table S19. (a) Relative electrical conductivity of H@M-N_nC_m-G_N with respect to 2.1% N-doped graphene [σ(H@M-N_nC_m-G_N)/σ^o)] upon H-adorption. (RPBE(U)+TS results are given for 3d metals).

Transition Metal	C_4	N_1C_3	N_2C_2	N_2C_2[b]	N_2C_2[c]	N_3C_1	N_4
Sc	0.002	0.593	0.029	0.043	0.002	0.619	0.016
Ti	0.230	0.016	0.524	0.391	0.411	0.023	0.006
V	0.050	0.503	0.572	0.210	0.417	0.039	0.019
Cr	0.001	0.036	0.476	0.538	0.413	0.014	0.024
Mn	0.008	0.445	0.025	0.013	0.001	0.026	0.025
Fe	0.002	0.529	0.031	0.015	0.001	0.013	0.026
Co	0.004	0.000	0.622	0.532	0.000	0.016	0.021
Ni	0.665	0.000	0.551	0.498	0.446	0.010	0.583
Cu	0.001	0.400	0.005	0.002	0.000	0.143	0.014
Zn	0.223	0.001	0.622	0.570	0.562	0.009	0.821
Y	0.003	0.540	0.032	0.024	0.002	0.609	0.016
Zr	0.280	0.002	0.603	0.441	0.429	0.021	0.925
Nb	0.043	0.474	0.003	0.002	0.004	0.046	0.591
Mo	0.184	0.468	0.002	0.014	0.016	0.019	0.289
Tc	0.561	0.005	0.151	0.004	0.001	0.008	0.013
Ru	0.717	0.000	0.267	0.068	0.000	0.002	0.148
Rh	0.009	0.045	0.003	0.002	0.000	0.320	0.033
Pd	0.432	0.000	0.696	0.572	0.487	0.003	0.625
Ag	0.001	0.555	0.004	0.003	0.000	0.453	0.050
Cd	0.195	0.002	0.004	0.003	0.014	0.001	0.830
La	0.005	0.063	0.011	0.468	0.001	0.575	0.027
Hf	0.760	0.001	0.093	0.456	0.412	0.018	0.843
Ta	0.041	0.495	0.007	0.006	0.019	0.242	0.000
W	0.498	0.354	0.004	0.018	0.017	0.065	0.009
Re	0.670	0.002	0.023	0.072	0.002	0.006	0.018
Os	0.788	0.000	0.370	0.389	0.001	0.003	0.012
Ir	0.007	0.041	0.002	0.005	0.000	0.277	0.013
Pt	0.387	0.000	0.484	0.403	0.385	0.002	0.657
Au	0.000	0.348	0.001	0.003	0.000	0.530	0.007
Hg	0.166	0.001	0.002	*	0.194	*	*

"*" denotes unstable systems.

Table S19. (b) Relative electrical conductivity of O@M-N_nC_m-G_N, HO@M-N_nC_m-G_N, and HOO@M-N_nC_m-G_N with respect to 2.1% N-doped graphene [σ(O@M-N_nC_m-G_N)/σ^o)], [σ(HO@M-N_nC_m-G_N)/σ^o)], and [σ(HOO@M-N_nC_m-G_N)/σ^o)] upon O^-, OH^-, OOH^- adorption]. (RPBE(U)+TS results are given for 3d metals).

TM	C_4			N_1C_3			N_2C_2			N_2C_2[b]			N_2C_2[c]			N_3C_1			N_4		
	*O	*OH	*OOH	*O	*OH	*OOH	*O	*OH	*OOH	*O	*OH	*OOH	*O	*OH	*OOH	*O	*OH	*OOH	*O	*OH	*OOH
Sc	0.100	0.030	0.020	0.004	0.030	0.010	0.008	0.010	0.000	0.010	0.036	0.045	0.059	0.000	0.000	0.193	0.620	0.620	0.385	0.010	0.010
Ti	0.450	0.870	0.900	0.046	0.870	0.000	0.006	0.490	0.510	0.052	0.408	0.454	0.000	0.450	0.370	0.614	0.020	0.030	0.034	0.010	0.010
V	1.060	0.050	0.040	0.003	0.050	0.000	0.573	0.560	0.550	**0.022**	**0.421**	**0.109**	**0.000**	**0.000**	**0.000**	0.568	0.030	0.080	**0.000**	**0.340**	**0.050**
Cr	1.030	0.720	0.120	0.006	0.720	0.040	0.547	0.520	0.500	**0.001**	**0.087**	**0.041**	**0.022**	**0.000**	**0.000**	0.028	0.100	0.110	0.019	0.030	0.030
Mn	0.150	0.120	0.370	0.007	0.120	0.060	0.508	0.280	0.280	0.476	0.367	0.401	0.408	0.050	0.180	0.025	0.340	0.270	0.016	0.130	0.140
Fe	0.040	0.140	0.100	0.056	0.140	0.070	0.491	0.440	0.210	0.330	0.470	0.296	**0.000**	**0.000**	**0.000**	0.424	0.470	0.450	0.068	0.060	0.080
Co	0.020	0.620	0.920	0.137	0.620	0.000	0.182	0.010	0.190	0.399	0.015	0.210	0.131	0.000	0.000	0.295	0.420	0.490	0.483	0.070	0.030
Ni	0.840	0.160	0.900	0.441	0.160	0.010	0.029	0.140	0.160	0.014	0.234	0.173	0.001	0.000	0.000	0.199	0.360	0.340	0.085	0.120	0.080
Cu	0.780	0.600	0.830	0.232	0.600	0.170	0.024	0.020	0.020	0.094	0.007	0.228	0.028	0.000	0.010	0.248	0.630	0.340	0.564	0.580	0.200
Zn	0.660	0.670	0.610	0.001	0.670	0.000	0.007	0.010	0.010	0.004	0.002	0.007	0.000	0.240	0.290	0.013	0.000	0.280	0.791	0.820	0.470
Y	0.230	0.020	0.010	0.003	0.020	0.000	0.004	0.010	0.000	0.004	0.048	0.032	0.621	0.000	0.000	0.453	0.610	0.620	0.362	0.020	0.010
Zr	0.100	0.730	0.720	0.019	0.730	0.000	0.008	0.610	0.580	0.013	0.441	0.445	0.000	0.420	0.410	0.614	0.020	0.030	0.017	0.940	0.870
Nb	0.990	0.040	0.040	0.001	0.040	0.430	0.632	0.000	0.000	0.482	0.003	0.002	**0.467**	**0.000**	**0.000**	**0.016**	**0.320**	**0.370**	**0.531**	**0.090**	**0.030**
Mo	0.040	0.020	0.070	0.374	0.020	0.500	**0.004**	**0.410**	**0.000**	**0.002**	**0.024**	**0.014**	**0.003**	**0.010**	**0.000**	**0.015**	**0.070**	**0.150**	**0.000**	**0.380**	**0.110**
Tc	0.080	0.800	0.820	**0.009**	**0.800**	**0.000**	**0.238**	**0.030**	**0.000**	**0.394**	**0.023**	**0.038**	**0.075**	**0.010**	**0.020**	**0.003**	**0.070**	**0.110**	**0.158**	**0.020**	**0.020**
Ru	0.020	0.990	1.110	0.524	0.990	0.000	**0.005**	**0.410**	**0.030**	**0.013**	**0.279**	**0.230**	**0.001**	**0.140**	**0.210**	**0.003**	**0.000**	**0.000**	**0.000**	**0.550**	**0.830**
Rh	0.830	0.000	0.000	0.002	0.000	0.330	0.004	0.000	0.000	0.005	0.006	0.006	0.000	0.020	0.000	0.018	0.070	0.050	0.022	0.040	0.030
Pd	0.030	0.190	0.330	0.002	0.190	0.000	0.003	0.400	0.370	0.185	0.297	0.196	0.052	0.010	0.000	0.039	0.000	0.010	0.055	0.110	0.110
Ag	0.120	0.010	0.060	0.009	0.010	0.120	0.071	0.020	0.470	0.002	0.002	0.502	0.000	0.000	0.340	0.434	0.670	0.170	0.480	0.080	0.040
Cd	0.010	0.000	0.550	0.009	0.000	0.000	0.003	0.000	0.010	0.004	0.003	0.002	0.052	0.010	0.070	0.001	0.000	0.010	0.397	0.780	0.640
La	0.550	0.010	0.020	0.002	0.010	0.160	0.005	0.000	0.000	0.565	0.035	0.038	0.744	0.000	0.000	0.758	0.640	0.640	0.599	0.020	0.020
Hf	0.470	0.800	0.780	0.182	0.800	0.000	0.010	0.600	0.580	0.010	0.437	0.501	0.000	0.400	0.390	0.589	0.020	0.020	0.015	0.890	0.870
Ta	0.990	0.040	0.040	0.002	0.040	0.440	**0.474**	**0.000**	**0.000**	**0.504**	**0.015**	**0.000**	0.398	0.010	0.010	0.009	0.300	0.290	**0.328**	**0.070**	**0.030**
W	0.040	0.530	0.650	0.452	0.530	0.490	**0.001**	**0.010**	**0.010**	**0.009**	**0.137**	**0.229**	**0.009**	**0.060**	**0.330**	**0.017**	**0.000**	**0.080**	**0.094**	**0.780**	**0.870**
Re	**0.040**	**0.550**	**0.220**	**0.003**	**0.550**	**0.010**	**0.122**	**0.010**	**0.010**	**0.151**	**0.109**	**0.044**	**0.026**	**0.020**	**0.020**	**0.002**	**0.020**	**0.150**	**0.020**	**0.020**	**0.020**
Os	**0.010**	**0.990**	**0.330**	**0.566**	**0.990**	**0.000**	**0.006**	**0.240**	**0.240**	**0.003**	**0.256**	**0.218**	**0.017**	**0.110**	**0.190**	**0.053**	**0.000**	**0.000**	**0.000**	**0.700**	**0.940**
Ir	0.940	0.000	0.000	0.000	0.000	0.280	0.005	0.000	0.000	0.008	0.009	0.008	**0.063**	**0.000**	**0.020**	**0.012**	**0.020**	**0.010**	0.028	0.030	0.020
Pt	0.040	0.110	0.550	0.006	0.110	0.000	0.002	0.490	0.470	0.084	0.500	0.305	0.040	0.430	0.000	0.077	0.000	0.000	0.021	0.050	0.130
Au	0.130	0.550	0.550	0.008	0.550	0.060	0.049	0.010	0.480	0.182	0.010	0.403	0.130	0.000	0.350	0.177	0.530	0.130	0.407	0.300	0.520
Hg	0.690	0.770	0.560	0.001	0.770	0.000	0.031	0.000	0.000	*	*	*	0.001	0.000	0.000	*	*	*	*	*	*

"*" denotes unstable systems.

Table S20. H-adsorption energy [$\Delta E_{H^*} = E_{H@M-G_N} - (E_{M-G_N} + E_{H_2}/2)$ in eV] for M-N_nC_m-G_N. (RPBE(U)+TS results are in parentheses).

Transition Metal	C_4	N_1C_3	N_2C_2	N_2C_2[b]	N_2C_2[c]	N_3C_1	N_4
Sc	-0.70 (-0.62)	-0.10 (-0.05)	0.13 (0.14)	0.06 (0.10)	-0.05 (-0.03)	0.18 (0.07)	-0.53 (-0.66)
Ti	0.08 (0.16)	-0.20 (-0.26)	-0.35 (-0.42)	-0.36 (-0.42)	-0.37 (-0.44)	-0.76 (-0.60)	-0.73 (-0.64)
V	-0.24 (0.06)	-0.30 (0.10)	-0.27 (-0.02)	-0.32 (-0.03)	-0.30 (-0.06)	-0.43 (-0.16)	-0.33 (-0.04)
Cr	-0.11 (-0.30)	-0.56 (-0.72)	-0.05 (-0.77)	-0.56 (-0.75)	-0.19 (-0.73)	-0.30 (-0.50)	0.11 (0.47)
Mn	-1.28 (-1.29)	-0.60 (-0.45)	-0.04 (-0.26)	0.07 (-0.11)	-0.23 (-0.27)	0.03 (-0.12)	0.29 (0.80)
Fe	-0.50 (-1.01)	-0.70 (0.80)	-0.42 (-0.18)	-0.12 (-0.01)	-0.14 (-0.14)	-0.11 (-0.18)	0.20 (0.87)
Co	-0.45 (-0.92)	-0.88 (-0.56)	-0.55 (-0.47)	-0.37 (-0.37)	-0.17 (-0.13)	-0.21 (-0.16)	0.07 (0.53)
Ni	-0.95 (-1.11)	-0.91 (-1.20)	-0.34 (-0.45)	-0.19 (-0.31)	0.10 (0.02)	-0.06 (-0.08)	1.23 (1.21)
Cu	-0.62 (-0.69)	-0.09 (-0.14)	-0.21 (-0.26)	-0.09 (-0.12)	-0.23 (-0.25)	-0.25 (-0.32)	0.92 (1.04)
Zn	-0.15 (-0.21)	-1.05 (-1.17)	-0.42 (-0.52)	-0.81 (-0.90)	-0.72 (-0.78)	-0.61 (-0.62)	0.94 (0.69)
Y	-0.53	-0.07	0.13	0.10	-0.03	0.30	-0.41
Zr	0.18	-0.22	-0.36	-0.37	-0.40	-0.93	-1.12
Nb	-0.48	-0.47	-0.88	-0.83	-0.92	-0.87	-0.96
Mo	-0.35	-0.34	-0.70	-0.65	-0.83	-0.69	-0.58
Tc	-0.02	-0.40	-0.40	-0.43	-0.69	-0.68	-0.62
Ru	0.05	-0.72	-0.18	-0.69	-0.70	-0.78	-0.69
Rh	-0.28	-0.24	-0.40	-0.60	-0.70	-0.49	-0.35
Pd	-0.96	-1.14	-0.58	-0.31	-0.01	-0.04	1.18
Ag	-1.06	-0.33	-0.17	-0.07	-0.21	1.13	0.19
Cd	0.91	-0.13	0.26	-1.30	-0.24	-0.15	-0.27
La	-0.47	1.06	0.11	0.10	-0.02	0.85	0.25
Hf	0.01	-0.56	-0.73	-0.73	-0.80	-1.20	-1.32
Ta	-0.92	-0.99	-1.24	-1.26	-1.29	-1.24	-1.24
W	-0.81	-0.85	-1.09	-1.09	-1.16	-1.09	-1.07
Re	-0.59	-0.86	-0.86	-0.84	-1.03	-1.02	-1.08
Os	-0.28	-1.05	-0.71	-0.99	-1.05	-1.05	-0.89
Ir	-0.46	-0.44	-0.51	-0.73	-1.00	-0.64	-0.48
Pt	-0.43	-0.73	-0.30	0.07	0.23	0.09	1.28
Au	-0.78	-0.12	-0.04	0.04	-0.12	0.36	1.31
Hg	-0.04	0.06	0.42	*	-0.45	*	*

"*" denotes unstable systems.

Table S21. HER free energies ($\Delta G_{H^*} = \Delta E_{H^*} + \Delta ZPE - T\Delta S$ in eV) for M-N_nC_m-G_N. (RPBE(U)+TS results are in parentheses).[a]

Transition Metal	C_4	N_1C_3	N_2C_2	N_2C_2[b]	N_2C_2[c]	N_3C_1	N_4
Sc	-0.31	0.28	0.46	0.44	0.34	0.38	-0.34
	(-0.23)	(0.33)	(0.48)	(0.49)	(0.35)	(0.27)	(-0.47)
Ti	0.43	0.01	-0.12	-0.15	-0.17	-0.54	-0.53
	(0.51)	(-0.05)	(-0.18)	(-0.21)	(-0.23)	(-0.38)	(-0.44)
V	-0.01	-0.09	-0.06	-0.12	-0.10	-0.21	-0.13
	(0.29)	(0.30)	(0.19)	(0.18)	(0.14)	(0.06)	(0.16)
Cr	0.24	-0.21	0.30	-0.21	0.16	0.05	0.32
	(0.05)	(-0.37)	(-0.42)	(-0.40)	(-0.38)	(-0.15)	(0.68)
Mn	-0.90	-0.22	0.29	0.46	0.15	0.37	0.53
	(-0.90)	(-0.06)	(0.06)	(0.27)	(0.11)	(0.22)	(1.04)
Fe	-0.15	-0.34	-0.07	0.23	0.22	0.21	0.44
	(-0.65)	(1.48)	(0.16)	(0.34)	(0.21)	(0.14)	(1.11)
Co	-0.10	-0.53	-0.20	-0.03	0.17	0.12	0.32
	(-0.57)	(-0.21)	(-0.13)	(-0.11)	(0.13)	(0.17)	(0.78)
Ni	-0.60	-0.56	0.00	0.02	0.45	0.27	1.61
	(-0.76)	(-0.84)	(-0.10)	(-0.09)	(0.23)	(0.25)	(1.59)
Cu	-0.27	0.26	0.14	0.25	0.13	0.13	1.31
	(-0.34)	(0.21)	(0.09)	(0.22)	(0.09)	(0.06)	(1.43)
Zn	0.21	-0.69	-0.06	-0.45	-0.36	-0.25	1.15
	(0.15)	(-0.81)	(-0.15)	(-0.54)	(-0.43)	(-0.26)	(0.90)
Y	-0.15	0.31	0.45	0.49	0.35	0.48	-0.22
Zr	0.51	0.00	-0.14	-0.15	-0.18	-0.71	-0.90
Nb	-0.24	-0.25	-0.66	-0.60	-0.70	-0.65	-0.74
Mo	-0.10	-0.12	-0.46	-0.43	-0.61	-0.48	-0.37
Tc	0.20	-0.16	-0.17	-0.18	-0.44	-0.43	-0.36
Ru	0.30	-0.46	0.06	-0.43	-0.44	-0.51	-0.43
Rh	0.06	0.11	-0.06	-0.32	-0.42	-0.19	-0.07
Pd	-0.60	-0.79	-0.25	0.05	0.35	0.29	1.57
Ag	-0.70	0.03	0.17	0.28	0.14	1.49	0.36
Cd	1.09	0.08	0.62	-0.93	0.15	0.06	-0.06
La	-0.09	1.44	0.44	0.48	0.36	1.01	0.43
Hf	0.23	-0.33	-0.50	-0.50	-0.57	-0.98	-1.09
Ta	-0.67	-0.75	-1.00	-1.02	-1.04	-0.99	-1.00
W	-0.56	-0.61	-0.85	-0.84	-0.91	-0.86	-0.82
Re	-0.34	-0.61	-0.61	-0.57	-0.78	-0.78	-0.83
Os	-0.06	-0.82	-0.45	-0.75	-0.82	-0.76	-0.66
Ir	-0.21	-0.17	-0.23	-0.46	-0.73	-0.36	-0.21
Pt	-0.09	-0.39	0.03	0.41	0.57	0.43	1.68
Au	-0.42	0.23	0.30	0.39	0.22	0.71	1.71
Hg	-0.18	0.44	0.78	*	-0.21	*	*

[a] ΔG_{H^*} values for the most stable adsorption site are presented here. The small values of $|\Delta G_{H^*}|$<0.1 eV are marked in red letter dubbed with yellow color. "*" denotes unstable systems.

Table S22. ΔG_{H^*} (in eV) in the absence and presence of water and the solvent effect for selected M-N_2C_2-G_N systems which have $|\Delta G_{H^*}| \leq 0.1$ eV. RPBE(U)+TS results are given for 3d metals. {The results from the explicit solvent models are in braces}.

Transition Metal	ΔG_{H^*}	ΔG_{H^*} in water: implicit(explicit)	Solvent effect
Mn	0.06	-0.03[a] {0.03}	-0.09 {-0.03}
Ni	-0.10	-0.10	0.00
Cu	0.09	0.07	-0.02
Ru	0.06	0.10 {0.09}	0.04 {0.03}
Rh	-0.06	-0.08	-0.02
Pt	0.03	0.04	0.01

[a] In the case of Mn, the difference between ΔG_{H^*}'s without and with implicit solvation models is slightly larger than other cases. This is because the magnetic moment of metal atoms are changed in the implicit solvation model. Thus, we also studied an explicit model which uses an implicit model in the presence of one water molecule added explicitly to each system. Then, in this explicit model, this spin change is not observed and the solvent effect is still negligible. Thus, the real solvent effect is found to be insignificant (no more than 0.03 eV).

Table S23. Volmer–Kubas reaction free energies and adsorption energies (in eV) for selected (M-N_2C_2)-G_N systems. (RPBE(U)+TS results are given in parentheses).

Transition Metal	ΔE_{H^*}	$\Delta E^{Kubas}_{2H^*/2}$	ΔG_{H^*}	$\Delta G^{Kubas}_{2H^*/2}$	$\Delta G_{2nd-H^*} = 2\Delta G^{Kubas}_{2H^*/2} - \Delta G_{H^*}$	H-H (Å)	M-H1 (Å)	M-H2 (Å)
Sc	0.13 (0.14)	-0.07 (-0.07)	0.46 (0.48)	0.12 (0.12)	-0.23 (-0.24)	0.75	2.65	2.67
Ti	-0.35 (-0.42)	-0.10 (-0.08)	-0.12 (-0.18)	0.09 (0.10)	0.30 (0.39)	0.77	2.26	2.23
V	-0.27 (-0.02)	0.10 (0.37)	-0.06 (0.19)	0.31 (0.58)	0.68 (0.97)	1.58	1.64	1.64
Cr	-0.05 (-0.77)	0.22 (-0.03)	0.30 (-0.42)	0.43 (0.18)	0.56 (0.78)	1.48	1.60	1.60
Mn	-0.04 (-0.26)	-0.03 (-0.03)	0.29 (0.06)	0.16 (0.16)	0.02 (0.26)	0.76	2.32	2.33
Fe	-0.42 (-0.18)	-0.04 (-0.03)	-0.07 (0.16)	0.16 (0.18)	0.40 (0.20)	0.79	1.88	1.88
Co	-0.55 (-0.47)	-0.12 (-0.03)	-0.20 (-0.13)	0.09 (0.18)	0.38 (0.48)	0.85	1.65	1.65
Ni	-0.34 (-0.45)	-0.02 (-0.02)	0.00 (-0.10)	0.19 (0.19)	0.39 (0.49)	0.75	3.12	3.84
Cu	-0.21 (-0.26)	-0.02 (-0.02)	0.14 (0.09)	0.19 (0.19)	0.23 (0.28)	0.75	3.29	3.32
Zn	-0.42 (-0.52)	-0.01 (-0.01)	-0.06 (-0.15)	0.18 (0.18)	0.43 (0.52)	0.75	3.40	3.45
Y	0.13	-0.05	0.45	0.17	-0.12	0.75	2.92	2.91
Zr	-0.36	-0.08	-0.14	0.12	0.38	0.76	2.49	2.50
Nb	-0.88	-0.24	-0.66	-0.01	0.63	2.03	1.80	1.77
Mo	-0.70	-0.26	-0.46	-0.02	0.43	1.88	1.72	1.72
Tc	-0.40	-0.42	-0.17	-0.19	-0.21	1.81	1.67	1.67
Ru	-0.18	-0.34	0.06	-0.12	-0.29	1.70	1.63	1.63
Rh	-0.40	0.08	-0.06	0.30	0.65	1.67	1.59	1.59
Pd	-0.58	-0.04	-0.25	0.19	0.63	0.75	3.23	3.96
Ag	-0.17	-0.03	0.17	0.19	0.21	0.75	3.58	3.61
Cd	0.26	-0.01	0.62	0.20	-0.23	0.75	3.45	3.47
La	0.11	-0.05	0.44	0.19	-0.05	0.75	3.31	3.30
Hf	-0.73	-0.08	-0.50	0.12	0.75	0.76	2.49	2.48
Ta	-1.24	-0.44	-1.00	-0.21	0.58	2.17	1.80	1.80
W	-1.09	-0.70	-0.85	-0.44	-0.04	1.93	1.71	1.74
Re	-0.86	-0.80	-0.61	-0.56	-0.50	1.92	1.69	1.69
Os	-0.71	-0.77	-0.45	-0.52	-0.59	1.94	1.66	1.66
Ir	-0.51	-0.29	-0.23	-0.05	0.13	1.94	1.61	1.61
Pt	-0.30	0.11	0.03	0.35	0.67	2.08	1.63	1.63
Au	-0.04	-0.03	0.30	0.20	0.11	0.75	3.75	3.52
Hg	0.42	0.09	0.78	0.31	-0.17	2.30	1.65	1.65

The values of $\Delta G_{2nd\text{-}H^*} < 0$ eV are marked in red letter and the small values of $|\Delta G| < 0.1$ eV are highlighted with yellow color.

Table S24. Adsorption free energies (eV) of OER/ORR inetermediates (OH*, O*, OOH*) for M-N_nC_m-G_N in the absence of solvent. (RPBE(U)+TS results are in parentheses).

TM	C_4			N_1C_3			N_2C_2			N_2C_2[b]			N_2C_2[c]			N_3C_1			N_4		
	ΔG_{OH*}	ΔG_{O*}	ΔG_{OOH*}	ΔG_{OH*}	ΔG_{O*}	ΔG_{OOH*}	ΔG_{OH*}	ΔG_{O*}	ΔG_{OOH*}	ΔG_{OH*}	ΔG_{O*}	ΔG_{OOH*}	ΔG_{OH*}	ΔG_{O*}	ΔG_{OOH*}	ΔG_{OH*}	ΔG_{O*}	ΔG_{OOH*}	ΔG_{OH*}	ΔG_{O*}	ΔG_{OOH*}
Sc	-0.78 (-0.79)	1.01 (1.15)	2.34 (2.40)	-0.63 (-0.67)	1.45 (1.59)	2.52 (2.47)	-0.92 (-0.97)	1.51 (1.56)	2.24 (2.19)	-0.90 (-0.94)	1.51 (1.59)	2.40 (2.29)	-1.11 (-1.15)	0.95 (1.07)	2.13 (2.07)	-1.25 (-1.05)	0.92 (1.27)	1.95 (2.16)	-1.98 (-2.06)	0.04 (0.13)	1.24 (1.16)
Ti	-1.11 (-1.12)	0.30 (0.48)	2.06 (2.08)	-1.59 (-1.65)	-0.30 (-0.19)	1.64 (1.59)	-1.74 (-1.77)	-0.97 (-0.87)	1.44 (1.43)	-1.73 (-1.77)	-0.97 (-0.85)	1.55 (1.50)	-1.72 (-1.80)	-1.13 (-0.98)	1.69 (1.54)	-2.13 (-1.85)	-1.75 (-1.29)	1.35 (1.42)	-2.09 (-2.04)	-2.40 (-2.06)	1.34 (1.21)
V	-1.38 (-1.08)	-1.02 (-0.63)	1.84 (2.19)	-1.46 (-1.20)	-1.57 (-1.08)	1.81 (2.07)	-1.61 (-1.31)	-1.63 (-0.92)	1.78 (2.05)	**0.84 (0.41)**	**0.29 (0.44)**	**3.03 (3.63)**	**0.73 (0.51)**	**0.59 (0.60)**	**3.42 (3.72)**	-1.75 (-0.87)	-1.82 (-0.73)	1.65 (2.56)	**2.79 (0.65)**	**3.19 (2.15)**	**4.23 (3.41)**
Cr	-0.93 (-0.43)	-0.82 (-0.15)	2.31 (2.84)	-1.09 (-0.56)	-1.24 (-0.33)	2.16 (2.71)	-0.83 (-0.41)	-1.11 (-0.19)	2.31 (2.76)	**0.98 (0.26)**	**1.07 (1.07)**	**4.24 (3.51)**	**1.14 (0.43)**	**1.43 (1.44)**	**4.36 (3.67)**	-0.67 (0.62)	-0.94 (0.93)	2.43 (3.79)	-0.17 (0.19)	-0.32 (0.95)	2.92 (3.31)
Mn	-0.92 (-0.01)	-0.76 (0.71)	2.30 (3.23)	-0.15 (0.44)	0.13 (1.80)	3.00 (3.63)	-0.11 (0.44)	0.13 (1.71)	3.06 (3.60)	0.08 (0.51)	0.29 (1.89)	3.16 (3.65)	0.05 (0.32)	0.34 (1.87)	3.25 (3.48)	-0.03 (0.26)	0.34 (1.86)	3.14 (3.45)	0.37 (0.24)	0.82 (1.86)	3.53 (3.43)
Fe	-0.04 (0.11)	0.21 (1.49)	3.12 (3.31)	0.18 (0.42)	0.44 (1.84)	3.25 (3.96)	0.02 (0.58)	0.54 (1.73)	3.00 (3.80)	0.27 (0.60)	0.84 (2.02)	3.41 (3.89)	**0.59 (-0.55)**	**1.79 (1.11)**	**3.77 (2.87)**	0.19 (0.78)	0.94 (2.28)	3.40 (3.96)	0.56 (0.70)	1.17 (1.95)	3.44 (3.91)
Co	0.18 (0.57)	1.04 (1.71)	3.16 (3.71)	0.21 (0.84)	1.08 (1.92)	3.18 (3.90)	0.55 (0.79)	1.43 (1.98)	3.36 (3.82)	0.61 (0.84)	1.50 (1.99)	3.67 (3.92)	0.49 (0.68)	1.53 (1.89)	3.54 (3.74)	0.77 (1.36)	1.68 (2.51)	3.66 (4.34)	0.98 (1.37)	2.31 (2.31)	4.08 (4.31)
Ni	0.88 (0.86)	2.23 (2.25)	3.79 (3.79)	1.28 (1.13)	2.81 (2.14)	4.18 (4.06)	1.58 (1.61)	3.10 (3.27)	4.44 (4.52)	1.59 (1.56)	3.22 (3.35)	4.47 (4.49)	1.58 (1.50)	3.17 (3.27)	4.43 (4.48)	1.56 (1.86)	3.45 (3.25)	4.45 (4.77)	1.95 (2.02)	3.97 (4.04)	4.71 (4.77)
Cu	1.99 (2.04)	3.92 (4.04)	4.82 (4.86)	2.16 (2.24)	4.13 (4.24)	4.86 (4.86)	1.72 (1.77)	3.79 (3.86)	4.62 (4.65)	1.82 (1.89)	4.02 (4.11)	4.68 (4.72)	1.67 (1.74)	3.78 (3.99)	4.63 (4.68)	1.74 (2.09)	3.88 (4.26)	4.64 (4.98)	1.76 (1.82)	3.64 (3.89)	4.64 (4.67)
Zn	1.66 (1.56)	3.82 (3.74)	4.53 (4.47)	1.01 (0.96)	3.25 (3.21)	4.15 (4.13)	1.46 (1.44)	3.57 (3.56)	4.52 (4.49)	0.94 (0.93)	3.26 (3.26)	4.10 (4.07)	0.93 (0.89)	0.19 (0.11)	4.12 (4.09)	0.69 (1.52)	2.93 (3.79)	3.90 (4.70)	0.56 (0.65)	3.03 (2.99)	3.78 (3.87)
Y	-0.76	1.03	2.31	-0.65	1.50	2.55	-0.76	1.62	2.39	-0.78	1.62	2.54	-1.03	1.13	2.15	-1.12	1.19	2.09	-1.85	0.47	1.38
Zr	-1.02	0.58	2.11	-1.53	0.01	1.65	-1.70	-0.50	1.49	-1.73	-0.58	1.52	-1.76	-0.70	1.53	-2.33	-1.41	0.93	-2.57	-2.32	0.67
Nb	-1.55	-0.90	1.62	-1.59	-1.46	1.61	-2.12	-2.06	0.96	-2.07	-2.09	1.19	**0.46**	**0.38**	**3.70**	**0.89**	**0.30**	**4.30**	**-0.16**	**0.93**	**2.85**
Mo	-1.27	-1.40	1.95	-1.34	-1.24	2.03	**1.78**	**1.28**	**3.79**	**1.29**	**0.85**	**4.66**	**1.22**	**1.12**	**4.53**	**1.64**	**1.37**	**4.09**	**0.73**	**3.14**	**3.89**
Tc	-0.63	-0.80	2.56	**1.79**	**1.44**	**4.43**	**1.87**	**2.08**	**4.49**	**1.55**	**1.67**	**4.87**	**1.60**	**1.94**	**4.85**	**1.66**	**1.86**	**4.58**	**1.40**	**3.39**	**4.35**
Ru	-0.34	-0.24	2.82	-0.88	-0.65	2.09	**2.08**	**2.45**	**4.69**	**1.03**	**1.98**	**4.23**	**1.23**	**2.46**	**4.39**	**0.97**	**1.98**	**4.09**	**2.41**	**2.70**	**3.65**
Rh	-0.06	0.83	3.02	0.42	1.03	3.49	0.31	1.38	3.26	0.33	1.21	3.30	0.84	1.27	3.25	0.81	1.81	3.72	0.96	2.37	3.90
Pd	1.06	2.40	4.02	1.08	2.78	4.04	1.65	3.47	4.52	1.86	3.57	4.70	1.92	3.61	4.71	1.94	3.88	4.78	2.27	4.31	4.88
Ag	1.71	3.74	4.65	2.08	4.19	4.83	1.92	4.05	4.74	1.83	4.02	4.82	1.60	3.71	4.72	2.00	3.73	4.74	0.70	2.73	3.74
Cd	1.24	3.47	4.28	0.18	2.53	3.54	0.83	3.15	4.15	0.20	2.49	3.47	0.11	2.38	3.37	0.10	2.44	3.40	0.16	2.63	3.42
La	-0.57	0.29	2.62	-0.39	0.81	2.80	-0.31	1.40	2.84	-0.35	1.42	2.88	-0.59	0.51	2.61	-0.68	0.96	2.48	-1.32	0.34	1.87
Hf	-1.43	0.46	1.79	-1.97	-0.20	1.28	-2.14	-0.74	1.09	-2.18	-0.83	1.19	-2.23	-0.96	1.14	-2.65	-1.51	0.65	-2.79	-2.32	0.53
Ta	-2.07	-1.32	1.40	-2.16	-1.92	1.18	**-0.11**	**0.59**	**3.37**	**0.42**	**0.17**	**2.11**	-2.53	-2.47	0.93	**0.91**	**0.47**	**4.13**	**-0.46**	**1.03**	**2.69**
W	-1.67	-1.96	1.55	-1.87	-1.99	1.58	**1.88**	**1.22**	**3.32**	**1.23**	**0.77**	**3.52**	**0.33**	**0.36**	**3.59**	**1.66**	**1.46**	**4.26**	**0.17**	**0.12**	**3.42**
Re	**1.50**	**1.44**	**4.43**	**2.31**	**1.42**	**4.51**	**0.83**	**2.17**	**3.98**	**1.66**	**1.67**	**4.43**	**1.03**	**0.91**	**1.27**	**2.21**	**2.37**	**5.17**	**2.41**	**3.44**	**4.27**
Os	**1.85**	**2.25**	**4.91**	**1.75**	**2.30**	**4.74**	**2.26**	**2.79**	**4.51**	**1.61**	**2.44**	**4.87**	**1.65**	**1.97**	**4.97**	**1.37**	**2.07**	**4.61**	**2.56**	**2.91**	**3.66**
Ir	-0.46	-0.18	2.59	0.13	0.23	3.13	0.06	0.72	2.98	0.31	0.82	3.20	**1.74**	**2.41**	**4.97**	0.78	1.47	3.71	1.04	2.07	3.97
Pt	1.01	1.99	4.23	1.05	2.47	4.07	1.52	3.04	4.45	1.88	3.36	4.79	1.88	3.31	4.76	1.85	3.83	4.79	2.30	4.19	4.90
Au	0.79	3.95	4.80	2.35	4.02	4.88	2.17	4.28	4.86	2.16	4.23	4.87	1.99	3.74	4.81	2.26	4.41	4.82	2.02	4.20	4.62
Hg	1.98	4.13	4.59	1.13	3.26	4.31	-0.29	1.93	3.82	*	*	*	0.33	2.20	3.64	*	*	*	*	*	*

The SAC systems with blue marked values have unstable *OOH reaction intermediate and these are calculated by associative pathway-Path-II shown in Figure 6.

Table S25. Adsorption free energies (eV) of OER/ORR inetermediates (OH*, O*, OOH*) for M-N_nC_m-G_N in the presence of solvent (dielectric constant: 80)**.** (RPBE(U)+TS results are in parentheses).

TM	C_4			N_1C_3			N_2C_2			N_2C_2[b]			N_2C_2[c]			N_3C_1			N_4		
	ΔG_{OH*}	ΔG_{O*}	ΔG_{OOH*}	ΔG_{OH*}	ΔG_{O*}	ΔG_{OOH*}	ΔG_{OH*}	ΔG_{O*}	ΔG_{OOH*}	ΔG_{OH*}	ΔG_{O*}	ΔG_{OOH*}	ΔG_{OH*}	ΔG_{O*}	ΔG_{OOH*}	ΔG_{OH*}	ΔG_{O*}	ΔG_{OOH*}	ΔG_{OH*}	ΔG_{O*}	ΔG_{OOH*}
Sc	0.27 (0.31)	0.24 (0.26)	3.52 (3.69)	0.46 (0.54)	1.59 (1.61)	3.60 (3.66)	0.46 (0.53)	2.33 (2.47)	3.63 (3.74)	0.53 (0.56)	2.31 (2.49)	3.61 (3.70)	0.26 (0.27)	1.32 (1.47)	3.31 (3.67)	0.32 (0.40)	1.98 (2.18)	3.39 (3.51)	-0.20 (-0.10)	1.40 (1.74)	2.89 (3.03)
Ti	0.44 (0.67)	1.64 (2.03)	3.50 (3.75)	-0.05 (0.13)	1.04 (1.35)	3.07 (3.25)	-0.15 (0.02)	0.39 (0.67)	2.89 (3.07)	-0.11 (0.00)	0.42 (0.68)	2.96 (3.15)	-0.17 (-0.04)	0.23 (0.51)	3.14 (3.27)	-0.67 (-0.56)	-0.46 (-0.19)	2.43 (2.57)	-0.77 (-0.35)	-1.14 (-0.50)	2.65 (2.74)
V	-0.46 (-0.16)	-0.14 (0.26)	2.69 (3.05)	-0.53 (-0.22)	-0.63 (-0.10)	2.65 (2.96)	-0.60 (-0.34)	-0.60 (0.01)	2.67 (2.92)	**0.78 (0.38)**	**0.32 (0.32)**	**2.61 (3.49)**	**0.77 (0.44)**	**0.63 (0.50)**	**2.89 (3.65)**	-1.09 (-0.58)	-1.25 (-0.55)	2.27 (2.73)	**1.32 (1.72)**	**3.29 (3.32)**	**4.16 (4.63)**
Cr	-0.43 (0.07)	-0.30 (0.35)	2.76 (3.28)	-0.74 (-0.37)	-0.85 (-0.10)	2.45 (2.84)	-0.61 (-0.24)	-0.85 (0.04)	2.47 (2.90)	**0.91 (0.14)**	**1.06 (0.92)**	**4.06 (3.26)**	**1.09 (0.35)**	**1.44 (1.37)**	**4.22 (3.53)**	-0.73 (-0.27)	-0.93 (0.13)	2.34 (2.84)	-0.37 (1.04)	-0.41 (0.82)	2.66 (3.03)
Mn	-0.31 (-0.13)	-0.08 (0.65)	2.85 (3.04)	-0.24 (0.34)	0.11 (1.76)	2.89 (3.49)	-0.17 (0.31)	0.13 (1.73)	2.95 (3.44)	0.01 (0.40)	0.29 (1.92)	3.00 (3.50)	-0.05 (0.21)	0.36 (1.91)	3.03 (3.34)	-0.20 (0.10)	0.31 (1.86)	2.92 (3.24)	0.18 (0.02)	0.73 (1.68)	3.29 (3.15)
Fe	-0.13 (0.02)	0.22 (1.48)	3.01 (3.18)	0.01 (0.19)	0.34 (1.67)	3.05 (3.73)	-0.13 (0.42)	0.37 (1.54)	2.72 (3.54)	0.10 (0.40)	0.70 (1.70)	3.25 (3.58)	**0.51 (-0.47)**	**1.58 (1.03)**	**3.64 (2.87)**	-0.01 (0.06)	0.76 (1.50)	3.16 (3.21)	0.38 (0.24)	0.91 (1.64)	3.20 (3.62)
Co	0.09 (0.44)	0.94 (1.53)	3.01 (3.54)	0.07 (0.68)	0.91 (1.72)	2.99 (3.67)	0.34 (0.57)	1.22 (1.69)	3.14 (3.55)	0.44 (0.62)	1.32 (1.73)	3.45 (3.64)	0.45 (-0.07)	1.30 (1.08)	3.32 (2.99)	0.58 (0.03)	1.48 (1.15)	3.43 (2.97)	0.75 (0.38)	2.00 (1.12)	3.60 (3.32)
Ni	0.70 (0.75)	2.04 (2.10)	3.58 (3.62)	1.11 (1.08)	2.62 (2.07)	3.98 (3.97)	1.37 (1.31)	2.91 (3.06)	4.22 (4.22)	1.37 (1.30)	3.02 (3.14)	4.22 (4.21)	1.35 (1.30)	2.98 (3.05)	4.20 (4.20)	1.27 (1.37)	3.15 (2.76)	4.14 (4.26)	1.61 (1.68)	3.40 (3.74)	4.38 (4.46)
Cu	1.73 (1.77)	3.69 (3.81)	4.57 (4.60)	1.84 (1.93)	3.86 (3.98)	4.60 (4.61)	1.35 (1.39)	3.43 (3.49)	4.27 (4.30)	1.47 (1.52)	3.65 (3.73)	4.32 (4.35)	1.29 (1.34)	3.44 (3.53)	4.26 (4.30)	1.42 (1.43)	3.45 (3.61)	4.31 (4.31)	1.33 (1.37)	3.25 (3.48)	4.29 (4.32)
Zn	1.34 (1.25)	3.51 (3.42)	4.30 (4.24)	0.70 (0.66)	2.92 (2.89)	3.81 (3.79)	1.11 (1.09)	3.18 (3.18)	4.19 (4.18)	0.70 (0.67)	2.94 (2.93)	3.81 (3.78)	0.61 (0.57)	-0.03 (-0.09)	3.80 (3.76)	0.37 (0.44)	2.55 (2.65)	3.55 (3.62)	0.21 (0.31)	2.44 (2.89)	3.40 (3.51)
Y	-0.04	0.30	3.30	0.21	1.48	3.36	0.44	2.23	3.47	0.46	2.28	3.59	0.24	1.24	3.32	0.26	2.12	3.45	-0.14	1.61	3.03
Zr	0.60	1.12	3.68	0.08	1.27	3.17	-0.02	0.87	3.19	0.05	1.25	3.24	-0.08	0.71	3.05	-0.45	0.27	2.64	-0.84	-0.67	2.23
Nb	-0.18	0.37	2.90	-0.36	-0.12	2.78	-0.91	-0.76	2.25	-0.77	-0.80	2.25	**0.58**	**0.54**	**3.83**	**0.99**	**0.47**	**4.30**	**-0.15**	**0.44**	**2.74**
Mo	-0.56	-0.62	2.60	-0.68	-0.52	2.42	**1.63**	**1.31**	**4.71**	**1.23**	**1.02**	**4.44**	**1.19**	**1.16**	**4.35**	**1.57**	**1.40**	**4.79**	**0.77**	**3.07**	**3.69**
Tc	-0.41	-0.49	2.75	**1.65**	**1.42**	**4.22**	**1.71**	**2.02**	**4.82**	**1.45**	**1.67**	**4.60**	**1.54**	**1.96**	**4.67**	**1.92**	**2.19**	**4.49**	**1.21**	**3.27**	**4.06**
Ru	-0.31	-0.13	2.60	-0.84	-0.55	1.98	**1.88**	**2.17**	**5.01**	**0.92**	**1.91**	**3.98**	**1.14**	**2.44**	**4.36**	**0.91**	**1.87**	**3.88**	**1.27**	**3.12**	**3.68**
Rh	-0.17	0.72	2.82	0.27	0.89	3.31	0.10	1.21	3.03	0.16	1.00	3.02	0.66	1.07	3.13	0.67	1.68	3.55	0.79	2.13	3.68
Pd	0.88	2.20	3.76	0.87	2.57	3.81	1.43	3.26	4.28	1.66	3.28	4.46	1.68	3.40	4.46	1.62	3.57	4.46	1.91	3.94	4.53
Ag	1.44	3.47	4.39	1.76	3.77	4.55	1.47	3.62	4.38	1.59	3.72	4.46	1.26	3.40	4.32	1.54	3.20	4.39	0.24	2.28	3.30
Cd	0.97	2.84	4.06	-0.10	2.18	3.17	0.60	2.87	3.84	0.22	2.38	3.44	-0.61	1.44	2.64	0.06	2.34	3.28	0.05	2.25	3.27
La	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Hf	0.47	1.30	3.56	-0.06	1.21	3.11	-0.20	0.85	3.16	-0.15	0.97	3.03	-0.24	0.66	2.91	-0.68	0.25	2.47	-1.06	-0.63	2.16
Ta	-0.40	0.24	2.93	-0.62	-0.39	2.61	**1.38**	**0.62**	**4.61**	**0.41**	**0.22**	**3.74**	-1.21	-1.17	2.11	**0.91**	**0.63**	**4.36**	**-0.59**	**0.47**	**2.53**
W	-0.95	-1.16	2.17	-1.09	-1.14	-0.73	**1.73**	**1.27**	**4.79**	**1.23**	**0.84**	**3.06**	**0.44**	**0.50**	**3.62**	**1.64**	**1.50**	**3.61**	**0.30**	**-0.17**	**3.29**
Re	**1.64**	**1.45**	**4.77**	**2.10**	**1.41**	**4.33**	**1.83**	**2.17**	**4.94**	**1.59**	**1.70**	**4.20**	**1.02**	**0.94**	**3.18**	**2.12**	**2.38**	**4.67**	**1.13**	**3.33**	**2.96**
Os	**1.64**	**2.25**	**4.81**	**1.61**	**2.27**	**4.42**	**2.08**	**2.48**	**4.58**	**1.49**	**2.41**	**4.63**	**1.60**	**1.99**	**4.89**	**1.28**	**1.82**	**4.14**	**0.93**	**2.92**	**3.58**
Ir	-0.61	-0.29	2.41	-0.03	0.11	2.53	-0.12	0.56	2.78	0.19	0.69	2.89	**1.64**	**2.17**	**4.77**	0.66	1.35	3.55	0.88	1.84	3.78
Pt	0.85	1.86	4.04	0.88	2.30	3.86	1.35	2.75	4.23	1.71	3.18	4.56	1.70	3.14	4.53	1.64	3.37	4.51	2.00	3.71	4.55
Au	0.62	3.72	4.50	2.00	3.70	4.54	1.73	3.90	4.50	1.80	3.92	4.51	1.58	3.43	4.45	1.79	3.99	4.47	1.41	3.63	4.25
Hg	1.58	3.80	4.34	0.92	2.97	4.02	-0.55	1.58	2.72	*	*	*	0.27	1.68	3.32	*	*	*	*	*	*

The SAC systems with blue marked values have unstable *OOH reaction intermediate and these are calculated by associative pathway-Path-II shown in Figure 6. The data for the La case in solvent are not provided due to the convergency issue.

Table S26. Reaction free energy (eV) of elementary steps for OER/ORR in the absence of solvent for M-N_nC_m-G_N DV-systems. (RPBE(U)+TS results are in parentheses). (*OOH is unstable, breaking into *O and *OH in path-I; so, for these systems the OER/ORR reaction free energies marked in blue are calculated for path-II shown in Figure 6).

TM	C_4								N_1C_3								N_2C_2							
	OER				ORR				OER				ORR				OER				ORR			
	ΔG_1	ΔG_2	ΔG_3	ΔG_4	ΔG_5	ΔG_6	ΔG_7	ΔG_8	ΔG_1	ΔG_2	ΔG_3	ΔG_4	ΔG_5	ΔG_6	ΔG_7	ΔG_8	ΔG_1	ΔG_2	ΔG_3	ΔG_4	ΔG_5	ΔG_6	ΔG_7	ΔG_8
Sc	-0.78 (-0.79)	1.79 (1.94)	1.33 (1.24)	2.58 (2.52)	-2.58 (-2.52)	-1.33 (-1.24)	-1.79 (-1.94)	0.78 (0.79)	-0.63 (-0.67)	2.08 (2.26)	1.07 (0.89)	2.40 (2.45)	-2.40 (-2.45)	-1.07 (-0.89)	-2.08 (-2.26)	0.63 (0.67)	-0.92 (-0.97)	2.43 (2.54)	0.73 (0.63)	2.68 (2.73)	-2.68 (-2.73)	-0.73 (-0.63)	-2.43 (-2.54)	0.92 (0.97)
Ti	-1.11 (-1.12)	1.41 (1.60)	1.76 (1.61)	2.86 (2.84)	-2.86 (-2.84)	-1.76 (-1.61)	-1.41 (-1.60)	1.11 (1.12)	-1.59 (-1.65)	1.29 (1.45)	1.93 (1.78)	3.28 (3.33)	-3.28 (-3.33)	-1.93 (-1.78)	-1.29 (-1.45)	1.59 (1.65)	-1.74 (-1.77)	0.77 (0.90)	2.42 (2.31)	3.48 (3.49)	-3.48 (-3.49)	-2.42 (-2.31)	-0.77 (-0.90)	1.74 (1.77)
V	-1.38 (-1.08)	0.35 (0.45)	2.86 (2.81)	3.08 (2.73)	-3.08 (-2.73)	-2.86 (-2.81)	-0.35 (-0.45)	1.38 (1.08)	-1.46 (-1.20)	-0.12 (0.12)	3.39 (3.15)	3.11 (2.85)	-3.11 (-2.85)	-3.39 (-3.15)	0.12 (-0.12)	1.46 (1.20)	-1.61 (-1.31)	-0.02 (0.40)	3.41 (2.97)	3.14 (2.87)	-3.14 (-2.87)	-3.41 (-2.97)	0.02 (-0.40)	1.61 (1.31)
Cr	-0.93 (-0.43)	0.12 (0.29)	3.12 (2.99)	2.61 (2.08)	-2.61 (-2.08)	-3.12 (-2.99)	-0.12 (-0.29)	0.93 (0.43)	-1.09 (-0.56)	-0.15 (0.23)	3.40 (3.04)	2.76 (2.21)	-2.76 (-2.21)	-3.40 (-3.04)	0.15 (-0.23)	1.09 (0.56)	-0.83 (-0.41)	-0.28 (0.22)	3.42 (2.94)	2.61 (2.16)	-2.61 (-2.16)	-3.42 (-2.94)	0.28 (-0.22)	0.83 (0.41)
Mn	-0.92 (-0.01)	0.16 (0.72)	3.06 (2.52)	2.62 (1.69)	-2.62 (-1.69)	-3.06 (-2.52)	-0.16 (-0.72)	0.92 (0.01)	-0.15 (0.44)	0.28 (1.36)	2.87 (1.83)	1.92 (1.29)	-1.92 (-1.29)	-2.87 (-1.83)	-0.28 (-1.36)	0.15 (-0.44)	-0.11 (0.44)	0.24 (1.26)	2.93 (1.89)	1.86 (1.32)	-1.86 (-1.32)	-2.93 (-1.89)	-0.24 (-1.26)	0.11 (-0.44)
Fe	-0.04 (0.11)	0.26 (1.38)	2.91 (1.82)	1.80 (1.61)	-1.80 (-1.61)	-2.91 (-1.82)	-0.26 (-1.38)	0.04 (-0.11)	0.18 (0.42)	0.26 (1.42)	2.81 (2.13)	1.67 (0.94)	-1.67 (-0.94)	-2.81 (-2.13)	-0.26 (-1.42)	-0.18 (-0.42)	0.02 (0.58)	0.52 (1.15)	2.47 (2.08)	1.92 (1.12)	-1.92 (-1.12)	-2.47 (-2.08)	-0.52 (-1.15)	-0.02 (-0.58)
Co	0.18 (0.57)	0.86 (1.13)	2.12 (2.01)	1.76 (1.21)	-1.76 (-1.21)	-2.12 (-2.01)	-0.86 (-1.13)	-0.18 (-0.57)	0.21 (0.84)	0.87 (1.08)	2.11 (1.98)	1.74 (1.02)	-1.74 (-1.02)	-2.11 (-1.98)	-0.87 (-1.08)	-0.21 (-0.84)	0.55 (0.79)	0.87 (1.19)	1.93 (1.84)	1.56 (1.10)	-1.56 (-1.10)	-1.93 (-1.84)	-0.87 (-1.19)	-0.55 (-0.79)
Ni	0.88 (0.86)	1.35 (1.39)	1.56 (1.54)	1.13 (1.13)	-1.13 (-1.13)	-1.56 (-1.54)	-1.35 (-1.39)	-0.88 (-0.86)	1.28 (1.13)	1.53 (1.00)	1.37 (1.92)	0.74 (0.86)	-0.74 (-0.86)	-1.37 (-1.92)	-1.53 (-1.00)	-1.28 (-1.13)	1.58 (1.61)	1.51 (1.65)	1.35 (1.25)	0.48 (0.40)	-0.48 (-0.40)	-1.35 (-1.25)	-1.51 (-1.65)	-1.58 (-1.61)
Cu	1.99 (2.04)	1.93 (2.01)	0.91 (0.81)	0.10 (0.06)	-0.10 (-0.06)	-0.91 (-0.81)	-1.93 (-2.01)	-1.99 (-2.04)	2.16 (2.24)	1.97 (2.00)	0.73 (0.62)	0.06 (0.06)	-0.06 (-0.06)	-0.73 (-0.62)	-1.97 (-2.00)	-2.16 (-2.24)	1.72 (1.77)	2.07 (2.09)	0.83 (0.79)	0.30 (0.27)	-0.30 (-0.27)	-0.83 (-0.79)	-2.07 (-2.09)	-1.72 (-1.77)
Zn	1.66 (1.56)	2.16 (2.19)	0.71 (0.73)	0.39 (0.45)	-0.39 (-0.45)	-0.71 (-0.73)	-2.16 (-2.19)	-1.66 (-1.56)	1.01 (0.96)	2.24 (2.25)	0.91 (0.92)	0.77 (0.79)	-0.77 (-0.79)	-0.91 (-0.92)	-2.24 (-2.25)	-1.01 (-0.96)	1.46 (1.44)	2.11 (2.13)	0.95 (0.92)	0.40 (0.43)	-0.40 (-0.43)	-0.95 (-0.92)	-2.11 (-2.13)	-1.46 (-1.44)
Y	-0.76	1.79	1.28	2.61	-2.61	-1.28	-1.79	0.76	-0.65	2.15	1.05	2.37	-2.37	-1.05	-2.15	0.65	-0.76	2.38	0.78	2.53	-2.53	-0.78	-2.38	0.76
Zr	-1.02	1.60	1.52	2.81	-2.81	-1.52	-1.60	1.02	-1.53	1.54	1.64	3.27	-3.27	-1.64	-1.54	1.53	-1.70	1.20	1.99	3.43	-3.43	-1.99	-1.20	1.70
Nb	-1.55	0.65	2.52	3.30	-3.30	-2.52	-0.65	1.55	-1.59	0.13	3.07	3.31	-3.31	-3.07	-0.13	1.59	-2.12	0.05	3.02	3.96	-3.96	-3.02	-0.05	2.12
Mo	-1.27	-0.13	3.36	2.97	-2.97	-3.36	0.13	1.27	-1.34	0.09	3.27	2.89	-2.89	-3.27	-0.09	1.34	**1.78**	**-0.50**	**2.51**	**1.13**	**-1.13**	**-2.51**	**0.50**	**-1.78**
Tc	-0.63	-0.17	3.36	2.36	-2.36	-3.36	0.17	0.63	**1.79**	**-0.35**	**2.99**	**0.49**	**-0.49**	**-2.99**	**0.35**	**-1.79**	**1.87**	**0.21**	**2.42**	**0.43**	**-0.43**	**-2.42**	**-0.21**	**-1.87**
Ru	-0.34	0.09	3.07	2.10	-2.10	-3.07	-0.09	0.34	-0.88	0.23	2.74	2.83	-2.83	-2.74	-0.23	0.88	**2.08**	**0.37**	**2.24**	**0.23**	**-0.23**	**-2.24**	**-0.37**	**-2.08**
Rh	-0.06	0.89	2.19	1.90	-1.90	-2.19	-0.89	0.06	0.42	0.61	2.46	1.43	-1.43	-2.46	-0.61	-0.42	0.31	1.08	1.88	1.66	-1.66	-1.88	-1.08	-0.31
Pd	1.06	1.34	1.62	0.90	-0.90	-1.62	-1.34	-1.06	1.08	1.70	1.27	0.88	-0.88	-1.27	-1.70	-1.08	1.65	1.82	1.05	0.40	-0.40	-1.05	-1.82	-1.65
Ag	1.71	2.03	0.91	0.27	-0.27	-0.91	-2.03	-1.71	2.08	2.11	0.64	0.09	-0.09	-0.64	-2.11	-2.08	1.92	2.13	0.68	0.18	-0.18	-0.68	-2.13	-1.92
Cd	1.24	2.24	0.81	0.64	-0.64	-0.81	-2.24	-1.24	0.18	2.35	1.01	1.38	-1.38	-1.01	-2.35	-0.18	0.83	2.32	1.00	0.77	-0.77	-1.00	-2.32	-0.83
La	-0.57	0.87	2.33	2.30	-2.30	-2.33	-0.87	0.57	-0.39	1.20	1.99	2.12	-2.12	-1.99	-1.20	0.39	-0.31	1.71	1.44	2.08	-2.08	-1.44	-1.71	0.31
Hf	-1.43	1.88	1.33	3.13	-3.13	-1.33	-1.88	1.43	-1.97	1.77	1.49	3.64	-3.64	-1.49	-1.77	1.97	-2.14	1.40	1.83	3.83	-3.83	-1.83	-1.40	2.14
Ta	-2.07	0.75	2.73	3.52	-3.52	-2.73	-0.75	2.07	-2.16	0.24	3.10	3.74	-3.74	-3.10	-0.24	2.16	**-0.11**	**0.70**	**2.78**	**1.55**	**-1.55**	**-2.78**	**-0.70**	**0.11**
W	-1.67	-0.28	3.51	3.37	-3.37	-3.51	0.28	1.67	-1.87	-0.12	3.57	3.34	-3.34	-3.57	0.12	1.87	**1.88**	**-0.66**	**2.10**	**1.60**	**-1.60**	**-2.10**	**0.66**	**-1.88**
Re	**1.50**	**-0.06**	**2.99**	**0.49**	**-0.49**	**-2.99**	**0.06**	**-1.50**	**2.31**	**-0.89**	**3.09**	**0.41**	**-0.41**	**-3.09**	**0.89**	**-2.31**	**0.83**	**1.34**	**1.81**	**0.94**	**-0.94**	**-1.81**	**-1.34**	**-0.83**
Os	**1.85**	**0.40**	**2.66**	**0.01**	**-0.01**	**-2.66**	**-0.40**	**-1.85**	**1.75**	**0.55**	**2.44**	**0.18**	**-0.18**	**-2.44**	**-0.55**	**-1.75**	**2.26**	**0.52**	**1.72**	**0.41**	**-0.41**	**-1.72**	**-0.52**	**-2.26**
Ir	-0.46	0.29	2.77	2.33	-2.33	-2.77	-0.29	0.46	0.13	0.10	2.90	1.79	-1.79	-2.90	-0.10	-0.13	0.06	0.66	2.26	1.94	-1.94	-2.26	-0.66	-0.06
Pt	1.01	0.98	2.25	0.69	-0.69	-2.25	-0.98	-1.01	1.05	1.42	1.60	0.85	-0.85	-1.60	-1.42	-1.05	1.52	1.52	1.40	0.47	-0.47	-1.40	-1.52	-1.52
Au	0.79	3.16	0.84	0.12	-0.12	-0.84	-3.16	-0.79	2.35	1.67	0.86	0.04	-0.04	-0.86	-1.67	-2.35	2.17	2.11	0.58	0.06	-0.06	-0.58	-2.11	-2.17
Hg	1.98	2.15	0.46	0.33	-0.33	-0.46	-2.15	-1.98	1.13	2.13	1.05	0.61	-0.61	-1.05	-2.13	-1.13	-0.29	2.22	1.89	1.10	-1.10	-1.89	-2.22	0.29

Table S26. Continued...

TM	N_2C_2[b]								N_2C_2[c]								N_3C_1							
	OER				ORR				OER				ORR				OER				ORR			
	ΔG_1	ΔG_2	ΔG_3	ΔG_4	ΔG_5	ΔG_6	ΔG_7	ΔG_8	ΔG_1	ΔG_2	ΔG_3	ΔG_4	ΔG_5	ΔG_6	ΔG_7	ΔG_8	ΔG_1	ΔG_2	ΔG_3	ΔG_4	ΔG_5	ΔG_6	ΔG_7	ΔG_8
Sc	-0.90	2.41	0.90	2.52	-2.52	-0.90	-2.41	0.90	-1.11	2.06	1.18	2.79	-2.79	-1.18	-2.06	1.11	-1.25	2.17	1.03	2.97	-2.97	-1.03	-2.17	1.25
	(-0.94)	(2.53)	(0.70)	(2.63)	(-2.63)	(-0.70)	(-2.53)	(0.94)	(-1.15)	(2.21)	(1.00)	(2.85)	(-2.85)	(-1.00)	(-2.21)	(1.15)	(-1.05)	(2.32)	(0.89)	(2.76)	(-2.76)	(-0.89)	(-2.32)	(1.05)
Ti	-1.73	0.77	2.52	3.37	-3.37	-2.52	-0.77	1.73	-1.72	0.59	2.81	3.23	-3.23	-2.81	-0.59	1.72	-2.13	0.38	3.10	3.57	-3.57	-3.10	-0.38	2.13
	(1.77)	(0.92)	(2.36)	(3.42)	(-3.42)	(-2.36)	(-0.92)	(1.77)	(-1.80)	(0.82)	(2.52)	(3.38)	(-3.38)	(-2.52)	(-0.82)	(1.80)	(-1.85)	(0.56)	(2.70)	(3.50)	(-3.50)	(-2.70)	(-0.56)	(1.85)
V	**0.84**	**-0.55**	**2.74**	**1.89**	**-1.89**	**-2.74**	**0.55**	**-0.84**	**0.73**	**-0.14**	**2.84**	**1.50**	**-1.50**	**-2.84**	**0.14**	**-0.73**	-1.75	-0.08	3.47	3.27	-3.27	-3.47	0.08	1.75
	(0.41)	**(0.03)**	**(3.19)**	**(1.29)**	**(-1.29)**	**(-3.19)**	**(-0.03)**	**(-0.41)**	**(0.51)**	**(0.08)**	**(3.13)**	**(1.20)**	**(-1.20)**	**(-3.13)**	**(-0.08)**	**(-0.51)**	(-0.87)	(0.14)	(3.29)	(2.36)	(-2.36)	(-3.29)	(-0.14)	(0.87)
Cr	**0.98**	**0.09**	**3.16**	**0.68**	**-0.68**	**-3.16**	**-0.09**	**-0.98**	**1.14**	**0.29**	**2.93**	**0.56**	**-0.56**	**-2.93**	**-0.29**	**-1.14**	-0.67	-0.27	3.37	2.49	-2.49	-3.37	0.27	0.67
	(0.26)	**(0.81)**	**(2.43)**	**(1.41)**	**(-1.41)**	**(-2.43)**	**(-0.81)**	**(-0.26)**	**(0.43)**	**(1.01)**	**(2.23)**	**(1.25)**	**(-1.25)**	**(-2.23)**	**(-1.01)**	**(-0.43)**	(0.62)	(0.31)	(2.85)	(1.13)	(-1.13)	(-2.85)	(-0.31)	(-0.62)
Mn	0.08	0.22	2.86	1.76	-1.76	-2.86	-0.22	-0.08	0.05	0.29	2.91	1.67	-1.67	-2.91	-0.29	-0.05	-0.03	0.38	2.80	1.78	-1.78	-2.80	-0.38	0.03
	(0.51)	(1.38)	(1.76)	(1.27)	(-1.27)	(-1.76)	(-1.38)	(-0.51)	(0.32)	(1.55)	(1.60)	(1.44)	(-1.44)	(-1.60)	(-1.55)	(-0.32)	(0.26)	(1.60)	(1.58)	(1.47)	(-1.47)	(-1.58)	(-1.60)	(-0.26)
Fe	0.27	0.57	2.57	1.51	-1.51	-2.57	-0.57	-0.27	**0.59**	**1.19**	**1.98**	**1.15**	**-1.15**	**-1.98**	**-1.19**	**-0.59**	0.19	0.76	2.46	1.52	-1.52	-2.46	-0.76	-0.19
	(0.60)	(1.42)	(1.86)	(1.03)	(-1.03)	(-1.86)	(-1.42)	(-0.60)	**(-0.55)**	**(1.65)**	**(1.77)**	**(2.05)**	**(-2.05)**	**(-1.77)**	**(-1.65)**	**(0.55)**	(0.78)	(1.50)	(1.69)	(0.96)	(-0.96)	(-1.69)	(-1.50)	(-0.78)
Co	0.61	0.89	2.17	1.25	-1.25	-2.17	-0.89	-0.61	0.49	1.04	2.01	1.38	-1.38	-2.01	-1.04	-0.49	0.77	0.91	1.98	1.26	-1.26	-1.98	-0.91	-0.77
	(0.84)	(1.15)	(1.93)	(1.00)	(-1.00)	(-1.93)	(-1.15)	(-0.84)	(0.68)	(1.21)	(1.86)	(1.18)	(-1.18)	(-1.86)	(-1.21)	(-0.68)	(1.36)	(1.15)	(1.84)	(0.58)	(-0.58)	(-1.84)	(-1.15)	(-1.36)
Ni	1.59	1.63	1.25	0.45	-0.45	-1.25	-1.63	-1.59	1.58	1.59	1.27	0.49	-0.49	-1.27	-1.59	-1.58	1.56	1.89	1.00	0.47	-0.47	-1.00	-1.89	-1.56
	(1.56)	(1.79)	(1.14)	(0.43)	(-0.43)	(-1.14)	(-1.79)	(-1.56)	(1.50)	(1.78)	(1.20)	(0.44)	(-0.44)	(-1.20)	(-1.78)	(-1.50)	(1.86)	(1.39)	(1.52)	(0.15)	(-0.15)	(-1.52)	(-1.39)	(-1.86)
Cu	1.82	2.20	0.66	0.24	-0.24	-0.66	-2.20	-1.82	1.67	2.11	0.85	0.29	-0.29	-0.85	-2.11	-1.67	1.74	2.14	0.77	0.28	-0.28	-0.77	-2.14	-1.74
	(1.89)	(2.22)	(0.60)	(0.20)	(-0.20)	(-0.60)	(-2.22)	(-1.89)	(1.74)	(2.26)	(0.68)	(0.24)	(-0.24)	(-0.68)	(-2.26)	(-1.74)	(2.09)	(2.17)	(0.72)	(-0.06)	(0.06)	(-0.72)	(-2.17)	(-2.09)
Zn	0.94	2.31	0.84	0.82	-0.82	-0.84	-2.31	-0.94	0.93	-0.75	3.93	0.80	-0.80	-3.93	0.75	-0.93	0.69	2.24	0.96	1.02	-1.02	-0.96	-2.24	-0.69
	(0.93)	(2.33)	(0.81)	(0.85)	(-0.85)	(-0.81)	(-2.33)	(-0.93)	(0.89)	(-0.78)	(3.98)	(0.83)	(-0.83)	(-3.98)	(0.78)	(-0.89)	(1.52)	(2.27)	(0.91)	(0.22)	(-0.22)	(-0.91)	(-2.27)	(-1.52)
Y	-0.78	2.40	0.91	2.38	-2.38	-0.91	-2.40	0.78	-1.03	2.16	1.01	2.77	-2.77	-1.01	-2.16	1.03	-1.12	2.32	0.89	2.83	-2.83	-0.89	-2.32	1.12
Zr	-1.73	1.16	2.10	3.40	-3.40	-2.10	-1.16	1.73	-1.76	1.06	2.23	3.39	-3.39	-2.23	-1.06	1.76	-2.33	0.91	2.34	3.99	-3.99	-2.34	-0.91	2.33
Nb	-2.07	-0.03	3.28	3.73	-3.73	-3.28	0.03	2.07	**0.46**	**-0.08**	**3.33**	**1.22**	**-1.22**	**-3.33**	**0.08**	**-0.46**	**0.89**	**-0.58**	**4.00**	**0.62**	**-0.62**	**-4.00**	**0.58**	**-0.89**
Mo	**1.29**	**-0.45**	**3.82**	**0.26**	**-0.26**	**-3.82**	**0.45**	**-1.29**	**1.22**	**-0.10**	**3.41**	**0.39**	**-0.39**	**-3.41**	**0.10**	**-1.22**	**1.64**	**-0.26**	**2.71**	**0.83**	**-0.83**	**-2.71**	**0.26**	**-1.64**
Tc	**1.55**	**0.11**	**3.20**	**0.05**	**-0.05**	**-3.20**	**-0.11**	**-1.55**	**1.60**	**0.34**	**2.91**	**0.07**	**-0.07**	**-2.91**	**-0.34**	**-1.60**	**1.66**	**0.19**	**2.72**	**0.34**	**-0.34**	**-2.72**	**-0.19**	**-1.66**
Ru	**1.03**	**0.95**	**2.25**	**0.69**	**-0.69**	**-2.25**	**-0.95**	**-1.03**	**1.23**	**1.23**	**1.93**	**0.53**	**-0.53**	**-1.93**	**-1.23**	**-1.23**	**0.97**	**1.00**	**2.12**	**0.83**	**-0.83**	**-2.12**	**-1.00**	**-0.97**
Rh	0.33	0.88	2.09	1.62	-1.62	-2.09	-0.88	-0.33	0.84	0.43	1.98	1.67	-1.67	-1.98	-0.43	-0.84	0.81	0.99	1.92	1.20	-1.20	-1.92	-0.99	-0.81
Pd	1.86	1.71	1.13	0.22	-0.22	-1.13	-1.71	-1.86	1.92	1.70	1.09	0.21	-0.21	-1.09	-1.70	-1.92	1.94	1.94	0.90	0.14	-0.14	-0.90	-1.94	-1.94
Ag	1.83	2.19	0.80	0.10	-0.10	-0.80	-2.19	-1.83	1.60	2.11	1.01	0.20	-0.20	-1.01	-2.11	-1.60	2.00	1.73	1.02	0.18	-0.18	-1.02	-1.73	-2.00
Cd	0.20	2.29	0.98	1.45	-1.45	-0.98	-2.29	-0.20	0.11	2.27	0.98	1.55	-1.55	-0.98	-2.27	-0.11	0.10	2.34	0.96	1.52	-1.52	-0.96	-2.34	-0.10
La	-0.35	1.78	1.46	2.04	-2.04	-1.46	-1.78	0.35	-0.59	1.10	2.10	2.31	-2.31	-2.10	-1.10	0.59	-0.68	1.64	1.52	2.44	-2.44	-1.52	-1.64	0.68
Hf	-2.18	1.35	2.01	3.73	-3.73	-2.01	-1.35	2.18	-2.23	1.27	2.09	3.78	-3.78	-2.09	-1.27	2.23	-2.65	1.15	2.16	4.27	-4.27	-2.16	-1.15	2.65
Ta	**0.42**	**-0.25**	**1.94**	**2.81**	**-2.81**	**-1.94**	**0.25**	**-0.42**	-2.53	0.07	3.40	3.99	-3.99	-3.40	-0.07	2.53	**0.91**	**-0.44**	**3.66**	**0.79**	**-0.79**	**-3.66**	**0.44**	**-0.91**
W	**1.23**	**-0.46**	**2.76**	**1.40**	**-1.40**	**-2.76**	**0.46**	**-1.23**	**1.03**	**-0.12**	**0.36**	**3.65**	**-3.65**	**-0.36**	**0.12**	**-1.03**	**1.66**	**-0.20**	**2.80**	**0.66**	**-0.66**	**-2.80**	**0.20**	**-1.66**
Re	**1.66**	**0.01**	**2.76**	**0.49**	**-0.49**	**-2.76**	**-0.01**	**-1.66**	**1.65**	**0.33**	**3.00**	**-0.05**	**0.05**	**-3.00**	**-0.33**	**-1.65**	**2.21**	**0.17**	**2.80**	**-0.25**	**0.25**	**-2.80**	**-0.17**	**-2.21**
Os	**1.61**	**0.83**	**2.43**	**0.05**	**-0.05**	**-2.43**	**-0.83**	**-1.61**	**1.74**	**0.67**	**2.55**	**-0.05**	**0.05**	**-2.55**	**-0.67**	**-1.74**	**1.37**	**0.71**	**2.53**	**0.31**	**-0.31**	**-2.53**	**-0.71**	**-1.37**
Ir	0.31	0.51	2.38	1.72	-1.72	-2.38	-0.51	-0.31	**0.61**	**1.30**	**2.05**	**0.96**	**-0.96**	**-2.05**	**-1.30**	**-0.61**	0.78	0.70	2.24	1.21	-1.21	-2.24	-0.70	-0.78
Pt	1.88	1.48	1.43	0.13	-0.13	-1.43	-1.48	-1.88	1.88	1.43	1.44	0.16	-0.16	-1.44	-1.43	-1.88	1.85	1.98	0.97	0.13	-0.13	-0.97	-1.98	-1.85
Au	2.16	2.07	0.64	0.05	-0.05	-0.64	-2.07	-2.16	1.99	1.75	1.07	0.11	-0.11	-1.07	-1.75	-1.99	2.26	2.15	0.42	0.10	-0.10	-0.42	-2.15	-2.26
Hg	*	*	*	*	*	*	*	*	0.33	1.87	1.44	1.28	-1.28	-1.44	-1.87	-0.33	*	*	*	*	*	*	*	*

Table S26. Continued...

TM	N_4							
	OER				ORR			
	ΔG_1	ΔG_2	ΔG_3	ΔG_4	ΔG_5	ΔG_6	ΔG_7	ΔG_8
Sc	-1.98 (-2.06)	2.02 (2.19)	1.20 (1.03)	3.68 (3.76)	-3.68 (-3.76)	-1.20 (-1.03)	-2.02 (-2.19)	1.98 (2.06)
Ti	-2.09 (-2.04)	-0.31 (-0.02)	3.74 (3.27)	3.58 (3.71)	-3.58 (-3.71)	-3.74 (-3.27)	0.31 (0.02)	2.09 (2.04)
V	**2.79** **(0.65)**	**0.40** **(1.50)**	**1.04** **(1.26)**	**0.69** **(1.51)**	**-0.69** **(-1.51)**	**-1.04** **(-1.26)**	**-0.40** **(-1.50)**	**-2.79** **(-0.65)**
Cr	-0.17 (0.19)	-0.15 (0.76)	3.24 (2.35)	2.00 (1.61)	-2.00 (-1.61)	-3.24 (-2.35)	0.15 (-0.76)	0.17 (-0.19)
Mn	0.37 (0.24)	0.45 (1.62)	2.71 (1.57)	1.39 (1.49)	-1.39 (-1.49)	-2.71 (-1.57)	-0.45 (-1.62)	-0.37 (-0.24)
Fe	0.56 (0.70)	0.60 (1.26)	2.27 (1.96)	1.48 (1.01)	-1.48 (-1.01)	-2.27 (-1.96)	-0.60 (-1.26)	-0.56 (-0.70)
Co	0.98 (1.37)	1.33 (0.94)	1.76 (2.00)	0.84 (0.61)	-0.84 (-0.61)	-1.76 (-2.00)	-1.33 (-0.94)	-0.98 (-1.37)
Ni	1.95 (2.02)	2.02 (2.03)	0.74 (0.73)	0.21 (0.15)	-0.21 (-0.15)	-0.74 (-0.73)	-2.02 (-2.03)	-1.95 (-2.02)
Cu	1.76 (1.82)	1.88 (2.07)	1.00 (0.77)	0.28 (0.25)	-0.28 (-0.25)	-1.00 (-0.77)	-1.88 (-2.07)	-1.76 (-1.82)
Zn	0.56 (0.65)	2.47 (2.34)	0.75 (0.88)	1.14 (1.05)	-1.14 (-1.05)	-0.75 (-0.88)	-2.47 (-2.34)	-0.56 (-0.65)
Y	-1.85	2.31	0.91	3.54	-3.54	-0.91	-2.31	1.85
Zr	-2.57	0.24	3.00	4.25	-4.25	-3.00	-0.24	2.57
Nb	**-0.16**	**1.09**	**1.92**	**2.07**	**-2.07**	**-1.92**	**-1.09**	**0.16**
Mo	**0.73**	**2.41**	**0.75**	**1.03**	**-1.03**	**-0.75**	**-2.41**	**-0.73**
Tc	**1.40**	**1.99**	**0.96**	**0.57**	**-0.57**	**-0.96**	**-1.99**	**-1.40**
Ru	**2.41**	**0.29**	**0.95**	**1.27**	**-1.27**	**-0.95**	**-0.29**	**-2.41**
Rh	0.96	1.41	1.53	1.02	-1.02	-1.53	-1.41	-0.96
Pd	2.27	2.04	0.57	0.04	-0.04	-0.57	-2.04	-2.27
Ag	0.70	2.03	1.01	1.18	-1.18	-1.01	-2.03	-0.70
Cd	0.16	2.46	0.80	1.50	-1.50	-0.80	-2.46	-0.16
La	-1.32	1.66	1.53	3.05	-3.05	-1.53	-1.66	1.32
Hf	-2.79	0.47	2.85	4.39	-4.39	-2.85	-0.47	2.79
Ta	**-0.46**	**1.48**	**1.67**	**2.23**	**-2.23**	**-1.67**	**-1.48**	**0.46**
W	**0.17**	**-0.05**	**3.30**	**1.50**	**-1.50**	**-3.30**	**0.05**	**-0.17**
Re	**2.41**	**1.03**	**0.84**	**0.65**	**-0.65**	**-0.84**	**-1.03**	**-2.41**
Os	**2.56**	**0.35**	**0.75**	**1.26**	**-1.26**	**-0.75**	**-0.35**	**-2.56**
Ir	1.04	1.03	1.90	0.95	-0.95	-1.90	-1.03	-1.04
Pt	2.30	1.89	0.71	0.02	-0.02	-0.71	-1.89	-2.30
Au	2.02	2.18	0.41	0.30	-0.30	-0.41	-2.18	-2.02
Hg	*	*	*	*	*	*	*	*

Table S27. Reaction free energy (eV) of elementary steps for OER/ORR in the presence of solvent (dielectric constant : 80) for M-N_nC_m-G_N DV-systems. (RPBE(U)+TS results are in parentheses). (*OOH is unstable, breaking into *O and *OH in pathway-I; so, for these systems the OER/ORR reacttion free energies marked in blue are calculated for path-II shown in Figure 6). The data for the La case in solvent are not provided due to the convergency problem.

TM	C_4								N_1C_3								N_2C_2							
	OER				ORR				OER				ORR				OER				ORR			
	ΔG_1	ΔG_2	ΔG_3	ΔG_4	ΔG_5	ΔG_6	ΔG_7	ΔG_8	ΔG_1	ΔG_2	ΔG_3	ΔG_4	ΔG_5	ΔG_6	ΔG_7	ΔG_8	ΔG_1	ΔG_2	ΔG_3	ΔG_4	ΔG_5	ΔG_6	ΔG_7	ΔG_8
Sc	0.27	-0.03	3.28	1.40	-1.40	-3.28	0.03	-0.27	0.46	1.13	2.01	1.32	-1.32	-2.01	-1.13	-0.46	0.46	1.87	1.29	1.29	-1.29	-1.29	-1.87	-0.46
	(0.31)	(-0.05)	(3.43)	(1.23)	(-1.23)	(-3.43)	(0.05)	(-0.31)	(0.54)	(1.07)	(2.06)	(1.26)	(-1.26)	(-2.06)	(-1.07)	(-0.54)	(0.53)	(1.94)	(1.27)	(1.18)	(-1.18)	(-1.27)	(-1.94)	(-0.53)
Ti	0.44	1.20	1.86	1.42	-1.42	-1.86	-1.20	-0.44	-0.05	1.09	2.03	1.85	-1.85	-2.03	-1.09	0.05	-0.15	0.54	2.50	2.03	-2.03	-2.50	-0.54	0.15
	(0.67)	(1.36)	(1.71)	(1.17)	(-1.17)	(-1.71)	(-1.36)	(-0.67)	(0.13)	(1.23)	(1.89)	(1.67)	(-1.67)	(-1.89)	(-1.23)	(-0.13)	(0.02)	(0.65)	(2.41)	(1.85)	(-1.85)	(-2.41)	(-0.65)	(-0.02)
V	-0.46	0.32	2.83	2.23	-2.23	-2.83	-0.32	0.46	-0.53	-0.10	3.28	2.27	-2.27	-3.28	0.10	0.53	-0.60	0.00	3.28	2.25	-2.25	-3.28	0.00	0.60
	(-0.16)	(0.42)	(2.79)	(1.87)	(-1.87)	(-2.79)	(-0.42)	(0.16)	(-0.22)	(0.12)	(3.07)	(1.96)	(-1.96)	(-3.07)	(-0.12)	(0.22)	(-0.34)	(0.35)	(2.91)	(2.00)	(-2.00)	(-2.91)	(-0.35)	(0.34)
Cr	-0.43	0.13	3.06	2.16	-2.16	-3.06	-0.13	0.43	-0.74	-0.11	3.31	2.47	-2.47	-3.31	0.11	0.74	-0.61	-0.24	3.32	2.45	-2.45	-3.32	0.24	0.61
	(0.07)	(0.27)	(2.94)	(1.64)	(-1.64)	(-2.94)	(-0.27)	(-0.07)	(-0.37)	(0.27)	(2.94)	(2.08)	(-2.08)	(-2.94)	(-0.27)	(0.37)	(-0.24)	(0.28)	(2.85)	(2.02)	(-2.02)	(-2.85)	(-0.28)	(0.24)
Mn	-0.31	0.22	2.93	2.07	-2.07	-2.93	-0.22	0.31	-0.24	0.34	2.78	2.03	-2.03	-2.78	-0.34	0.24	-0.17	0.30	2.82	1.97	-1.97	-2.82	-0.30	0.17
	(-0.13)	(0.77)	(2.39)	(1.88)	(-1.88)	(-2.39)	(-0.77)	(0.13)	(0.34)	(1.42)	(1.73)	(1.43)	(-1.43)	(-1.73)	(-1.42)	(-0.34)	(0.31)	(1.42)	(1.71)	(1.48)	(-1.48)	(-1.71)	(-1.42)	(-0.31)
Fe	-0.13	0.35	2.79	1.91	-1.91	-2.79	-0.35	0.13	0.01	0.33	2.71	1.87	-1.87	-2.71	-0.33	-0.01	-0.13	0.50	2.35	2.20	-2.20	-2.35	-0.50	0.13
	(0.02)	(1.46)	(1.70)	(1.74)	(-1.74)	(-1.70)	(-1.46)	(-0.02)	(0.19)	(1.48)	(2.06)	(1.19)	(-1.19)	(-2.06)	(-1.48)	(-0.19)	(0.42)	(1.12)	(2.00)	(1.38)	(-1.38)	(-2.00)	(-1.12)	(-0.42)
Co	0.09	0.85	2.07	1.91	-1.91	-2.07	-0.85	-0.09	0.07	0.84	2.08	1.93	-1.93	-2.08	-0.84	-0.07	0.34	0.88	1.92	1.78	-1.78	-1.92	-0.88	-0.34
	(0.44)	(1.09)	(2.01)	(1.38)	(-1.38)	(-2.01)	(-1.09)	(-0.44)	(0.68)	(1.04)	(1.95)	(1.25)	(-1.25)	(-1.95)	(-1.04)	(-0.68)	(0.57)	(1.12)	(1.87)	(1.37)	(-1.37)	(-1.87)	(-1.12)	(-0.57)
Ni	0.70	1.34	1.54	1.34	-1.34	-1.54	-1.34	-0.70	1.11	1.52	1.36	0.94	-0.94	-1.36	-1.52	-1.11	1.37	1.53	1.32	0.70	-0.70	-1.32	-1.53	-1.37
	(0.75)	(1.34)	(1.52)	(1.30)	(-1.30)	(-1.52)	(-1.34)	(-0.75)	(1.08)	(0.99)	(1.89)	(0.95)	(-0.95)	(-1.89)	(-0.99)	(-1.08)	(1.31)	(1.76)	(1.15)	(0.70)	(-0.70)	(-1.15)	(-1.76)	(-1.31)
Cu	1.73	1.96	0.88	0.35	-0.35	-0.88	-1.96	-1.73	1.84	2.02	0.74	0.32	-0.32	-0.74	-2.02	-1.84	1.35	2.08	0.84	0.65	-0.65	-0.84	-2.08	-1.35
	(1.77)	(2.04)	(0.79)	(0.32)	(-0.32)	(-0.79)	(-2.04)	(-1.77)	(1.93)	(2.05)	(0.63)	(0.31)	(-0.31)	(-0.63)	(-2.05)	(-1.93)	(1.39)	(2.11)	(0.81)	(0.62)	(-0.62)	(-0.81)	(-2.11)	(-1.39)
Zn	1.34	2.17	0.80	0.62	-0.62	-0.80	-2.17	-1.34	0.70	2.22	0.89	1.11	-1.11	-0.89	-2.22	-0.70	1.11	2.07	1.01	0.73	-0.73	-1.01	-2.07	-1.11
	(1.25)	(2.17)	(0.82)	(0.68)	(-0.68)	(-0.82)	(-2.17)	(-1.25)	(0.66)	(2.23)	(0.91)	(1.13)	(-1.13)	(-0.91)	(-2.23)	(-0.66)	(1.09)	(2.09)	(1.00)	(0.74)	(-0.74)	(-1.00)	(-2.09)	(-1.09)
Y	-0.04	0.34	3.00	1.62	-1.62	-3.00	-0.34	0.04	0.21	1.28	1.88	1.56	-1.56	-1.88	-1.28	-0.21	0.44	1.79	1.24	1.45	-1.45	-1.24	-1.79	-0.44
Zr	0.60	0.52	2.57	1.24	-1.24	-2.57	-0.52	-0.60	0.08	1.19	1.90	1.75	-1.75	-1.90	-1.19	-0.08	-0.02	0.89	2.31	1.73	-1.73	-2.31	-0.89	0.02
Nb	-0.18	0.55	2.53	2.02	-2.02	-2.53	-0.55	0.18	-0.36	0.25	2.90	2.14	-2.14	-2.90	-0.25	0.36	-0.91	0.16	3.01	2.67	-2.67	-3.01	-0.16	0.91
Mo	-0.56	-0.05	3.22	2.32	-2.32	-3.22	0.05	0.56	-0.68	0.17	2.94	2.50	-2.50	-2.94	-0.17	0.68	**1.63**	**-0.32**	**3.39**	**0.21**	**-0.21**	**-3.39**	**0.32**	**-1.63**
Tc	-0.41	-0.08	3.24	2.17	-2.17	-3.24	0.08	0.41	**1.65**	**-0.23**	**2.80**	**0.70**	**-0.70**	**-2.80**	**0.23**	**-1.65**	**1.71**	**0.31**	**2.80**	**0.10**	**-0.10**	**-2.80**	**-0.31**	**-1.71**
Ru	-0.31	0.19	2.72	2.32	-2.32	-2.72	-0.19	0.31	-0.84	0.30	2.53	2.94	-2.94	-2.53	-0.30	0.84	**1.88**	**0.29**	**2.84**	**-0.09**	**0.09**	**-2.84**	**-0.29**	**-1.88**
Rh	-0.17	0.89	2.10	2.10	-2.10	-2.10	-0.89	0.17	0.27	0.61	2.42	1.61	-1.61	-2.42	-0.61	-0.27	0.10	1.11	1.82	1.89	-1.89	-1.82	-1.11	-0.10
Pd	0.88	1.32	1.56	1.16	-1.16	-1.56	-1.32	-0.88	0.87	1.70	1.24	1.11	-1.11	-1.24	-1.70	-0.87	1.43	1.82	1.02	0.64	-0.64	-1.02	-1.82	-1.43
Ag	1.44	2.03	0.92	0.53	-0.53	-0.92	-2.03	-1.44	1.76	2.00	0.79	0.37	-0.37	-0.79	-2.00	-1.76	1.47	2.16	0.76	0.54	-0.54	-0.76	-2.16	-1.47
Cd	0.97	1.87	1.23	0.86	-0.86	-1.23	-1.87	-0.97	-0.10	2.28	0.99	1.75	-1.75	-0.99	-2.28	0.10	0.60	2.27	0.97	1.08	-1.08	-0.97	-2.27	-0.60
La	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Hf	0.47	0.83	2.26	1.36	-1.36	-2.26	-0.83	-0.47	-0.06	1.27	1.90	1.81	-1.81	-1.90	-1.27	0.06	-0.20	1.05	2.31	1.76	-1.76	-2.31	-1.05	0.20
Ta	-0.40	0.65	2.68	1.99	-1.99	-2.68	-0.65	0.40	-0.62	0.23	3.00	2.31	-2.31	-3.00	-0.23	0.62	**1.38**	**-0.76**	**4.00**	**0.31**	**-0.31**	**-4.00**	**0.76**	**-1.38**
W	-0.95	-0.21	3.33	2.75	-2.75	-3.33	0.21	0.95	-1.09	-0.04	0.40	5.65	-5.65	-0.40	0.04	1.09	**1.73**	**-0.46**	**3.52**	**0.13**	**-0.13**	**-3.52**	**0.46**	**-1.73**
Re	**1.64**	**-0.19**	**3.32**	**0.15**	**-0.15**	**-3.32**	**0.19**	**-1.64**	**2.10**	**-0.68**	**2.92**	**0.59**	**-0.59**	**-2.92**	**0.68**	**-2.10**	**1.83**	**0.35**	**2.76**	**-0.02**	**0.02**	**-2.76**	**-0.35**	**-1.83**
Os	**1.64**	**0.61**	**2.56**	**0.11**	**-0.11**	**-2.56**	**-0.61**	**-1.64**	**1.61**	**0.65**	**2.15**	**0.50**	**-0.50**	**-2.15**	**-0.65**	**-1.61**	**2.08**	**0.40**	**2.10**	**0.34**	**-0.34**	**-2.10**	**-0.40**	**-2.08**
Ir	-0.61	0.31	2.71	2.51	-2.51	-2.71	-0.31	0.61	-0.03	0.14	2.41	2.39	-2.39	-2.41	-0.14	0.03	-0.12	0.68	2.21	2.14	-2.14	-2.21	-0.68	0.12
Pt	0.85	1.01	2.17	0.88	-0.88	-2.17	-1.01	-0.85	0.88	1.42	1.56	1.06	-1.06	-1.56	-1.42	-0.88	1.35	1.40	1.48	0.69	-0.69	-1.48	-1.40	-1.35
Au	0.62	3.10	0.78	0.42	-0.42	-0.78	-3.10	-0.62	2.00	1.70	0.84	0.38	-0.38	-0.84	-1.70	-2.00	1.73	2.16	0.60	0.42	-0.42	-0.60	-2.16	-1.73
Hg	1.58	2.22	0.54	0.58	-0.58	-0.54	-2.22	-1.58	0.92	2.06	1.05	0.90	-0.90	-1.05	-2.06	-0.92	-0.55	2.13	1.13	2.20	-2.20	-1.13	-2.13	0.55

Table S27. Continued...

TM	N_2C_2[b]								N_2C_2[c]								N_3C_1							
	OER				ORR				OER				OER				OER				OER			
	ΔG_1	ΔG_2	ΔG_1	ΔG_1	ΔG_1	ΔG_1	ΔG_1	ΔG_1	ΔG_1	ΔG_1	ΔG_3	ΔG_4	ΔG_5	ΔG_6	ΔG_7	ΔG_8	ΔG_1	ΔG_2	ΔG_3	ΔG_4	ΔG_5	ΔG_6	ΔG_7	ΔG_8
Sc	0.53	1.77	1.30	1.31	-1.31	-1.30	-1.77	-0.53	0.26	1.07	1.99	1.61	-1.61	-1.99	-1.07	-0.26	0.32	1.67	1.41	1.53	-1.53	-1.41	-1.67	-0.32
	(0.56)	(1.94)	(1.21)	(1.22)	(-1.22)	(-1.21)	(-1.94)	(-0.56)	(0.27)	(1.20)	(2.20)	(1.25)	(-1.25)	(-2.20)	(-1.20)	(-0.27)	(0.40)	(1.78)	(1.33)	(1.41)	(-1.41)	(-1.33)	(-1.78)	(-0.40)
Ti	-0.11	0.53	2.54	1.96	-1.96	-2.54	-0.53	0.11	-0.17	0.40	2.92	1.78	-1.78	-2.92	-0.40	0.17	-0.67	0.21	2.89	2.49	-2.49	-2.89	-0.21	0.67
	(0.00)	(0.68)	(2.46)	(1.77)	(-1.77)	(-2.46)	(-0.68)	(0.00)	(-0.04)	(0.54)	(2.76)	(1.65)	(-1.65)	(-2.76)	(-0.54)	(0.04)	(-0.56)	(0.37)	(2.77)	(2.35)	(-2.35)	(-2.77)	(-0.37)	(0.56)
V	**0.78**	**-0.46**	**2.29**	**2.31**	**-2.31**	**-2.29**	**0.46**	**-0.78**	**0.77**	**-0.15**	**2.26**	**2.03**	**-2.03**	**-2.26**	**0.15**	**-0.77**	-1.09	-0.16	3.52	2.65	-2.65	-3.52	0.16	1.09
	(0.38)	**(0.05)**	**(3.17)**	**(1.43)**	**(-1.43)**	**(-3.17)**	**(0.05)**	**(-0.38)**	**(0.44)**	**(0.06)**	**(3.15)**	**(1.27)**	**(-1.27)**	**(-3.15)**	**(-0.06)**	**(-0.44)**	(-0.58)	(0.03)	(3.28)	(2.19)	(-2.19)	(-3.28)	(-0.03)	(0.58)
Cr	**0.91**	**0.15**	**3.00**	**0.86**	**-0.86**	**-3.00**	**-0.15**	**-0.91**	**1.09**	**0.35**	**2.78**	**0.70**	**-0.70**	**-2.78**	**-0.35**	**-1.09**	-0.73	-0.19	3.27	2.58	-2.58	-3.27	0.19	0.73
	(0.14)	**(0.78)**	**(2.35)**	**(1.66)**	**(-1.66)**	**(-2.35)**	**(-0.78)**	**(-0.14)**	**(0.35)**	**(1.02)**	**(2.16)**	**(1.39)**	**(-1.39)**	**(-2.16)**	**(-1.02)**	**(-0.35)**	(-0.27)	(0.39)	(2.72)	(2.08)	(-2.08)	(-2.72)	(-0.39)	(0.27)
Mn	0.01	0.29	2.71	1.92	-1.92	-2.71	-0.29	-0.01	-0.05	0.41	2.67	1.89	-1.89	-2.67	-0.41	0.05	-0.20	0.50	2.61	2.00	-2.00	-2.61	-0.50	0.20
	(0.40)	(1.51)	(1.59)	(1.42)	(-1.42)	(-1.59)	(-1.51)	(-0.40)	(0.21)	(1.70)	(1.43)	(1.58)	(-1.58)	(-1.43)	(-1.70)	(-0.22)	(0.10)	(1.76)	(1.38)	(1.68)	(-1.68)	(-1.38)	(-1.76)	(-0.10)
Fe	0.10	0.61	2.55	1.67	-1.67	-2.55	-0.61	-0.10	**0.51**	**1.07**	**2.07**	**1.28**	**-1.28**	**-2.07**	**-1.07**	**-0.51**	-0.01	0.77	2.40	1.76	-1.76	-2.40	-0.77	0.01
	(0.4)	(1.30)	(1.88)	(1.34)	(-1.34)	(-1.88)	(-1.30)	(-0.40)	**(-0.47)**	**(1.50)**	**(1.84)**	**(2.05)**	**(-2.05)**	**(-1.84)**	**(-1.50)**	**(0.47)**	(0.06)	(1.44)	(1.71)	(1.71)	(-1.71)	(-1.71)	(-1.44)	(-0.06)
Co	0.44	0.87	2.13	1.47	-1.47	-2.13	-0.87	-0.44	0.45	0.85	2.02	1.60	-1.60	-2.02	-0.85	-0.45	0.58	0.90	1.95	1.49	-1.49	-1.95	-0.90	-0.58
	(0.62)	(1.11)	(1.91)	(1.28)	(1.28)	(-1.91)	(-1.11)	(-0.62)	(-0.07)	(1.15)	(1.91)	(1.93)	(-1.93)	(-1.91)	(-1.15)	(0.07)	(0.03)	(1.12)	(1.82)	(1.95)	(-1.95)	(-1.82)	(-1.12)	(-0.03)
Ni	1.37	1.65	1.20	0.70	-0.70	-1.20	-1.65	-1.37	1.35	1.63	1.23	0.72	-0.72	-1.23	-1.63	-1.35	1.27	1.87	0.99	0.78	-0.78	-0.99	-1.87	-1.27
	(1.30)	(1.84)	(1.08)	(0.71)	(-0.71)	(-1.08)	(-1.84)	(-1.30)	(1.30)	(1.76)	(1.15)	(0.72)	(-0.72)	(-1.15)	(-1.76)	(-1.30)	(1.37)	(1.39)	(1.50)	(0.66)	(-0.66)	(-1.50)	(-1.39)	(-1.37)
Cu	1.47	2.18	0.67	0.60	-0.60	-0.67	-2.18	-1.47	1.29	2.15	0.82	0.66	-0.66	-0.82	-2.15	-1.29	1.42	2.03	0.85	0.61	-0.61	-0.85	-2.03	-1.42
	(1.52)	(2.21)	(0.62)	(0.57)	(-0.57)	(-0.62)	(-2.21)	(-1.52)	(1.34)	(2.19)	(1.77)	(0.62)	(-0.62)	(-0.77)	(-2.19)	(-1.34)	(1.43)	(2.18)	(0.70)	(0.61)	(-0.61)	(-0.70)	(-2.18)	(-1.43)
Zn	0.70	2.25	0.86	1.11	-1.11	-0.86	-2.25	-0.70	0.61	-0.64	3.83	1.12	-1.12	-3.83	0.64	-0.61	0.37	2.18	1.00	1.37	-1.37	-1.00	-2.18	-0.37
	(0.67)	(2.26)	(0.84)	(1.14)	(-1.14)	(-0.84)	(-2.26)	(-0.67)	(0.57)	(-0.66)	(3.85)	(1.16)	(-1.16)	(-3.85)	(0.66)	(-0.57)	(0.44)	(2.22)	(0.97)	(1.30)	(-1.30)	(-0.97)	(-2.22)	(-0.44)
Y	0.46	1.82	1.31	1.33	-1.33	-1.31	-1.82	-0.46	0.24	0.99	2.09	1.60	-1.60	-2.09	-0.99	-0.24	0.26	1.86	1.33	1.47	-1.47	-1.33	-1.86	-0.26
Zr	0.05	1.21	1.98	1.68	-1.68	-1.98	-1.21	-0.05	-0.08	0.80	2.34	1.87	-1.87	-2.34	-0.80	0.08	-0.45	0.71	2.37	2.28	-2.28	-2.37	-0.71	0.45
Nb	-0.77	-0.03	3.05	2.67	-2.67	-3.05	0.03	0.77	**0.58**	**-0.05**	**3.29**	**1.09**	**-1.09**	**-3.29**	**0.05**	**-0.58**	**0.99**	**-0.52**	**3.83**	**0.62**	**-0.62**	**-3.83**	**0.52**	**-0.99**
Mo	**1.23**	**-0.21**	**3.43**	**0.48**	**-0.48**	**-3.43**	**0.21**	**-1.23**	**1.19**	**-0.04**	**3.19**	**0.57**	**-0.57**	**-3.19**	**0.04**	**-1.19**	**1.57**	**-0.17**	**3.39**	**0.13**	**-0.13**	**-3.39**	**0.17**	**-1.57**
Tc	**1.45**	**0.22**	**2.92**	**0.32**	**-0.32**	**-2.92**	**-0.22**	**-1.45**	**1.54**	**0.42**	**2.71**	**0.25**	**-0.25**	**-2.71**	**-0.42**	**-1.54**	**1.92**	**0.27**	**2.30**	**0.43**	**-0.43**	**-2.30**	**-0.27**	**-1.92**
Ru	**0.92**	**1.00**	**2.06**	**0.94**	**-0.94**	**-2.06**	**-1.00**	**-0.92**	**1.14**	**1.31**	**1.91**	**0.56**	**-0.56**	**-1.91**	**-1.31**	**-1.14**	**0.91**	**0.96**	**2.01**	**1.04**	**-1.04**	**-2.01**	**-0.96**	**-0.91**
Rh	0.16	0.84	2.02	1.90	-1.90	-2.02	-0.84	-0.16	0.66	0.41	2.06	1.79	-1.79	-2.06	-0.41	-0.66	0.67	1.00	1.87	1.37	-1.37	-1.87	-1.00	-0.67
Pd	1.66	1.62	1.18	0.46	-0.46	-1.18	-1.62	-1.66	1.68	1.72	1.06	0.46	-0.46	-1.06	-1.72	-1.68	1.62	1.95	0.89	0.46	-0.46	-0.89	-1.95	-1.62
Ag	1.59	2.13	0.75	0.46	-0.46	-0.75	-2.13	-1.59	1.26	2.14	0.92	0.60	-0.60	-0.92	-2.14	-1.26	1.54	1.67	1.18	0.53	-0.53	-1.18	-1.67	-1.54
Cd	0.22	2.16	1.05	1.48	-1.48	-1.05	-2.16	-0.22	-0.61	2.05	1.19	2.28	-2.28	-1.19	-2.05	0.61	0.06	2.28	0.94	1.64	-1.64	-0.94	-2.28	-0.06
La	10.88	-3.82	10.95	-13.09	13.09	-10.95	3.82	-10.88	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Hf	-0.15	1.12	2.05	1.89	-1.89	-2.05	-1.12	0.15	-0.24	0.91	2.25	2.01	-2.01	-2.25	-0.91	0.24	-0.68	0.93	2.22	2.45	-2.45	-2.22	-0.93	0.68
Ta	**0.41**	**-0.19**	**3.52**	**1.18**	**-1.18**	**-3.52**	**0.19**	**-0.41**	-1.21	0.04	3.27	2.81	-2.81	-3.27	-0.04	1.21	**0.91**	**-0.29**	**3.73**	**0.56**	**-0.56**	**-3.73**	**0.29**	**-0.91**
W	**1.23**	**-0.40**	**2.22**	**1.86**	**-1.86**	**-2.22**	**0.40**	**-1.23**	**1.02**	**-0.09**	**2.24**	**1.74**	**-1.74**	**-2.24**	**0.09**	**-1.02**	**1.64**	**-0.14**	**2.11**	**1.31**	**-1.31**	**-2.11**	**0.14**	**-1.64**
Re	**1.59**	**0.11**	**2.50**	**0.72**	**-0.72**	**-2.50**	**-0.11**	**-1.59**	**1.60**	**0.40**	**2.89**	**0.03**	**-0.03**	**-2.89**	**-0.40**	**-1.60**	**2.12**	**0.25**	**2.29**	**0.25**	**-0.25**	**-2.29**	**-0.25**	**-2.12**
Os	**1.49**	**0.91**	**2.22**	**0.29**	**-0.29**	**-2.22**	**-0.91**	**-1.49**	**1.64**	**0.52**	**2.60**	**0.15**	**-0.15**	**-2.60**	**-0.52**	**-1.64**	**1.28**	**0.54**	**2.32**	**0.78**	**-0.78**	**-2.32**	**-0.54**	**-1.28**
Ir	0.19	0.50	2.20	2.03	-2.03	-2.20	-0.50	-0.19	**0.58**	**1.14**	**2.12**	**1.08**	**-1.08**	**-2.12**	**-1.14**	**-0.58**	0.66	0.69	2.20	1.37	-1.37	-2.20	-0.69	-0.66
Pt	1.71	1.47	1.38	0.36	-0.36	-1.38	-1.47	-1.71	1.70	1.44	1.39	0.39	-0.39	-1.39	-1.44	-1.70	1.64	1.72	1.14	0.41	-0.41	-1.14	-1.72	-1.64
Au	1.80	2.12	0.59	0.41	-0.41	-0.59	-2.12	-1.80	1.58	1.85	1.02	0.47	-0.47	-1.02	-1.85	-1.58	1.79	2.20	0.49	0.45	-0.45	-0.49	-2.20	-1.79
Hg	*	*	*	*	*	*	*	*	0.27	1.41	1.63	1.60	-1.60	-1.63	-1.41	-0.27	*	*	*	*	*	*	*	*

Table S27. Continued...

TM	N_4							
	OER				ORR			
	ΔG_1	ΔG_2	ΔG_3	ΔG_4	ΔG_5	ΔG_6	ΔG_7	ΔG_8
Sc	-0.20 (-0.10)	1.60 (1.84)	1.49 (1.29)	2.03 (1.89)	-2.03 (-1.89)	-1.49 (-1.29)	-1.60 (-1.84)	0.20 (0.10)
Ti	-0.77 (-0.35)	-0.37 (-0.15)	3.80 (3.24)	2.27 (2.18)	-2.27 (-2.18)	-3.80 (-3.24)	0.37 (0.15)	0.77 (0.35)
V	**1.32** **(1.72)**	**1.97** **(1.60)**	**0.86** **(1.31)**	**0.76** **(0.29)**	**-0.76** **(-0.29)**	**-0.86** **(-1.31)**	**-1.97** **(-1.60)**	**-1.32** **(-1.72)**
Cr	-0.37 (1.04)	-0.05 (-0.22)	3.08 (2.21)	2.26 (1.89)	-2.26 (-1.89)	-3.08 (-2.21)	0.05 (0.22)	0.37 (-1.04)
Mn	0.18 (0.02)	0.54 (1.66)	2.56 (1.47)	1.63 (1.77)	-1.63 (-1.77)	-2.56 (-1.47)	-0.54 (1.66)	-0.18 (-0.02)
Fe	0.38 (0.24)	0.53 (1.40)	2.29 (1.98)	1.72 (1.30)	-1.72 (-1.30)	-2.29 (-1.98)	-0.53 (-1.40)	-0.38 (-0.24)
Co	0.75 (0.38)	1.25 (0.74)	1.61 (2.20)	1.32 (1.60)	-1.32 (-1.60)	-1.61 (-2.20)	-1.25 (-0.74)	-0.75 (-0.38)
Ni	1.61 (1.68)	1.78 (2.05)	0.99 (0.72)	0.54 (0.46)	-0.54 (-0.46)	-0.99 (-0.72)	-1.78 (-2.05)	-1.61 (-1.68)
Cu	1.33 (1.37)	1.92 (2.12)	1.04 (0.84)	0.63 (0.60)	-0.63 (-0.60)	-1.04 (-0.84)	-1.92 (-2.12)	-1.33 (-1.37)
Zn	0.21 (0.31)	2.23 (2.28)	0.96 (0.92)	1.52 (1.41)	-1.52 (-1.41)	-0.96 (-0.92)	-2.23 (-2.28)	-0.21 (-0.31)
Y	-0.14	1.76	1.42	1.89	-1.89	-1.42	-1.76	0.14
Zr	-0.84	0.17	2.90	2.69	-2.69	-2.90	-0.17	0.84
Nb	**-0.15**	**0.59**	**2.30**	**2.18**	**-2.18**	**-2.30**	**-0.59**	**0.15**
Mo	**0.77**	**2.30**	**0.62**	**1.23**	**-1.23**	**-0.62**	**-2.30**	**-0.77**
Tc	**1.21**	**2.06**	**0.79**	**0.86**	**-0.86**	**-0.79**	**-2.06**	**-1.21**
Ru	**1.27**	**1.85**	**0.56**	**1.24**	**-1.24**	**-0.56**	**-1.85**	**-1.27**
Rh	0.79	1.34	1.55	1.24	-1.24	-1.55	-1.34	-0.79
Pd	1.91	2.03	0.59	0.39	-0.39	-0.59	-2.03	-1.91
Ag	0.24	2.04	1.02	1.62	-1.62	-1.02	-2.04	-0.24
Cd	0.05	2.19	1.02	1.65	-1.65	-1.02	-2.19	-0.05
La	-	-	-	-	-	-	-	-
Hf	-1.06	0.43	2.79	2.76	-2.76	-2.79	-0.43	1.06
Ta	**-0.59**	**1.06**	**2.06**	**2.39**	**-2.39**	**-2.06**	**-1.06**	**0.59**
W	**0.30**	**-0.47**	**3.47**	**1.63**	**-1.63**	**-3.47**	**0.47**	**-0.30**
Re	**1.13**	**2.20**	**-0.37**	**1.96**	**-1.96**	**0.37**	**-2.20**	**-1.13**
Os	**0.93**	**1.99**	**0.67**	**1.34**	**-1.34**	**-0.67**	**-1.99**	**-0.93**
Ir	0.88	0.96	1.93	1.14	-1.14	-1.93	-0.96	-0.88
Pt	2.00	1.70	0.84	0.37	-0.37	-0.84	-1.70	-2.00
Au	1.41	2.22	0.62	0.67	-0.67	-0.62	-2.22	-1.41
Hg	*	*	*	*	*	*	*	*

Table S28. Theoretical overpotential of OER in the absence (vacuum) and presence of solvent (by implicit solvation model) for divalency systems. (RPBE(U)+TS results are in parentheses). The solvent effect is added: $\Delta\eta = \eta_{sol} - \eta_{vac}$.

TM	C_4			N_1C_3			N_2C_2			N_2C_2[b]			N_2C_2[c]			N_3C_1			N_4		
	$\eta^{OER-vac}$	$\eta^{OER-sol}$	$\Delta\eta^{OER}$	$\eta^{OER-vac}$	$\eta^{OER-sol}$	$\Delta\eta^{OER}$	$\eta^{OER-vac}$	$\eta^{OER-sol}$	$\Delta\eta^{OER}$	$\eta^{OER-vac}$	$\eta^{OER-sol}$	$\Delta\eta^{OER}$	$\eta^{OER-vac}$	$\eta^{OER-sol}$	$\Delta\eta^{OER}$	$\eta^{OER-vac}$	$\eta^{OER-sol}$	$\Delta\eta^{OER}$	$\eta^{OER-vac}$	$\eta^{OER-sol}$	$\Delta\eta^{OER}$
Sc	1.35	2.05	0.70	1.17	0.78	-0.39	1.45	0.64	-0.81	1.29	0.54	-0.74	1.56	0.76	-0.80	1.74	0.44	-1.30	2.45	0.80	-1.65
	(1.29)	(2.20)	(0.91)	(1.22)	(0.83)	(-0.39)	(1.50)	(0.71)	(-0.79)	(1.40)	(0.71)	(-0.69)	(1.62)	(0.97)	(-0.65)	(1.53)	(0.55)	(-0.99)	(2.53)	(0.66)	(-1.87)
Ti	1.63	0.63	-1.00	2.05	0.80	-1.25	2.25	1.27	-0.98	2.14	1.31	-0.83	2.00	1.69	-0.32	2.34	1.66	-0.68	2.51	2.57	0.05
	(1.61)	(0.48)	(-1.12)	(2.10)	(0.66)	(-1.44)	(2.26)	(1.18)	(-1.08)	(2.19)	(1.23)	(-0.95)	(2.15)	(1.53)	(-0.62)	(2.27)	(1.54)	(-0.74)	(2.48)	(2.01)	(-0.46)
V	1.85	1.60	-0.25	2.16	2.05	-0.11	2.18	2.05	-0.13	**1.51**	**1.08**	**-0.44**	**1.61**	**1.03**	**-0.57**	2.24	2.29	0.05	**1.56**	**0.74**	**-0.82**
	(1.58)	(1.56)	(-0.03)	(1.92)	(1.84)	(-0.08)	(1.74)	(1.68)	(-0.06)	**(1.96)**	**(1.94)**	**(-0.02)**	**(1.90)**	**(1.92)**	**(0.03)**	(2.06)	(2.05)	(-0.01)	**(0.28)**	**(0.49)**	**(0.21)**
Cr	1.89	1.83	-0.06	2.17	2.08	-0.09	2.19	2.09	-0.10	**1.93**	**1.77**	**-0.16**	**1.70**	**1.55**	**-0.15**	2.14	2.04	-0.10	2.01	1.85	-0.16
	(1.76)	(1.71)	(-0.05)	(1.81)	(1.71)	(-0.10)	(1.71)	(1.62)	(-0.09)	**(1.20)**	**(1.12)**	**(-0.09)**	**(1.00)**	**(0.93)**	**(-0.07)**	(1.62)	(1.49)	(-0.14)	(1.12)	(0.98)	(-0..15)
Mn	1.83	1.70	-0.12	1.64	1.55	-0.09	1.70	1.59	-0.11	1.63	1.48	-0.15	1.68	1.44	-0.24	1.57	1.38	-0.19	1.48	1.33	-0.14
	(1.29)	(1.16)	(-0.12)	(0.60)	(0.50)	(-0.10)	(0.66)	(0.48)	(-0.18)	(0.53)	(0.36)	(-0.17)	(0.37)	(0.47)	(0.09)	(0.37)	(0.53)	(0.16)	(0.39)	(0.54)	(0.15)
Fe	1.68	1.56	-0.12	1.58	1.48	-0.10	1.24	1.12	-0.12	1.34	1.32	-0.02	**0.75**	**0.84**	**0.08**	1.23	1.17	-0.06	1.04	1.06	0.02
	(0.59)	(0.51)	(-0.08)	(0.90)	(0.83)	(-0.07)	(0.85)	(0.77)	(-0.07)	(0.63)	(0.65)	(0.02)	**(0.82)**	**(0.82)**	**(0.00)**	(0.46)	(0.48)	(0.03)	(0.73)	(0.75)	(0.02)
Co	0.89	0.84	-0.05	0.88	0.85	-0.02	0.70	0.69	-0.01	0.94	0.90	-0.03	0.78	0.79	0.01	0.75	0.72	-0.03	0.53	0.38	-0.15
	(0.78)	(0.78)	(0.00)	(0.75)	(0.72)	(-0.03)	(0.61)	(0.64)	(0.03)	(0.70)	(0.68)	(-0.01)	(0.63)	(0.70)	(0.07)	(0.61)	(0.72)	(0.12)	(0.77)	(0.97)	(0.20)
Ni	0.33	0.31	-0.02	0.30	0.29	-0.02	0.35	0.30	-0.05	0.40	0.42	0.02	0.36	0.40	0.04	0.66	0.64	-0.02	0.79	0.55	-0.24
	(0.31)	(0.29)	(-0.02)	(0.69)	(0.66)	(-0.03)	(0.42)	(0.53)	(0.10)	(0.56)	(0.61)	(0.05)	(0.55)	(0.53)	(-0.02)	(0.63)	(0.27)	(-0.36)	(0.80)	(0.82)	(0.03)
Cu	0.76	0.73	-0.03	0.93	0.79	-0.15	0.84	0.85	0.01	0.97	0.95	-0.02	0.88	0.92	0.04	0.91	0.80	-0.11	0.65	0.69	0.04
	(0.81)	(0.81)	(0.00)	(1.01)	(0.82)	(-0.18)	(0.86)	(0.88)	(0.02)	(0.99)	(0.98)	(-0.01)	(1.03)	(0.96)	(-0.07)	(0.94)	(0.95)	(0.00)	(0.84)	(0.89)	(0.04)
Zn	0.93	0.94	0.01	1.01	0.99	-0.01	0.88	0.84	-0.03	1.08	1.02	-0.07	2.70	2.60	-0.10	1.01	0.95	-0.06	1.24	1.00	-0.24
	(0.96)	(0.94)	(-0.02)	(1.02)	(1.00)	(-0.02)	(0.90)	(0.86)	(-0.04)	(1.10)	(1.03)	(-0.07)	(2.75)	(2.62)	(-0.13)	(1.04)	(0.99)	(-0.06)	(1.11)	(1.05)	(-0.07)
Y	1.38	1.77	0.40	1.14	0.65	-0.50	1.30	0.56	-0.73	1.17	0.59	-0.58	1.54	0.86	-0.69	1.60	0.63	-0.98	2.31	0.66	-1.65
Zr	1.58	1.34	-0.25	2.04	0.67	-1.37	2.20	1.08	-1.12	2.17	0.75	-1.42	2.16	1.11	-1.05	2.76	1.14	-1.62	3.02	1.67	-1.34
Nb	2.07	1.30	-0.76	2.08	1.67	-0.41	2.73	1.78	-0.95	2.50	1.82	-0.69	**2.10**	**2.06**	**-0.03**	**2.77**	**2.60**	**-0.17**	**0.84**	**1.07**	**0.23**
Mo	2.13	1.99	-0.14	2.04	1.71	-0.34	**1.28**	**2.16**	**0.88**	**2.59**	**2.20**	**-0.39**	**2.18**	**1.96**	**-0.22**	**1.48**	**2.16**	**0.67**	**1.18**	**1.07**	**-0.11**
Tc	2.13	2.01	-0.12	**1.76**	**1.57**	**-0.19**	**1.19**	**1.57**	**0.39**	**1.97**	**1.69**	**-0.28**	**1.68**	**1.48**	**-0.20**	**1.49**	**1.07**	**-0.42**	**0.76**	**0.83**	**0.07**
Ru	1.84	1.49	-0.34	1.60	1.71	0.11	**1.01**	**1.61**	**0.60**	**1.02**	**0.83**	**-0.19**	**0.70**	**0.68**	**-0.02**	**0.89**	**0.78**	**-0.10**	**1.18**	**0.62**	**-0.57**
Rh	0.96	0.87	-0.09	1.23	1.19	-0.05	0.65	0.66	0.01	0.86	0.79	-0.07	0.75	0.83	0.08	0.69	0.64	-0.05	0.30	0.32	0.02
Pd	0.39	0.33	-0.05	0.47	0.47	0.00	0.59	0.59	0.00	0.63	0.43	-0.20	0.69	0.49	-0.20	0.71	0.72	0.01	1.04	0.80	-0.24
Ag	0.80	0.80	0.00	0.88	0.77	-0.11	0.90	0.93	0.03	0.96	0.90	-0.06	0.88	0.91	0.03	0.77	0.44	-0.33	0.80	0.81	0.01
Cd	1.01	0.64	-0.37	1.12	1.05	-0.07	1.09	1.04	-0.05	1.06	0.93	-0.13	1.04	1.05	0.01	1.11	1.05	-0.06	1.23	0.96	-0.27
La	1.10	-	-	0.89	-	-	0.85	-	-	0.81	-	-	1.08	-	-	1.21	-	-	1.82	-	-
Hf	1.90	1.03	-0.87	2.41	0.67	-1.74	2.60	1.08	-1.51	2.50	0.82	-1.68	2.55	1.02	-1.54	3.04	1.22	-1.82	3.16	1.56	-1.59
Ta	2.29	1.45	-0.83	2.51	1.77	-0.74	**1.55**	**2.77**	**1.21**	**1.58**	**2.29**	**0.71**	2.76	2.04	-0.72	**2.43**	**2.50**	**0.07**	**1.00**	**1.16**	**0.17**
W	2.28	2.10	-0.18	2.34	4.42	2.08	**0.87**	**2.29**	**1.42**	**1.53**	**0.99**	**-0.54**	**2.42**	**1.01**	**-1.41**	**1.57**	**0.88**	**-0.69**	**2.07**	**2.24**	**0.17**
Re	**1.76**	**2.09**	**0.33**	**1.86**	**1.69**	**-0.17**	**0.58**	**1.53**	**0.95**	**1.53**	**1.27**	**-0.26**	**1.77**	**1.66**	**-0.10**	**1.57**	**1.06**	**-0.51**	**1.18**	**0.97**	**-0.20**
Os	**1.43**	**1.33**	**-0.09**	**1.21**	**0.92**	**-0.29**	**1.03**	**0.87**	**-0.16**	**1.20**	**0.99**	**-0.21**	**1.32**	**1.37**	**0.05**	**1.30**	**1.09**	**-0.21**	**1.33**	**0.76**	**-0.58**
Ir	1.54	1.48	-0.06	1.67	1.18	-0.49	1.03	0.98	-0.05	1.15	0.97	-0.18	**0.82**	**0.89**	**0.07**	1.01	0.97	-0.04	0.67	0.70	0.03
Pt	1.02	0.94	-0.07	0.37	0.33	-0.04	0.29	0.25	-0.04	0.65	0.48	-0.17	0.65	0.47	-0.18	0.75	0.49	-0.25	1.07	0.77	-0.30
Au	1.93	1.87	-0.06	1.12	0.77	-0.35	0.94	0.93	-0.01	0.93	0.89	-0.05	0.76	0.62	-0.14	1.03	0.97	-0.06	0.95	0.99	0.04
Hg	0.92	0.99	0.07	0.90	0.83	-0.07	0.99	0.97	-0.02	*	*	*	0.64	0.40	-0.24	*	*	*	*	*	*

The SAC systems with blue marked values have unstable *OOH reaction intermediates which are calculated for the associative pathway-Path-II. * denotes unstable.

Table S29. Theoretical overpotential of ORR in the absence (vacuum) and presence of solvent (by implicit solvation model) for divacancy systems (RPBE(U)+TS results are in parentheses). The solvent effect is added: $\Delta\eta = \eta_{sol} - \eta_{vac}$).

TM	C_4			N_1C_3			N_2C_2			N_2C_2[b]			N_2C_2[c]			N_3C_1			N_4		
	$\eta^{ORR\text{-}vac}$	$\eta^{ORR\text{-}sol}$	$\Delta\eta^{ORR}$	$\eta^{ORR\text{-}vac}$	$\eta^{ORR\text{-}sol}$	$\Delta\eta^{ORR}$	$\eta^{ORR\text{-}vac}$	$\eta^{ORR\text{-}sol}$	$\Delta\eta^{ORR}$	$\eta^{ORR\text{-}vac}$	$\eta^{ORR\text{-}sol}$	$\Delta\eta^{ORR}$	$\eta^{ORR\text{-}vac}$	$\eta^{ORR\text{-}sol}$	$\Delta\eta^{ORR}$	$\eta^{ORR\text{-}vac}$	$\eta^{ORR\text{-}sol}$	$\Delta\eta^{ORR}$	$\eta^{ORR\text{-}vac}$	$\eta^{ORR\text{-}sol}$	$\Delta\eta^{ORR}$
Sc	2.01 (2.02)	1.26 (1.28)	-0.75 (-0.74)	1.86 (1.90)	0.77 (0.69)	-1.09 (-1.21)	2.15 (2.20)	0.77 (0.70)	-1.38 (-1.50)	2.13 (2.17)	0.70 (0.67)	-1.43 (-1.50)	2.34 (2.38)	0.97 (0.96)	-1.36 (-1.42)	2.48 (2.28)	0.91 (0.83)	-1.57 (-1.45)	3.21 (3.29)	1.43 (1.33)	-1.78 (-1.95)
Ti	2.34 (2.35)	0.79 (0.56)	-1.55 (-1.79)	2.82 (2.88)	1.28 (1.10)	-1.54 (-1.77)	2.97 (3.00)	1.38 (1.21)	-1.59 (-1.79)	2.96 (3.00)	1.34 (1.23)	-1.62 (-1.78)	2.95 (3.03)	1.40 (1.27)	-1.55 (-1.77)	3.36 (3.08)	1.90 (1.79)	-1.45 (-1.29)	3.32 (3.27)	2.00 (1.58)	-1.33 (-1.69)
V	2.61 (2.31)	1.69 (1.39)	-0.92 (-0.92)	2.69 (2.43)	1.76 (1.45)	-0.93 (-0.98)	2.84 (2.54)	1.83 (1.57)	-1.01 (-0.97)	**1.78 (1.20)**	**1.69 (1.28)**	**-0.09 (0.08)**	**1.37 (1.15)**	**1.38 (1.17)**	**0.01 (0.03)**	2.98 (2.10)	2.32 (1.81)	-0.65 (-0.29)	**0.83 (0.58)**	**0.47 (0.94)**	**-0.36 (0.36)**
Cr	2.16 (1.66)	1.66 (1.16)	-0.50 (-0.50)	2.32 (1.79)	1.97 (1.60)	-0.35 (-0.19)	2.06 (1.64)	1.84 (1.47)	-0.22 (-0.17)	**1.14 (0.97)**	**1.08 (1.09)**	**-0.06 (0.12)**	**0.94 (0.80)**	**0.88 (0.88)**	**-0.06 (0.08)**	1.90 (0.92)	1.96 (1.50)	0.06 (0.58)	1.40 (1.04)	1.60 (1.45)	0.20 (0.41)
Mn	2.15 (1.24)	1.54 (1.36)	-0.62 (0.12)	1.38 (0.79)	1.47 (0.89)	0.08 (0.10)	1.34 (0.79)	1.40 (0.92)	0.06 (0.13)	1.15 (0.72)	1.22 (0.83)	0.07 (0.11)	1.18 (0.91)	1.28 (1.02)	0.10 (0.11)	1.26 (0.97)	1.43 (1.13)	0.16 (0.16)	0.86 (0.99)	1.05 (1.21)	0.19 (0.22)
Fe	1.27 (1.12)	1.36 (1.21)	0.08 (0.10)	1.05 (0.81)	1.22 (1.04)	0.18 (0.23)	1.21 (0.65)	1.36 (0.81)	0.15 (0.16)	0.96 (0.63)	1.13 (0.83)	0.18 (0.20)	**0.64 (1.78)**	**0.72 (1.70)**	**0.08 (-0.07)**	1.04 (0.45)	1.24 (1.17)	0.20 (0.72)	0.67 (0.53)	0.85 (0.99)	0.18 (0.46)
Co	1.05 (0.66)	1.14 (0.79)	0.09 (0.14)	1.02 (0.39)	1.16 (0.55)	0.14 (0.16)	0.68 (0.44)	0.89 (0.66)	0.21 (0.22)	0.62 (0.39)	0.79 (0.61)	0.17 (0.22)	0.74 (0.55)	0.78 (1.30)	0.04 (0.74)	0.46 (0.65)	0.65 (1.20)	0.20 (0.55)	0.39 (0.62)	0.48 (0.85)	0.09 (0.23)
Ni	0.35 (0.37)	0.53 (0.48)	0.18 (0.11)	0.49 (0.37)	0.29 (0.28)	-0.20 (-0.10)	0.75 (0.83)	0.53 (0.53)	-0.22 (-0.30)	0.78 (0.80)	0.53 (0.52)	-0.25 (-0.27)	0.74 (0.79)	0.51 (0.51)	-0.23 (-0.28)	0.76 (1.08)	0.45 (0.57)	-0.32 (-0.51)	1.02 (1.08)	0.69 (0.77)	-0.32 (-0.31)
Cu	1.13 (1.17)	0.88 (0.91)	-0.25 (-0.26)	1.17 (1.17)	0.91 (0.92)	-0.26 (-0.26)	0.93 (0.96)	0.58 (0.61)	-0.35 (-0.35)	0.99 (1.03)	0.63 (0.66)	-0.36 (-0.36)	0.94 (0.99)	0.57 (0.61)	-0.37 (-0.38)	0.95 (1.29)	0.62 (0.62)	-0.33 (-0.67)	0.95 (0.98)	0.60 (0.63)	-0.35 (-0.35)
Zn	0.84 (0.78)	0.61 (0.55)	-0.23 (-0.23)	0.46 (0.44)	0.53 (0.57)	0.07 (0.13)	0.83 (0.80)	0.50 (0.49)	-0.33 (-0.30)	0.41 (0.42)	0.53 (0.56)	0.12 (0.14)	1.98 (2.01)	1.87 (1.89)	-0.11 (-0.12)	0.54 (1.01)	0.86 (0.79)	0.32 (-0.21)	0.67 (0.58)	1.02 (0.92)	0.35 (0.34)
Y	1.99	1.27	-0.72	1.88	1.02	-0.86	1.99	0.79	-1.20	2.01	0.77	-1.24	2.26	0.99	-1.27	2.35	0.97	-1.38	3.08	1.37	-1.70
Zr	2.25	0.71	-1.53	2.76	1.15	-1.60	2.93	1.25	-1.69	2.96	1.18	-1.78	2.99	1.31	-1.67	3.56	1.68	-1.88	3.80	2.07	-1.72
Nb	2.78	1.41	-1.37	2.82	1.59	-1.23	3.35	2.14	-1.20	3.30	2.00	-1.29	**1.31**	**1.28**	**-0.04**	**1.81**	**1.75**	**-0.07**	**1.39**	**1.38**	**-0.01**
Mo	2.50	1.79	-0.71	2.57	1.91	-0.66	**1.73**	**1.55**	**-0.18**	**1.68**	**1.44**	**-0.24**	**1.33**	**1.27**	**-0.06**	**1.49**	**1.40**	**-0.09**	**0.50**	**0.61**	**0.12**
Tc	1.86	1.64	-0.22	**1.58**	**1.46**	**-0.12**	**1.02**	**1.13**	**0.11**	**1.18**	**1.01**	**-0.17**	**1.16**	**0.98**	**-0.18**	**1.04**	**0.96**	**-0.08**	**0.66**	**0.44**	**-0.22**
Ru	1.57	1.54	-0.02	2.11	2.07	-0.04	**1.00**	**1.32**	**0.32**	**0.54**	**0.31**	**-0.22**	**0.70**	**0.67**	**-0.03**	**0.40**	**0.32**	**-0.09**	**0.94**	**0.67**	**-0.27**
Rh	1.29	1.40	0.11	0.81	0.96	0.15	0.92	1.13	0.21	0.90	1.07	0.17	0.80	0.82	0.03	0.42	0.56	0.14	0.27	0.44	0.17
Pd	0.33	0.35	0.02	0.35	0.36	0.01	0.83	0.59	-0.24	1.01	0.77	-0.24	1.02	0.77	-0.24	1.09	0.77	-0.32	1.19	0.84	-0.35
Ag	0.96	0.70	-0.26	1.14	0.86	-0.28	1.05	0.69	-0.35	1.13	0.77	-0.36	1.03	0.63	-0.40	1.05	0.70	-0.36	0.53	0.99	0.46
Cd	0.59	0.37	-0.22	1.05	1.33	0.28	0.46	0.63	0.17	1.03	1.01	-0.03	1.12	1.84	0.73	1.13	1.17	0.05	1.07	1.18	0.11
La	1.80	-	-	1.62	-	-	1.54	-	-	1.58	-	-	1.82	-	-	1.91	-	-	2.55	-	-
Hf	2.66	0.76	-1.89	3.20	1.29	-1.91	3.37	1.43	-1.94	3.41	1.38	-2.03	3.46	1.47	-1.99	3.88	1.91	-1.97	4.02	2.29	-1.73
Ta	3.30	1.63	-1.66	3.39	1.85	-1.54	**1.34**	**1.99**	**0.65**	**1.48**	**1.42**	**-0.06**	3.76	2.44	-1.33	**1.67**	**1.52**	**-0.15**	**1.69**	**1.82**	**0.14**
W	2.90	2.18	-0.73	3.10	2.32	-0.78	**1.89**	**1.69**	**-0.20**	**1.69**	**1.63**	**-0.07**	**1.35**	**1.32**	**-0.03**	**1.43**	**1.37**	**-0.06**	**1.28**	**1.70**	**0.42**
Re	**1.29**	**1.42**	**0.12**	**2.12**	**1.91**	**-0.21**	**0.40**	**1.25**	**0.85**	**1.22**	**1.12**	**-0.10**	**1.28**	**1.20**	**-0.08**	**1.48**	**0.98**	**-0.50**	**0.58**	**1.60**	**1.02**
Os	**1.22**	**1.12**	**-0.10**	**1.05**	**0.73**	**-0.32**	**0.82**	**0.89**	**0.07**	**1.18**	**0.94**	**-0.25**	**1.28**	**1.08**	**-0.20**	**0.92**	**0.69**	**-0.23**	**0.88**	**0.56**	**-0.32**
Ir	1.69	1.84	0.14	1.13	1.26	0.13	1.17	1.35	0.18	0.92	1.04	0.12	**0.62**	**0.65**	**0.03**	0.53	0.57	0.03	0.28	0.35	0.06
Pt	0.54	0.38	-0.17	0.38	0.35	-0.03	0.76	0.54	-0.22	1.10	0.87	-0.22	1.07	0.84	-0.23	1.10	0.82	-0.28	1.21	0.86	-0.35
Au	1.11	0.81	-0.30	1.19	0.85	-0.34	1.17	0.81	-0.36	1.18	0.82	-0.35	1.12	0.76	-0.36	1.13	0.78	-0.35	0.93	0.61	-0.31
Hg	0.90	0.69	-0.21	0.62	0.33	-0.29	1.52	1.78	0.26	*	*	*	0.90	0.96	0.06	*	*	*	*	*	*

The SAC systems with blue marked values have unstable *OOH reaction intermediates which are calculated for the associative pathway-Path-II. "*" denotes unstable.

Table S30. Predicted probable HER/OER/ORR overpotentials with error bound ($\eta \pm \varepsilon$) for M-N_nC_m moitifs.

$\eta = (\eta_{vac} + \eta_{sol})/2$ and $\varepsilon = \sqrt{\varepsilon_{vac-sol}{}^2 + \varepsilon_{vac}{}^2 + \varepsilon_{sol}{}^2}$ where $\eta_{vac/sol} = (\eta^{\circ}_{vac/sol} + \eta^{U}_{vac/sol})/2$, $\varepsilon_{vac/sol} = (|\eta^{\circ}_{vac/sol} - \eta^{U}_{vac/sol}|)/2$, and $\varepsilon_{vac-sol} = (|\eta_{vac} - \eta_{sol}|)/2$. Subscript "$_{vac}$" and "$_{sol}$" denote the cases in vacuum and solvent (ϵ=80), respectively, and superscripts "$^{\circ}$" and "^{U}" denote the cases without and with Hubbard effective parameter *U* (which was used only for 3d metals), respectively. In case of HER, η°_{vac} and η^{U}_{vac} are (ΔG_{H^*})/e without and with Hubbard effective parameter U at vacuum, respectively.

[Since the value of U in DFT calculations is used to obtain a reasonable band structure (but not a thermodynamic energy), the (U) needs be considered for the band structure, electronic properties and conductivity. Given that the electronic state becomes better, the thermodynamic energy is likely to be better-corrected toward the right direction, but in certain cases a biased electron density modification towards better electronic properties could degrade the quantitative energy evaluation due to a possible bias. In this regard, in the evaluation of $\boldsymbol{\eta}$, we may consider both U=0, and U values as the upper/lower limits, or simply consider their mid value with the error bar (ε) as the half of their difference. Regarding the solvent effect, in a bulk dielectric medium the implicit solvent model would be reliable. However, in ~1 nm-sized confined medium, the Debye screening is far from complete, and thus, the dielectric constant is much reduced due to the limited polarization arising from almost forbidden rotational mode of solvent molecules.[8] Indeed, the dielectric constant for the interfacial water can be reduced to only ~2.[9] Similarly, the rotationally forbidden surface water molecules at catalytic sites may not play the proper role of Debye screening and thus the water dielectric effect needs to be much reduced. In this regard, the implicit solvent effect could overestimate the effect of interfacial solvents on $\boldsymbol{\eta}$ of (TM)SA catalysts embedded on G_N/Gr. In this regard, we again consider both vacuum and implicit solvent models as the upper/lower limits, or the mid value with the error bar as the half of their difference.**]**

(a). $\eta^{HER} \pm \varepsilon$

Transition Metal	C_4	N_1C_3	N_2C_2	$N_2C_2{}^b$	$N_2C_2{}^c$	N_3C_1	N_4
Sc	-0.27 ± 0.04	-0.31 ± 0.03	-0.47 ± 0.01	-0.46 ± 0.02	-0.35 ± 0.00	-0.33 ± 0.06	-0.41 ± 0.07
Ti	-0.47 ± 0.04	-0.03 ± 0.02	-0.15 ± 0.03	-0.18 ± 0.03	-0.20 ± 0.03	-0.46 ± 0.08	-0.49 ± 0.05
V	-0.15 ± 0.14	-0.20 ± 0.11	-0.13 ± 0.07	-0.15 ± 0.03	-0.12 ± 0.02	-0.14 ± 0.08	-0.15 ± 0.02
Cr	-0.15 ± 0.10	-0.29 ± 0.08	-0.36 ± 0.06	-0.30 ± 0.10	-0.27 ± 0.11	-0.10 ± 0.05	-0.50 ± 0.18
Mn	-0.90 ± 0.00	-0.14 ± 0.08	-0.18 ± 0.12	-0.36 ± 0.09	-0.13 ± 0.02	-0.30 ± 0.08	-0.79 ± 0.26
Fe	-0.40 ± 0.25	-0.91 ± 0.57	-0.12 ± 0.05	-0.29 ± 0.06	-0.22 ± 0.01	-0.18 ± 0.04	-0.78 ± 0.34
Co	-0.34 ± 0.24	-0.37 ± 0.16	-0.17 ± 0.04	-0.07 ± 0.04	-0.15 ± 0.02	-0.15 ± 0.03	-0.55 ± 0.23
Ni	-0.68 ± 0.08	-0.70 ± 0.14	-0.05 ± 0.05	-0.06 ± 0.03	-0.34 ± 0.11	-0.26 ± 0.01	-1.60 ± 0.01
Cu	-0.31 ± 0.04	-0.24 ± 0.03	-0.12 ± 0.03	-0.24 ± 0.02	-0.11 ± 0.02	-0.10 ± 0.04	-1.37 ± 0.06
Zn	-0.18 ± 0.03	-0.75 ± 0.06	-0.11 ± 0.05	-0.50 ± 0.04	-0.40 ± 0.04	-0.26 ± 0.01	-1.03 ± 0.13
Y	-0.15	-0.31	-0.45	-0.28	-0.35	-0.48	-0.22
Zr	-0.51	0.00	-0.14	-0.36	-0.18	-0.71	-0.90
Nb	-0.24	-0.25	-0.66	-0.81	-0.70	-0.65	-0.74
Mo	-0.10	-0.12	-0.46	-0.64	-0.61	-0.48	-0.37
Tc	-0.20	-0.16	-0.17	-0.39	-0.44	-0.43	-0.36
Ru	-0.30	-0.46	-0.06	-0.64	-0.44	-0.51	-0.43
Rh	-0.06	-0.11	-0.06	-0.53	-0.42	-0.19	-0.07
Pd	-0.60	-0.79	-0.25	-0.16	-0.35	-0.29	-1.57
Ag	-0.70	-0.03	-0.17	-0.08	-0.14	-1.49	-0.36
Cd	-1.09	-0.08	-0.62	-0.16	-0.15	-0.06	-0.06
La	-0.09	-1.44	-0.44	-0.48	-0.36	-1.01	-0.43
Hf	-0.23	-0.33	-0.50	-0.71	-0.57	-0.98	-1.09
Ta	-0.67	-0.75	-1.00	-1.22	-1.04	-0.99	-1.00
W	-0.56	-0.61	-0.85	-1.04	-0.91	-0.86	-0.82
Re	-0.34	-0.61	-0.61	-0.77	-0.78	-0.78	-0.83
Os	-0.06	-0.82	-0.45	-0.96	-0.82	-0.76	-0.66
Ir	-0.21	-0.17	-0.23	-0.67	-0.73	-0.36	-0.21
Pt	-0.09	-0.39	-0.03	-0.20	-0.57	-0.43	-1.68
Au	-0.42	-0.23	-0.30	-0.18	-0.22	-0.71	-1.71
Hg	-0.18	-0.44	-0.78	*	-0.21	*	*

Table S30 (b). $\eta^{OER} \pm \varepsilon$

Transition Metal	C_4	N_1C_3	N_2C_2	$N_2C_2^b$	$N_2C_2^c$	N_3C_1	N_4
Sc	1.73 ± 0.41	1.00 ± 0.20	1.07 ± 0.40	0.98 ± 0.37	1.23 ± 0.38	1.06 ± 0.58	1.61 ± 0.88
Ti	1.09 ± 0.53	1.41 ± 0.68	1.74 ± 0.52	1.72 ± 0.45	1.84 ± 0.26	1.95 ± 0.36	2.39 ± 0.29
V	1.65 ± 0.15	1.99 ± 0.17	1.91 ± 0.29	**1.62 ± 0.50**	**1.62 ± 0.49**	2.16 ± 0.15	**0.77 ± 0.67**
Cr	1.80 ± 0.10	1.94 ± 0.26	1.90 ± 0.34	**1.51 ± 0.49**	**1.30 ± 0.47**	1.82 ± 0.38	1.49 ± 0.63
Mn	1.50 ± 0.39	1.07 ± 0.74	1.11 ± 0.76	1.00 ± 0.79	0.99 ± 0.82	0.96 ± 0.73	0.93 ± 0.67
Fe	1.08 ± 0.76	1.20 ± 0.47	**1.84 ± 0.90**	0.98 ± 0.49	**0.81 ± 0.04**	0.84 ± 0.52	0.90 ± 0.22
Co	0.82 ± 0.07	0.80 ± 0.09	0.66 ± 0.05	0.80 ± 0.16	0.72 ± 0.09	0.70 ± 0.07	0.66 ± 0.32
Ni	0.31 ± 0.02	0.49 ± 0.27	0.40 ± 0.12	0.50 ± 0.12	0.46 ± 0.11	0.55 ± 0.21	0.74 ± 0.15
Cu	0.78 ± 0.05	0.89 ± 0.09	0.86 ± 0.02	0.97 ± 0.02	0.95 ± 0.08	0.90 ± 0.08	0.77 ± 0.14
Zn	0.94 ± 0.01	1.00 ± 0.01	0.87 ± 0.02	1.06 ± 0.04	2.67 ± 0.06	1.00 ± 0.04	1.10 ± 0.10
Y	1.58 ± 0.20	0.89 ± 0.25	0.93 ± 0.80	0.88 ± 0.29	1.20 ± 0.34	1.12 ± 0.49	1.49 ± 0.82
Zr	1.46 ± 0.12	1.36 ± 0.69	1.64 ± 0.56	1.46 ± 0.71	1.63 ± 0.52	1.95 ± 0.81	2.34 ± 0.67
Nb	1.69 ± 0.38	1.87 ± 0.21	2.26 ± 0.47	2.16 ± 0.34	**2.08 ± 0.02**	**2.69 ± 0.09**	**0.95 ± 0.11**
Mo	2.06 ± 0.07	1.88 ± 0.17	**1.72 ± 0.44**	**2.39 ± 0.19**	**2.07 ± 0.11**	**1.82 ± 0.34**	**1.12 ± 0.05**
Tc	2.07 ± 0.06	**1.67 ± 0.09**	**1.38 ± 0.19**	**1.83 ± 0.14**	**1.58 ± 0.10**	**1.28 ± 0.21**	**0.80 ± 0.03**
Ru	1.66 ± 0.17	1.65 ± 0.06	**1.31 ± 0.30**	**0.92 ± 0.09**	**0.69 ± 0.01**	**0.83 ± 0.05**	0.90 ± 0.28
Rh	0.91 ± 0.05	1.21 ± 0.02	0.65 ± 0.01	0.83 ± 0.03	0.79 ± 0.04	0.66 ± 0.02	0.31 ± 0.01
Pd	0.36 ± 0.03	0.47 ± 0.00	0.59 ± 0.00	0.53 ± 0.10	0.59 ± 0.10	0.72 ± 0.00	0.92 ± 0.12
Ag	0.80 ± 0.00	0.83 ± 0.05	0.91 ± 0.01	0.93 ± 0.03	0.89 ± 0.01	0.60 ± 0.16	0.80 ± 0.01
Cd	0.82 ± 0.18	1.08 ± 0.03	1.07 ± 0.03	1.00 ± 0.06	1.05 ± 0.01	1.08 ± 0.03	1.10 ± 0.14
La	1.10^{vac}	0.89^{vac}	0.88^{vac}	0.81^{vac}	1.08^{vac}	1.21^{vac}	1.82^{vac}
Hf	1.46 ± 0.44	1.54 ± 0.87	1.84 ± 0.76	1.66 ± 0.84	1.79 ± 0.77	2.13 ± 0.91	2.36 ± 0.80
Ta	1.87 ± 0.42	2.14 ± 0.37	**2.16 ± 0.61**	**1.93 ± 0.36**	2.40 ± 0.36	**2.47 ± 0.04**	**1.08 ± 0.08**
W	2.19 ± 0.09	3.38 ± 1.04	**1.58 ± 0.71**	**1.26 ± 0.27**	**1.72 ± 0.70**	**1.23 ± 0.34**	**2.15 ± 0.08**
Re	**1.92 ± 0.16**	**1.78 ± 0.09**	**1.06 ± 0.48**	**1.40 ± 0.13**	**1.72 ± 0.05**	**1.31 ± 0.25**	**1.08 ± 0.10**
Os	**1.38 ± 0.05**	**1.07 ± 0.15**	**0.95 ± 0.08**	**1.10 ± 0.11**	**1.35 ± 0.02**	**1.20 ± 0.11**	**1.04 ± 0.29**
Ir	1.51 ± 0.03	1.43 ± 0.24	1.01 ± 0.03	1.06 ± 0.09	**0.86 ± 0.04**	0.99 ± 0.02	0.69 ± 0.02
Pt	0.98 ± 0.04	0.35 ± 0.02	0.27 ± 0.02	0.57 ± 0.09	0.56 ± 0.09	0.62 ± 0.13	0.92 ± 0.15
Au	1.90 ± 0.03	0.94 ± 0.18	0.94 ± 0.01	0.91 ± 0.02	0.69 ± 0.07	1.00 ± 0.03	0.97 ± 0.02
Hg	0.96 ± 0.04	0.86 ± 0.03	0.98 ± 0.01	*	0.52 ± 0.12	*	*

Table S30 (c). $\eta^{ORR} \pm \varepsilon$

Transition Metal	C_4	N_1C_3	N_2C_2	$N_2C_2^b$	$N_2C_2^c$	N_3C_1	N_4
Sc	1.64 ± 0.37	1.31 ± 0.58	1.46 ± 0.72	1.42 ± 0.73	1.66 ± 0.70	1.63 ± 0.76	2.32 ± 0.93
Ti	1.51 ± 0.84	2.02 ± 0.83	2.14 ± 0.85	2.13 ± 0.85	2.16 ± 0.83	2.53 ± 0.70	2.55 ± 0.78
V	2.00 ± 0.51	2.08 ± 0.52	2.20 ± 0.53	**1.49 ± 0.35**	**1.27 ± 0.15**	2.30 ± 0.56	**0.71 ± 0.27**
Cr	1.66 ± 0.43	1.92 ± 0.35	1.75 ± 0.29	**1.07 ± 0.09**	**0.88 ± 0.07**	1.57 ± 0.57	1.37 ± 0.25
Mn	1.57 ± 0.48	1.13 ± 0.42	1.11 ± 0.37	0.98 ± 0.30	1.10 ± 0.20	1.20 ± 0.22	1.03 ± 0.15
Fe	1.24 ± 0.12	1.03 ± 0.18	**1.01 ± 0.40**	0.89 ± 0.25	**1.21 ± 0.75**	0.98 ± 0.38	0.76 ± 0.19
Co	0.91 ± 0.27	0.78 ± 0.45	0.67 ± 0.20	0.60 ± 0.17	0.84 ± 0.34	0.74 ± 0.34	0.58 ± 0.23
Ni	0.43 ± 0.08	0.36 ± 0.10	0.66 ± 0.13	0.66 ± 0.13	0.64 ± 0.13	0.72 ± 0.27	0.89 ± 0.17
Cu	1.02 ± 0.13	1.04 ± 0.13	0.77 ± 0.18	0.83 ± 0.18	0.78 ± 0.19	0.87 ± 0.30	0.79 ± 0.18
Zn	0.70 ± 0.12	0.50 ± 0.06	0.65 ± 0.16	0.48 ± 0.07	1.94 ± 0.06	0.80 ± 0.24	0.80 ± 0.19
Y	1.63 ± 0.36	1.45 ± 0.43	1.39 ± 0.60	1.39 ± 0.62	1.62 ± 0.63	1.66 ± 0.69	2.23 ± 0.85
Zr	1.48 ± 0.77	1.95 ± 0.80	2.09 ± 0.84	2.07 ± 0.89	2.15 ± 0.84	2.62 ± 0.94	2.93 ± 0.86
Nb	2.09 ± 0.69	2.21 ± 0.61	2.74 ± 0.60	2.65 ± 0.65	**1.29 ± 0.02**	**1.78 ± 0.03**	**1.38 ± 0.00**
Mo	2.15 ± 0.35	2.24 ± 0.33	**1.64 ± 0.09**	**1.56 ± 0.12**	**1.30 ± 0.03**	**1.45 ± 0.05**	**0.55 ± 0.06**
Tc	1.75 ± 0.11	**1.52 ± 0.06**	**1.08 ± 0.06**	**1.09 ± 0.09**	**1.07 ± 0.09**	**1.00 ± 0.04**	**0.55 ± 0.11**
Ru	1.56 ± 0.01	2.09 ± 0.02	**1.16 ± 0.16**	**0.42 ± 0.11**	**0.68 ± 0.01**	**0.36 ± 0.04**	**0.80 ± 0.14**
Rh	1.34 ± 0.05	0.88 ± 0.07	1.03 ± 0.10	0.99 ± 0.09	0.81 ± 0.01	0.49 ± 0.07	0.36 ± 0.08
Pd	0.34 ± 0.01	0.36 ± 0.00	0.71 ± 0.12	0.89 ± 0.12	0.89 ± 0.12	0.93 ± 0.16	1.01 ± 0.17
Ag	0.83 ± 0.13	1.00 ± 0.14	0.87 ± 0.18	0.95 ± 0.18	0.83 ± 0.20	0.88 ± 0.18	0.76 ± 0.23
Cd	0.48 ± 0.11	1.19 ± 0.14	0.55 ± 0.08	1.02 ± 0.01	1.48 ± 0.36	1.15 ± 0.02	1.12 ± 0.05
La	1.80^{vac}	1.62^{vac}	1.54^{vac}	1.58^{vac}	1.82^{vac}	1.91^{vac}	2.55^{vac}
Hf	1.71 ± 0.95	2.25 ± 0.96	2.40 ± 0.97	2.39 ± 1.02	2.47 ± 0.99	2.90 ± 0.99	3.15 ± 0.87
Ta	2.46 ± 0.83	2.62 ± 0.77	**1.66 ± 0.32**	**1.45 ± 0.03**	3.10 ± 0.66	**1.59 ± 0.07**	**1.76 ± 0.07**
W	2.54 ± 0.36	2.71 ± 0.39	**1.79 ± 0.10**	**1.66 ± 0.03**	**1.33 ± 0.02**	**1.40 ± 0.03**	**1.49 ± 0.21**
Re	**1.36 ± 0.06**	**2.02 ± 0.10**	**0.82 ± 0.42**	**1.17 ± 0.05**	**1.24 ± 0.04**	**1.23 ± 0.25**	**1.09 ± 0.51**
Os	**1.17 ± 0.05**	**0.89 ± 0.16**	**0.86 ± 0.04**	**1.06 ± 0.12**	**1.18 ± 0.10**	**0.80 ± 0.11**	**0.72 ± 0.16**
Ir	1.77 ± 0.07	1.20 ± 0.07	1.26 ± 0.09	0.98 ± 0.06	**0.64 ± 0.02**	0.55 ± 0.02	0.31 ± 0.03
Pt	0.46 ± 0.08	0.37 ± 0.02	0.65 ± 0.11	0.99 ± 0.11	0.95 ± 0.11	0.96 ± 0.14	1.04 ± 0.18
Au	0.96 ± 0.15	1.02 ± 0.17	0.99 ± 0.18	1.00 ± 0.18	0.94 ± 0.18	0.96 ± 0.18	0.77 ± 0.16
Hg	0.79 ± 0.10	0.48 ± 0.14	1.65 ± 0.13	*	0.93 ± 0.03	*	*

Table S31. Promising M-N_nC_m moities for high-performing HER/OER/ORR and their predicted properties. Only outstanding properies are listed based on **Table S31** (Criteria: $|\eta^{HER}| < 0.1$V, $\eta^{OER/ORR} < 0.7$ V. Only thermodynamically/electrochemically stable systems are considered).

Bi/Tri-functional						
System	$E_{emb/coh}$	U_{ds}	η^{HER} (V)	η^{OER} (V)	η^{ORR} (V)	σ/σ^0 (pristine)
Ni-C_4	-1.15	0.32		0.31±0.02	0.43±0.08	0.750
Pd-C_4	-1.26	1.58		0.36±0.03	0.34±0.01	0.262
Pt-C_4	-1.85	2.10	-0.09±0.00		0.46±0.08	
Ni-N_1C_3	-2.48	0.98		0.49±0.27	0.36±0.10	
Pd-N_1C_3	-2.29	2.09		0.47±0.00	0.36±0.00	0.364
Pt-N_1C_3	-2.72	2.54		0.35±0.02	0.37±0.02	0.293
Co-N_2C_2	-3.48	1.46		0.66±0.05	0.67±0.20	
Ni-N_2C_2	-3.60	1.54	-0.05±0.05	0.40±0.12	0.66±0.13	
Rh-N_2C_2	-2.45	1.82	-0.06±0.00	0.65±0.01		0.687
Pt-N_2C_2	-3.19	2.77	-0.03±0.00	0.27±0.02	0.65±0.11	
Ni-N_2C_2[b]	-3.10	1.29	-0.06±0.03	0.50±0.12	0.66±0.13	
Ni-N_2C_2[c]	-2.49	0.98		0.46±0.11	0.64±0.13	
Rh-N_3C_1	-1.45	1.33		0.66±0.02	0.49±0.07	
Co-N_4	-2.45	0.95		0.66±0.32	0.58±0.23	
Rh-N_4	-0.76	0.98	-0.07±0.00	0.31±0.01	0.36±0.08	0.237
Ir-N_4	-0.44	1.31		0.69±0.02	0.31±0.03	0.239
Monofunctional						
System	$E_{emb/coh}$	U_{ds}	η^{HER} (V)	η^{OER} (V)	η^{ORR} (V)	σ/σ^0 (pristine)
Zn-C_4	-1.55	0.01			0.70±0.12	
Rh-C_4	-1.26	1.23	-0.06±0.00			0.111
Os-C_4	-0.48	0.90	-0.06±0.00			
Zn-N_1C_3	-2.14	0.31			0.50±0.06	0.261
Ag-N_1C_3	-1.35	2.15	-0.03±0.00			
Hg-N_1C_3	-0.49	1.29			0.48±0.14	0.542
Zn-N_2C_2	-3.16	0.82			0.65±0.16	
Ru-N_2C_2	-1.69	1.31	-0.06±0.00			
Pd-N_2C_2	-3.02	2.46		0.59±0.00		
Cd-N_2C_2	-1.67	0.43			0.55±0.08	
Co-N_2C_2[b]	-3.00	1.22	-0.07±0.04		0.60±0.17	
Zn-N_2C_2[b]	-2.24	0.36			0.48±0.07	
Ru-N_2C_2[b]	-0.60	0.76			0.42±0.11	
Pd-N_2C_2[b]	-2.66	2.28		0.53±0.10		
Ag-N_2C_2[b]	-1.31	2.11	-0.08±0.00			
Pt-N_2C_2[b]	-2.91	2.63		0.57±0.09		
Pd-N_2C_2[c]	-1.98	1.94		0.59±0.10		
Ir-N_2C_2[c]	-0.71	1.40			0.64±0.02	0.179
Pt-N_2C_2[c]	-2.15	2.26		0.56±0.09		
Au-N_2C_2[c]	-1.43	1.98		0.69±0.07		0.475
Cr-N_3C_1	-3.52	0.85	-0.10±0.05			0.476
Co-N_3C_1	-3.08	1.26		0.70±0.07		0.487
Ni-N_3C_1	-3.12	1.30		0.55±0.21		0.510
Cu-N_3C_1	-1.99	1.34	-0.10±0.04			
Ru-N_3C_1	-0.21	0.57			0.36±0.04	
Ag-N_3C_1	-0.67	1.47		0.60±0.16		
Cd-N_3C_1	-0.87	0.03	-0.06±0.00			0.576
Ir-N_3C_1	-1.28	1.59			0.55±0.02	
Pt-N_3C_1	-2.55	2.45		0.62±0.13		0.383

Table S32. HER: Mean and standard deviation values of the Area Under the Receiver Operating Characteristic Curve (ROC AUC) for different machine learning methods: KNN—k-nearest neighbors , RNC—Radius Neighbors Classifier, RF—Random Forest, ET—Extremely Randomized Trees, GB—Gradient Boosting, SVC—Support Vector Classification, NN—Neural Network, DT—Decision Trees, LR—Logistic Regression, NB—Gaussian Naïve Bayes.

Classifier	Elemental properties		DFT-optimized geometry	
	ROC AUC mean	ROC AUC sd	ROC AUC mean	ROC AUC sd
RF	0.79	0.05	0.88	0.05
SVC	0.77	0.06	0.86	0.06
KNN	0.76	0.07	0.91	0.04
RNC	0.66	0.09	0.90	0.04
ET	0.76	0.08	0.87	0.05
GB	0.76	0.07	0.87	0.06
NN	0.75	0.07	0.82	0.07
DT	0.71	0.08	0.76	0.07
LR	0.7	0.08	0.71	0.06
NB	0.64	0.09	0.64	0.08

Table S33. OER: Mean and standard deviation values of the Area Under the Receiver Operating Characteristic Curve (ROC AUC) for different machine learning methods: KNN—k-nearest neighbors , RNC—Radius Neighbors Classifier, RF—Random Forest, ET—Extremely Randomized Trees, GB—Gradient Boosting, SVC—Support Vector Classification, NN—Neural Network, DT—Decision Trees, LR—Logistic Regression, NB—Gaussian Naïve Bayes.

Classifier	Elemental properties		DFT-optimized geometry					
	ROC AUC		*O - ROC AUC		*O, *OH - ROC AUC		*O, *OH, *OOH - ROC AUC	
	mean	sd	mean	sd	mean	sd	mean	sd
RF	0.87	0.06	0.86	0.07	0.87	0.07	0.89	0.06
SVC	0.84	0.08	0.87	0.06	0.87	0.08	0.84	0.07
KNN	0.80	0.07	0.87	0.06	0.86	0.06	0.83	0.07
RNC	0.79	0.09	0.87	0.06	0.84	0.07	0.82	0.08
ET	0.85	0.07	0.84	0.07	0.86	0.07	0.86	0.07
GB	0.87	0.05	0.84	0.07	0.85	0.07	0.86	0.08
NN	0.79	0.08	0.83	0.09	0.80	0.08	0.80	0.08
DT	0.84	0.07	0.84	0.07	0.85	0.07	0.80	0.07
LR	0.75	0.08	0.79	0.09	0.78	0.08	0.78	0.09
NB	0.64	0.11	0.75	0.09	0.81	0.08	0.80	0.08

Table S34. ORR: Mean and standard deviation values of the Area Under the Receiver Operating Characteristic Curve (ROC AUC) for different machine learning methods: KNN—k-nearest neighbors , RNC—Radius Neighbors Classifier, RF—Random Forest, ET—Extremely Randomized Trees, GB—Gradient Boosting, SVC—Support Vector Classification, NN—Neural Network, DT—Decision Trees, LR—Logistic Regression, NB—Gaussian Naïve Bayes.

Classifier	Elemental properties		DFT-optimized geometry					
	ROC AUC		*O - ROC AUC		*O, *OH - ROC AUC		*O, *OH, *OOH - ROC AUC	
	mean	sd	mean	sd	mean	sd	mean	sd
RF	0.81	0.08	0.81	0.08	0.82	0.07	0.87	0.06
SVC	0.80	0.07	0.81	0.07	0.83	0.07	0.84	0.07
KNN	0.79	0.08	0.84	0.07	0.84	0.07	0.82	0.07
RNC	0.72	0.10	0.85	0.07	0.84	0.06	0.82	0.09
ET	0.81	0.07	0.83	0.06	0.83	0.07	0.84	0.07
GB	0.82	0.08	0.78	0.07	0.81	0.07	0.86	0.07
NN	0.78	0.08	0.79	0.09	0.78	0.07	0.80	0.09
DT	0.79	0.08	0.76	0.08	0.80	0.09	0.82	0.10
LR	0.80	0.07	0.81	0.08	0.83	0.08	0.82	0.07
NB	0.72	0.10	0.77	0.10	0.81	0.07	0.83	0.09

References

1. Y.-W. Tan, Y. Zhang, K. Bolotin, Y. Zhao, S. Adam, E. H. Hwang, S. Das Sarma, H. L. Stormer and P. Kim, *Phys. Rev. Lett.* 2007, **99**, 10.
2. J. K. Nørskov, T. Bligaard, A. Logadottir, J. R. Kitchin, J. G. Chen, S. Pandelov and U. Stimming, *J. Electrochem. Soc.* 2005, **152**, J23.
3. L. M. Mentel, *Mendeleev - A Python resource for properties of chemical elements, ions and isotopes*, https://github.com/lmmentel/mendeleev, (accessed April 2021).
4. M. Rupp, A. Tkatchenko, K.-R. Müller and O. A. von Lilienfeld, *Phys. Rev. Lett.* 2012, **108**, 058301.
5. M. Dwass, *Ann. Math. Stat.* 1957, **28**, 181.
6. F. Pedregosa, G. Varoquaux, A. Gramfort, V. Michel, B. Thirion, O. Grisel, M. Blondel, P. Prettenhofer, R. Weiss, V. Dubourg, J. Vanderplas, A. Passos, D. Cournapeau, M. Brucher and M. Perrot, *J. Mach. Learn. Res.* 2011, **12**, 2825.
7. D. K. McClish, *Med. Decis. Making*. 1989, **9** (3), 190.
8. M. H. Motevaselian and N. R. Aluru, *ACS Nano* 2020, **14**, 12761−12770.
9. L. Fumagalli, A. Esfandiar , R. Fabregas, S. Hu, P. Ares, A. Janardanan, Q. Yang, B. Radha, T. Taniguchi , K. Watanabe, G. Gomila, K. S. Novoselov and A. K. Geim, *Science* 2018, **360**, 1339–1342.