Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Supplementary Materials

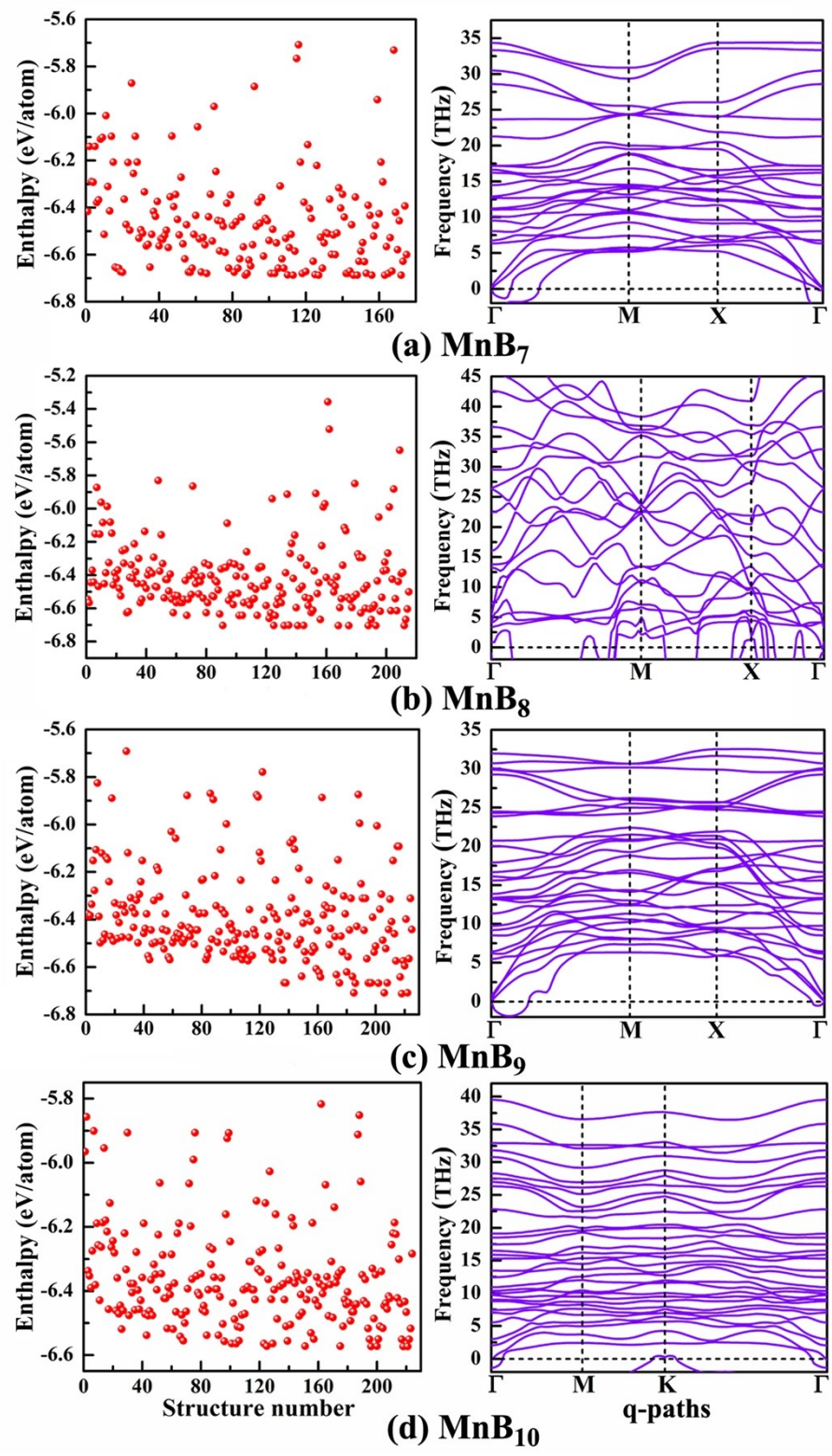

Figure S1 Typical enthalpy evolutions for the MnB_n (n=7~10) during evolutionary structural searches. In each panel, the calculated phonon spectrum of the lowest energy structure is also provided, which suggests the dynamic unstability due to the imaginary phonon modes.

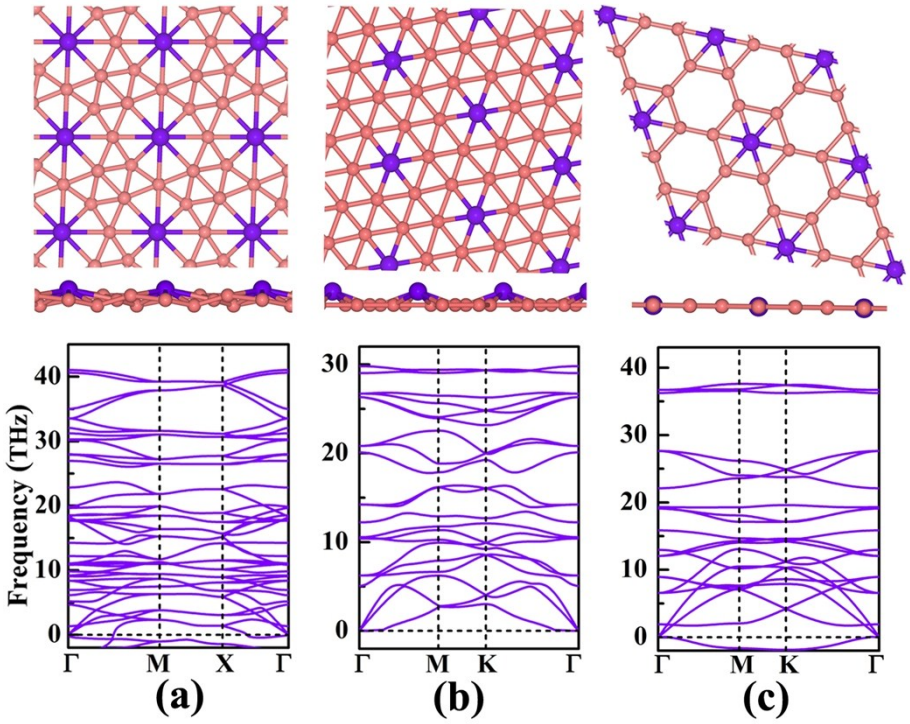

Figure S2 Structures of the MnB_6 optimized after replacing Fe with Mn in the α-, β-, and γ-phases of FeB_6 are shown in (a), (b), and (c), respectively. The corresponding phonon spectra are also calculated. Only β-phase configuration is dynamically stable for MnB_6.

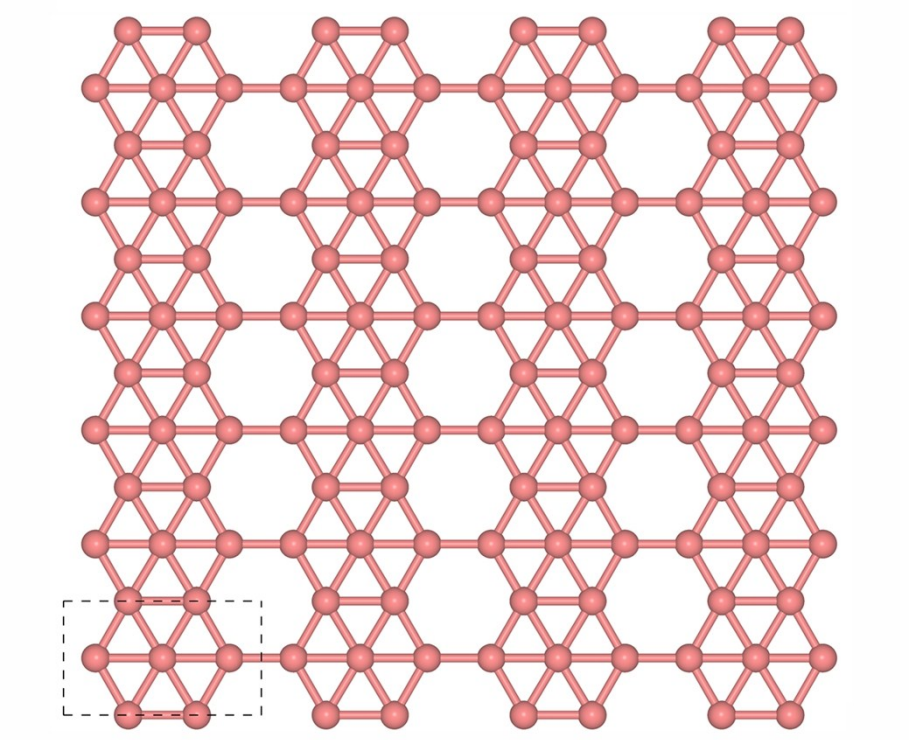

Figure S3 Geometrical configuration of the experimentally fabricated borophene. The lowest energy free-standing structure proposed by Wu and coworkers [*Nat. Chem.* **8**, 563 (2016)] was adopted in our studies as reference.

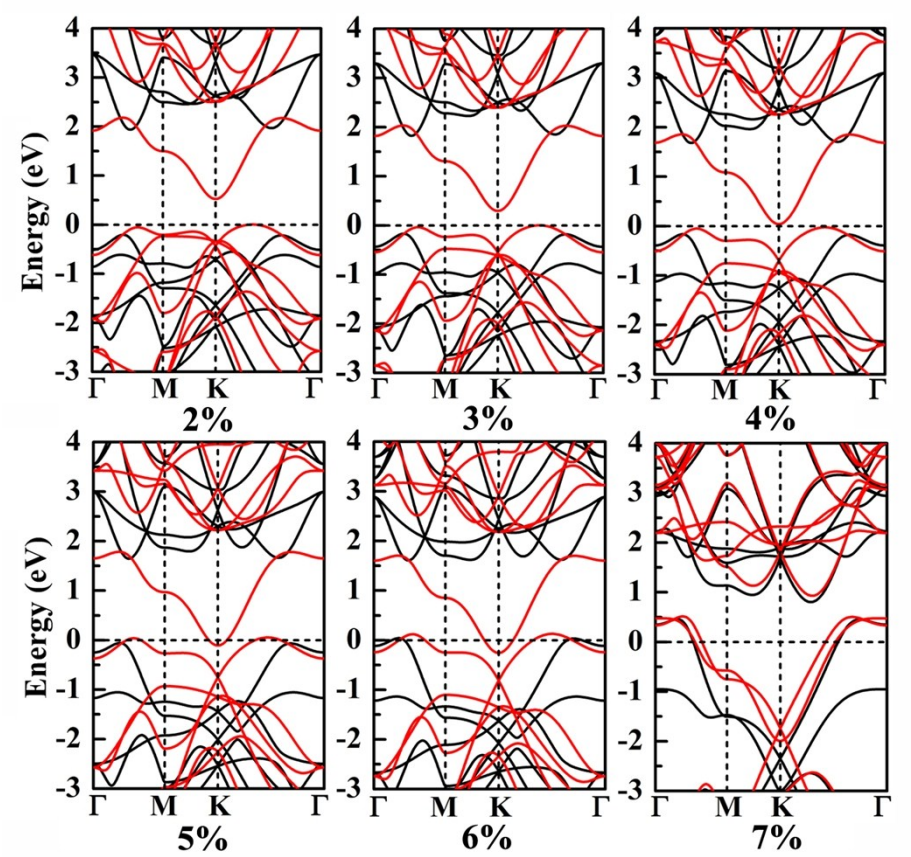

Figure S4 Spin-resolved bandstructures of the strain engineered MnB_6 in FM state. The black and red lines are for the spin-up and spin-down electrons, respectively. The transport properties can be changed from semiconducting to metallic.

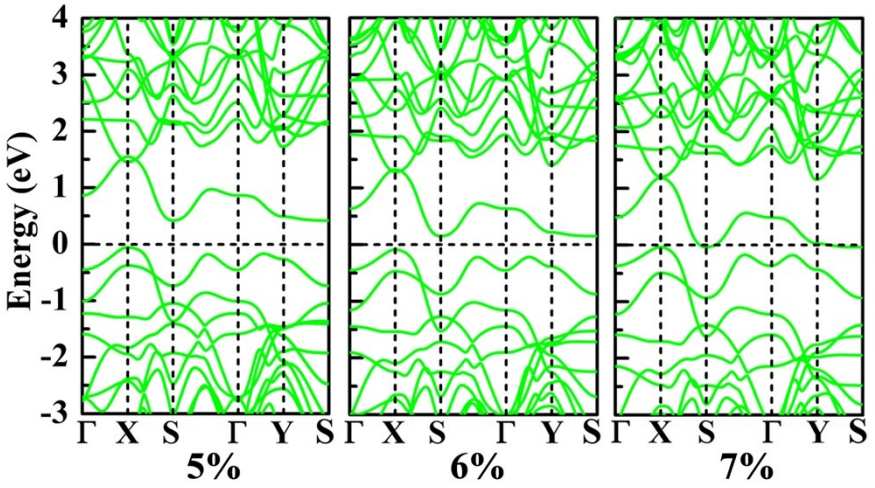

Figure S5 Bandstructures of the strain engineered MnB_6 in AFM state. Only the cases under 5%, 6%, and 7% strains are provided for illustration.

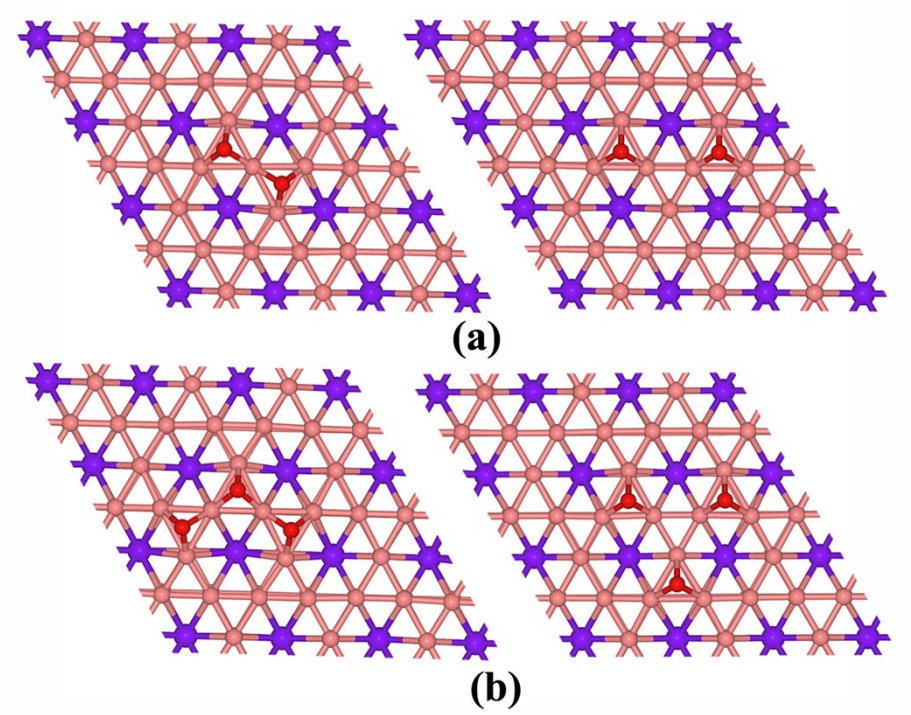

Figure S6 Adsorption configurations of two O atoms (a) and three O atoms (b) on MnB_6 sheet. Compared to the nearest neighboring adsorptions, the next-nearest neighboring adsorptions are lower in total energy by 0.43 and 0.94 eV for two and three O atoms, respectively.

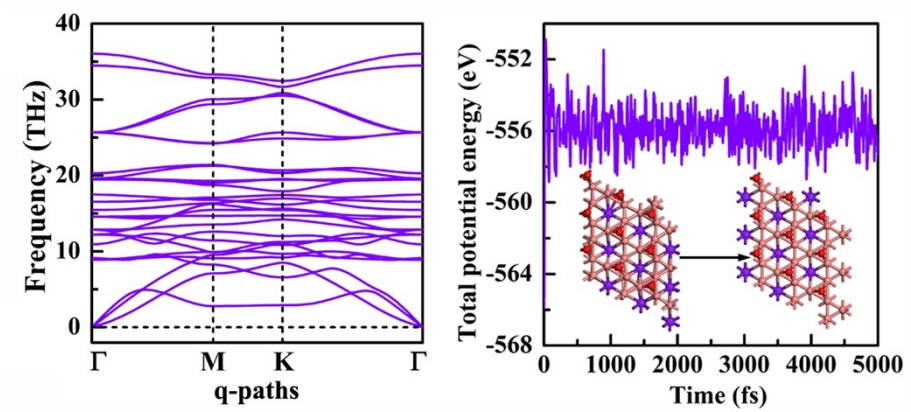

Figure S7 The left is the calculated phonon spectrum for MnB_6O_2, suggesting dynamic stability attributed to the positive phonon modes. The right is the evolution of potential energy versus simulation time. The molecular dynamics simulation at the temperature of 1000 K is shown. Due to the intensive computing load, the 3×3×1 supercell was adopted. The insets are the structure obtained at the end of simulation at 1000 K and the corresponding equilibrium structure obtained after structural optimization at 0 K. The blue, brown, and red balls represent Mn, B, and O atoms, respectively.

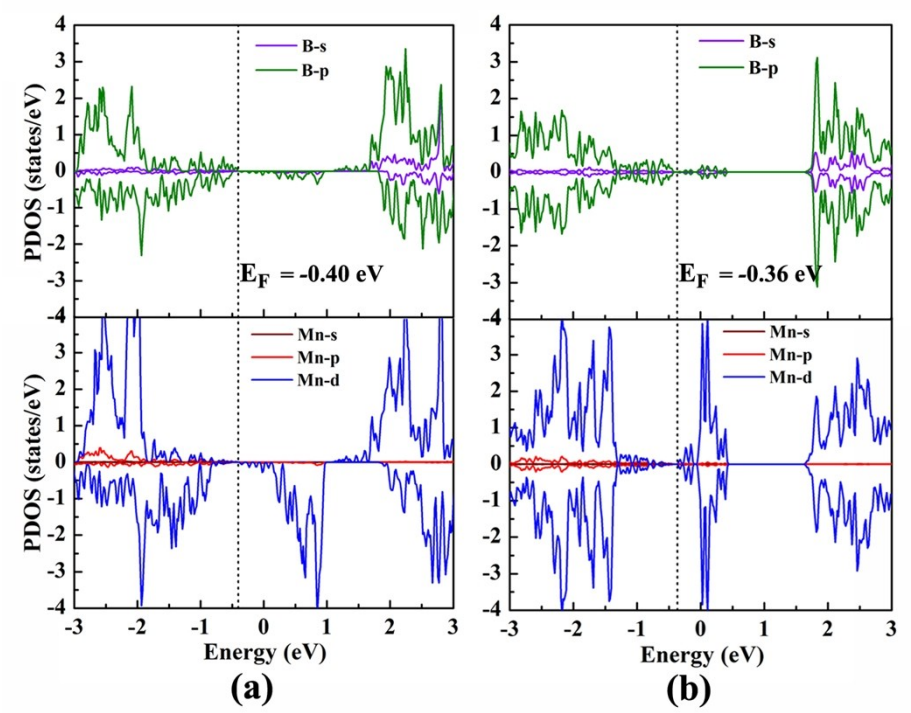

Figure S8 The partial densities of states of B and Mn atoms of the MnB_6O_2 structure in FM state (a) and AFM state (b). The positive and negative densities are for the spin-up and spin-down electrons, respectively. The lower E_F energy of FM state is an indication of slightly stronger orbital hybridization between B 2p and Mn 3d.