Ti (III) catalyzed synthesis of exocyclic allenes and development of new titanocene complexes /

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Monografía

This thesis deals with the Ti(III) catalyzed synthesis of exocyclic allenes and the development of new titanocene complexes. It is structured in six chapters:Chapter 1 is a general introduction about the generation and reactivity of [TiCp2Cl]. A review about the different [TiCp2Cl] catalyzed or promoted reactions reported to date, is included.Chapter 2 is divided in introduction and results and discussion. In the introduction, anoverview about the promoted or catalyzed methodologies, already available for the synthesis of α-allenols, is included. Results and discussion section is sub-divided in other two parts. The first one describes the synthesis of carbocyclic or nitrogen heterocyclic precursors bearing a propargyl halide and a carbonyl group. Next, the [TiCp2Cl] catalyzed synthesis of exocyclic allenols is studied. Additionally, a mechanistic study through deuterium incorporation and reaction of secondarypropargyl halide precursors is performed. In the second part, the preparation of oxygen precursors and its cyclization are studied.In chapter 3, there is an introduction about enantioselective syntheses using chiral titanocene catalysts. The results and discussion section deals with the enantioselective cyclization of some of the precursors previously prepared using precatalyst (R,R)-ethylenebis(4,5,6,7-tetrahydro-1-indenyl) titanium(IV), also known as Brintzinger complex. A formal synthesis of the alkaloid (})-stemoamide is carried out in chapter 4, being the key step for this synthesis the generation of an α-allenol derivative through a [TiCp2Cl] catalyzedcyclization. This chapter is also divided in an introduction, in which a review about the different methods of synthesis of stemoamide are included, and results and discussion.Chapter 5 compiles the project that I have developed during my research stay at University of Bonn, under the supervision of Prof. Gansauer. The target of this project was to develop an azide functionalized short linker titanocene derivative with the aim of linking it to a surface by click chemistry. The chapter is divided in introduction and results and discussion. In the introduction the different methodologies for the synthesis of titanocene derivatives are reviewed. This section also includes the background in which this project is based.Chapter 6 includes the experimental part. In this chapter all reactions performed during the development of this thesis, as well as the spectroscopic characterization of compounds, are compiled.

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