

the dihedral angle between π orbitals that may have to interact [*i.e.*, those spanning C(3i)—C(4i)—C(9i)—N(1i) and spanning C(5)—C(4)—N(3)—C(2)] is 133.4° in (1) and 67.8° in (2). If the predominant solution conformations of (1) and (2) resemble the conformations in the crystal, then this stereoelectronic effect might be responsible for differences in photosplitting efficiencies of (1) and (2).

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Structure of 9-Benzyl-1,3-dimethyl-2,4,8-trioxo-1,3,6,7,9-pentahydro-1,3-diazino[2,1-f]purine

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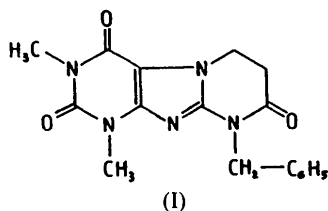
(Received 13 November 1990; accepted 24 January 1991)

Abstract. C₁₇H₁₇N₅O₃, $M_r = 339.35$, orthorhombic, *Pbca*, $a = 7.521$ (1), $b = 38.635$ (4), $c = 11.008$ (1) Å, $V = 3198.6$ (6) Å³, $Z = 8$, $D_m = 1.40$ (2), $D_x = 1.409$ g cm⁻³, $\lambda(\text{Cu } K\alpha) = 1.54178$ Å, $\mu = 7.88$ cm⁻¹, $F(000) = 1424$, $T = 293$ K, final $R = 0.0514$ for 1157 observed reflections. The purine fused-ring system is planar. The six-membered 1,3-diazinone ring adopts a half-chair conformation. Theophylline skeletons show significant stacking with mean separation between molecular planes of 3.34 (4) Å.

Introduction. Theophylline, as natural methylxanthine, is a well known pharmacological agent of

great therapeutic interest. The structure determination of 9-benzyl-1,3-dimethyl-2,4,8-trioxo-1,3,6,7,9-pentahydro-1,3-diazino[2,1-f]purine (I) is part of a larger X-ray and pharmacological study on tricyclic theophylline derivatives (Karolak-Wojciechowska & Pawłowski, 1990). The annelation of the six-membered lactam ring at the 7,8-position of theophylline changes the profile of its activity to neurosedative action, probably due to the chemical properties of the third lactam ring (Pawłowski, Drabczyńska, Gorczyca, Malec & Modzelewski, 1991). Incorporation of this ring allows us to expect a higher lipophilicity and a better absorption by the central nervous system. These pharmacological

results prompted us to investigate the molecular structure of the title compound as an aspect of its biological activity.



Experimental. Colourless crystals of the title compound were obtained from ethyl alcohol by slow evaporation. The density was determined by flotation in KI solution. Space group and initial cell parameters were obtained from Weissenberg photographs (produced for [100] and [001] directions): systematic absence $0kl$, $k \neq 2n$, $h0l$ $l \neq 2n$, $hk0$ $h \neq 2n$ determined the space group as $Pbca$ (No. 61). Crystal size $0.37 \times 0.12 \times 0.10$ mm, KM-4 automatic four-circle diffractometer, $\text{Cu K}\alpha$, graphite monochromator. Unit-cell parameters from setting angles of 23 reflections in range $25 < \theta < 43^\circ$. 2921 reflections collected with $2 \leq \theta \leq 65^\circ$; index range $0 \leq h \leq 8$, $0 \leq k \leq 45$, $0 \leq l \leq 12$; ω - 2θ scan method; intensities of three standard reflections measured every 70 reflections showed no significant fluctuations; no absorption correction. 2495 unique reflections, 1157 of which with $|F| > 3\sigma(F)$. Structure solved by direct methods using *SHELXS86* (Sheldrick, 1986) with $R(E) = 0.23$. *E* map revealed positions of all non-H atoms. Full-matrix least-squares refinement on *F*'s with *SHELX76* (Sheldrick, 1976); anisotropic thermal parameters applied to non-H atoms. All H atoms successively located from difference Fourier maps and their positional parameters refined with isotropic thermal parameters taken as 1.5 times those of the values for the respective parent C atoms. Final $R = 0.0514$, $wR = 0.0421$ with $w = 1.4845/[\sigma^2(F_o) + 0.000504F_o^2]$; empirical isotropic extinction parameter $g = 2.5(3) \times 10^{-7}$, where $F_c^* = F_c(1 - gF_c/\sin\theta)$; max. $\Delta/\sigma = 0.007$; max. difference peak $+0.20$, min. difference peak $-0.21 \text{ e } \text{\AA}^{-3}$. Atomic scattering factors as supplied by the program. The geometrical calculations were carried out using *CSU* (Vicković, 1988) and *PARST* (Nardelli, 1983) programs.

Discussion. Final atomic coordinates and equivalent isotropic thermal parameters are given in Table 1,†

† Lists of structure factors, anisotropic thermal parameters, least-squares planes, dihedral angles and H-atom parameters have been deposited with the British Library Document Supply Centre as Supplementary Publication No. SUP 53944 (14 pp.). Copies may be obtained through The Technical Editor, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.

Table 1. Fractional atomic coordinates and equivalent isotropic temperature factors (\AA^2) with e.s.d.'s in parentheses

U_{eq} is defined as one third of the trace of the orthogonalized U_{ij} tensor.

	x	y	z	U_{eq}^*
N(1)	-0.0506 (4)	0.40420 (9)	0.8490 (3)	0.038 (1)
N(3)	0.0910 (4)	0.45850 (9)	0.8340 (3)	0.040 (1)
N(5)	-0.0162 (4)	0.41944 (9)	0.5361 (3)	0.038 (1)
N(9)	-0.1355 (5)	0.36967 (9)	0.4403 (3)	0.046 (1)
N(11)	-0.1239 (4)	0.37721 (8)	0.6563 (3)	0.040 (1)
O(2)	0.0305 (4)	0.43376 (8)	1.0179 (2)	0.056 (1)
O(4)	0.1522 (4)	0.48438 (8)	0.6509 (3)	0.056 (1)
O(8)	-0.1692 (6)	0.36874 (9)	0.2364 (3)	0.090 (1)
C(1)	-0.1136 (8)	0.3745 (1)	0.9191 (4)	0.052 (1)
C(2)	0.0227 (6)	0.4321 (1)	0.9068 (4)	0.040 (1)
C(3)	0.1707 (8)	0.4884 (1)	0.8951 (5)	0.055 (1)
C(4)	0.0915 (5)	0.4592 (1)	0.7061 (4)	0.039 (1)
C(6)	0.0370 (7)	0.4351 (1)	0.4211 (4)	0.047 (1)
C(7)	-0.0929 (7)	0.4229 (1)	0.3251 (4)	0.055 (2)
C(8)	-0.1342 (7)	0.3851 (1)	0.3273 (4)	0.058 (1)
C(10)	-0.0963 (5)	0.3884 (1)	0.5431 (4)	0.040 (1)
C(12)	-0.0544 (5)	0.4033 (1)	0.7243 (3)	0.034 (1)
C(13)	0.0141 (5)	0.4296 (1)	0.6561 (3)	0.035 (1)
C(14)	-0.1779 (7)	0.3325 (1)	0.4555 (5)	0.059 (1)
C(15)	-0.0157 (8)	0.3121 (1)	0.4908 (4)	0.055 (1)
C(16)	0.1227 (9)	0.3081 (1)	0.4098 (5)	0.073 (2)
C(17)	0.2715 (11)	0.2889 (2)	0.4413 (9)	0.105 (3)
C(18)	0.2827 (12)	0.2740 (2)	0.5512 (10)	0.111 (3)
C(19)	0.1458 (14)	0.2781 (2)	0.6343 (7)	0.101 (3)
C(20)	-0.0015 (9)	0.2971 (1)	0.6040 (5)	0.069 (2)

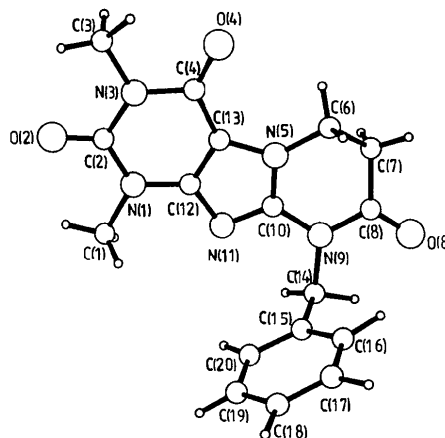


Fig. 1. A view of the molecule with the atomic labelling.

bond distances, valence angles and selected torsion angles in Table 2. A view of the molecule with numbering of the atoms is shown (*PLUTO*; Motherwell & Clegg, 1978) in Fig. 1.

The effect of thermal motion on bond lengths was examined (Busing & Levy, 1964). Bond lengths corrected for libration do not differ significantly from the uncorrected values and the latter values will be used in the discussion.

Table 2. Bond lengths (Å), angles (°) and torsion angles (°) in the 1,3-diazinone ring with *e.s.d.*'s in parentheses

N(1)—C(1)	1.461 (5)	O(2)—C(2)	1.226 (4)
N(1)—C(2)	1.369 (5)	O(4)—C(4)	1.233 (4)
N(1)—C(12)	1.374 (5)	O(8)—C(8)	1.213 (5)
N(3)—C(2)	1.394 (5)	C(4)—C(13)	1.398 (5)
N(3)—C(3)	1.464 (6)	C(6)—C(7)	1.515 (7)
N(3)—C(4)	1.409 (5)	C(7)—C(8)	1.491 (6)
N(5)—C(6)	1.459 (5)	C(12)—C(13)	1.364 (5)
N(5)—C(10)	1.346 (5)	C(14)—C(15)	1.503 (7)
N(5)—C(13)	1.396 (5)	C(15)—C(16)	1.379 (7)
N(9)—C(8)	1.380 (5)	C(15)—C(20)	1.378 (7)
N(9)—C(10)	1.374 (5)	C(16)—C(17)	1.387 (9)
N(9)—C(14)	1.481 (5)	C(17)—C(18)	1.341 (10)
N(11)—C(10)	1.335 (5)	C(18)—C(19)	1.387 (10)
N(11)—C(12)	1.359 (5)	C(19)—C(20)	1.369 (9)
C(1)—N(1)—C(2)	120.3 (3)	N(9)—C(8)—O(8)	121.0 (4)
C(1)—N(1)—C(12)	120.0 (4)	N(9)—C(8)—C(7)	116.1 (4)
C(2)—N(1)—C(12)	119.6 (4)	O(8)—C(8)—C(7)	122.9 (4)
C(2)—N(3)—C(3)	117.5 (4)	N(5)—C(10)—N(9)	121.2 (4)
C(2)—N(3)—C(4)	126.2 (4)	N(5)—C(10)—N(11)	114.3 (4)
C(3)—N(3)—C(4)	116.3 (4)	N(9)—C(10)—N(11)	124.5 (4)
C(6)—N(5)—C(10)	122.9 (4)	N(1)—C(12)—N(11)	125.3 (4)
C(6)—N(5)—C(13)	131.2 (3)	N(1)—C(12)—C(13)	121.5 (4)
C(10)—N(5)—C(13)	105.6 (3)	N(11)—C(12)—C(13)	113.2 (3)
C(8)—N(9)—C(10)	120.9 (4)	N(5)—C(13)—C(4)	132.1 (4)
C(8)—N(9)—C(14)	121.6 (4)	N(5)—C(13)—C(12)	104.5 (3)
C(10)—N(9)—C(14)	117.6 (3)	C(4)—C(13)—C(12)	123.4 (4)
C(10)—N(11)—C(12)	102.4 (3)	N(9)—C(14)—C(15)	111.3 (4)
N(1)—C(2)—N(3)	117.2 (4)	C(14)—C(15)—C(16)	120.3 (5)
N(1)—C(2)—O(2)	121.6 (4)	C(14)—C(15)—C(20)	121.1 (5)
N(3)—C(2)—O(2)	121.2 (4)	C(16)—C(15)—C(20)	118.6 (6)
N(3)—C(4)—O(4)	120.6 (4)	C(15)—C(16)—C(17)	120.5 (6)
N(3)—C(4)—C(13)	112.1 (4)	C(16)—C(17)—C(18)	120.4 (7)
O(4)—C(4)—C(13)	127.3 (4)	C(17)—C(18)—C(19)	119.9 (8)
N(5)—C(6)—C(7)	107.3 (4)	C(18)—C(19)—C(20)	120.1 (7)
C(6)—C(7)—C(8)	115.4 (4)	C(15)—C(20)—C(19)	120.5 (6)
N(5)—C(6)—C(7)—C(8)	-45.5 (5)	N(9)—C(10)—N(5)—C(6)	-1.0 (6)
N(5)—C(10)—N(9)—C(8)	-15.8 (6)	C(7)—C(6)—N(5)—C(10)	30.6 (5)
N(9)—C(8)—C(7)—C(6)	33.5 (6)	C(7)—C(8)—N(9)—C(10)	-1.6 (6)

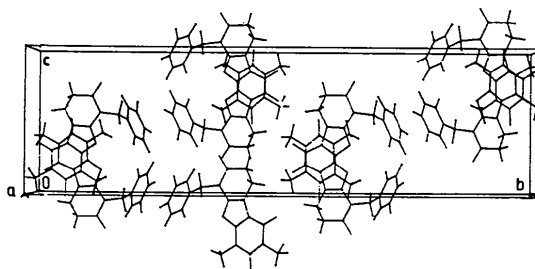


Fig. 2. Unit-cell packing.

angles C(8)—N(9)—C(14)—C(15) and N(9)—C(14)—C(15)—C(16) of 109.4 (5) and -67.4 (6)° respectively.

Packing of the molecules is shown in Fig. 2. The theophylline moieties are stacked in the *a* direction. This stacking can be characterized by the dipolar overlap of the hetero-bonds [N(3)—C(3) and O(4)—C(4), N(3)—C(4), N(1)—C(1) and N(11)—C(12)] and the mean distance between molecular planes of 3.34 (4) Å.

This research was supported by Project RP.II.10 from the Polish Ministry of Education.

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