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Hydrazine borane and derivatives for solid state hydrogen storage

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Solid state hydrogen storage is one of the main roadblocks to the development of a suitable hydrogen economy. Among the numerous solutions to overcome this problem, boron- nitrogen-based materials have emerged owing to their high gravimetric and volumetric hydrogen contents. A typical example is ammonia-borane (NH_3BH_3, 19.6wt%H) intensively studied since the early 2000s. A more recent material (2009) envisaged is hydrazine-borane ($N_2H_4BH_3$, 15.3wt%H). Both materials are attractive for hydrogen storage purpose; nevertheless two main drawbacks have been encountered, namely, high temperatures of dehydrogenation (100-200°C) and emission of impurities (borazine, hydrazine, ammonia …). Consequently, to overcome these issues, several strategies have been proposed. Chemical destabilization of these materials belongs to the solutions. Herein, we present our solutions to chemically destabilize hydrazine-borane leading to the synthesis of two novel materials so-called metal hydrazinidoboranes. In a first part, the description of the synthetic methods, to obtain first hydrazine-borane, with a straightforward and relatively low price synthesis optimization (purity ≥ 99% and yield ≈ 80%), followed by the mechano-chemical synthesis of lithium and sodium hydrazinidoboranes ($MN_2H_3BH_3$, M=Li 11.6wt%H and M=Na 8.8 wt%H), are given. Here also, novel in-situ methods, using PXRD and pressure measurements, to follow the reaction between sodium hydride (NaH) and hydrazine-borane are exposed. Then, the physicochemical characterization of these novel materials are drawn, with crystal structure resolution and refinement of sodium and lithium hydrazinidoboranes, ^{11}B MAS NMR results are exposed and compared as well as FTIR spectrum. We have put forward the metastability of lithium hydrazinidoborane leading to two polymorphs and this aspect is described in details. In a final part, we will discuss about thermal characterization, TGA, DSC, isothermal decomposition as well as in- and ex-situ results. Based on the aforementioned results the dehydrogenation mechanisms and optimization of the performances are discussed.

Keywords: hydrogen storage; hydrazine-borane; crystal structure; in-situ XRD; dehydrogenation

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