Supporting Information

On the Origin of Surface Reconstruction via Oxygen Termination and Improved Hydrogen Evolution Reaction in MXenes

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Contents

Supplementary Notes

References

Supplementary Figures

Supplementary Tables

Supplementary Multimedia

Supplementary Notes

Supplementary Note 1: Description of hydrogen adsorption sites

Screening of the catalyst was done only by considering the ground state configuration of both the termination groups. After determining the ground state configuration for termination groups for all MXenes, we can distribute the MXene in three categories based on the placement of the termination groups as on-top type, fcc type, and hcp type MXene. In figure S2, we have shown three representative cases from each category to understand their structure and available sites for the hydrogen adsorption.

Figure S2 (a-b) is $Mo_3C_2F_2$, in which -F is terminated over the top of metal atoms from the upper layer. In this type of MXene, hydrogen adsorption will occur over three sites, as marked in the figure. Since this is on-top terminated MXene hydrogen adsorption over termination atom is on-top adsorption as marked 1, while the 2 and 3 correspond to hydrogen adsorption over the fcc site (over the middle layer metal atom) and hcp site (over the carbon atom). Figure S2 (c-d) shows $Mo_3C_2O_2$, in which -O is terminated over the top of the carbon atom (hcp site). In this case, hydrogen adsorption over -O termination would be the same as adsorption over the hcp site. While 2 and 3 markings denote the hydrogen adsorption over the on-top and fcc sites. At last, Figure S2 (e-f) is $Ti_3C_2O_2$, in which -O is terminated over the middle layer transition metal atom, also known as the fcc site. In this scenario, hydrogen adsorption over the -O termination would be similar to adsorption over the fcc site as marked as 1, while 2 and 3 correspond to hydrogen adsorption over the on-top site and hcp site, respectively.

Supplementary Note 2: Calculation of adsorbate-adsorbate interaction parameter using DFT derived cluster energies

$$E_* + \left(\mu(O) + E_{ad}^{(O)} \right) + \delta E = E_0 \qquad \text{[S1]}$$

$$E_* + 2\left(\mu(O) + E_{ad}^{(O)} \right) + J_{21} + \delta E = E_{21} \qquad \text{[S2]}$$

$$E_* + 2\left(\mu(O) + E_{ad}^{(O)}\right) + J_{22} + \delta E = E_{22} \qquad\qquad\text{[S3]}$$

$$E_* + 2\left(\mu(O) + E_{ad}^{(O)}\right) + J_{23} + \delta E = E_{23} \qquad\qquad\text{[S4]}$$

$$E_* + 2\left(\mu(O) + E_{ad}^{(O)}\right) + J_{24} + \delta E = E_{24} \qquad\qquad\text{[S5]}$$

$$E_* + 2\left(\mu(O) + E_{ad}^{(O)}\right) + J_{25} + \delta E = E_{25} \qquad\qquad\text{[S6]}$$

$$E_* + 3\left(\mu(O) + E_{ad}^{(O)}\right) + 3J_{21} + J_{31} + \delta E = E_{31} \qquad\qquad\text{[S7]}$$

$$E_* + 3\left(\mu(O) + E_{ad}^{(O)}\right) + 2J_{21} + J_{22} + J_{32} + \delta E = E_{32} \qquad\qquad\text{[S8]}$$

$$E_* + 3\left(\mu(O) + E_{ad}^{(O)}\right) + 2J_{21} + J_{23} + J_{33} + \delta E = E_{33} \qquad\qquad\text{[S9]}$$

$$E_* + 3\left(\mu(O) + E_{ad}^{(O)}\right) + 3J_{21} + J_{34} + \delta E = E_{34} \qquad\qquad\text{[S10]}$$

$$E_* + 3\left(\mu(O) + E_{ad}^{(O)}\right) + J_{21} + J_{22} + J_{24} + J_{35} + \delta E = E_{35} \qquad\qquad\text{[S11]}$$

$$E_* + 4\left(\mu(O) + E_{ad}^{(O)}\right) + 5J_{21} + J_{22} + J_{31} + 2J_{32} + J_{34} + J_{41} + \delta E = E_{41} \qquad\qquad\text{[S12]}$$

Supplementary Note 3: Monte Carlo simulation to Compute the heat of formation

To determine the quantity, like heat of formation, we have used the Monte Carlo simulation to explore the configuration space of oxygen distribution using series expansion of energy functional. To determine the energy of each configuration of oxygen, various singlet, doublet (pairwise J_{ij}), triplet (three-body J_{ijk}), and quadruplet (four-body (J_{ijkl})) clusters are used as shown in figure S2. The detailed algorithm of Monte Carlo simulation (Figure S7) contains the subroutine to compute the energy of the system with a given distribution of oxygen adsorbates. Calculation of energy of given configuration $\sigma(i, j)$ requires the adsorbate-adsorbate interaction parameter, which can be derived from the set of equations S4 to S15 using DFT derived cluster energies.

The main program has two different loops to evaluate the quantities, like ΔH_f and θ_{eq}. The program systematically scans θ to evaluate energy $E(\theta)$. The initial loop mainly evaluates the

energy cost to fill each and every lattice site by oxygen adsorbate for given θ. In this loop, each and every site is filled by oxygen adsorbate to compute the energy to fill a particular lattice site. Later, this site is reset as an empty site.

In the final loop, a randomly lattice site is selected and filled by oxygen adsorbate. This final loop goes over a statistically large run. In each run, a randomly different site filled with oxygen adsorbate and energy and a lattice site is stored if subsequent runs lead to an energetically more stable configuration. The final loop determines the energetically favourable configuration for given θ. At last, the final loop finishes by updating the lattice configuration corresponding to an energetically favourable lattice site. At the end, quantities like $E(\sigma)$ and $\Delta H_f(\sigma)$ are determined corresponding to given θ. This program again restarts with new θ after resetting quantities like E(i, j), and E_{min}. This program ends after reaching the condition of full oxygen coverage.

Supplementary Note 4: Zero-point energy calculation using mass weighted hessian matrix

To calculate the Gibbs free energy of hydrogen adsorption or any other adsorbate includes the calculation of the change in zero-point energy for the adsorbate. Zero-point energy of adsorbate in gas phase can be calculated by phonon calculation using vibrational modes associated with gas molecules. But over the surface of the catalyst, the entire phonon calculation could become computationally costly and more complex. Hence, we have manually computed the required block of hessian matrix to calculate the associated vibrational modes of adsorbate over the surface of the catalyst. In this method, we consider the catalyst surface as static and only compute the hessian corresponding to adsorbate via the finite difference method. In relation to the total energy of the system, the second derivative can be defined as,

$$k_{ij} = \left(\frac{\partial^2 E}{\partial q_i \partial q_j} \right) \qquad \text{[S13]}$$

Harmonic vibrational frequencies $\{v_i\}$ of an adsorbate or a molecule over the surface can be evaluated using,

$$v_i = \left(\frac{\lambda_i^{1/2}}{2\pi} \right) \qquad \text{[S14]}$$

Here, λ_i are eigenvalues corresponding to the mass-weighted Hessian matrix (K'),

$$K'_{ij} = \frac{1}{\sqrt{m_i m_j}} k_{ij} \qquad [S15]$$

By diagonalization of a mass weighted Hessian matrix, one can compute the eigenvalues of the Hessian matrix to evaluate the vibrational modes of an adsorbate or a molecule over a catalyst surface. In this method, we actually compute a sub-block of the Hessian matrix[1] and from that, one can compute the zero-point energy of an adsorbate or a molecule over the surface of the catalyst. Apart from zero-point energy calculation, partial Hessian vibrational analysis can be conducted to determine the stable equilibrium of the adsorbate over a specific site of the catalyst.

Supplementary Note 5: Additional Computational details

In order to initially screen the M_3C_2 MXene, hydrogen evolution reaction (HER) activity was determined for pristine as well as -F and -O terminated MXenes. To consider the effect of hydrogen coverage, two different hydrogen coverages were used during the calculation of hydrogen adsorption energy. 2×2×1 supercell and unitcell were used for low hydrogen coverage (θ=1/4) and high hydrogen coverage (θ=1), respectively. Wave function cutoff, charge density cutoff, k-mesh, and vacuum are consistent, as mentioned in the main manuscript.

References

1. Li, H. and Jensen, J.H., 2002. Partial Hessian vibrational analysis: the localization of the molecular vibrational energy and entropy. Theoretical Chemistry Accounts, **107**, pp.211-219.

Supplementary Figures
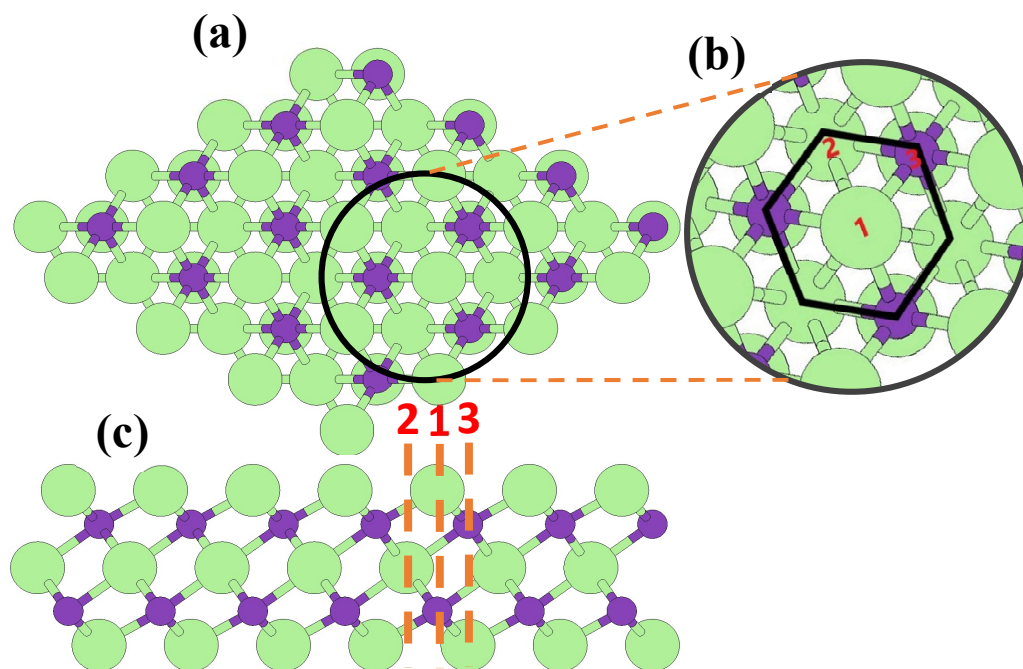
Figure S1 Schematic Diagram of (a) top view of M_3C_2 MXene, (b) Pop up shows the possible sites for the placement of the termination groups (1) on-top site (2) fcc site (3) hcp site; (c) side view of M_3C_2 MXene [here, green colour atom is early transition metal atom and purple colour atom is carbon.]
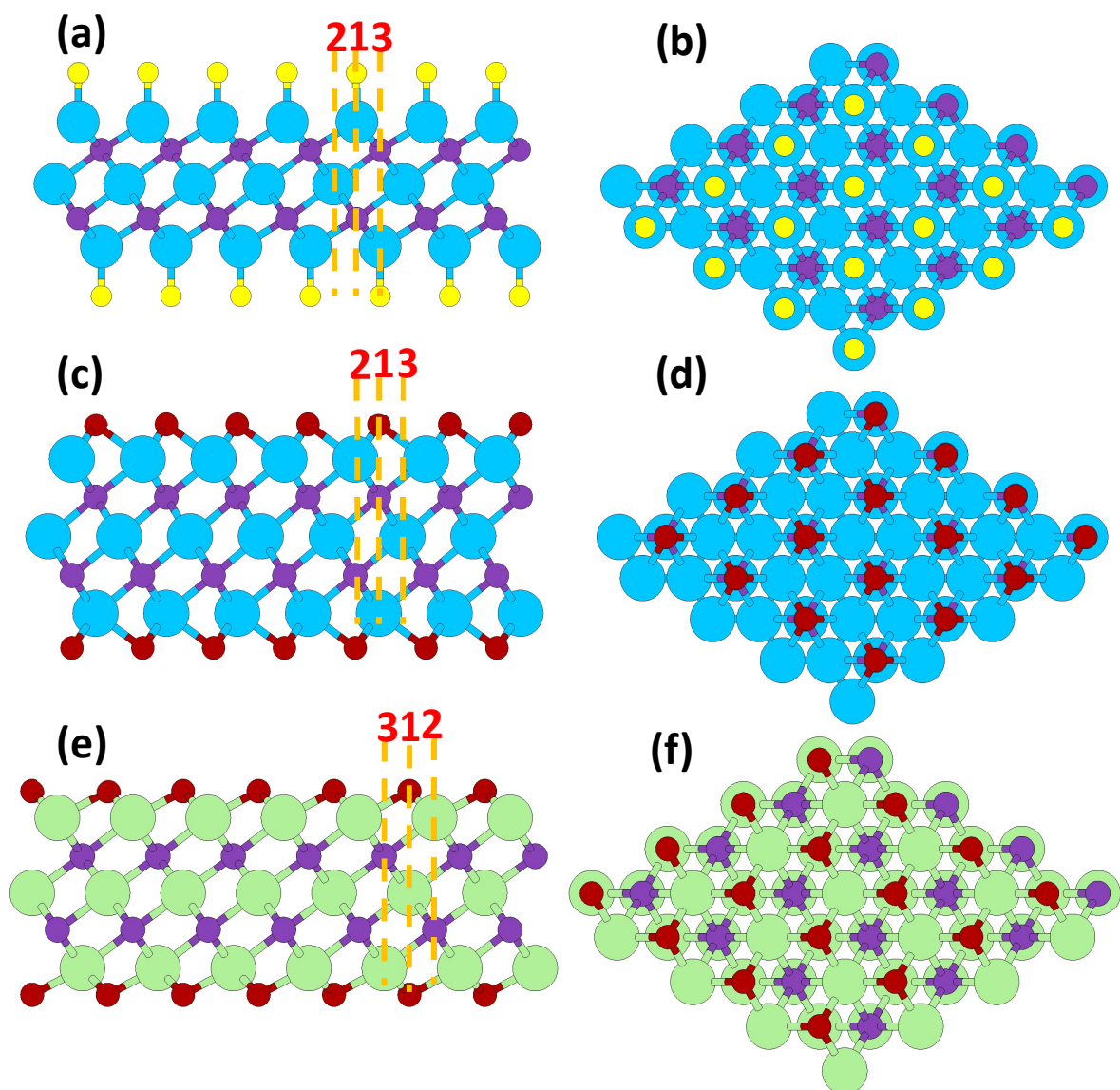
Figure S2 2D Schematic Diagram of $Mo_3C_2F_2$ with (a) side view (b) top view; $Mo_3C_2O_2$ with (c) side view (d) top view; $Ti_3C_2O_2$ with (e) side view (f) top view [sky blue, light green, purple, red, yellow are Mo, Ti, C, O, F, respectively].
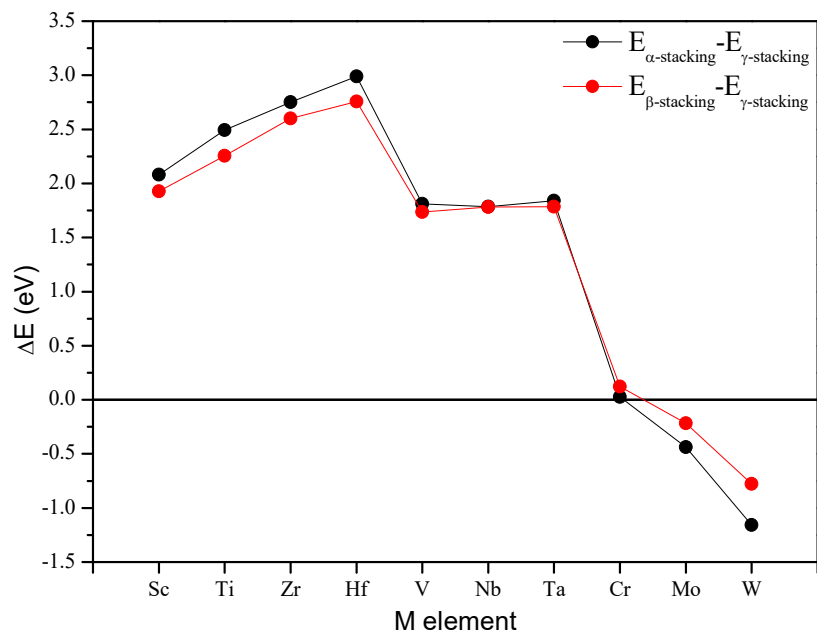
Figure S3 Comparative stacking energy of different stackings for early transition-metal based M_3C_2 MXenes.
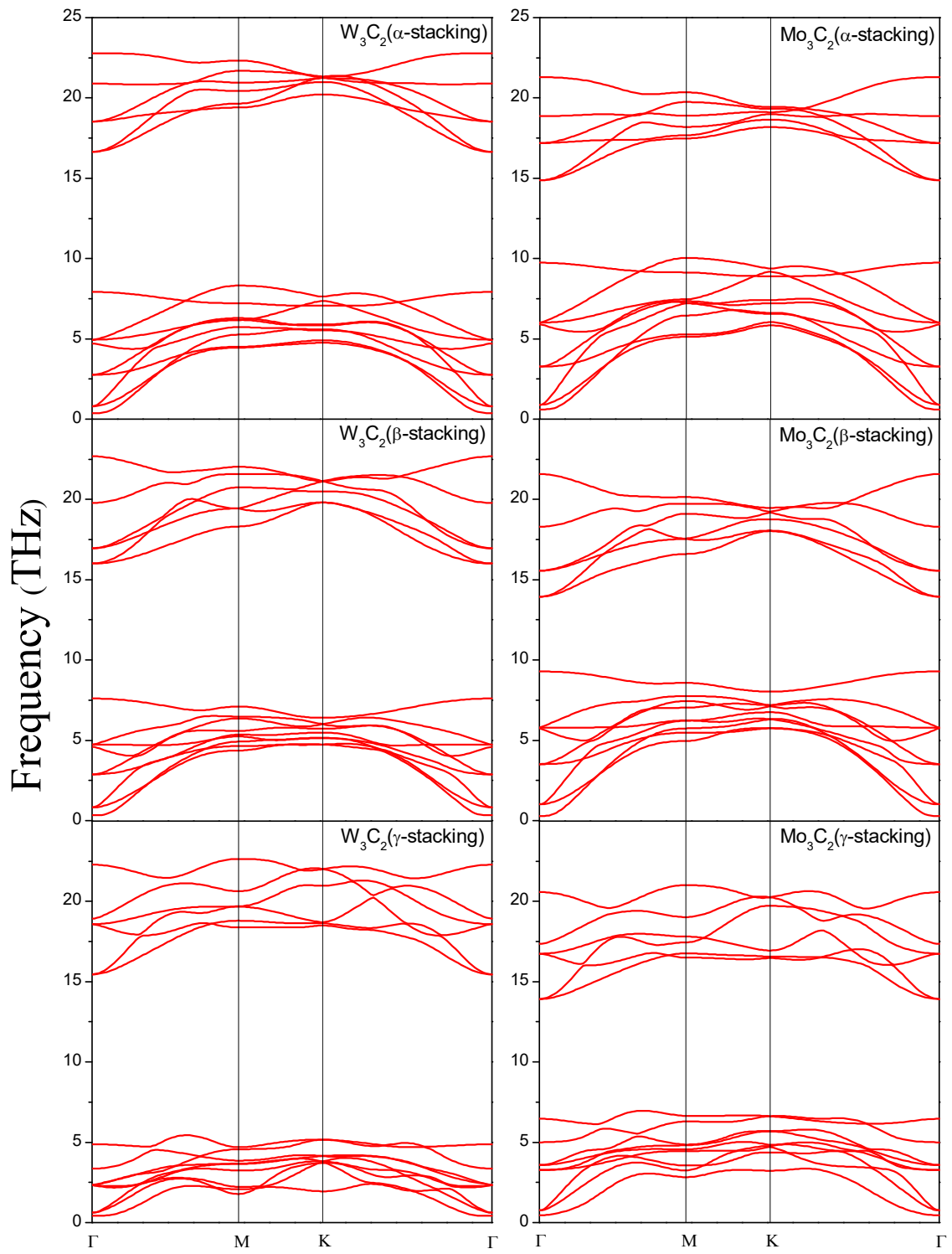
Figure S4: Phonon dispersion of three different stackings of pristine W_3C_2 and Mo_3C_2 MXenes.
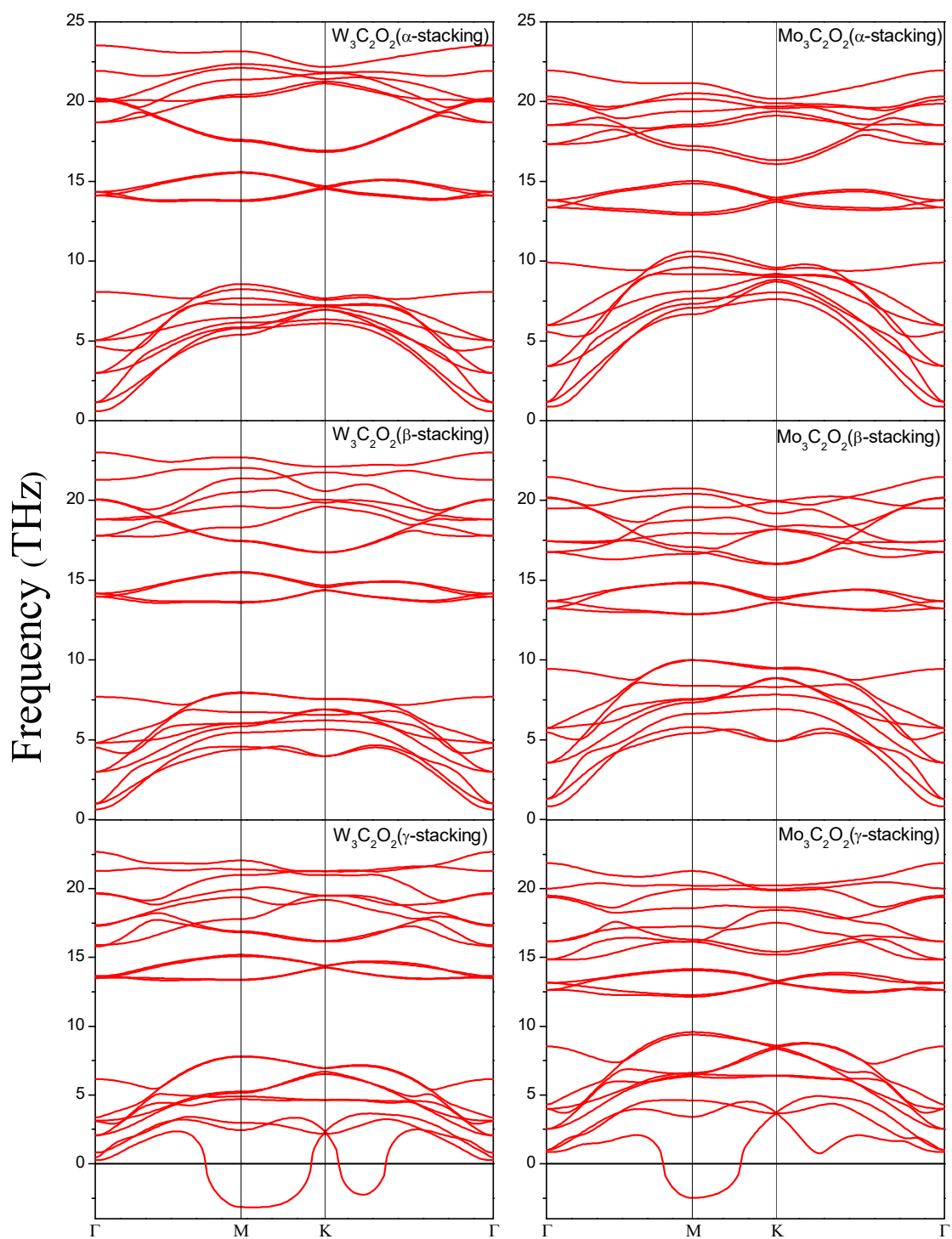
Figure S5: Phonon dispersion of three different stackings of pristine $W_3C_2O_2$ and $Mo_3C_2O_2$ MXenes.
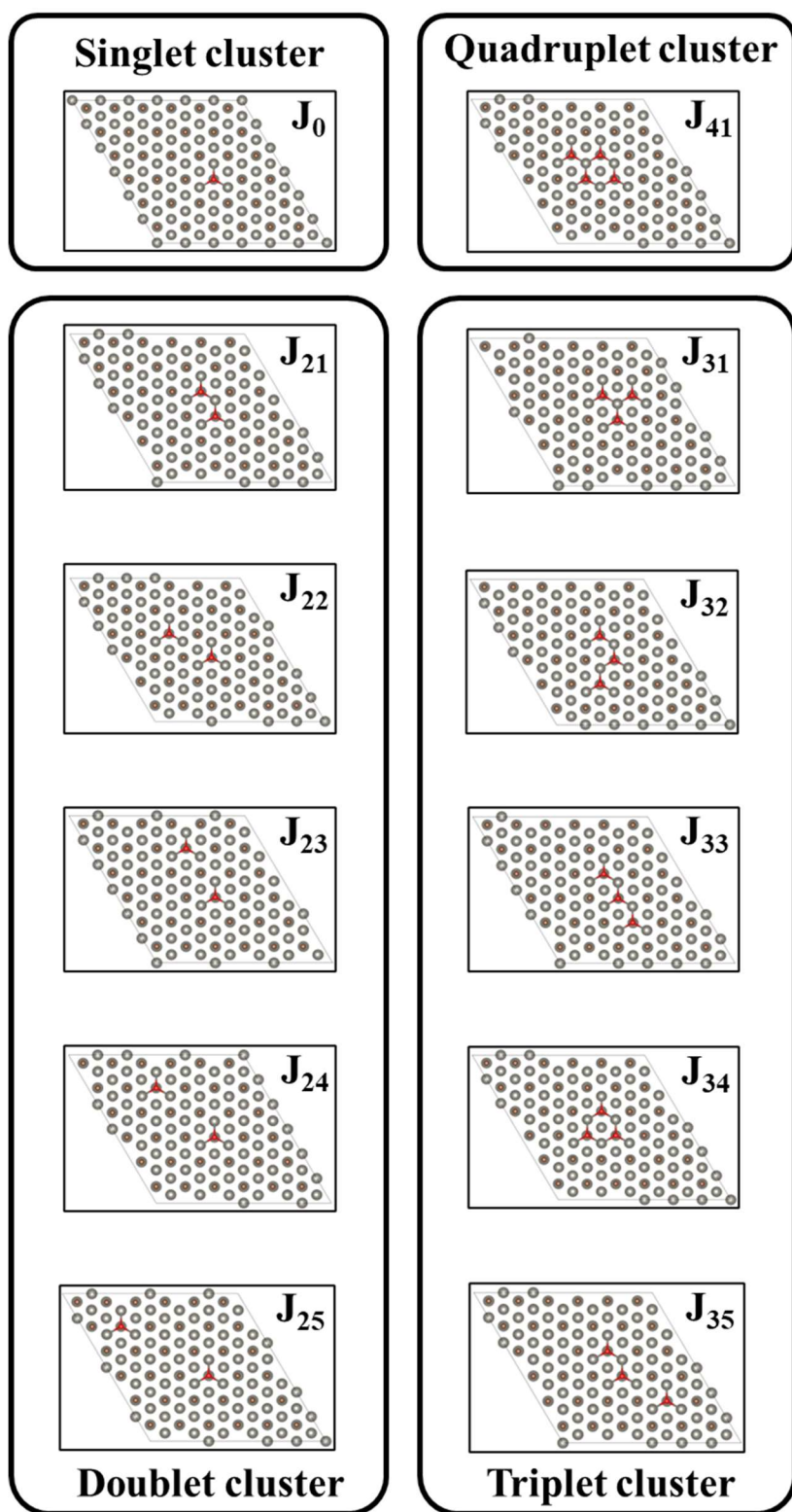
Figure S6: Considered singlet, doublet (pairwise J_{ij}), triplet (three-body J_{ijk}), and quadruplet (four-body (J_{ijkl})) clusters for adsorbate-adsorbate interaction parameters for oxygen over the surface of MXene.
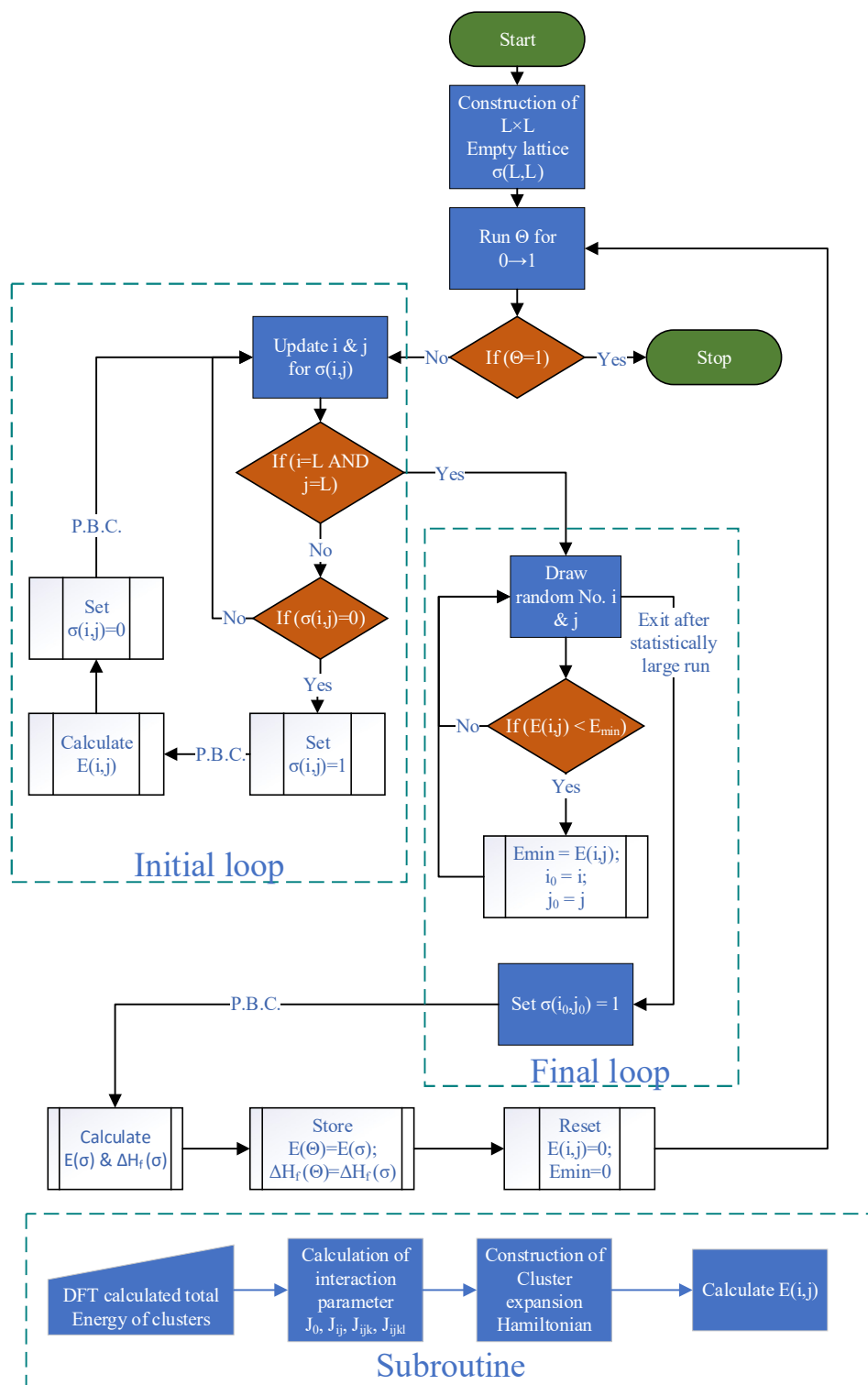
Figure S7: Algorithm to compute the Heat of formation (ΔH_f) using interaction parameters derived via density functional calculation. [Here, Θ is adsorbate coverage, and $\sigma(i,j)$ defines the lattice sites. P.B.C. refers to periodic boundary condition.]
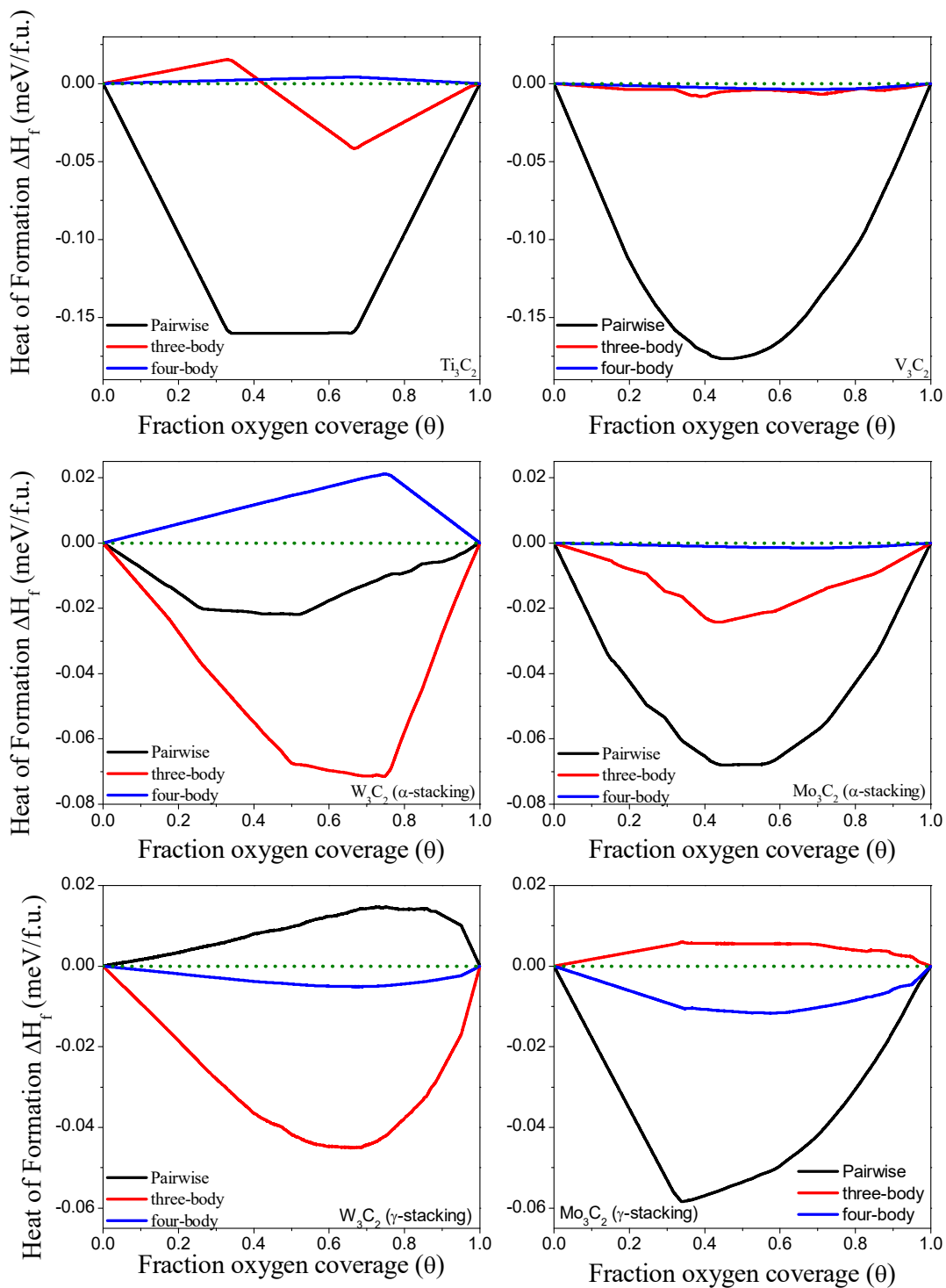
Figure S8: Distribution of Heat of formation in its pairwise, three-body, and four-body components with fractional oxygen coverage over surface of pristine MXene [calculated using interactive lattice simulation with 80×80 lattice of MXene.]

γ-Ti$_3$C$_2$O$_{2\theta}$

$\theta_{eq} = 0.659$

γ-V$_3$C$_2$O$_{2\theta}$

$\theta_{eq} = 0.450$

γ-W$_3$C$_2$O$_{2\theta}$

$\theta_{eq} = 0.568$

γ-Mo$_3$C$_2$O$_{2\theta}$

$\theta_{eq} = 0.338$

α-W$_3$C$_2$O$_{2\theta}$

$\theta_{eq} = 0.509$

α-Mo$_3$C$_2$O$_{2\theta}$

$\theta_{eq} = 0.443$
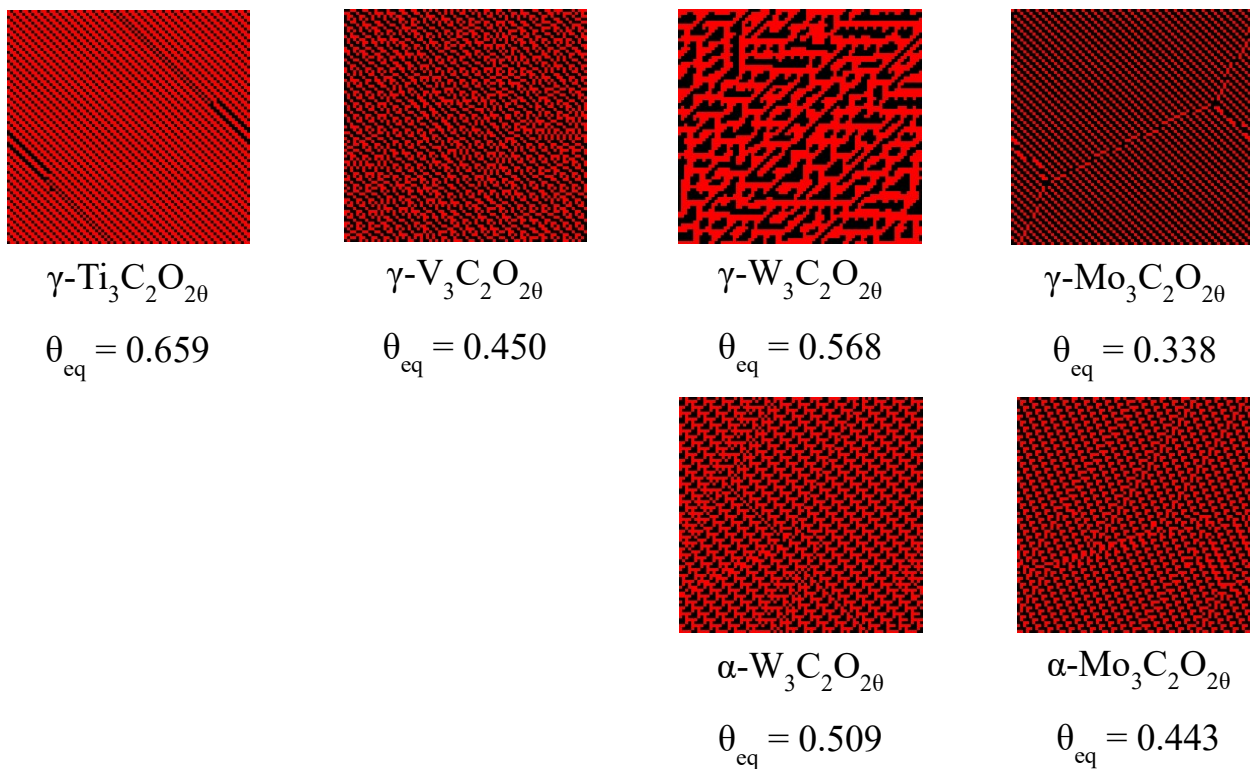
Figure S9: Calculated surface configuration oxygen over MXene at equilibrium coverage. [Here, the black colour surface is a suitable lattice site for oxygen termination, and the red dot indicates the presence of oxygen at the lattice site. For Ti$_3$C$_2$O$_{2\theta}$ and V$_3$C$_2$O$_{2\theta}$, a suitable lattice site refers to the fcc site, while for W$_3$C$_2$O$_{2\theta}$ and Mo$_3$C$_2$O$_{2\theta}$, it indicates the hcp site.]
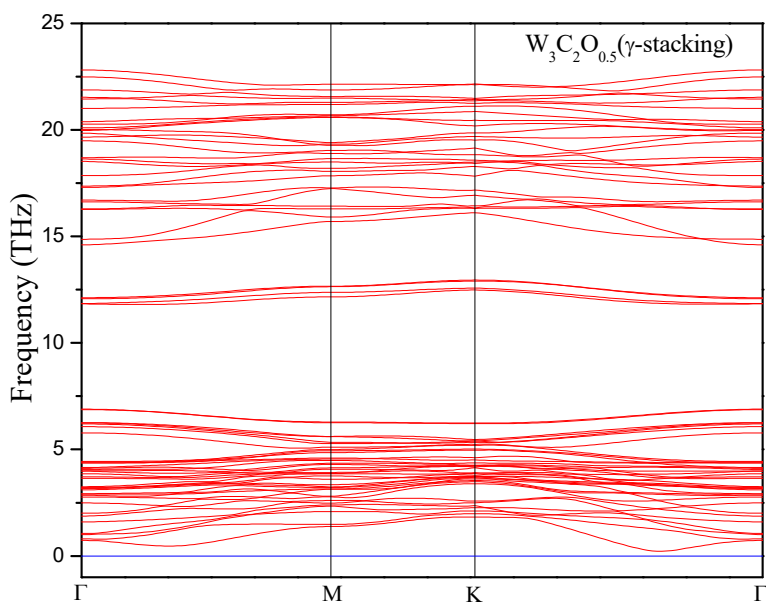
Figure S10 Phonon dispersion of γ-stackings of W$_3$C$_2$O$_{0.5}$ MXenes.
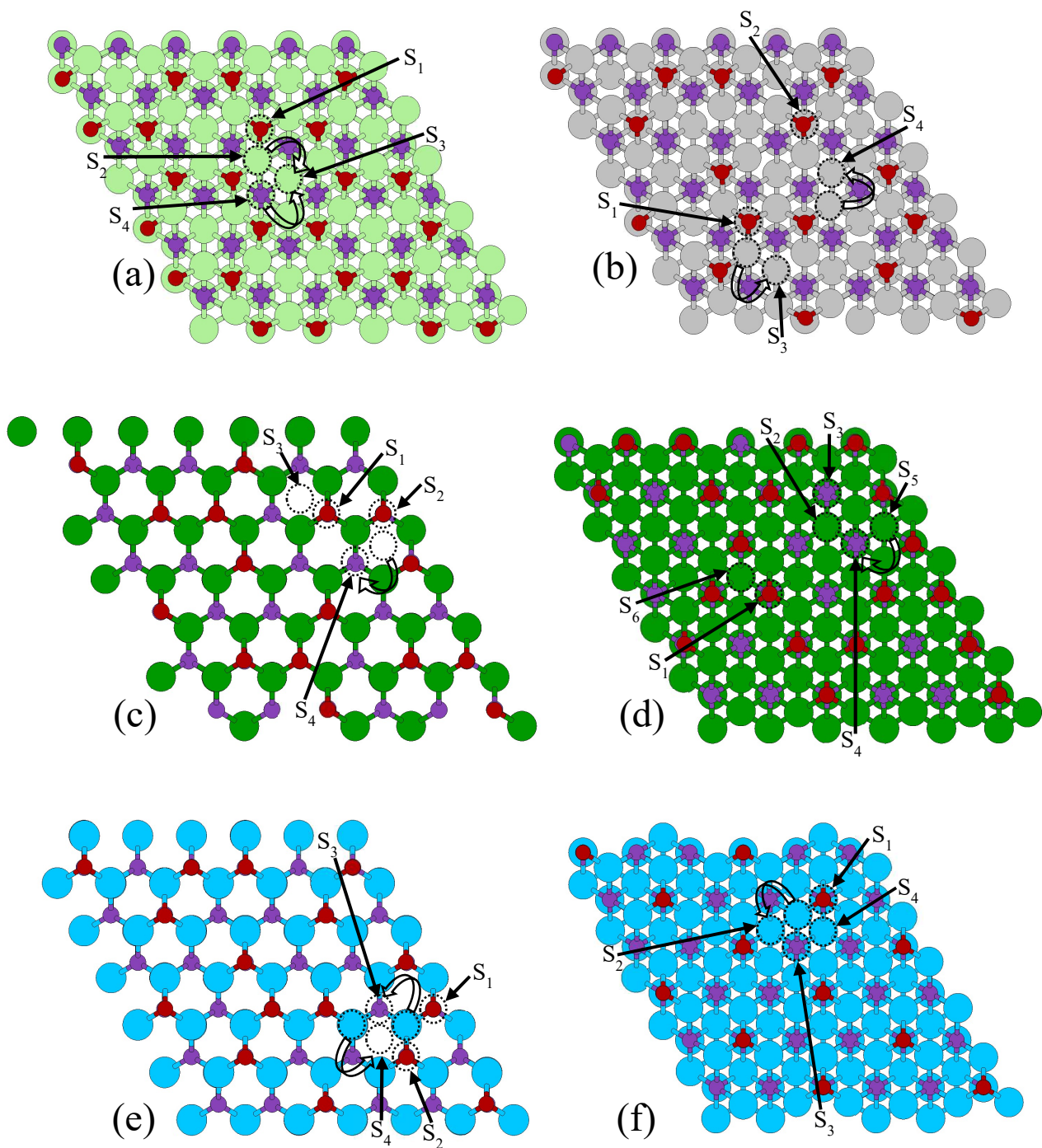
Figure S11: Final optimized structure of MXene with equilibrium oxygen coverage derived from lattice model along with different HER sites. [Actual calculation is performed over the 3×3 supercell of MXene. To properly understand the symmetry of oxygen configuration 6×6 supercell is shown above. Here, Light green, grey, green, sky blue, violet, and red represents Ti, V, W, Mo, C, and O atoms, respectively.]

Supplementary Tables

Table S1: Total energy and lattice parameter of γ-stacking type MXenes with different placement of -F and -O termination groups. [Here, blue, green, and orange represent fcc, hcp, and the on-top site as energetically favourable sites for termination groups, as also verified via minimum energy depicted in bold letters.]

| Early transition metal group	MXene	Lattice Parameter 'a' of pristine MXene (Å)	Placement of termination group	T_x = -O termination		T_x = -F termination	
				Lattice parameter a (Å)	Total Energy E (eV)	Lattice parameter a (Å)	Total Energy E (eV)
3	$Sc_3C_2T_x$	3.359	top site (T_t)	3.342	-5173.114	3.379	-5637.548
			hcp (T_h)	3.405	-5180.093	3.194	-5639.130
			fcc (T_f)	**3.268**	**-5180.418**	**3.239**	**-5640.359**
4	$Ti_3C_2T_x$	3.081	top site (T_t)	3.198	-6069.671	3.157	-6531.086
			hcp (T_h)	3.004	-6073.256	3.005	-6531.957
			fcc (T_f)	**3.028**	**-6074.767**	**3.061**	**-6532.687**
	$Zr_3C_2T_x$	3.326	top site (T_t)	3.464	-5239.092	3.400	-5700.991
			hcp (T_h)	3.256	-5243.892	3.255	-5702.403
			fcc (T_f)	**3.297**	**-5245.572**	**3.320**	**-5702.942**
	$Hf_3C_2T_x$	3.257	top site (T_t)	3.411	-6279.120	3.346	-6741.231
			hcp (T_h)	3.196	-6284.475	3.200	-6742.253
			fcc (T_f)	**3.240**	**-6286.412**	**3.265**	**-6742.836**
5	$V_3C_2T_x$	2.937	top site (T_t)	3.086	-7122.245	3.025	-7581.496
			hcp (T_h)	2.853	-7123.629	**2.846**	**-7582.136**
			fcc (T_f)	**2.905**	**-7124.252**	2.956	-7581.991
	$Nb_3C_2T_x$	3.131	top site (T_t)	3.299	-5999.529	3.203	-6458.914
			hcp (T_h)	3.073	-6002.157	**3.062**	**-6459.973**
			fcc (T_f)	**3.141**	**-6002.668**	3.098	-6459.387
	$Ta_3C_2T_x$	3.094	top site (T_t)	3.270	-6908.073	3.154	-7367.705
			hcp (T_h)	3.055	-6911.345	**3.041**	**-7368.260**
			fcc (T_f)	**3.123**	**-6911.867**	3.073	-7367.539
6	$Cr_3C_2T_x$	2.820	top site (T_t)	3.016	-8321.096	**2.953**	**-8780.214**
			hcp (T_h)	2.728	-8322.390	2.811	-8780.018
			fcc (T_f)	**2.867**	**-8322.471**	2.879	-8779.982
	$Mo_3C_2T_x$	3.007	top site (T_t)	3.167	-6872.953	**3.094**	**-7332.050**
			hcp (T_h)	**2.911**	**-6875.253**	2.998	-7331.812
			fcc (T_f)	3.053	-6874.135	3.044	-7331.757
	$W_3C_2T_x$	2.989	top site (T_t)	3.092	-7677.793	**3.035**	**-8136.950**
			hcp (T_h)	**2.904**	**-7680.440**	2.994	-8135.946
			fcc (T_f)	3.069	-7679.176	3.037	-8135.817

Table S2: Hydrogen adsorption energy for $M_3C_2T_x$ MXenes with/without termination groups for low hydrogen coverage ($\theta = \frac{1}{4}$) and high hydrogen coverage ($\theta = 1$). [Here, blue, green, and orange represent fcc, hcp, and the on-top site as energetically favourable sites for termination groups.]

Early transition metal group	MXene	Termination group	Adsorption energy E for hydrogen coverage $\theta = \frac{1}{4}$ (eV)			Adsorption energy E for hydrogen coverage $\theta = 1$ (eV)		
			S_1	S_2	S_3	S_1	S_2	S_3
3	$Sc_3C_2T_x$	pristine	-0.049	-0.655	-0.977	0.653	-0.361	-0.825
		-O	-	-	-1.087	-	-0.452	-0.760
		-F	2.810	-0.478	-	-	-	-
4	$Ti_3C_2T_x$	pristine	0.042	-0.965	-1.280	0.235	-0.933	-1.275
		-O	2.481	-	-0.435	-	-	0.096
		-F	1.949	-	-	-	-	-
	$Zr_3C_2T_x$	pristine	0.225	-0.991	-1.286	0.287	-1.078	-1.385
		-O	2.431	-	-0.047	2.520	-	0.423
		-F	1.260	0.640	-	2.068	1.529	-
	$Hf_3C_2T_x$	pristine	0.190	-1.057	-1.433	0.181	-1.142	-1.474
		-O	2.438	-	0.293	-	-	0.800
		-F	0.804	0.304	-	1.879	1.477	-
5	$V_3C_2T_x$	pristine	-0.13	-0.857	-1.074	0.11	-0.773	-1.001
		-O	0.848	-	-0.689	-	-	-0.007
		-F	-	1.567	-	-	-	-
	$Nb_3C_2T_x$	pristine	-0.02	-1.056	-1.219	0.13	-1.038	-1.19
		-O	1.163	-	0.048	1.729	-	0.467
		-F	1.77	-	0.757	-	-	-
	$Ta_3C_2T_x$	pristine	-0.061	-1.187	-1.332	0.003	-1.116	-1.26
		-O	1.015	-	0.485	1.6	-	0.872
		-F	1.154	-	0.389	-	-	-
6	$Cr_3C_2T_x$	pristine	-0.563	-0.801	-0.863	-0.155	-0.514	-0.774
		-O	-	-	-0.581	-	-0.264	-0.181
		-F	-	-	-	-	-	-
	$Mo_3C_2T_x$	pristine	-0.306	-0.636	-0.817	-0.251	-0.462	-0.654
		-O	1.622	-0.583	1.452	-	0.435	-
		-F	0.394	0.542	0.55	1.116	0.406	-
	$W_3C_2T_x$	pristine	-0.426	-0.485	-0.648	-0.516	-0.335	-0.532
		-O	1.65	-0.318	-	-	0.919	-
		-F	0.053	0.018	0.053	-	0.209	-

S_1-over metal site; S_2- over hcp site; S_3-over fcc site. Missing data represents either H is not adsorbed over that site or it shifted to other site.

Table S3 Relative total energy for placement of -O termination group, lattice parameter, and thickness of studied MXenes.

MXene	Stacking Configuration	Placement of termination group (lower-upper)	Relative Total Energy (eV/f.u.)	Lattice Parameter a (Å)	Thickness d (Å)
W₃C₂O₂	α	**hcp-hcp**	**0.000**	**2.89**	**8.11**
		hcp-fcc	0.796	2.90	8.12
		fcc-fcc	1.491	2.91	8.12
		top-hcp	2.220	2.92	8.66
		top-fcc	2.943	2.93	8.65
		top-top	4.374	2.96	9.21
	β	**hcp-hcp**	**0.000**	**2.89**	**8.14**
		hcp-fcc	0.509	2.90	8.15
		fcc-fcc	1.150	2.90	8.17
		top-hcp	2.101	2.92	8.70
		top-fcc	2.627	2.93	8.68
		top-top	4.120	2.96	9.23
	γ	**hcp-hcp**	**0.000**	**2.90**	**7.98**
		hcp-fcc	0.791	2.94	7.84
		fcc-fcc	1.263	3.07	7.14
		top-hcp	1.434	2.99	8.17
		top-fcc	1.998	3.08	7.80
		top-top	2.646	3.09	8.48
Mo₃C₂O₂	α	**hcp-hcp**	**0.000**	**2.88**	**8.04**
		hcp-fcc	0.615	2.89	8.03
		fcc-fcc	1.132	2.90	8.02
		top-hcp	2.137	2.91	8.59
		top-fcc	2.686	2.93	8.55
		top-top	4.232	2.95	9.12
	β	**hcp-hcp**	**0.000**	**2.88**	**8.08**
		hcp-fcc	0.384	2.89	8.07
		fcc-fcc	0.867	2.89	8.07
		top-hcp	2.053	2.91	8.63
		top-fcc	2.426	2.92	8.59
		top-top	4.020	2.95	9.16
	γ	**hcp-hcp**	**0.000**	**2.91**	**7.81**
		hcp-fcc	0.652	2.94	7.70
		fcc-fcc	1.118	3.05	7.13
		top-hcp	1.277	3.00	8.03
		top-fcc	1.757	3.07	7.74
		top-top	2.300	3.17	8.13

Table S4 Calculated change in zero-point energy and entropic correction between adsorbed and gas state over considered MXenes ['Im' signifies Imaginary vibration modes associated to hydrogen atom over catalytic site.]

MXene	Stacking Configuration	Termination group	Δ*ZPE* (eV)			Δ*ZPE* − *T*Δ*S* (eV)		
			top site	hcp site	fcc site	top site	hcp site	fcc site
W_3C_2	α	Pristine	Im	0.043	0.063	-	0.248	0.268
		-O	-	0.168	Im	-	0.373	-
	β	Pristine	Im	0.040	0.065	-	0.245	0.270
		-O	-	0.167	-	-	0.372	-
	γ	Pristine	0.024	0.000	0.022	0.229	0.205	0.227
		-O	0.042	0.173	-	0.247	0.378	-
Mo_3C_2	α	Pristine	Im	0.036	0.060	-	0.241	0.265
		-O	Im	0.168	-	-	0.373	-
	β	Pristine	Im	0.034	0.063	-	0.239	0.268
		-O	-	0.167	-	-	0.372	-
	γ	Pristine	-0.002	0.002	0.025	0.203	0.207	0.230
		-O	Im	0.174	Im	-	0.379	-
Ti_3C_2	γ	Pristine	Im	0.003	0.031	-	0.208	0.236
		-O	-0.005	-	0.166	0.200	-	0.371
V_3C_2	γ	Pristine	Im	0.019	0.044	-	0.224	0.249
		-O	0.043	-	0.173	0.248	-	0.378

Table S5 Calculated change in Gibbs free energy of hydrogen and description of HER active site over studied MXene at equilibrium oxygen coverage, as shown in figure S11.

MXene $(M_3C_2O_{2(0)})$	Site	ΔG_{H^*} (eV)	Initial Site	Final Site	Remarks
γ-Ti$_3$C$_2$O$_{2(0.66)}$	S$_1$	0.566	over oxygen (fcc)	over oxygen (fcc)	-
	S$_2$	-0.155	over Ti (top)	over fcc hollow (fcc)	migration of H
	S$_3$	-0.154	over fcc hollow (fcc)	over fcc hollow (fcc)	-
	S$_4$	-0.154	over C (hcp)	over fcc hollow (fcc)	migration of H
γ-V$_3$C$_2$O$_{2(0.44)}$	S$_1$	0.305	over oxygen (fcc)	over oxygen (fcc)	-
	S$_2$	0.518	over oxygen (fcc)	over oxygen (fcc)	-
	S$_3$	-0.450	over V (top)	over fcc hollow (fcc)	migration of H
	S$_4$	-0.619	over V (top)	over fcc hollow (fcc)	migration of H
α-W$_3$C$_2$O$_{2(0.44)}$	S$_1$	1.216	over oxygen (hcp)	over oxygen (hcp)	-
	S$_2$	0.668	over oxygen (hcp)	over oxygen (hcp)	-
	S$_3$	-0.006	over fcc hollow (fcc)	over fcc hollow (fcc)	-
	S$_4$	-0.615	over fcc hollow (fcc)	over C (hcp)	migration of H
γ-W$_3$C$_2$O$_{2(0.55)}$	S$_1$	0.638	over oxygen (hcp)	over oxygen (hcp)	-
	S$_2$	-0.375	over fcc hollow (fcc)	over fcc hollow (fcc)	-
	S$_3$	-0.242	over C (hcp)	over C (hcp)	-
	S$_4$	-0.269	over C (hcp)	over C (hcp)	-
	S$_5$	-0.268	over fcc hollow (fcc)	over C (hcp)	migration of H
	S$_6$	1.604	over fcc hollow (fcc)	over fcc hollow (fcc)	Absorption in first layer
α-Mo$_3$C$_2$O$_{2(0.44)}$	S$_1$	0.496	over oxygen (hcp)	over oxygen (hcp)	-
	S$_2$	0.725	over oxygen (hcp)	over oxygen (hcp)	-
	S$_3$	-0.567	over Mo (top)	over C (hcp)	migration of H
	S$_4$	0.023	over Mo (top)	over fcc hollow (fcc)	migration of H
γ-Mo$_3$C$_2$O$_{2(0.33)}$	S$_1$	0.646	over oxygen (hcp)	over oxygen (hcp)	-
	S$_2$	0.064	over Mo (top)	over fcc hollow (fcc)	migration of H
	S$_3$	-0.231	over C (hcp)	over C (hcp)	-
	S$_4$	0.065	over fcc hollow (fcc)	over fcc hollow (fcc)	-

Supplementary Multimedia

Multimedia 1.mp4

Multimedia S1: Changes in surface configuration of MXene with change in oxygen coverage from $\Theta=0$ to $\Theta=1$ [Here, the black colour surface is a suitable lattice site for oxygen termination, and the red dot indicates the presence of oxygen at the lattice site. For Ti_3C_2 and V_3C_2, a suitable lattice site refers to the fcc site, while for W_3C_2 and Mo_3C_2, it indicates the hcp site.]