Supplementary Materials for

Design of 2D materials – $MSi_2C_xN_{4-x}$ (M = Cr, Mo, and W; x = 1 and 2) with tunable electronic and magnetic properties

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Table S1. Structural parameters, including lattice constants, bonding angle, and corresponding space group for MSi_2N_4 and $MSi_2C_xN_{4-x}$.

Structure	**a (Å)**	**b (Å)**	**c (Å)**	**θ1 (°)**	**θ2 (°)**	**Space Group**
$CrSi_2N_4$	2.84531	2.84531	24.53985	90.45	69.90	P3m1
$CrSi_2CN_3$	2.93316	2.93316	23.09139	90.58	70.06	P3m1
$CrSi_2N_2C_2$	3.02460	3.02460	23.64520	94.02	64.73	$P\bar{6}m2$
$CrSi_2N_3C$	2.92179	2.92179	25.33855	92.24	67.78	P3m1
$MoSi_2N_4$	2.91155	2.91155	25.51660	88.14	73.13	P3m1
$MoSi_2CN_3$	2.92303	2.92303	23.25167	87.53	73.25	P3m1
$MoSi_2N_2C_2$	3.04500	3.04500	23.32958	91.26	68.73	$P\bar{6}m2$
$MoSi_2N_3C$	2.97916	2.97916	24.37215	90.20	70.75	P3m1
WSi_2N_4	2.91605	2.91605	23.36308	87.87	73.51	P3m1
WSi_2CN_3	2.93970	2.93970	22.98874	87.40	72.97	P3m1
$WSi_2N_2C_2$	3.04615	3.04615	23.31187	91.03	69.07	$P\bar{6}m2$
WSi_2N_3C	2.98016	2.98016	24.35565	90.03	89.20	P3m1

Table S2. Calculated ground energy (eV/supercell), band gap (E_g) (eV) and conducting propoerties (CP) of MSi_2N_4 systems in non-magnetic (NM), anti-ferromagnetic (AFM) and ferromagnetic (FM) states. S – Semiconductor.

Structure	NM	AFM	FM	E_g	CP
$CrSi_2N_4$	-238.344	-238.344	-238.344	0.49	S
$MoSi_2N_4$	-246.916	-246.916	-246.916	1.74	S
WSi_2N_4	-255.028	-255.028	-255.028	2.08	S

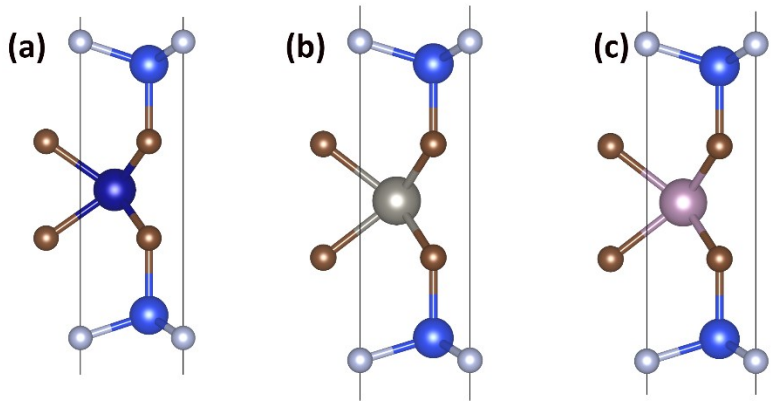

Figure S1. Structures for (a) $CrSi_2C_2N_2$, (b) $MoSi_2C_2N_2$, and (c) $WSi_2C_2N_2$.

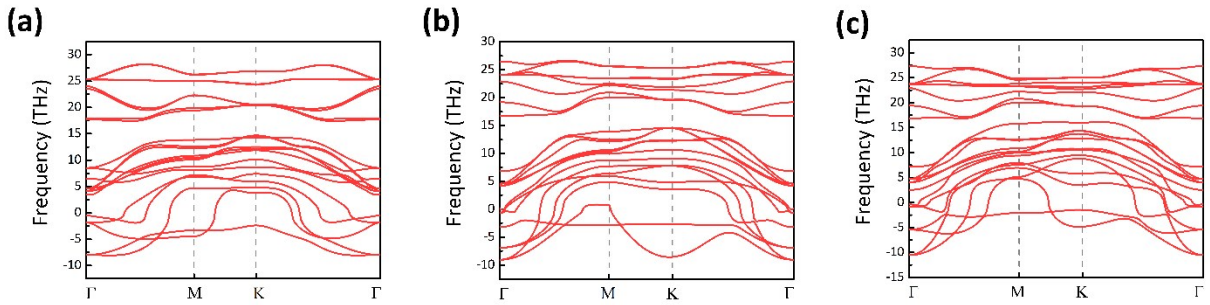

Figure S2. Calculated phonon dispersions for: (a) $CrSi_2C_2N_2$, (b) $MoSi_2C_2N_2$, and (c) $WSi_2C_2N_2$.

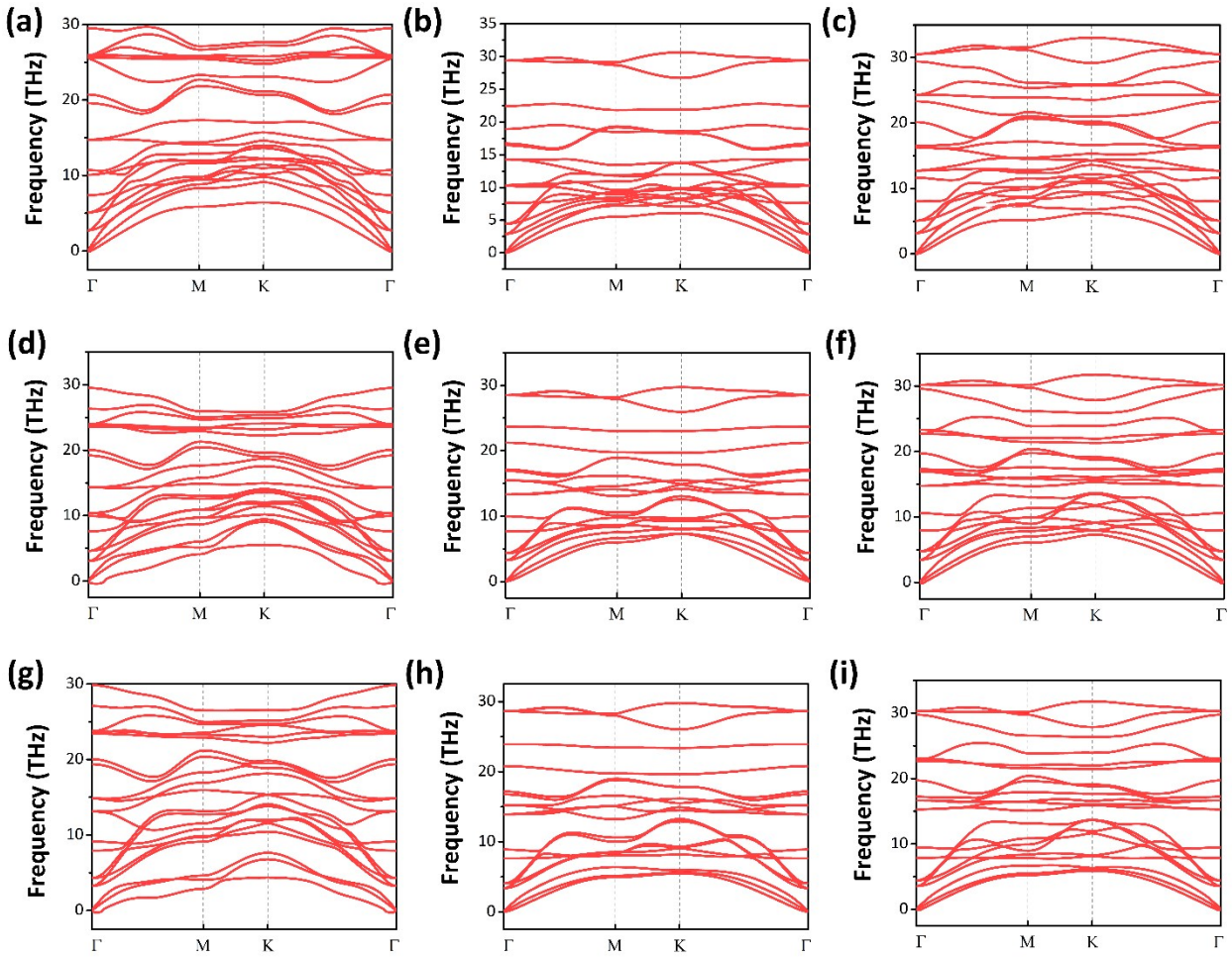

Figure S3. Phonon dispersions for (a) $CrSi_2CN_3$, (b) $CrSi_2N_2C_2$, (c) $CrSi_2N_3C$, (d) $MoSi_2CN_3$, (e) $MoSi_2N_2C_2$, (f) $MoSi_2N_3C$, (g) WSi_2CN_3, (h) $WSi_2N_2C_2$, and (i) WSi_2N_3C.

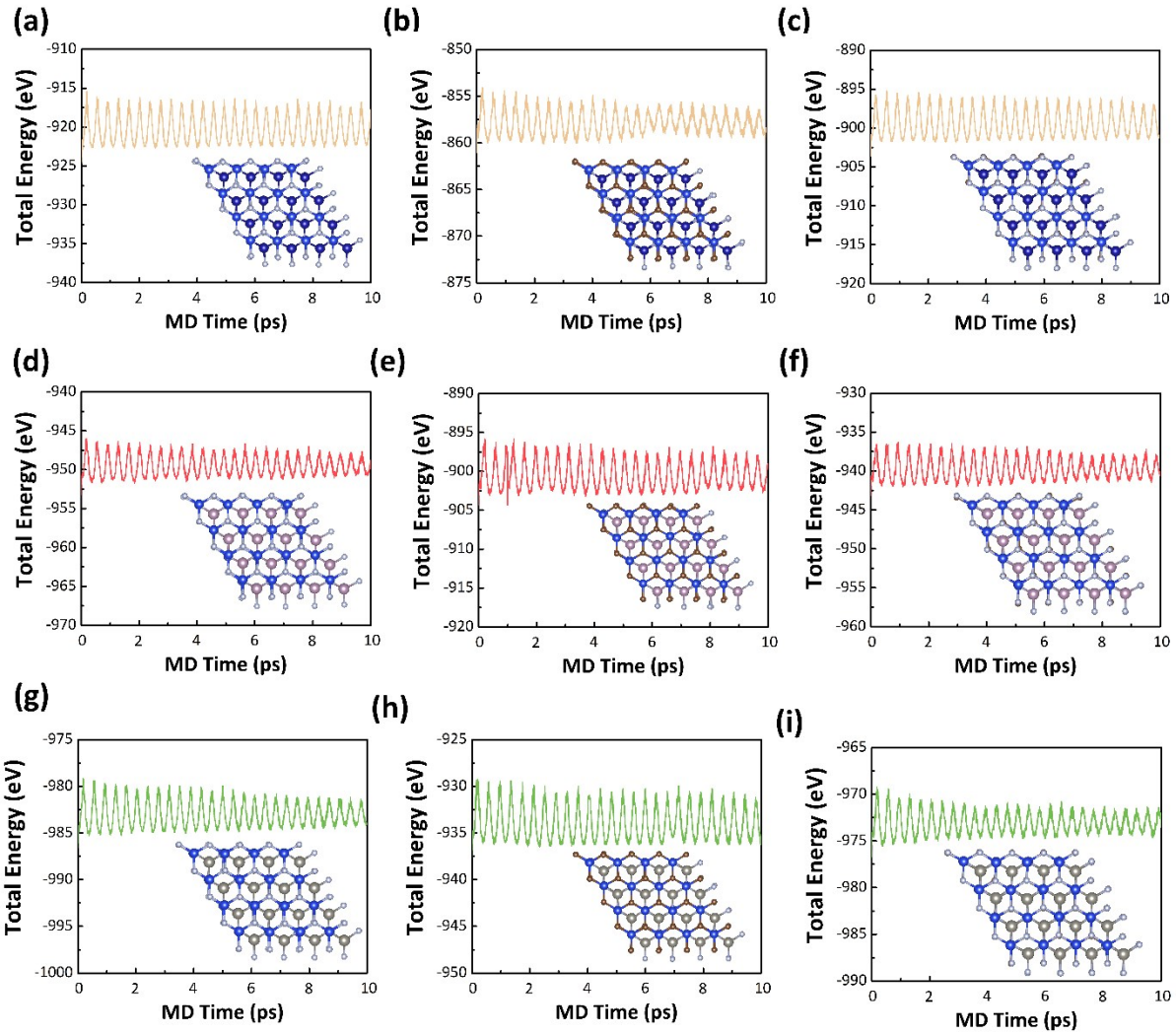

Figure S4. The energy vibrations and final structures after the AIMD test for (a) $CrSi_2CN_3$, (b) $CrSi_2N_2C_2$, (c) $CrSi_2N_3C$, (d) $MoSi_2CN_3$, (e) $MoSi_2N_2C_2$, (f) $MoSi_2N_3C$, (g) WSi_2CN_3, (h) $WSi_2N_2C_2$, and (i) WSi_2N_3C.

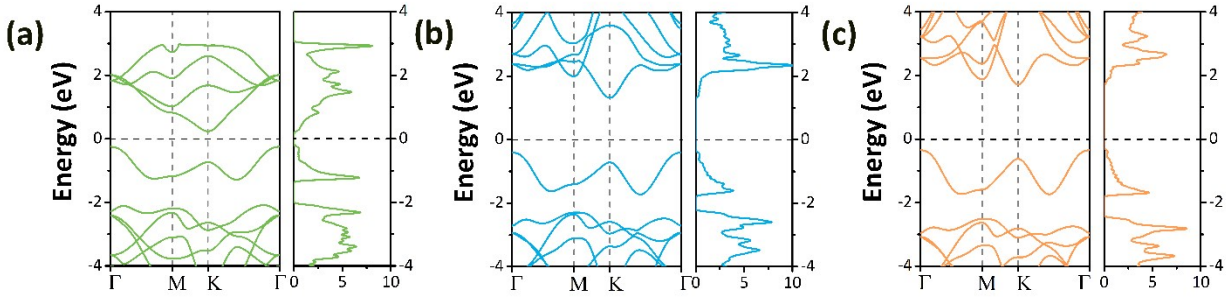

Figure S5. Band structures and TDOSs: (a) $CrSi_2N_4$, (b) $MoSi_2N_4$, and (c) WSi_2N_4.

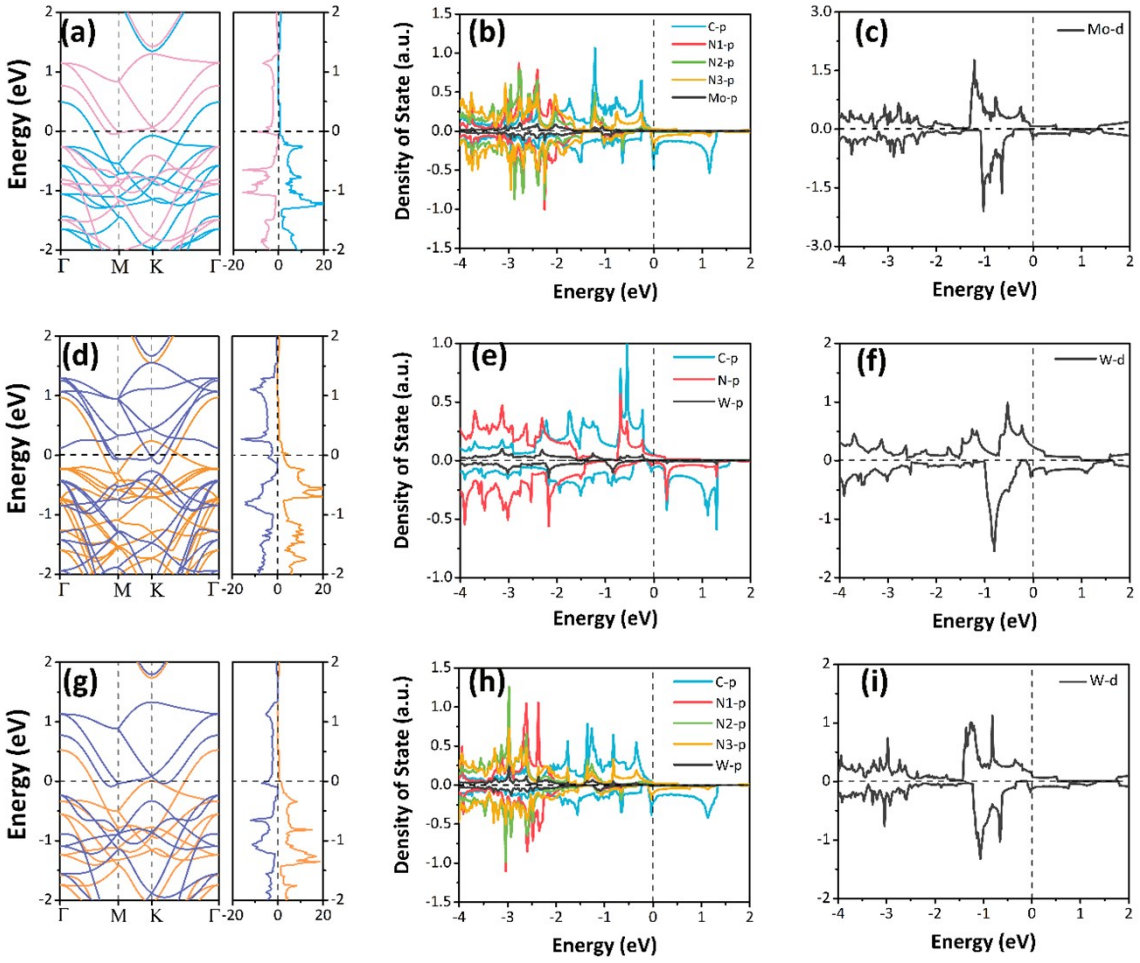

Figure S6. Calculated electronic properties of FM $MoSi_2N_3C$, $WSi_2N_2C_2$, and WSi_2N_3C: (a) band structure and TDOSs of $MoSi_2N_3C$; (b) PDOSs of C, N, and Mo atom's p-electrons in $MoSi_2N_3C$; (c) PDOSs of Mo d-electrons in $MoSi_2N_3C$; (d) band structure and TDOSs of $WSi_2N_2C_2$; (e) PDOSs of C, N, and W atom's p-electrons in $WSi_2N_2C_2$; (f) PDOSs of W d-electrons in $WSi_2N_2C_2$; (g) band structure and TDOSs of WSi_2N_3C; (h) PDOSs of C, N, and W atom's p-electrons in WSi_2N_3C; and (i) PDOSs of W d-electrons in WSi_2N_3C.

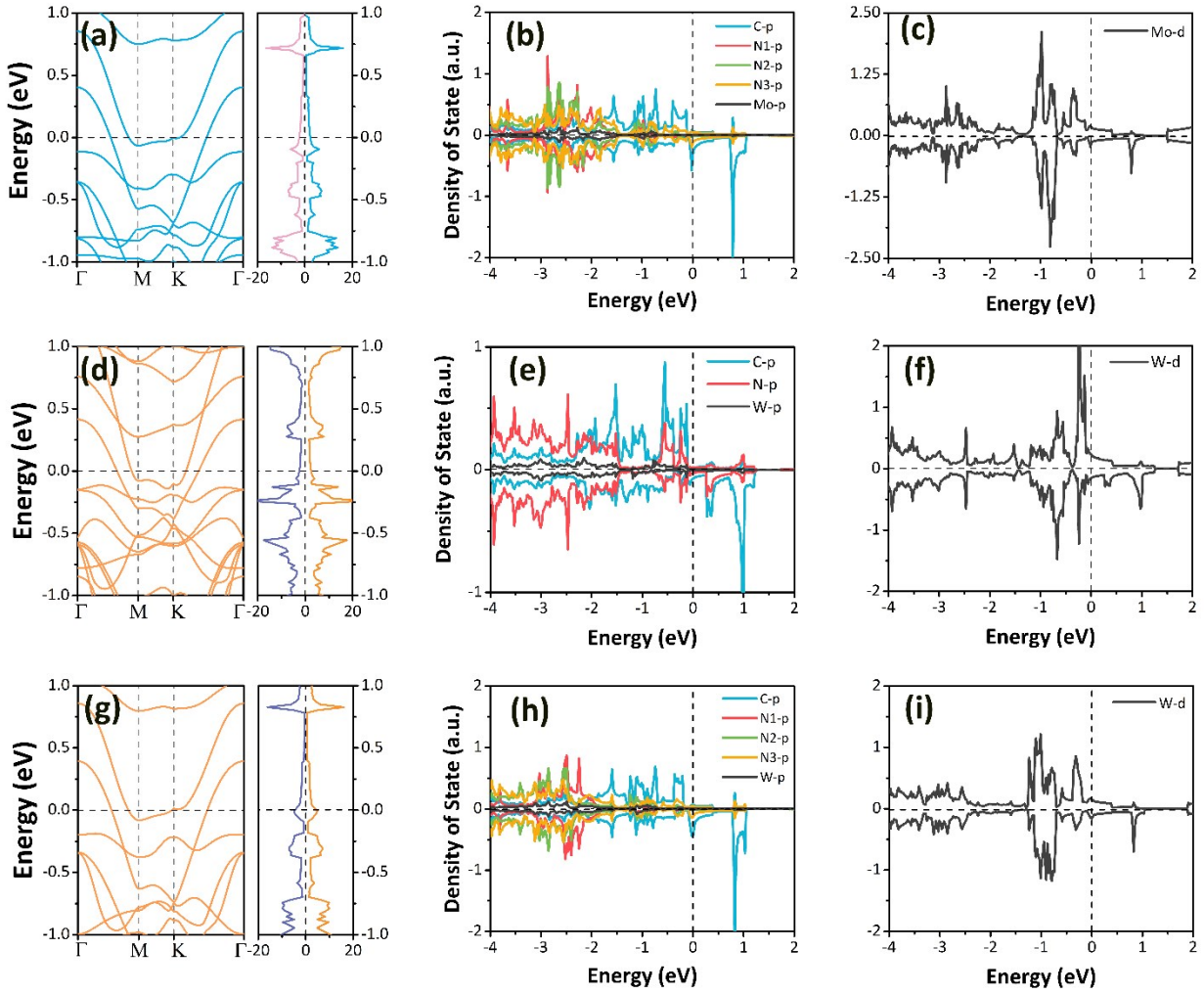

Figure S7. Calculated electronic properties of AFM $MoSi_2N_3C$, $WSi_2N_2C_2$, and WSi_2N_3C: (a) band structure and TDOSs of $MoSi_2N_3C$; (b) PDOSs of C, N, and Mo atom's p-electrons in $MoSi_2N_3C$; (c) PDOSs of Mo d-electrons in $MoSi_2N_3C$; (d) band structure and TDOSs of $WSi_2N_2C_2$; (e) PDOSs of C, N, and W atom's p-electrons in $WSi_2N_2C_2$; (f) PDOSs of W d-electrons in $WSi_2N_2C_2$; (g) band structure and TDOSs of WSi_2N_3C; (h) PDOSs of C, N, and W atom's p-electrons in WSi_2N_3C; and (i) PDOSs of W d-electrons in WSi_2N_3C.

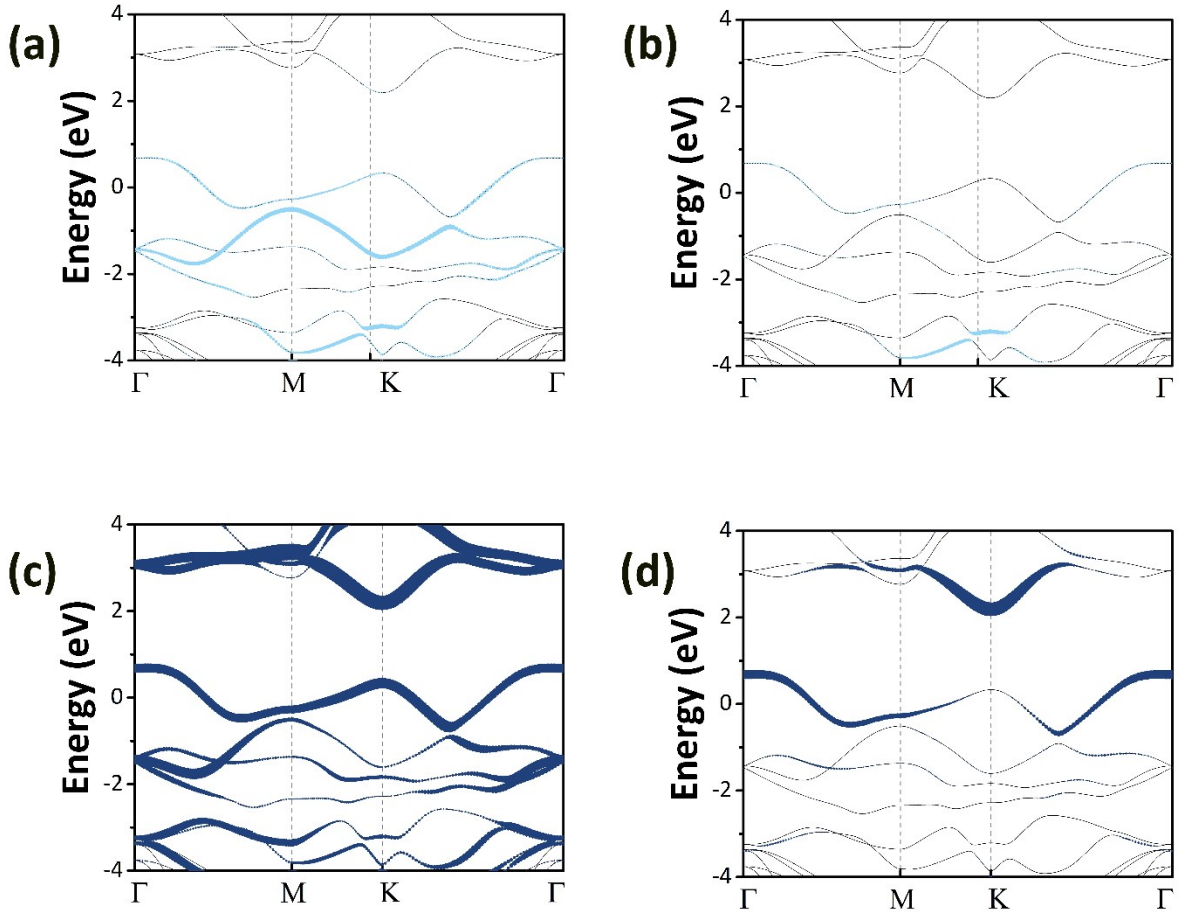

Figure S8. Contributions to PBANDs for $MoSi_2CN_3$: (a) total electrons of C atoms, (b) pz-orbital electrons of C atoms, (c) total electrons of Mo atoms, and (d) dz^2-orbital electrons of Mo atoms.

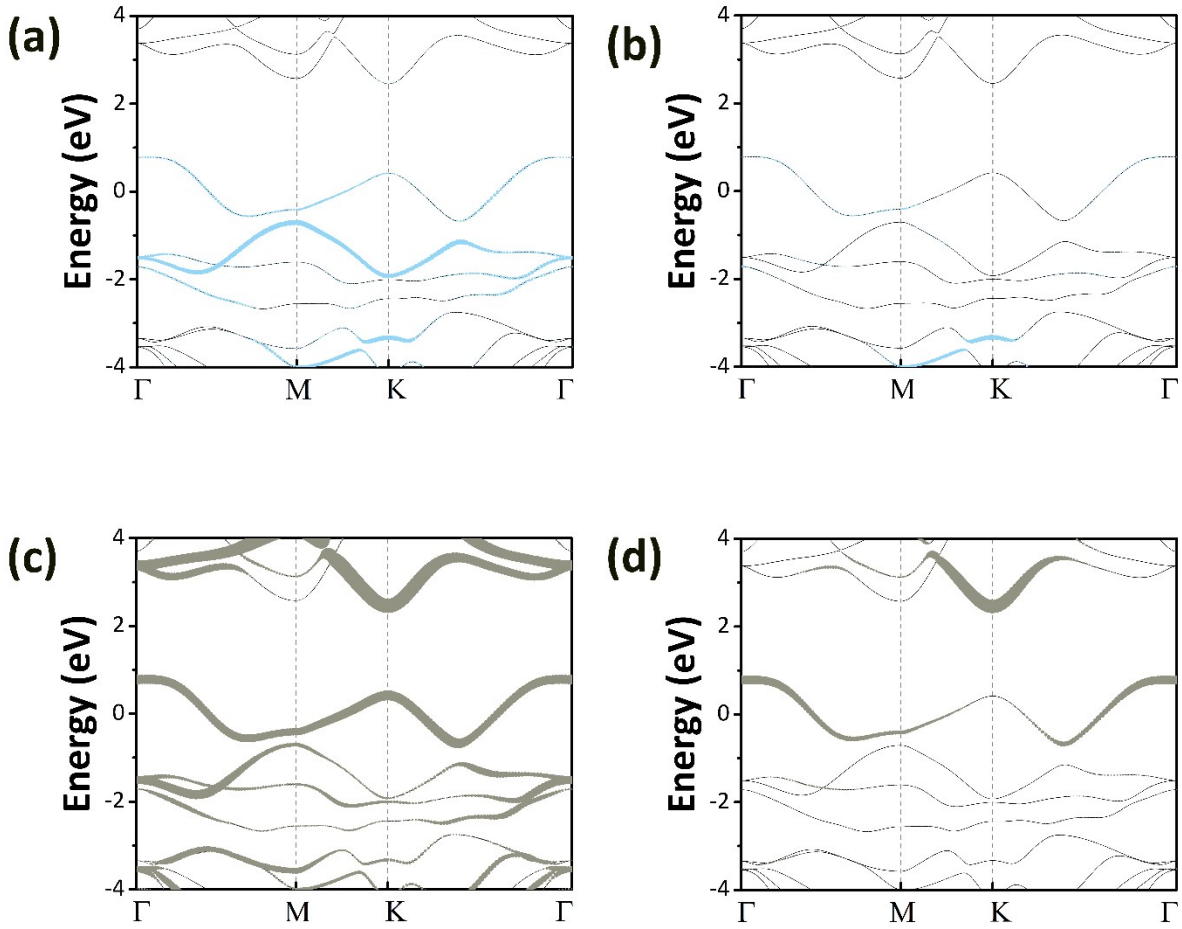

Figure S9. Contributions to PBANDs for WSi_2CN_3: (a) total electrons of C atoms, (b) pz-orbital electrons of C atoms, (c) total electrons of W atoms, and (d) dz^2-orbital electrons of W atoms.

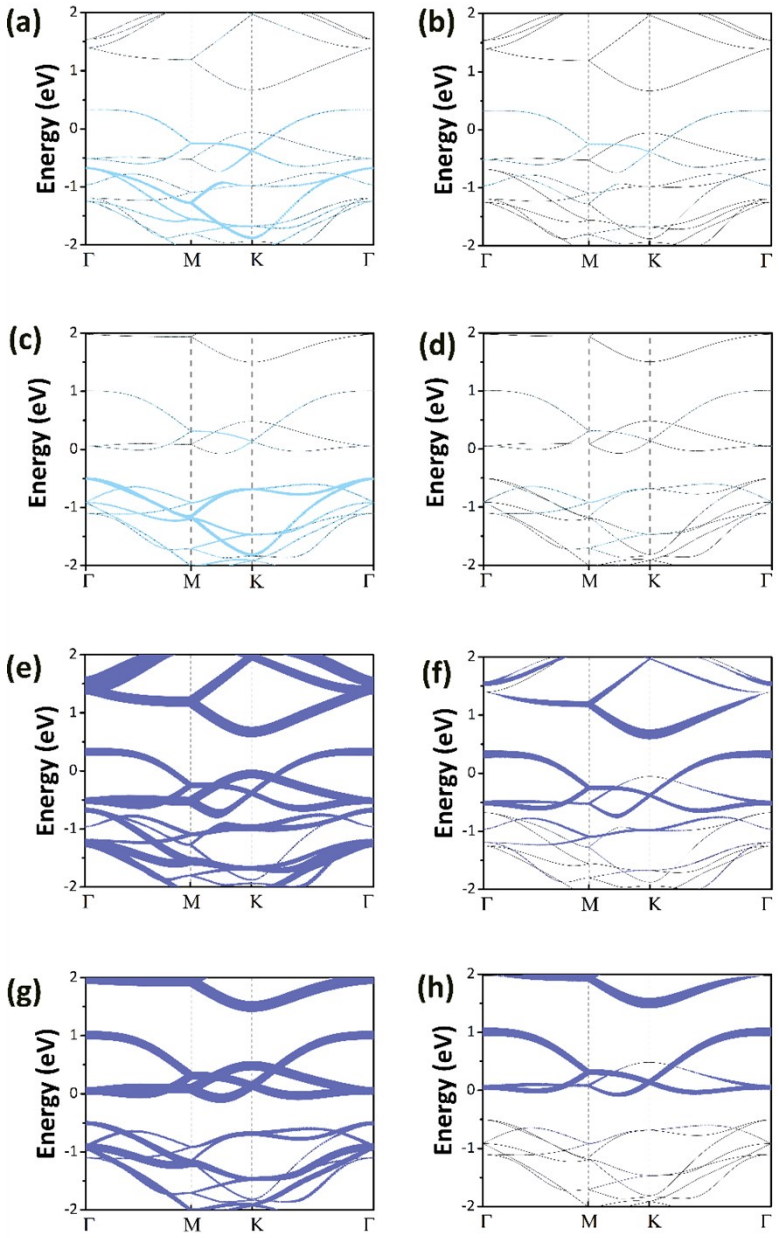

Figure S10. Contributions to PBANDs for FM $CrSi_2CN_3$: (a) total spin-up electrons of C atoms, (b) pz-orbital spin-up electrons of C atoms, (c) total spin-down electrons of C atoms, (d) pz-orbital spin-down electrons of C atoms, (e) total spin-up electrons of Cr atoms, (f) dz^2-orbital spin-up electrons of Cr atoms, (g) total spin-down electrons of Cr atoms, and (h) dz^2-orbital spin-down electrons of Cr atoms.

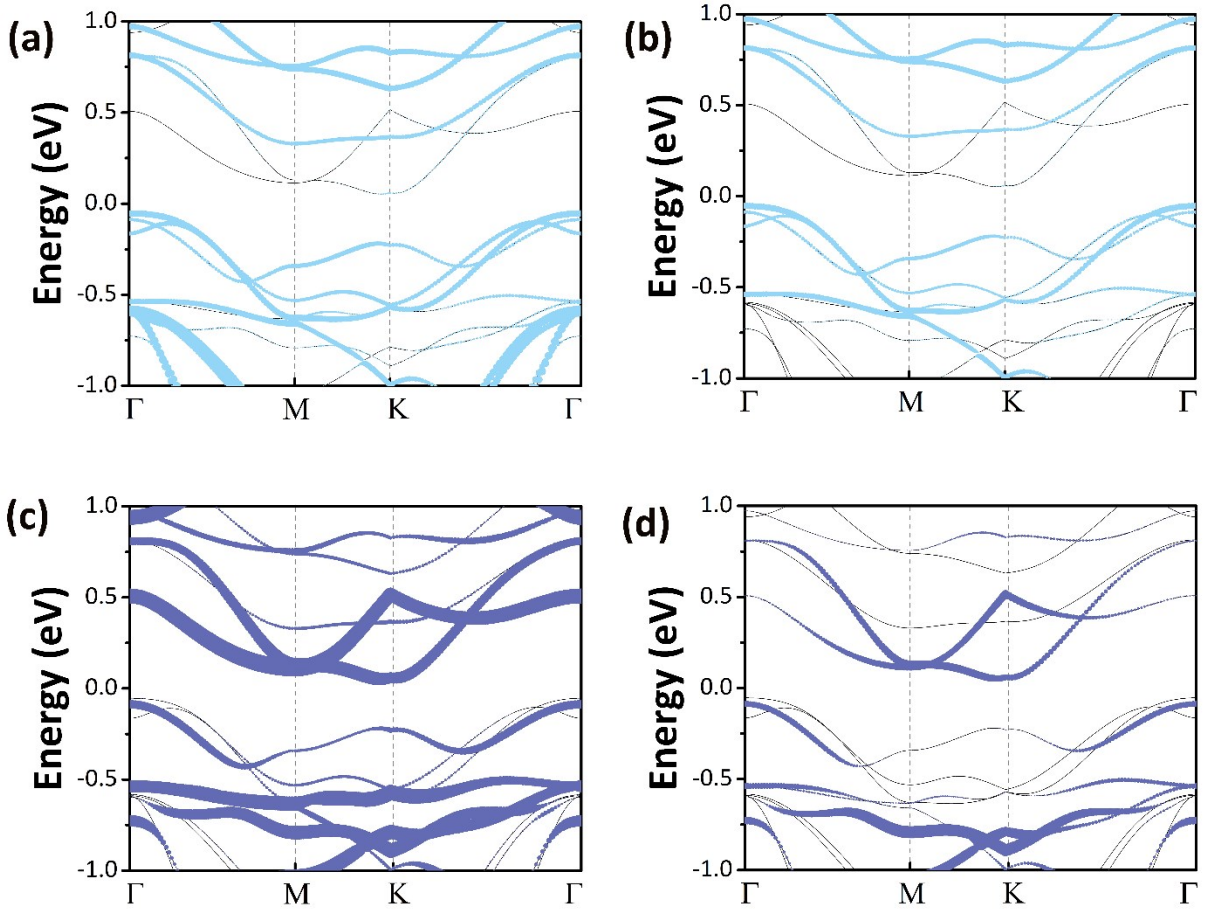

Figure S11. Contributions to PBANDs for AFM $CrSi_2N_2C_2$: (a) total electrons of C atoms, (b) pz-orbital electrons of C atoms, (c) total electrons of Cr atoms, and (d) dz^2-orbital electrons of Cr atoms.

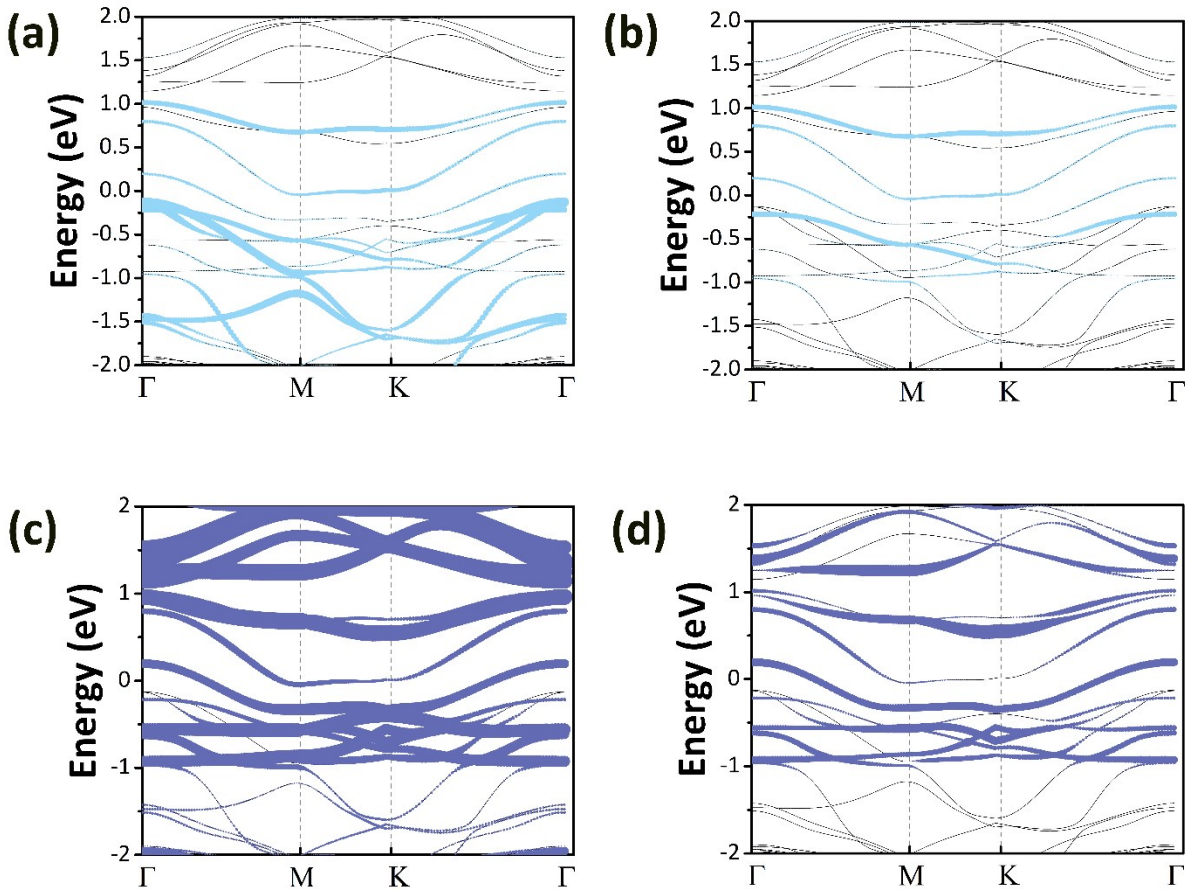

Figure S12. Contributions to PBANDs for AFM $CrSi_2N_3C$: (a) total electrons of C atoms, (b) pz-orbital electrons of C atoms, (c) total electrons of Cr atoms, and (d) dz^2-orbital electrons of Cr atoms.

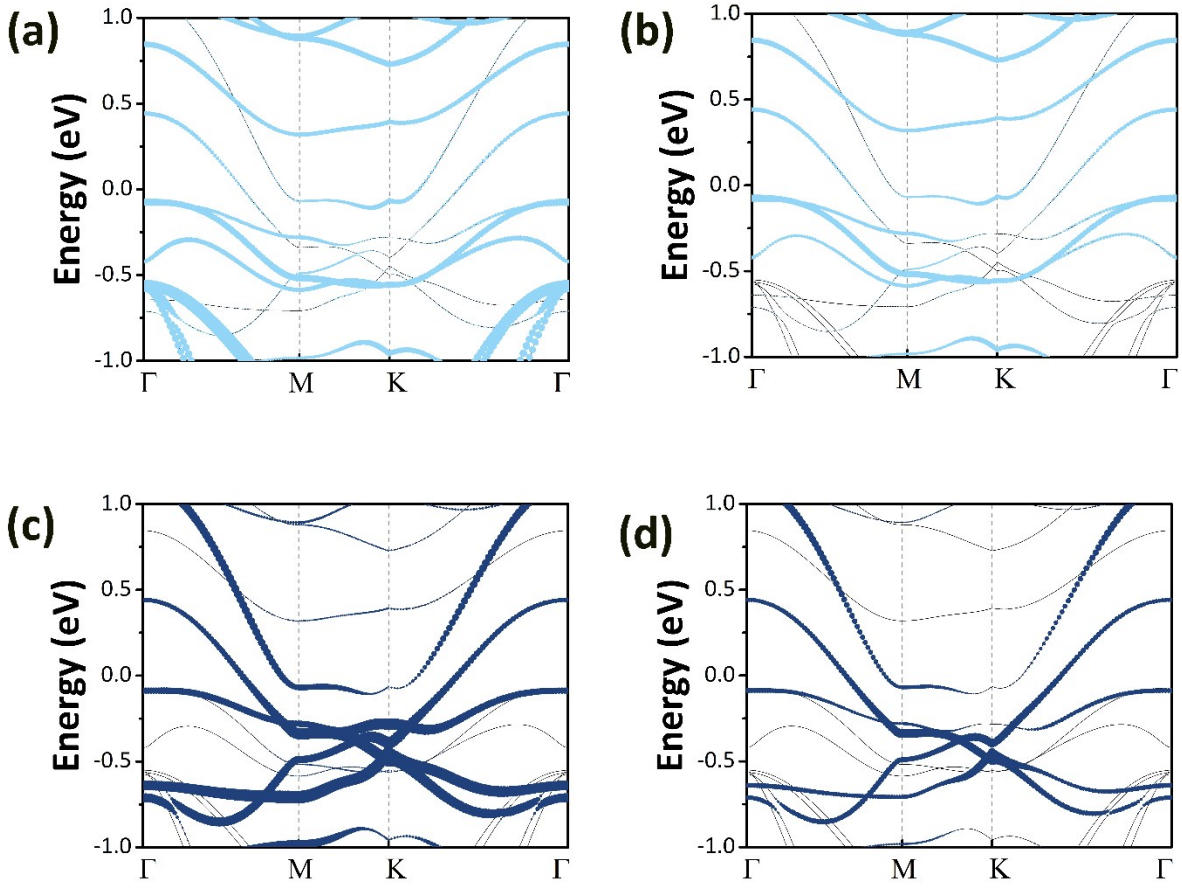

Figure S13. Contributions to PBANDs for AFM $MoSi_2N_2C_2$: (a) total electrons of C atoms, (b) pz-orbital electrons of C atoms, (c) total electrons of Mo atoms, and (d) dz^2-orbital electrons of Mo atoms.

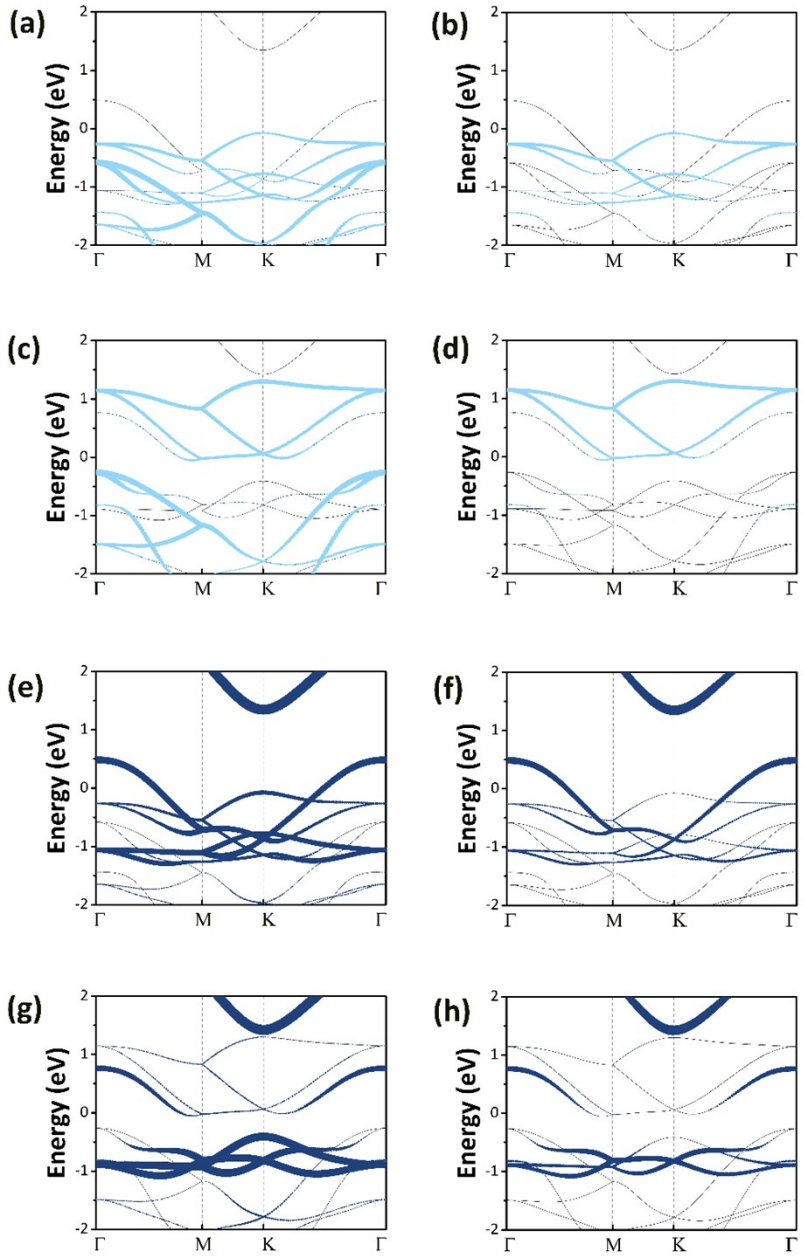

Figure S14. Contributions to PBANDs for FM $MoSi_2N_3C$: (a) total spin-up electrons of C atoms, (b) pz-orbital spin-up electrons of C atoms, (c) total spin-down electrons of C atoms, (d) pz-orbital spin-down electrons of C atoms, (e) total spin-up electrons of Mo atoms, (f) dz^2-orbital spin-up electrons of Mo atoms, (g) total spin-down electrons of Mo atoms, and (h) dz^2-orbital spin-down electrons of Mo atoms.

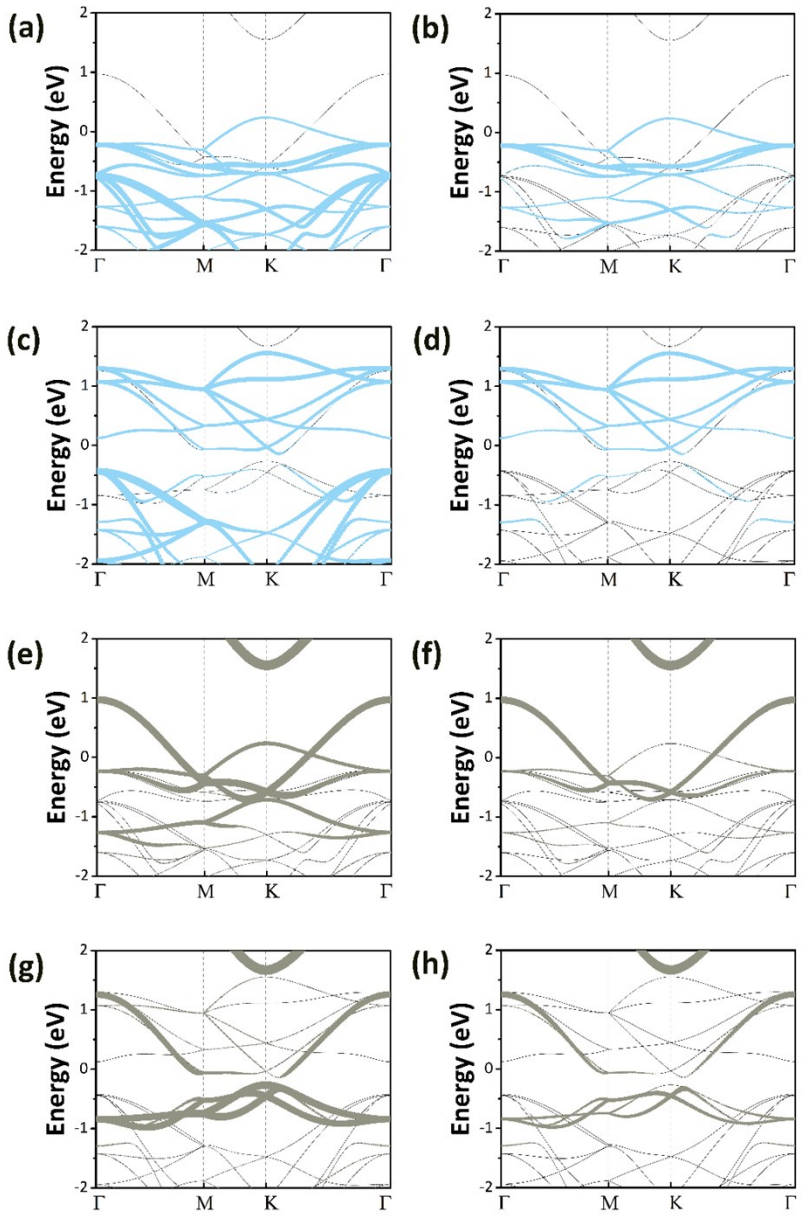

Figure S15. Contributions to PBANDs for FM $WSi_2N_2C_2$: (a) total spin-up electrons of C atoms, (b) pz-orbital orbital spin-up electrons of C atoms, (c) total spin-down electrons of C atoms, (d) pz-orbital spin-down electrons of C atoms (e) total spin-up electrons of W atoms, (f) dz^2-orbital spin-up electrons of W atoms, (g) total spin-down electrons of W atoms, and (h) dz^2-orbital spin-down electrons of W atoms.

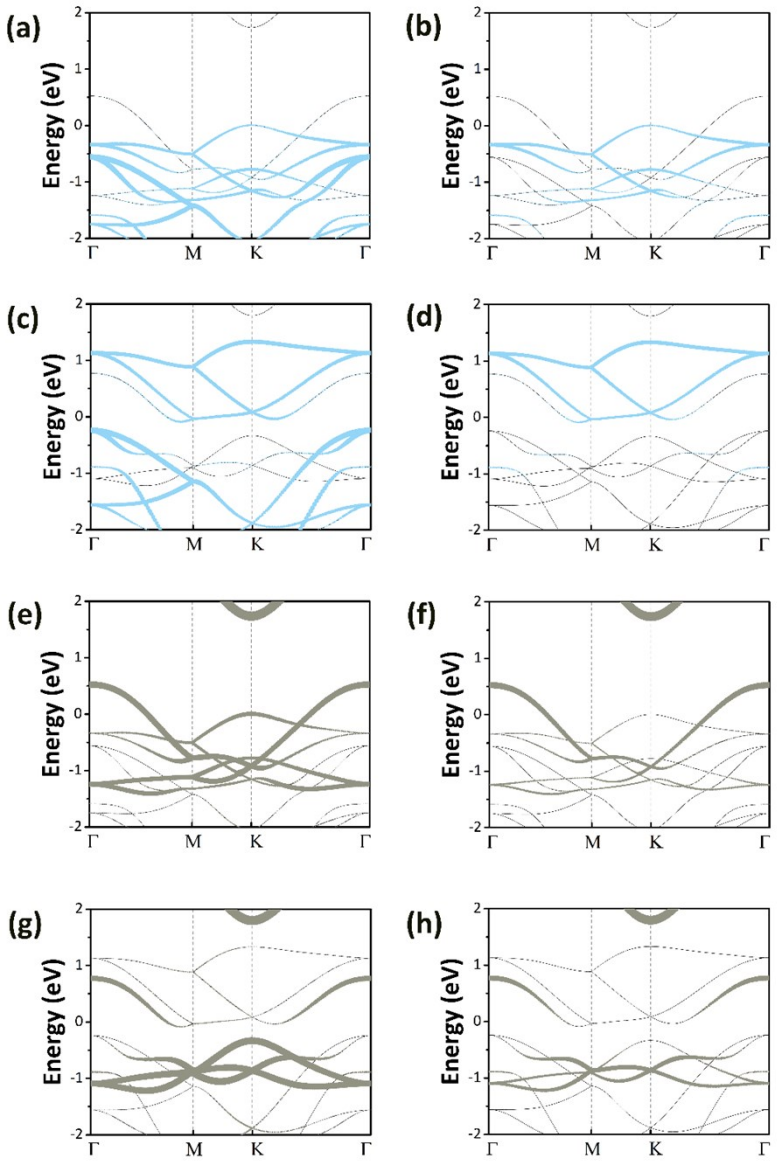

Figure S16. Contributions to PBANDs for FM WSi_2N_3C : (a) total spin-up electrons of C atoms, (b) pz-orbital spin-up electrons of C atoms, (c) total spin-down electrons of C atoms, (d) pz-orbital spin-down electrons of C atoms, (e) total spin-up electrons of W atoms, (f) dz^2-orbital spin-up electrons of W atoms, (g) total spin-down electrons of W atoms, and (h) dz^2-orbital spin-down electrons of W atoms.

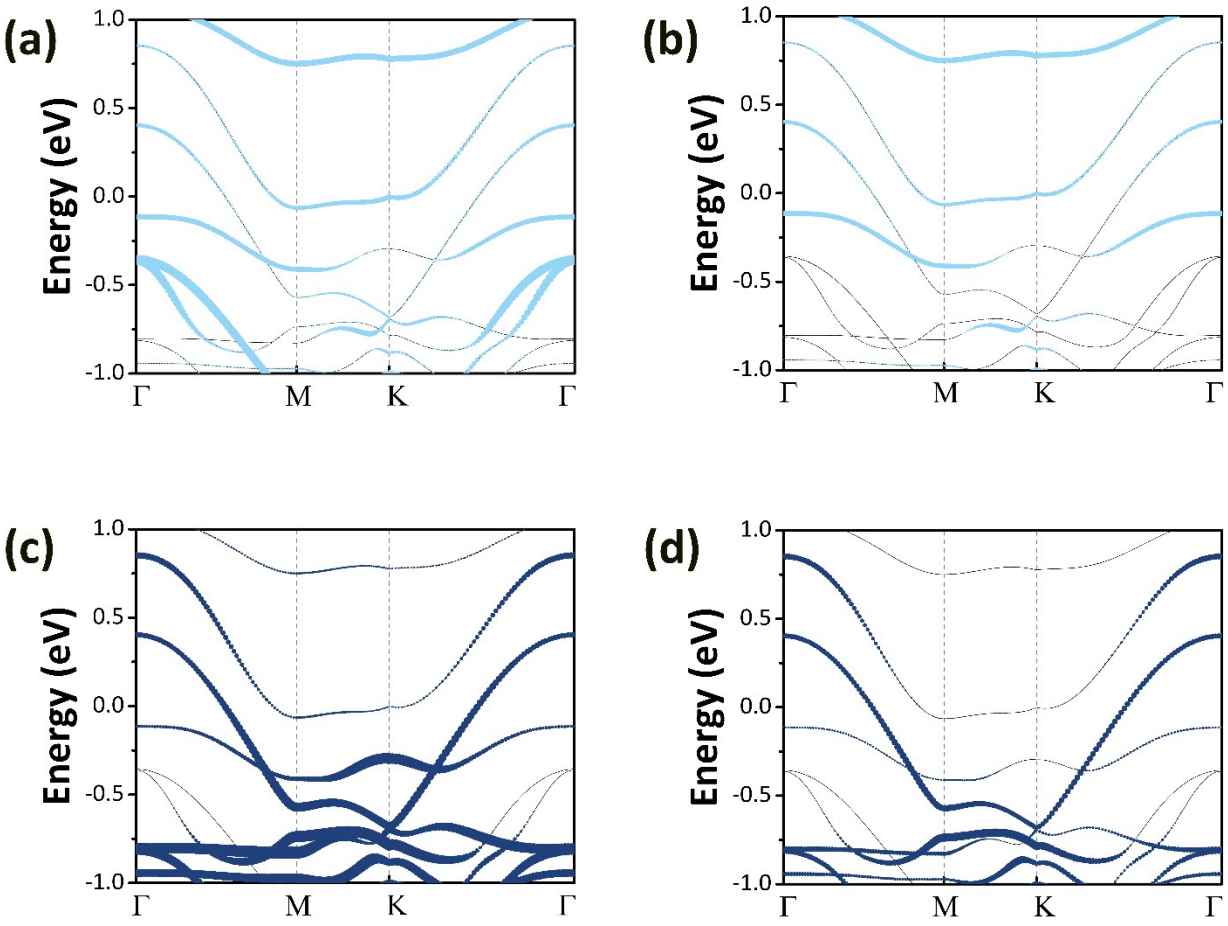

Figure S17. Contributions to PBANDs for AFM $MoSi_2N_3C$: (a) total electrons of C atoms, (b) pz-orbital electrons of C atoms, (c) total electrons of Mo atoms, and (d) dz^2-orbital electrons of Mo atoms.

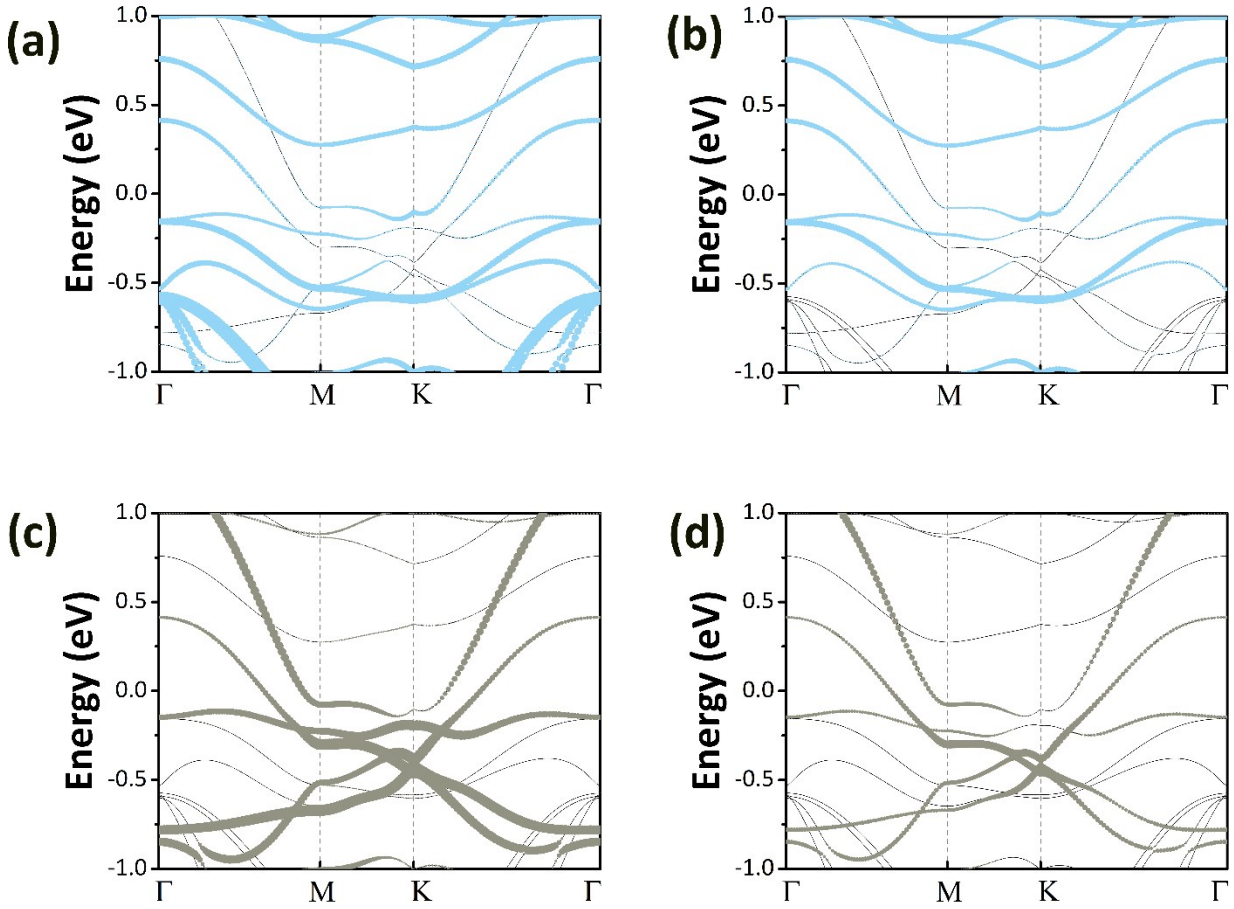

Figure S18. Contributions to PBANDs for AFM $WSi_2N_2C_2$: (a) total electrons of C atoms, (b) pz-orbital electrons of C atoms, (c) total electrons of W atoms, and (d) dz^2-orbital electrons of W atoms.

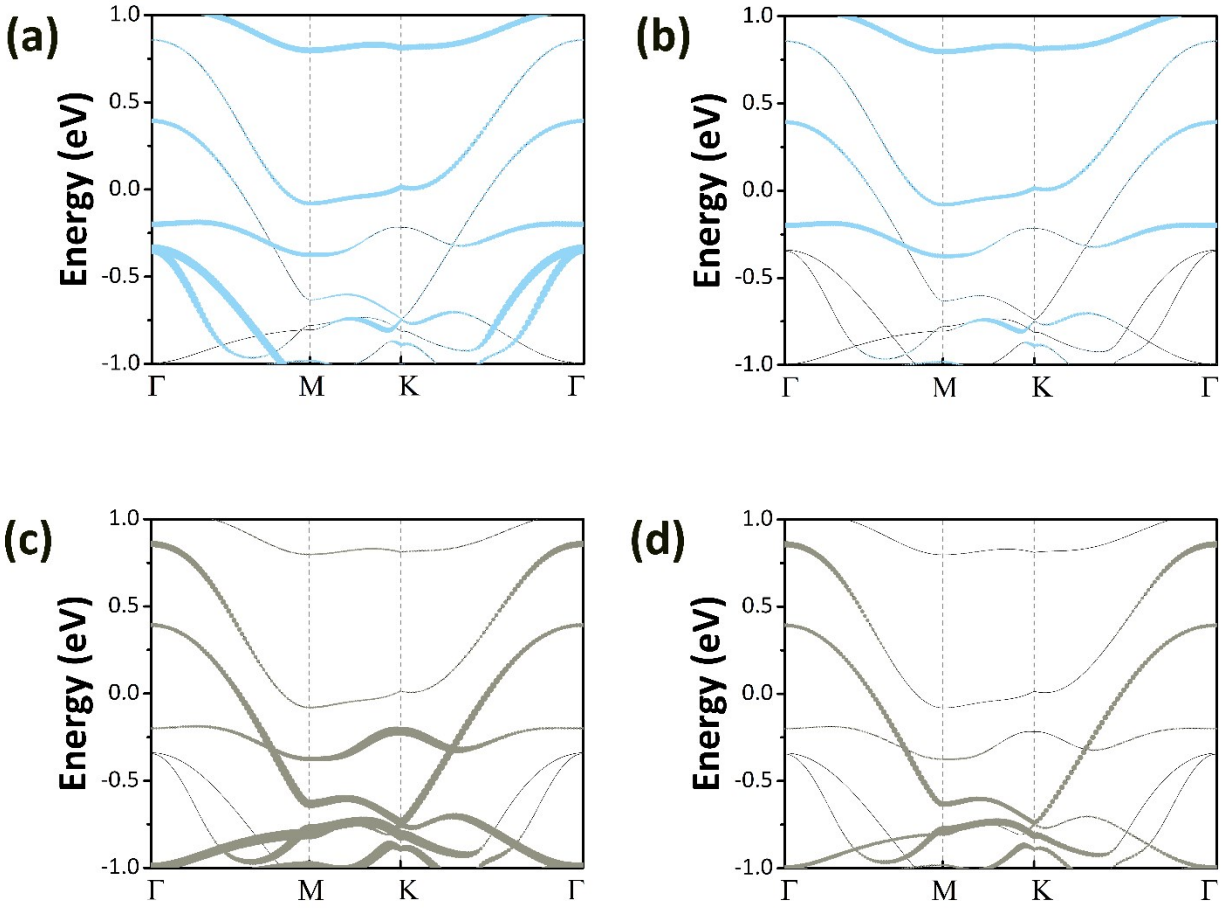

Figure S19. Contributions to PBANDs for AFM WSi_2N_3C : (a) total electrons of C atoms, (b) pz-orbital electrons of C atoms, (c) total electrons of W atoms, and (d) dz^2-orbital electrons of W atoms.

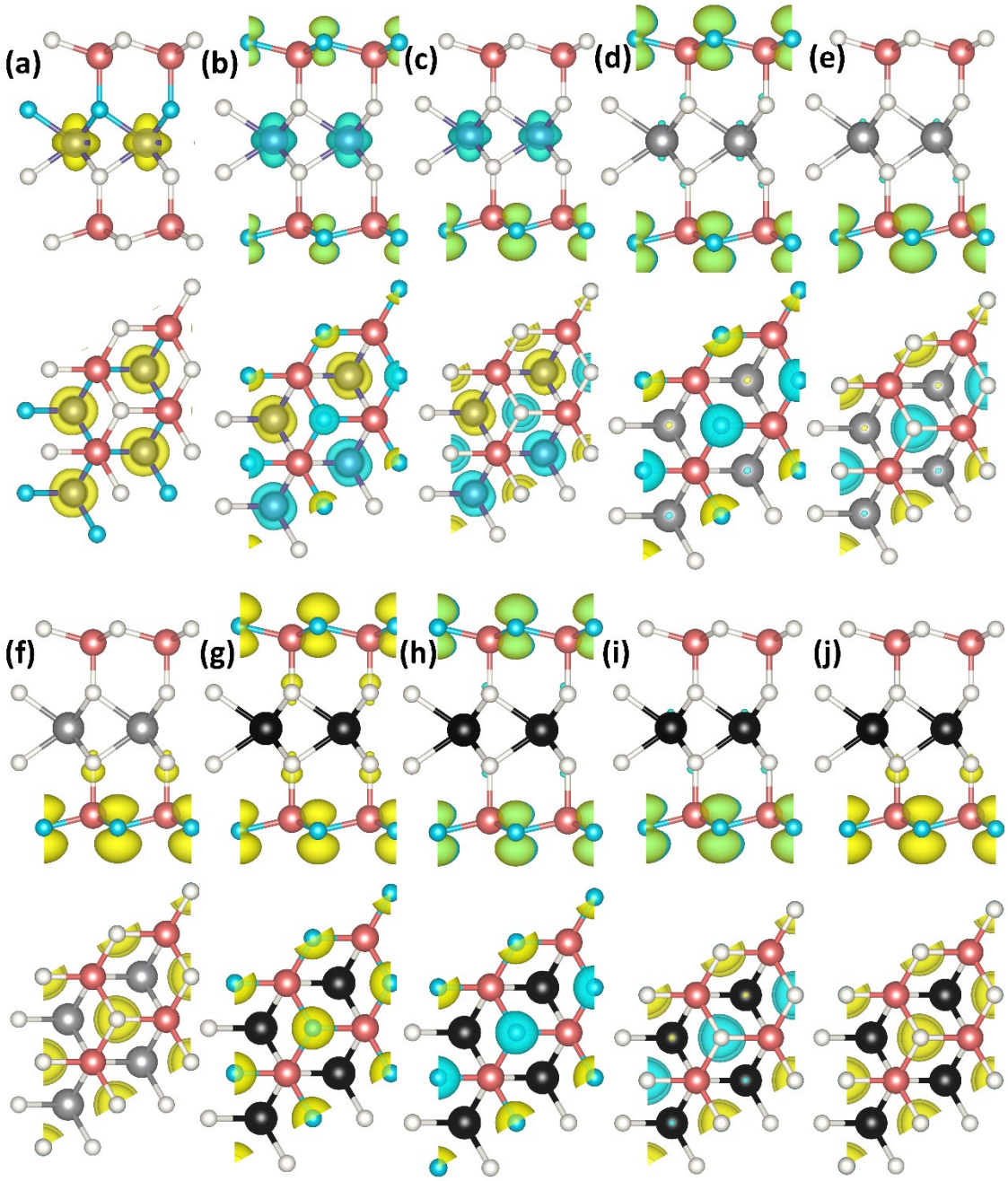

Figure S20. Calculated spin-density for: (a) FM $CrSi_2CN_3$, (b) AFM $CrSi_2N_2C_2$, (c) AFM $CrSi_2N_3C$, (d) AFM $MoSi_2N_2C_2$, (e) AFM $MoSi_2N_3C$, (f) FM $MoSi_2N_3C$, (g) FM $WSi_2N_2C_2$, (h) AFM $WSi_2N_2C_2$, (i) AFM WSi_2N_3C, and (j) FM WSi_2N_3C. Yellow and azure indicate spin-up state and spin-down states, respectively.

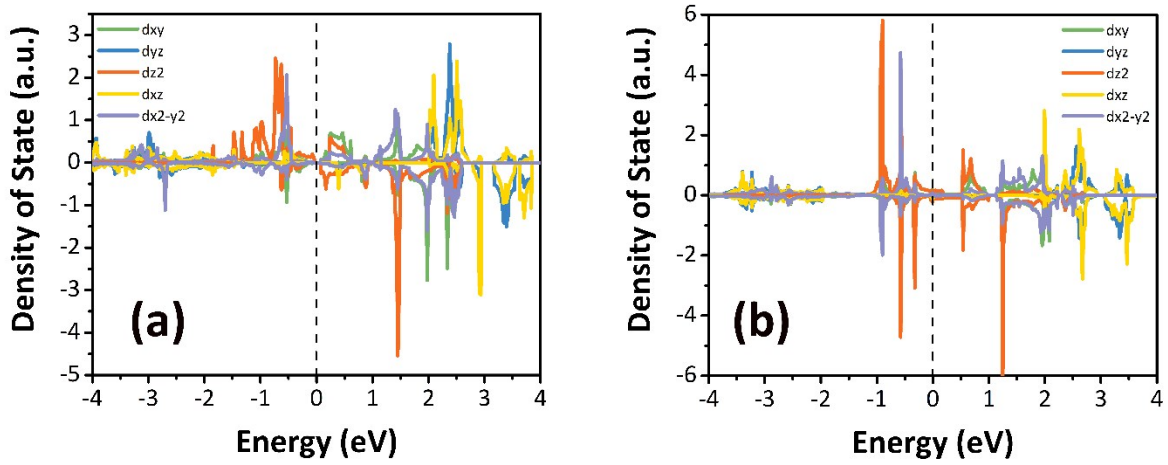

Figure S21. PDOSs of Cr-d electrons in (a) $CrSi_2N_2C_2$ and (b) $CrSi_2N_3C$.

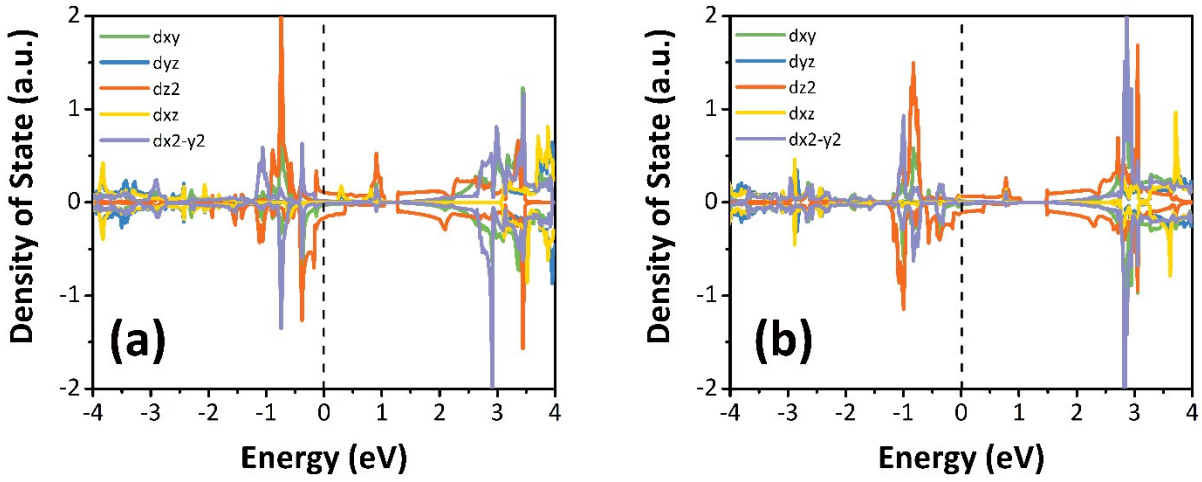

Figure S22. PDOS of Mo-d electrons in (a) $MoSi_2N_2C_2$ and (b) $MoSi_2N_3C$.