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First-principles studies of intermediate products in the decomposition of metal amidoboranes YONGSHENG ZHANG, Department of Materials Science & Engineering, Northwestern University, TOM AUTREY, Pacific Northwest National Laboratory, CHRIS WOLVERTON, Department of Materials Science & Engineering, Northwestern University — Metal amidoboranes [MAB, M=metal cation] form an interesting class of recently-discovered hydrogen storage compounds. However, the decomposition products remain largely unknown. Armed with the combination of the prototype electrostatic ground state search and density-functional theory methodology (PEGS+DFT), we have searched for crystal structures of possible reaction products with $[NHBH_2]^-$, $[NBH]^-$, $[NBH_5]^-$, polymer-$[NHBH_2]$ anion groups in the decomposition of LiAB and CaAB. All these reaction pathways are significantly endothermic, which is in disagreement with the experimentally measured enthalpies in these systems, which are found to be nearly thermoneutral [$-3 \sim -5$ kJ/(mol H_2) in LiAB and 3.5 kJ/(mol H_2) in CaAB]. Using newly developed dianion group $[NHBHNHBH_3]^{2-}$, our PEGS+DFT methodology predicts structures and energies of Li/Ca-dianion compounds. Including vibrational thermodynamics and zero-point effects, we successfully obtain a nearly thermoneutral enthalpy of decomposition into these dianion compounds. This agreement lends strong support to the dianion phases as energetically preferred products in the decomposition of metal amidoboranes.

Yongsheng Zhang
Dept of Materials Science & Engineering,
Northwestern University

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