ABSTRACT

The thesis entitled "***Organocatalytic Stereoselective C-C Bond Formation Through Desymmetrization, Michael, and Aldol Condensation Reactions***" is divided into five chapters which are mainly focused on the development of organocatalytic stereoselective C-C bond formation. In chapters two and three, the concept of enantioselective desymmetrization has been demonstrated. Chapter four realizes the concept of remote C-H functionalization through bifunctional organocatalysis. Finally, in chapter five, stereoselective synthesis of 3-carboxamidecoumarins tethered α-trifluoromethyl styrenes has been described by employing the concept of an aldol condensation. Chapter 1 discloses a brief description of general organic chemistry, chirality, and its role in day to day life, asymmetric catalysis where special emphasis has been made on organocatalysis, and various activation modes of organocatalysis. Finally, the concept of enantioselective desymmetrization has been elaborated with the main focus on the desymmetrization of cyclopent-4-ene-1,3-dione. In chapter 2**,** a L-*tert*-leucine derived thiourea catalyzed enantioselective homologating annulation of cyclopent-4-ene-dione with 3-cyano-4-methylcoumarins giving access to a wide range of enantioriched polycyclic multisubstituted amino pentafulvene cores has been demonstrated. Importantly, our catalytic system efficiently catalyzed this single-step transformation involving substrates tethered with natural products and drug candidates, providing the complex homologated adducts in high yields with excellent stereocontrol. The cytotoxicity and cellular uptake experiments revealed that the enantiopurity of this novel class of polycyclic aminopentafulvenes has significant effects on their photophysical properties and cell viability. In chapter 4, we have disclosed a L-tert-leucine dipeptide thiourea catalyzed enantioselective (4+1) carbospiroannulation between cyclohexenylidene malononitriles and cyclopent-4-ene-1,3-diones. The developed protocol can successfully be applied to a wide range of cyclopent-4-ene-1,3-diones and a few cyclohexenylidene malononitriles and the carbospiroannulated adducts could be achieved in good to high yields with moderate to excellent stereocontrol. Pleasingly, this methodology realizes an unprecedented enantioselective vinylogous carbospiroannulation for the formation of chiral [4,4]nonane frameworks. Chapter 4 deals with the development of a catalytic asymmetric vinylogous Michael addition of 3-cyano-4-methylcoumarins by using maleimides as Michael acceptor. Catalyzed by L- *tert*-leucine based thiourea, this reaction led to the densely functionalized product in good yields with moderate to

excellent enantioselectivities. This report represents the first non-covalent organocatalytic asymmetric vinylogous Michael addition reaction of 3-cyano-4-methylcoumarins. In chapter 5, an efficient base catalyzed methodology for the synthesis of 3-carboxamidecoumarins tethered α-Trifluoromethyl styrenes by reacting 3-cyano-4 methylcoumarin and trifluoromethyl ketones has been developed. The corresponding products could be achieved in satisfactory yields and good to excellent stereoselectivies.