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Title:	Fine-Tuning of Lewis Acidity: The Case of Borenium Hydride Complexes Derived from Bis(phosphinimino)amide Boron Precursors
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Keywords:	Amides Boranes Cations Hydrides Lewis acids
Issue Date:	2016
Publisher:	Wiley-VCH Verlag
Citation:	Chemistry - A European Journal, 22(31), pp. 11035-11041
Abstract:	Reactions of bis(phosphinimino)amines LH and L'H with Me <sub>2</sub> S-BH <sub>2</sub> Cl afforded chloroborane complexes LBHCl (1) and L'BHCl (2), and the reaction of L'H with BH <sub>3</sub> -Me <sub>2</sub> S gave a dihydridoborane complex L'BH <sub>2</sub> (3) (LH=[{(2,4,6-Me <sub>3</sub> C <sub>6</sub> H <sub>2</sub> N)P(Ph <sub>2</sub> )} <sub>2</sub> N]H and L'H=[{(2,6-iPr <sub>2</sub> C <sub>6</sub> H <sub>3</sub> N)P(Ph <sub>2</sub> )} <sub>2</sub> N]H). Furthermore, abstraction of a hydride ion from L'BH <sub>2</sub> (3) and LBH <sub>2</sub> (4) mediated by Lewis acid B(C <sub>6</sub> F <sub>5</sub> ) <sub>3</sub> or the weakly coordinating ion pair [Ph <sub>3</sub> C][B(C <sub>6</sub> F <sub>5</sub> ) <sub>4</sub> ] smoothly yielded a series of borenium hydride cations: [L'BH] <sup>+</sup> [HB(C <sub>6</sub> F <sub>5</sub> ) <sub>3</sub> ] <sup>-</sup> (5), [L'BH] <sup>+</sup> [B(C <sub>6</sub> F <sub>5</sub> ) <sub>4</sub> ] <sup>-</sup> (6), [LBH] <sup>+</sup> [HB(C <sub>6</sub> F <sub>5</sub> ) <sub>3</sub> ] <sup>-</sup> (7), and [LBH] <sup>+</sup> [B(C <sub>6</sub> F <sub>5</sub> ) <sub>4</sub> ] <sup>-</sup> (8). Synthesis of a chloroborenium species [LBCl] <sup>+</sup> [BCl <sub>4</sub> ] <sup>-</sup> (9) without involvement of a weakly coordinating anion was also demonstrated from a reaction of LBH <sub>2</sub> (4) with three equivalents of BCl <sub>3</sub> . It is clear from this study that the sterically bulky strong donor bis(phosphinimino)amide ligand plays a crucial role in facilitating the synthesis and stabilization of these three-coordinated cationic species of boron. Therefore, the present synthetic approach is not dependent on the requirement of weakly coordinating anions; even simple BCl <sub>4</sub> <sup>-</sup> can act as a counteranion with borenium cations. The high Lewis acidity of the boron atom in complex 8 enables the formation of an adduct with 4-dimethylaminopyridine (DMAP), [LBH-(DMAP)] <sup>+</sup> [B(C <sub>6</sub> F <sub>5</sub> ) <sub>4</sub> ] <sup>-</sup> (10). The solid-state structures of complexes 1, 5, and 9 were investigated by means of single-crystal X-ray structural analysis.
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