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Theoretical prediction of decomposition paths for $Ca(BH_4)_2$ and $Mg(BH_4)_2$ YONGSHENG ZHANG, CHRIS WOLVERTON, ERIC MAJZOUB, VIDVUDS OZOLINS, MATERIALS SCIENCE & ENGINEERING, NORTHWESTERN UNIVERSITY TEAM, PHYSICS & ASTRONOMY, UNIVERSITY OF MISSOURI COLLABORATION, MATERIALS SCIENCE & ENGINEERING, UNIVERSITY OF CALIFORNIA, LA COLLABORATION — Experimental and theoretical studies on $Ca(BH_4)_2$ indicate that the decomposition pathway of this compound is not a simple one-step reaction to final products, but instead may be a multi-step decomposition path (though the reaction pathway is currently unknown). We have studied the decomposition pathways of both Ca- and Mg-borohydride using Density Functional Theory (DFT) as well as a Monte Carlo-based crystal structure prediction method called PEGS. We find that a recently experimentally proposed CaB_2H_2 intermediate is extremely high in energy and hence very unlikely to form. We then symmetrically studied the low-energy structures of CaB_2H_4 and CaB_2H_6 stoichiometries by PEGS+DFT simulations. Based on all these possible reaction paths, the $Ca(BH_4)_2$ decomposition convex hull using our calculated reaction enthalpies finds a new compound, CaB_2H_6, which forms a nearly degenerate pathway to the previously-proposed $CaB_{12}H_{12}$ phase. Similar calculations for the Mg system show that the MgB_2H_6 predicted structure does not form a stable intermediate in the decomposition of $Mg(BH_4)_2$.

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