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Title:	Hydrosilylation of Carbonyls Catalyzed by Hydridoborenium Borate Salts: Lewis Acid Activation and Anion Mediated Pathways
Authors:	Rawat, S. (/jspui/browse?type=author&value=Rawat%2C+S.) Bhandari, M. (/jspui/browse?type=author&value=Bhandari%2C+M.) Porwal, Vishal Kumar (/jspui/browse?type=author&value=Porwal%2C+Vishal+Kumar) Singh, Sandhya (/jspui/browse?type=author&value=Singh%2C+Sandhya)
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Abstract:	The electronically unsaturated three-coordinated hydridoborenium cations [LBH]+[HB(C6F5)3]-(1) and [LBH]+[B(C6F5)4]-(2), supported by a bis(phosphinimino)amide ligand, were found to be excellent catalysts for hydrosilylation of a range of aliphatic and aromatic aldehydes and ketones under mild reaction conditions (L = [(2,4,6-Me3C6H2N)P(Ph2)]2N). The key steps of the catalytic cycle for hydrosilylation of PhCHO were monitored via in situ multinuclear NMR measurements for catalysts 1 and 2. The combined effect of carbonyl activation via the Lewis acidic hydridoborenium cation and the hydridic nature of the borate counteranion in 1 makes it a more efficient catalyst in comparison to that of carbonyl activation via the predominant Lewis acid activation pathway operating with catalyst 2. The catalytic cycle of 1 showed hydride transfer from the borate moiety [HB(C6F5)3]- to PhCHO in the first step, forming [PhCH2-O-B(C6F5)3]-, which subsequently underwent σ -bond metathesis with Et3SiH to form the product, PhCH2-O-SiEt3. Quantum chemical calculations also support the borate anion mediated mechanism with 1. In contrast, the reaction catalyzed by 2 proceeds predominantly via the Lewis acid activation of the carbonyl group involving [LB(H)-OC(H)Ph]+[B(C6F5)4]- as the transition state and [LBOCH2Ph]+[B(C6F5)4]- as the intermediate.
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