Hydrogen Activation by complex aluminates

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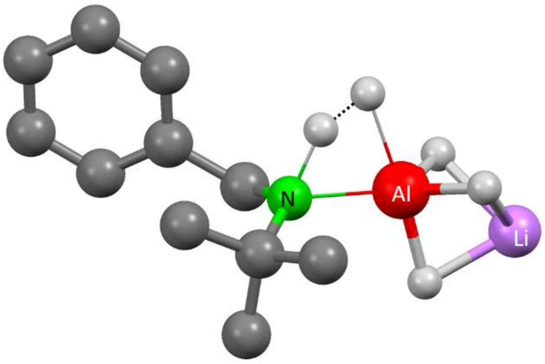
Traditionally lithium aluminum hydride ($LiAlH_4$) is used as a stoichiometric reducing agent for among other organic multiple bonds followed by aqueous work up. This process leads to a considerable amount of waste, since $LiAlH_4$ is employed in an excess. Early main group metal complexes are known to react catalytically with C-C multiple bonds and imines in the presence of molecular hydrogen to yield the reduced substrate. [1,2] Presently we were able to observe that aluminate complexes were able to cleave hydrogen heterolytically, resulting in the formation of an aluminum hydride.

This work focuses on the reaction of $LiAlH_4$ with imines leading to the formation of an aluminum amide complex and its subsequent reaction with molecular hydrogen. Various model systems were modeled by DFT methods (B3PW91/6-311++G**) to gain an insight into the molecular processes. Understanding the reaction steps might lead to the development of a highly atom economic catalytic process.



Calculated transition state of the hydrogen cleavage

[1] J. Spielmann, F. Buch, S. Harder, *Angew. Chem. Int. Ed.* **2008**, 47, 9434-9438

[2] H. Bauer, M. Alonso, C. Färber, H. Elsen, J. Pahl, A. Causero, G. Ballmann, F. De Proft, S. Harder, *Nature Catalysis*, **2018**, 1, 40-47