INDIAN INSTITUTE OF TECHNOLOGY GUWAHATI
SHORT ABSTRACT OF THESIS

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Thesis Title: **Experimental and Molecular Modeling Insights on the Thermolytic Dehydrogenation of Amine Boranes with Ionic Liquids and Deep Eutectic Solvents**

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SHORT ABSTRACT

The scientific community has highlighted hydrogen energy as a potential fuel for the foreseeable future. The search for materials capable of storing hydrogen efficiently while staying compact and lightweight is one of the most challenging issues facing the burgeoning hydrogen economy. Using solid chemical hydrides with high gravimetric and volumetric hydrogen densities may circumvent the challenges of hydrogen storage. It is generally known that B-N compounds include hydrogen bonded to the B and N moieties, which can be desorbed at a reasonable temperature in the presence of a solvent or a catalytic system. In the amine borane family, ammonia borane (AB) is the most fundamental component; it has been identified as a promising chemical hydride storage medium due to its high hydrogen weight percentage (19.6 wt%) and possible regenerability. AB has a greater gravimetric density than the vast majority of other chemical systems that have been reported. This capacity and stability have reignited interest in amine boranes as hydrogen storage carriers. The solid-phase nature of these hydrogen storage systems offers numerous advantages, including simplicity of discharge, enhanced kinetics, and temperature-controlled release of hydrogen equivalents.

However, solid-state dehydrogenation of AB has significant restrictions, including the need to modify the structure of AB molecules by employing carbon derivatives or adducts. Ionic liquids (ILs) and Deep Eutectic Solvents (DESs), a class of environmentally friendly solvents, have several potential usages because of their physicochemical features. ILs and DESs can be utilized to sidestep the issue of thermolysis of AB and its adducts due to their low vapor pressure, superior thermal stability, and molecular tunability. Due to their solubilizing capabilities, ILs/DESs tend to stabilize the polar and transition intermediates produced during the dehydrogenation process, and they effectively inhibit the onset phase of the dehydrogenation process of AB and its adducts. Consequently, they foster a conducive environment for dehydrogenation to take place.

This thesis discusses the use of ILs and their analogues, specifically DESs, as catalytic reaction media for the thermal dehydrogenation of amine borane complexes. The dehydrogenation of these specific chemical compounds facilitates their utilization as hydrogen storage carriers for automobiles and transportation infrastructure. Both solvent classes utilized in this research are non-toxic and accelerate hydrogen gas emission at lower temperatures. Effectively covering the principles of the usage of these specialized solvents, this work paves the way for future efforts to design solvents suitable for comparable applications. In addition, the thesis elaborates on the numerous

thermal and chemical characteristics of the dehydrogenation process, such as the screening and tuning of solvents, the comprehension of their formation mechanism, and other crucial aspects. The research also investigates the applicability of Quantum Chemical (QC) calculations to the design of novel solvents, the search for new hydrogen storage carriers, and the onset of dehydrogenation. These characteristics were thoroughly investigated through experimental investigations and validated with the density functional theory-based transition state search, enabling us to investigate and predict the reaction mechanism of novel hydrogen storage carriers in the presence and absence of studied exotic solvents (ILs and DESs). The thesis effectively addresses hitherto undiscovered issues by demonstrating amine boranes as a suitable hydrogen storage carrier and the application of sustainable solvents such as ILs and DESs in the thermal dehydrogenation process.
