Supporting Information

Metallic CrP_2 Monolayer: Potential Applications in Energy Storage and Conversion

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Computational Details

The local structural relaxations and electronic properties calculations were performed in the framework of the density functional theory (DFT) within the generalized gradient approximation (GGA) as implemented in the VASP code.[1] The $3d^5 4s^1$ and $3s^2 3p^3$ atomic orbitals were treated as valence states for Cr and P, respectively. The cut-off energy for the expansion of wave functions into plane waves was set to 500 eV in the calculations.

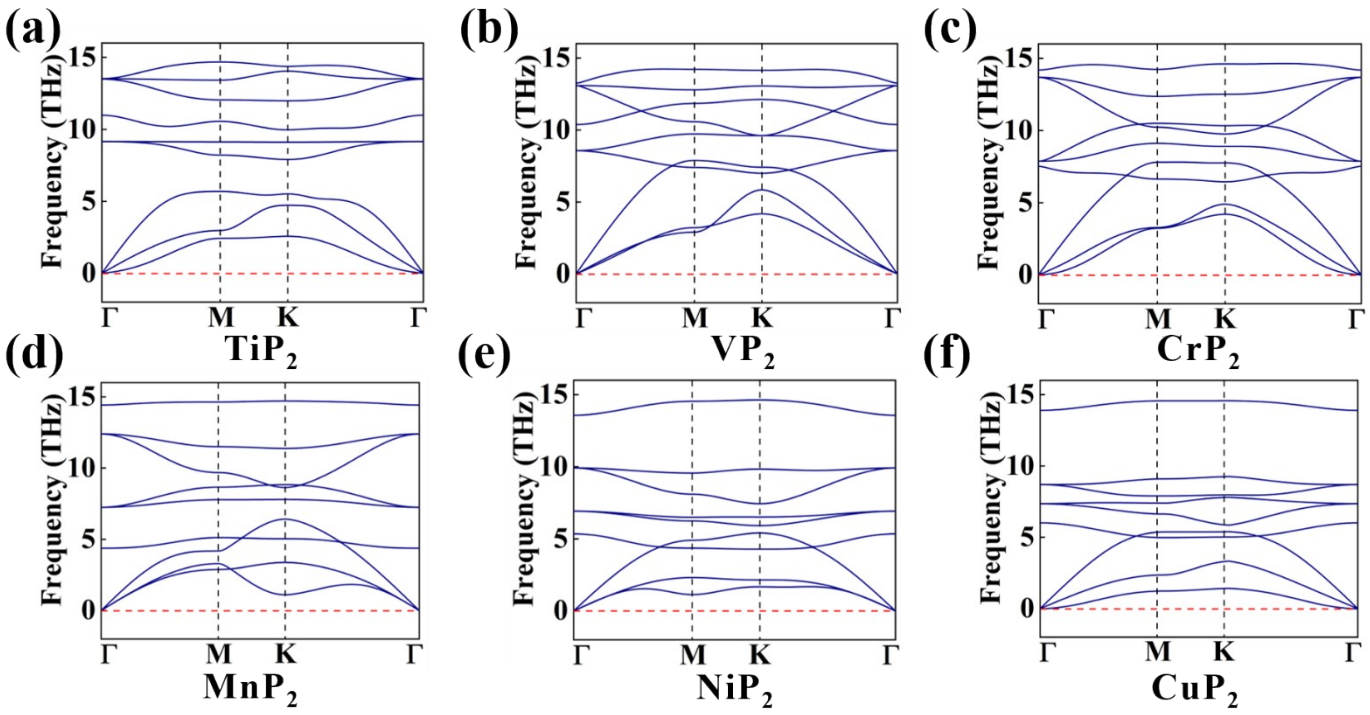

Figure S1. The calculated phonon dispersion curves of TiP_2, VP_2, CrP_2, MnP_2, NiP_2, and CuP_2 monolayers.

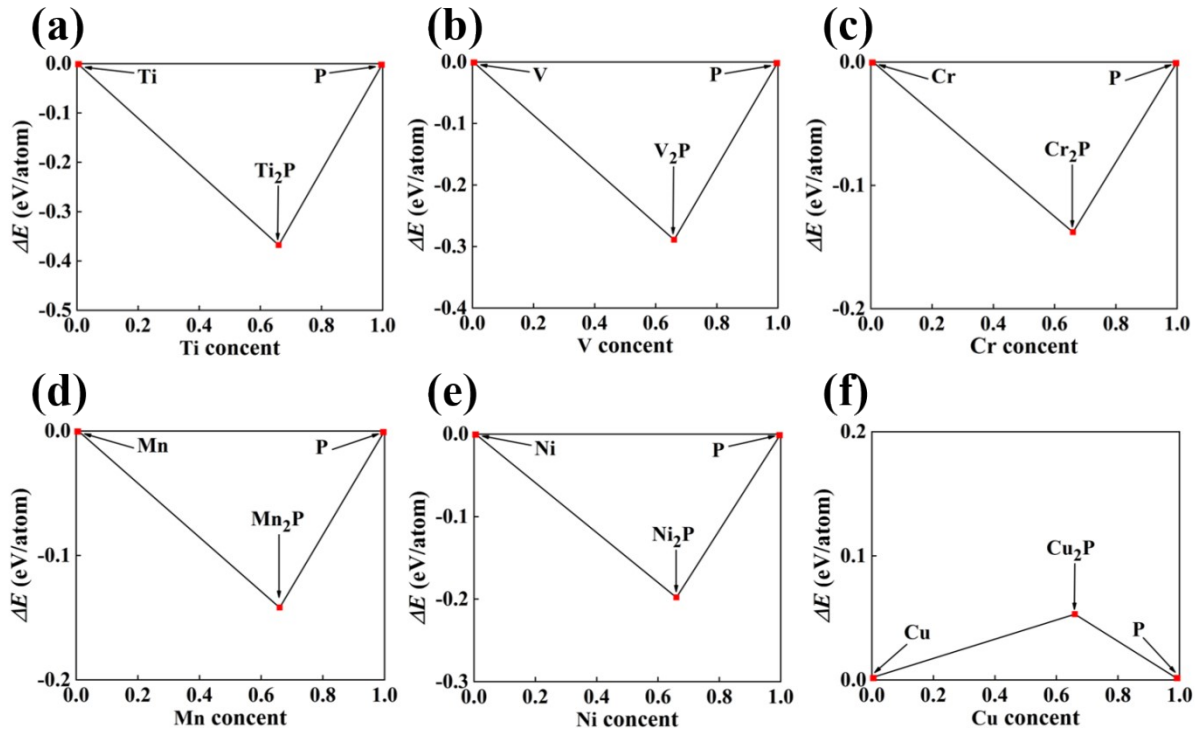

Figure S2. The formation energies of TMPs monolayers with respect to the transition metal solids and black phosphorus (*Cmca*).[2] The transition metal solids and their space groups are Ti ($P6_3/mmc$),[3] V (*Im*-3*m*),[4] Cr (*Im*-3*m*),[5] Mn (*I*-43*m*),[6] Ni ($P6_3/mmc$)[7] and Cu(*Fm*-3*m*).[8]

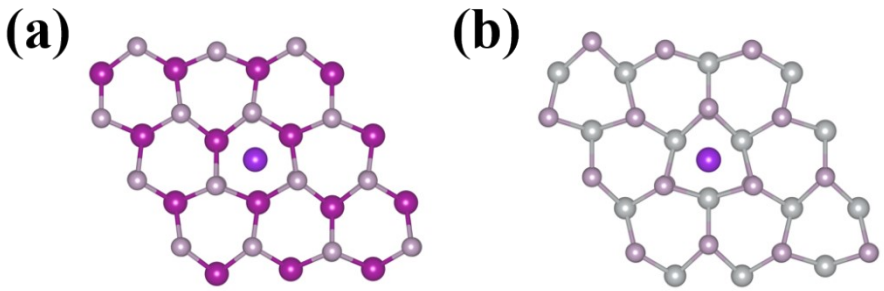

Figure S3. The structures of the MnP_2 (a) and NiP_2 (b) monolayers after K adsorption.

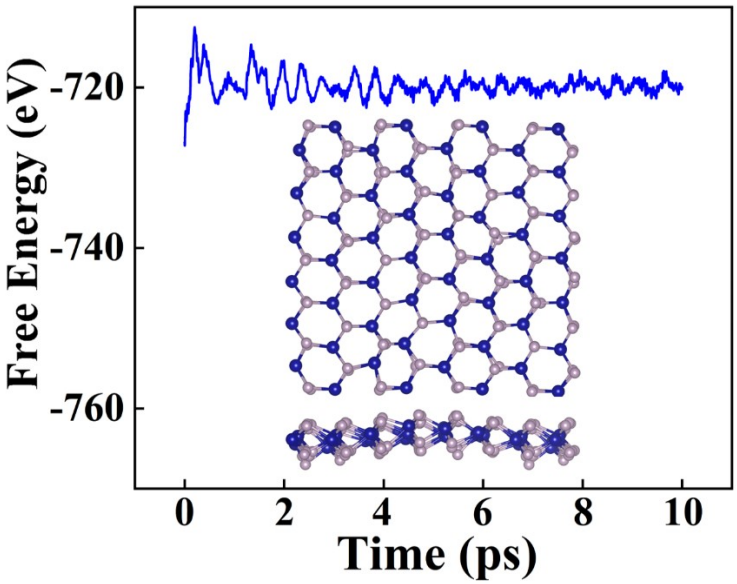

Figure S4. First-principles molecular dynamics (MD) simulations were performed at a temperature of 500 K with the free energy variation and final structure of the CrP_2 monolayer within a duration of 10 ps.

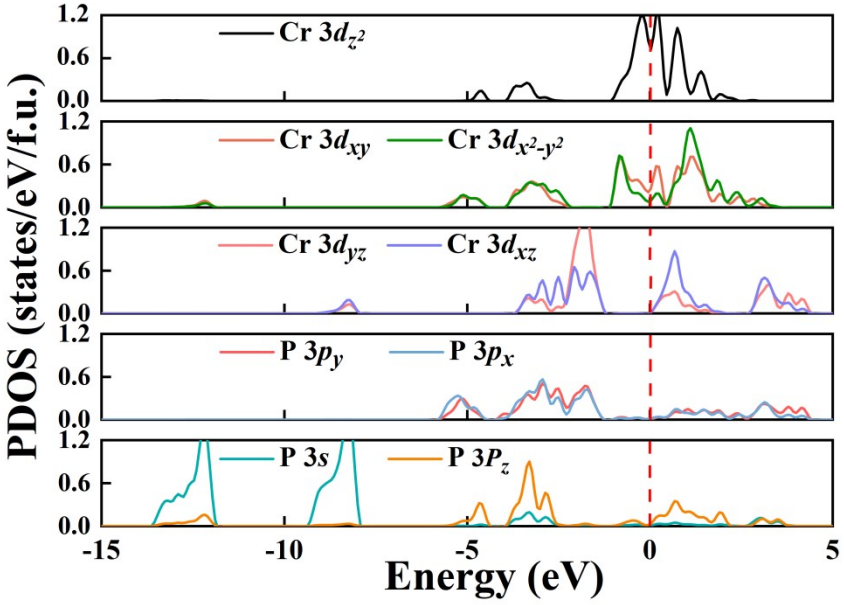

Figure S5. Projected density of states (PDOS) of the CrP_2 monolayer.

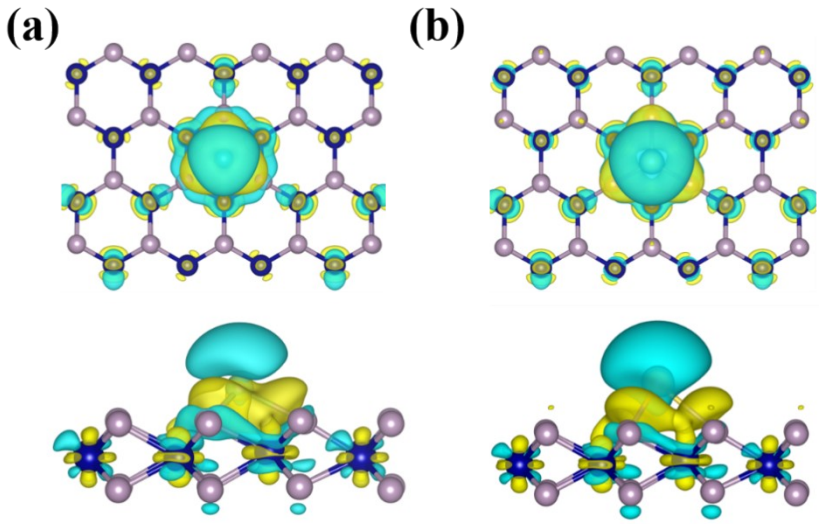

Figure S6. The top and side views of charge density difference (CDD) when a Li (a) and Na (b) are adsorbed on the CrP_2 monolayer.

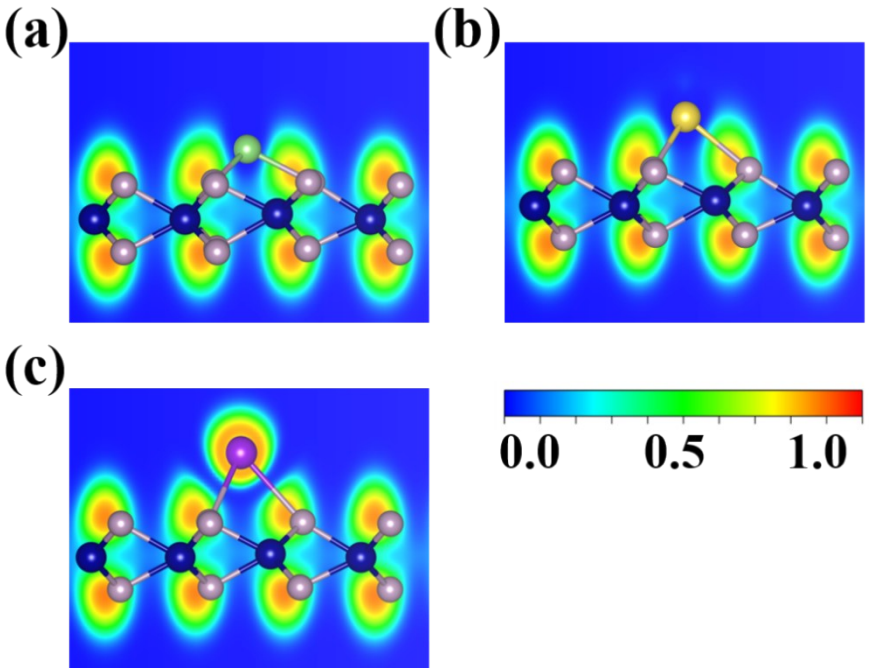

Figure S7. The electron localization function (ELF) maps when a Li (a), Na (b) and K(c) are adsorbed on the CrP_2 monolayer.

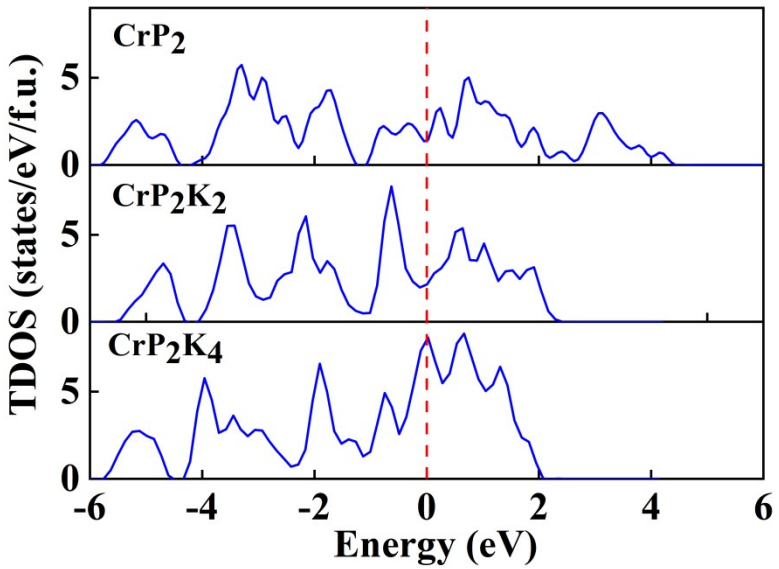

Figure S8. Total density of states (TDOS) of CrP_2K_2 and CrP_2K_4.

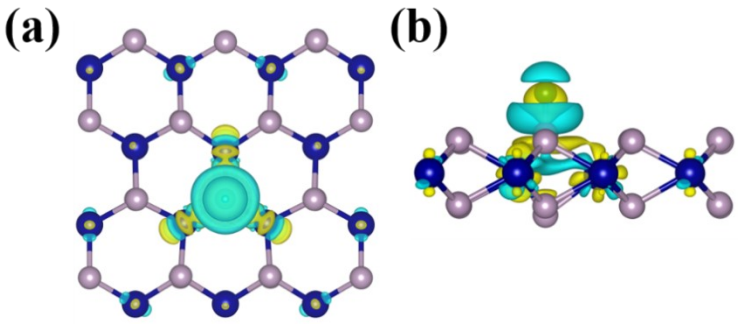

Figure S9. The top (a) and side (b) views of charge density difference (CDD) when H adsorbed on the CrP_2 monolayer.

Table S1. Structural information of the CrP_2 monolayer.

Structure	Space Group	Lattice Parameters (Å, °)	Atom positions			
				x	*y*	*z*
CrP_2	*P*-6*m*2	$a = b = 3.4764$	Cr	0.33333	0.66667	0.50000
		$c = 20.0000$	P	0.00000	0.00000	0.42707
		$\alpha = \beta = 90.000$	P	0.00000	0.00000	0.57293
		$\gamma = 120.000$				

Table S2. The adsorption energies (eV) of Li, Na, and K atoms on the TiP_2, VP_2 and CrP_2 monolayers.

Structures	Li	Na	K
TiP_2	-0.909	-0.626	-0.919
VP_2	-0.644	-0.390	-0.731
CrP_2	-1.105	-0.878	-1.204

Table S3. Elastic constants of the CrP_2 monolayers (in N/m).

Structure	C_{11}	C_{12}	C_{22}	C_{66}
CrP_2	76.96	43.18	76.96	16.89

Based on the Born-Huang criterion: $C_{11}C_{22} - C_{12}C_{12} > 0$ and $C_{66} > 0$, the CrP_2 monolayer is mechanically stable.[9]

Table S4. Relative energies among different magnetic ground states of the CrP_2 monolayer.

Magnetic ground states	AFM	FM	FIM
Relative energies (eV)	0	0.063	0.234

Table S5. The distances between the metal atoms (Li, Na, and K) and the CrP_2 monolayer, and atomic radius of the three considered metal atoms.

Metal atoms	Distance (Å)	Atomic radius (pm)
Li	1.18	152
Na	1.75	186
K	2.32	232

Table S6. The Mulliken charge and Lowdin charge of single atom (Li, Na, and K) adsorbed on the CrP_2 monolayer.

Metal atoms	Mulliken charge (e^-)	Lowdin charge (e^-)
Li	1.00	0.75
Na	0.87	0.70
K	0.98	0.87

References

1 N. A. W. Holzwarth, A. R. Tackett and G. E. Matthews, *Comput. Phys. Commun.*, 2001, **135**, 329-347.

2 R. S. Brown A, *Acta Crystallogr. B*, 1965, **19**, 684-685.

3 R. M. Wood, *Proc. Phys. Soc.*, 1962, **80**, 783.

4 S. A. Bradford, and O. N. Carlson, *Trans. Am. Soc. Met.*, 1962, **55**, 169-172.

5 A. W. Hull and W. P. Davey, *Phys. Rev.*, 1919, **14**, 540-540.

6 J. A. Oberteuffer and J. A. Ibers, *Acta Crystallogr. B*, 1970, **26**, 1499-1504.

7 H. P. Weik H, *Bull. Am. Phys. Soc.*, 1965, **10**, 1140.

8 H. M. Otte, *J. Appl. Phys.*, 2004, **32**, 1536-1546.

9 M. B. Kanoun, S. Goumri-Said and M. Jaouen, *Phys. Rev. B*, 2007, **76**, 134109.