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Understanding the tunable sodium storage performance in pillared MXene: A first-principles study

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Supplementary Material

Table S1. Layer spacing before and after enforcing the optimization process of selected structures.

Layer spacing (Å)	**Layer spacing (Å) (after optimization)**			
(Initial)	**Ti_2CO_2-SL**	**Ti_2CO_2-DL**	**$Ti_3C_2O_2$-SL**	**$Ti_3C_2O_2$-DL**
3	3.10	3.09	3.08	3.06
4	4.07	4.08	4.06	4.05
5	5.05	5.06	5.05	5.06
6	6.04	6.04	6.08	6.04
7	7.02	7.03	7.03	7.03
8	8.03	8.02	8.02	8.03
9	9.01	9.01	9.00	9.02
10	10.00	10.01	10.01	10.01
12	12.00	12.01	12.00	12.01

Table S2. The adsorption energy of Na on selected structures with different layer spacing.

Layer spacing (Å)	E_a(eV per Na)							
	Ti_2CO_2-SL				$Ti_3C_2O_2$-SL			
	1/16 ML	1/9 ML	1/4 ML	1 ML	1/16 ML	1/9 ML	1/4 ML	1 ML
3	4.16	4.20	3.81	3.72	4.61	4.53	4.27	3.82
3.5	4.54	4.35	4.06	3.93	4.89	4.82	4.46	3.97
4	4.89	4.66	4.50	4.07	5.07	5.00	4.70	4.13
4.5	4.57	4.51	4.05	3.49	4.92	4.90	4.51	3.56
5	4.21	4.11	3.58	3.21	4.63	4.58	4.32	3.35
5.5	3.89	3.82	3.49	2.37	4.41	4.38	4.10	2.83
6	3.82	3.73	3.41	2.01	4.33	4.31	3.96	2.35
7	3.75	3.69	3.39	1.97	4.21	4.12	3.74	2.26
8	3.73	3.63	3.39	1.96	4.18	4.08	3.69	2.24
9	3.73	3.63	3.37	1.95	4.17	4.06	3.66	2.24

Table S3. Calculated in-plane planar elastic constants C_{11} and Poisson's ratio (v) of MXene-Na system at different layer spacing. The data in brackets correspond to the double Na-atomic layers adsorption system.

Layer	**C_{11} (N/m)**		**v**	
spacing (Å)	**Ti_2CO_2-Na**	**$Ti_3C_2O_2$-Na**	**Ti_2CO_2-Na**	**$Ti_3C_2O_2$-Na**
3	349	421	0.296	0.279
4	280	388	0.278	0.257
5	278 (336)	381 (438)	0.212 (0.328)	0.208 (0.298)
6	286 (314)	385 (406)	0.236 (0.288)	0.214 (0.286)
7	287 (301)	393 (389)	0.242 (0.269)	0.231 (0.272)
8	291 (286)	395 (375)	0.250 (0.260)	0.239 (0.255)
9	292 (274)	394 (379)	0.255 (0.259)	0.243 (0.257)

Table S4. The adsorption energy of Na on selected structures with different layer spacing at 1 ML Na coverage.

Layer spacing (Å)	E_a(eV per Na)	
	Ti_2CO_2(C-Ti stacking)	$Ti_3C_2O_2$(C-C stacking)
3	3.21	3.89
4	3.66	4.13
5	2.69	3.34
6	1.93	2.3
7	1.91	2.19
8	1.9	2.18

Table S5. The normalized values of all the characteristic indicators by the Min-max standardization method. The normalized equation for E_a and C_{11} is X = (x - min)/(max - min), and for EB_d and v is X = (max - x)/(max - min). All EB_d data (C-C stacking and C-Ti stacking) were combined together for normalized calculation.

MXene	**Layer spacing (Å)**	**E_a**	**EB_d (C-C stacking)**	**EB_d (C-Ti stacking)**	**C_{11}**	**v**
Ti_2CO_2	5	0.48	0	1	1	0
	6	0.76	0.55	0.90	0.57	0.59
	7	1	0.65	0.75	0.30	0.87
	8	0	0.68	0.71	0	1
$Ti_3C_2O_2$	5	0.44	0	1	1	0
	6	1	0.67	0.85	0.50	0.28
	7	0	0.71	0.81	0.22	0.60
	8	0.33	0.75	0.77	0	1

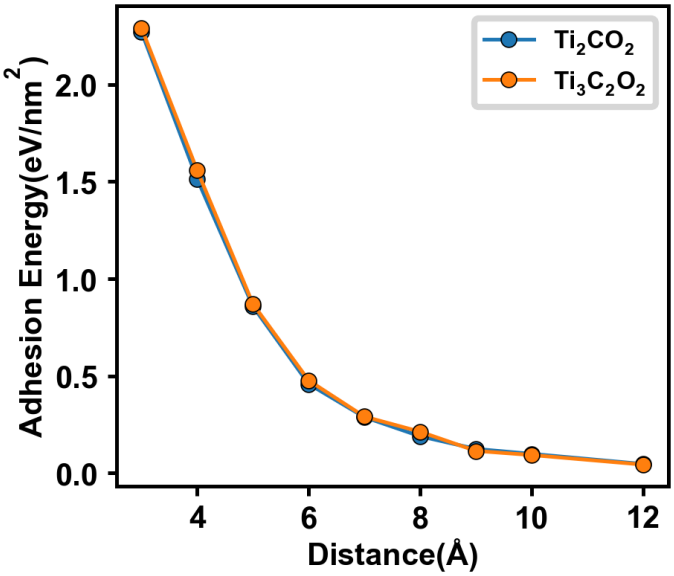

Fig. S1. Distribution of the adhesion energy between MXene layers with different layer spacing for double-layer MXene models.

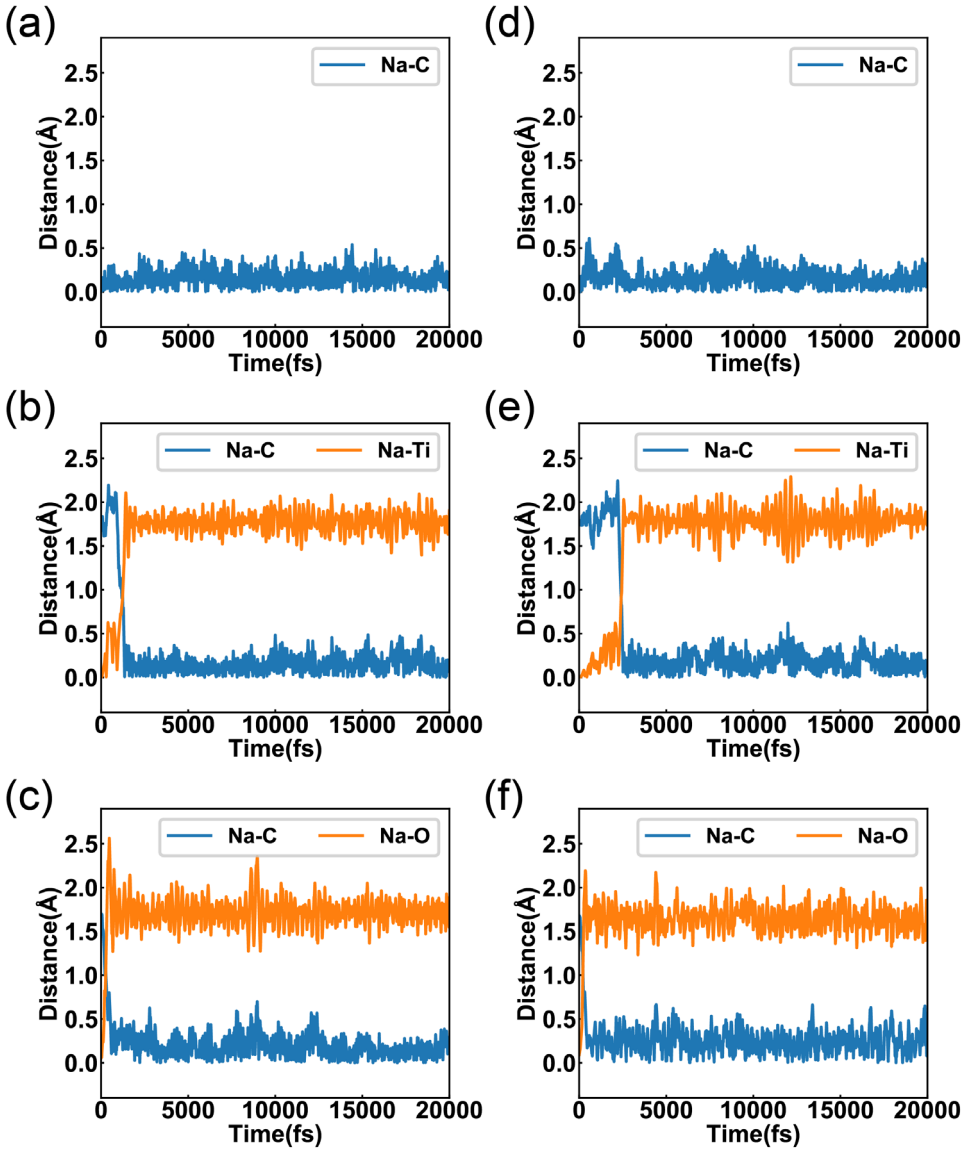

Fig. S2. Change of the distance in the x−y plane between Na and the initial and final adsorption site along the MD simulation time. (a) The initial and final adsorption site is C in Ti_2CO_2, (b) The initial adsorption site is Ti and the final adsorption site is C site in Ti_2CO_2, (c) The initial adsorption site is O and the final adsorption site is C in Ti_2CO_2, (d) The initial and final adsorption site is C in $Ti_3C_2O_2$, (e) The initial adsorption site is Ti and the final adsorption site is C site in $Ti_3C_2O_2$, (f) The initial adsorption site is O and the final adsorption site is C in $Ti_3C_2O_2$.

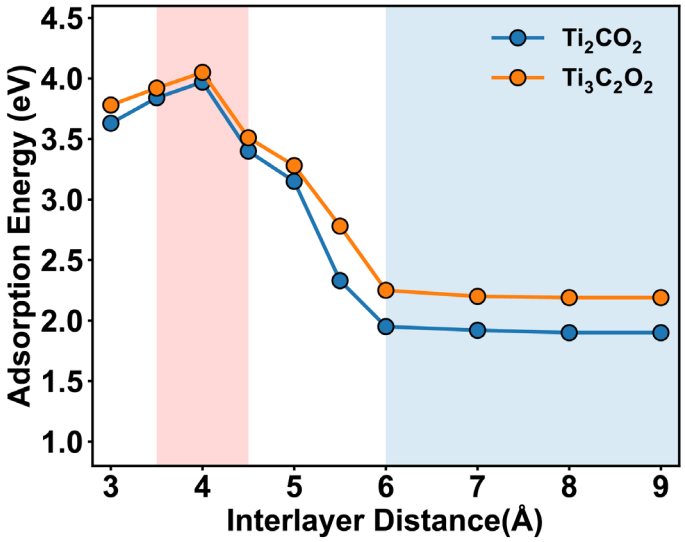

Fig. S3. Change of the Na adsorption energy with layer spacing in double-layer MXene models.

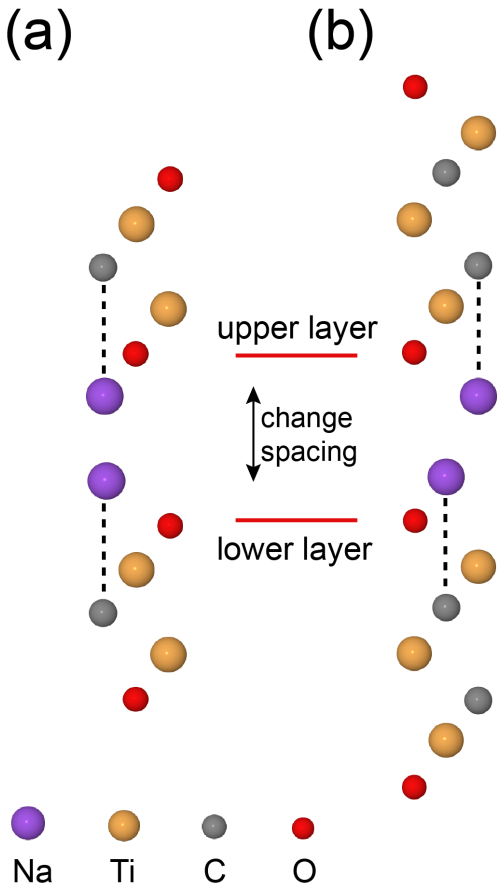

Fig. S4. Schematic diagram illustrating double Na-atomic layers adsorption in 1 x 1 MXene model (a) Ti_2CO_2 and (b) $Ti_3C_2O_2$.

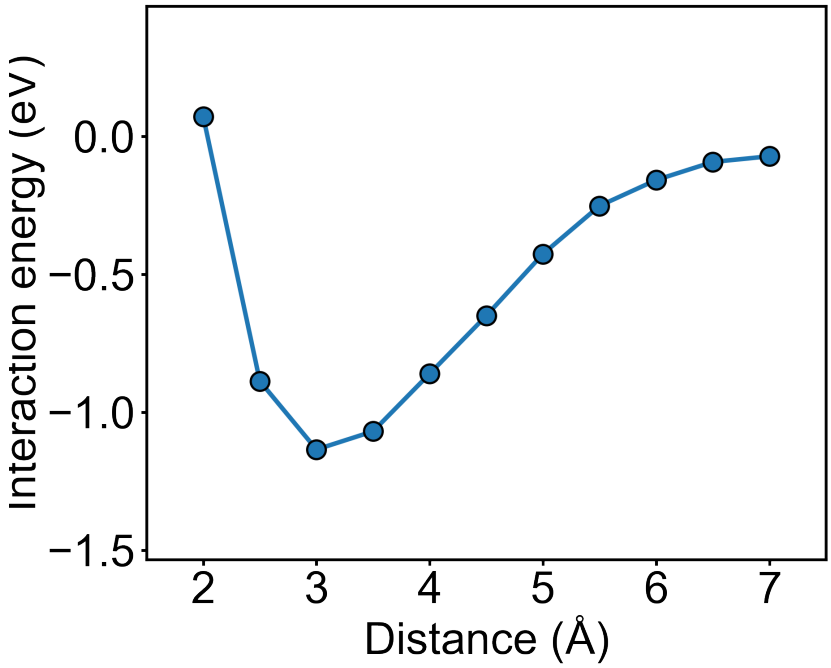

Fig. S5. The interaction energy between two Na atoms as a function of Na-Na distance.