Table 2. *Selected bond lengths* (Å) *and angles* (°)

Cr—Cr′	2.446 (2)	Cr—O(3)	2.243 (5)
Cr—O(11)	2.016 (4)	Cr—O(21)	2.027 (3)
Cr—O(12′)	2.022 (4)	Cr—O(22′)	2.019 (3)
Zn—Cl(1)	2.295 (2)	Zn—Cl(2)	2.241 (2)
Zn—Cl(3)	2.243 (2)	Zn—Cl(4)	2.252 (2)

O(11)—Cr—Cr′	86.3 (1)	O(21)—Cr—Cr′	85.7 (1)
O(3)—Cr—Cr′	174.6 (1)	Cr′—Cr—O(12′)	87.7 (1)
Cr′—Cr—O(22′)	88.0 (1)	O(11)—Cr—O(3)	92.4 (2)
O(21)—Cr—O(3)	89.1 (2)	O(3)—Cr—O(12′)	93.7 (2)
O(3)—Cr—O(22′)	97.2 (2)	O(11)—Cr—O(21)	92.3 (1)
O(11)—Cr—O(12′)	173.9 (2)	O(21)—Cr—O(12′)	88.1 (1)
O(11)—Cr—O(22′)	89.0 (1)	O(21)—Cr—O(22′)	173.5 (2)
O(12′)—Cr—O(22′)	90.0 (1)	Cl(1)—Zn—Cl(2)	108.2 (1)
Cl(1)—Zn—Cl(3)	109.5 (1)	Cl(2)—Zn—Cl(3)	113.1 (1)
Cl(1)—Zn—Cl(4)	108.3 (1)	Cl(2)—Zn—Cl(4)	110.0 (1)
Cl(3)—Zn—Cl(4)	107.7 (1)		

Symmetry operation for primed atoms: $1-x$, $1-y$, $1-z$.

The complex was prepared from chromium(II) chloride, zinc amalgam, betaine and dilute HCl in a Jones reductor-column apparatus under a nitrogen atmosphere and crystals were obtained by solvent-volume reduction and cooling (60% yield, satisfactory chemical analyses). As the product is air sensitive, the crystal for X-ray study was sealed in a nitrogen-filled Lindemann capillary with traces of mother liquor.

Data collection and reduction: Stoe *DIF*4 software 1984. Program used to solve, refine and display structure: *SHELXTL* (Sheldrick, 1985). H atoms constrained [C—H 0.96 Å, H—C—H 109.5°, $U(H) = 1.2U_{eq}(C)$], no H atoms for H_2O. The scan range was from 0.51° below α_1 to 0.51° above α_2 with scan times of 14-56 s.

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Lists of structure factors, anisotropic thermal parameters, H-atom coordinates and complete geometry have been deposited with the British Library Document Supply Centre as Supplementary Publication No. SUP 55210 (20 pp.). Copies may be obtained through The Technical Editor, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England. [CIF reference: MU1005]

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Structure of 2-[(4-Methylphenylimino)methyl]-1-nitrobenzene

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Abstract

The benzylideneaniline moiety adopts a distorted planar conformation with a dihedral angle of 3.9 (5)° between the two phenyl rings. The nitro group is twisted from the phenyl ring by 27.9 (5)°.

Comment

Although benzylideneaniline adopts an energetically favoured non-planar conformation (Bürgi & Dunitz, 1970; Bernstein, Engel & Hagler, 1981), some of its derivatives with homo or hetero *para* substituents prefer a planar conformation (Bar & Bernstein, 1983). Different kinds of chemical substitutions at different positions in these derivatives result in distorted planar conformations (Fun, Teo, Teoh & Yeap, 1991). The title compound, which is extensively employed in the synthesis and characterization of adducts (Srivastava & Chauhan, 1977), is found to exhibit a slightly distorted planar conformation

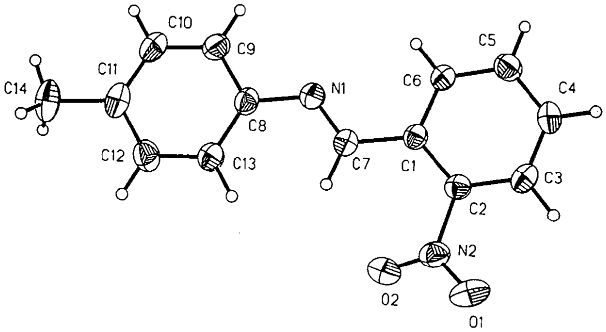

Fig. 1. A view of the molecule showing the labelling of the non-H atoms. Thermal ellipsoids are shown at 50% probability levels.

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[C7—N1—C8—C13 = 3.9 (4) and N1—C7—C1—C6 = −5.0 (4)°]. The NO_2 plane is twisted out of the plane of the phenyl ring by 27.9 (5)° which minimizes the steric hindrance between the O atoms and both H7 and H3 but allows some electron delocalization with the phenyl ring. The close O1···H3 and O2···H7 contacts of 2.419 (4) and 2.179 (3) Å respectively are a result of this compromise.

Experimental

Crystal data

$C_{14}H_{12}N_2O_2$
$M_r = 240.26$
Monoclinic
$C2/c$
$a = 11.362$ (2) Å
$b = 9.962$ (1) Å
$c = 22.113$ (3) Å
$\beta = 104.01$ (1)°
$V = 2428.5$ (7) Å^3
$Z = 8$
$D_x = 1.314$ Mg m^{-3}
$D_m = 1.309$ Mg m^{-3}
Mo $K\alpha$ radiation
$\lambda = 0.71069$ Å
Cell parameters from 30 reflections
$\theta = 35$-$45°$
$\mu = 0.08$ mm^{-1}
$T = 298$ K
Needle
0.3 × 0.2 × 0.2 mm
Yellow

Data collection

Syntex $P2_1$ diffractometer
$2\theta/\theta$ scans
Absorption correction: empirical
$T_{min} = 0.8443$, $T_{max} = 0.9986$
2730 measured reflections
2461 independent reflections
1840 observed reflections [$I>2.5\sigma(I)$]
$R_{int} = 0.0369$
$\theta_{max} = 52.2°$
$h = 0 \rightarrow 10$
$k = 0 \rightarrow 14$
$l = -12 \rightarrow 12$
1 standard reflection monitored every 50 reflections
intensity variation: none

Refinement

Refinement on F
Final $R = 0.0526$
$wR = 0.0610$
$S = 1.46$
1840 reflections
175 parameters
Only H-atom U's refined
$w = 3.0743/[\sigma^2(F)+0.000531F^2]$
$(\Delta/\sigma)_{max} = 0.003$
$\Delta\rho_{max} = 0.214$ e Å^{-3}
$\Delta\rho_{min} = -0.171$ e Å^{-3}
Atomic scattering factors from *SHELX*76

Table 1. *Fractional atomic coordinates and equivalent isotropic thermal parameters* (Å^2)

$$B_{eq} = \tfrac{8}{3}\pi^2\Sigma_i\Sigma_jU_{ij}a_i^*a_j^*\mathbf{a}_i.\mathbf{a}_j.$$

	x	y	z	B_{eq}
N1	0.9206 (2)	0.3023 (2)	0.5004 (1)	5.69 (6)
N2	0.7330 (2)	0.2584 (2)	0.3113 (1)	5.17 (6)
O1	0.6546 (2)	0.2153 (2)	0.2687 (1)	7.42 (6)
O2	0.7374 (2)	0.3757 (2)	0.3272 (1)	8.40 (7)
C1	0.8939 (2)	0.1885 (2)	0.4046 (1)	3.88 (5)
C2	0.8305 (2)	0.1674 (1)	0.3429 (1)	4.03 (5)
C3	0.8549 (2)	0.0615 (2)	0.3076 (1)	4.64 (6)
C4	0.9446 (2)	−0.0277 (2)	0.3333 (1)	5.12 (6)
C5	1.0078 (2)	−0.0119 (2)	0.3945 (1)	5.20 (7)
C6	0.9831 (2)	0.0946 (2)	0.4290 (1)	4.51 (6)
C7	0.8705 (2)	0.2970 (2)	0.4453 (1)	4.96 (6)
C8	0.8995 (2)	0.4069 (2)	0.5398 (1)	4.55 (6)
C9	0.9679 (2)	0.4047 (2)	0.6009 (1)	5.25 (6)
C10	0.9524 (2)	0.5017 (2)	0.6427 (1)	5.81 (7)
C11	0.8690 (2)	0.6032 (2)	0.6256 (1)	5.12 (7)
C12	0.8016 (2)	0.6056 (2)	0.5647 (1)	5.77 (7)
C13	0.8165 (2)	0.5094 (2)	0.5223 (1)	5.65 (7)
C14	0.8509 (3)	0.7091 (3)	0.6715 (1)	7.21 (9)

Table 2. *Geometric parameters* (Å, °)

N1—C7	1.215 (3)	C4—C5	1.379 (3)
N1—C8	1.415 (3)	C5—C6	1.375 (3)
N2—O1	1.208 (3)	C8—C9	1.386 (3)
N2—O2	1.218 (3)	C8—C13	1.380 (3)
N2—C2	1.470 (3)	C9—C10	1.378 (3)
C1—C2	1.396 (3)	C10—C11	1.375 (3)
C1—C6	1.389 (3)	C11—C12	1.379 (3)
C1—C7	1.471 (3)	C11—C14	1.512 (4)
C2—C3	1.380 (3)	C12—C13	1.379 (3)
C3—C4	1.368 (3)		
C7—N1—C8	122.8 (2)	C1—C6—C5	122.1 (2)
O2—N2—C2	118.7 (2)	N1—C7—C1	122.4 (2)
O1—N2—C2	118.7 (2)	N1—C8—C13	125.3 (2)
O1—N2—O2	122.5 (2)	N1—C8—C9	116.7 (2)
C6—C1—C7	118.7 (2)	C9—C8—C13	117.9 (2)
C2—C1—C7	125.6 (2)	C8—C9—C10	120.7 (2)
C2—C1—C6	115.7 (2)	C9—C10—C11	121.5 (2)
N2—C2—C1	121.5 (2)	C10—C11—C14	121.7 (2)
C1—C2—C3	122.8 (2)	C10—C11—C12	117.7 (2)
N2—C2—C3	115.7 (2)	C12—C11—C14	120.6 (2)
C2—C3—C4	119.5 (2)	C11—C12—C13	121.4 (2)
C3—C4—C5	119.5 (2)	C8—C13—C12	120.7 (2)
C4—C5—C6	120.3 (2)		

Program used to solve the structure: *SHELXS*86 (Sheldrick, 1985). Program used to refine structure: *SHELX*76 (Sheldrick, 1976). Program used for absorption correction: *ABSORB* (Ugozzoli, 1987). Program used for geometrical calculations: *XANADU* (Robert & Sheldrick, 1975). Program used for molecular drawing: *ORTEP* (Johnson, 1965). A variable scan speed of 5.0-29.3° min^{-1} was used for data collection. Idealized riding H atoms (C—H 0.96 Å).

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Structure of an Oxonium Salt Crystal Complex

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Abstract

The title complex is composed of two compounds, oxonium 1-cyano-2-hydrazino-2-oxoethane-1-nitronate and 3-amino-4-nitro-1,2-dihydro-5-pyrazolone monohydrate (1). The anion is nearly planar and is stabilized by delocalization of the negative charge. The oxonium ion is involved in an O—H···N hydrogen bond to the terminal N atom in the hydrazide group. The pyrazolone rings and the anions stack nearly parallel to the 101 plane in the crystal structure which is stabilized by intermolecular hydrogen bonds.

Comment

2-Cyano-2-nitroacetohydrazide monohydrate (2) was first synthesized and described by Darapsky & Hillers (1915); there are five plausible tautomeric forms. The present investigation is part of a study of aliphatic nitro compounds with acid properties and has been undertaken in order to determine the actual tautomeric form of (2). The investigation revealed that the title complex (1) is a crystal composed of two compounds, (2) and the isomeric 3-amino-4-nitro-1,2-dihydro-5-pyrazolone (3). This is comprehensible as (2) easily rearranges to (3) by heating or by standing in solution at room temperature (Darupsky & Hillers, 1915).

Bond distances and bond angles for the anion are given in Table 2. The structural formula with the atomic numbering scheme is shown in Fig. 1. The sum of the angles around C2 is 360 (1)° and a least-squares plane defined by all non-H atoms in (2) apart from N4 reveals a nearly planar system with a maximum distance of −0.06 Å (for O3) from the least-squares plane. Hence C2 is sp^2-hybridized. In Table 3, a comparison is made between selected bond distances in (1) and the corresponding bonds in related compounds. The table shows the effect on cyanoacetohydrazide bond lengths of substituting H with a nitro group at C2: C1—C2 and C2—C3 are shortened, C3—N3 is elongated, C1—O1 is unchanged and C1—N1 is probably extended by 0.02 Å. The C2—N2 length [1.357 (8) Å] is characteristic of a C—N partial double bond and is comparable to the corresponding bond [1.327 (4) Å] in the nitromalonamide anion (Simonsen, 1981). Three resonance formulae are proposed to describe the delocalization of the negative charge in (2), each with

Table 1. *Fractional atomic coordinates and equivalent isotropic thermal parameters* (Å^2)

$$U_{\rm eq} = \tfrac{1}{3}\Sigma_i\Sigma_jU_{ij}a_i^*a_j^*\mathbf{a}_i.\mathbf{a}_j.$$

	x	y	z	$U_{\rm eq}$
O1	0.4313 (8)	0.4197 (6)	0.4031 (3)	0.036 (4)
O2	0.6718 (8)	0.7273 (6)	0.1461 (4)	0.043 (5)
O3	0.7342 (9)	0.4724 (6)	0.0753 (3)	0.045 (5)
O4†	0.0683 (9)	0.6443 (7)	0.5591 (4)	0.048 (5)
O15	0.9282 (8)	0.9296 (6)	−0.3253 (3)	0.038 (4)
O41‡	0.7478 (9)	0.9089 (7)	0.4645 (4)	0.043 (5)
O141	0.9440 (9)	1.3127 (7)	−0.2909 (4)	0.055 (5)
O142	0.8259 (9)	1.3751 (7)	−0.1409 (4)	0.051 (5)
N1	0.502 (1)	0.7107 (7)	0.3326 (4)	0.033 (5)
N2	0.673 (1)	0.5501 (8)	0.1529 (4)	0.030 (5)
N3	0.606 (1)	0.0896 (8)	0.2398 (4)	0.041 (6)
N4	0.3988 (9)	0.7958 (7)	0.4165 (4)	0.030 (5)
N11	0.861 (1)	0.7620 (7)	−0.1679 (4)	0.033 (5)
N12	0.800 (1)	0.8042 (8)	−0.0718 (4)	0.031 (5)
N13	0.743 (1)	1.081 (1)	0.0062 (4)	0.043 (6)
N14	0.8736 (9)	1.2625 (8)	−0.2005 (5)	0.035 (5)
C1	0.505 (1)	0.5236 (9)	0.3305 (5)	0.023 (5)
C2	0.597 (1)	0.4493 (9)	0.2399 (5)	0.025 (6)
C3	0.603 (1)	0.251 (1)	0.2393 (5)	0.030 (6)
C13	0.796 (1)	0.9907 (9)	−0.0722 (5)	0.029 (6)
C14	0.856 (1)	1.0708 (9)	−0.1717 (5)	0.024 (5)
C15	0.890 (1)	0.9257 (9)	−0.2328 (5)	0.030 (6)

†H_2O.
‡H_3O^+.

Table 2. *Geometric parameters* (Å, °)

(2)			
O1—C1	1.220 (7)	N2—C2	1.357 (8)
O2—N2	1.260 (7)	N3—C3	1.157 (9)
O3—N2	1.272 (7)	C1—C2	1.446 (9)
N1—N4	1.419 (8)	C2—C3	1.42 (1)
N1—C1	1.346 (8)		
N4—N1—C1	121.1 (5)	N1—C1—C2	118.2 (5)
O2—N2—O3	118.3 (5)	N2—C2—C1	126.9 (6)
O2—N2—C2	119.9 (5)	N2—C2—C3	115.4 (6)
O3—N2—C2	121.7 (6)	C1—C2—C3	117.7 (5)
O1—C1—N1	120.8 (6)	N3—C3—C2	179.2 (7)
O1—C1—C2	121.0 (6)		
(3)			
O15—C15	1.268 (9)	N12—C13	1.334 (9)
O141—N14	1.273 (8)	N13—C13	1.33 (1)
O142—N14	1.223 (8)	N14—C14	1.385 (8)
N11—N12	1.399 (8)	C13—C14	1.422 (9)
N11—C15	1.376 (8)	C14—C15	1.42 (1)
N12—N11—C15	110.4 (5)	N13—C13—C14	127.6 (6)
N11—N12—C13	109.3 (5)	N14—C14—C13	123.1 (6)
O141—N14—O142	122.7 (6)	N14—C14—C15	127.4 (6)
O141—N14—C14	116.5 (6)	C13—C14—C15	109.5 (6)
O142—N14—C14	120.9 (6)	O15—C15—N11	123.3 (7)
N12—C13—N13	125.7 (6)	O15—C15—C14	132.6 (6)
N12—C13—C14	106.7 (6)	N11—C15—C14	104.0 (6)

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