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Reaction Pathways in the Reactive Composite $Mg(NH_2)_2$ + LiH
DENIZ CAKIR, University of Twente, GILLES A. DE WIJS, Radboud University Nijmegen, GEERT BROCKS — Chen *et al* [1] reported reversible hydrogen storage in a mixture of LiH + $LiNH_2$ with a storage capacity of 6.5 wt %. However, this system requires an operating temperature in excess of 250 C to achieve a hydrogen pressure of 1 bar. Several efforts including cation substitution have been considered in order to improve the operating conditions, which is necessary for onboard applications. For instance, replacing LiH with MgH_2 markedly reduces the operating temperature through the reaction $MgH_2 + 2LiNH_2 \rightarrow Li_2Mg(NH)_2 + 2H_2 \leftrightarrow Mg(NH_2)_2 + 2LiH$. Recent experimental results however indicate that the latter is not a simple one-step reaction and full hydrogenation of $Li_2Mg(NH)_2$ occurs in a two-step sequence via an intermediate $Li_2Mg_2(NH)_3$ [2]. In this work we examine the stability and structure of possible intermediates compounds, namely $Li_{2-2x}Mg_xNH$, $Li_{1-2x}Mg_xNH_2$, and $Li_{2-x}Mg(NH)_{2-x}(NH_2)_x$, by means of first-principles DFT calculations. All intermediate compounds are thermodynamically stable with respect to the elements. The hydrogenation reaction of $Li_2Mg(NH)_2$ via the intermediate imides $Li_{2-2x}Mg_xNH$ is energetically favorable compared to other intermediates.
Ref : [1] Nature **420**, 302 (2002). [2] J. Phys. Chem. C **113**, 15772 (2009).

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