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CYCLOPENTANE-1,2-DIONE BIS(TERT-BUTYLDIMETHYLSILYL) ENOL ETHER IN ASYMMETRIC ORGANOCATALYTIC MUKAIYAMA–MICHAEL REACTIONS

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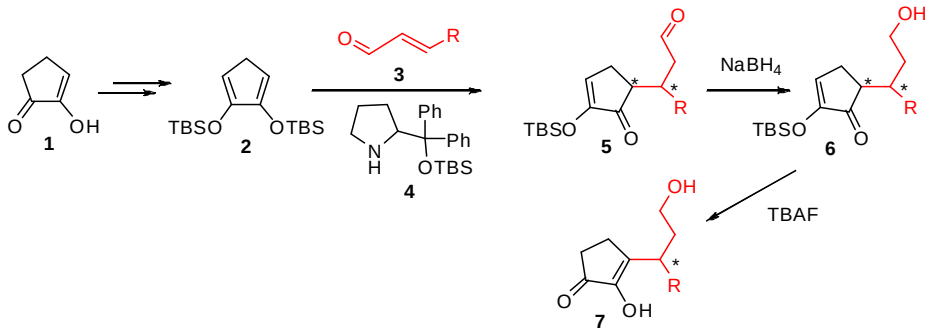
Cyclopentane-1,2-diones are a versatile group of organic compounds that can be transformed into a variety of products, including food flavourings[1] and biologically active compounds, for example antiviral nucleoside analogues.[2] Despite such versatility, cyclopentane-1,2-diones have not received widespread attention as a synthetic feedstock in the preparation of complex compounds. This is partly due to the lack of convenient methods for the preparation of cyclopentane-1,2-diones. Most of the available methods require inefficient and waste producing synthesis and there is only one prior example in the literature that allows to prepare enantiomerically enriched chiral 3-alkylcyclopentane-1,2-diones.

Herein we reveal a novel organocatalytic method that uses a common low molecular weight secondary amine catalyst for the preparation of chiral 3-alkylcyclopentane-1,2-diones. Unsubstituted cyclopentane-1,2-dione was prepared by a known procedure and activated by converting it to a bis-silyldienolate. The latter was used as a nucleophile in an iminium catalytic Mukaiyama-Michael reaction with α,β-unsaturated aldehydes. The primary adduct was reduced *in situ* into a more stable alcohol. Following the removal of the remaining TBS group the product isomerized to its more stable keto-enol form, eliminating one of the initially formed stereogenic centres and producing 3-alkylcyclopentane-1,2-diones in good yields and enantioselectivities. The method constitutes the first example for the organocatalytic preparation of chiral 3-alkylcyclopentane-1,2-diones.



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