Abstract Submitted
for the MAR12 Meeting of
The American Physical Society

Theoretical prediction of intermediates in the decomposition of Mg(BH$_4$)$_2$ ERIC MAJZOUB, Center for Nanoscience, and Department of Physics and Astronomy, University of Missouri, YONGSHENG ZHANG, Materials Science and Engineering, Northwestern University, VIDVUDS OZOLINS, Materials Science and Engineering, University of California, Los Angeles, CHRISTOPHER WOLVERTON, Materials Science and Engineering, Northwestern University — We have studied the decomposition pathway of Mg-borohydride using density-functional theory (DFT) calculations of the free energy (including vibrational contributions) in conjunction with a Monte Carlo-based crystal structure prediction method, the prototype electrostatic ground state (PEGS) method. We find that a recently proposed $Mg(B_3H_8)_2$ intermediate [Chong, etc, *Chem. Commun.* **47**, 1330, (2011)] is energetically highly unfavorable with respect to decomposition into $MgB_{12}H_{12}$. We systematically search for low-energy structures of Mg-triboranes [$Mg(B_3H_8)_2$, MgB_3H_7, and $Mg_3(B_3H_6)_2$], closoborane MgB_nH_n (n=6,7,8,9,10,11), and $Mg(B_{11}H_{14})_2$ compounds using PEGS+DFT simulations. We find that only the reaction enthalpy to $Mg_3(B_3H_6)_2$ is close to the stable $MgB_{12}H_{12}$ pathway, and falls within the thermodynamic conditions for reversibility [e.g., Δ H = 20∼50 kJ/(mol H_2)]. Careful control over experimental conditions might allow for $Mg_3(B_3H_6)_2$ as a possible intermediate in the decomposition of $Mg(BH_4)_2$, and might allow $Mg_3(B_3H_6)_2$ to be rehydrided back to $Mg(BH_4)_2$ under modest $H_2(T, p)$ conditions.

Yongsheng Zhang
Materials Science & Engineering, Northwestern University

Date submitted: 10 Nov 2011 Electronic form version 1.4