4-Acetonylidene-1-ethyl-2,3,4,5-tetrahydro-1H-1,5-benzodiazepin-2-one

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The title compound, $C_{14}H_{16}N_2O_2$, contains a diazepine ring, which appears in a boat conformation. An intramolecular hydrogen bond is formed between the NH group of the diazepine ring and a carbonyl O atom of one of the side chains.

Comment

As part of our research on the use of 4-hydroxy-6-methyl-pyran-2-one in heterocyclic synthesis (El Abassi *et al.*, 1987, 1997; Essassi *et al.*, 1987), we have reinspected the condensation of *o*-phenylenediamine with γ-pyrone with the aim of confirming the structure of the obtained product. In a previous work (El Abassi *et al.*, 1987), we have shown that the reaction of *o*-phenylenediamine with γ-pyrone leads to 4-acetonylidene-1,5-benzodiazepin-2-one. In order to confirm the structure of the reaction product, we have converted 4-acetonylidene-1,5-benzodiazepin-2-one with ethyl bromide to afford the title compound, (I), and carried out an X-ray structure analysis.

(I)
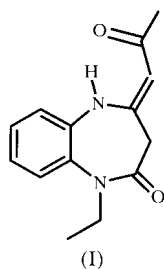
The geometry of the title compound shows no unusual features. The diazepine ring shows a boat conformation. The seven-membered ring can be described as being composed of three planes: a bow plane (C2, C3 and C4), a central plane (N1, C2, C4 and N5; r.m.s. deviation 0.019 Å) and a stern plane (N1 C11, C6 and N5; r.m.s. deviation 0.013 Å). The dihedral angle between central and bow planes is 58.70 (8)°, and there is an angle of 36.31 (5)° between central and stern planes.

The title compound shows an intramolecular hydrogen bond between the NH group of the diazepine ring and a carbonyl O atom of one of the side chains. Furthermore, the other carbonyl O atom shows three short C—H⋯O contacts.

Experimental

To a solution of 4-acetonylidene-1,5-benzodiazepin-2-one (1 mmol), in benzene (60 ml), tetra-*n*-butylammonium bromide (20 mmol) in NaOH (30 ml) was added as an alkylating agent. After dilution and decantation, the organic phase was washed with a hydrochloric acid solution (10%) and dried. The benzene was evaporated under reduced pressure and the residue was chromatographied on a silicate column with hexane as eluant. Analysis measured (calculated): C 68.83 (68.85), H 6.52 (6.56), N 11.43% (11.47%).

Crystal data

$C_{14}H_{16}N_2O_2$	$D_x = 1.249$ Mg m^{-3}
$M_r = 244.29$	Mo $K\alpha$ radiation
Monoclinic, $P2_1/n$	Cell parameters from 8192
$a = 8.730$ (1) Å	reflections
$b = 10.052$ (1) Å	$\theta = 1–25°$
$c = 14.998$ (2) Å	$\mu = 0.085$ mm^{-1}
$\beta = 99.36$ (1)°	$T = 173$ (2) K
$V = 1298.6$ (3) Å^3	Block, colourless
$Z = 4$	$0.60 \times 0.40 \times 0.30$ mm

Data collection

Siemens CCD three-circle diffractometer	$R_{int} = 0.034$
	$\theta_{max} = 28.70°$
ω scans	$h = -11 \rightarrow 11$
Absorption correction: empirical	$k = -13 \rightarrow 13$
(*SADABS*; Sheldrick, 1996)	$l = -20 \rightarrow 20$
$T_{min} = 0.951$, $T_{max} = 0.975$	8192 standard reflections
28 486 measured reflections	frequency: 1080 min
3350 independent reflections	intensity decay: none
2865 reflections with $I > 2\sigma(I)$	

Refinement

Refinement on F^2	$w = 1/[\sigma^2(F_o^2) + (0.0635P)^2$
$R[F^2 > 2\sigma(F^2)] = 0.039$	$+ 0.2665P]$
$wR(F^2) = 0.113$	where $P = (F_o^2 + 2F_c^2)/3$
$S = 1.068$	$(\Delta/\sigma)_{max} < 0.001$
3350 reflections	$\Delta\rho_{max} = 0.29$ e Å^{-3}
164 parameters	$\Delta\rho_{min} = -0.26$ e Å^{-3}
H-atom parameters constrained	

Table 1
Hydrogen-bonding geometry (Å, °).

$D—H\cdots A$	$D—H$	$H\cdots A$	$D\cdots A$	$D—H\cdots A$
C10—H10⋯O41[i]	0.95	2.52	3.3367 (13)	144
C21—H21⋯O41[ii]	0.95	2.58	3.4457 (14)	152
C3—H3A⋯O41[ii]	0.99	2.66	3.5948 (14)	157
N1—H1⋯O22	0.88	1.98	2.6640 (12)	133

Symmetry codes: (i) $1 + x, y, z$; (ii) $-x, 1 - y, 1 - z$.

All H atoms were initially located by difference Fourier synthesis. Subsequently their positions were idealized and constrained to ride on their parent atoms with C—H(aromatic) = 0.95, C—H(secondary) = 0.99, C—H(methyl) = 0.98 or N—H = 0.88 Å, and fixed individual displacement parameters [U(H) = $1.5U_{eq}$(methyl C),

$U(H) = 1.2U_{eq}(C)$ or $U(H) = 1.2U_{eq}(N)$]. The C23 methyl group was allowed to rotate about its local threefold axis.

Data collection: *SMART* (Siemens, 1995); cell refinement: *SMART*; data reduction: *SAINT* (Siemens, 1995); program(s) used to solve structure: *SHELXS*97 (Sheldrick, 1990); program(s) used to refine structure: *SHELXL*97 (Sheldrick, 1997).

References

El Abbassi, M., Essassi, E. M. & Fifani, J. (1987). *Tetrahedron Lett.* **28**, 1389–1390.

El Abbassi, M., Essassi, E. M. & Fifani, J. (1997). *Bull. Soc. Chim. Belg.* **106**, 205–210.

Essassi, E. M., El Abbassi, M. & Fifani, J. (1987). *Bull. Soc. Chim. Belg.* **96**, 225–228.

Sheldrick, G. M. (1990). *Acta Cryst.* A**46**, 467–473.

Sheldrick, G. M. (1996). *SADABS*. University of Göttingen, Germany.

Sheldrick, G. M. (1997). *SHELXL*97. University of Göttingen, Germany.

Siemens (1995). *SMART* and *SAINT*. Siemens Analytical X-ray Instruments Inc., Madison, Wisconsin, USA.