MXene-Specific Machine-Learned Potential — A DFT Surrogate for Structure Relaxation and Surface Chemistry

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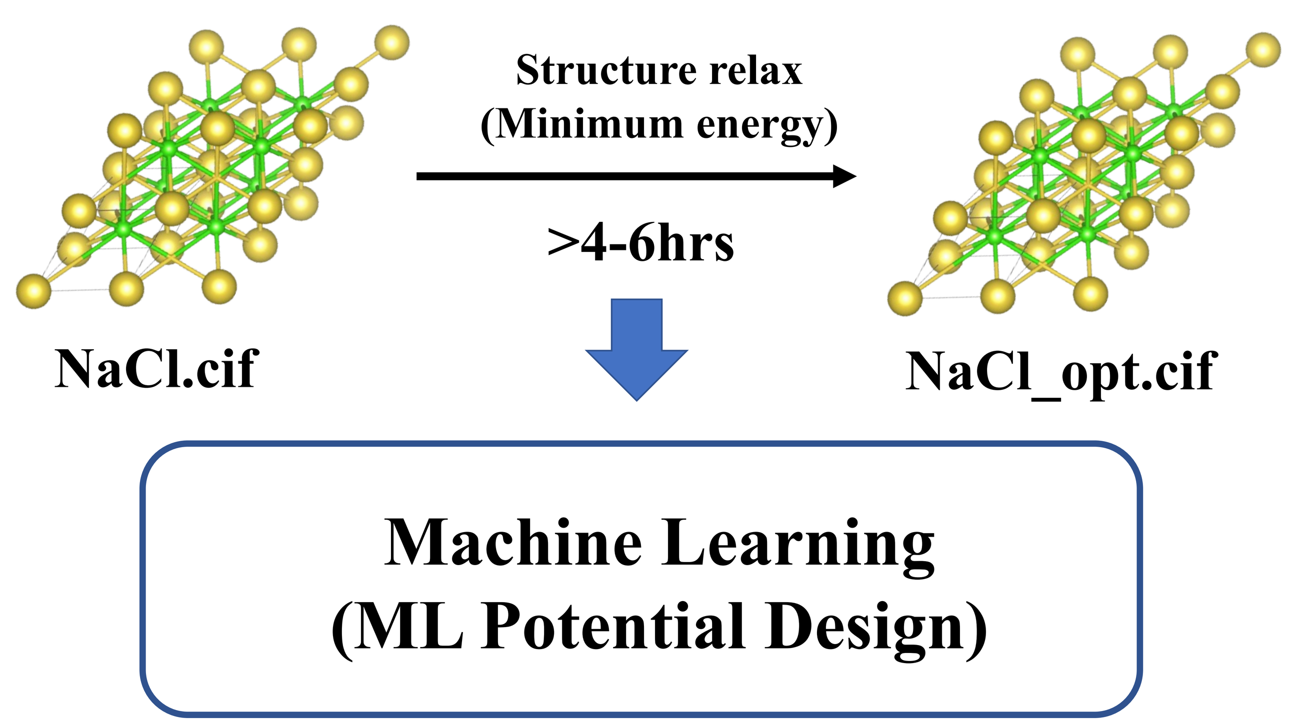
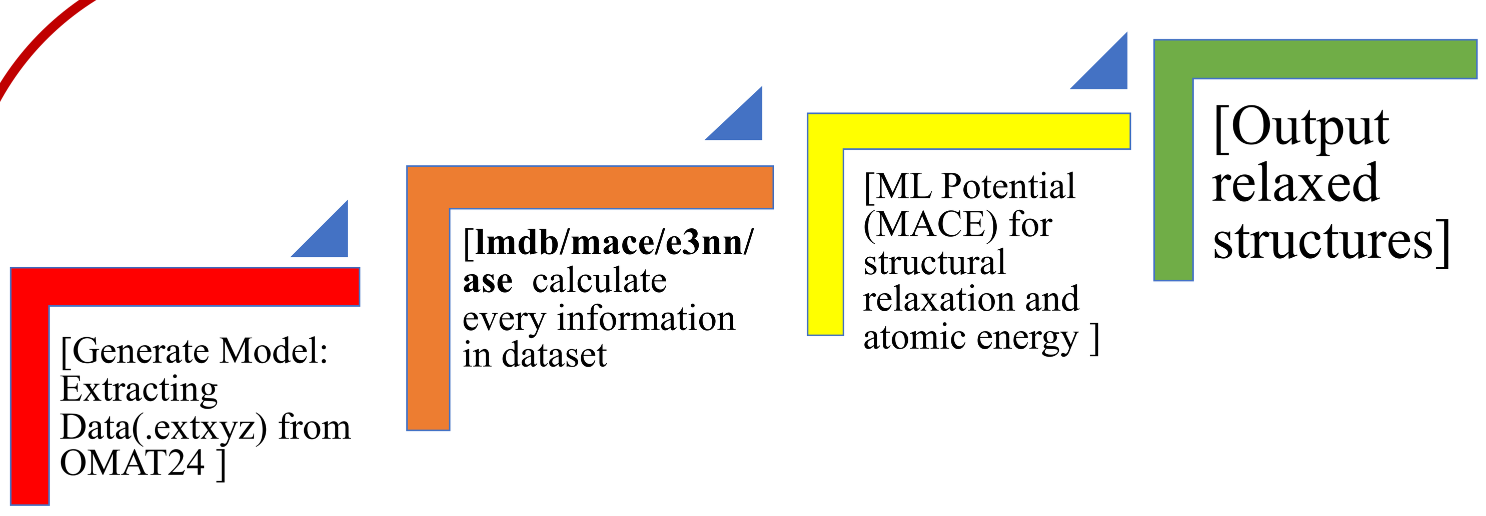
Abstract

Thousands of MXene-like structures were curated from Meta Open Materials 2024 (OMat24) Dataset, filtered into composition/termination-balanced subsets, and converted to .extxyz with DFT energies, forces, and virials. An E(3)-equivariant graph neural network (MACE) is then trained as a **MXene-specific machine-learned potential** to perform structure relaxation and surface-chemistry predictions. Uncertainty ensembling and an active-learning loop are used to flag out-of-distribution cases and to prioritize new DFT labels. Deployed as an ASE calculator, the model accelerates geometry optimization and screening while preserving DFT-level trends in lattice parameters, interlayer spacing, work-function shifts, and adsorption energies across Ti/C/N-based MXenes. This MXene-focused surrogate enables rapid, large-scale exploration of composition–termination–adsorbate space for electrocatalysis and sensing in the future.

Motivation

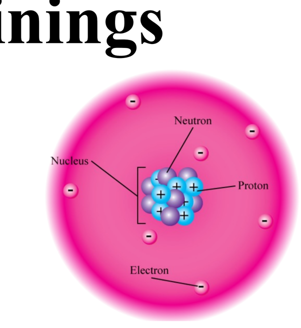


Experimental & Simulation Method



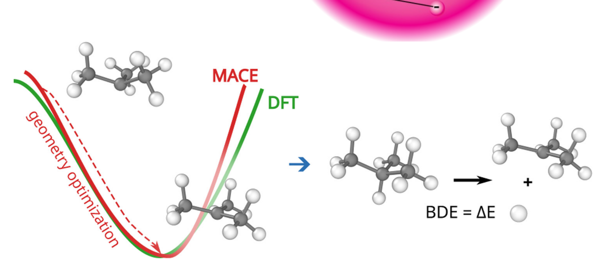
Classical formulas used for ML trainings

$$E = \sum_{i=1}^{N} \varepsilon_i(\{\mathbf{R}_j - \mathbf{R}_i, Z_j\})$$ Total energy decomposition



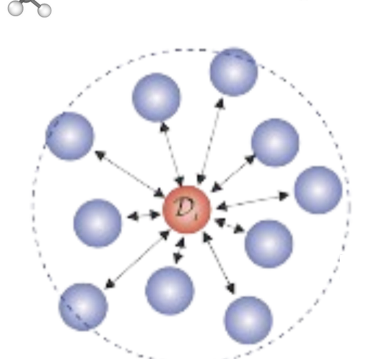
$$\mathbf{F}_i = -\frac{\partial E}{\partial \mathbf{R}_i}$$ Atomic forces

$$\boldsymbol{\sigma} = \frac{1}{V}\frac{\partial E}{\partial \boldsymbol{\varepsilon}} \approx -\frac{1}{V}\sum_i \mathbf{R}_i \otimes \mathbf{F}_i$$ Stress tensor



$$\mathrm{MSE} = \tfrac{1}{n}\sum_i (y_i - \hat{y}_i)^2, \qquad \mathrm{MAE} = \tfrac{1}{n}\sum_i |y_i - \hat{y}_i|$$ MSE / MAE

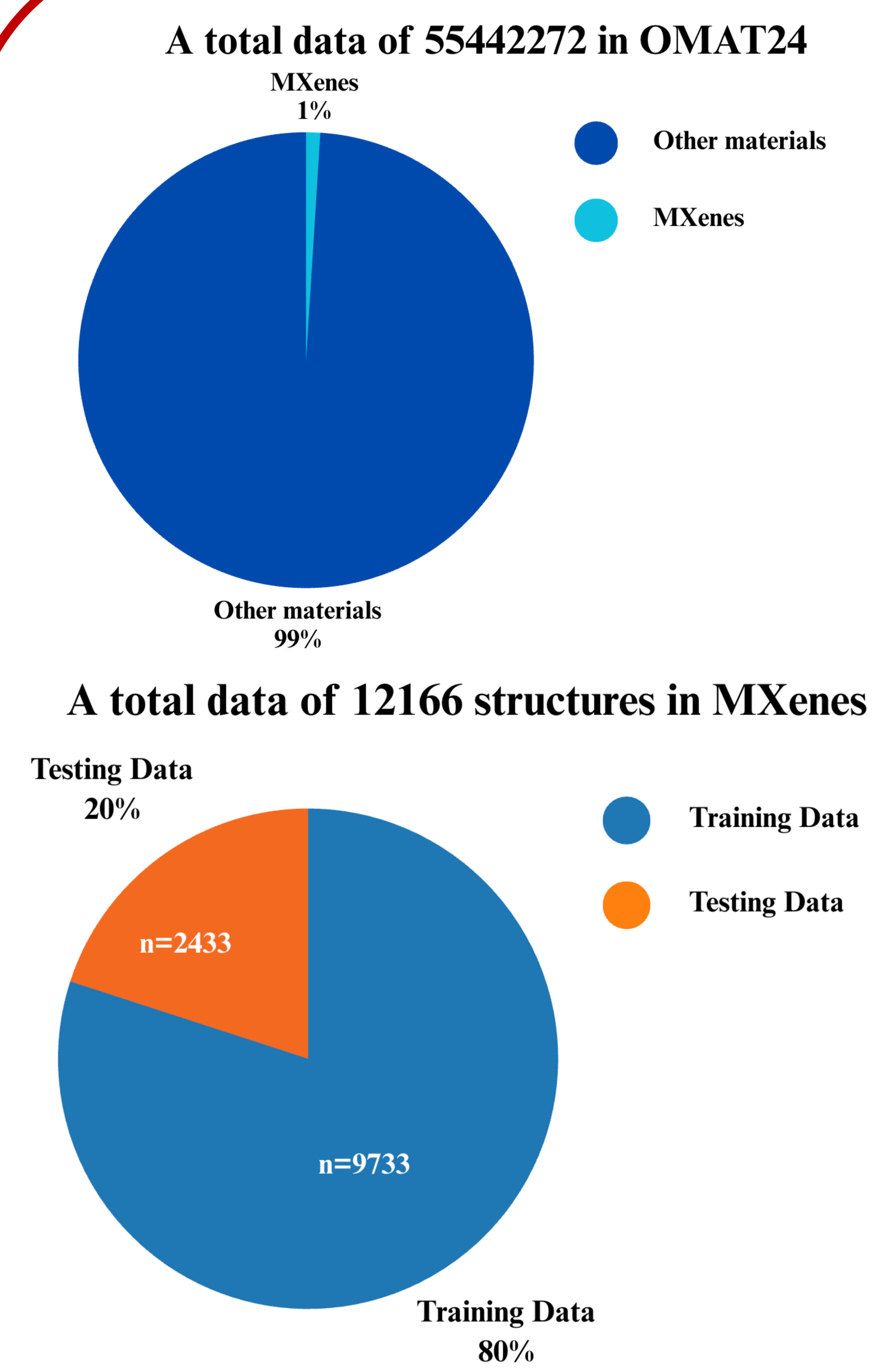
$$E_{\mathrm{pred}} = \sum_i \varepsilon_\theta(\{\, r_{ij},\, \hat{\mathbf{r}}_{ij},\, Z_j \,\}_{j\neq i})$$ Energy as sum of atomic contributions



$$\mathcal{L} = w_E \frac{(E_{\mathrm{pred}} - E_{\mathrm{DFT}})^2}{N} + w_F \frac{1}{3N}\sum_i \|\mathbf{F}_{i,\mathrm{pred}} - \mathbf{F}_{i,\mathrm{DFT}}\|_2^2 + w_\sigma \|\boldsymbol{\sigma}_{\mathrm{pred}} - \boldsymbol{\sigma}_{\mathrm{DFT}}\|_F^2$$

Multi-task loss (typical weighting)

Results & Discussion



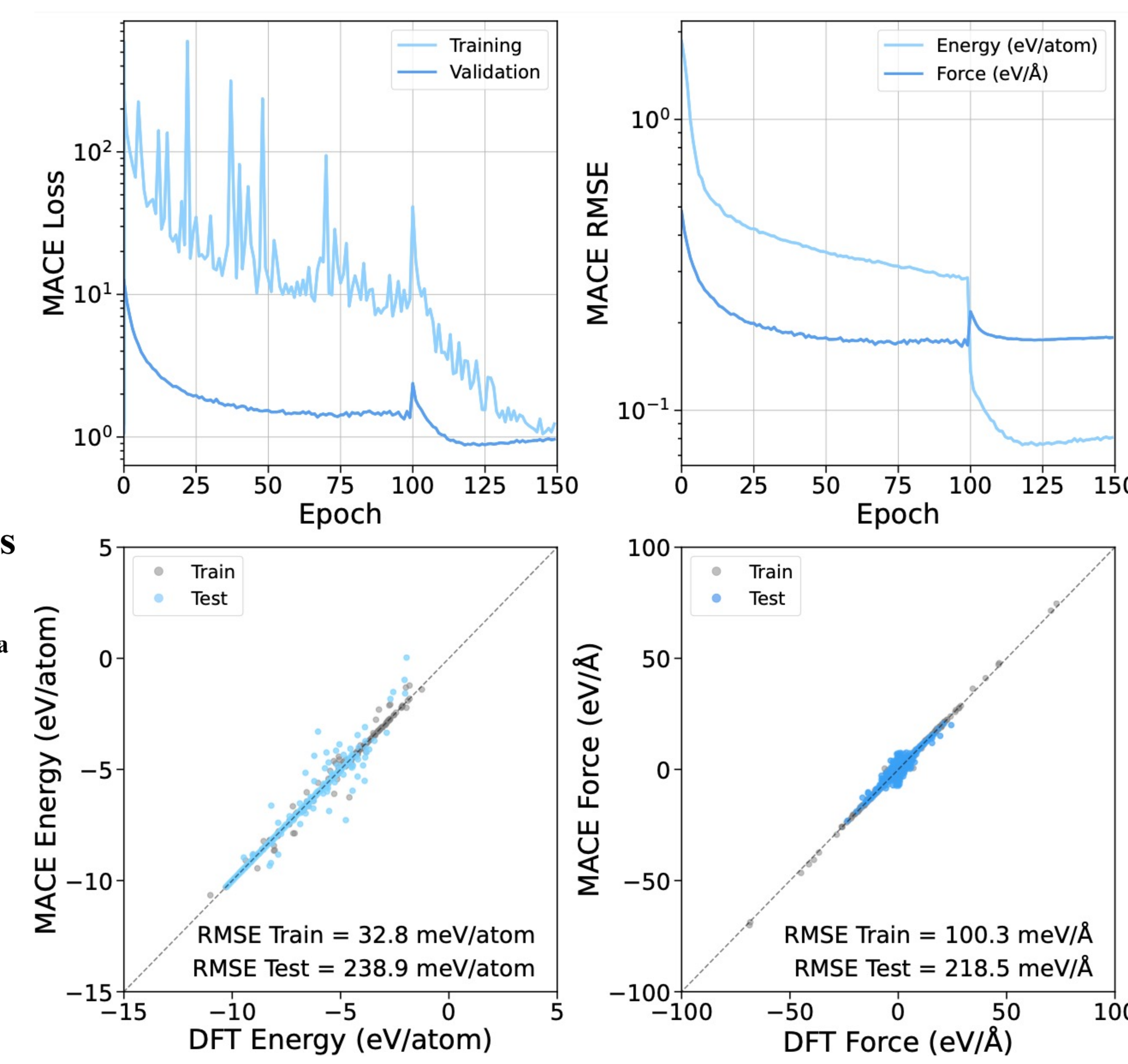


Table 1, CPU time spent (hour∗core) comparison between DFT calculation, Universal Model and our model

Methods / Numbers of atoms	DFT	Universal Model	Our Model
$Ti_3C_2Cl_2$ (Unit cell)(29 atoms)	3-5	~8*10^{-4}	~8*10^{-4}
$Ti_3C_2Cl_2$ (Supercell: 4*4*1)(157 atoms)	16-48	~8*10^{-4}	~8*10^{-4}

Table 2, Formation Energy (E_f) comparison between DFT calculation, Universal Model and our model

Compounds	DFT (eV)	Universal Model (eV)	Our Model (eV)
$Ti_2S_4Br_4$	-58.60	-48.08	-47.83
$Ti_2S_4Cl_4$	-59.83	-50.49	-50.33
$Ti_2Se_4Br_4$	-57.2	-45.09	-44.815
$Ti_2Se_4Cl_4$	-58.42	-47.7	-47.36
Ti_3C_2	-93	-91	-90.5
$Ti_3C_2Cl_2$	-54.82	-53.3	-53.5
$Ti_3C_2F_2$	-59.08	-58.4	-57.92
$Ti_3C_2O_2$	-64.185	-63.54	-63.3
$Ti_3C_2OH_2$	-64.15	-70.1	-70.4
$Ti_4Se_4I_4$	-63.94	-62.35	-62.02

Conclusion

- Built from ~10,000 curated MXene entries, the initial model generalizes across compositions and surface terminations.
- Trained an MXene-specific machine-learned potential that matches DFT trends, keeping per-structure total-energy errors < 10 eV on the held-out set.
- Expand data and labels (E/F/σ), add uncertainty-aware active learning, and extend to adsorption energies and work-function shifts—delivering a robust ASE-deployable surrogate for high-throughput screening.