Table S1. Calculated total energies (E_{tot}), energy differences (ΔE_{tot}) of HCP and FCC models, lattice parameters (a), slab thicknesses (L), M-C bond lengths ($d_{M\text{-}C}$), M-F bond lengths ($d_{M\text{-}F}$) of different M_2CF_2 configurations.

M_2CF_2	F sites	E_{tot} (eV/atom)	ΔE_{tot} (eV/atom)	a (Å)	L (Å)	$d_{M\text{-}C}$ (Å)	$d_{M\text{-}F}$ (Å)
Sc_2CF_2	HCP	-7.434		6.389	5.227	2.268	2.253
Sc_2CF_2	FCC	-7.679	-0.245	6.531	4.832	2.274	2.206
Sc_2CF_2	TOP	-7.106		6.965	6.049	2.340	1.827
Y_2CF_2	HCP	-7.574		6.907	5.526	2.447	2.405
Y_2CF_2	FCC	-7.778	-0.203	7.079	5.104	2.461	2.360
Y_2CF_2	TOP	-7.130		7.509	6.554	2.533	1.966
Ti_2CF_2	HCP	-7.966		5.945	5.072	2.100	2.170
Ti_2CF_2	FCC	-8.083	-0.117	6.101	4.782	2.098	2.160
Ti_2CF_2	TOP	-7.744		6.392	5.738	2.146	1.77
Zr_2CF_2	HCP	-8.414		6.459	5.409	2.277	2.330
Zr_2CF_2	FCC	-8.481	-0.067	6.597	5.139	2.273	2.322
Zr_2CF_2	TOP	-8.085		6.788	6.364	2.334	1.914
Hf_2CF_2	HCP	-10.260		6.316	5.411	2.236	2.305
Hf_2CF_2	FCC	-10.321	-0.062	6.534	5.055	2.233	2.310
Hf_2CF_2	TOP	-10.010		6.798	6.285	2.324	1.897
V_2CF_2	HCP	-7.945		5.574	5.143	2.038	2.082
V_2CF_2	FCC	-8.015	-0.069	5.962	4.601	2.001	2.145
V_2CF_2	TOP	-7.833		6.338	5.312	2.043	1.748
Nb_2CF_2	HCP	-8.565		6.004	5.523	2.195	2.237
Nb_2CF_2	FCC	-8.602	-0.037	6.412	4.936	2.153	2.302
Nb_2CF_2	TOP	-8.364		6.342	6.223	2.208	1.876
Ta_2CF_2	HCP	-9.271		5.916	5.633	2.190	2.238
Ta_2CF_2	FCC	-9.255	0.016	6.398	4.947	2.140	2.313
Ta_2CF_2	TOP	-9.157		6.200	6.320	2.201	1.878
Cr_2CF_2	HCP	-7.621		5.490	5.041	1.962	2.091
Cr_2CF_2	FCC	-7.711	-0.089	6.000	4.310	1.953	2.136
Cr_2CF_2	TOP	-7.642		5.837	5.465	1.963	1.726
Mo_2CF_2	HCP	-8.176		5.915	5.446	2.271	2.271
Mo_2CF_2	FCC	-8.242	-0.066	6.535	4.515	2.109	2.299
Mo_2CF_2	TOP	-8.206		6.169	6.024	2.117	1.867

Table S2. Formation energies (E_{f1} and E_{f2}, eV/atom) of M_2CF_2 configurations relative to the combination of M, C and F elemental materials and the combination of bare M_2C MXene and F.

	E_{tot}	E_M	E_C	E_F	E_{f1}	E_{f2}
Sc_2CF_2	-7.68	-6.63	-9.38	-1.84	-2.42	-2.45
Y_2CF_2	-7.78	-7.03	-9.38	-1.84	-2.36	-2.46
Ti_2CF_2	-8.08	-12.30	-9.38	-1.84	-0.56	-2.24
Zr_2CF_2	-8.48	-8.60	-9.38	-1.84	-2.44	-2.27
Hf_2CF_2	-10.32	-14.19	-9.38	-1.84	-2.04	-2.26
V_2CF_2	-8.01	-9.37	-9.38	-1.84	-1.66	-1.90
Nb_2CF_2	-8.60	-10.86	-9.38	-1.84	-1.65	-1.94
Ta_2CF_2	-9.25	-13.20	-9.38	-1.84	-1.37	-1.84
Cr_2CF_2	-7.71	-10.02	-9.38	-1.84	-1.10	-1.61
Mo_2CF_2	-8.24	-11.58	-9.38	-1.84	-1.01	-1.61

Table S3. Calculated total energy differences (eV/atom) of ZXY-$M_2CF_2Li_4$ configurations to the most stable configuration.

ZXY	*M*=Sc	Y	Ti	Zr	Hf	V	Nb	Ta
HHH	0.124	0.105	0.285	0.245	0.328	0.469	0.458	0.577
FHH	0.093	0.063	0.263	0.212	0.297	0.453	0.433	0.553
THH	0.093	0.062	0.263	0.212	0.297	0.453	0.433	0.553
HHF	0.038	0.032	0.035	0.013	0.016	0.026	0.041	0.042
FHF	0.066	0.069	0.117	0.114	0.134	0.131	0.124	0.142
THF	0.005	0.044	0.013	0.005	0.006	0.010	0.017	0.021
HHT	0.034	0.032	0.025	0.013	0.036	0.017	0.030	0.030
FHT	0	0.017	0	0	0	0	0.003	0.006
THT	0.063	0.064	0.108	0.101	0.110	0.115	0.102	0.113
HFH	0.067	0.072	0.118	0.115	0.132	0.128	0.119	0.137
FFH	0.040	0.039	0.036	0.022	0.065	0.028	0.039	0.038
TFH	0.008	0.047	0.014	0.013	0.032	0.013	0.015	0.016
HFF	0.012	0.000	0.211	0.187	0.273	0.398	0.412	--
FFF	0.047	0.048	0.238	0.224	0.313	0.421	--	--
TFF	0.012	0	0.211	0.187	0.274	0.398	0.412	--
HFT	0.009	0.021	0.003	0.009	0.004	0.003	0	0
FFT	0.077	0.039	0.027	0.015	0.052	0.019	0.027	0.024
TFT	0.053	0.066	0.179	0.102	0.139	0.114	0.099	0.110
HTH	0.079	0.083	0.140	0.154	0.173	0.157	0.157	0.190
FTH	0.079	0.069	0.040	--	0.090	0.048	0.062	0.094
TTH	0.060	--	0.063	0.148	0.120	0.063	0.086	0.115
HTF	0.028	0.068	0.041	--	0.090	0.049	0.062	0.093
FTF	0.079	0.083	0.139	0.154	0.173	0.158	0.159	0.192
TTF	0.061	--	0.063	0.148	--	0.064	0.086	0.115
HTT	0.149	0.164	0.284	0.262	0.306	0.394	0.384	0.447
FTT	0.149	0.164	0.284	0.262	0.306	0.394	0.384	0.447
TTT	0.183	0.180	0.308	0.293	0.336	0.412	0.408	0.469

X=Li site, Y=F site, Z=Li site; H=HCP site, F=FCC site, T=TOP site.

"--" means that structural relaxation does not converge.

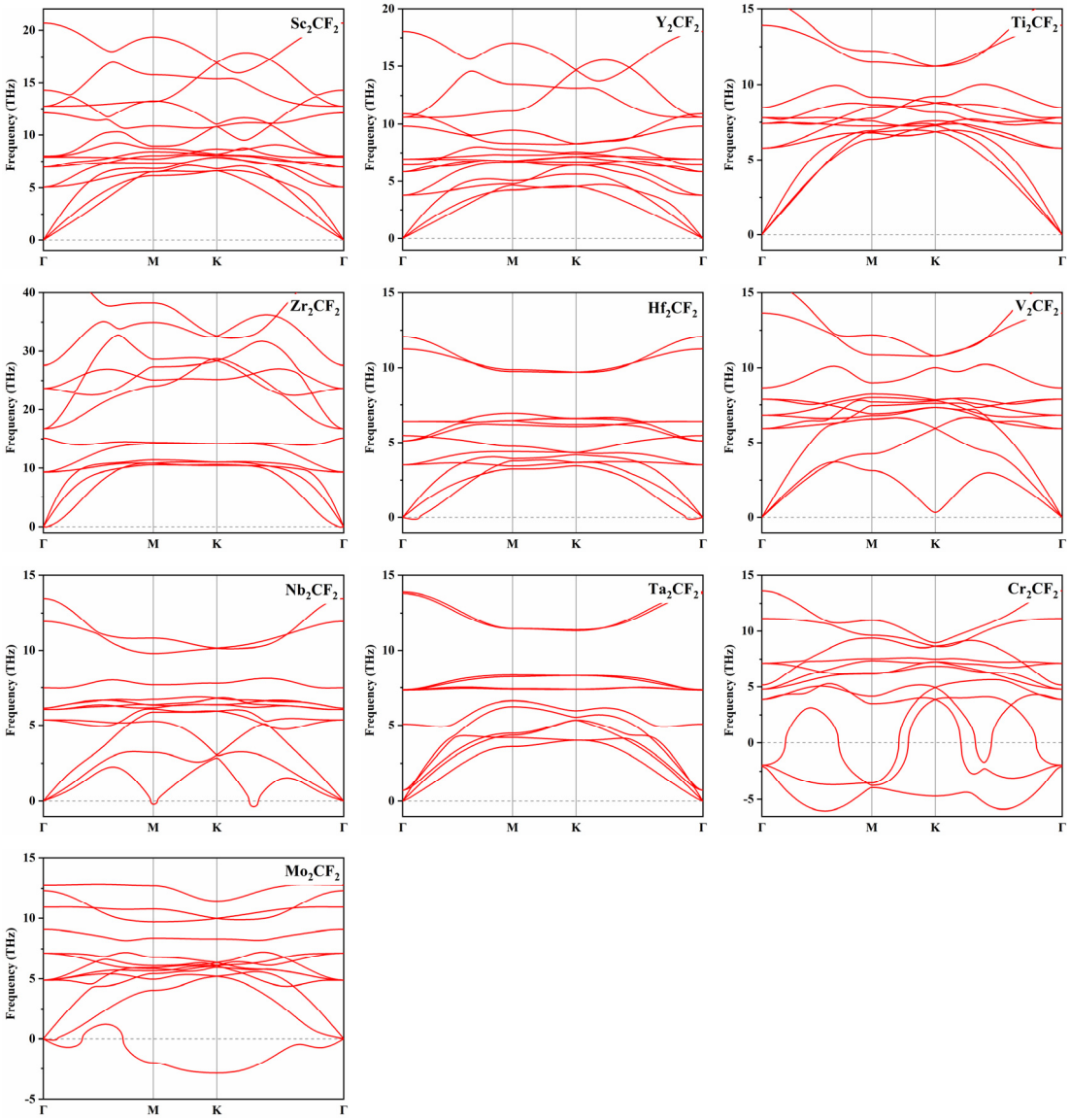


Fig. S1. Phonon dispersions of the most stable M_2CF_2 configurations.

Notes: Only a little imaginary frequencies exist for the Nb_2CF_2, which attribute to the inadequate supercell for phonon calculation.

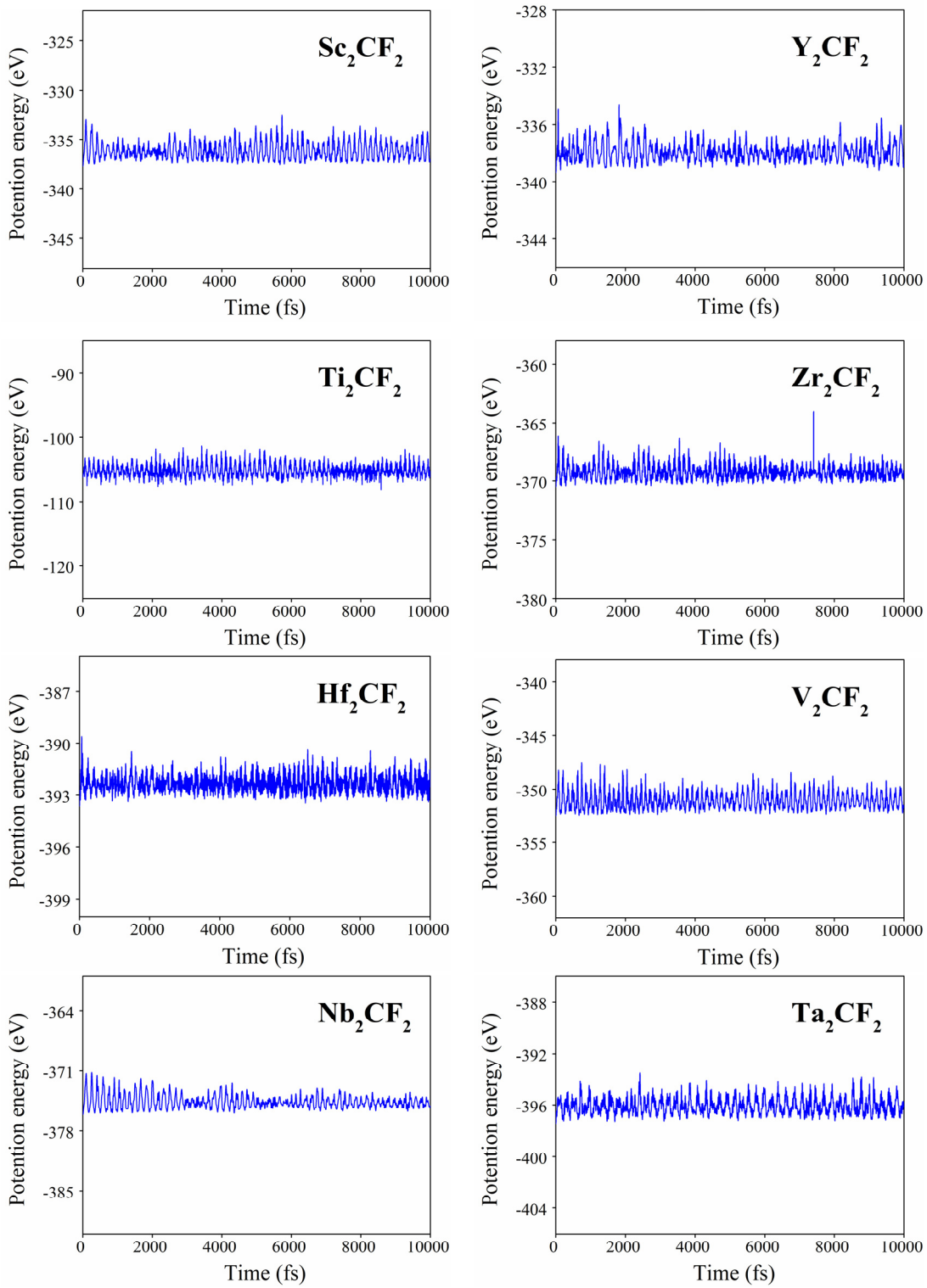
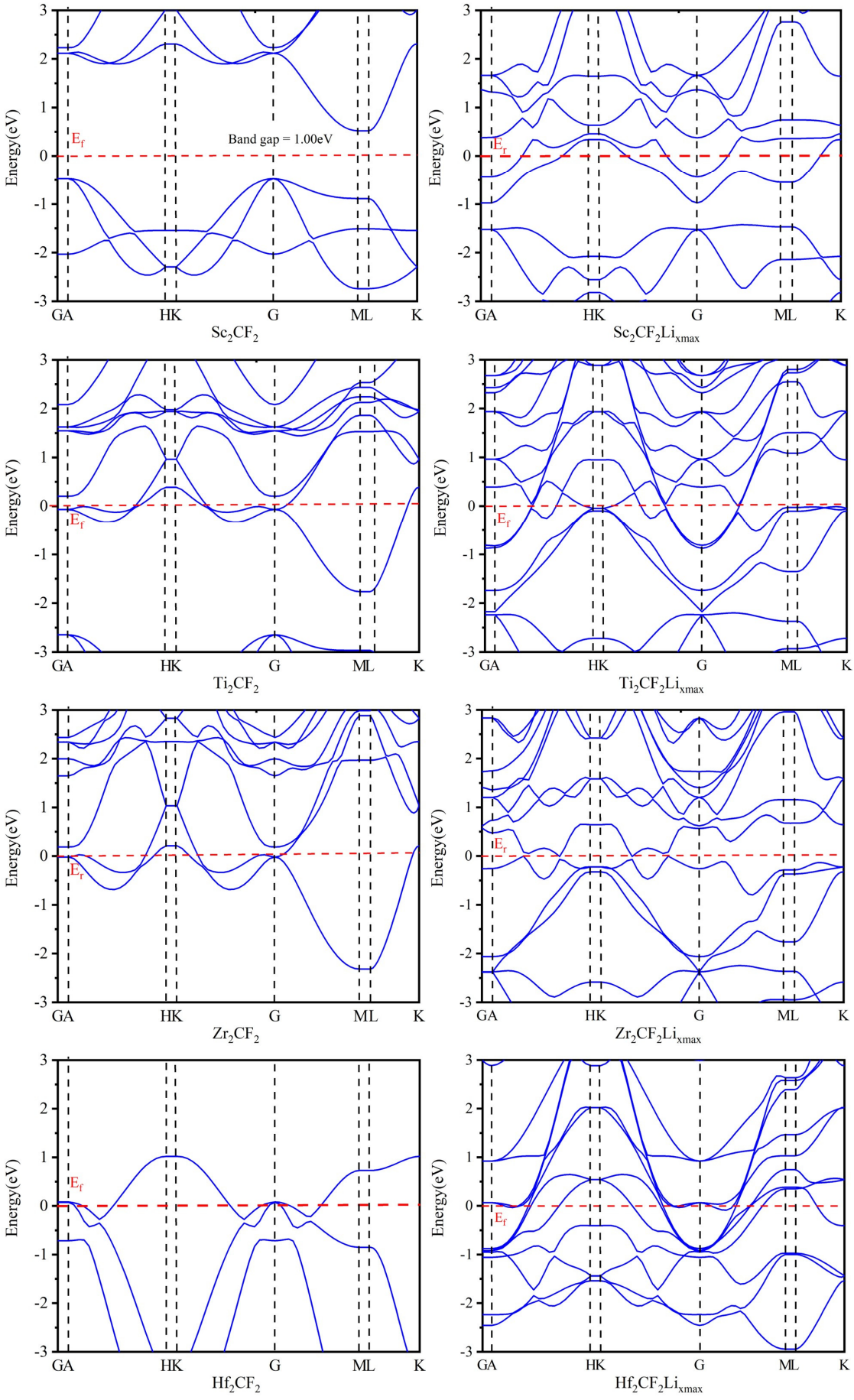


Fig. S2. Potential energy profiles of different M_2CF_2 configurations during the AIMD simulations.


Energy(eV)
E_f
Band gap = 1.00eV
GA
HK
G
ML
K
Sc2CF2
Sc2CF2Lixmax
Ti2CF2
Ti2CF2Lixmax
Zr2CF2
Zr2CF2Lixmax
Hf2CF2
Hf2CF2Lixmax

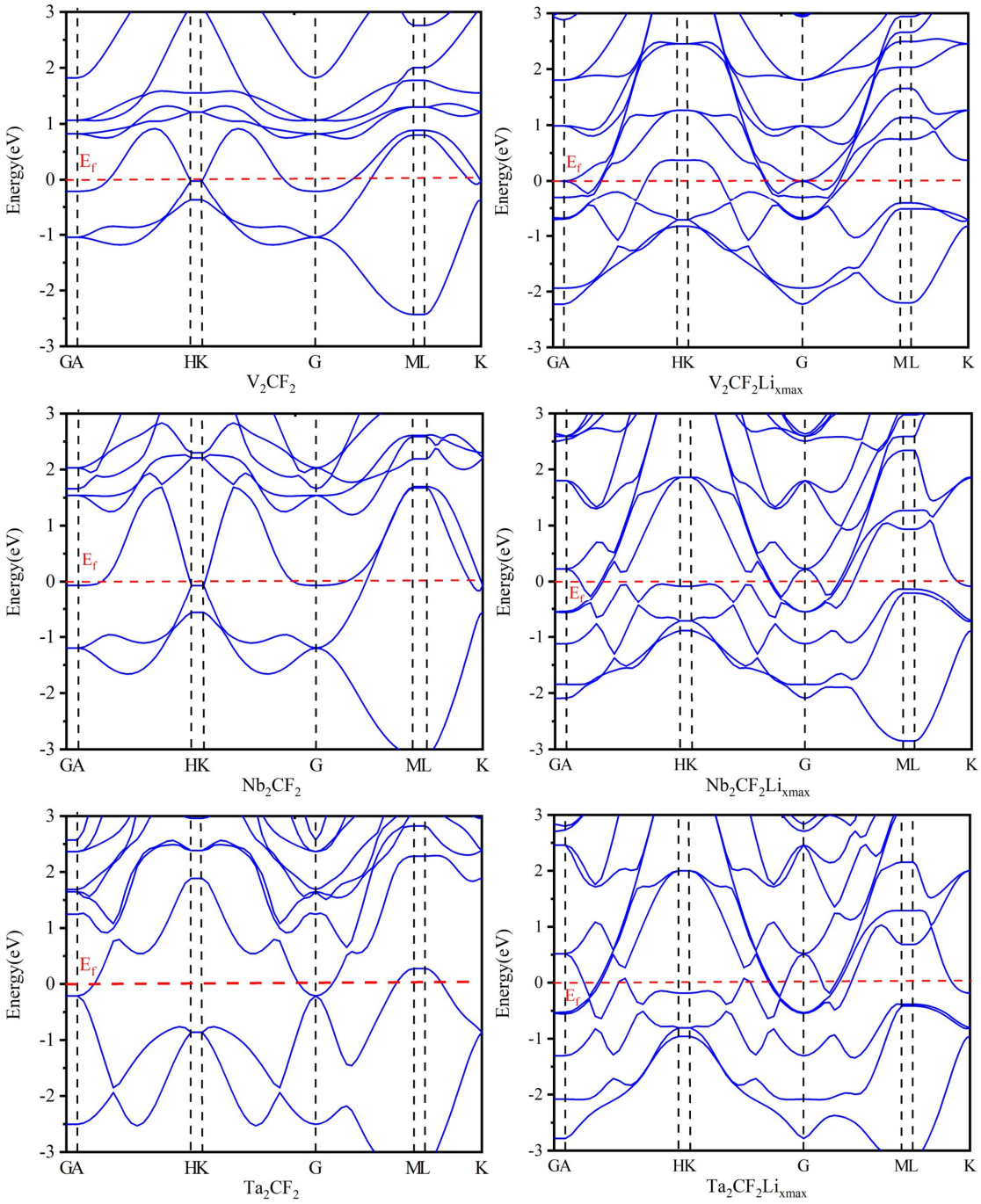
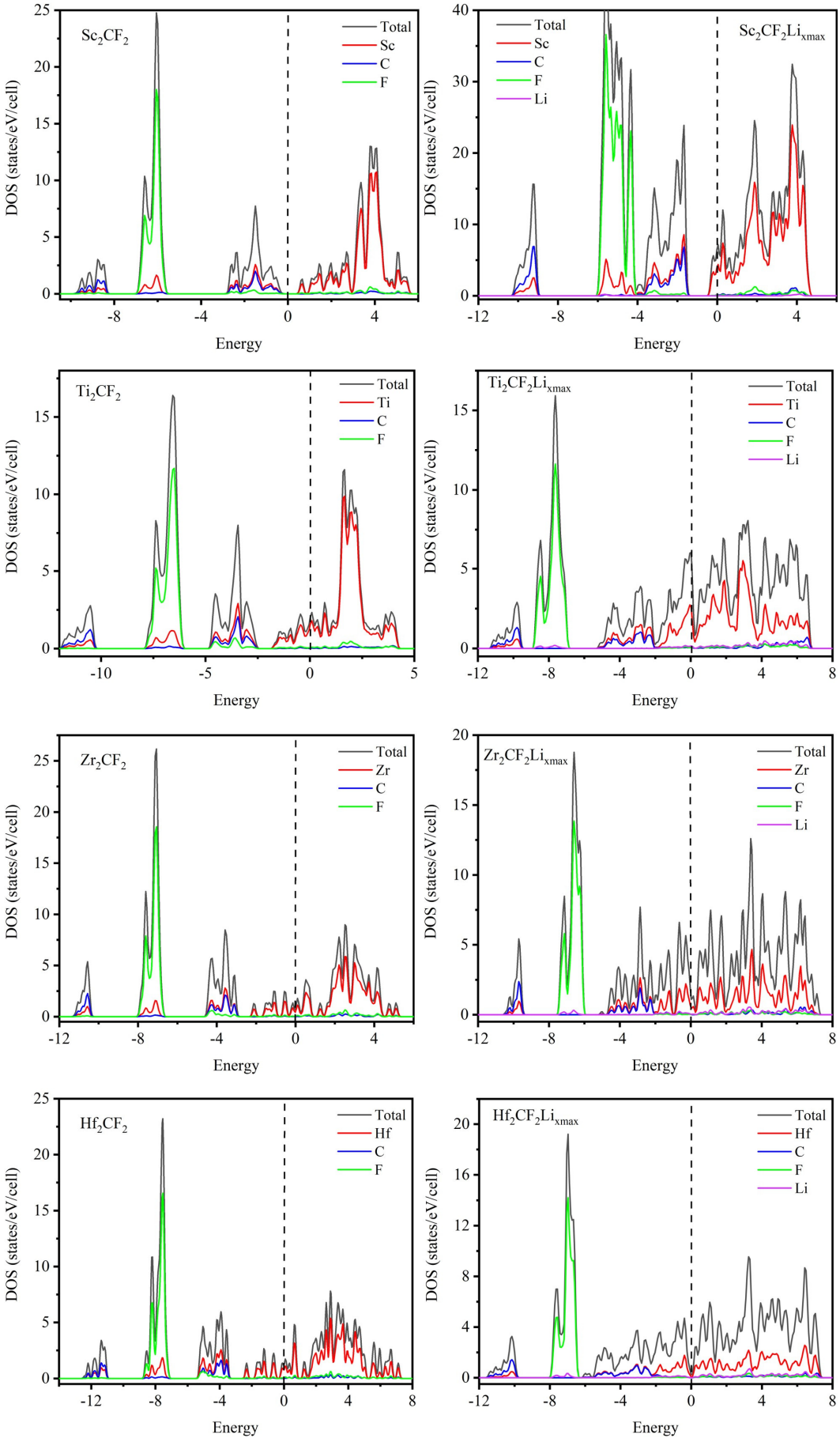


Fig. S3. Band structures of different MXenes before and after lithiation. Xmax indicates the maximum Li content.


Sc2CF2
Total
Sc
C
F
Sc2CF2Lixmax
Total
Sc
C
F
Li
Ti2CF2
Total
Ti
C
F
Ti2CF2Lixmax
Total
Ti
C
F
Li
Zr2CF2
Total
Zr
C
F
Zr2CF2Lixmax
Total
Zr
C
F
Li
Hf2CF2
Total
Hf
C
F
Hf2CF2Lixmax
Total
Hf
C
F
Li
DOS (states/eV/cell)
Energy

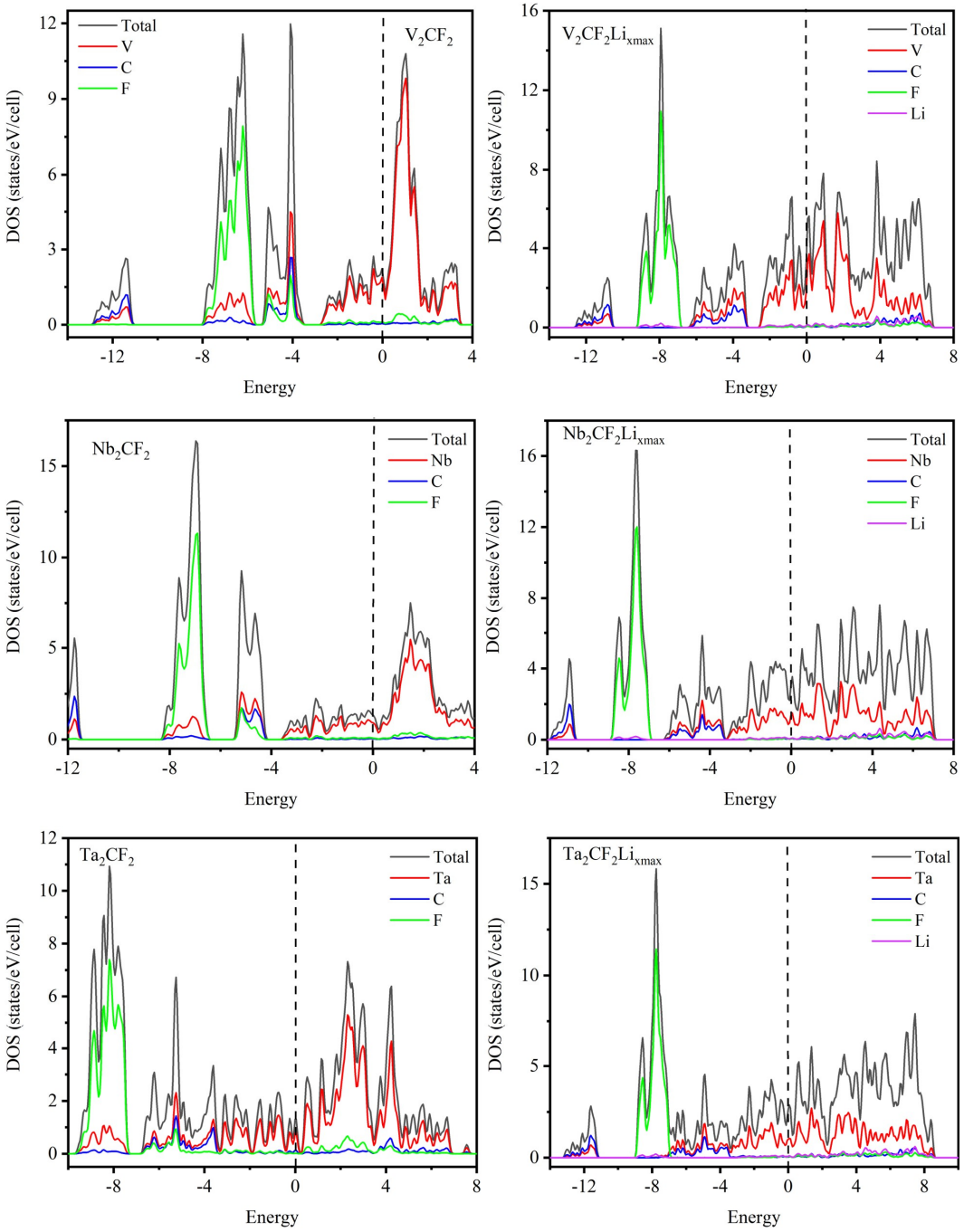


Fig. S4. Density of states of different MXenes before and after lithiation. Xmax indicates the maximum Li content.