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## 3,5,5-Triphenyl-4,5-dihydro-1,2,4-oxadiazole

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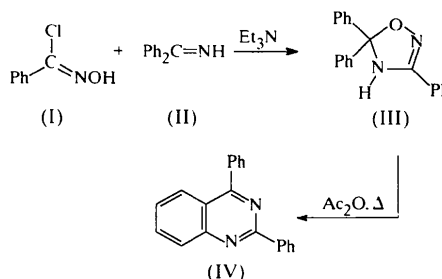
(Received 20 July 1999; accepted 30 September 1999)

## Abstract

Reaction of benzophenone imine with benzeneoximoyl chloride performed in the presence of triethylamine in diethyl ether affords the title compound, C<sub>20</sub>H<sub>16</sub>N<sub>2</sub>O. In the crystalline state, the product exists in two conformations differing in the orientation of the 3-phenyl substituent with respect to the heterocyclic ring. N—H···N hydrogen bonds link the molecules into chains parallel to the *b* axis.

## Comment

Benzeneoximoyl chloride, (I), in the presence of triethylamine yields benzonitrile *N*-oxide which, by undergoing a 1,3-dipolar cycloaddition reaction to benzophenone imine, (II), should easily give 3,5,5-triphenyl-4,5-dihydro-1,2,4-oxadiazole, (III) (De Micheli *et al.*, 1974). The latter when treated with hot acetic anhydride gave 2,4-triphenylquinazoline, (IV), in good yield. As it was expected that (III) would be acylated on the N atom, we suspected that transformation of (III) to (IV) either must have involved *O*-acetylation or the assumption concerning the structure of (III) must have been incorrect. The rather unexpected result of the overall reaction encouraged us to verify unequivocally the identity of (III) using crystallographic methods.



X-ray diffraction analysis of (III) has ultimately proved its structure as 3,5,5-triphenyl-4,5-dihydro-1,2,4-oxadiazole. Compound (III) exists as two conformers in the crystalline state. The key difference is the orientation of the phenyl substituent at C1 with respect to the five-membered ring. The dihedral angles between the phenyl and heterocyclic rings are 31.1 (1) and 27.1 (1)° for the first (C1–N5 and C6–C11) and the second conformer (C1A–N5A and C6A–C11A), respectively. The molecules also show slight differences in the C1–N2–C3 valence angle and in the C1–N2 bond length. It is worth noting that the latter is considerably shortened compared with a C–N single bond and can be classified as aromatic. This effect may be ascribed to the ‘background delocalization phenomenon’ (Słowikowska *et al.*, 1998) associated with the presence of a C=N double bond at C1. In both conformers, the heterocyclic ring is not planar but adopts a three-envelope conformation, as confirmed by the asymmetry parameters ( $\Delta C_3^3 = 0.3$ ,  $\Delta C_2^1 = 8.386$ ;  $\Delta C_5^{3A} = 1.651$ ,  $\Delta C_2^{5A} = 8.065^\circ$ ; Duax & Norton, 1975).

The molecules are involved in intermolecular N—H···N hydrogen bonds connecting translationally related molecules in infinite chains parallel to the *b* axis. Along *a*, chains formed alternately by the two conformers are ordered in an *A–B–A–B* fashion running, accordingly, in opposite directions. Additionally, several weaker intermolecular C—H···O, C—H···N and N—H···O contacts occur. Based on the criteria given by Pedireddi & Desiraju (1992), most of them also qualify as very weak hydrogen bonds.

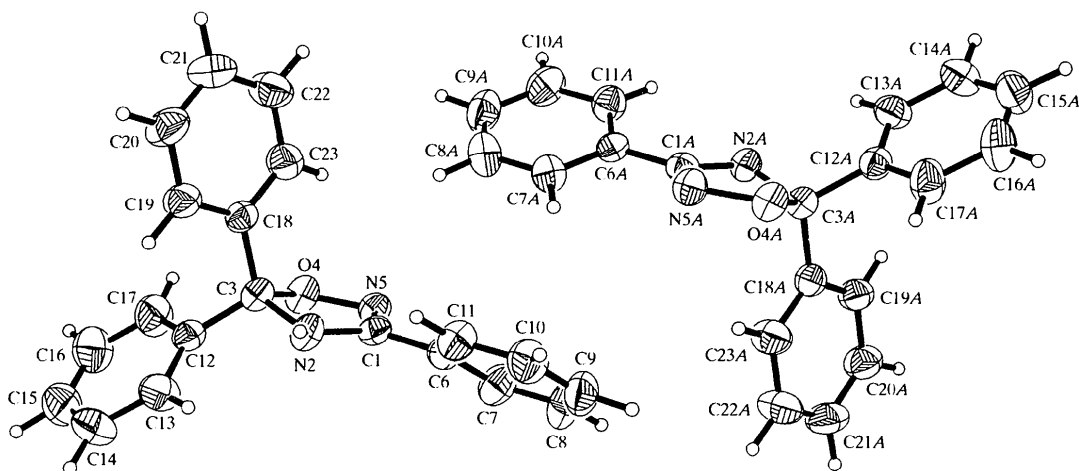


Fig. 1. The molecular structure of (III) showing 50% probability displacement ellipsoids.

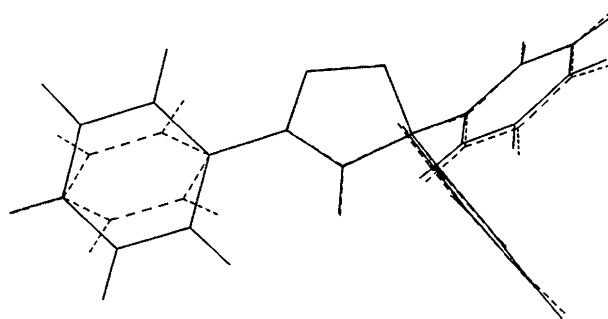


Fig. 2. Superposition of the two conformers. Fitting was performed on all non-H atoms.

A rather high value for the valence angle H2N—N2—C1 of 122.3 (19)° may be indicative of low nucleophilicity of the N atom. This can be helpful in explaining the unexpected course of the reaction of (III) with acetic anhydride. Details of the novel transformation of oxadiazolines to quinazolines will be discussed elsewhere.

## Experimental

A solution of triethylamine (0.01 mol) in diethyl ether (20 ml) was added to a stirred solution of freshly prepared benzene-oximoyl chloride, obtained from 0.01 mol of  $\alpha$ -benzaloxime (Baruah *et al.*, 1988) and dissolved in diethyl ether (50 ml) at a temperature below 273 K. This was followed by the addition of a benzophenone imine (0.01 mol) (Cristau *et al.*, 1998) solution in diethyl ether (30 ml). The resulting mixture was stirred for 24 h without cooling, extracted with water (30 ml), 15% hydrochloric acid (3  $\times$  30 ml), and again with water. The organic layer was collected, dried over anhydrous magnesium sulfate and evaporated under reduced pressure. The residue was recrystallized from methanol to give (III) in 58% yield (m.p. 450–451 K).  $M_r^* = 300$ ;  $^1\text{H NMR}$  ( $\text{CDCl}_3$ ):  $\delta$  5.16 (s, 1H, NH), 7.33–7.43 (m, 9H, all *m*- and *p*-protons), 7.52–7.55 (m, 4H, *o*-protons), 7.73–7.76 (m, 2H, *o*-protons).

## Crystal data

$\text{C}_{20}\text{H}_{16}\text{N}_2\text{O}$   
 $M_r = 300.35$   
 Monoclinic  
 $P2_1/c$   
 $a = 16.3000$  (8) Å  
 $b = 5.4550$  (10) Å  
 $c = 34.976$  (2) Å  
 $\beta = 90.667$  (4)°  
 $V = 3109.7$  (6) Å<sup>3</sup>  
 $Z = 8$   
 $D_x = 1.283$  Mg m<sup>-3</sup>  
 $D_m$  not measured

Cu K $\alpha$  radiation  
 $\lambda = 1.54178$  Å  
 Cell parameters from 46 reflections  
 $\theta = 5.97$ –26.34°  
 $\mu = 0.632$  mm<sup>-1</sup>  
 $T = 293$  (2) K  
 Plate  
 0.50  $\times$  0.25  $\times$  0.15 mm  
 Colourless

## Data collection

KM-4 (Kuma Diffraction) diffractometer  
 $\omega$ -2 $\theta$  scans  
 Absorption correction: none  
 6705 measured reflections  
 6482 independent reflections  
 2686 reflections with  $I > 2\sigma(I)$

$R_{\text{int}} = 0.025$   
 $\theta_{\text{max}} = 78.22^\circ$   
 $h = 0 \rightarrow 20$   
 $k = 0 \rightarrow 6$   
 $l = -44 \rightarrow 44$   
 3 standard reflections every 150 reflections  
 intensity decay: 0.67%

## Refinement

Refinement on  $F^2$   
 $R[F^2 > 2\sigma(F^2)] = 0.040$   
 $wR(F^2) = 0.151$   
 $S = 0.937$   
 5150 reflections  
 544 parameters  
 All H atoms refined  
 $w = 1/[\sigma^2(F_o^2) + (0.0690P)^2]$   
 where  $P = (F_o^2 + 2F_c^2)/3$   
 $(\Delta/\sigma)_{\text{max}} < 0.001$

$\Delta\rho_{\text{max}} = 0.202$  e Å<sup>-3</sup>  
 $\Delta\rho_{\text{min}} = -0.173$  e Å<sup>-3</sup>  
 Extinction correction: SHELXL93 (Sheldrick, 1993)  
 Extinction coefficient: 0.0016 (2)  
 Scattering factors from *International Tables for Crystallography* (Vol. C)

Table 1. Selected geometric parameters (Å, °)

C1—N5	1.294 (3)	C1A—N5A	1.290 (3)
C1—N2	1.358 (3)	C1A—N2A	1.372 (3)
C1—C6	1.469 (3)	C1A—C6A	1.470 (3)
N2—C3	1.469 (3)	N2A—C3A	1.465 (3)
N2—H2N	0.89 (3)	N2A—H2NA	0.92 (3)

C3—O4	1.459 (3)	C3A—O4A	1.456 (3)
C3—C12	1.512 (3)	C3A—C12A	1.517 (3)
C3—C18	1.520 (3)	C3A—C18A	1.522 (3)
O4—N5	1.436 (2)	O4A—N5A	1.438 (2)
N5—C1—N2	114.5 (2)	N5A—C1A—N2A	114.6 (2)
N5—C1—C6	121.8 (2)	N5A—C1A—C6A	121.7 (2)
N2—C1—C6	123.7 (2)	N2A—C1A—C6A	123.7 (2)
C1—N2—C3	107.6 (2)	C1A—N2A—C3A	106.4 (2)
C1—N2—H2N	122.3 (19)	C1A—N2A—H2NA	118.2 (15)
C3—N2—H2N	121.3 (19)	C3A—N2A—H2NA	117.9 (16)
O4—C3—N2	100.1 (2)	O4A—C3A—N2A	100.4 (2)
O4—C3—C12	107.1 (2)	O4A—C3A—C12A	107.3 (2)
N2—C3—C12	114.5 (2)	N2A—C3A—C12A	113.9 (2)
O4—C3—C18	110.2 (2)	O4A—C3A—C18A	109.6 (2)
N2—C3—C18	112.3 (2)	N2A—C3A—C18A	112.8 (2)
C12—C3—C18	111.9 (2)	C12A—C3A—C18A	112.1 (2)
N5—O4—C3	109.3 (2)	N5A—O4A—C3A	109.1 (2)
C1—N5—O4	105.5 (2)	C1A—N5A—O4A	105.2 (2)
N5—C1—C6—C7	-32.2 (4)	N5A—C1A—C6A—C7A	-25.0 (4)

Table 2. Hydrogen-bonding geometry (Å, °)

D—H...A	D—H	H...A	D...A	D—H...A
N2—H2N...N5 <sup>i</sup>	0.89 (3)	2.44 (3)	3.287 (3)	160 (3)
N2A—H2NA...N5A <sup>ii</sup>	0.92 (3)	2.45 (3)	3.303 (3)	155 (2)

Symmetry codes: (i)  $x, 1 + y, z$ ; (ii)  $x, y - 1, z$ .

The asymmetric unit contained two molecules of (III) related approximately by a centre of symmetry and differing in the orientation of the phenyl ring at C1. However, inspection of the *hkl* intensities did not allow a transformation to a smaller unit cell. Refined C—H distances are in the range 0.92 (3)–1.07 (4) Å.

Data collection: *KM4B8* (Gałdecki *et al.*, 1997a). Cell refinement: *KM4B8*. Data reduction: *DATAPROC* (Gałdecki *et al.*, 1997b). Program(s) used to solve structure: *SHELXS86* (Sheldrick, 1990a). Program(s) used to refine structure: *SHELXL93* (Sheldrick, 1993). Molecular graphics: *SHELXTL/PC* (Sheldrick, 1990b). Software used to prepare material for publication: *SHELXL93*.

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Supplementary data for this paper are available from the IUCr electronic archives (Reference: SK1322). Services for accessing these data are described at the back of the journal.

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## 2-Tosylaminobenzaldehyd†

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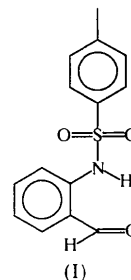
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## Abstract

The title molecule, C<sub>14</sub>H<sub>13</sub>NO<sub>3</sub>S, is not planar; the angle between the two aromatic rings is 81.85 (6)°. The conformation of the molecule in the solid state is determined by an intramolecular N—H...O hydrogen bond. This interaction modifies the electrophilic character of the C atom in the carboxaldehyde group, rendering it more suitable for a nucleophilic attack.

## Comment

The preparation of the title compound, (I), is the first step in the synthesis of dianionic Schiff bases (Bailey *et al.*, 1974; Bailey & McKenzie, 1980) that could act as polydentate ligands through both N and O atoms.



The crystal structures of the ligands *N,N'*-bis(2-tosylaminobenzylidene)-1,3-diaminopropane (Mahía *et al.*, 1999) and 2-isopropylidene-1-(*p*-toluenesulfonyl)-hydrazide (Ojala *et al.*, 1998), and of the mononuclear

† Alternative name: *N*-(*o*-formylphenyl)-4-toluenesulfonamide.