measurements, these results provide a basis for the calculations needed in this work.

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Inclusion Behavior of 2-Hydroxy-3,5-dinitro-*N'*-(5-nitrofurfurylidene)benzohydrazide, $C_{12}H_7N_5O_9$. Crystal Structures with Acetonitrile and Acetic Acid

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Abstract. Acetonitrile (1:1) complex: $M_r = 406{\cdot}3$, monoclinic, $P2_1/c$, $a = 12{\cdot}977$ (2), $b = 8{\cdot}021$ (4), $c = 16{\cdot}830$ (3) Å, $\beta = 106{\cdot}95$ (1)°, $V = 1675{\cdot}7$ Å^3, $Z = 4$, $D_x = 1{\cdot}610$ g cm^{-3}, $F(000) = 832$, λ(Mo $K\alpha$) = 0·7107 Å, μ(Mo $K\alpha$) = 1·5 cm^{-1}, room temperature. Acetic acid (1:1) complex: $M_r = 425{\cdot}3$, triclinic, $P\bar{1}$, $a = 8{\cdot}665$ (1), $b = 8{\cdot}696$ (7), $c = 12{\cdot}077$ (2) Å, $\alpha = 90{\cdot}82$ (3), $\beta = 99{\cdot}82$ (1), $\gamma = 105{\cdot}15$ (3) Å, $V = 863{\cdot}8$ Å^3, $Z = 2$, $D_x = 1{\cdot}635$ g cm^{-3}, $F(000) = 436$, μ(Mo $K\alpha$) = 1·6 cm^{-1}, room temperature. The two structures were determined by a combination of direct and Fourier methods and refined to R values of respectively 0·048 and 0·052 for 2062 and 1690 observed reflections [$F_o^2 \geq 3\sigma(F_o^2)$]. The observed dense packing arrangements are dominated by characteristic dipole–dipole interactions between molecular entities located across crystallographic centers of symmetry. The solvent guest species are included in pseudo-channels formed between layers of the hydrazide molecules. Structural differences between the two compounds are mainly due to the different functionality of the solvent environments as revealed by observed hydrogen-bonding patterns. They reflect a considerable influence of the solvation forces on the host conformation, as a result of which polymorphic crystal-structure types are formed.

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Introduction. Organic compounds containing polar surfaces frequently exhibit interesting inclusion properties upon crystallization from various environments, which are associated with the appearance of channels or cages within the crystal lattice (*e.g.* MacNicol, McKendrick & Wilson, 1978). Such phenomena of co-crystallization with other species into a multicomponent system, which can be directly related to the energetics of intermolecular interactions, are even more pronounced when the polar substituents act also as good donors or acceptors of hydrogen bonds. The crystal packing of dipolar species is usually stabilized by a combination of attractive and repulsive forces. However, an optimized accommodation of all interactions in the solid structure may not always be consistent with an efficient packing of the molecular entities (particularly those characterized by an irregular shape), inducing in such case voids within the host lattice. To achieve a thermally stable crystal this space must be filled by another (guest) component with complementary geometric and functional properties, requirements often giving rise to selective molecular complexations (*e.g.* Iwamoto, 1979). In inclusion structures with imperfect geometric relationships and lacking specific bindings between the complex constituents the guest species are either disordered or thermally smeared within the rigid host lattice (Allcock, Allen, Bissell, Smeltz & Teeter, 1976; Goldberg, 1982). Solvation

forces, undoubtedly, play a major role in determining the crystal-structure type of the inclusion compound formed. If they are sufficiently strong in comparison with other intermolecular interactions, the solvating species will be carried out from the solution into the crystal along with the solvated substance. As a result, structural domains that dominate dynamic equilibria in the liquid phase may persist also, at least to some extent, in the solid inclusion system.

In the course of our investigations into the structural properties and elementary patterns of intermolecular interactions in inclusion compounds we have recently characterized a series of channel-type clathrates capable of accommodating various guest molecules (Goldberg, 1982). These complexes were found to be stabilized mainly by dipole–dipole forces. In another example the inclusion phenomenon has been used as an aid in studying the relationships between solvation patterns and structural polymorphism (Goldberg, 1983). It has been shown that it is possible to induce different conformations in a flexible polar system by a suitable modification of the solvating media. To provide additional structural data and to further our understanding of the inclusion behavior we have characterized the structural properties of 2-hydroxy-3,5-dinitro-*N'*-(5-nitrofurfurylidene)benzohydrazide (BAH). This compound contains a large number of polar functions as well as sites available for hydrogen bonding, and has an elongated flat shape. It reveals remarkable tendency to form inclusion complexes when crystallized from various solvents; consequently, sizeable crystals of the pure species have not yet been obtained. The present report is thus concerned with the crystal structures of two representative 1:1 inclusion complexes of BAH with aprotic acetonitrile (I) and protic acetic acid (II) solvents. Interactions which determine the corresponding structures are discussed in detail with an emphasis on the variations effected by the different nature of the solvating moieties.

Experimental. BAH kindly supplied by TEVA Pharmaceutical Industries, Ltd. Single crystals of (I) and (II) suitable for crystallographic study obtained upon recrystallization from acetonitrile and glacial acetic acid respectively; crystals used ~0·2 × 0·2 × 0·4 mm (I) and 0·1 × 0·3 × 0·4 mm (II); cell constants by least-squares procedure applied to 24 reflections with $11 < \theta < 13°$ for (I) and to 20 reflections with $10 < \theta < 13°$ for (II). Diffraction data measured at *ca* 291 K, Enraf–Nonius CAD-4 diffractometer equipped with a graphite monochromator, employing Mo $K\alpha$ ($\lambda_{mean} = 0{\cdot}71069$ Å) radiation and ω–2θ scans, scan rate 1–5° min^{-1}. For (I) 4092 reflections collected to $\theta_{max} = 27°$ of which 3039 were unique with $I > 0$ ($R_{int} = 0{\cdot}030$). For (II) 2593 reflections measured to $\theta = 23°$ of which 2187 were unique and had a positive intensity ($R_{int} = 0{\cdot}029$). In both cases intensity variation of three standard reflections from different zones of the reciprocal space negligible during the measurements, corrections for Lorentz and polarization effects, but not for absorption or secondary extinction. Final refinement calculations based on 2062 reflections ($-16 \le h \le 15$, $0 \le k \le 10$, $0 \le l \le 21$) (I) and 1690 ($-9 \le h \le 9$, $-9 \le k \le 9$, $0 \le l \le 13$) (II) [$F_o^2 \ge 3\sigma(F_o^2)$]; atomic scattering factors from *International Tables for X-ray Crystallography* (1974).

The two structures were solved by a combination of direct methods (Main, Hull, Lessinger, Germain, Declercq & Woolfson, 1978; Main, Fiske, Hull, Lessinger, Germain, Declercq & Woolfson, 1980) and Fourier techniques. Subsequent refinement included the positional and anisotropic thermal parameters of all the nonhydrogen atoms. During this procedure it became apparent that the 2-hydroxybenzoic acid fragment exhibits in compound (II) a twofold rotational disorder with respect to an axis joining the carbonyl carbon and positions 1 and 4 of the benzoic acid ring. This has been taken into account in the structural model by assuming that the hydroxyl group is disordered between two sites, and refining the relative occupancies. The refined occupancy factor was 0·33 (4) for the minor site which corresponds to the observed molecular conformation in (I), and 0·67 for the major site. The disorder is also reflected to some extent in the relatively large thermal parameters of the two nitro groups substituted on the benzoic acid, but the positions of the disordered individual atoms appear to overlap and could not be resolved. All H atoms in (I) and (II) (except that of the disordered hydroxyl) were located directly from electron density difference maps and assigned isotropic temperature factors. The atomic parameters of the H atoms were not refined except for their partial adjustment at an intermediate stage with low-order data ($\sin\theta/\lambda < 0{\cdot}50$ Å^{-1}). Least-squares calculations minimized $\sum w(\Delta F)^2$, $w = 1/\sigma^2(F_o)$. At convergence $(\Delta/\sigma) < 0{\cdot}3$. The final $R = 0{\cdot}048$ and $wR = 0{\cdot}058$ for (I) and $R = 0{\cdot}052$ and $wR = 0{\cdot}066$ for (II).

The final difference Fourier maps showed no indications of incorrectly placed or missing atoms; $(\Delta\rho)_{max}$ and $(\Delta\rho)_{min}$ respectively 0·36 and −0·33 e Å^{-3} in (I) and 0·21 and −0·27 e Å^{-3} in (II). Computations performed with an extensively modified version of *ORFLS* (Busing, Martin & Levy, 1962), and with *PARST* (Nardelli, 1983). Figures drawn with *ORTEP* (Johnson, 1976).*

* Lists of structure factors and anisotropic thermal parameters as well as Fig. 3′, an illustration of the stacking interactions and modes of intermolecular overlap in (I), and Fig. 4′, an edge-on stereoview, approximately down **a**, of a single stack of interacting molecules in (II) have been deposited with the British Library Lending Division as Supplementary Publication No. SUP 38908 (17 pp.). Copies may be obtained through The Executive Secretary, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.

Table 1. *Atomic coordinates and isotropic thermal parameters*

U_{eq} for the nonhydrogen atoms is one third of the trace of the orthogonalized U^{ij}.

(I) Acetonitrile complex

	x	*y*	*z*	U_{eq}/U(Å^2)
C(1)	0·4213 (2)	0·6746 (4)	0·1408 (2)	0·0358
C(2)	0·4652 (2)	0·7073 (4)	0·0798 (2)	0·0451
C(3)	0·3941 (3)	0·6398 (4)	0·0082 (2)	0·0450
C(4)	0·3126 (2)	0·5679 (4)	0·0310 (2)	0·0346
O(5)	0·3276 (1)	0·5901 (2)	0·1146 (1)	0·0332
N(6)	0·4538 (2)	0·7198 (3)	0·2263 (2)	0·0437
O(7)	0·5375 (2)	0·7995 (3)	0·2509 (2)	0·0671
O(8)	0·3968 (2)	0·6792 (3)	0·2696 (1)	0·0612
C(9)	0·2178 (2)	0·4782 (4)	−0·0152 (2)	0·0327
N(10)	0·1538 (2)	0·4179 (3)	0·0222 (1)	0·0318
N(11)	0·0671 (2)	0·3316 (3)	−0·0275 (1)	0·0311
C(12)	−0·0004 (2)	0·2588 (3)	0·0094 (2)	0·0297
O(13)	0·0145 (2)	0·2710 (3)	0·0851 (1)	0·0468
C(14)	−0·0939 (2)	0·1629 (3)	−0·0430 (2)	0·0269
C(15)	−0·1609 (2)	0·0813 (4)	−0·0016 (2)	0·0301
C(16)	−0·2497 (2)	−0·0068 (4)	−0·0505 (2)	0·0299
C(17)	−0·2730 (2)	−0·0175 (4)	−0·1354 (2)	0·0311
C(18)	−0·2071 (2)	0·0644 (3)	−0·1732 (2)	0·0304
C(19)	−0·1187 (2)	0·1535 (3)	−0·1287 (2)	0·0286
O(20)	−0·1411 (2)	0·0894 (3)	0·0799 (1)	0·0451
N(21)	−0·3250 (2)	−0·0905 (3)	−0·0134 (2)	0·0414
O(22)	−0·2973 (2)	−0·1251 (4)	0·0581 (2)	0·0888
O(23)	−0·4105 (3)	−0·1293 (5)	−0·0573 (2)	0·1101
N(24)	−0·2329 (2)	0·0605 (3)	−0·2639 (1)	0·0384
O(25)	−0·1796 (2)	0·1458 (3)	−0·2967 (1)	0·0538
O(26)	−0·3071 (2)	−0·0286 (3)	−0·3022 (1)	0·0545
N(27)	−0·0758 (2)	−0·2163 (4)	−0·2966 (2)	0·0568
C(28)	−0·1256 (3)	−0·3098 (4)	−0·2730 (2)	0·0436
C(29)	−0·1907 (3)	−0·4281 (5)	−0·2439 (2)	0·0585
H(2)	0·533	0·765	0·090	0·070
H(3)	0·397	0·640	−0·049	0·062
H(9)	0·202	0·472	−0·074	0·045
H(11)	0·059	0·322	−0·083	0·042
H(17)	−0·331	−0·077	−0·166	0·040
H(19)	−0·078	0·211	−0·156	0·021
H(20)	−0·078	0·151	0·102	0·075
H(29*A*)	−0·174	−0·427	−0·186	0·090
H(29*B*)	−0·259	−0·391	−0·263	0·100
H(29*C*)	−0·160	−0·536	−0·258	0·111

(II) Acetic acid complex

	x	*y*	*z*	U_{eq}/U(Å^2)
C(1)	0·7430 (5)	0·0541 (5)	0·9132 (3)	0·0399
C(2)	0·6338 (6)	−0·0792 (5)	0·9318 (3)	0·0462
C(3)	0·5072 (5)	−0·1058 (5)	0·8389 (3)	0·0455
C(4)	0·5496 (5)	0·0145 (4)	0·7690 (3)	0·0371
O(5)	0·6976 (3)	0·1145 (3)	0·8146 (2)	0·0410
N(6)	0·8894 (5)	0·1394 (4)	0·9812 (3)	0·0508
O(7)	0·9745 (4)	0·2576 (4)	0·9457 (3)	0·0696
O(8)	0·9255 (4)	0·0902 (4)	1·0749 (3)	0·0671
C(9)	0·4624 (5)	0·0487 (5)	0·6644 (3)	0·0394
N(10)	0·5279 (4)	0·1705 (4)	0·6135 (2)	0·0397
N(11)	0·4303 (4)	0·1957 (4)	0·5168 (3)	0·0393
C(12)	0·4967 (5)	0·3126 (4)	0·4533 (3)	0·0369
O(13)	0·6419 (4)	0·3823 (3)	0·4719 (2)	0·0524
C(14)	0·3860 (5)	0·3561 (4)	0·3580 (3)	0·0355
C(15)	0·4625 (5)	0·4497 (4)	0·2803 (3)	0·0386
C(16)	0·3650 (5)	0·5007 (4)	0·1918 (3)	0·0398
C(17)	0·2007 (5)	0·4662 (5)	0·1794 (3)	0·0421
C(18)	0·1281 (5)	0·3769 (5)	0·2583 (3)	0·0408
C(19)	0·2175 (5)	0·3202 (4)	0·3480 (3)	0·0405
O(20)	0·1447 (5)	0·2440 (5)	0·4237 (3)	0·0487
O(20*)†	0·6193 (11)	0·4955 (10)	0·2930 (7)	0·0504
N(21)	0·4464 (5)	0·6030 (4)	0·1114 (3)	0·0529
O(22)	0·5894 (5)	0·6598 (5)	0·1343 (3)	0·1112
O(23)	0·3628 (5)	0·6281 (5)	0·0271 (3)	0·0877
N(24)	−0·0461 (5)	0·3481 (5)	0·2479 (3)	0·0564
O(25)	−0·1205 (4)	0·4060 (5)	0·1744 (3)	0·0791
O(26)	−0·1139 (4)	0·2609 (5)	0·3145 (3)	0·0814
C(27)	−0·0724 (5)	0·1894 (6)	0·6060 (4)	0·0497
O(28)	−0·0973 (4)	0·3334 (4)	0·6094 (3)	0·0692
C(29)	0·0711 (7)	0·1783 (6)	0·6868 (4)	0·0761
O(30)	−0·1594 (4)	0·0861 (4)	0·5420 (3)	0·0742
H(2)	0·642	−0·135	1·004	0·078
H(3)	0·390	−0·191	0·823	0·059
H(9)	0·338	−0·030	0·632	0·047
H(11)	0·320	0·122	0·488	0·060
H(17)	0·127	0·484	0·111	0·060
H(28)	−0·203	0·326	0·557	0·105
H(29*A*)	0·070	0·233	0·767	0·100
H(29*B*)	0·186	0·256	0·667	0·100
H(29*C*)	0·076	0·071	0·694	0·100

† O(20*) represents the minor site of the disordered hydroxyl oxygen.

Discussion. Table 1 gives atomic parameters for both structures. Figs. 1 and 2 illustrate the molecular structures of the two compounds; the hydrogen bonding between the component species is also shown. The covalent bond lengths and angles obtained for the hydrazide moiety are compared in Table 2. Most of the individual parameters have similar values in the two structures, exhibiting no extraordinary features. Normal geometries also occur in the acetonitrile [C≡N 1·135 (5) Å, C–C 1·448 (5) Å, N≡C–C 179·0 (3)°] and acetic acid [C=O 1·183 (5) Å, C–O 1·326 (6) Å, C–C 1·468 (7) Å, C–C–O 112·7 (4)°, C–C=O 126·4 (5)°, O–C=O 120·9 (4)°] guest molecules.

In the acetonitrile complex the hydrazide framework is approximately planar. The displacements of individual atoms from the least-squares molecular plane are within ±0·15 Å, excluding the O atoms of the three peripheral nitro groups. The latter are significantly twisted about the exocyclic C–N bonds out of the (almost perfect) planes of the benzene and furan rings, by 3·3 (4)° about C(1)–N(6), 18·3 (3)° about C(16)–N(21) and 6·3 (1)° about C(18)–N(24). On the other hand, the molecular framework of BAH in the acetic acid complex deviates considerably from planarity. This is reflected in the gradual decrease of torsion angles about bonds which bridge between the two rings: C(4)–C(9)=N(10)–N(11) 177·2 (4), C(9)=N(10)–N(11)–C(12) 173·9 (4), N(10)–N(11)–C(12)–C(14) 171·4 (3), N(11)–C(12)–C(14)–C(15) 166·2 (4) and N(11)–C(12)–C(14)–C(19) −19·0 (6)°. As a result, an angle of 32·5 (2)° is formed between the normals to the furan and benzene rings. Minor deviations of the nitro substituents from coplanarity with the individual rings exist in this structure as well, the dihedral angles about the respective C–NO_2 bonds varying between 4·3 (2) and 13·6 (4)° (however, these values correspond to averaged positions of the nitro groups in the rotationally disordered benzoic acid fragment described above).

Two aspects of the molecular structure deserve particular attention as they have a direct influence on the conformation adopted by BAH, the packing modes it exhibits and its inclusion behavior.

The first feature involves the hydrogen-bonding capability of BAH. In this respect the hydrazide molecule contains functional groups of two kinds, those that are good donors (O–H and N–H) and those that

are good acceptors (C=O) of hydrogen bonds. In the crystals obtained from acetonitrile, an aprotic environment, the molecular structure is characterized by an internal O–H(20)···O=C(12) bond. This is associated with the formation of a six-membered ring (Fig. 1) and appears to be an important factor in stabilizing the planarity of the molecular framework. The NH(11) group is approached by the nitrogen end of acetonitrile, providing a link between the host and the solvent species *via* an N–H···N bond. Geometric parameters related to these interactions are in Table 3.

Crystallization from a protic acetic acid solvent leads to a different pattern of hydrogen bonding and intermolecular arrangement in the solid. Thus, in the acetic acid complex the BAH host interacts simultaneously with two guest moieties approaching from opposite sides. The NH(11) function donates its proton to one acid molecule, while the carbonyl group acts as acceptor from the carboxylic group of another molecule. Correspondingly (considering the host–guest stoichiometric ratio of 1:1), each molecule of acetic acid is bound to two adjacent hosts, thus forming an H-bond stabilized cluster of four molecules (two hosts and two guests) around the crystallographic inversion

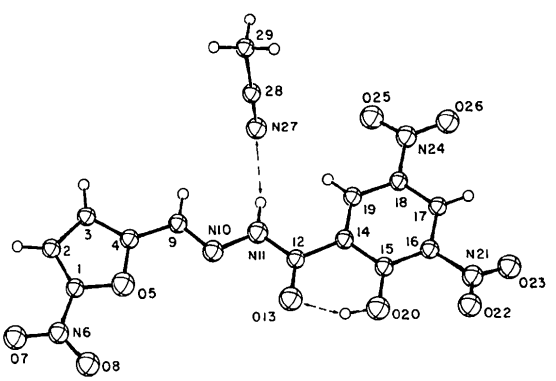

Fig. 1. Molecular structure of the 1:1 complex of BAH with acetonitrile showing the atom-numbering scheme.

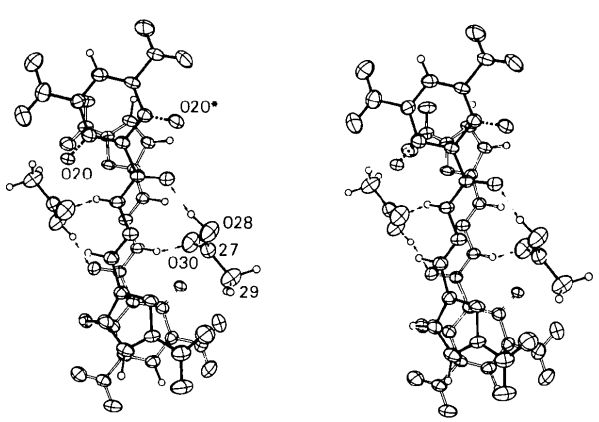

Fig. 2. Stereoview of the 1:1 complex of BAH with acetic acid showing the hydrogen-bonded dimers of centrosymmetrically related entities.

Table 2. *Bond distances* (Å) *and angles* (°) *for* (I) *and* (II) *with e.s.d.'s in parentheses*

	(I)	(II)
C(1)–C(2)	1·338 (5)	1·340 (5)
C(1)–O(5)	1·349 (3)	1·347 (4)
C(1)–N(6)	1·423 (4)	1·403 (5)
C(2)–C(3)	1·396 (4)	1·399 (6)
C(3)–C(4)	1·355 (5)	1·368 (5)
C(4)–O(5)	1·375 (3)	1·367 (4)
C(4)–C(9)	1·441 (4)	1·434 (5)
N(6)–O(7)	1·224 (4)	1·228 (5)
N(6)–O(8)	1·223 (4)	1·234 (5)
C(9)–N(10)	1·274 (4)	1·283 (5)
N(10)–N(11)	1·376 (3)	1·377 (4)
N(11)–C(12)	1·346 (4)	1·348 (5)
C(12)–O(13)	1·236 (3)	1·225 (5)
C(12)–C(14)	1·489 (4)	1·489 (6)
C(14)–C(15)	1·421 (4)	1·391 (5)
C(14)–C(19)	1·385 (4)	1·394 (6)
C(15)–C(16)	1·397 (4)	1·397 (6)
C(15)–O(20)	1·323 (3)	1·293 (9)*
C(16)–C(17)	1·375 (4)	1·357 (6)
C(16)–N(21)	1·467 (4)	1·471 (5)
C(17)–C(18)	1·373 (4)	1·370 (6)
C(18)–C(19)	1·374 (4)	1·395 (6)
C(18)–N(24)	1·466 (3)	1·448 (6)
N(21)–O(22)	1·185 (4)	1·189 (6)
N(21)–O(23)	1·181 (4)	1·203 (5)
N(24)–O(25)	1·214 (4)	1·202 (6)
N(24)–O(26)	1·221 (3)	1·230 (6)
C(19)–O(20)		1·292 (6)†

	(I)	(II)
O(5)–C(1)–C(2)	113·1 (2)	112·6 (3)
N(6)–C(1)–C(2)	131·3 (3)	130·1 (4)
N(6)–C(1)–O(5)	115·5 (3)	117·3 (3)
C(1)–C(2)–C(3)	105·3 (3)	105·7 (4)
C(2)–C(3)–C(4)	107·1 (3)	106·7 (3)
C(3)–C(4)–O(5)	110·2 (2)	109·8 (3)
C(3)–C(4)–C(9)	132·3 (3)	130·3 (3)
C(9)–C(4)–O(5)	117·4 (3)	119·8 (3)
C(4)–O(5)–C(1)	104·2 (2)	105·2 (3)
C(1)–N(6)–O(7)	116·5 (3)	119·3 (3)
C(1)–N(6)–O(8)	118·8 (3)	117·3 (3)
O(8)–N(6)–O(7)	124·7 (3)	123·4 (3)
C(4)–C(9)–N(10)	120·0 (3)	119·0 (3)
C(9)–N(10)–N(11)	115·2 (2)	114·0 (3)
N(10)–N(11)–C(12)	117·7 (2)	117·2 (3)
N(11)–C(12)–O(13)	121·0 (2)	122·5 (4)
N(11)–C(12)–C(14)	118·6 (2)	117·3 (3)
C(14)–C(12)–O(13)	120·4 (3)	120·2 (3)
C(12)–C(14)–C(15)	117·2 (2)	115·3 (3)
C(12)–C(14)–C(19)	123·1 (3)	124·5 (3)
C(19)–C(14)–C(15)	119·7 (2)	119·9 (4)
C(14)–C(15)–C(16)	117·6 (2)	117·9 (4)
C(14)–C(15)–O(20)	121·6 (2)	121·0 (5)*
O(20)–C(15)–C(16)	120·8 (3)	121·0 (5)*
C(15)–C(16)–C(17)	122·5 (3)	123·4 (4)
C(15)–C(16)–N(21)	121·3 (2)	117·8 (4)
N(21)–C(16)–C(17)	116·2 (2)	118·7 (4)
C(16)–C(17)–C(18)	118·2 (2)	117·7 (4)
C(17)–C(18)–C(19)	122·1 (2)	122·0 (4)
C(17)–C(18)–N(24)	119·3 (2)	117·4 (4)
N(24)–C(18)–C(19)	118·7 (3)	120·6 (4)
C(18)–C(19)–C(14)	120·0 (3)	119·0 (4)
C(16)–N(21)–O(22)	119·8 (3)	118·7 (4)
C(16)–N(21)–O(23)	118·4 (3)	117·5 (4)
O(23)–N(21)–O(22)	121·7 (3)	123·7 (4)
C(18)–N(24)–O(25)	118·3 (2)	120·0 (4)
C(18)–N(24)–O(26)	117·8 (3)	117·9 (4)
O(26)–N(24)–O(25)	123·8 (2)	122·0 (4)
C(14)–C(19)–O(20)		121·1 (4)†
C(18)–C(19)–O(20)		119·7 (4)†

* Bond length and angles corresponding to the minor [O(20*)] site of the disordered hydroxyl group in (II).

† Bond length and angles corresponding to the major site of the disordered hydroxyl group in (II).

Table 3. *Geometry of the hydrogen bonds*

Donor *RH*	Acceptor *R'*	*R*—H (Å)	*R*···*R'* (Å)	H···*R'* (Å)	*R*—H···*R'* (°)
(I) Acetonitrile complex					
NH(11)···N(27i)		0·91	3·019 (4)	2·12	168
OH(20)···O(13)		0·93	2·470 (3)	1·63	148
(II) Acetic acid complex					
NH(11)···O(30ii)		0·99	2·924 (6)	1·96	155
O(20)···O(30ii)†		—	2·936 (6)	—	—
O(20*)···O(13)†		—	2·376 (9)	—	—
OH(28)···O(13iii)		1·00	2·700 (5)	1·74	161

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

† The hydrogen atom of the disordered hydroxyl group has not been located.

center at $(\frac{1}{2}, 0, \frac{1}{2})$. This interaction pattern between the constituent species is associated with the observed deviation from planarity of BAH (see above) and consequent distortion of the intramolecular O—H···O bond found in the previous example. The presence of the twofold rotational disorder of the dinitrohydroxyphenyl fragment with respect to the C(12)—C(14) bond reflects a decreased affinity of the C=O(13) site for the O—H(20) proton; the conformation in which these two groups are parallel to each other is, in fact, less populated than a rotated conformation where the OH(20) group is located close to carbonyl O(30) of an adjacent molecule of acetic acid (Fig. 2). Of the two pairs of proton-donating and proton-accepting sites in this structure, NH(11) and O(30) form weaker hydrogen bonds than OH(28) and O(13) (Table 3).

The second feature is related to the crystal packing. Intermolecular arrangement of planar or nearly planar molecules containing highly polar groups was found to be dominated frequently by characteristic dipole–dipole interactions between molecules located across crystallographic centers of symmetry. Indeed, the crystal structure of the acetonitrile complex reveals many features that have recently been observed in crystals of related species (Goldberg, 1982). It can conveniently be described as composed of stacks of host molecules that extend parallel to **b**. Within the stacks adjacent molecules appear to associate by dipolar interactions in two different modes of intermolecular overlap. The average interplanar distance between molecules related by inversion at $(0, \frac{1}{2}, 0)$, which overlap almost completely, is 3·29 Å; the mean separation between hosts related by inversion at (0, 0, 0) and characterized by only partial overlap is 3·41 Å. Along the **a** translation these stacks form efficiently packed layers which are lined on both sides by the polar NO_2 groups. The packing of adjacent layers, related to each other by the glide symmetry and separated by about 8 Å along **c**, is thus strongly affected by repulsive interactions between the negative poles of the peripheral nitro substituents. Correspondingly, all interlayer interatomic contacts are larger than the sum of the respective van der Waals radii, *e.g.* O···O > 3·2 Å and O···C > 3·3 Å. Pseudochannels between the layers are filled by the acetonitrile guest moieties (Fig. 3). These extend along **b**, at $x = 0{\cdot}0$, $z = 0{\cdot}23$ and $x = 0{\cdot}0$, $z = 0{\cdot}73$.

The crystal structure of the acetic acid complex (Fig. 4), although of different symmetry, exhibits similar features. It also consists of layers of host molecules parallel to (101). Each layer is composed of stacks of molecules which are displaced with respect to each other along a direction roughly parallel to the largest dimension of the host. Along the stacking axes there are two different modes of interaction between the centrosymmetrically related and overlapping species. As described above, in addition to the dipole–dipole attractions the BAH molecules form hydrogen-bonded dimers through the acetic acid linkage, the mean separation between the overlapping regions of adjacent pairs being 3·44 Å. Side packing of the layered domains induces pseudochannels also in this structure. The acetic acid molecules included in the crystal are arranged in an alternating (zigzag) manner along the channel axis at $x = 0, z = \frac{1}{2}$.

The above observations supplement and are in agreement with the results of our previous study of solvation patterns and their relationship to structural polymorphism (Goldberg, 1983).

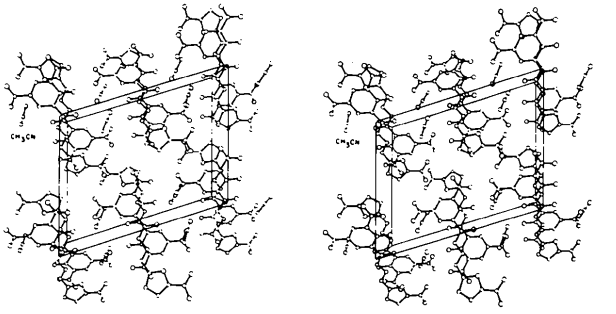

Fig. 3. Stereoview of the inclusion structure formed by BAH with acetonitrile, approximately down **b**.

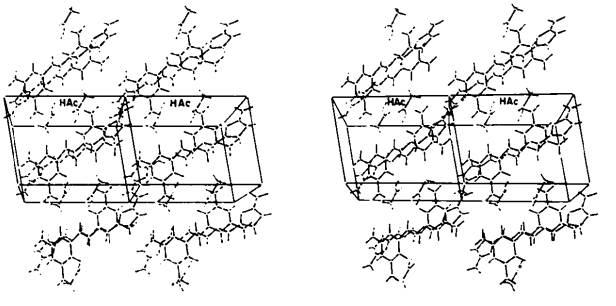

Fig. 4. Stereoview of the crystal structure of the acetic acid compound approximately down **b** showing enclosure of the solvent species in channel-type voids.

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Structure of 2,2′-Bis(4-chlorophenyl)-5,5′-dimethyl-[3,3′-bi-1,3-thiazolidine]-4,4′-dione, $C_{20}H_{18}Cl_2N_2O_2S_2$

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Abstract. $M_r = 453{\cdot}4$, monoclinic, $P2_1/n$, $a = 13{\cdot}857\ (2)$, $b = 12{\cdot}527\ (2)$, $c = 12{\cdot}199\ (2)$ Å, $\beta = 94{\cdot}58\ (3)°$, $Z = 4$, $U = 2110{\cdot}8\ (6)$ Å^3, $D_m = 1{\cdot}41$ (flotation), $D_x = 1{\cdot}426$ g cm^{-3}, μ(Mo $K\alpha$) $= 5{\cdot}2$ cm^{-1}, $F(000) = 936$, room temperature. Final conventional R value is 0·069 ($R_w = 0{\cdot}058$) for 1178 diffractometer-measured intensities. The molecular structure is characterized by the intramolecular interaction between the two parallel phenyl groups (mean distance 3·3 Å) and by the different conformations of the two thiazolidine rings, half-chair and envelope. Both enantiomers 2*R*,5*S*,2′*S*,5′*S* and 2*S*,5*R*,2′*R*,5′*S* are present in the structure.

Introduction. Substituted 2-aryl-4-thiazolidinones exhibit several interesting therapeutical properties. In particular some of them have antibacterial, antifungal, antitubercular, myorelaxant, and antiviral activity. In addition, when the H atom bound to the nitrogen in position 3 of the pentaatomic ring is substituted by the nitrogen of another 2-aryl-4-thiazolidinone ring the compound obtained exhibits properties like those of an anti-inflammatory drug instead of those previously quoted (Fenech, 1972–73). The crystal and molecular structure of the title compound (F2TD2, hereinafter) has been determined as part of an investigation of the structural features of some substituted thiazolidinones, which we are examining in an attempt to correlate molecular conformation and pharmacological activity.

Experimental. Colourless single crystals of F2TD2, suitable for an X-ray analysis, obtained by slow evaporation of their ethanolic solution; approximate unit-cell parameters estimated from preliminary Weissenberg and precession photographs, crystal size 0·2 × 0·2 × 0·3 mm, Siemens–Stoe four-circle diffractometer, graphite monochromator, Mo $K\alpha$ radiation ($\lambda = 0{\cdot}71069$ Å), accurate unit-cell parameters and crystal-orientation matrices (together with their estimated standard errors) obtained from least-squares refinement of the 2θ, ω, χ and φ values of 20 carefully centred high-angle (θ range 6–12°) reflections; ω/θ scan, scan width 1·4°, scan speed 0·04° s^{-1}, 2θ range 6–50°, two standard reflections (154 and $\bar{5}01$)

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