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学位論文題目	Development of Novel Methodology for the Synthesis of Heterocyclic-Fluorine containing Compounds under Catalytic and Non-Catalytic Conditions （触媒あるいは非触媒下における含フッ素複素環化合物の新規合成法の開発）
論文審査委員	主査　教授　山下　啓司 教授　柴田　哲男 教授　林　秀敏 （名古屋市立大学）

論文内容の要旨

Fluorine substituted synthetic organic molecules gained special attention during the last few decades as it either improves or alter the physical, chemical and biological properties of original molecules. On the other hand, in synthetic organic chemistry, construction of medium size carbo- and hetero-cyclic rings in an efficient way remains a challenging research theme because such rings form the structural core of a large number of biologically active natural products and medicinally important synthetic compounds. This thesis describes our exploration in the construction of fluorinated small and medium-sized heterocyclic ring skeletons via transition metal catalyzed decarboxylative pathway. Finally, we have developed a synthetic methodology for the construction of trifluoromethanesulfonyl pyrazole by using diazo-triflone as a building block.

Chapter 1 mainly focused on the general introduction of palladium catalyzed decarboxylation reactions to construct various cyclic and acyclic molecules via palladium-π-allyl complex formation.

In chapter 2, we have disclosed a novel synthetic methodology to construct trifluoromethyl-substituted nine-membered ring structures via [5+4] cycloaddition reaction under palladium catalysis. The idea for our novel [5+4] cycloaddition reaction consists of double decarboxylation of two independent substrates viz. trifluoromethyl benzoxazinanones and vinylethylene carbonates involving *in-situ* generated Pd-π-allyl intermediate. A variety of trifluoromethyl benzoxazinanones and vinylethylene carbonates were smoothly transformed to trifluoromethyl-substituted tetrahydrobenzoxazonine in good to excellent yields.

Chapter 3 mainly focused on a novel method to synthesize diversely substituted CF_3-indolines from trifluoromethyl benzoxazinanones and sulfur ylides under palladium catalysis in a highly diastereoselective fashion. A series of CF_3-benzoxazinanones was smoothly reacted with sulfur ylides under palladium catalysis in the presence of tricyclohexylphosphine to provide pharmaceutically attractive variously substituted CF_3-indolines in high to excellent yields with high diastereoselectivities.

In chapter 4 we mainly focused on the synthesis of pyrazole triflones via the reaction of 2-diazo-1-phenyl-2-((trifluoromethyl)sulfonyl)ethan-1-one and differently substituted nitrostyrenes under basic reaction conditions. The protocol is the first report to introduce a trifluoromethyl sulfonyl group at the 3-position of pyrazole ring by using diazo-triflone as a building block.

Chapter 5 mainly focused on the synthesis of 5-nitro-pyrazole triflones via [3+2] cycloaddition reaction and its further application for potential insecticide.

In chapter 6, summary of the thesis was described.