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[4+2] and [4+3] cycloaddition reactions and Lewis acid catalyzed cycloisomerisation of malonyl epoxides

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Donor-acceptor cyclopropanes have been extensively used in synthetic chemistry in [3+2] and [3+3] cycloaddition reactions for the preparation of highly substituted carbo- and heterocyclic products. This methodology is further extended to donor-acceptor cyclobutane in [4+2] and [4+3] cycloaddition reactions for the synthesis of highly substituted carbo- and heterocyclic products. Initial work carried out makes use of cyclobutanes substituted with a metal-alkyne complex towards the synthesis of tetrahydropyrans in good yields and with acceptable diastereoselectivity. The initial aim of the project was to improve and expand the scope of the previous work carried out within the group on [4+2] cycloaddition reaction. For example, [4+2] and [4+3] cycloaddition reaction were carried out by using diester cyclobutanes having an alkene and phenyl π -donors. The [4+3] cycloaddition reaction of cyclobutane with nitrene did not work but [4+2] cycloaddition was successful when aldehydes were used as trapping reagents. Lower yields of the cycloadducts were observed due to formation of (\pm)-dimethyl-2-methyl-6-phenylcyclohex-3-ene-1,1-dicarboxylate and 2,6-diphenyl-4,8-dipropenylcyclooctane-1,1,5,5-tetracarboxylic acid tetramethyl ester. During the synthesis of a precursor cyclobutane a novel cycloisomerisation of malonyl epoxide under Lewis acidic conditions to 6,8-dioxabicyclo[3.2.1]octane derivatives was developed. This reaction has opened a new pathway for the synthesis of 6,8-dioxabicyclo[3.2.1] octanes in a diastereoselective fashion using malonyl epoxides as precursors. A wide range of malonyl epoxides were cycloisomerised under Lewis acidic conditions and the cycloisomerisation of syn and anti malonyl epoxides were stereospecific. The diastereoselectivity of the process was proven by nOe and X-ray analysis. The cycloisomerisation of malonyl diepoxide has also been investigated towards the formation of 5,5-dimethoxy-6,6,8,8-tetraoxa4,4-spirobi[bicyclo[3.2.1]octane].

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