Table S1-The optimized geometric and electronic structure parameters of MA_2Z_4 monolayer including A-Z (d_1, d_2) and M-Z (d_3) bond lengths; A-Z-A (θ_1), Z-A-Z (θ_2), and Z-M-Z (θ_3, θ_4) angles.

Structure	d_1 (Å)	d_2 (Å)	d_3 (Å)	θ_1 (°)	θ_2 (°)	θ_3 (°)	θ_4 (°)
$MoSi_2N_4$	1.75	1.74	2.09	111.96	106.82	87.85	73.54
$MoSi_2P_4$	2.24	2.23	2.45	100.71	117.20	89.73	70.91
$MoSi_2As_4$	2.35	2.33	2.55	98.97	118.60	89.08	71.83
$MoGe_2N_4$	1.86	1.89	2.12	108.74	110.22	90.73	69.50
$MoGe_2P_4$	2.32	2.32	2.46	99.12	118.51	91.46	68.48
$MoGe_2As_4$	2.43	2.43	2.57	98.04	119.35	91.02	69.05
WSi_2N_4	1.75	1.74	2.09	111.99	106.82	87.74	73.71
WSi_2P_4	2.24	2.23	2.45	100.96	117.05	89.94	70.61
WSi_2As_4	2.36	2.35	2.55	99.24	118.60	89.33	71.22
WGe_2N_4	1.86	1.89	2.13	108.99	109.90	90.82	69.35
WGe_2P_4	3.34	5.61	3.90	64.25	142.12	54.15	116.60
WGe_2As_4	2.44	2.46	2.59	98.46	119.01	91.30	68.68
VSi_2N_4	1.73	1.74	2.02	111.12	107.81	90.26	70.17
VSi_2P_4	2.25	2.22	2.41	101.47	116.58	92.16	67.44
VSi_2As_4	2.35	2.34	2.52	99.17	118.45	90.77	69.44
VGe_2N_4	1.85	1.89	2.06	107.83	111.07	93.28	65.82
VGe_2P_4	2.32	2.34	2.43	99.43	118.35	93.86	64.99
VGe_2As_4	2.43	2.44	2.53	97.99	119.39	92.62	66.79

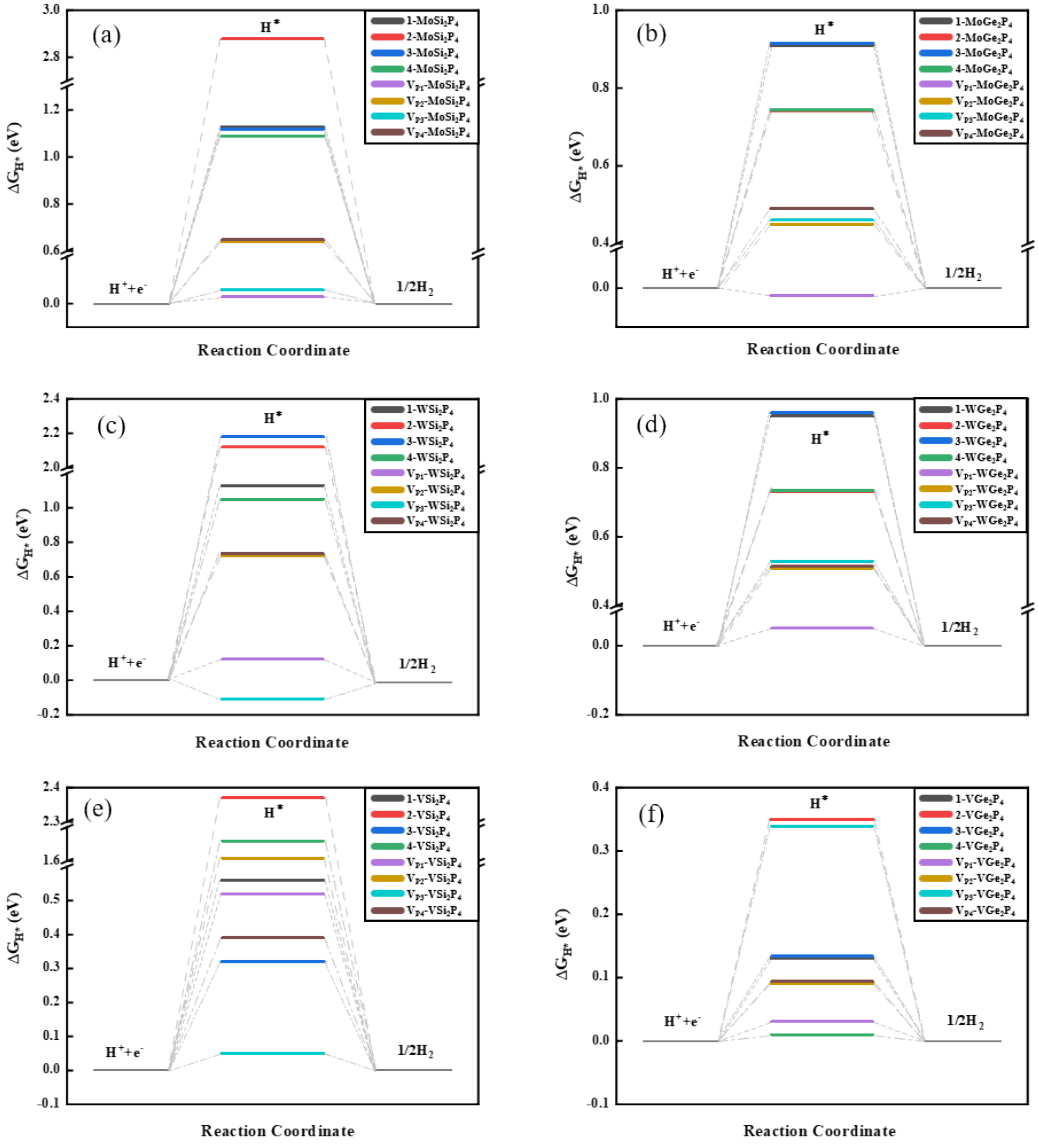

Figure S1. The Gibbs free energy of (a) $MoSi_2P_4$, (b) WSi_2P_4, (c) VSi_2P_4, (d) $MoGe_2P_4$, (e) WGe_2P_4, and (f) VGe_2P_4 for intrinsic on site 1-4 and the Gibbs free energy of them for hydrogen evolution after V_p on site1-4.