Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Support information

Leveraging all-fixed transfer framework to predict interpretable formation energy of MXene with hybrid terminals

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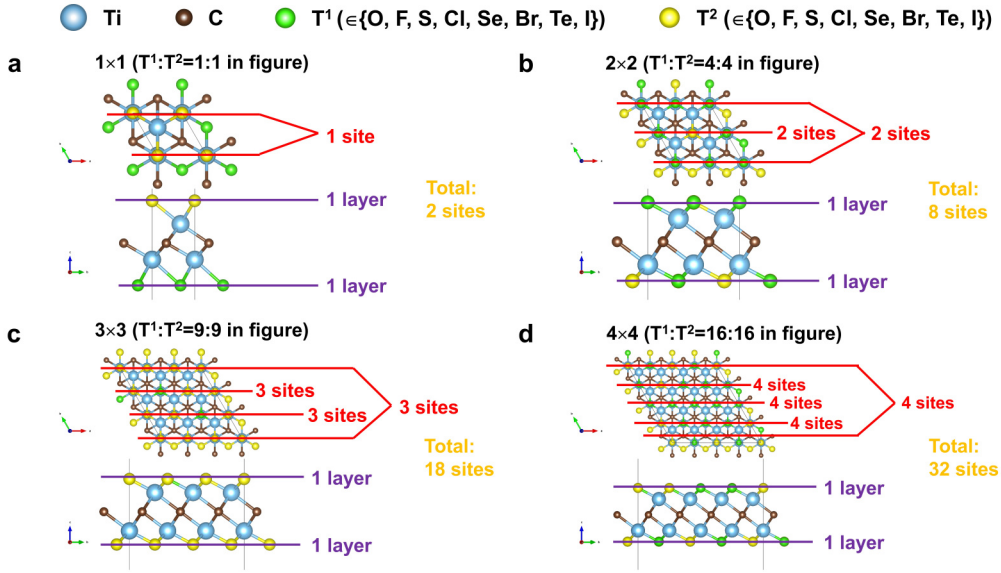

Fig. S1: The crystal structures of Ti_2CT_2 in different lattice. (a) 1×1 lattice with 2 sites. T^1:T^2 can only be 1:1. (b) 2×2 lattice with 8 sites. T^1:T^2 can range from 1:7 to 7:1. (c) 3×3 lattice with 18 sites. T^1:T^2 can range from 1:17 to 17:1. (d) 4×4 lattice with 32 sites. T^1:T^2 can range from 1:31 to 31:1.

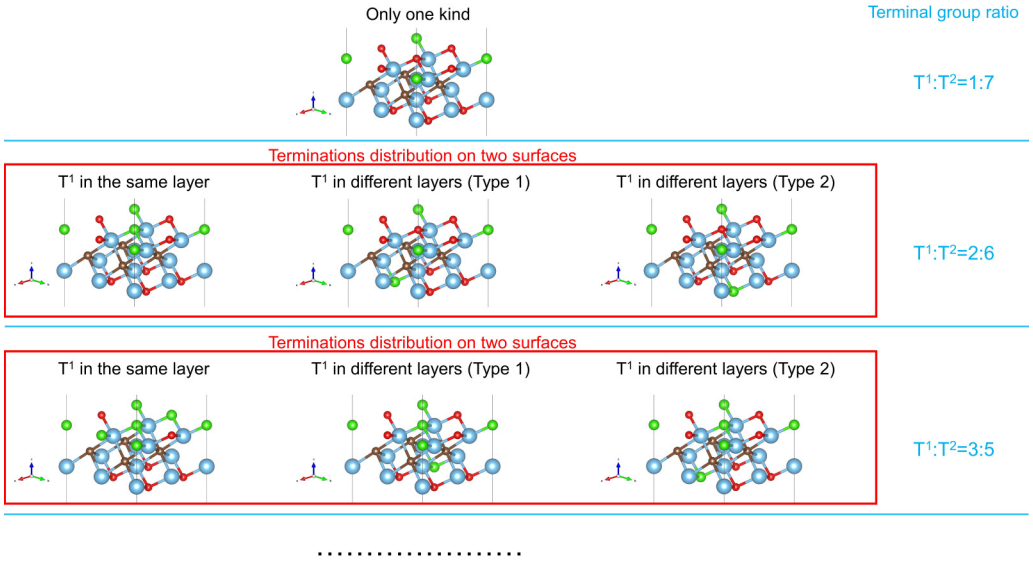

Fig. S2: The detailed displaying of terminal group ratio and terminations distribution on two surfaces.

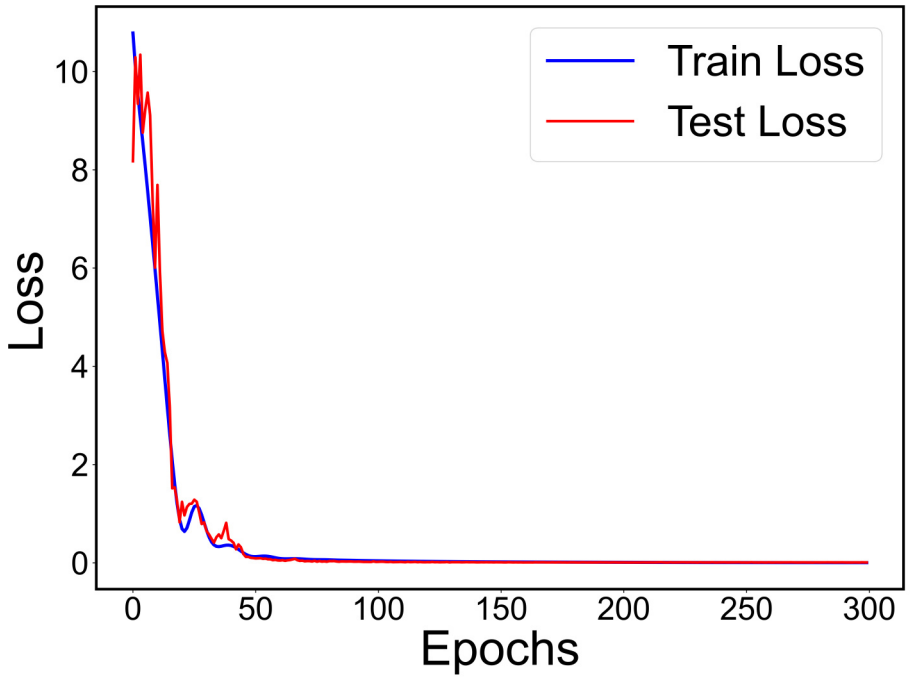

Fig. S3: The learning rate of the transferred model in the first transfer. The target training set is 1×1 Ti_2CT_2 lattice that has 36 individuals.

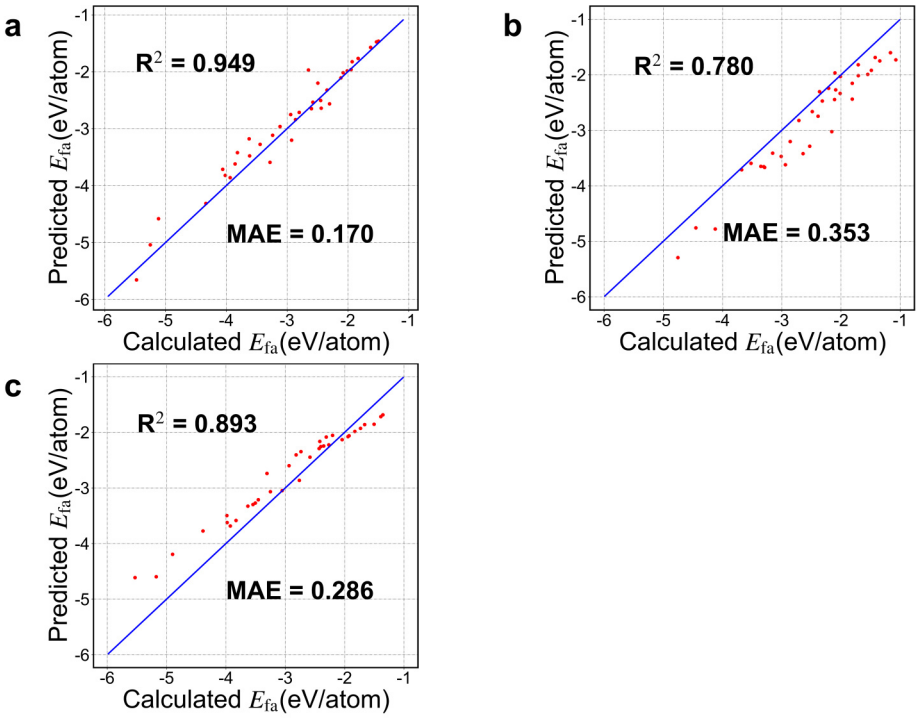

Fig. S4: R^2 of the model trained on 1×1 Ti_2CT_2 lattice dataset and predicts optimized 1×1 lattice of (a) Ti_2NT_2, (b) V_2CT_2, (c) $Ti_3C_2T_2$.

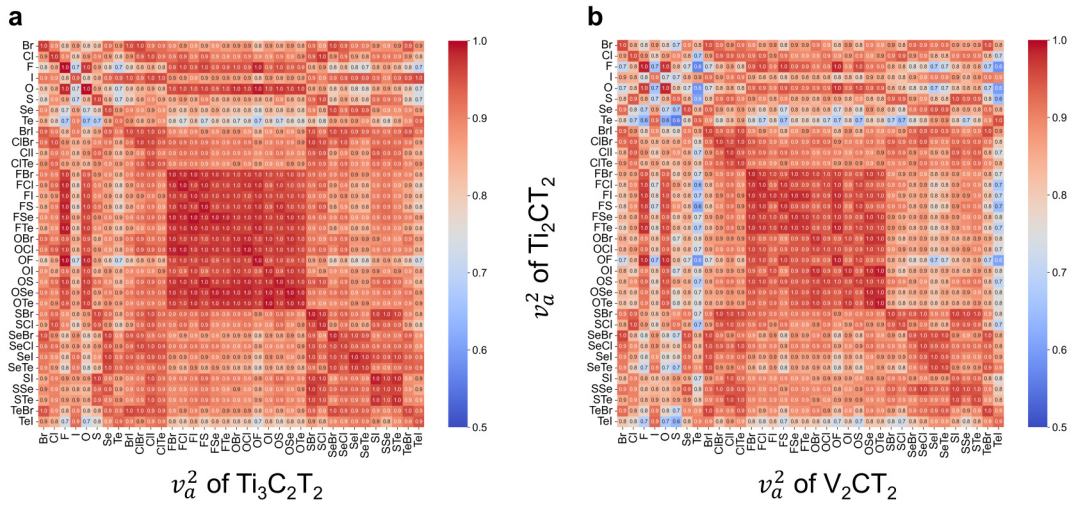

Fig. S5: The Pearson correlations of the features v_a^2 of 1×1 Ti_2CT_2 and 1×1 V_2CT_2, 1×1 Ti_2CT_2 and 1×1 $Ti_3C_2T_2$ from the last layer of our framework. (a) The Pearson correlations of the features v_a^2 of 1×1 Ti_2CT_2 and 1×1 V_2CT_2. (b) The Pearson correlations of the features v_a^2 of 1×1 Ti_2CT_2 and 1×1 $Ti_3C_2T_2$.

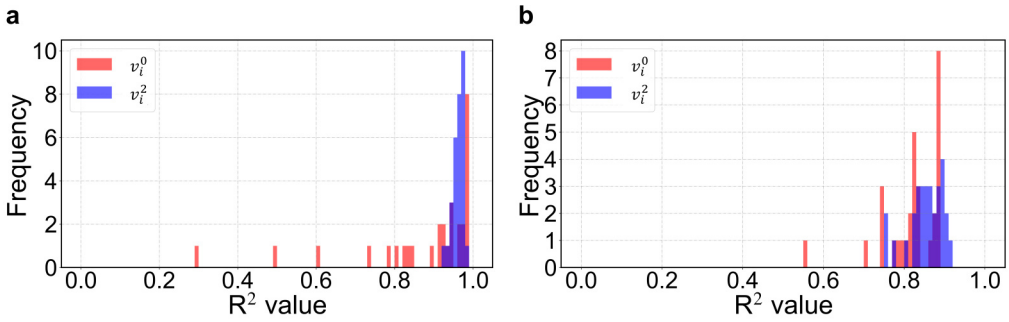

Fig. S6: The frequency distribution of the performance of feature v_i^0 and feature v_i^2. Both of them were used to train on the 1×1 Ti_2CT_2 lattice and to predict the 1×1 (a) Ti_2NT_2 and (b) $Ti_3C_2T_2$ lattice. This process is conducted totally 30 times in both (a) and (b).

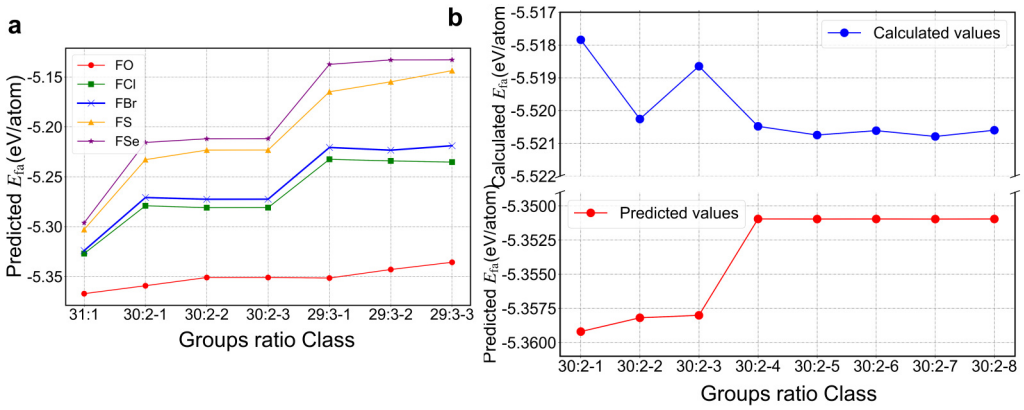

Fig. S7: The E_{fa} of screening results and their comparing with calculated results. (a) Screening results of the 4×4 $Ti_3C_2T_2$ lattice with the lowest E_{fa}. "30:2-1, 30:2-2, 30:2-3" are sampled from all kinds of structures under the ratio of 30:2. The same with "29:3-1, 29:3-2, 29:3-3". (b) Results comparing the prediction of all kinds structures of 4×4 $Ti_3C_2T_2$ FO lattice to calculation of those under the ratio of 30:2.

Table. S1: The training and testing sets of different kinds of MXene.

Training set			Testing set			Corresponding figures
Lattice size	MXene type	Number of data	Lattice size	MXene type	Number of data	\
1×1	Ti_2C	36	1×1	Ti_2N	36	Fig. S3a
				V_2C	36	Fig. S3b
				Ti_3C_2	36	Fig. S3c
			2×2	Ti_2C	560	Fig. 4a
			3×3	Ti_2C	72	Fig. 4b
			4×4	Ti_2C	144	Fig. 4c
				Ti_2C (without optimization)	144	Fig. 4d
2×2	Ti_2C	560	3×3	Ti_2C	72	Fig. 5a
			4×4	Ti_2C	144	Fig. 5b
				Ti_2C (without optimization)	144	Fig. 5c

Table. S2: The reference values of E_{fa} of non-optimized 4×4 $Ti_{n+1}C_nT^1_xT^2_{2-x}$(n=1,2) lattice(values in () is the calculated values).

Base and Pair		Elements ratio				
		31:1	30:2	29:3	27:5	24:8
Ti_2C	FO	-5.89(-5.58)	-5.84(-5.57)	-5.78(-5.56)	-5.68(-5.53)	-5.54(-5.48)
	FCl	-5.84(-5.53)	-5.75(-5.46)	-5.65(-5.4)	-5.46(-5.27)	-5.17(-5.06)
	FBr	-5.8(-5.51)	-5.66(-5.43)	-5.52(-5.35)	-5.23(-5.19)	-4.83(-4.92)
	FS	-5.8(-5.51)	-5.66(-5.43)	-5.52(-5.34)	-5.24(-5.17)	-4.83(-4.9)
	FSe	-5.79(-5.5)	-5.63(-5.41)	-5.48(-5.31)	-5.17(-5.12)	-4.7(-4.82)
Ti_3C_2	FO	-5.37(-5.53)	-5.35(-5.52)	-5.34(-5.51)	-5.32(-5.48)	-5.28(-5.43)
	FCl	-5.33(-5.47)	-5.28(-5.41)	-5.23(-5.34)	-5.14(-5.21)	-5.02(-5)
	FBr	-5.32(-5.46)	-5.27(-5.38)	-5.22(-5.3)	-5.13(-5.13)	-4.99(-4.85)
	FS	-5.3(-5.46)	-5.22(-5.38)	-5.15(-5.3)	-5.01(-5.12)	-4.8(-4.85)
	FSe	-5.3(-5.45)	-5.21(-5.36)	-5.13(-5.27)	-4.98(-5.07)	-4.74(-4.78)

The Supporting information of the calculation method of lithium storage capacity:

Table. S3: The formation energy of a single Li atom adsorbed on T_C, T_M, T_T and T_t sites.

Element pair	Adsorbed sites	Formation energy
FO	T_M	-0.76484
	T_C	-0.94663
	T_T	unfavorable
	T_t	-0.93221
FCl	T_M	-0.73536
	T_C	-0.91498
	T_T	-0.22675
	T_t	0.33698
FBr	T_M	-0.72826
	T_C	-0.91475
	T_T	-0.21825
	T_t	0.48174
FS	T_M	-0.78245
	T_C	-0.99712
	T_T	unfavorable
	T_t	-0.43601
FSe	T_M	-0.77543
	T_C	-0.99603
	T_T	unfavorable
	T_t	-0.27374

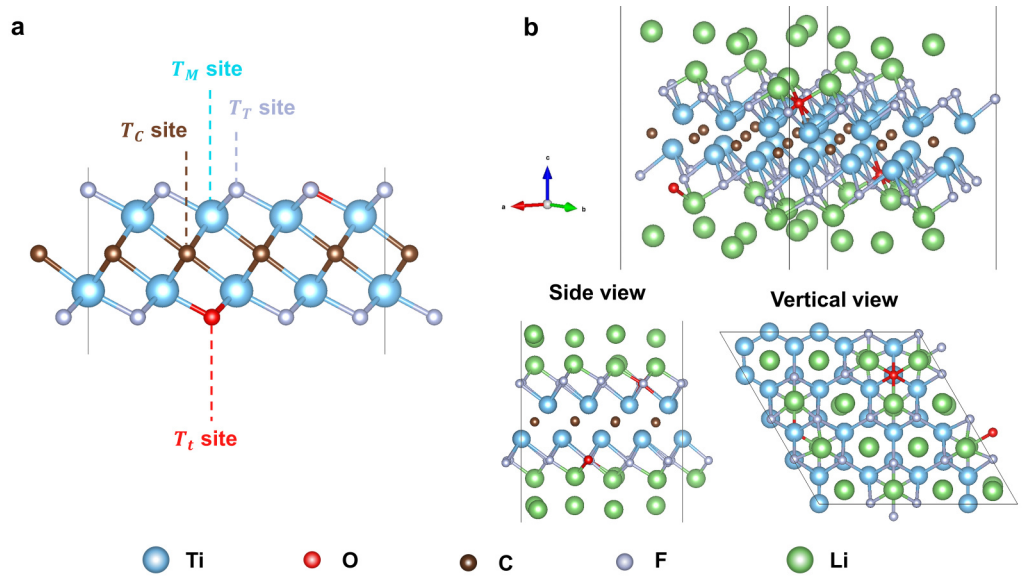

Fig. S8: The verification of the possible adsorbed sites on the surface of MXene. (a) The chosen of the possible adsorbed sites. (b) The calculated structure of FO MXene that fully covers Li atoms on the T_C sites.

Investigating the formation energy of a single Li atom adsorbed on different sites of 4×4 Ti_2C FX MXenes(X represents O, Cl, Br, S Se), we found that Li prefers to occupy the T_C site compared with T_M site or T_T/T_t site, as shown in Table. S3 and Fig. S7a. Thus, we step further to investigate the structures of the same MXenes that fully occupied by Li atoms in all T_C sites. Fig. S7b shows the structure of the lowest total energy of FO MXene adsorbing Li atoms(FCl, FBr FS, FSe are shown in Fig. S8), but such structure can't form since its formation energy is positive. Apart from FSe and FBr MXenes, most of the Li atoms which are on the surface of MXene can still settle on T_C sites with the increasing of Li atoms' number. It inspired us to take a new way to add the adsorbed atoms that adheres to the four principles below:

1. The formation energy produced by adding a new Li into the structure must be negative. Known as the E_{fi} in $E_{fi} = E_0^1 - E_0^2 - n\mu_{Li}$ must be negative. (E_0^1 and E_0^2 represent the structures that have or haven't added the new atoms. n is the number of the newly added atoms one time while μ_{Li} is the chemical potential of a single Li atom.)

2. After adding the new atoms, the structures that have been optimized by VASP once again should not decompose.

3. The newly added atoms should occupy the T_C sites as much as possible.

4. The newly added atoms should be adsorbed on the first layer as much as possible.

If any of these four principles don't satisfied in one adding cycle, the newly added atoms will change the original sites to another random sites and the structure will be recalculated, until all the sites have been tried but the newly added Li can't occupy the T_C or be adsorbed on the first layer while the E_{fi} is still negative. When the E_{fi} becomes positive or the structure can't afford to avoid decomposing after an adding cycle, the total number of the adsorbed Li atoms before this cycle will be treated as the theoretical capacity.

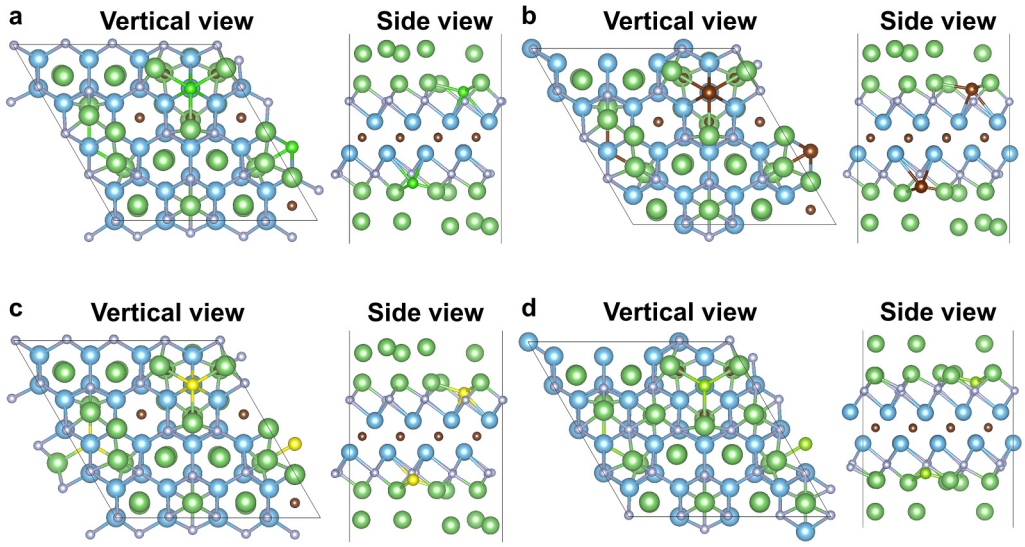

Fig. S8: The optimized structure of (a) FCl, (b) FBr, (c) FS, and (d) FSe MXene fully adsorbing Li atoms on T_C sites.

Meanwhile, in order to increase efficiency, we chose the element ratio of F:X equals to 30:2 and 24:8, where the "X" atoms can be evenly divided into two sides of the Ti_2C. So, we can add two Li atoms one time, with the first Li in one side and the second in the other side, utilizing the symmetrical equivalence to reduce errors.