

# Synthesis and Single Crystal X-Ray Structure of 2-(1,3,4-Oxadiazol-2-yl)aniline

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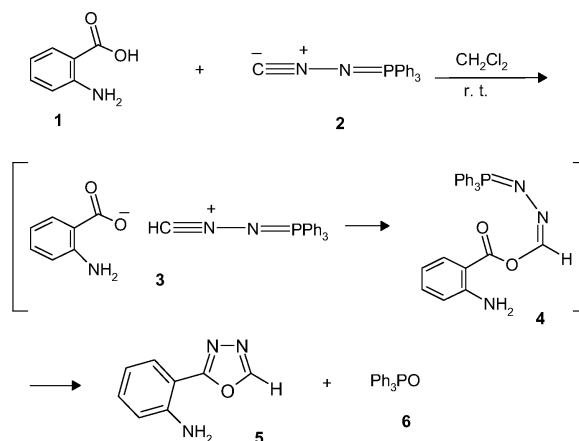
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Reaction of 2-aminobenzoic acid with (*N*-isocyanimino)triphenylphosphorane proceeds smoothly at r. t. to afford 2-(1,3,4-oxadiazol-2-yl)aniline in high yield. The structure of this compound was confirmed by IR, <sup>1</sup>H, and <sup>13</sup>C NMR spectroscopy, mass spectrometry, and single crystal X-ray structure determination. The X-ray structural analysis of the product indicated that its aromatic rings are approximately co-planar.

**Key words:** 2-Aminobenzoic Acid, (*N*-Isocyanimino)triphenylphosphorane, 2-(1,3,4-Oxadiazol-2-yl)aniline, *Aza*-Wittig Reaction, Crystal Structure Determination

## Introduction

1,3,4-Oxadiazoles have attracted interest in medicinal chemistry as surrogates of carboxylic acids, esters, and carboxamides. They are an important class of heterocyclic compounds that have a wide range of pharmaceutical and biological activities being, among others, anti-microbial, anti-fungal, anti-inflammatory, and anti-hypertensive [1–5]. Several methods have been reported in the literature for the synthesis of 1,3,4-oxadiazoles. These protocols are routinely multi-step in nature [6–11]. The most general method involves the cyclization of diacylhydrazides with a variety of reagents, such as thionyl chloride, phosphorus oxychloride, and sulfuric acid, usually under harsh reaction conditions. Few reliable and operationally facile processes have been reported for the one step synthesis of 1,3,4-oxadiazoles, especially from readily available carboxylic acids and acid hydrazides [12–16]. In the last years several synthetic methods have been reported for the preparation of (*N*-isocyanimino)triphenylphosphorane (CNNPPH<sub>3</sub>) (**2**) (Scheme 1) [17–18]. There are several reports for the use of (*N*-isocyanimino)triphenylphosphorane (**2**) in the synthesis of metal complexes [17–18]. However, application of (*N*-isocyanimino)triphenylphosphorane (**2**) in the synthesis of organic compounds has not been reported. As part of our ongoing program to develop efficient and robust methods for the preparation of het-



Scheme 1.

erocyclic compounds [19–21], we sought to examine the reaction of 2-aminobenzoic acid (**1**) with (*N*-isocyanimino)triphenylphosphorane (**2**) (Scheme 1). In this paper, we describe the synthesis and crystal structure of 2-(1,3,4-oxadiazol-2-yl)aniline.

## Results and Discussion

2-Aminobenzoic acid (**1**) and (*N*-isocyanimino)triphenylphosphorane (**2**) in dichloromethane undergo a smooth 1:1 addition reaction at r. t. to produce 2-(1,3,4-oxadiazol-2-yl)aniline (**5**) and triphenylphosphine oxide (**6**) (Scheme 1). The reaction proceeds

Table 1. Crystal data and structure refinement details for **5**.

Crystal data:	
Empirical formula	C <sub>8</sub> H <sub>7</sub> N <sub>3</sub> O
Formula weight, g mol <sup>-1</sup>	161.17
Crystal system, space group	monoclinic, <i>P</i> 2 <sub>1</sub> / <i>n</i>
<i>a</i> [Å]	10.547(3)
<i>b</i> [Å]	9.801(3)
<i>c</i> [Å]	14.460(4)
$\beta$ [deg]	101.07(3)
<i>V</i> [Å <sup>3</sup> ]	1466.9(7)
<i>Z</i>	8
<i>D</i> <sub>calc</sub> [g cm <sup>-3</sup> ]	1.459
$\mu$ [mm <sup>-1</sup> ]	0.102
<i>F</i> (000) [e]	672
Crystal size [mm <sup>3</sup> ]	0.50 × 0.50 × 0.42
Crystal color and form	yellowish block
Data collection:	
Diffractometer	Kuma KM4CCD
Data collection method	$\omega$ scans
Monochromator	graphite
Radiation type, $\lambda$ [Å]	MoK $\alpha$ , 0.71073
<i>T</i> , K	100(2)
$\theta$ Range [deg]	3.37–29.98
<i>h</i> , <i>k</i> , <i>l</i> Ranges	–13 ≤ <i>h</i> ≤ 14, –12 ≤ <i>k</i> ≤ 12, –19 ≤ <i>l</i> ≤ 20
Measured reflections	12283
Independent reflections	3866
Observed refl. [ <i>I</i> ≥ 2 $\sigma$ ( <i>I</i> )]	2897
Completeness to $\theta$ = 30.00°	0.998
Refinement:	
Refinement on	<i>F</i> <sup>2</sup>
Data/restraints/parameters	3866/0/273
<i>R</i> [ <i>F</i> <sub>o</sub> <sup>2</sup> ≥ 2 $\sigma$ ( <i>F</i> <sub>o</sub> <sup>2</sup> )]	<i>R</i> 1 = 0.0405, <i>wR</i> 2 = 0.1039
<i>R</i> (all data)	<i>R</i> 1 = 0.0557, <i>wR</i> 2 = 0.1094
Goof = <i>S</i>	1.008
Weighting parameter <i>a/b</i>	0.0740/0.0
$\Delta\rho_{\max}/\Delta\rho_{\min}$ [e Å <sup>-3</sup> ]	0.29/–0.28

$R1 = \sum||F_o| - |F_c|| / \sum|F_o|$ ;  $wR2 = \sqrt{\sum[w(F_o^2 - F_c^2)^2] / \sum[w(F_o^2)^2]}$ ; weighting scheme:  $w = 1 / [\sigma^2(F_o^2) + (aP)^2 + bP]$  where  $P = (F_o^2 + 2F_c^2) / 3$ .

smoothly and cleanly under mild conditions and no side reactions were observed. The mechanism of the reaction between **1** and **2** has not been established experimentally. However, a possible explanation is proposed in Scheme 1. On the basis of the well established chemistry of isocyanides [22], it is reasonable to assume that protonation of the zwitterionic isocyanide **2** by 2-aminobenzoic acid (**1**), followed by quenching of the cationic center by the conjugate base of the acid, can generate the iminophosphorane **4** [23]. Intramolecular *aza*-Wittig [23] reaction of the iminophosphorane **4** leads to the formation of the 2-(1,3,4-oxadiazol-2-yl)aniline (**5**) and triphenylphosphine oxide (**6**). The structure of the 2-(1,3,4-oxadiazol-2-yl)aniline (**5**) was confirmed by IR, <sup>1</sup>H, and <sup>13</sup>C NMR spectroscopy, mass spectrometry, and single crystal X-ray structure

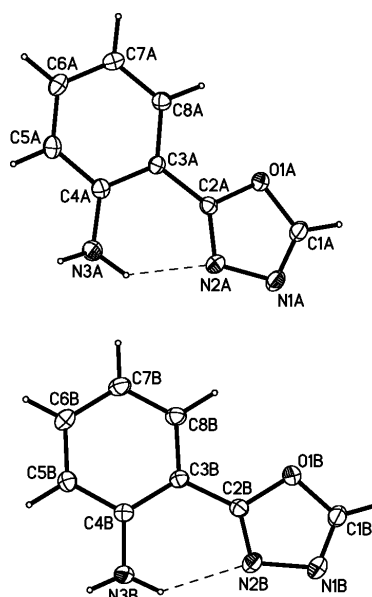


Fig. 1. The molecular structures of the two crystallographically independent molecules (denoted as A and B) present in the crystal of **5**, showing the atom numbering scheme and the intramolecular N–H...N hydrogen contacts forming *S*(6) motifs (dashed lines). Displacement ellipsoids are shown at the 40% probability level.

determination (see Experimental Section).

#### Description of the crystal structure of **5**

The asymmetric unit of the crystal of compound **5** contains two chemically identical molecules of 2-(1,3,4-oxadiazol-2-yl)aniline (denoted as A and B). The molecular structures of A and B are shown in Fig. 1 and a summary of the experimental details is given in Table 1. The molecules are planar with r.m.s. deviations of fitted atoms at 0.051 Å for A and 0.062 Å for B. In both molecules maximum deviation from the least-squares planes is observed for atom O(1) [0.094(1) Å in A and 0.103(1) Å in B], which was also observed for the close analogue of **5**, its 2-carbonyl derivative 5-(2-aminophenyl)-1,3,4-oxadiazol-2(3*H*)-one [24].

The two parts of each molecule are almost exactly planar [with r.m.s. of fitted atoms of 0.001, 0.005, 0.002 and 0.009 Å for oxadiazole and aminophenyl moieties of A and B, respectively, with maximum deviations less than 0.014(1) Å, and with an angle between the least-squares planes through oxadiazole and the aminophenyl moieties of 6.2(1)° for A and 7.2(1)° for B]. The geometry of the two molecules is the same

Table 2. Selected interatomic distances (Å), valence angles (deg) and torsion angles (deg) in **5**.

	Molecule A	Molecule B
<i>Bond lengths:</i>		
O(1)–C(1)	1.357(2)	1.358(2)
O(1)–C(2)	1.371(2)	1.372(2)
N(1)–N(2)	1.416(2)	1.412(2)
N(1)–C(1)	1.280(2)	1.280(2)
N(2)–C(2)	1.302(2)	1.300(2)
N(3)–C(4)	1.376(2)	1.376(2)
C(2)–C(3)	1.450(2)	1.450(2)
C(3)–C(4)	1.416(2)	1.419(2)
<i>Bond angles:</i>		
C(1)–O(1)–C(2)	102.59(8)	102.41(9)
C(1)–N(1)–N(2)	105.44(9)	105.41(9)
C(2)–N(2)–N(1)	106.61(9)	106.81(9)
N(1)–C(1)–O(1)	113.87(10)	113.88(10)
N(2)–C(2)–O(1)	111.49(9)	111.49(10)
O(1)–C(2)–C(3)	118.72(8)	119.36(9)
N(2)–C(2)–C(3)	129.78(9)	129.15(10)
<i>Torsion angles:</i>		
N(2)–C(2)–C(3)–C(8)	173.2(1)	172.4(1)
O(1)–C(2)–C(3)–C(4)	175.3(1)	176.1(1)
C(8)–C(3)–C(4)–N(3)	179.0(1)	177.9(1)
C(2)–C(3)–C(4)–N(3)	–2.2(2)	–4.7(2)
C(2)–C(3)–C(4)–C(5)	179.6(1)	177.5(1)

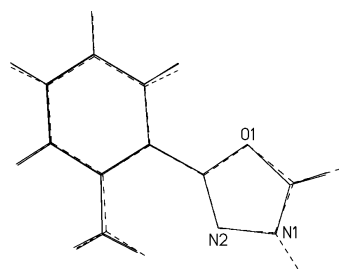


Fig. 2. Comparison of the molecular structures of the two crystallographically independent molecules of 2-(1,3,4-oxadiazol-2-yl)aniline (**5**; solid lines) with its 2-carbonyl derivative: 5-(2-aminophenyl)-1,3,4-oxadiazol-2(3H)-one [24] (atomic coordinates from Ref. [25]; dashed lines). The common reference points are O(1), N(1) and N(2).

within experimental error, which is clearly indicated by the geometrical parameters listed in Table 2 and is easy to see when superimposing the molecular structures of A and B (see Fig. 2). Additionally, which is also shown in Fig. 2, the overall geometry of A and B is very close to that observed in 5-(2-aminophenyl)-1,3,4-oxadiazol-2(3H)-one [24, 25].

The pyramidalization of the amino group is not so expressed as observed in crystalline aniline [26]. Both N(3) atoms lie nearly exactly in the phenyl ring planes (they are coplanar within 0.02 Å in A and 0.03 Å in B), whereas the hydrogen atoms are slightly displaced from these planes [the distances of atoms H(31A),

H(32A), H(31B) and H(32B) from the respective Ph ring planes are in the range of 0.12–0.23 Å]. The dihedral angle between the planes defined by the amino group and the phenyl ring is 22(2)° in A and 28(2)° in B, compared to 37–38° in the crystal structure of the free aniline [26] and about 16° in 5-(2-aminophenyl)-1,3,4-oxadiazol-2(3H)-one [24, 25]. The sum of the angles about N(3) is 355(2)° in A and 351(2)° in B, compared to 360° typical for a planar, trigonal environment with a  $sp^2$ -hybridized atom, about 330° at a tetrahedral  $sp^3$ -hybridized N, about 340° in crystalline aniline [26], or about 356° in 5-(2-aminophenyl)-1,3,4-oxadiazol-2(3H)-one [24, 25]. All that indicates a rather small degree of pyramidalization of the amine N(3) atoms in compound **5**, as well as in its close analogue, the 2-carbonyl derivative. There is also a slight shortening of N(3)–C(4) (1.376(2) Å in both A and B, and 1.373(4) Å in 5-(2-aminophenyl)-1,3,4-oxadiazol-2(3H)-one [24]) as compared to typical  $C_{ar}$ –NH<sub>2</sub>(pyramidal) systems [27].

The overall planar geometry of the molecules is stabilized by the intramolecular N(3)–H(31)···N(2) hydrogen bond forming the usual six-membered *S*(6) motif, also shown in Fig. 1. It is possible that the intramolecular N–H···N hydrogen bond may be assisted by resonance, which is often observed when the hydrogen-bond donor and acceptor are connected by a short chain of conjugated single and double bonds [28] [compare the geometry of the fragment involved in the *S*(6) ring motif: H(31)–N(3)–C(4)–C(3)–C(2)–N(2); Tables 2 and 3]. Similar intramolecular N–H···N contacts are observed in 5-(2-aminophenyl)-1,3,4-oxadiazol-2(3H)-one [24].

In the crystals of **5**, adjacent molecules A and B are linked by a rather extensive network of N–H···N hydrogen bonds and C–H···N/O close interactions to form two independent chains (denoted as chain A and chain B), both parallel to the *b* axis, as shown in Fig. 3. Most of the interactions are formed within the respective chain A or B, however two types of them, *i.e.* C(1B)–H(1B)···N(3A)<sup>iv</sup> and C(5B)–H(5B)···O(1A)<sup>v</sup> [equivalent C(5B)<sup>ii</sup>–H(5B)<sup>ii</sup>···O(1A)<sup>i</sup>] is shown in Fig. 3), act as linkers between the independent chains A and B. The other weak contact, C(1A)–H(1A)···N(1A)<sup>iii</sup>, is formed between two adjacent chains of the same type A. Despite very similar overall structures of the respective chains A and B, the network of hydrogen contacts is quite different, which is easy to see in Fig. 3. A short non-bonding, rather repulsive, inter D–H···H–X con-

Table 3. Geometry of proposed hydrogen bonds and N–H···N, C–H···N/O close contacts, and  $\pi$ ·· $\pi$  stacking interactions for **5** (Å, deg).

D–H···A	D–H (Å)	H···A (Å)	D···A (Å)	D–H···A (deg)	$\pi$ ·· $\pi$ stacking interactions	Cg···Cg (Å)	dihedral angle (deg)
N(3A)–H(31A)···N(2A)	0.90(2)	2.08(2)	2.788(2)	135(2)	Cg(1)···Cg(2) <sup>vii</sup>	3.583(2)	6.03
N(3A)–H(32A)···N(1A) <sup>i</sup>	0.88(2)	2.53(2)	3.400(2)	166(2)	Cg(2)···Cg(1) <sup>vii</sup>	3.583(2)	6.03
N(3B)–H(31B)···N(2B)	0.90(2)	2.09(2)	2.782(2)	133(1)	Cg(2)···Cg(4) <sup>i</sup>	3.710(2)	7.27
N(3B)–H(32B)···N(1B) <sup>ii</sup>	0.90(2)	2.35(2)	3.248(2)	174(2)	Cg(3)···Cg(4) <sup>viii</sup>	3.582(2)	6.83
C(1A)–H(1A)···N(1A) <sup>iii</sup>	0.97(2)	2.78(2)	3.503(2)	132(1)	Cg(4)···Cg(2) <sup>ix</sup>	3.710(2)	7.27
C(5A)–H(5A)···N(2A) <sup>j</sup>	0.99(2)	2.79(2)	3.687(2)	150(1)	Cg(4)···Cg(3) <sup>viii</sup>	3.582(2)	6.83
C(5A)–H(5A)···N(3A) <sup>j</sup>	0.99(2)	2.78(2)	3.667(2)	149(1)			
C(1B)–H(1B)···N(3A) <sup>iv</sup>	0.94(2)	2.68(2)	3.344(2)	128(1)			
C(5B)–H(5B)···O(1A) <sup>v</sup>	0.98(2)	2.73(2)	3.251(2)	114(1)			
C(5B)–H(5B)···N(2B) <sup>ii</sup>	0.98(2)	2.65(2)	3.463(2)	140(1)			
C(6B)–H(6B)···N(1B) <sup>vi</sup>	0.96(2)	2.76(2)	3.624(2)	150(1)			

Symmetry codes: <sup>i</sup>  $-x+1/2, y-1/2, -z+3/2$ ; <sup>ii</sup>  $-x+3/2, y-1/2, -z+3/2$ ; <sup>iii</sup>  $-x, -y+1, -z+1$ ; <sup>iv</sup>  $x, y+1, z$ ; <sup>v</sup>  $x+1, y, z$ ; <sup>vi</sup>  $x, y-1, z$ ; <sup>vii</sup>  $-x, -y, -z+1$ ; <sup>viii</sup>  $-x+1, -y+1, -z+1$ ; <sup>ix</sup>  $-x+1/2, y+1/2, -z+3/2$ . Cg(1), Cg(2), Cg(3) and Cg(4) are the centroids of the 1,3,4-oxadiazolyl and aniline phenyl rings from molecules A and B, respectively, *i. e.* O(1A)/C(1A)/N(1A)/N(2A)/C(2A), C(3A)–C(8A) and O(1B)/C(1B)/N(1B)/N(2B)/C(2B), C(3B)–C(8B).

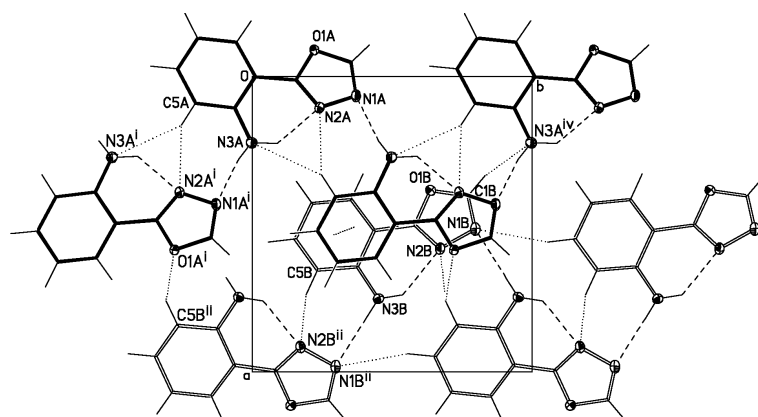


Fig. 3. The arrangement of the molecules A (solid lines) and B (open lines) within the chains running parallel to the *b* axis; viewed down the *c* axis. Intra- and intermolecular N–H···N hydrogen bonds are shown with dashed lines, and intermolecular C–H···N/O close contacts and weak  $\pi$ ·· $\pi$  stacking interactions with dotted lines. Symmetry codes are given in Table 3.

tact is observed in chain A. It is formed between atoms H(5A) and H(31A)<sup>i</sup> [with a H(5A)···H(31A)<sup>i</sup> distance of 2.04(2) Å] and accompanies the weak, bifurcated C(5A)–H(5A)···N(2A)<sup>j</sup>/N(3A)<sup>j</sup> interaction, which in combination with intramolecular N(3A)–H(31A)···N(2A) hydrogen bonds, gives rise to  $R_2^2$  (4) rings formed between two adjacent molecules A in chain A.

Beside different intra- and interchain N–H···N and C–H···N/O interactions, the molecules of adjacent chains A and B are stacked through  $\pi$ ·· $\pi$  interactions along the *c* axis, with a ring-centroid separation of 3.582(2)–3.710(2) Å and the angles between the planes of the stacking rings in the range of 6.03–7.27° (Table 3, Fig. 3).

## Conclusions

We believe the reported method offers a mild, simple and efficient route for the preparation of 2-(1,3,4-

oxadiazol-2-yl)aniline (**5**). The ease of work up, high yield and fairly mild reaction conditions make it a useful addition to modern synthetic methodologies. Other aspects of this process are under investigation. The X-ray structural analysis of the product indicated that the two aromatic rings of the molecule are approximately co-planar.

## Experimental Section

### General procedures

Freshly distilled solvents were used throughout, and anhydrous solvents were dried according to Perrin and Armarego [29]. Elemental analyses were performed using a Heraeus CHN-O-Rapid analyzer. <sup>1</sup>H (250 MHz) and <sup>13</sup>C (62.5 MHz) NMR measurements were recorded on a Bruker 250 spectrometer in CDCl<sub>3</sub> with tetramethylsilane as internal standard. IR spectra were measured on a Shimadzu IR-460 spectrometer. Mass spectra were

recorded on a FINNIGAN-MAT 8430 mass spectrometer operating at an ionization potential of 20 eV. Melting points were measured on an Electrothermal 9100 apparatus and are uncorrected. (*N*-Isocyanimino)triphenylphosphorane (**2**) was prepared based on a reported procedure [17]. Other starting materials and solvents were obtained from Merck (Germany) and Fluka (Switzerland) and were used without further purification. Flash chromatography columns were prepared from Merck silica gel powder.

#### 2-(1,3,4-Oxadiazol-2-yl)aniline (**5**)

To a magnetically stirred solution of (*N*-isocyanimino)triphenylphosphorane **2** (0.302 g, 1 mmol) in dry CH<sub>2</sub>Cl<sub>2</sub> (8 mL) was added dropwise a mixture of 2-amino benzoic acid **1** (0.137 g, 1 mmol) in dry CH<sub>2</sub>Cl<sub>2</sub> (7 mL) over 15 min. The mixture was stirred for 6 h at r. t. The solvent was removed under reduced pressure and the viscous residue was purified by flash column chromatography [silica gel; petroleum ether-ethyl acetate (10:2)]. The solvent was removed under reduced pressure and the product **4** was obtained as light yellow crystals. Yield: 140 mg (86%); m. p. 145.3 °C. – IR (KBr):  $\nu = 3431, 3339, 3039, 2939, 2931, 2931, 1623, 1269, 1108, 754 \text{ cm}^{-1}$ . – <sup>1</sup>H NMR (CDCl<sub>3</sub>):  $\delta = 5.69$  (br. s, 2 H, NH<sub>2</sub>), 6.73 (s, 1 H, arom.), 6.79 (t, 1 H, <sup>3</sup>J = 7.5 Hz, arom.), 7.75 (t, 1 H, <sup>3</sup>J = 7.8 Hz, arom.), 7.76 (d, 1 H, <sup>3</sup>J = 6.0 Hz, arom.), 8.41 (s, 1 H, oxadiazole). – <sup>13</sup>C NMR (CDCl<sub>3</sub>):  $\delta = 116.28, 116.91, 127.99$  and  $132.79$  (4 CH, arom.);  $105.38$  and  $147.01$  (2 C, arom.);  $150.91$  (1 CH, oxadiazole);  $164.62$  (1 C, oxadiazole). – MS:  $m/z$  (%) = 161 (100) [M]<sup>+</sup>, 120 (50), 105 (22), 92 (25), 90 (15), 78 (15), 77 (34), 65 (28), 63 (21), 58 (22), 55 (11), 52 (20), 51 (31), 50 (18), 43 (28), 41 (21). – C<sub>8</sub>H<sub>7</sub>N<sub>3</sub>O (161.06): calcd. C 59.62, H 4.38, N 26.07; found C 59.57, H 4.34, N 26.14.

#### Preparation of single crystals of 2-(1,3,4-oxadiazol-2-yl)aniline (**5**)

Single crystals of **5** were prepared by using the branch tube method with *n*-hexane at 40 °C during two weeks [30]. The light yellow crystals were filtered off, washed with cold *n*-hexane and dried at r. t. (m. p. 145.5 °C).

#### Crystal structure determination of **5**

The crystallographic measurement was performed on a  $\kappa$ -geometry Kuma KM4CCD automated four-circle diffractometer with the graphite-monochromatized MoK $\alpha$  radiation. The data were collected at 100(2) K using the Oxford Cryosystems cooler. A summary of the conditions for the data collection and the structure refinement parameters is given in Table 1. The data were corrected for Lorentz and polarization effects. Data collection, cell refinement, and data reduction and analysis were carried out with the KM4CCD software (Oxford Diffraction, Poland): CrysAlis CCD and CrysAlis RED, respectively [31]. The structure was solved by Direct Methods using SHELXS-97 [32] and refined by full-matrix least-squares techniques with anisotropic displacement parameters for the non-H atoms using SHELXL-97 [33]. All H atoms were found in difference Fourier maps and were refined isotropically. All figures were made using the program XP [34].

#### Supplementary material

CCDC 635760 contains the supplementary crystallographic data for this paper. These data can be obtained free of charge from The Cambridge Crystallographic Data Centre via [www.ccdc.cam.ac.uk/data\\_request/cif](http://www.ccdc.cam.ac.uk/data_request/cif).

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- [1] W. R. Tully, C. R. Gardner, R. J. Gillespie, R. Westwood, *J. Med. Chem.* **1991**, *34*, 2060–2067.
- [2] C. Chen, C. H. Senanayake, T. J. Bill, R. D. Larsen, T. R. Verhoeven, P. T. Reider, *J. Org. Chem.* **1994**, *59*, 3738–3741.
- [3] B. S. Holla, R. Gonsalves, S. Shenoy, *Eur. J. Med. Chem.* **2000**, *35*, 267–271.
- [4] M. J. Crimmin, P. J. Hanlon, N. H. Rogers, G. Walker, *J. Chem. Soc., Perkin Trans. 1* **1989**, 2047–2056.
- [5] U. V. Laddi, S. R. Desai, R. S. Bennur, S. C. Bennur, *Ind. J. Heterocycl. Chem.* **2002**, *11*, 319–322.
- [6] I. R. Baxendale, S. V. Ley, M. Martinelli, *Tetrahedron* **2005**, *61*, 5323–5349.
- [7] S. Liras, M. P. Allen, B. E. Segelstein, *Synth. Commun.* **2000**, *30*, 437–443.
- [8] B. J. Brown, I. R. Clemens, J. K. Neesom, *Synlett* **2000**, *1*, 131–133.
- [9] F. T. Coppo, K. A. Evans, T. L. Graybill, G. Burton, *Tetrahedron Lett.* **2004**, *45*, 3257–3260.
- [10] C. T. Brain, J. M. Paul, Y. Loong, P. J. Oakley, *Tetrahedron Lett.* **1999**, *40*, 3275–3278.
- [11] C. T. Brain, S. A. Brunton, *Synlett* **2001**, *3*, 382–384.
- [12] V. K. Tandon, R. B. Chhor, *Synth. Commun.* **2001**, *31*, 1727–1732.
- [13] S. H. Mashraqui, S. G. Ghadigaonkar, R. S. Kenny, *Synth. Commun.* **2003**, *33*, 2541–2545.

- [14] F. Bentiss, M. Lagrenee, D. Barbry, *Synth. Commun.* **2001**, *31*, 935–938.
- [15] E. Jedlovska, J. Lesko, *Synth. Commun.* **1994**, *24*, 1879–1885.
- [16] Y. Wang, D. R. Sauer, S. W. Djuric, *Tetrahedron Lett.* **2006**, *47*, 105–108.
- [17] H. Stolzenberg, B. Weinberger, W. P. Fehlhammer, F. G. Pühlhofer, R. Weiss, *Eur. J. Inorg. Chem.* **2005**, *21*, 4263–4271.
- [18] T. W. Chiu, Y. H. Liu, K. M. Chi, Y. S. Wen, K. L. Lu, *Inorg. Chem.* **2005**, *44*, 6425–6430.
- [19] I. Yavari, A. Ramazani, A. Yahya-Zadeh, *Synth. Commun.* **1996**, *26*, 4495–4499.
- [20] A. Ramazani, E. Ahmadi, A. R. Kazemizadeh, L. Dolatyari, N. Noshiranzadeh, I. Eskandari, A. Souldozi, *Phosphorus, Sulphur, and Silicon* **2005**, *180*, 2419–2422.
- [21] A. Ramazani, E. Ahmadi, *Phosphorus, Sulfur, and Silicon* **2006**, *181*, 2725–2729.
- [22] A. Domling, *Chem. Rev.* **2006**, *106*, 17–89.
- [23] P. Molina, M. J. Vilaplana, *Synthesis* **1994**, 1197–1218.
- [24] J. S. Davidson, J. Trotter, *Acta Crystallogr.* **1996**, *C52*, 3236–3238.
- [25] Cambridge Structural Database (CSD); F. H. Allen, *Acta Crystallogr.* **2002**, *B58*, 380–388.
- [26] M. Fukuyo, K. Hirotsu, T. Higuchi, *Acta Crystallogr.* **1982**, *B38*, 640–643.
- [27] F. H. Allen, O. Kennard, D. G. Watson, L. Brammer, A. G. Orpen, R. Taylor in *International Tables for Crystallography*, Vol. C (Ed.: A. J. C. Wilson) Kluwer Academic Publishers, Dordrecht, **1992**, chapter 9.5.
- [28] V. Bertolasi, L. Pretto, G. Gilli, P. Gilli, *Acta Crystallogr.* **2006**, *B62*, 850–863 and refs. cited herein.
- [29] D. D. Perrin, W. L. F. Armarego, *Purification of Laboratory Chemicals*, Pergamon Press, Oxford, **1988**, pp. 20–35.
- [30] A. Ramazani, A. Souldozi, A. Morsali, A. R. Jalilian, *Z. Kristallogr. NCS* **2004**, *219*, 247–248.
- [31] Kuma KM4CCD software: CrysAlis CCD and CrysAlis RED (ver. 1.171) Oxford Diffraction (Poland) **1995–2003**.
- [32] G. M. Sheldrick, SHELXS-97. Program for the solution of crystal structures. University of Göttingen, Göttingen (Germany) **1997**.
- [33] G. M. Sheldrick, SHELXL-97. Program for the refinement of crystal structures. University of Göttingen, Göttingen (Germany) **1997**.
- [34] XP – Interactive molecular graphics (ver. 5.1), Bruker Analytical X-ray Systems, Karlsruhe (Germany) **1998**.