

6-Ethyl-5,7-dimethyl-6*H*-1,4-diazepine-2,3-dicarbonitrile

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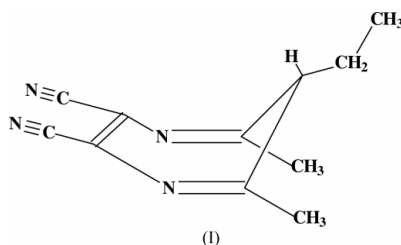
Key indicators

Single-crystal X-ray study
 $T = 295$ K
Mean $\sigma(\text{C}-\text{C}) = 0.002$ Å
 R factor = 0.043
 wR factor = 0.111
Data-to-parameter ratio = 17.1For details of how these key indicators were
automatically derived from the article, see
<http://journals.iucr.org/e>.

In the title compound, $\text{C}_{11}\text{H}_{12}\text{N}_4$, the diazepine ring exhibits a boat conformation, where the displacements of the C atoms attached to the nitrile groups and the C atom attached to the ethyl group are 0.591 (1), 0.585 (1) and 0.845 (1) Å from the base of the boat.

Comment

The present work is a continuation of our investigation of the chemical properties of diaminomaleonitrile (Sidorov *et al.*, 1999; Nesterov *et al.*, 2001). We have synthesized 6-ethyl-5,7-dimethyl-6*H*-1,4-diazepine-2,3-dicarbonitrile, (I), and investigated its structure (Fig. 1). Although such 1,4-diazepine derivatives have been known for a long time (Begland *et al.*, 1974; Ohtsuka, 1976), their structures have not yet been sufficiently investigated. In the Cambridge Structural Database (CSD; Allen, 2002), we found only two examples (Donzello *et al.*, 1999; Mague & Eduok, 2000). Such compounds can be used as starting materials in the synthesis of bi- and triheterocyclic systems (Essaber *et al.*, 1998). 5,7-Diphenyl-1,4-diazepine has been used as a dicyano monomeric precursor for the synthesis of a new class of porphyr-azine macrocycles (Donzello *et al.*, 1999). According to literature data (Begland *et al.*, 1974), such compounds can be readily oxidized to the corresponding dihydrodiazepines.



The geometric parameters in (I) are very similar to those found in 5,7-dimethyl-6*H*-1,4-diazepine-2,3-dicarbonitrile and 5,7-diphenyl-6*H*-1,4-diazepine-2,3-dicarbonitrile (Mague & Eduok, 2000; Donzello *et al.*, 1999). The diazepine ring in (I) exhibits a boat conformation. The displacements of atoms C2, C3 and C6 from the mean N1/N4/C5/C7 plane are 0.591 (1), 0.585 (1) and 0.845 (1) Å, respectively. The dihedral angles between the N1/N4/C5/C7 mean plane and the C5/C6/C7 and N1/C2/C3/N4 planes are 60.2 (1) and 32.0 (1)°, respectively. The ethyl substituent in (I) has an equatorial orientation, as can be seen in Fig. 1.

The bond lengths in the diazepine ring have values intermediate between single- and double-bond values (Allen *et al.*, 1987). This indicates that π -conjugation exists in this fragment of the molecule, despite the fact that the diazepine ring is not planar.

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Experimental

The title compound, (I), was obtained according to the following procedure, which is different from the literature method of Begland *et al.* (1974): diaminomaleonitrile (1.08 g, 0.010 mol) and 3-ethyl-2,4-pentanedione (1.34 ml, 0.010 mol) were heated to boiling in ethanol solution (20 ml), in the presence of a catalytic amount of acetic acid. The precipitate which formed was separated and recrystallized from ethanol (30 ml) [m.p. 461 K; yield 1.58 g (79%)]. Crystals were obtained by isothermal evaporation from an ethanol solution.

Crystal data

$C_{11}H_{12}N_4$	$D_x = 1.208 \text{ Mg m}^{-3}$
$M_r = 200.25$	Mo $K\alpha$ radiation
Monoclinic, $P2_1/n$	Cell parameters from 24 reflections
$a = 7.4160 (15) \text{ \AA}$	$\theta = 10\text{--}11^\circ$
$b = 13.344 (3) \text{ \AA}$	$\mu = 0.08 \text{ mm}^{-1}$
$c = 11.141 (2) \text{ \AA}$	$T = 295 (2) \text{ K}$
$\beta = 92.90 (3)^\circ$	Prism, colorless
$V = 1101.1 (4) \text{ \AA}^3$	$0.40 \times 0.30 \times 0.20 \text{ mm}$
$Z = 4$	

Data collection

Enraf–Nonius CAD-4 diffractometer	$\theta_{\text{max}} = 27.0^\circ$
$\theta/2\theta$ scans	$h = 0 \rightarrow 9$
Absorption correction: none	$k = 0 \rightarrow 17$
2560 measured reflections	$l = -14 \rightarrow 14$
2381 independent reflections	3 standard reflections every 97 reflections
1380 reflections with $I > 2\sigma(I)$	intensity decay: 3%
$R_{\text{int}} = 0.031$	

Refinement

Refinement on F^2	H-atom parameters constrained
$R[F^2 > 2\sigma(F^2)] = 0.043$	$w = 1/[\sigma^2(F_o^2) + (0.045P)^2]$
$wR(F^2) = 0.111$	where $P = (F_o^2 + 2F_c^2)/3$
$S = 1.18$	$(\Delta/\sigma)_{\text{max}} < 0.001$
2381 reflections	$\Delta\rho_{\text{max}} = 0.13 \text{ e \AA}^{-3}$
139 parameters	$\Delta\rho_{\text{min}} = -0.12 \text{ e \AA}^{-3}$

Table 1

Selected geometric parameters (\AA , $^\circ$).

N1—C7	1.288 (2)	C2—C3	1.367 (2)
N1—C2	1.376 (2)	C5—C6	1.511 (2)
N4—C5	1.290 (2)	C6—C7	1.504 (2)
N4—C3	1.370 (2)		
C7—N1—C2	120.58 (14)	N4—C5—C6	120.99 (15)
C5—N4—C3	120.18 (14)	C7—C6—C5	99.57 (12)
C3—C2—N1	125.88 (15)	N1—C7—C6	120.52 (15)
C2—C3—N4	126.26 (15)		
C7—N1—C2—C3	36.5 (2)	N4—C5—C6—C7	76.76 (18)
N1—C2—C3—N4	−0.3 (3)	C2—N1—C7—C6	11.1 (2)
C5—N4—C3—C2	−36.1 (2)	C5—C6—C7—N1	−76.88 (17)
C3—N4—C5—C6	−10.7 (2)		

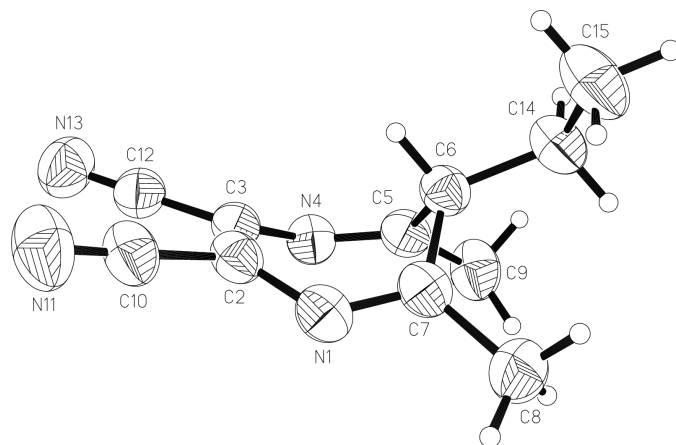


Figure 1

A view of compound (I), showing the atom numbering used. The non-H atoms are shown with displacement ellipsoids drawn at the 50% probability level.

Data collection: *CAD-4 Software* (Enraf–Nonius, 1989); cell refinement: *CAD-4 Software*; data reduction: *SHELXTL* (Sheldrick, 1994); program(s) used to solve structure: *SHELXS97* (Sheldrick, 1990); program(s) used to refine structure: *SHELXL97* (Sheldrick, 1997); molecular graphics: *SHELXTL*; software used to prepare material for publication: *SHELXL97*.

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