Supporting Information

First-Principles Screening of Single Transition Metal Atom Anchored on Two-Dimensional C_9N_4 for Nitrogen Reduction reaction

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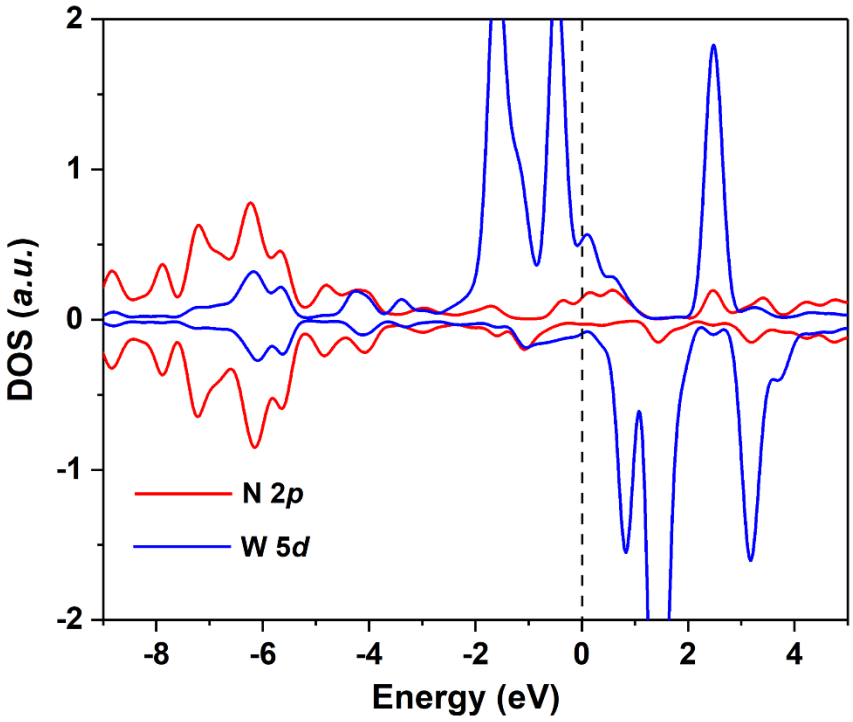

Figure S1. The calculated partial DOS projected on the 5*d* orbitals of W and 2*p* orbitals of the neighboring N atoms for W@C_9N_4.

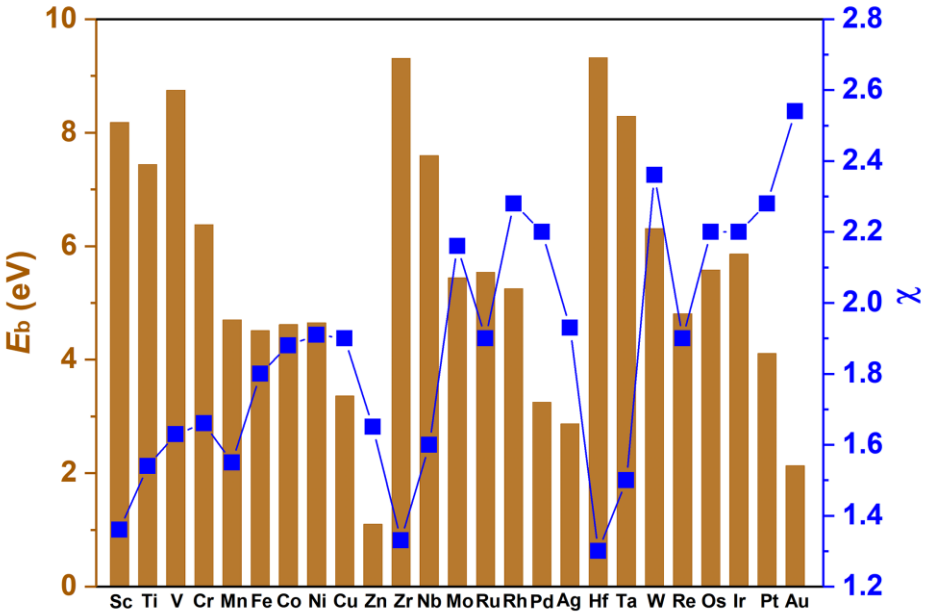

Figure S2. The relationship between binding energy (E_b) and electronegativity (χ) for the 25 metal atoms.

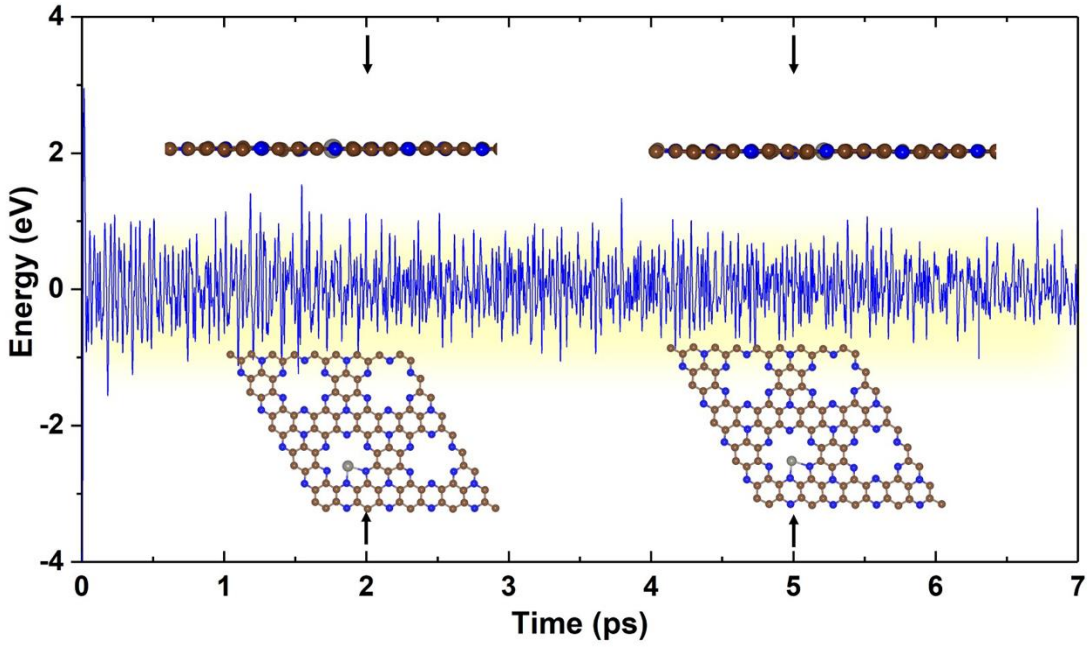

Figure S3. The *ab initio* molecular dynamics simulated energy evolution for W@C_9N_4 at a temperature of 300 K. Insets show the top and side views of the snapshots of atomic configurations at 2 ad 5 ps.

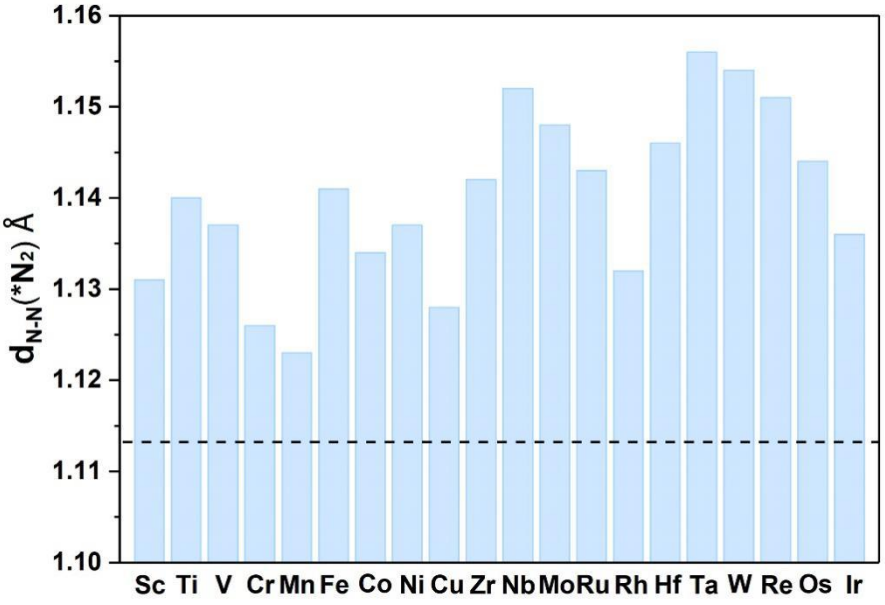

Figure S4. The calculated bond length of the adsorbed N_2 on M@C_9N_4. The black dotted line represents the bond length of N_2 in the gas phase.

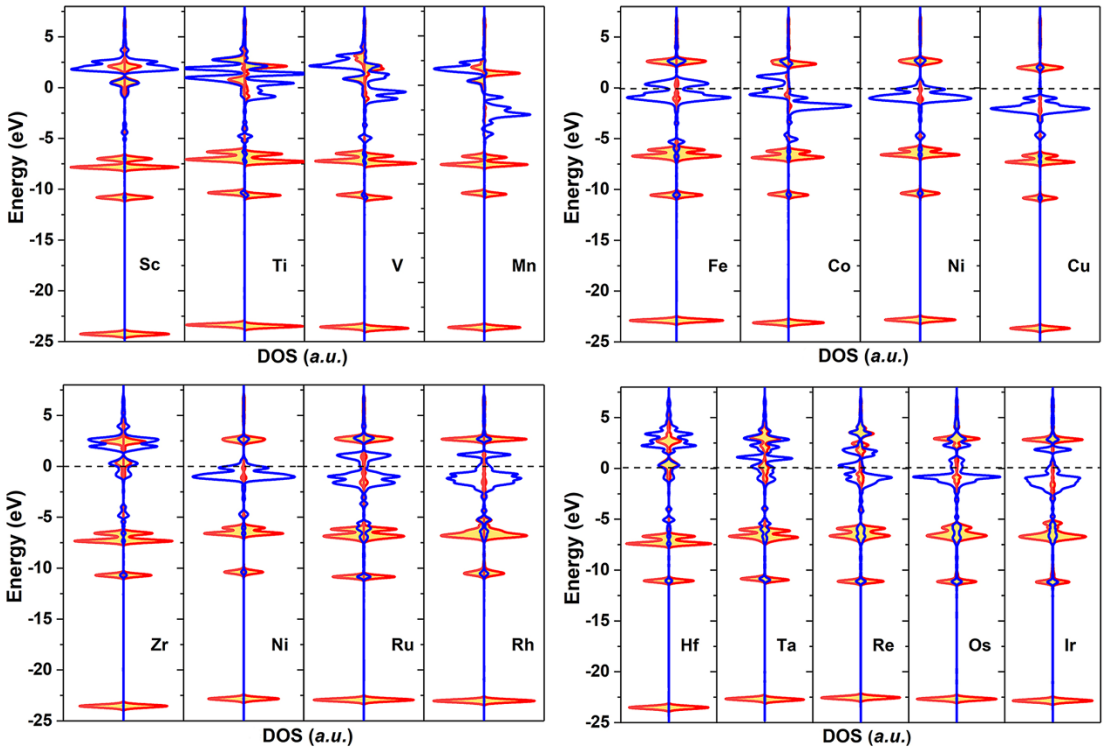

Figure S5. The calculated partial DOS projected on the molecular orbitals (red curves) of N_2 and the *d* orbitals (blue curves) of metal atoms for N_2 adsorption on Sc, Ti, V, Mn, Fe, Co, Ni, Cu, Zr, Ni, Ru, Rh, Hf, Ta, Re, Os and Ir@C_9N_4. Fermi level is set to zero.

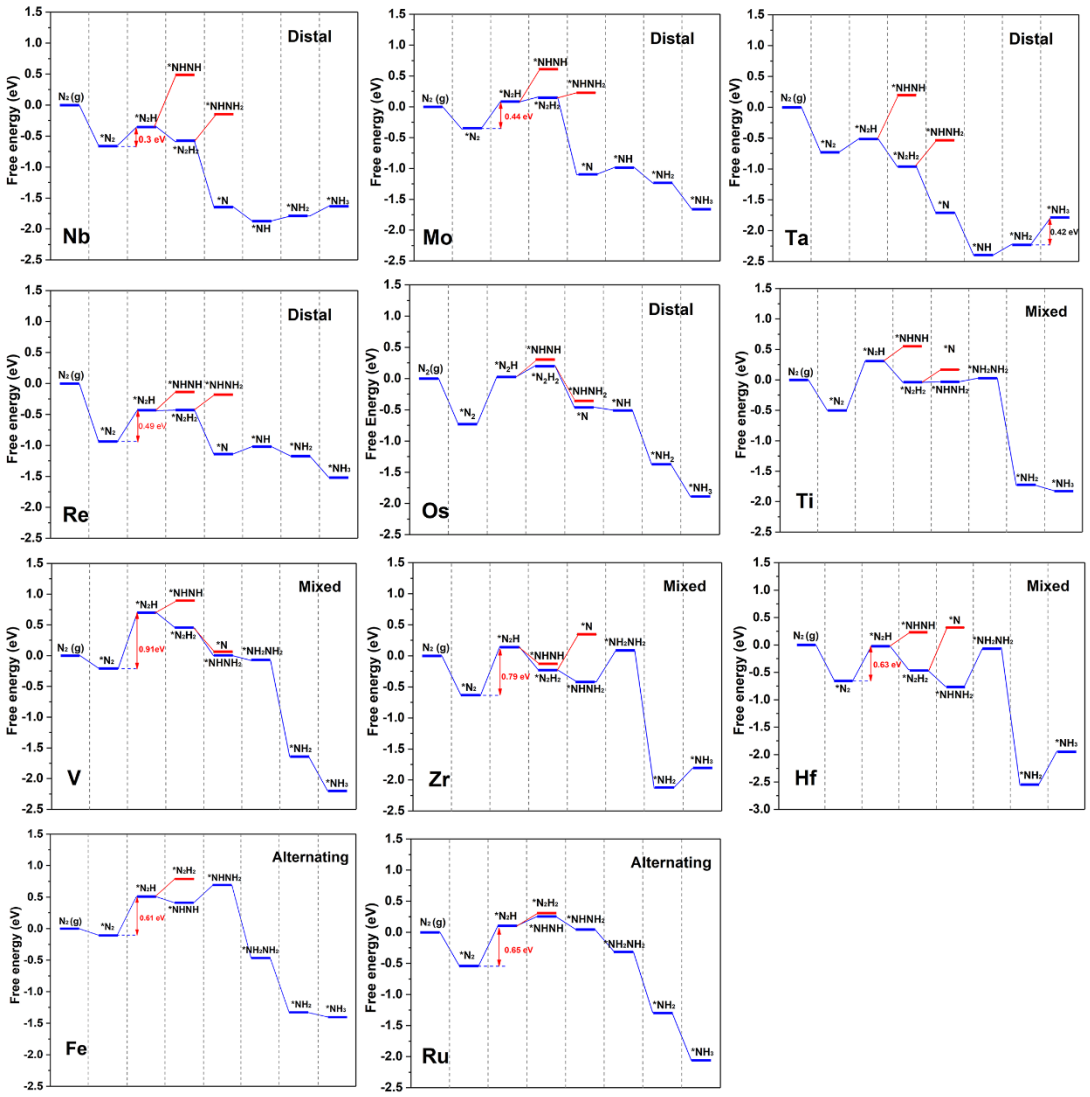

Figure S6. The calculated free energy diagrams of NRR on Nb, Mo, Ta, Re and Os@C_9N_4 along the distal pathway (top panel), Ti, V, Zr and Hf@C_9N_4 along the mixed pathway (middle panel), Fe and Ru@C_9N_4 along the alternating pathway (bottom panel).

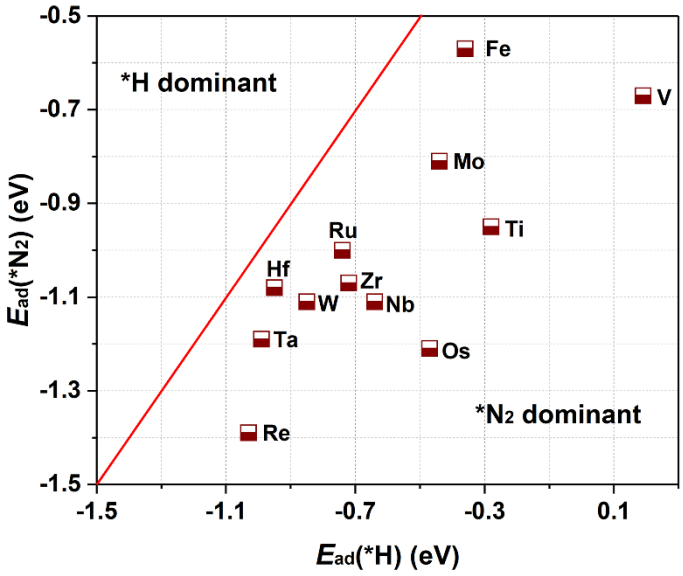

Figure S7. The calculated adsorption energy of N_2, $E_{ad}(^*N_2)$, with reference to the adsorption energy of H, $E_{ad}(^*H)$, on $M@C_9N_4$. The red line is indicated to guide the eye.