[Electronic Supplementary Information]

Title: **ψ-BCN Monolayers as Emerging 2D Materials: Effects of Hydrogen Passivation on Structure, Stability, and Functionality**

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Figure S1. Initial and optimized configuration with different ratio of B, C, and N atoms with relative energy (ΔE)

Figure S2. Hydrogen passivation on multiple different atomic sites (a) H-passivation on B and C atoms (b) H-passivation on B and N atoms

Figure S3. Polar plots of (a) directional Young's modulus and (b) directional Poisson's ratio for hydrogenated ψ-BCN monolayers in comparison with pristine ψ-BCN.

Figure S4. Total potential energy fluctuation of the ψ-BCN monolayer at 300 K, 600 K, and 1000 K during the AIMD simulations. The right panels display the final structural snapshots at each temperature, confirming the thermal stability and structural integrity of the monolayer.

Figure S5. Total potential energy fluctuation of the hydrogenated ψ-BCN monolayer at 1000 K during the AIMD simulations. The right panels display the final structural snapshots of each hydrogenated structure, confirming the thermal stability and structural integrity of the monolayer.

Figure S6. Electron localization function (ELF) maps of ψ-BCN monolayers: (a) pristine ψ-BCN, (b) ψ-B_HCN, (c) ψ-BC_HN, and (d) ψ-BCN_H. Nitrogen atoms are shown in blue, boron in green, and carbon in grey. The color scale (0.0-1.0) indicates the degree of electron localization, with higher values representing stronger localization. Hydrogenation at C and B maintains the overall ELF topology, whereas hydrogenation at N leads to pronounced localization around the N-H bond and noticeable perturbation of nearby bonding regions.

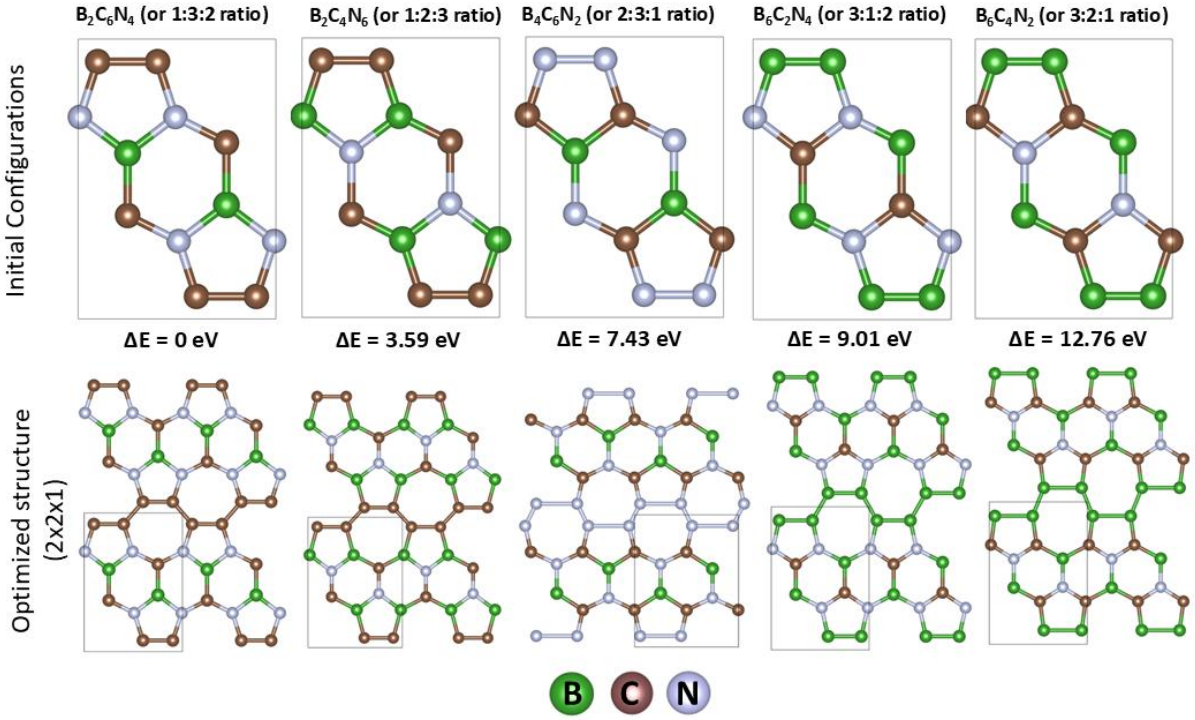

Figure S1

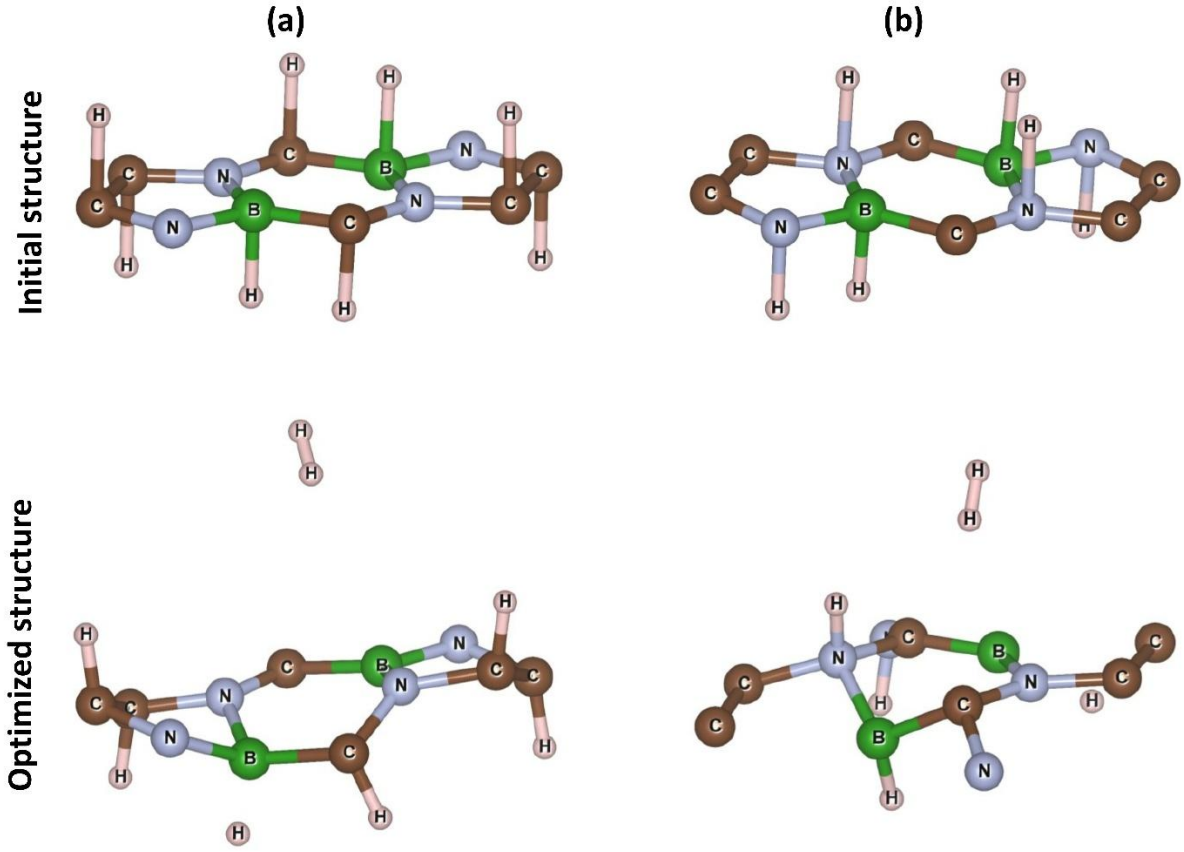

Figure S2

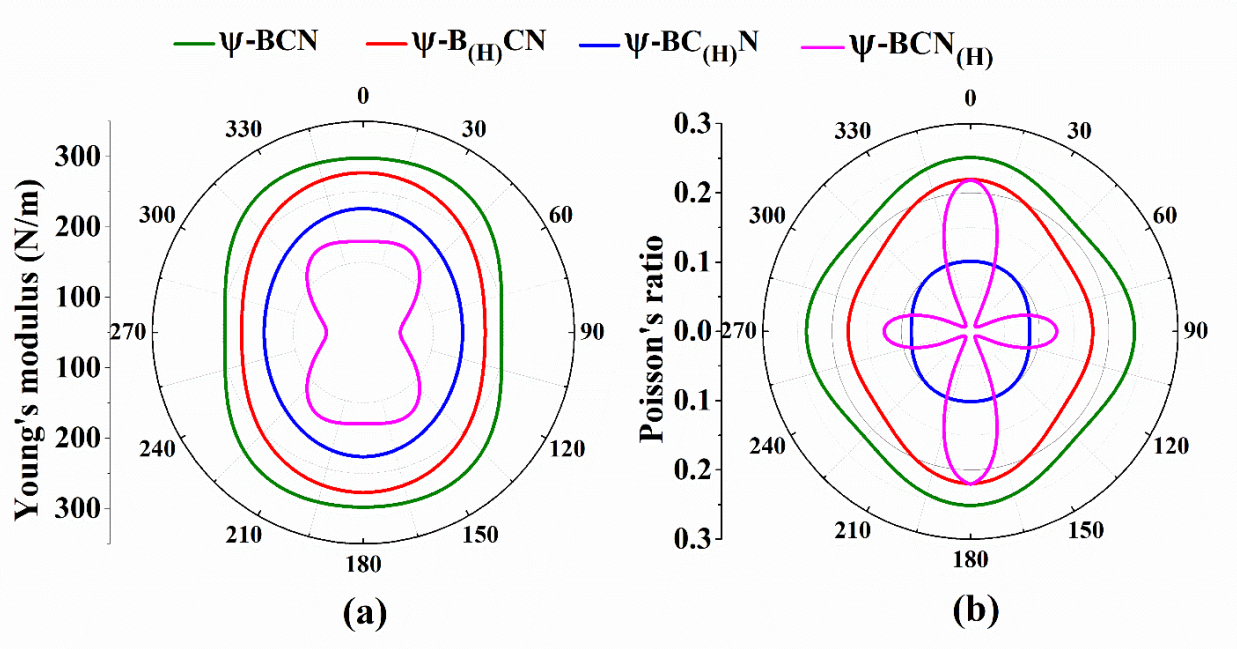

Figure S3

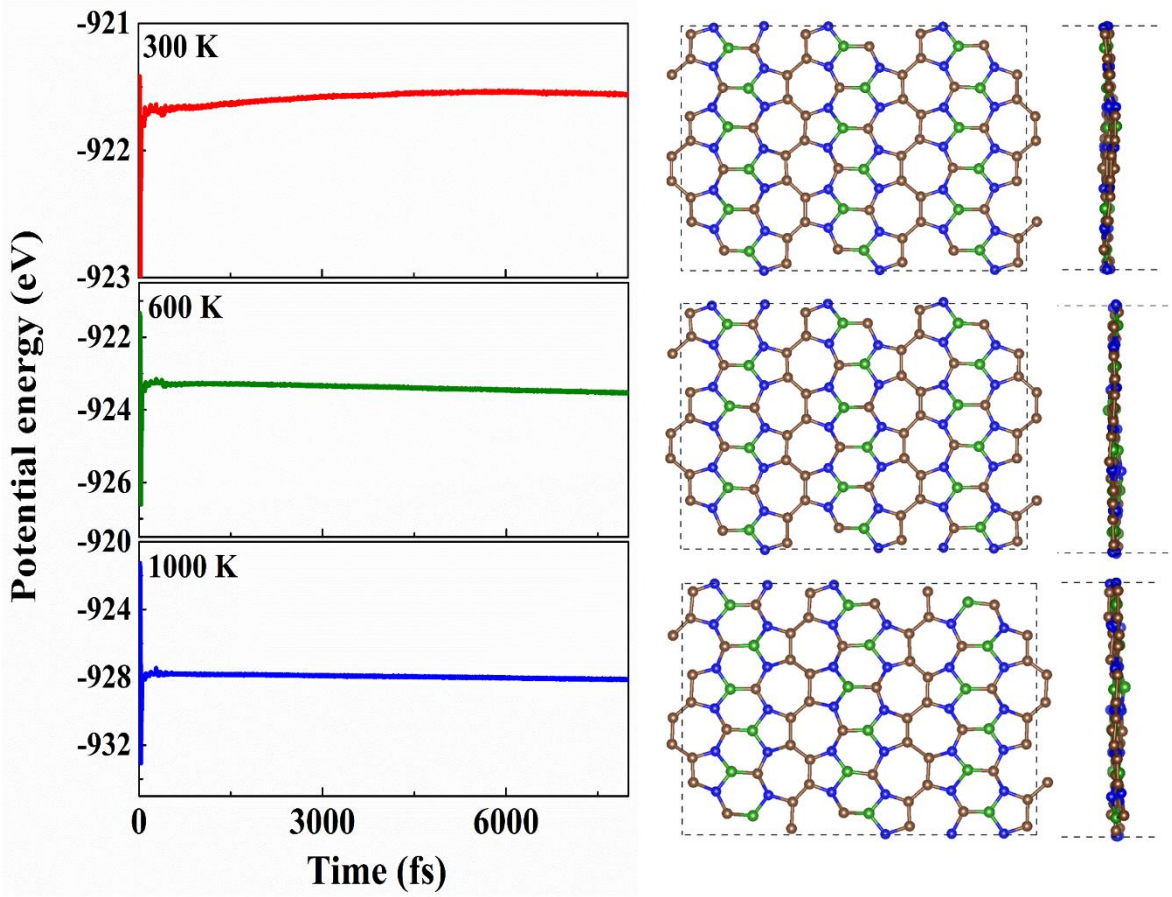

Figure S4

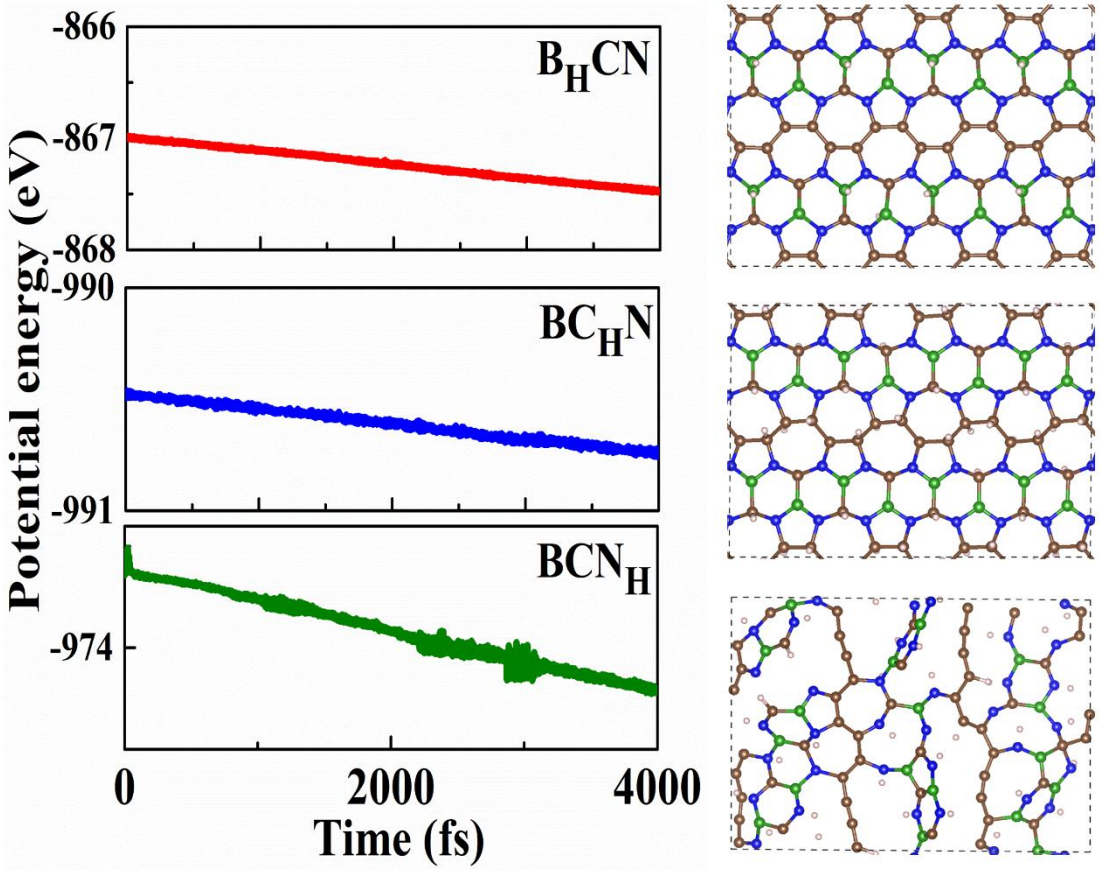

Figure S5

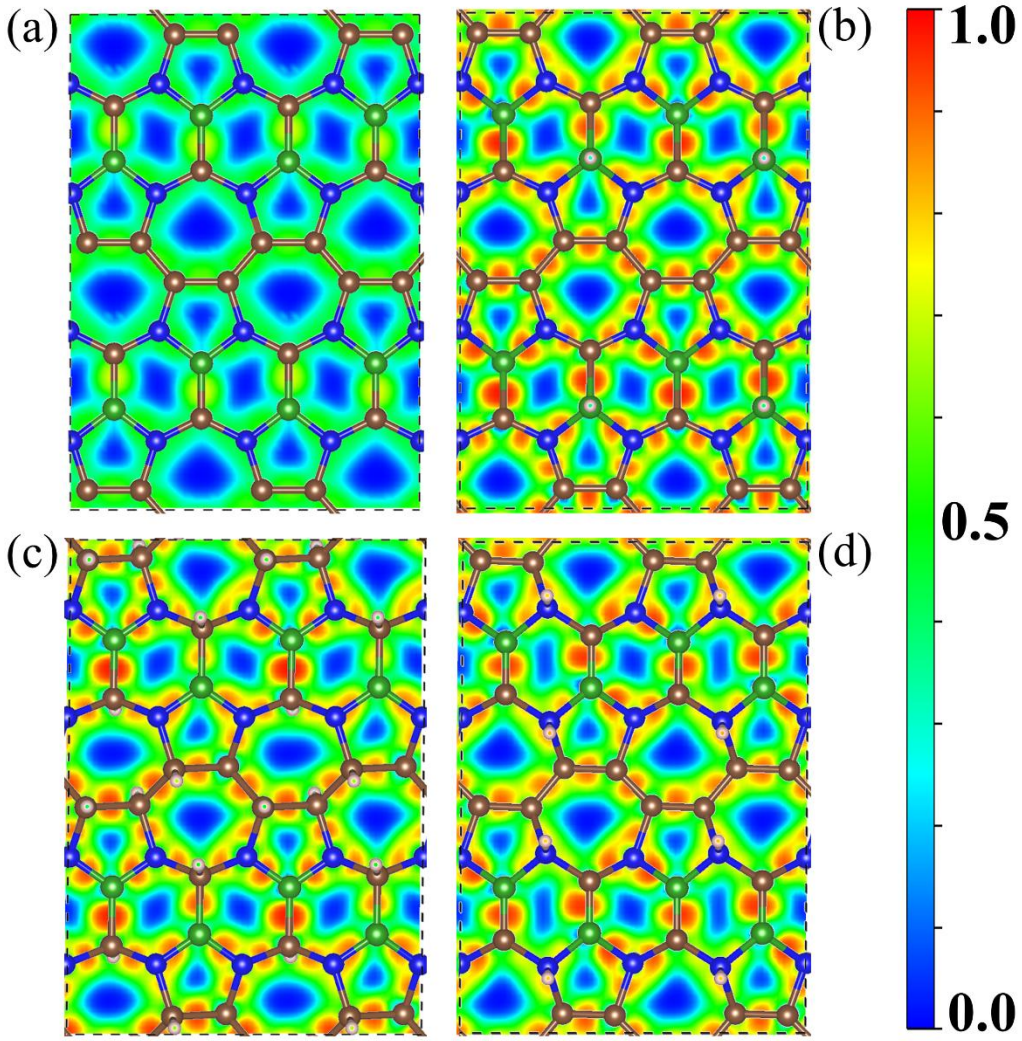

Figure S6