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Synergy Effect of Hydrogen Bonding and Steric Interactions for Reactivity and Selectivity of Organic, Inorganic and Organometallic Compounds

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Non-covalent interactions are a highly promising tool for the development of transition-metal-free chemospecific synthetic transformations. Here we demonstrated the implementation of synergy between hydrogen bonding and steric interactions as a simple, precise and flexible synthetic toolbox, allowing the controlled transformation of organic, inorganic and organometallic species into hard-to-reach products.

We show the transformation of *ortho*-dimethylaminoaryloximes and imines into nitriles and various nitrogen heterocycles under mild conditions. This diverse reactivity is activated via hydrogen bonding and facilitated via the buttressing effect of the substituents next to the NMe2 group. All transformations require only simple and easily available acids and solvents, which generally provide precise control over the direction of the reaction.

We introduce the first example of H-bond-assisted chalcogen exchange between arsine oxides and phosphine selenides under mild conditions, providing a powerful approach for the synthesis of arsine selenides. This newly discovered reaction is applicable to various arsine oxides and phosphine selenides, although the use of reagents with bulky substituents significantly hinders its efficiency.

We utilize the non-covalent Li⁻⁻H interaction as a tool for the second lithiation of various lithionaphthalenes. A series of previously inaccessible 4,5-disubstituted derivatives of 1,8-bis(dimethylamino)naphthalene has been prepared in a good to excellent yield.

Keywords

Hydrogen bond: weak, strong; State of system: solution; Organolithium, Pnictine chalcogenides, Nitrogen heterocycles

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Primary author: ANTONOV, Alexander (University of Regensburg)

Presenter: ANTONOV, Alexander (University of Regensburg)