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Unraveling Structure-Performance Relationships: Tailored d-band Centers in monolayer MSi_2N_4 and $MoSi_2Z_4$ by Atomic Substitution

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Table S1 The lattice constants (a), alterations in bond lengths (d_1-d_4), angles (θ_1-θ_4) and band gaps of monolayer MSi_2N_4 and $MoSi_2Z_4$

	a(Å)	d_1(Å)	d_2(Å)	d_3(Å)	d_4(Å)	θ_1(°)	θ_2(°)	θ_3(°)	θ_4(°)	E_g (eV)
$MoSi_2N_4$	2.91	6.99	1.75	1.75	2.09	112.09	106.71	88.22	73.01	1.70
$CrSi_2N_4$	2.84	6.86	1.72	1.75	2.00	110.74	108.17	90.36	70.02	0.51
WSi_2N_4	2.90	7.01	1.75	1.74	2.10	112.26	106.60	87.84	73.60	2.13
$TaSi_2N_4$	2.96	7.00	1.78	1.75	2.12	112.70	106.00	88.29	72.94	/
$TiSi_2N_4$	2.92	6.90	1.76	1.75	2.06	111.86	106.96	90.1	70.44	1.63
$ZrSi_2N_4$	3.03	7.04	1.81	1.75	2.18	113.27	105.34	88.26	72.97	1.57
$HfSi_2N_4$	3.02	6.99	1.80	1.75	2.18	112.95	105.65	88.80	72.12	1.63
$MoSi_2P_4$	3.46	9.36	2.25	2.23	2.45	100.73	117.22	89.75	70.88	0.69
$MoSi_2As_4$	3.61	9.90	2.36	2.34	2.56	99.43	118.25	89.56	71.15	0.60

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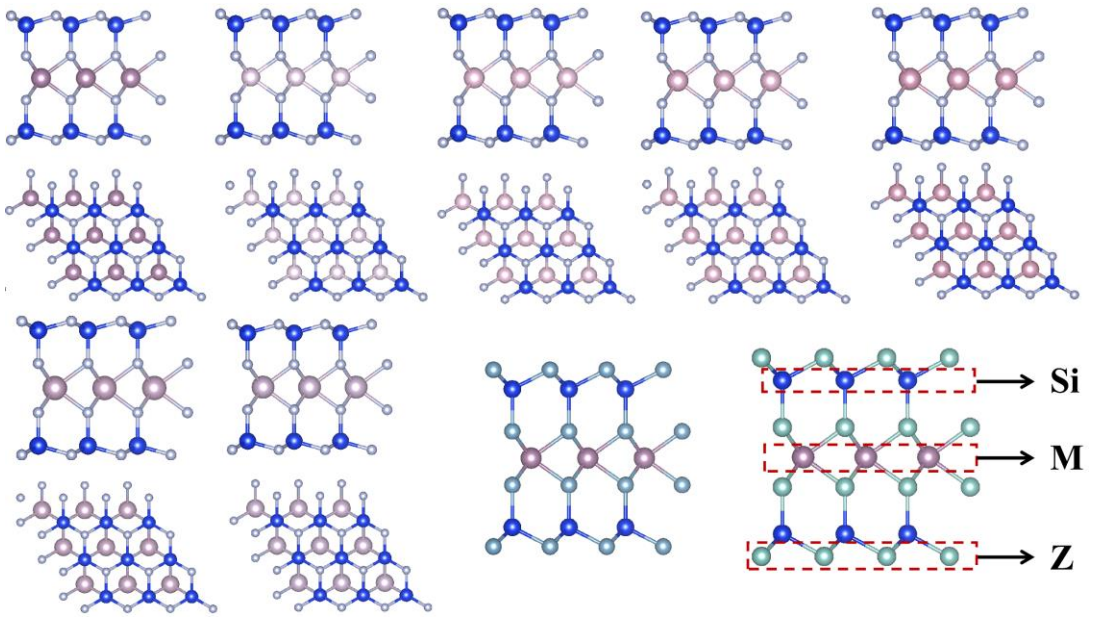

Fig. S1 Side view and top view of monolayer MSi_2N_4 (M= Mo, Cr, W, Ta, Ti, Zr and Hf) and $MoSi_2Z_4$ (Z= P and As)

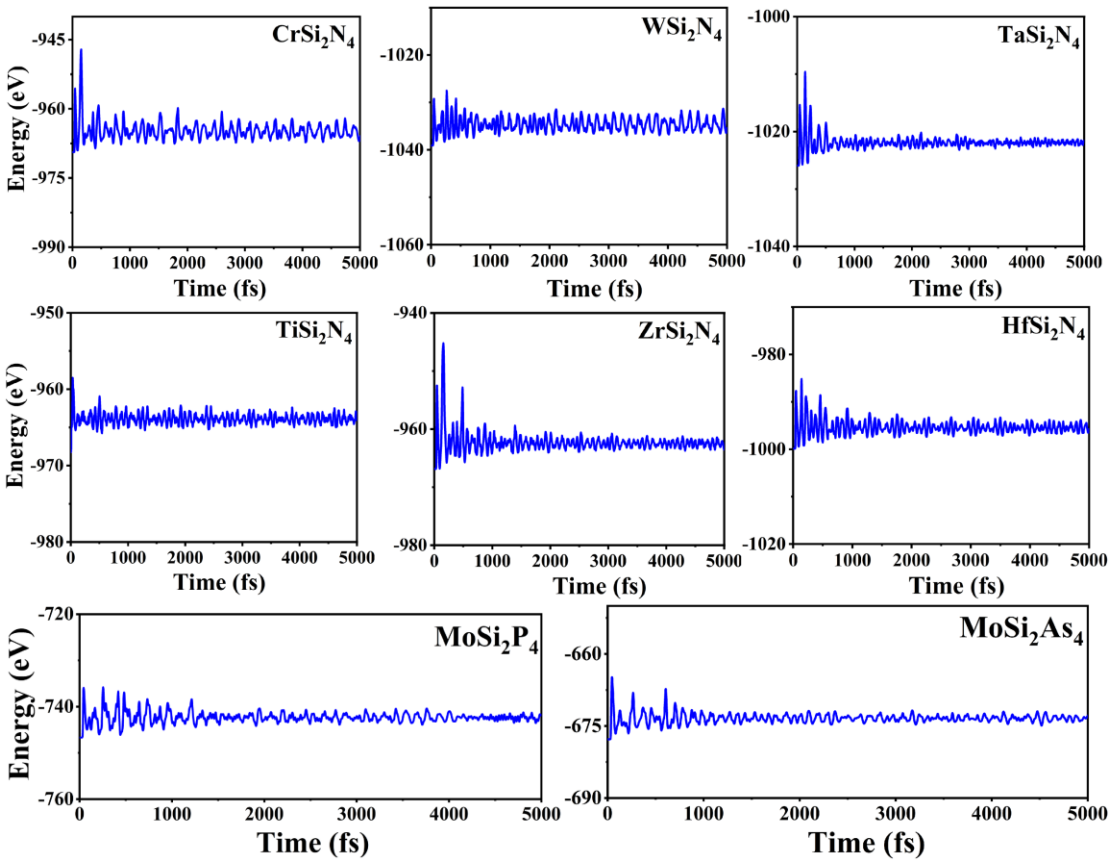

Fig. S2 The free energy and temperature over 5 ps during AIMD at 300 K of monolayer MSi_2N_4 and $MoSi_2Z_4$

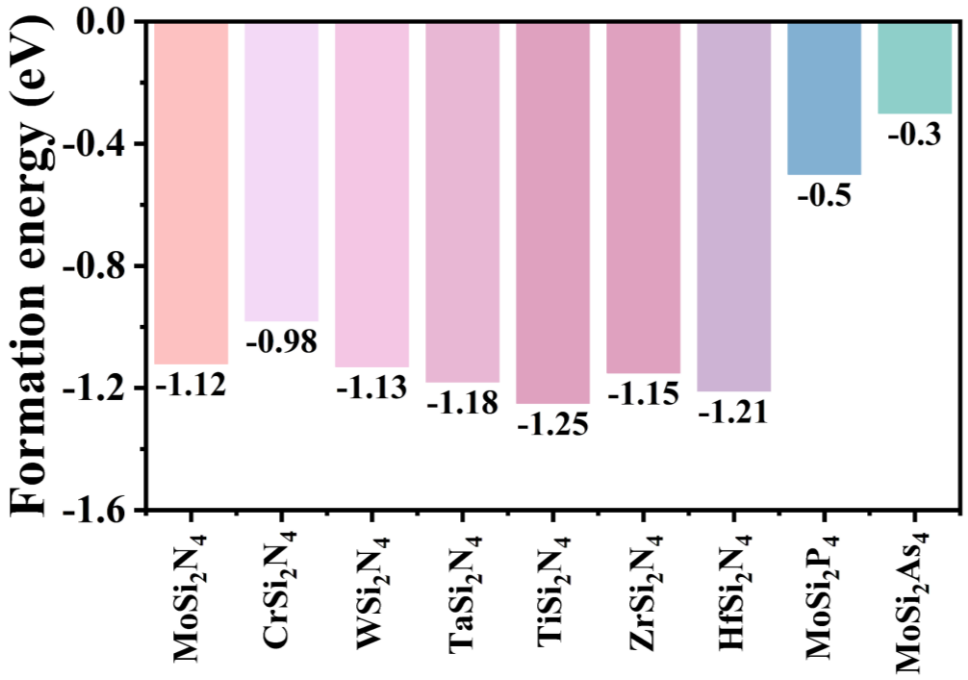

Fig. S3 The formation energy of monolayer MSi_2N_4 and $MoSi_2Z_4$

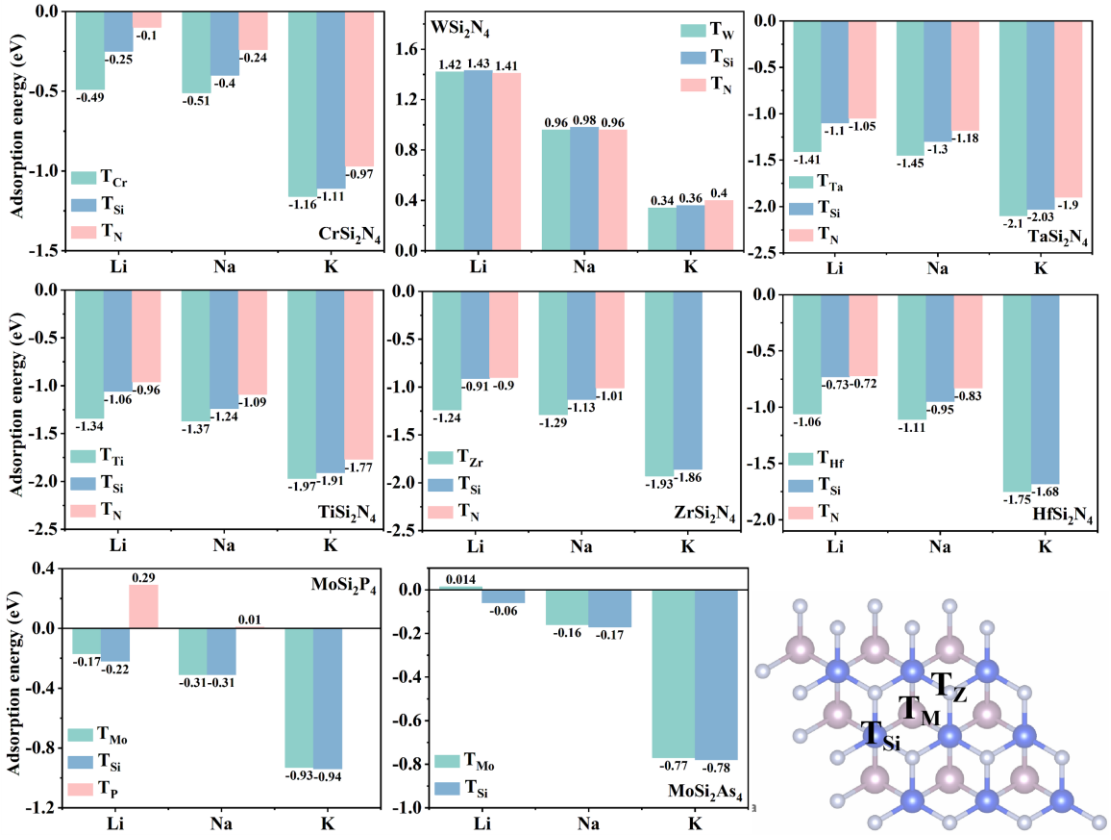

Fig. S4 The adsorption energy of stable adsorption site for alkali metal atoms of monolayer MSi_2N_4 and $MoSi_2Z_4$

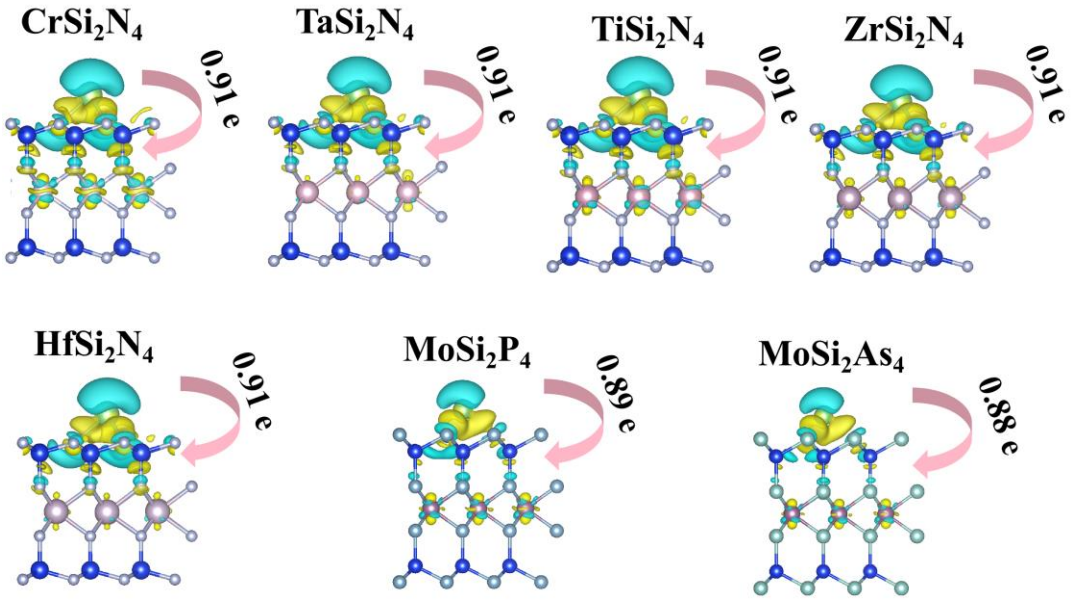

Fig.S5 The charge density difference at the most stable positions of Li adsorption on monolayer MSi_2N_4 and $MoSi_2Z_4$

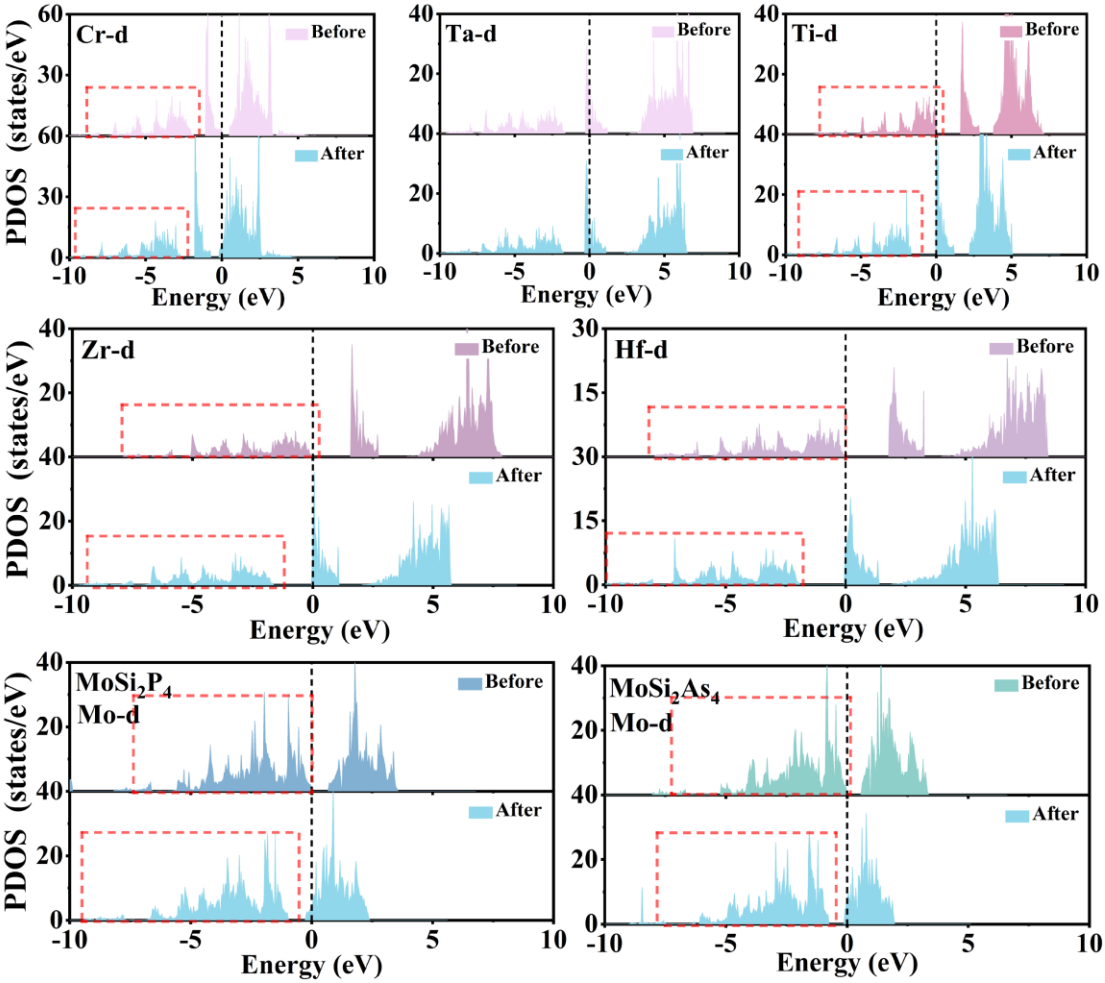

Fig.S6 The PDOS of TMs before and after Li adsorption

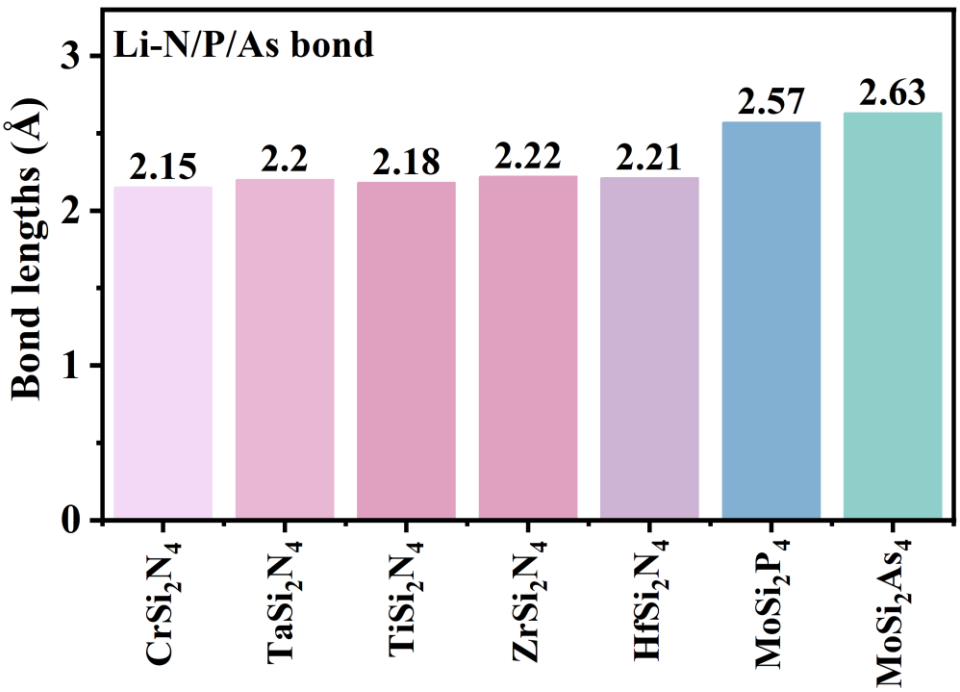

Fig. S7 The bond lengths at the most stable positions of Li adsorption on monolayer MSi_2N_4 and $MoSi_2Z_4$

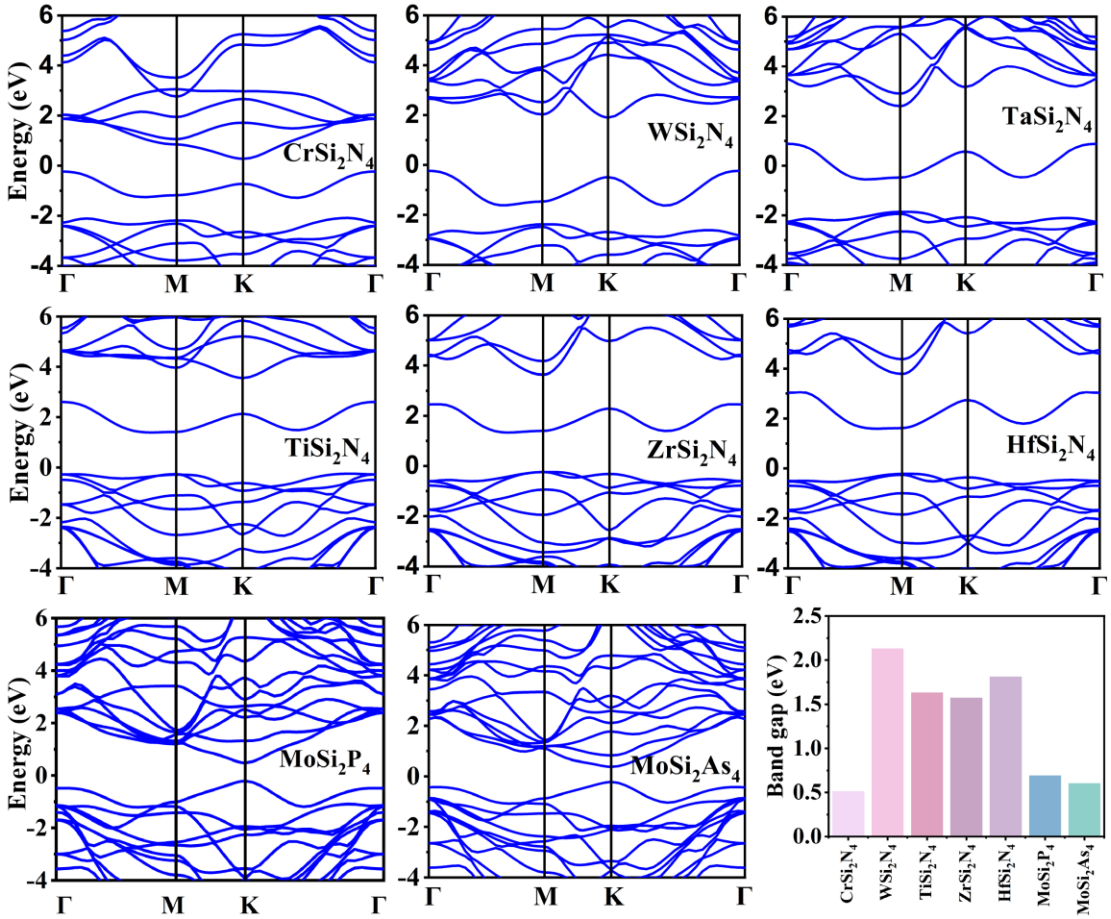

Fig. S8 The band structures of monolayer MSi_2N_4 and $MoSi_2Z_4$

Table S2 Elastic Properties of MSi_2N_4 and $MoSi_2Z_4$ (Elastic Constants C_{ij}, Shear Modulus G^{2D},Young's Modulus Y in N/m, and Poisson's Ratio ν)

	C_{11}	C_{12}	C_{22}	$C_{66}=G^{2D}$	$Y_{[x]}$	$Y_{[y]}$	$\nu_{[x]}$	$\nu_{[y]}$
$CrSi_2N_4$	523.71	154.37	523.71	184.67	478.21	478.21	0.30	0.30
WSi_2N_4	574.64	158.07	574.64	208.29	531.16	531.16	0.28	0.28
$TaSi_2N_4$	518.85	166.10	518.85	176.37	465.67	465.67	0.32	0.32
$TiSi_2N_4$	498.16	153.31	498.16	172.43	450.98	450.98	0.31	0.31
$ZrSi_2N_4$	437.89	138.86	437.89	149.52	393.85	393.85	0.32	0.32
$HfSi_2N_4$	459.25	156.23	459.25	151.51	406.11	406.11	0.34	0.34
$MoSi_2P_4$	218.90	55.57	218.90	81.67	204.80	204.80	0.25	0.25
$MoSi_2As_4$	181.46	52.83	181.46	64.32	166.08	166.08	0.29	0.29

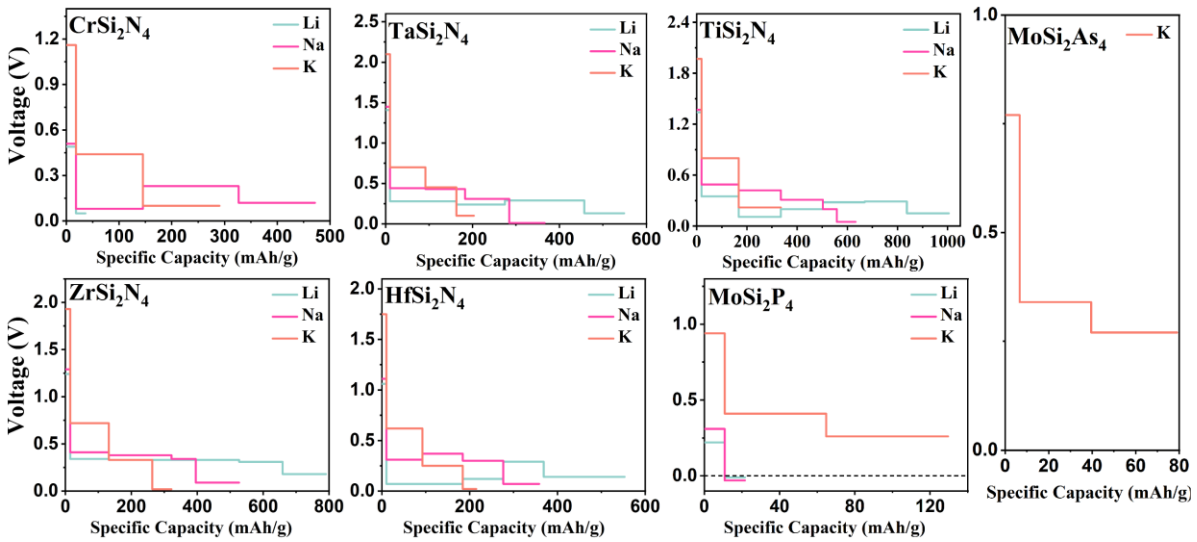

Fig. S9 The voltage profiles as a function of specific capacity of monolayer MSi_2N_4 and $MoSi_2Z_4$