

$$w = 1/[\sigma^2(F_o^2) + (0.0228P)^2 + 0.4177P]$$

where  $P = (F_o^2 + 2F_c^2)/3$

Scattering factors from  
*International Tables for  
Crystallography* (Vol. C)

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## 1,2,3,4-Tetrahydrobenzimidazo[2,1-*b*]-quinazoline

GALAL E. H. ELGEMEIE,<sup>a</sup> NAHED M. FATHY,<sup>b</sup> HENNING HOPF<sup>c</sup> AND PETER G. JONES<sup>d\*</sup>

<sup>a</sup>Chemistry Department, Faculty of Science, Helwan University, Helwan, Cairo, Egypt, <sup>b</sup>Photochemistry Laboratory, National Research Centre, Cairo, Egypt, <sup>c</sup>Institut für Organische Chemie, Technische Universität Braunschweig, Postfach 3329, 38023 Braunschweig, Germany, and <sup>d</sup>Institut für Anorganische und Analytische Chemie, Technische Universität Braunschweig, Postfach 3329, 38023 Braunschweig, Germany. E-mail: jones@xray36.anchem.nat.tu-bs.de

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### Abstract

The title compound, C<sub>14</sub>H<sub>13</sub>N<sub>3</sub>, was identified as one of four possible isomers from a condensation reaction. Three of the four rings are coplanar within 0.026 Å. The molecules stack in pairs across inversion centres, with a shortest C···C distance of 3.373 (3) Å.

### Comment

Chemically synthesized purine analogues find numerous applications in clinical medicine and medical research. The pharmacological approach involves analogues in which the heterocyclic ring system has been modified so as to induce toxic effects when the analogue is incorporated into specific cell constituents. As part of our program directed towards the synthesis of purines and other antimetabolites (Elgemeie *et al.*, 1996, 1997, 1998), we have recently reported various successful approaches to syntheses of purine, pyrimidine and folic acid analogues. Derivatives of these ring systems are of interest as antimetabolites in biochemical reactions (Elgemeie & Fathy, 1995; Elgemeie & Hussain, 1994; Elgemeie *et al.*, 1994).

We report here a novel one-pot synthesis of a purine analogue by reaction of the sodium salt of 2-(hydroxymethylene)-1-cyclohexanone, (3), with 2-aminobenzimidazole, (1) (see scheme below). The reaction between (1) and (3) in the presence of piperidine acetate gives an adduct for which four isomeric structures, (4)–(7), are possible. Initial nucleophilic attack by the amino group at the carbonyl carbon could be followed by cyclization and elimination of water to give compound (5) or (7); alternatively, attack of the amino group at the formyl group followed by cyclization leads to compound (4) or (6). Spectroscopic methods did not identify the product unambiguously and therefore an X-ray structure determination was carried out.

Table 1. Selected geometric parameters (Å, °)

|              |           |               |           |
|--------------|-----------|---------------|-----------|
| N1—C12       | 1.389 (3) | N2—C16        | 1.316 (3) |
| N1—C16       | 1.410 (3) | N2—C15        | 1.379 (3) |
| N1—C14       | 1.410 (3) | C14—C15       | 1.407 (4) |
| C12—N1—C16   | 121.8 (2) | N2—C15—C8     | 127.5 (2) |
| C12—N1—C14   | 133.5 (2) | N2—C15—C14    | 112.1 (2) |
| C16—N1—C14   | 104.7 (2) | C8—C15—C14    | 120.4 (2) |
| C16—N2—C15   | 104.3 (2) | N2—C16—N1     | 114.1 (2) |
| C11—C14—C15  | 121.0 (2) | N2—C16—C7     | 128.5 (2) |
| C11—C14—N1   | 134.1 (3) | N1—C16—C7     | 117.4 (2) |
| C15—C14—N1   | 104.9 (2) |               |           |
| C12—C1—C2—C3 | −82.9 (3) | C3—C4—C5—C13  | 78.7 (3)  |
| C1—C2—C3—C4  | 60.7 (4)  | C2—C1—C12—C13 | 71.5 (3)  |
| C2—C3—C4—C5  | −60.0 (4) | C4—C5—C13—C12 | −62.6 (4) |

H atoms were placed geometrically and thereafter allowed to ride on their parent atoms.

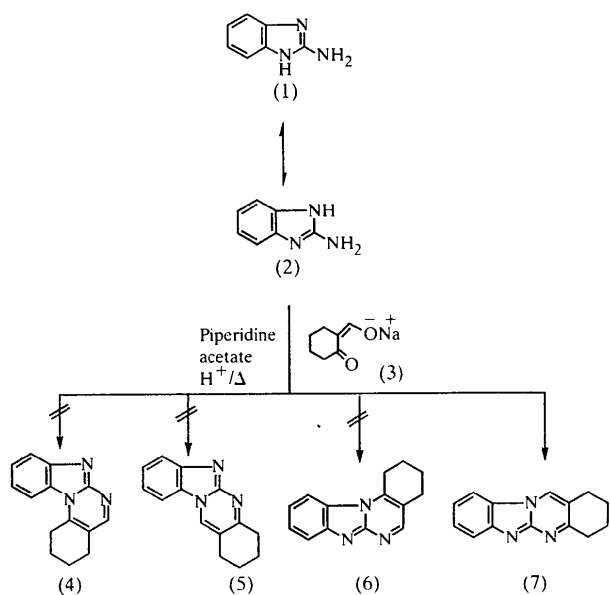
Data collection: *DIF4* (Stoe & Cie, 1992a). Cell refinement: *DIF4*. Data reduction: *REDU4* (Stoe & Cie, 1992b). Program(s) used to solve structure: *SHELXS86* (Sheldrick, 1990). Program(s) used to refine structure: *SHELXL93* (Sheldrick, 1993). Molecular graphics: *XP* (Siemens, 1994). Software used to prepare material for publication: *SHELXL93*.

We thank the Deutsche Forschungsgemeinschaft for supporting our collaboration, the Fonds der Chemischen Industrie for financial support and Mr A. Weinkauff for technical assistance.

Supplementary data for this paper are available from the IUCr electronic archives (Reference: BM1233). Services for accessing these data are described at the back of the journal.

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The structure was confirmed as (7) by X-ray methods (Fig. 1), with displacement parameters, bond lengths and located H atoms providing unambiguous evidence. The tricyclic ring system incorporating atoms N1–N3 and C5–C14 is planar (mean deviation 0.026 Å); atoms C1–C4 of the saturated ring deviate from this plane by 0.032 (3), 0.697 (4), 0.062 (3) and 0.166 (3) Å, respectively, all on the same side.

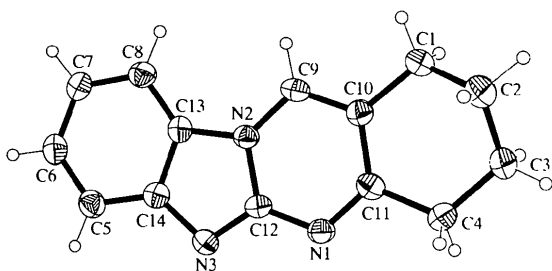


Fig. 1. The molecule of the title compound in the crystal. Ellipsoids represent 50% probability levels and H-atom radii are arbitrary.

Bond lengths and angles are as expected; the major angle deviations from ideal values are associated with the five-membered ring [e.g. C14–N3–C12 104.6 (2) and C8–C13–N2 132.1 (2)<sup>o</sup>]. A search of the Cambridge Structural Database (Allen & Kennard, 1993) revealed no other examples of the same tetraannular system; the corresponding compound with an aromatic rather than a saturated ring, *i.e.* 3,4-dihydro-2H-benzimidazolo[2,3-*b*]quinazoline, was investigated by Molina *et al.* (1994) but was disordered.

The molecules pack in stacked pairs related by inversion centres; the shortest C...C contact is one of 3.373 (3) Å involving C12...C13(1-x, 1-y, 1-z).

## Experimental

A solution of 2-aminobenzimidazole [(1); 1.33 g, 0.01 mol], the sodium salt of 2-(hydroxymethylene)-1-cyclohexanone [(3); 1.47 g, 0.01 mol], and piperidine acetate (1 ml) in water (50 ml) and ethanol (50 ml) was refluxed for 15 min. Acetic acid (1.5 ml) was added to the hot solution. The precipitated solid was collected by filtration and crystallized from benzene in 65% yield (m.p. 504 K).

## Crystal data

C<sub>14</sub>H<sub>13</sub>N<sub>3</sub>  
*M<sub>r</sub>* = 223.27  
 Monoclinic  
*P*2<sub>1</sub>/*n*  
*a* = 6.4542 (14) Å  
*b* = 12.388 (3) Å  
*c* = 13.501 (3) Å  
 $\beta$  = 93.20 (2)<sup>o</sup>  
*V* = 1077.8 (4) Å<sup>3</sup>  
*Z* = 4  
*D<sub>v</sub>* = 1.376 Mg m<sup>-3</sup>  
*D<sub>m</sub>* not measured

Mo K $\alpha$  radiation

$\lambda$  = 0.71073 Å

Cell parameters from 52 reflections

$\theta$  = 10.0–11.5<sup>o</sup>

$\mu$  = 0.085 mm<sup>-1</sup>

*T* = 143 (2) K

Needle

0.6 × 0.2 × 0.2 mm

Yellow

## Data collection

Stoe Stadi-4 diffractometer  
 $\omega/\theta$  scans  
 Absorption correction: none  
 3666 measured reflections  
 1913 independent reflections  
 1320 reflections with  
 $I > 2\sigma(I)$   
*R<sub>int</sub>* = 0.032

$\theta_{\max}$  = 25.04<sup>o</sup>

*h* = 0 → 7

*k* = -14 → 9

*l* = -16 → 16

3 standard reflections

frequency: 60 min

intensity decay: none

## Refinement

Refinement on *F*<sup>2</sup>  
*R* [*F*<sup>2</sup> > 2 $\sigma$ (*F*<sup>2</sup>)] = 0.051  
 $wR$ (*F*<sup>2</sup>) = 0.126  
*S* = 1.047  
 1913 reflections  
 154 parameters  
 H atoms: see below  
 $w = 1/[\sigma^2(F_o^2) + (0.0462P)^2 + 0.4705P]$   
 where  $P = (F_o^2 + 2F_c^2)/3$

( $\Delta/\sigma$ )<sub>max</sub> < 0.001

$\Delta\rho_{\max}$  = 0.36 e Å<sup>-3</sup>

$\Delta\rho_{\min}$  = -0.21 e Å<sup>-3</sup>

Extinction correction: none

Scattering factors from

*International Tables for Crystallography* (Vol. C)

Table 1. Selected geometric parameters (Å, <sup>o</sup>)

|              |             |              |           |
|--------------|-------------|--------------|-----------|
| N1—C11       | 1.322 (3)   | N3—C12       | 1.318 (3) |
| N1—C12       | 1.352 (3)   | N3—C14       | 1.390 (3) |
| N2—C9        | 1.366 (3)   | C9—C10       | 1.357 (3) |
| N2—C13       | 1.393 (3)   | C10—C11      | 1.432 (3) |
| N2—C12       | 1.397 (3)   | C13—C14      | 1.415 (3) |
| C11—N1—C12   | 117.8 (2)   | N1—C12—N2    | 120.7 (2) |
| C9—N2—C13    | 131.7 (2)   | C8—C13—N2    | 132.1 (2) |
| C9—N2—C12    | 121.5 (2)   | C8—C13—C14   | 123.8 (2) |
| C13—N2—C12   | 106.79 (18) | N2—C13—C14   | 104.1 (2) |
| C12—N3—C14   | 104.55 (19) | N3—C14—C5    | 130.3 (2) |
| N3—C12—N1    | 126.3 (2)   | N3—C14—C13   | 111.5 (2) |
| N3—C12—N2    | 113.0 (2)   | C5—C14—C13   | 118.2 (2) |
| C10—C1—C2—C3 | 54.1 (3)    | C2—C3—C4—C11 | 36.7 (3)  |
| C1—C2—C3—C4  | -59.8 (3)   |              |           |

H atoms were placed geometrically and allowed to ride on their parent atoms. Residual electron density of  $ca\ 0.3\ e\ \text{\AA}^{-3}$  near the C2 atom may indicate a very small degree of disorder.

Data collection: *DIF4* (Stoe & Cie, 1992a). Cell refinement: *DIF4*. Data reduction: *REDU4* (Stoe & Cie, 1992b). Program(s) used to solve structure: *SHELXS86* (Sheldrick, 1990). Program(s) used to refine structure: *SHELXL93* (Sheldrick, 1993). Molecular graphics: *XP* (Siemens, 1994). 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: BM1232). Services for accessing these data are described at the back of the journal.

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## A Triclinic Polymorph of Hexaphenylcyclotrisiloxane

BURKHARD ZIEMER,<sup>a</sup> MICHAEL BACKER<sup>a</sup> AND NORBERT AUNER<sup>b</sup>

<sup>a</sup>Institut für Chemie, Humboldt-Universität zu Berlin, Hessische Straße 1-2, D-10115 Berlin, Germany, and

<sup>b</sup>Institut für Anorganische Chemie, Johann Wolfgang Goethe Universität Frankfurt, Marie-Curie-Straße 11, D-60439 Frankfurt am Main, Germany. E-mail: burkhard=ziemer@chemie.hu-berlin.de

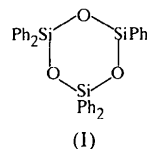
(Received 21 January 1998; accepted 23 February 1998)

## Abstract

In triclinic crystals of 2,2,4,4,6,6-hexaphenylcyclotrisiloxane,  $C_{36}H_{30}O_3Si_3$ , the endocyclic C—C—C bond angles in the phenyl groups at the C atoms bound to silicon are reduced to about  $117^\circ$ . The other geometric parameters do not show any significant differences from expected values.

## Comment

Crystals of the title compound, (I), a large scale precursor in silicone chemistry, were first prepared and investigated by Hyde *et al.* (1947). Two kinds of crystals were optically separated and were characterized as orthorhombic and triclinic polymorphs by X-ray powder diffraction. The structure of the orthorhombic polymorph was later determined by Bokii *et al.* (1972) and redetermined with higher precision by Tomlins *et al.* (1985). The redetermination revealed small but significant distortions of the phenyl rings from their ideal geometry. With the intention of generating diphenylsilanediol by reaction of dichlorodiphenylsilane with water in toluene in the presence of *tert*-amyl alcohol (Burkhard, 1945), hexaphenylcyclotrisiloxane, (I), was formed after the solution had been stored for more than two weeks at room temperature. The solid was recrystallized from warm 2-butanone and chloroform as large colorless blocks.



The present structure analysis of the triclinic polymorph also shows some significant distortions of the phenyl rings, most significantly, the decrease of the C—C—C bond angles to  $117.0(4)^\circ$  at those phenyl C atoms bound at silicon. A similar distortion observed in the structure of 2,2-diphenyl-2-sila-1,3,4-trihydro-