

Fig. 2. Crystal structure along the *c* axis. The dotted lines indicate hydrogen bonds.

screw axis. Those of molecules *B* and *C* are also related by another pseudo twofold screw axis. The crystallographic twofold screw axis and the two pseudo screw axes are roughly perpendicular to each other. The four unique molecules are connected by the two types of N...O intermolecular hydrogen

bonds: one type is that between the sulfamoyl groups [N(1)...O(1) or N(1)...O(2)] and the other is that between the sulfamoyl and amide groups [N(1)...O(4)]. Thus, a layer structure of these three groups perpendicular to the *b* axis is formed by a network of the hydrogen bonds. The layer is sandwiched by the other two layers of the methyl, pyrrolidine and butyl groups. In those two layers the constituent groups are in contact *via* van der Waals forces.

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Structure of 7-(*N*-Formyl-*N*-phenyl)amino-4,6-dinitrobenzofurazan 1-Oxide

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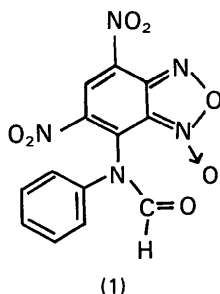
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Abstract. C₁₃H₇N₅O₇, *M_r* = 345.23, tetragonal, *I*4̄, *a* = 11.129 (3), *c* = 23.368 (6) Å, *V* = 2894 (1) Å³, *Z* = 8, *D_x* = 1.585 g cm⁻³, λ(Mo *K*α) = 0.71069 Å, μ = 1.2 cm⁻¹, *F*(000) = 1616, *T* = 293 K, *R* = 0.056 for

1198 reflections with *F_o* > 4σ(*F_o*). The N—O distance in the nitro groups is found to be significantly smaller (1.172–1.218 Å) compared with a typical value of 1.23 Å. The formyl group at N(1) introduces

considerable changes in the structure of the molecule. The molecules are held together by extensive van der Waals interactions.

Introduction. 4,6-Dinitrobenzofurazan derivatives are of special significance because of their extraordinary electrophilic character. In our investigations on this type of compound we have been interested in relationships between molecular structure, π -electron density distribution and chemical reactivity. Thus 4-anilino-5,7-dinitrobenzofurazan was found to have a partial polymethinic structure which is reflected by NMR chemical shifts, alternating π -electron densities and unusual chemical behaviour (Niclas, Göhrmann, Ramm & Schulz, 1990). In the present paper we report the crystal structure of 7-(*N*-formyl-*N*-phenyl)amino-4,6-dinitrobenzofurazan 1-oxide (1), prepared by nitric acid oxidation of silver 7-(*N*-formyl-*N*-phenyl)amino-4,6-dinitro-7,*x*-dihydrobenzofurazanide 1-oxide (Göhrmann & Niclas, 1989).



Experimental. Red crystals were obtained by recrystallization from a mixture of ethanol/acetone/ether (1:2:1) under concentration of the mother liquor by evaporation of the solvent at 331 K. Dimensions of the sample $0.5 \times 0.4 \times 0.4$ mm. Intensities measured on an Enraf-Nonius CAD-4 diffractometer with a graphite monochromator using ω - 2θ scans. Unit-cell dimensions from setting angles of 25 reflections having $9 < \theta < 14^\circ$. Data collected to $(\sin \theta)/\lambda = 0.594 \text{ \AA}^{-1}$, $h 0 \rightarrow 13$, $k -13 \rightarrow 13$, $l 0 \rightarrow 27$. Two standard reflections ($\bar{4}53$, $\bar{2}51$) measured every 3000 s, total loss in intensity -0.6% over 60.9 h. 2773 reflections measured, 1994 unique reflections ($R_{\text{int}} = 0.0171$), 1781 reflections observed with $I > 2\sigma(I)$. Data corrected for Lorentz and polarization factors, no absorption correction applied due to low absorption coefficient. Structure solved by direct and Fourier methods. Full-matrix least squares minimized $\sum w|F_o - F_c|^2$. H atoms located by difference Fourier synthesis and refined isotropically. Number of parameters refined 255, $R = 0.056$, $wR = 0.036$, $S = 7.5$ (2) for 1198 reflections, weighting scheme $w(F) = 1/\sigma^2(F)$. Final $(\Delta/\sigma)_{\text{max}} = 0.149$, $-0.25 \leq \Delta\rho \leq 0.25 \text{ e \AA}^{-3}$ on final difference map. Lowest F_c extinc-

tion correcting factor 0.91. All crystallographic computations were performed on a VAX/VMS V4-6 computer using programs *MITHRIL* (Gilmore, 1983), and *XTAL2.2* (Hall & Stewart, 1987) with the atomic scattering factors as included in the program. The *XTAL* version of *ORTEPII* (Johnson, 1971) was used to prepare the figures.

Discussion. The final positional parameters with equivalent isotropic temperature factors of the non-H atoms are given in Table 1.* A drawing of the molecule with the atomic numbering scheme is shown in Fig. 1. Bond lengths and angles are compiled in Table 2.

The six-membered ring contains two short bond distances [C(7)—C(12) 1.365 (7) and C(10)—C(11) 1.342 (7) Å] whereas the other bonds are significantly longer [1.398 (7)—1.428 (7) Å]. These values correspond to the canonical form of the molecule with double bonds between C(7)—C(12) and C(10)—C(11) and single bonds between the other C atoms.

The C—NO₂ bond C(12)—N(4) [1.446 (7) Å] is short when compared to normal aromatic nitro compounds (Coppens, 1964; Coppens & Schmidt, 1964) but (as to be expected) distinctly longer than those in the salts of dinitrobenzofuroxan (Lowe-Ma, 1986) or dinitrobenzofurazan (Mathew & Palenik, 1971). Compared to the typical N—O distance in nitro groups (1.23 Å) we have found significantly smaller values [1.172 (8)—1.218 (7) Å]. This can be explained by the high thermal motion of the oxygen atoms [*cf.* also N(2)—O(7) 1.198 (7) Å].

The N—C bonds in the furoxan ring are extended compared to the bond lengths of other furoxan rings (Britton & Noland, 1972; Britton & Olson, 1979; Calleri, Bonaccorti & Viterbo, 1977; Calleri, Chowdhury & Viterbo, 1976). The different N—O bond lengths [N(3)—O(1) 1.376 (8) Å; N(2)—O(1) 1.421 (7) Å] agree with corresponding values found in other furoxan rings. The five-membered ring is nearly planar [distance of the O atom O(7) from the least-squares plane is 0.04 Å] and twisted about 2.8 (2)° to the plane of the six-membered ring. The nitro groups at C(12) and C(10) are tilted against the six-membered ring at about 32.8 (2) and 2.4 (1)°, respectively. The benzofuroxan fragment and the phenyl ring C(1)—C(6) enclose a dihedral angle of 95.0 (2)°.

The packing of the molecules in the unit cell is shown in Fig. 2. The molecular arrangement is dominated by van der Waals interactions.

* Lists of structure factors, anisotropic thermal parameters and H-atom parameters have been deposited with the British Library Document Supply Centre as Supplementary Publication No. SUP 53870 (25 pp.). Copies may be obtained through The Technical Editor, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.

Table 1. Final positional and thermal parameters of non-H atoms ($\times 10^2 \text{ \AA}^2$) with e.s.d.'s in parentheses
$$U_{eq} = (1/3) \sum_i \sum_j U_{ij} a_i^* a_j^* a_i a_j$$

	x	y	z	U_{eq}
O(1)	0.6506 (4)	-0.0149 (4)	0.1327 (2)	8.4 (2)
O(2)	1.0666 (4)	0.3682 (4)	0.1890 (3)	10.4 (2)
O(3)	1.1318 (4)	0.2463 (4)	0.2505 (2)	9.2 (2)
O(4)	0.7051 (5)	-0.0651 (4)	0.3063 (2)	10.1 (2)
O(5)	0.8444 (6)	0.0210 (6)	0.3501 (2)	12.8 (3)
O(6)	1.1212 (4)	0.1453 (6)	0.1071 (2)	11.4 (2)
O(7)	0.7271 (4)	0.1001 (4)	0.0602 (2)	8.8 (2)
N(1)	0.9400 (4)	0.2396 (4)	0.1025 (2)	5.4 (2)
N(2)	0.7301 (5)	0.0727 (5)	0.1098 (3)	7.1 (2)
N(3)	0.6747 (5)	-0.0357 (5)	0.1896 (3)	7.8 (2)
N(4)	1.0592 (4)	0.2759 (5)	0.2142 (2)	6.1 (2)
N(5)	0.7904 (5)	0.0011 (5)	0.3081 (3)	6.8 (2)
C(1)	0.8683 (4)	0.3298 (5)	0.0736 (2)	5.0 (2)
C(2)	0.8082 (6)	0.4141 (6)	0.1040 (3)	7.7 (3)
C(3)	0.7428 (7)	0.4974 (7)	0.0747 (4)	9.3 (3)
C(4)	0.7443 (7)	0.5053 (7)	0.0168 (4)	8.6 (3)
C(5)	0.8018 (7)	0.4159 (7)	-0.0143 (3)	8.6 (3)
C(6)	0.8651 (6)	0.3293 (6)	0.0142 (3)	7.6 (3)
C(7)	0.8994 (4)	0.1852 (4)	0.1543 (2)	4.5 (2)
C(8)	0.8017 (4)	0.1054 (5)	0.1547 (2)	4.8 (2)
C(9)	0.7646 (4)	0.0408 (5)	0.2029 (2)	4.9 (2)
C(10)	0.8256 (5)	0.0599 (4)	0.2544 (2)	4.7 (2)
C(11)	0.9189 (5)	0.1363 (4)	0.2562 (2)	4.7 (2)
C(12)	0.9565 (4)	0.1982 (4)	0.2057 (2)	4.6 (2)
C(13)	1.0563 (6)	0.2125 (7)	0.0849 (3)	8.4 (3)

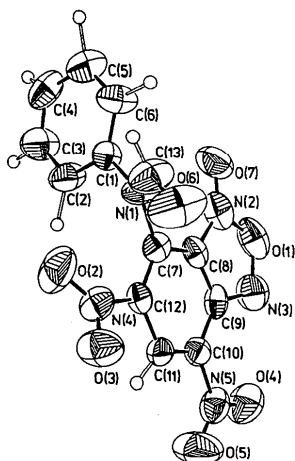


Fig. 1. ORTEP (Johnson, 1971) drawing of the molecular structure with crystallographic numbering scheme.

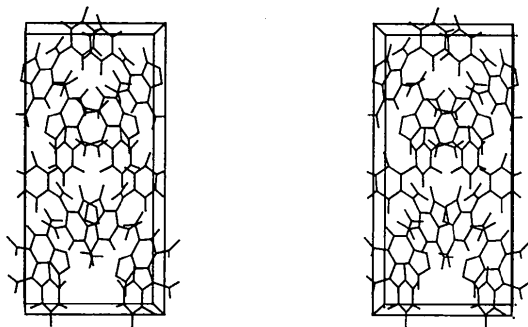


Fig. 2. Stereoview of the packing of the molecules in the unit cell.

Table 2. Bond lengths (\AA) and valence angles ($^\circ$) with e.s.d.'s in parentheses

O(1)—N(3)	1.376 (8)	C(4)—C(5)	1.39 (1)
N(1)—C(1)	1.449 (7)	C(5)—C(6)	1.37 (1)
N(1)—C(7)	1.429 (7)	C(7)—C(8)	1.403 (7)
N(1)—C(13)	1.390 (8)	C(7)—C(12)	1.365 (7)
N(2)—O(1)	1.421 (7)	C(8)—N(2)	1.367 (8)
N(2)—O(7)	1.198 (7)	C(8)—C(9)	1.400 (8)
N(4)—O(2)	1.186 (7)	C(9)—N(3)	1.350 (7)
N(4)—O(3)	1.218 (7)	C(9)—C(10)	1.398 (7)
N(5)—O(4)	1.202 (7)	C(10)—N(5)	1.467 (8)
N(5)—O(5)	1.172 (8)	C(10)—C(11)	1.342 (7)
C(1)—C(2)	1.353 (8)	C(11)—C(12)	1.428 (7)
C(1)—C(6)	1.389 (8)	C(12)—N(4)	1.446 (7)
C(2)—C(3)	1.36 (1)	C(13)—O(6)	1.162 (9)
C(3)—C(4)	1.36 (1)		
N(2)—O(1)—N(3)	110.9 (4)	C(4)—C(5)—C(6)	119.1 (7)
C(13)—N(1)—C(1)	121.7 (5)	C(1)—C(6)—C(5)	119.9 (6)
C(13)—N(1)—C(7)	116.9 (5)	N(1)—C(7)—C(8)	121.2 (5)
C(8)—N(2)—O(1)	104.9 (5)	N(1)—C(7)—C(12)	123.6 (4)
C(8)—N(2)—O(7)	133.7 (5)	C(8)—C(7)—C(12)	115.0 (5)
O(1)—N(2)—O(7)	121.4 (5)	C(9)—C(8)—N(2)	108.0 (5)
C(9)—N(3)—O(1)	105.2 (5)	C(7)—C(8)—N(2)	128.0 (5)
O(2)—N(4)—O(3)	122.2 (5)	C(7)—C(8)—C(9)	124.0 (5)
C(12)—N(4)—O(2)	120.4 (5)	C(8)—C(9)—N(3)	110.9 (5)
C(12)—N(4)—O(3)	117.2 (5)	C(8)—C(9)—C(10)	118.1 (5)
O(4)—N(5)—O(5)	123.3 (6)	C(10)—C(9)—N(3)	131.0 (5)
C(10)—N(5)—O(4)	117.1 (5)	C(9)—C(10)—N(5)	122.5 (5)
C(10)—N(5)—O(5)	119.6 (6)	C(11)—C(10)—N(5)	117.6 (5)
N(1)—C(1)—C(2)	120.6 (5)	C(9)—C(10)—C(11)	119.9 (5)
N(1)—C(1)—C(6)	118.4 (5)	C(10)—C(11)—C(12)	120.5 (5)
C(2)—C(1)—C(6)	120.9 (5)	C(7)—C(12)—N(4)	123.5 (4)
C(1)—C(2)—C(3)	118.2 (6)	C(11)—C(12)—N(4)	114.0 (4)
C(2)—C(3)—C(4)	122.6 (7)	C(7)—C(12)—C(11)	122.5 (4)
C(3)—C(4)—C(5)	118.8 (7)	N(1)—C(13)—O(6)	125.9 (7)

Our results show that the introduction of the formyl group at N(1) effects considerable changes in the structure of the molecule. The most striking differences are the bond lengths of C(7)—N(1) [cf. Niclas, Göhrmann, Ramm & Schulz (1990): C—NHPh 1.320 Å; here C—N(CHO)Ph 1.429 (7) Å] which cannot be explained by the additional N→O function.

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