Cooperativity in Catalysis: A Novel Method for Enantioselective Transformations

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Substrate activation is an important event in carrying out asymmetric transformations effectively. There are a large number of ways to achieve asymmetric induction with simple substrates. However, activation and highly effective asymmetric transformations with more complex substrates is challenging. In search of a general solution to this important problem, we have initiated a program where two different activators interacting with the substrate cooperatively provide the necessary activation for a variety of transformations. A general scheme of our new methodology is shown below. The substrate is attached to a template that contains both Lewis and Brønsted basic sites. Upon simultaneous activation with a combination of Lewis and Brønsted acids, the substrate is activated to undergo reactions effectively. Proof of principle experiments as well as the utility of this methodology in a number of asymmetric transformations such as Friedel-Crafts alkylation, conjugate amine addition, and dipolar cycloaddition will be presented. Mechanistic details based on solid state, solution phase, and computational studies will be presented.

References

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