Electronic Supplementary Material (ESI) for Nanoscale.

Supporting Information

Computational screening of pristine and functionalized ordered TiVC MXenes as highly efficient anode materials for lithium-ion batteries

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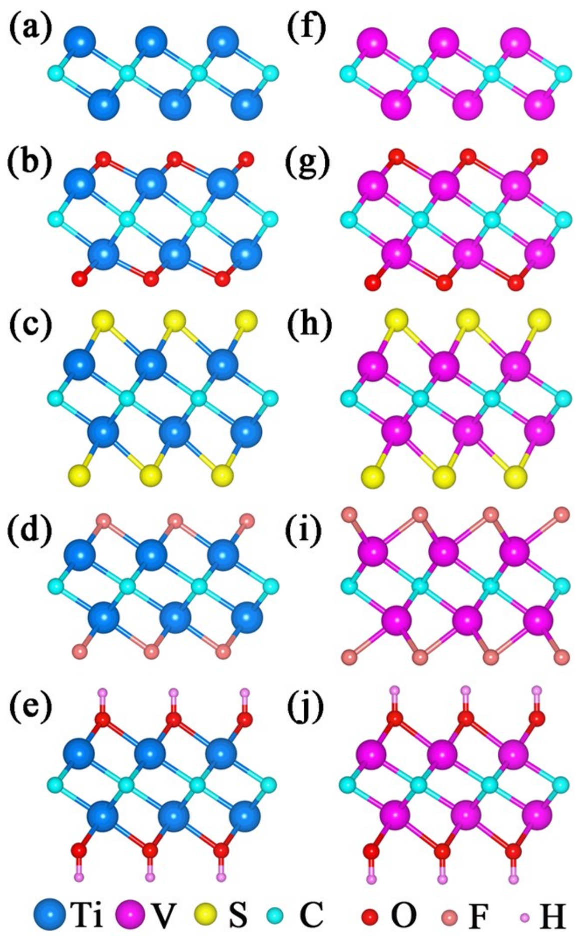

Figure S1. Side views for the most stable configurations of (a) Ti_2C, (b) Ti_2CO_2, (c) Ti_2CS_2, (d) Ti_2CF_2, (e) $Ti_2C(OH)_2$, and (f) V_2C, (g) V_2CO_2, (h) V_2CS_2, (i) V_2CF_2 and (j) $V_2C(OH)_2$ monolayers after relaxation, respectively (Referring to Figure 1 for interpretation of atomic color).

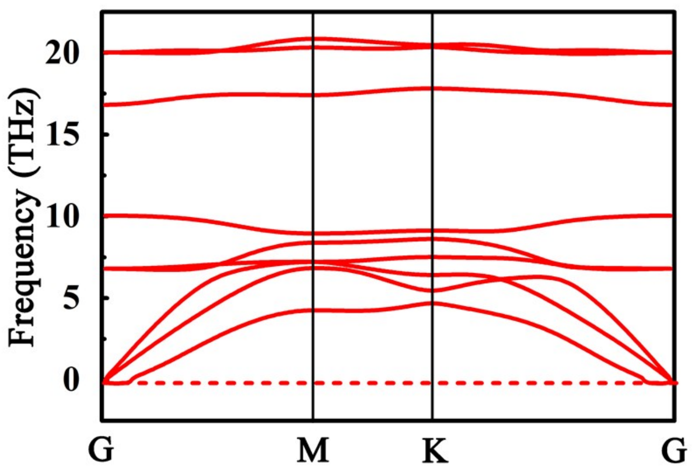

Figure S2. The calculated phonon dispersion curves of TiVC monolayer.

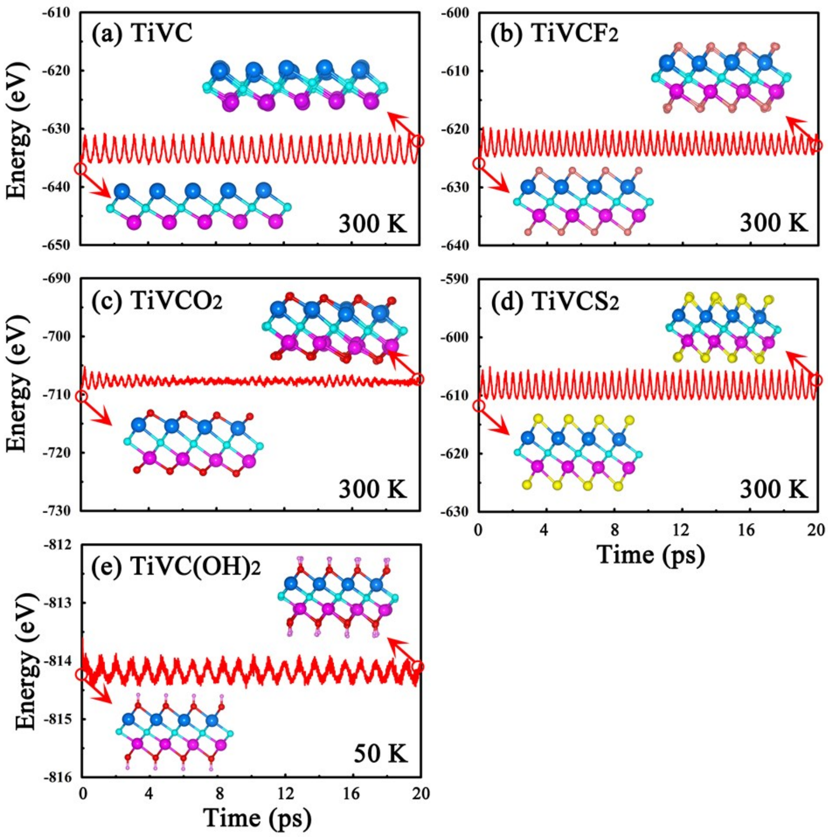

Figure S3. The pristine and functionalized TiVC MXenes after *ab initio* molecular dynamics simulations of 20 ps.

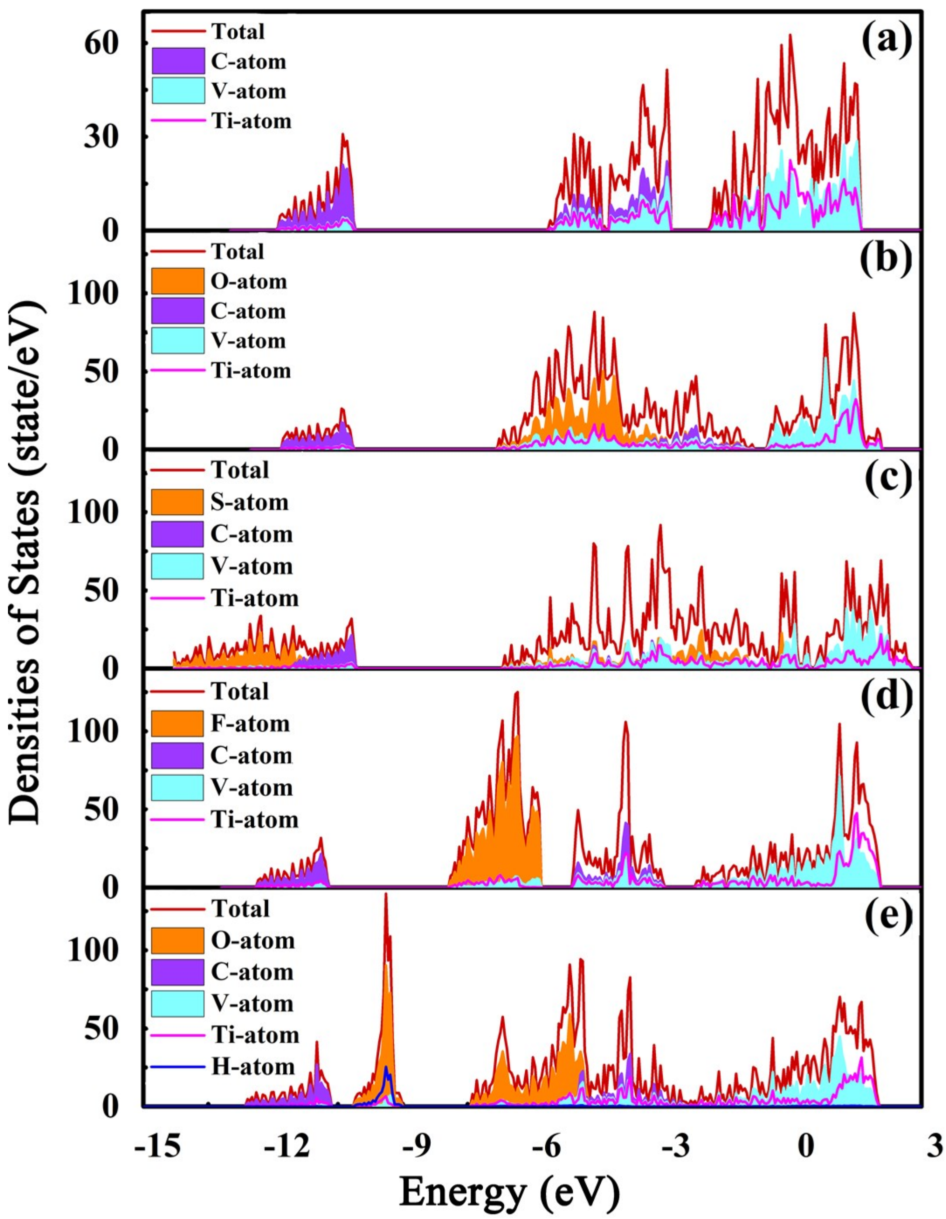

Figure S4. The density of states (DOS) for (a) TiVC, (b) $TiVCO_2$, (c) $TiVCS_2$, (d) $TiVCF_2$, and (e) $TiVC(OH)_2$ monolayers. Note that the Fermi levels are set to zero.

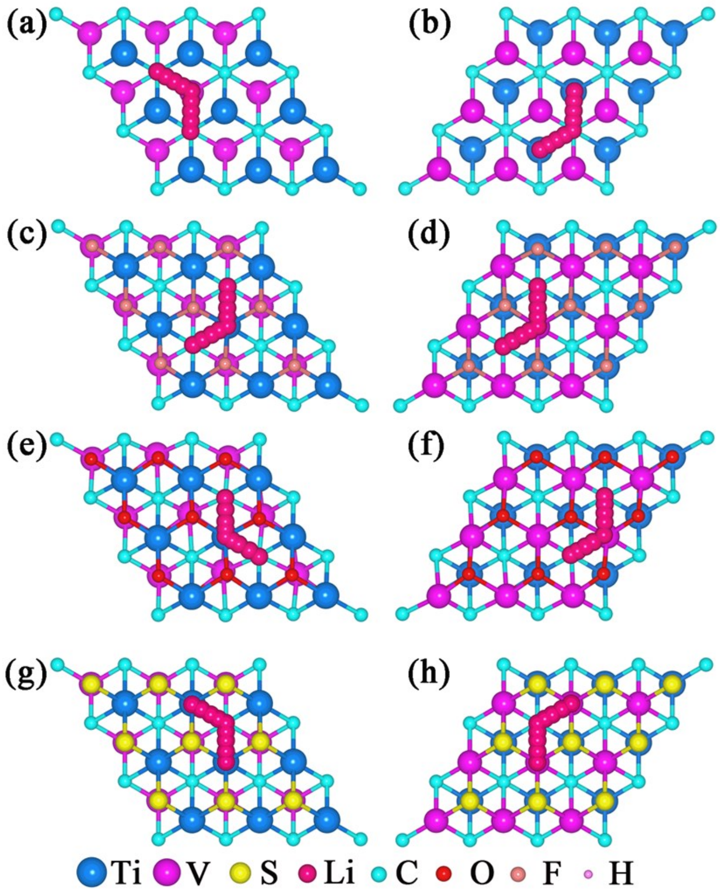

Figure S5. The diffusion pathway of Li ion on (a) Ti and (b) V surfaces of pristine TiVC monolayer, (c) Ti and (d) V surfaces of $TiVCF_2$ monolayer, (e) Ti and (f) V surfaces of $TiVCO_2$ monolayer, and (g) Ti and (h) V surfaces of $TiVCS_2$ monolayer (Referring to Figure 1 for interpretation of atomic color).

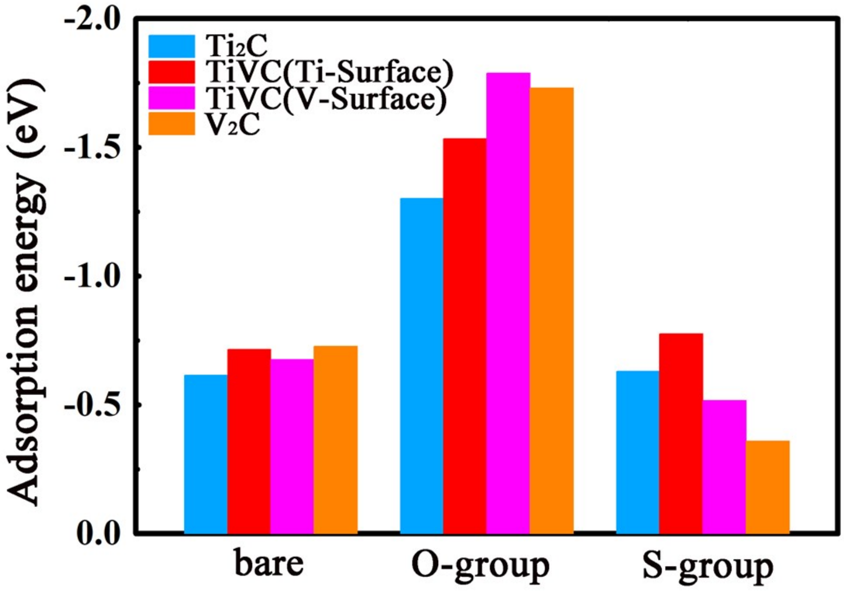

Figure S6. The average adsorption energy of multi-Li ions on favorable adsorption sites of TiVC, $TiVCO_2$ and $TiVCS_2$ monolayers.

Table S1 The structure characteristics and the amount of obtained charges, for T-groups on Ti_2CT_2, $TiVCT_2$ and V_2CT_2 (T = O, S, F, or OH) monolayers.

Materials		O-group	S-group	F-group	OH-group
Ti_2CT_2	charges	–1.097	–0.716	–0.747	–0.753
	$r_{(Ti-C)}, r_{(Ti-T)}$	2.188, 1.974	2.148, 2.381	2.097, 2.155	2.104, 2.163
$TiVCT_2$ (Ti-surface)	charges	–1.077	–0.697	–0.733	–0.744
	$r_{(Ti-C)}, r_{(Ti-T)}$	2.189, 1.932	2.114, 2.370	2.065, 2.134	2.072, 2.139
$TiVCT_2$ (V-surface)	charges	–0.990	–0.611	–0.715	–0.721
	$r_{(V-C)}, r_{(V-T)}$	2.152, 1.891	2.063, 2.325	2.013, 2.138	2.023, 2.130
V_2CT_2	charges	–0.984	–0.579	–0.674	–0.721
	$r_{(V-C)}, r_{(V-T)}$	2.057, 1.960	2.020, 2.336	2.073, 2.123	1.987, 2.111

Table S2 The adsorption energy of single Li ion on different sites of TiVC and $TiVCT_2$ (T = O, S, F, or OH) monolayers, and the bracket indicates the Li ion adsorption site after relaxation. The loss of charge amount after Li ion adsorption on surface of TiVC and $TiVCT_2$ (T = O, S, F, or OH) monolayers.

	Adsorption energy (eV)			Transfer charge (e^-)		
Adsorption Site	**C-site**	**H-site**	**T-site**	**C-site**	**H-site**	**T-site**
Ti_2C	–0.706	–0.721	–0.700(C-H)	0.835	0.835	0.835
TiVC (Ti-Surface)	–0.749	–0.735	–0.749(C)	0.837	0.838	0.837
TiVC (V-Surface)	–0.839	–0.845	–0.845(H)	0.836	0.835	0.836
V_2C	–0.911	–0.891	–0.911(C)	0.837	0.839	0.838
Ti_2CO_2	–1.974	–1.974(C)	–1.658	0.882	0.883	0.904
$TiVCO_2$ (Ti-Surface)	–2.450	–2.340(T)	–2.340	0.885	0.898	0.896
$TiVCO_2$ (V-Surface)	–2.643	–1.826	–2.446	0.885	0.915	0.897
V_2CO_2	–2.856	–2.974(C)	–2.685	0.887	0.886	0.898
Ti_2CS_2	–1.704	–1.786(T)	–1.786	0.882	0.874	0.875
$TiVCS_2$ (Ti-Surface)	–1.763	–1.865(T)	–1.865	0.881	0.872	0.872
$TiVCS_2$ (V-Surface)	–1.218	–1.340(T)	–1.340	0.887	0.874	0.874
V_2CS_2	–0.960	–0.960(C)	–1.085	0.888	0.888	0.873
Ti_2CF_2	–0.778	–0.778(C)	–0.625	0.896	0.896	0.905
$TiVCF_2$ (Ti-Surface)	–0.881	–0.769(T)	–0.769	0.898	0.905	0.904
$TiVCF_2$ (V-Surface)	–1.508	–1.390(T)	–1.390	0.894	0.902	0.902
V_2CF_2	–1.101(T)	–1.187	–1.094	0.901	0.897	0.901
$Ti_2C(OH)_2$	0.768	0.803	0.765	–0.101	0.072	–0.088
$TiVC(OH)_2$ (Ti-Surface)	0.720	0.750	0.717	–0.102	0.115	–0.096
$TiVC(OH)_2$ (V-Surface)	0.794	0.826	0.789	–0.013	0.131	0.010
$V_2C(OH)_2$	0.670	0.670	0.633	0.247	0.212	0.094

Table S3 The classical diffusion constant (k), with and without Wigner ZPE-tunneling corrected diffusion constant (kwig/tunn) and percentage of tunneling at three different temperatures (T=100, 200 and 300 K) for Li ion on pristine and functionalized Ti_2C, TiVC and V_2C monolayers.

Pristine	**T**	***K*** **(s^{-1})**	**$K_{wig/tunn}$ (s^{-1})**	**QMT (%)**
Ti_2C	100	2.007×10^{11}	2.032×10^{11}	1.230
	200	5.078×10^{11}	5.146×10^{11}	1.321
	300	6.920×10^{11}	6.976×10^{11}	0.803
TiVC (Ti-Surface)	100	3.754×10^{11}	4.031×10^{11}	6.872
	200	8.962×10^{11}	9.190×10^{11}	2.481
	300	1.198×10^{12}	1.213×10^{12}	1.237
TiVC (V-Surface)	100	6.062×10^{11}	6.646×10^{11}	8.787
	200	1.366×10^{12}	1.386×10^{12}	1.443
	300	1.791×10^{12}	1.799×10^{12}	0.445
V_2C	100	2.803×10^{11}	3.787×10^{11}	25.984
	200	8.945×10^{11}	9.990×10^{11}	10.460
	300	1.317×10^{12}	1.393×10^{12}	5.456

F-group	**T**	***k*** **in s^{-1}**	**$K_{wig/tunn}$ in s^{-1}**	**Percentage of QMT**
Ti_2CF_2	100	0.762	1.039	26.603
	200	1.277×10^{6}	1.341×10^{6}	4.773
	300	1.517×10^{8}	1.541×10^{8}	1.557
$TiVCF_2$ (Ti-Surface)	100	1.823×10^{2}	2.600×10^{2}	29.885
	200	3.781×10^{7}	4.076×10^{7}	7.237
	300	2.239×10^{9}	2.310×10^{9}	3.074
$TiVCF_2$(V-Surface)	100	5.481×10	7.539×10	27.298
	200	2.153×10^{7}	2.265×10^{7}	4.945
	300	1.577×10^{9}	1.604×10^{9}	1.683
V_2CF_2	100	2.385×10^{4}	6.500×10^{3}	/
	200	2.285×10^{8}	1.421×10^{8}	/
	300	4.854×10^{9}	3.829×10^{9}	/

O-group	T	k in s^{-1}	$K_{wig/tunn}$ in s^{-1}	Percentage of QMT
Ti_2CO_2	100	1.794×10^{-5}	1.902×10^{-3}	99.057
	200	2.942×10^{3}	1.172×10^{5}	97.490
	300	1.610×10^{6}	5.139×10^{7}	96.867
$TiVCO_2$ (Ti-Surface)	100	2.431×10	2.665×10	8.780
	200	1.137×10^{7}	1.129×10^{7}	/
	300	8.821×10^{8}	8.739×10^{8}	/
$TiVCO_2$ (V-Surface)	100	2.479	0.662	/
	200	1.094×10^{6}	6.880×10^{5}	/
	300	8.326×10^{7}	6.617×10^{7}	/
V_2CO_2	100	4.037×10	9.386×10	56.989
	200	1.680×10^{7}	2.143×10^{7}	21.605
	300	1.255×10^{9}	1.403×10^{9}	10.549

S-group	T	k in s^{-1}	$K_{wig/tunn}$ in s^{-1}	Percentage of QMT
Ti_2CS_2	100	1.823×10^{3}	2.659×10^{3}	31.440
	200	1.185×10^{8}	1.293×10^{8}	8.353
	300	4.766×10^{9}	4.946×10^{9}	3.639
$TiVCS_2$ (Ti-Surface)	100	1.412×10^{3}	1.893×10^{3}	25.409
	200	9.730×10^{7}	1.039×10^{8}	6.352
	300	3.989×10^{9}	4.101×10^{9}	2.731
$TiVCS_2$(V-Surface)	100	2.564×10^{3}	3.041×10^{3}	15.686
	200	1.247×10^{8}	1.278×10^{8}	2.426
	300	4.553×10^{9}	4.587×10^{9}	0.741
V_2CS_2	100	1.323×10^{4}	1.821×10^{4}	27.348
	200	2.857×10^{8}	3.099×10^{8}	7.809
	300	7.955×10^{9}	8.250×10^{9}	3.576