

Chapter 1 : Organic Chemistry – UTSA Chemistry

aspects of small ring chemistry, with emphasis being placed on the application in organic synthesis. "State of the art" reviews will be presented by individual authors.

The strain associated with the structure of these compounds is in general responsible for the good reactivity of the carbon-carbon double bond, even in sterically encumbered substrates. The key role of these compounds in organic synthesis resides in the exclusive, and by far the most useful, possibility of the introduction of a cyclopropyl ring in a more complex molecule through reactions onto the exocyclic double bond. For this reason the investigations on the synthesis and reactivity of these compounds has exploded in the last decade. This article will deal with all the reactions which involve the exocyclic double bond of alkylidenecyclopropanes in cycloaddition processes. Albeit most of the reactions are true pericyclic cycloaddition processes, also those reactions which lead to cycles by stepwise processes will be reported with comments on the mechanism. Cycloadditions onto Methylene- and Alkylidenecyclopropane Derivatives

1 Introduction Methylene- and alkylidenecyclopropanes are highly strained molecules, but at the same time most of them are surprisingly stable to allow their use in many synthetic applications. Being multifunctional reagents with high energy, they offer enormous potential in organic syntheses that has been only partially disclosed in the last decades. In the last decade other selective syntheses have been developed which gave easy access to compounds containing specific substitution patterns [3], to optically active derivatives [4], or more sophisticated derivatives like dicyclopropylideneethane 2 [5], bicyclopropylidene BCP 3 [6] and chloromethoxy carbonylmethylenecyclopropane 4 [1-7]. The highly strained nature of methylene- and alkylidenecyclopropanes has been evidenced by spectroscopic measurements and X-ray analysis. The presence of the exocyclic double bond imposes a lengthening of the C₂-C₃ bond as a result of an increase of the C₂-C₁-C₃ angle compared to cyclopropane. This chemistry, usually referred to as "TMM chemistry", has been the object of many studies and thoroughly reviewed by Binger and Biich [2] and Trost [8]. The second large group of reactions is characterised by the elaboration of the exocyclic double bond. This chemistry is particularly interesting because it allows the incorporation of a cyclopropyl ring in a more complex molecule, to take advantage of its peculiar feature as a reactive three-carbon functional group. Among these reactions, the cycloadditions play a central role because of their general application, and for the unequalled property of the possible introduction of two new carbon-carbon or carbon-heteroatom bonds in the same step or process. Despite the study of cycloadditions to alkylidenecyclopropanes being far from fully exploited, the results accumulated in the literature are enough for a comprehensive review which will present the state of the art. The present review will deal with all the processes of cycloadditions, without discriminating between the nature, concerted or stepwise, of the processes. It will cover those reactions which involve exclusively the exocyclic double bond of methylenecyclopropane and alkylidenecyclopropanes in the formation of three-, four-, five- and six-membered rings. Moreover, the few examples reported deal with rather sophisticated substrates such as 2,2-difluoromethylenecyclopropane, diarylmethylenecyclopropanaphthalenes, bicyclopropylidene and diacylmethylene cyclopropanes. The substitution of two, or more, ring protons with fluorine atoms, however, seems to improve dramatically the dienophilic reactivity of the exocyclic double bond. With cyclopentadiene 6 and furan 7, it formed two isomeric adducts Scheme 1 [9]. In both cases the adduct with the endo CF₂ group is the major isomer. Diphenylisobenzofuran 10 reacted with 5 to give only the endo isomer 11 Scheme 2 [9].

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