

X-ray Investigations of Bicyclic α -methylene- δ -valerolactones

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The α -methylene- δ -valerolactones moiety is present in various biologically active natural compounds, *e.g.* vernolepin, vernomenin, pentalenolactone *E*, teucriumlactone, artemisitene and crassin. However, work on isolation and synthesis of new α -methylene- δ -valerolactones has not led to a significant number of crystal structure investigations. A search of the CSD (version 5.26) shows that system in which δ -valerolactone ring is condensed with the cyclohexane moiety along the individual C δ -C γ single bond is unique among crystal structures examined to date. Investigated compounds represent a novel group of the optically active α -methylene- δ -valerolactones synthesized in a highly stereoselective Michael reaction. Recently we reported crystal structures of two compounds *i.e.* the 3-methylene-2-oxohexahydrochromene-4a-carboxylic acid ethyl ester [1] and the 4a-methyl-3-methylene-octahydro-chromen-2-one [2]. The six following crystal structures will be shown in detail. In all compounds the δ -valerolactone rings adopt a half-chair conformation. The highly polar character of the carbonyl group hinders π electron density delocalization within the O=C-C=C moiety. In the crystal, molecular conformation is stabilized by attractive interactions between the oppositely charged atoms. The mechanism of interactions has been investigated using *NBO* theory at the *MP2/6-31+G(d,p)* level.

[1] Krawczyk H., Śliwiński M., Wolf W.M., Bodalski R., *Synlett*, 2004, 1995.

[2] Krawczyk H., Śliwiński M., Wolf W.M., *Acta Cryst.*, 2004, **C60**, o897.

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