Enantioselective Construction of Carbocycles and Heterocycles Using N-Heterocyclic Carbene (NHC) Organocatalysis

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N-Heterocyclic carbene (NHC)-catalyzed umpolung of aldehydes leading to the formation of nucleophilic acyl anion intermediates and their subsequent interception with various electrophiles, like aldehydes, ketones, imines and activated, polarized C=C-double bonds are explored for the unconventional access to target molecules.[1] The NHC-catalyzed generation of α,β-unsaturated acyl azoliums followed by their interception with various nucleophiles, the formal [3+3] annulation reaction is also well documented. Recently we reported the enantioselective NHC-catalyzed annulations of 2-bromoenals with 1,3-dicarbonyl compounds and enamines via chiral α,β-unsaturated acyl azoliums. The reaction resulted in the asymmetric synthesis of synthetically and medicinally important dihydropyranones and dihydropyridinones.[2] Moreover, we demonstrated that enolizable aldehydes, and heterocyclic C-H acids as nucleophilic partners for chiral α,β-unsaturated acyl azoliums, and the reaction furnished functionalized dihydropyranones.[3] In addition, we also developed the utility of malonates having a ☐-aroxy group as the nucleophilic coupling partner for α,β-unsaturated acyl azoliums leading to the synthesis of chiral cyclopentenes.[4] Very recently, we have intercepted the α,β-unsaturated acyl azoliums with pyrazolones[5] and pyrazolinones.[6] The details of this work will be presented.

References