Acta Crystallographica Section E
Structure Reports Online
ISSN 1600-5368

5-[(Cyclohexylamino)methylene]-2,2-dimethyl-1,3-dioxane-4,6-dione

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Key indicators

Single-crystal X-ray study
T = 297 K
Mean σ(C–C) = 0.006 Å
R factor = 0.057
wR factor = 0.204
Data-to-parameter ratio = 7.7

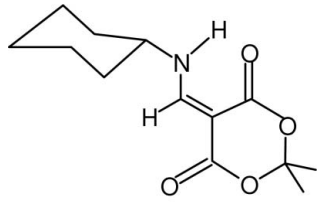
For details of how these key indicators were automatically derived from the article, see http://journals.iucr.org/e.

Received 5 June 2006
Accepted 10 July 2006

In the title enaminone, $C_{13}H_{19}N_1O_4$, the Meldrum's acid group has a distorted boat conformation. The two independent molecules of the asymmetric unit are dimerized about a pseudo inversion center through two intermolecular N—H···O hydrogen bonds, with N···O distances of 3.049 (4) and 3.093 (4) Å. The dimers are stacked along the [010] direction.

Comment

The synthesis of functionalized enaminones is a theme of ongoing interest owing to their potential in the synthesis of heterocyclic compounds (Negri *et al.*, 2004; Lue & Greenhill, 1997; Kuckländer, 1994; Ferraz & Pereira, 2004). A particular class of such compounds is derived from Meldrum's acid and the solid-state structural study of such derivatives has been described (Vencato *et al.*, 2004; Cunha *et al.*, 2003; Silva *et al.*, 2006; Silva *et al.*, 2005*a*,*b*, Joussef *et al.*, 2005*a*,*b*). Here we report our structural study of one enaminone, (I), derived from Meldrum's acid.



(I)

The structure has two independent molecules in the asymmetric unit (Fig. 1). Selected bond distances, angles and torsion angles are given in Table 1. In both molecules, the Meldrum's acid group has a distorted boat conformation, indicated by the Cremer & Pople (1975) parameters: O5*A*→C4*A*→ ···→C6*A* [Q = 0.429 (4) Å, θ = 61.6 (5)° and φ = 297.7 (6)°] and O5*B*→C4*B*→ ···→C6*B* [Q = 0.436 (4) Å, θ = 63.3 (5)° and φ = 298.4 (5)°].

There are small differences between the two molecules, as can be seen in the bond angles O7—C6—C12 of 106.7 (4) and 104.5 (3)° for molecules *A* and *B*, respectively. The bond distance O10*A*—C8*A* of 1.219 (4) Å is marginally longer than the corresponding O10*B*—C8*B* distance of 1.204 (4) Å, presumably due to the fact that atom O10*A* is involved in a hydrogen bond as acceptor.

There are two intramolecular hydrogen bonds (Table 2), providing two flattened H1—N1—C2—C3—C8—O10 rings for molecules *A* and *B*. The two independent molecules of the asymmetric unit are dimerized about a pseudo-inversion center through two intermolecular hydrogen bonds (Table 2). It is worth noting that atoms H1*A* and H1*B* belong to bifur-

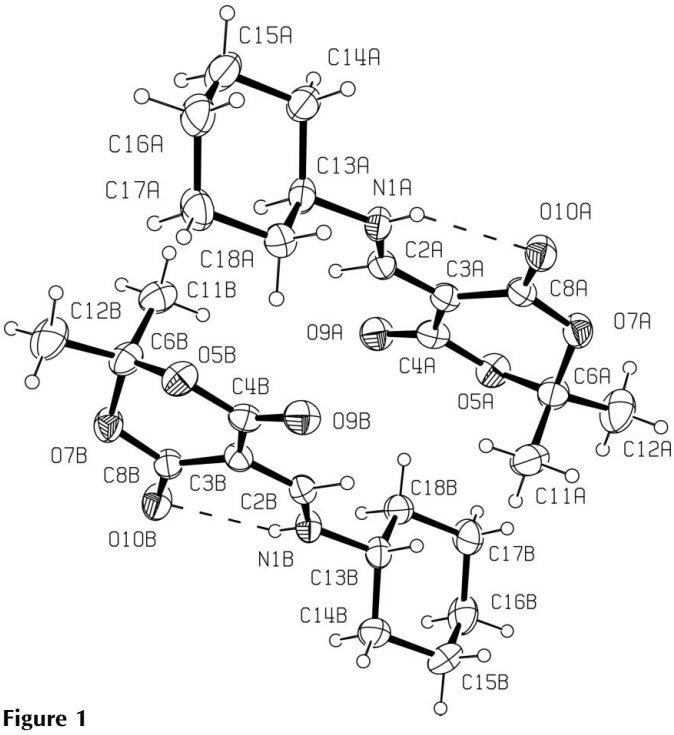


Figure 1
The asymmetric unit of (I), with the atom-numbering schemes for molecules *A* and *B*. Displacement ellipsoids are drawn at the 30% probability level. Dashed lines indicate hydrogen bonds.

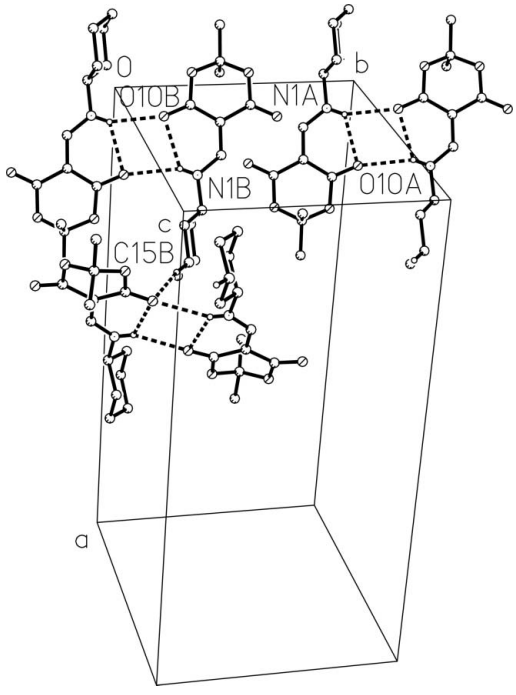


Figure 2
Hydrogen bonding of (I). Intra- and intermolecular hydrogen bonds are shown as dashed lines. Only the H atoms involved in hydrogen bonds are shown.

cated intra- and intermolecular hydrogen bonds. The hydrogen bonding is shown in Fig. 2. The molecular packing of (I) is shown in Fig. 3, with the geometrical parameters in Table 2. The non-classical intermolecular C—H···O hydrogen bond completes the packing, with the dimers stacked parallel to the [010] direction.

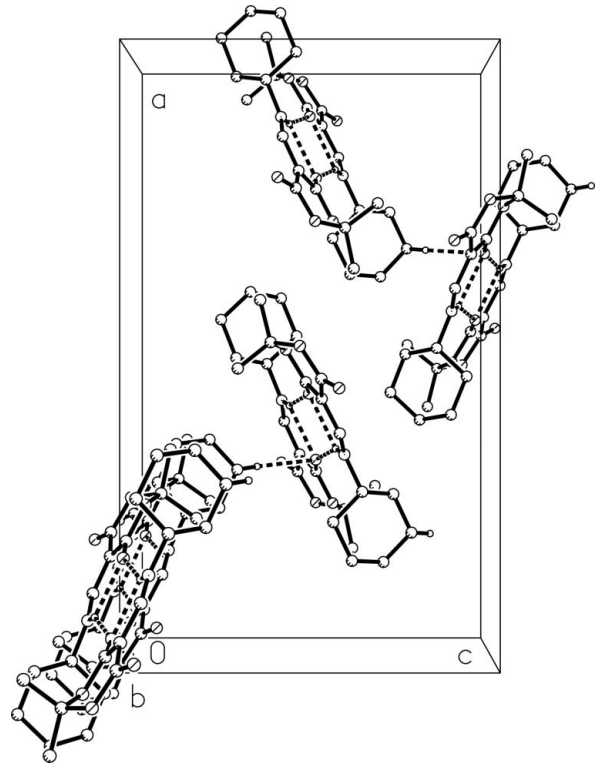


Figure 3
Partial packing diagram of (I), viewed down the *b* axis. Dashed lines indicate hydrogen bonds.

Experimental

A solution of 288 mg (2.0 mmol) of Meldrum's acid in 2 ml (26.1 mmol) of trimethyl orthoformate was heated at reflux for 3 h after which time the solvent was evaporated. The yellow solid that formed was added to 2.0 mmol of cyclohexylamine in 5 ml of CH_2Cl_2 and the solution was allowed to stand at room temperature with stirring for 72 h. The solvent was evaporated and the crude residue was purified by silica gel 60 (70–230 mesh) column chromatography (hexane/ethyl acetate 50%), afforded (I) as a yellow solid (yield 116.7 mg, 23%; m.p. 421–422 K). Single crystals were obtained by slow evaporation of a solution of (I) in CH_3Cl at room temperature.

Crystal data

$C_{13}H_{19}NO_4$
M_r = 253.29
Orthorhombic, $Pca2_1$
a = 20.905 (2) Å
b = 10.222 (1) Å
c = 12.516 (6) Å
V = 2674.6 (13) Å^3
Z = 8
D_x = 1.258 Mg m^{-3}
Cu $K\alpha$ radiation
μ = 0.77 mm^{-1}
T = 297 (2) K
Prism, yellow
0.35 × 0.35 × 0.30 mm

Data collection

Enraf–Nonius CAD-4 diffractometer
$\omega/2\theta$ scans
Absorption correction: none
2698 measured reflections
2558 independent reflections
2404 reflections with $I > 2\sigma(I)$
R_{int} = 0.025
θ_{max} = 68.0°
2 standard reflections
frequency: 120 min
intensity decay: 1%

Refinement

Refinement on F^2	$w = 1/[\sigma^2(F_o^2) + (0.1659P)^2 + 0.0648P]$ where $P = (F_o^2 + 2F_c^2)/3$
$R[F^2 > 2\sigma(F^2)] = 0.057$	$(\Delta/\sigma)_{max} = 0.005$
$wR(F^2) = 0.204$	$\Delta\rho_{max} = 0.39$ e Å^{-3}
$S = 1.20$	$\Delta\rho_{min} = -0.40$ e Å^{-3}
2558 reflections	Extinction correction: *SHELXL97*
332 parameters	Extinction coefficient: 0.054 (4)
H atoms treated by a mixture of independent and constrained refinement	

Table 1
Selected geometric parameters (Å, °).

N1*A*—C2*A*	1.294 (4)	N1*B*—C2*B*	1.287 (5)
N1*A*—C13*A*	1.469 (4)	N1*B*—C13*B*	1.472 (4)
C4*A*—O9*A*	1.208 (5)	C4*B*—O9*B*	1.213 (4)
C4*A*—O5*A*	1.373 (5)	C4*B*—O5*B*	1.368 (5)
O5*A*—C6*A*	1.441 (5)	O5*B*—C6*B*	1.429 (5)
C6*A*—O7*A*	1.435 (4)	C6*B*—O7*B*	1.441 (5)
O7*A*—C8*A*	1.364 (4)	O7*B*—C8*B*	1.373 (4)
C8*A*—O10*A*	1.219 (4)	C8*B*—O10*B*	1.204 (4)
C2*A*—N1*A*—C13*A*	123.1 (3)	C2*B*—N1*B*—C13*B*	122.8 (3)
O7*A*—C6*A*—C12*A*	106.7 (4)	O7*B*—C6*B*—C12*B*	104.5 (4)
C18*A*—C13*A*—C14*A*	111.1 (3)	C18*B*—C13*B*—C14*B*	111.8 (3)
C15*A*—C16*A*—C17*A*	112.7 (4)	C15*B*—C16*B*—C17*B*	111.4 (4)

Table 2
Hydrogen-bond geometry (Å, °).

D—H···*A*	*D*—H	H···*A*	*D*···*A*	*D*—H···*A*
N1*A*—H1*A*···O10*A*	0.80 (5)	2.27 (5)	2.791 (4)	124 (4)
N1*B*—H1*B*···O10*B*	0.77 (5)	2.27 (5)	2.796 (4)	126 (4)
N1*A*—H1*A*···O10*B*^{i}	0.80 (5)	2.31 (5)	3.049 (4)	154 (4)
N1*B*—H1*B*···O10*A*^{ii}	0.77 (5)	2.38 (5)	3.093 (4)	154 (4)
C15*B*—H15*D*···O10*A*^{iii}	0.97	2.58	3.536 (6)	170

Symmetry codes: (i) $x, y+1, z$; (ii) $x, y-1, z$; (iii) $-x+\frac{1}{2}, y-1, z+\frac{1}{2}$.

H atoms bonded to atoms N1*A* and N1*B* were found in a difference map and their positions were refined freely, with U_{iso}(H) = 1.2U_{eq}(N). All other H atoms were placed in calculated positions (C—H = 0.96 and 0.97 Å) and treated as riding atoms [U_{iso}(H) = 1.2 U_{eq}(C) or 1.5 U_{eq}(methyl C)]. Due to the absence of any significant anomalous scatterers in the compound, Friedel pairs were merged.

Data collection: *CAD-4-PC* (Enraf–Nonius, 1993); cell refinement: *CAD-4-PC*; data reduction: *XCAD4* (Harms & Wocadlo, 1995); program(s) used to solve structure: *SHELXL97* (Sheldrick, 1997); program(s) used to refine structure: *SHELXL97* (Sheldrick, 1997); molecular graphics: *ORTEP-3 for Windows* (Farrugia, 1997); software used to prepare material for publication: *WinGX* (Farrugia, 1999).

The authors gratefully acknowledge the financial support of Conselho Nacional de Desenvolvimento Científico e Tecnológico–CNPq (Edital Universal-2002, Processo 474592/03–0), Fundação de Amparo à Pesquisa do Estado da Bahia – FAPESB (Edital Fluxo Contínuo-2004, Processo 1431040046342 and Edital PRODOC-2004 Processo 1331040048671) and Fundação de Apoio à Pesquisa–FUNAPE/UFG. We also thank CAPES for a fellowship to JF, and CNPq for research fellowships to IV and SC.

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