Figure S1 Phase diagrams plotted for the calculated total energies versus the lattice ratios referring to the corresponding lattice constants of 3.46, 4.45, 4.75, and 5.36 Å for tri-, α-, β-, and γ-FeB₆, respectively. All of the studied structures contain one Fe and six B atoms in their corresponding primitive unit cells. These data were calculated on PBE level.
Figure S2 The optimized adsorption configurations of Fe$_4$ and B$_4$ clusters on the $tri$-FeB$_6$ sheet. The brown and dark purple balls stand for B and Fe atoms, respectively. These results suggest the tendency for Fe and B atoms to cluster into particle rather than to uniformly wet the $tri$-FeB$_6$ sheet.
Figure S3 The (a), (b), (c), and (d) show the electronic bandstructures of $\text{tri-FeB}_6$ under 2%, 3%, 4%, and 5% tensile strains, respectively. The corresponding ones for the FeB$_6$O$_2$ oxide sheet are presented in (e), (f), (g), and (h), respectively. The band gaps are marked for eye guidance.