

A computational and conceptual DFT study on the mechanism of hydrogen activation by novel frustrated Lewis pairs

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Abstract

A computational and conceptual density functional theory (DFT) study on the mechanism of molecular hydrogen activation by a set of three frustrated Lewis pairs (FLPs) was performed at the ω B97X-D/6-311G(d,p) level of theory. A reduced model and other two prototypes derived from experimental data, based on the donor nitrogen and acceptor boron atoms, were used. Analysis based on the energy results, geometries and the global electron density transfer at the TSs made it possible to obtain some interesting conclusions: (i) despite the well-known very low reactivity of molecular hydrogen, the catalytic effectiveness of the three FLPs produces reactions with almost unappreciable activation energies; (ii) the reactions, being exothermic, follow a one-step mechanism via polarised TSs; (iii) there are neither substituent effects on the kinetics nor on the thermodynamics of these reactions; (iv) the activation of molecular hydrogen seems to be attained when the N–B distance in the FLP derivatives is around 2.74 Å; and (v) the proposed FLP model is consistent with the behaviour of the experimental prototypes. Finally, the ability of the three FLPs as efficient catalysts was evaluated studying the hydrogenation of acetylene to yield ethylene.