Carbon-heteroatom containing molecules are indispensable substructures of synthetically valuable compounds ranging from agrochemicals and pharmaceuticals to fine chemicals. Therefore, their easy and atom-efficient synthesis starting from simple, commercially available precursors plays a significant role in modern synthetic chemistry [1]. The hydrogen borrowing strategy provides promising opportunities as it is an atom-efficient method for a variety of transformations of alcohols, in particular the formation of new carbon-carbon or carbon-nitrogen bonds [2]. In order to broaden the applicability of this method we are investigating suitable cascade reactions. We investigated, among others, the selective N-alkylation of anilines with benzylic alcohols catalyzed by a nitrile-ligated variant of Knölker’s complex [3]. While a hydrogen borrowing process is observed in a closed system under inert conditions, a dehydrogenative condensation occurs in an open system in air. The latter leads to a selective formation of reactive imines, which can be further functionalized. Hence, we report the one-pot three-component condensation of enantioenriched α-alkylamino phosphonates by a sequential iron-promoted dehydrogenative condensation followed by a hydrophosphonylation-step promoted by chiral BINOL-based phosphoric acids [4].


