

## **Electronic Supplementary Material (ESI) for**

# **Exploiting monolayer MoSi<sub>2</sub>N<sub>4</sub>, WSi<sub>2</sub>N<sub>4</sub> and WGe<sub>2</sub>N<sub>4</sub> for efficient photocatalytic overall water splitting across a broad pH range**

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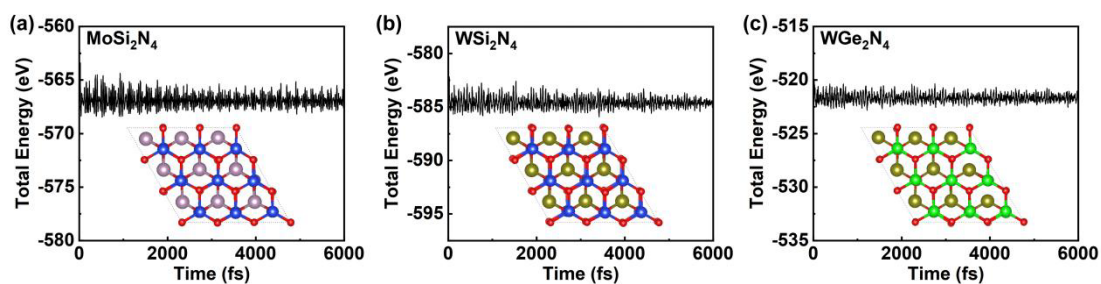


Fig. S1 AIMD of  $\text{MoSi}_2\text{N}_4$ ,  $\text{WSi}_2\text{N}_4$ , and  $\text{WGe}_2\text{N}_4$

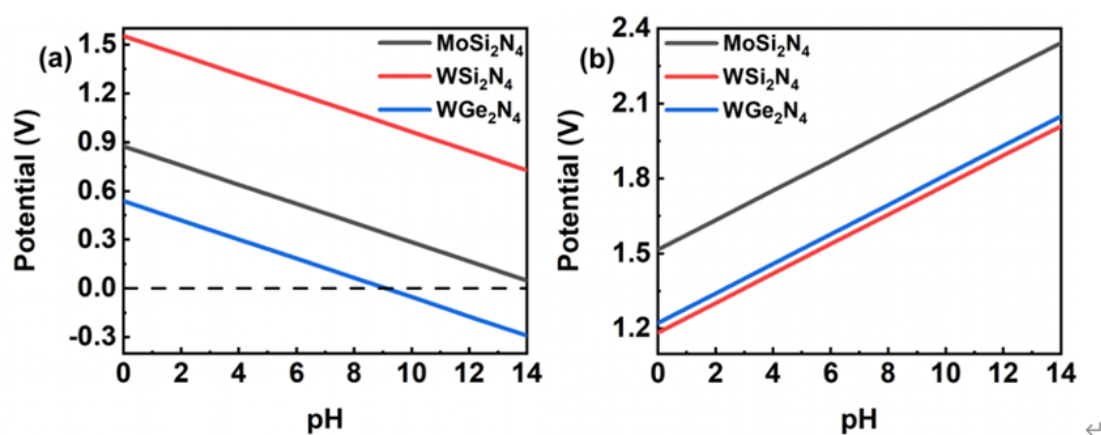


Fig. S2 The external potentials of (a)  $U_e$  and (b)  $U_h$  provided by  $\text{MoSi}_2\text{N}_4$ ,  $\text{WSi}_2\text{N}_4$ , and  $\text{WGe}_2\text{N}_4$ , respectively, where the negative value indicates that there is no photocatalytic activity.

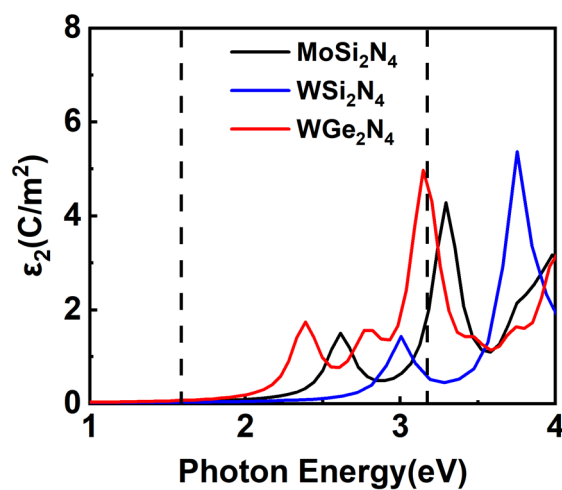


Fig. S3 Calculated imaginary part of dielectric function of  $\text{MoSi}_2\text{N}_4$ ,  $\text{WSi}_2\text{N}_4$ , and  $\text{WGe}_2\text{N}_4$  using HSE06.

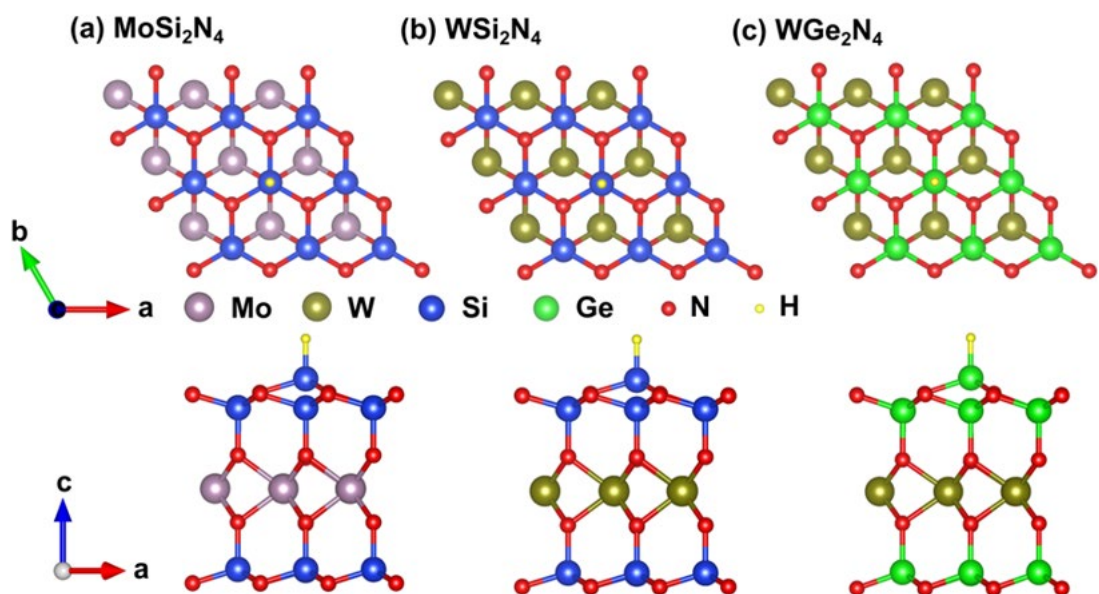


Fig. S4 The top and side view of (a)  $\text{MoSi}_2\text{N}_4$ , (b)  $\text{WSi}_2\text{N}_4$  and (c)  $\text{WGe}_2\text{N}_4$  of hydrogen atom adsorption, respectively.

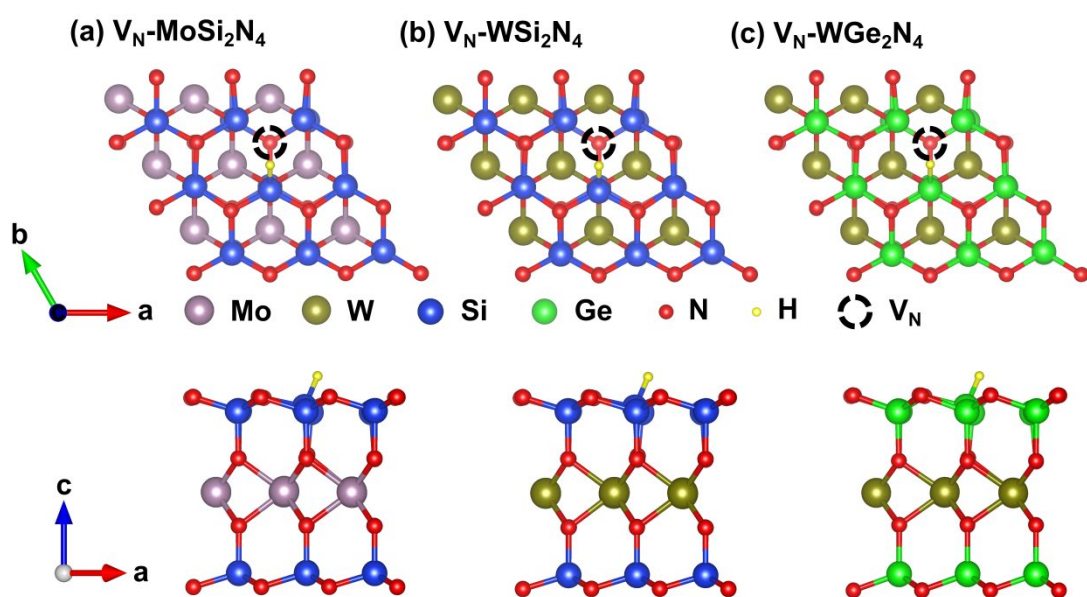


Fig. S5 The top and side view of (a)  $\text{V}_\text{N}$ - $\text{MoSi}_2\text{N}_4$ , (b)  $\text{V}_\text{N}$ - $\text{WSi}_2\text{N}_4$  and (c)  $\text{V}_\text{N}$ - $\text{WGe}_2\text{N}_4$  of hydrogen atom adsorption, respectively.

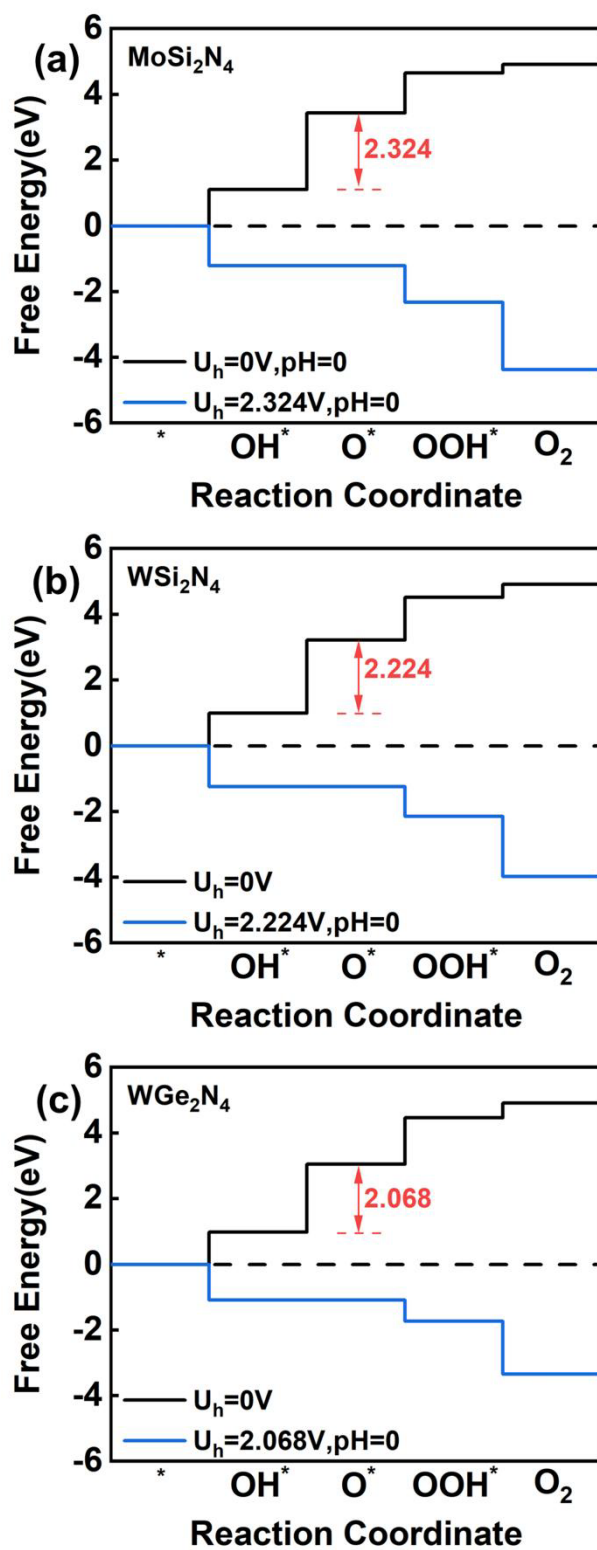


Fig. S6 The free energy for OER on the surface of (a)  $\text{MoSi}_2\text{N}_4$ , (b)  $\text{WSi}_2\text{N}_4$ , and (c)  $\text{WGe}_2\text{N}_4$ , respectively.

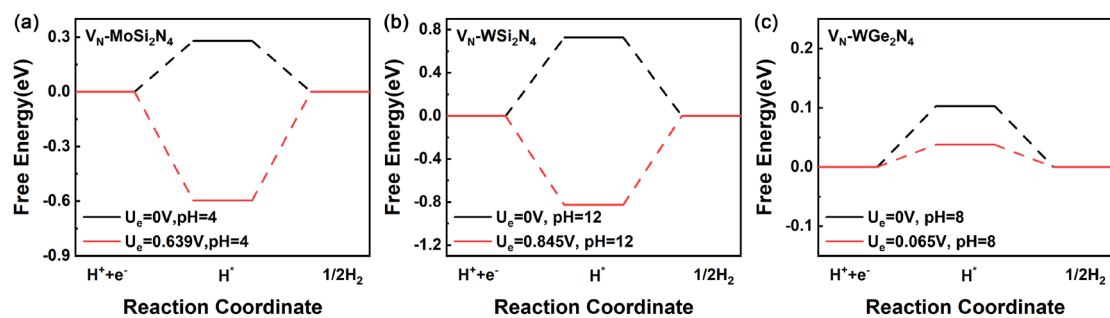


Fig. S7 The free energy for HER in (a)  $MoSi_2N_4$ , (b)  $WSi_2N_4$ , and (c)  $WGe_2N_4$  with considering the N vacancy defects. Black line corresponds the conditions absent of light irradiation at pH = 4, 12, and 8, the red line denotes the provision of light at pH = 4, 12, and 8 for  $MoSi_2N_4$ ,  $WSi_2N_4$ , and  $WGe_2N_4$  respectively.