

Catalytic Desymmetrization of Diarylmethines and Tandem Atroposelective Cyclodehydration

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Catalyst control over chemical reactions that can produce multiple stereoisomers is an intrinsic and persistent challenge in chemical synthesis, and in the preparation of bioactive molecules. Enormous advances have been achieved in enantioselective catalysis with a myriad of approaches contributing to solutions. However, catalysts that allow control of reactions that involve remote asymmetric induction, wherein remote stereogenic centers are created during asymmetric reactions are uncommon.

We have developed a new set of reactions for the desymmetrization of diarylmethine substrates, structures that are receiving a very high level of interest in the fields of organic synthesis, and as scaffolds for medicinal chemistry. The key to the successful reactions is the development of an unusual Cu-based catalyst system that is rendered chiral through the use of guanidinylated peptide-based ligands. Then, chiral phosphoric acid catalysts are employed to control the formation of an axis of chirality embedded in the scaffold. In conclusion, the systematic study of two widely different types of catalysts on the enantioselective cyclodehydration reaction has been carried out.