

$wR = 0.039$
 1060 reflections
 149 parameters
 Only coordinates of H atoms refined
 Unit weights applied
 $(\Delta/\sigma)_{\max} = 0.001$

Extinction correction: Gaussian (Zachariasen, 1968)
 Extinction coefficient: 75 (7)
 Atomic scattering factors from *International Tables for X-ray Crystallography* (1974, Vol. IV, Table 2.3.1)

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 Morino, Y., Kuchitsu, K. & Yokoseki, A. (1967). *Bull. Chem. Soc. Jpn.*, **40**, 1552.
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 Watkin, D. J., Carruthers, J. R. & Betteridge, P. W. (1985). *CRYSTALS. An Advanced Crystallographic Computer Program*. Chemical Crystallographic Laboratory, Univ. of Oxford, England.
 Zachariasen, W. H. (1968). *Acta Cryst.* **A24**, 212-216.

Table 1. Fractional atomic coordinates and equivalent isotropic displacement parameters (\AA^2)

$$U_{\text{eq}} = (1/3)\sum_i \sum_j U_{ij} a_i^* a_j^* \mathbf{a}_i \cdot \mathbf{a}_j$$

	x	y	z	U_{eq}
C(1)	0.7854 (4)	0.0713 (2)	0.0988 (2)	0.0347
C(2)	0.6530 (4)	0.1314 (2)	0.1716 (2)	0.0343
C(3)	0.4618 (5)	0.2134 (3)	0.1247 (2)	0.0431
C(4)	0.5457 (6)	0.3331 (3)	0.1530 (3)	0.0541
C(5)	0.5708 (5)	0.3439 (3)	0.2560 (3)	0.0514
C(6)	0.5038 (5)	0.2322 (2)	0.3019 (2)	0.0423
C(7)	0.6795 (4)	0.1423 (2)	0.2792 (2)	0.0347
C(8)	0.8442 (5)	0.0882 (2)	0.3648 (2)	0.0375
C(9)	0.3082 (5)	0.1987 (3)	0.2118 (3)	0.0510
N(11)	0.8857 (4)	-0.0262 (2)	0.1312 (2)	0.0386
O(11)	0.7947 (3)	0.1135 (2)	0.0089 (1)	0.0440
N(81)	0.8950 (5)	0.1472 (2)	0.4558 (2)	0.0470
O(81)	0.9266 (3)	-0.0070 (2)	0.3534 (1)	0.0425

Table 2. Selected geometric parameters (\AA , $^\circ$)

C(1)—C(2)	1.479 (3)	C(6)—C(7)	1.545 (4)
C(1)—N(11)	1.323 (3)	C(6)—C(9)	1.544 (4)
C(1)—O(11)	1.241 (3)	C(7)—C(8)	1.481 (3)
C(2)—C(3)	1.538 (4)	C(8)—N(81)	1.329 (3)
C(2)—C(7)	1.341 (3)	C(8)—O(81)	1.236 (3)
C(3)—C(4)	1.511 (4)	N(11)···O(81)	2.772(3)
C(4)—C(5)	1.285 (4)	O(81)···H(111)	1.89 (4)
C(5)—C(6)	1.509 (4)		
N(11)—C(1)—C(2)	118.7 (2)	C(7)—C(6)—C(5)	106.8 (2)
O(11)—C(1)—C(2)	118.6 (2)	C(9)—C(6)—C(5)	99.0 (2)
O(11)—C(1)—N(11)	122.7 (2)	C(9)—C(6)—C(7)	98.7 (2)
C(3)—C(2)—C(1)	119.9 (2)	C(6)—C(7)—C(2)	106.0 (2)
C(7)—C(2)—C(1)	132.8 (2)	C(8)—C(7)—C(2)	130.5 (2)
C(7)—C(2)—C(3)	106.7 (2)	C(8)—C(7)—C(6)	123.4 (2)
C(4)—C(3)—C(2)	106.8 (2)	N(81)—C(8)—C(7)	116.0 (2)
C(9)—C(3)—C(2)	98.6 (2)	O(81)—C(8)—C(7)	122.3 (2)
C(9)—C(3)—C(4)	98.6 (2)	O(81)—C(8)—N(81)	121.7 (2)
C(5)—C(4)—C(3)	108.0 (3)	C(6)—C(9)—C(3)	91.3 (2)
C(6)—C(5)—C(4)	107.6 (3)	N(11)—H(111)···O(81)	151 (3)

Data collection: *CAD-4 Software* (Enraf-Nonius, 1989). Cell refinement: *CAD-4 Software CELDIM*. Data reduction: *CRYSTALS RC85* (Watkin, Carruthers & Betteridge, 1985). Program(s) used to solve structure: *CRYSTALS*; *SHELXS86* (Sheldrick, 1985). Program(s) used to refine structure: *CRYSTALS*. Molecular graphics: *ORTEPII* (Johnson, 1976). Software used to prepare material for publication: *CRYSTALS*.

Lists of structure factors, anisotropic displacement parameters, H-atom coordinates and complete geometry have been deposited with the British Library Document Supply Centre as Supplementary Publication No. SUP 71686 (12 pp.). Copies may be obtained through The Technical Editor, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England. [CIF reference: PA1037]

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Acta Cryst. (1994). **C50**, 415-417

Dimethyl 9-Phenyl-1,4-dihydro-1,4-ethenoanthracene-11,12-dicarboxylate

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Abstract

The molecule $\text{C}_{26}\text{H}_{20}\text{O}_4$ has normal geometry and dimensions. The methoxycarbonyl group that is remote from the phenyl substituent is conjugated with the $\text{C}11=\text{C}12$ double bond [$\text{C}=\text{C}-\text{C}=\text{O}$ torsion angle = $-163.2(4)^\circ$, $\cos^2(\text{angle}) = 0.92$], while the adjacent group is only very slightly conjugated [$\text{C}=\text{C}-\text{C}=\text{O}$ torsion angle = $68.1(5)^\circ$, $\cos^2(\text{angle}) = 0.14$].

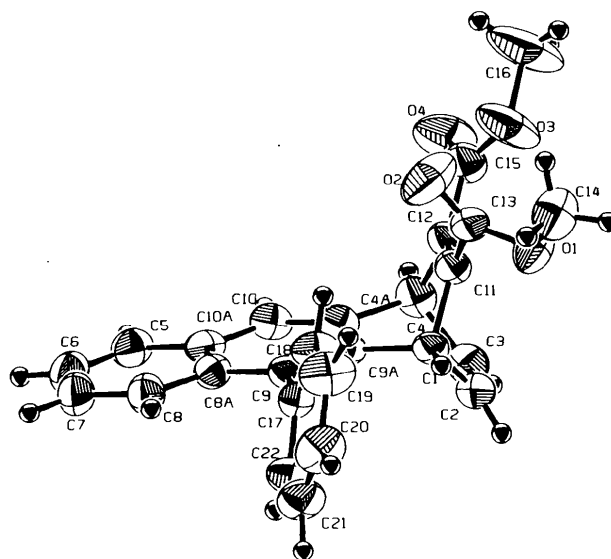
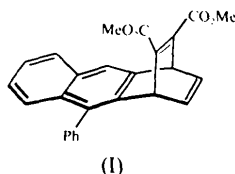


Fig. 1. View of the molecule with 50% probability ellipsoids.

Comment

The title compound (I) is a by-product in the synthesis of the 9,10-etheno isomer (Pokkuluri, Scheffer & Trotter, 1993).

**Experimental**

The compound was prepared according to the procedure described by Pokkuluri, Scheffer & Trotter (1993).

*Crystal data*C₂₆H₂₀O₄*M_r* = 396.44

Orthorhombic

P2₁2₁2₁*a* = 8.100 (2) Å*b* = 31.729 (5) Å*c* = 7.848 (3) Å*V* = 2017 (1) Å³*Z* = 4*D_x* = 1.305 Mg m⁻³

Cu Kα radiation

λ = 1.5418 Å

Cell parameters from 19 reflections

θ = 13–24°

μ = 0.67 mm⁻¹*T* = 294 K

Plate

0.4 × 0.4 × 0.1 mm

Colourless

Data collection

Rigaku AFC-6 diffractometer

ω/2θ scans

Absorption correction:
not applied*(T_{min}* = 0.90, *T_{max}* = 1.00)

2347 measured reflections

2347 independent reflections

1862 observed reflections

[*I* > 3σ(*I*)]*Refinement*Refinement on *F*²*R* = 0.040*wR* = 0.056*S* = 1.81

1862 reflections

272 + 56 H parameters

w = 1/σ²(*F*)(Δ/σ)_{max} = 0.005Δρ_{max} = 0.26 e Å⁻³Δρ_{min} = -0.18 e Å⁻³θ_{max} = 77.6°*h* = 0 → 10*k* = 0 → 40*l* = 0 → 93 standard reflections
monitored every 150 reflections

intensity variation: 0.1%

Extinction correction:

TEXSAN (Molecular
Structure Corporation,
1990)

Extinction coefficient:

3.4 (6) × 10⁻⁶Atomic scattering factors
from *International Tables*
for *X-ray Crystallography*
(1974, Vol. IV)

Table 1. Fractional atomic coordinates and equivalent isotropic displacement parameters (Å²)

$$U_{eq} = (1/3)\sum_i\sum_j U_{ij}a_i^*a_j^*a_i \cdot a_j$$

	<i>x</i>	<i>y</i>	<i>z</i>	<i>U_{eq}</i>
O1	0.2730 (3)	0.23235 (6)	0.5008 (3)	0.064
O2	0.0694 (4)	0.20758 (9)	0.3433 (5)	0.096
O3	0.3514 (6)	0.20769 (7)	0.0843 (3)	0.104

O4	0.4129 (5)	0.14816 (9)	-0.0433 (3)	0.089
C1	0.3234 (4)	0.13803 (8)	0.5514 (4)	0.040
C2	0.5050 (4)	0.12643 (9)	0.5698 (4)	0.048
C3	0.5695 (4)	0.10927 (10)	0.4330 (5)	0.054
C4	0.4486 (4)	0.10469 (9)	0.2851 (4)	0.050
C4A	0.3060 (4)	0.07819 (8)	0.3570 (4)	0.044
C5	0.0524 (5)	-0.01856 (9)	0.3222 (5)	0.055
C6	-0.0784 (5)	-0.03752 (10)	0.4021 (5)	0.061
C7	-0.1565 (4)	-0.01760 (10)	0.5369 (5)	0.061
C8	-0.0993 (4)	0.02055 (9)	0.5961 (5)	0.053
C8A	0.0383 (4)	0.04033 (8)	0.5213 (4)	0.043
C9	0.1016 (4)	0.07988 (8)	0.5839 (3)	0.040
C9A	0.2342 (4)	0.09719 (8)	0.5032 (3)	0.041
C10	0.2506 (4)	0.04093 (8)	0.2968 (4)	0.048
C10A	0.1147 (4)	0.02073 (8)	0.3786 (4)	0.046
C11	0.3059 (4)	0.16527 (8)	0.3933 (3)	0.042
C12	0.3757 (4)	0.14815 (9)	0.2547 (4)	0.047
C13	0.2044 (4)	0.20387 (8)	0.4032 (4)	0.046
C14	0.1828 (5)	0.27069 (10)	0.5294 (6)	0.073
C15	0.3829 (5)	0.16717 (10)	0.0825 (4)	0.055
C16	0.3431 (12)	0.22883 (14)	-0.0757 (6)	0.143
C17	0.0260 (4)	0.10091 (8)	0.7350 (4)	0.041
C18	-0.0606 (5)	0.13828 (10)	0.7147 (5)	0.056
C19	-0.1263 (6)	0.15927 (12)	0.8535 (5)	0.072
C20	-0.1062 (6)	0.14289 (13)	1.0156 (5)	0.068
C21	-0.0214 (5)	0.10628 (12)	1.0363 (4)	0.063
C22	0.0445 (4)	0.08505 (10)	0.8991 (4)	0.054

Table 2. Selected geometric parameters (Å, °)

C1—C2	1.523 (4)	C9—C17	1.492 (4)
C1—C9A	1.531 (4)	C10—C10A	1.426 (5)
C1—C11	1.519 (4)	C11—C12	1.341 (4)
C2—C3	1.312 (5)	C11—C13	1.477 (4)
C3—C4	1.526 (5)	C12—C15	1.481 (4)
C4—C4A	1.536 (4)	C17—C18	1.387 (4)
C4—C12	1.519 (4)	C17—C22	1.391 (4)
C4A—C9A	1.421 (4)	C18—C19	1.383 (5)
C4A—C10	1.350 (4)	C19—C20	1.384 (6)
C5—C6	1.370 (5)	C20—C21	1.359 (5)
C5—C10A	1.416 (4)	C21—C22	1.378 (5)
C6—C7	1.385 (5)	C13—O1	1.308 (4)
C7—C8	1.377 (4)	C13—O2	1.196 (4)
C8—C8A	1.407 (4)	C14—O1	1.437 (4)
C8A—C9	1.442 (4)	C15—O3	1.311 (4)
C8A—C10A	1.423 (4)	C15—O4	1.182 (4)
C9—C9A	1.362 (4)	C16—O3	1.425 (4)
C2—C1—C9A	105.9 (2)	C5—C10A—C8A	118.4 (3)
C2—C1—C11	107.8 (2)	C5—C10A—C10	122.0 (3)
C9A—C1—C11	103.6 (2)	C8A—C10A—C10	119.6 (2)
C1—C2—C3	114.0 (3)	C1—C11—C12	113.1 (2)
C2—C3—C4	114.0 (3)	C1—C11—C13	118.7 (2)
C3—C4—C4A	104.8 (3)	C12—C11—C13	127.8 (3)
C3—C4—C12	106.4 (2)	C4—C12—C11	113.9 (3)
C4A—C4—C12	105.2 (3)	C4—C12—C15	119.9 (3)
C4—C4A—C9A	111.9 (2)	C11—C12—C15	126.2 (3)
C4—C4A—C10	126.9 (3)	C11—C13—O1	111.5 (3)
C9A—C4A—C10	121.2 (3)	C11—C13—O2	124.7 (3)
C6—C5—C10A	121.3 (3)	O1—C13—O2	123.4 (3)
C5—C6—C7	120.2 (3)	C12—C15—O3	112.5 (3)
C6—C7—C8	120.3 (3)	C12—C15—O4	124.2 (3)
C7—C8—C8A	121.2 (3)	O3—C15—O4	123.3 (3)
C8—C8A—C9	121.8 (3)	C9—C17—C18	119.9 (3)
C8—C8A—C10A	118.6 (3)	C9—C17—C22	122.0 (2)
C9—C8A—C10A	119.6 (3)	C18—C17—C22	118.1 (3)
C8A—C9—C9A	118.2 (3)	C17—C18—C19	121.1 (3)
C8A—C9—C17	120.9 (2)	C18—C19—C20	119.9 (3)
C9A—C9—C17	120.8 (2)	C19—C20—C21	119.4 (3)
C1—C9A—C4A	111.4 (2)	C20—C21—C22	121.4 (3)
C1—C9A—C9	126.8 (2)	C17—C22—C21	120.3 (3)
C4A—C9A—C9	121.8 (2)	C13—O1—C14	117.4 (3)
C4A—C10—C10A	119.5 (3)	C15—O3—C16	117.5 (3)

The structure was determined by direct methods. H-atom positions were refined isotropically except for methyl H atoms which were fixed. Data collection, cell refinement, data reduction, structure solution and refinement, and graphical represen-

tation were performed using *TEXSAN* (Molecular Structure Corporation, 1990).

We thank the Natural Sciences and Engineering Research Council of Canada for financial support.

Lists of structure factors, anisotropic displacement parameters, H-atom coordinates and complete geometry, along with stereo molecular and packing diagrams have been deposited with the British Library Document Supply Centre as Supplementary Publication No. SUP 71667 (26 pp.). Copies may be obtained through The Technical Editor, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England. [CIF reference: BR1057]

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 Pokkuluri, P. R., Scheffer, J. R. & Trotter, J. (1993). *Acta Cryst.* **B49**, 754–760.

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Intermolecular Hydrogen-Bonded Structure of Bis(4-methoxyphenylmethine)carbonylhydrazide

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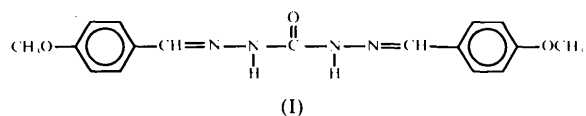
Abstract

The first reported X-ray crystal structure of a carbazone is described. [For a discussion of CAS/IUPAC nomenclature of carbazones see Grasselli & Weast (1989). In *Handbook of Data on Organic Compounds*, 2nd edition, Vol. I, Introduction, §§190, 252. Florida: CRC Press.] Crystalline 1,5-bis(4-methoxyphenylmethine)carbonylhydrazide, (I), $C_{17}H_{18}N_4O_3$, exists as a non-centrosymmetric infinite hydrogen-bonded chain in which both amidic H

atoms of one molecule are tightly hydrogen bonded to the carbonyl O atom of another molecule, forming intermolecular six-membered ring linkages. The intermolecular hydrogen-bond distances for N—H \cdots O are 1.91 Å for H \cdots O and 2.759 (5) Å for N \cdots O, and the N—H \cdots O angles are 148.0°.

Comment

IR spectra of a series of semicarbazones in the solid state were found to exhibit unexpectedly high carbonyl frequencies (Kolb, Stupar, Janota & Duax, 1989; Kolb, Janota, Dantzman, Kozenski, Strommen, Snyder & Tipsword, 1992). Through a combination of Raman spectroscopic and X-ray crystallographic studies these unpredicted frequency shifts were attributed to vibrational coupling of the C=O bands in the solid-state hydrogen-bonded network (Kolb, Janota, Dantzman, Kozenski, Strommen, Snyder & Tipsword, 1992; Kolb, Dantzman, Kozenski & Strommen, 1993). The Raman and IR carbonyl frequencies of a series of related carbazones in the solid state were then examined in an attempt to deduce their structure (Kolb, Janota, Dantzman, Kozenski & Strommen, 1992). To investigate more specifically the role played by hydrogen bonding and to correlate the carbonyl frequencies with patterns of hydrogen bonding, we undertook X-ray studies of the crystal structures of these carbazones (Kolb, Meyers & Robinson, 1993). Although the crystal structure of carbazide itself, $NH_2NHC(O)NHNH_2$, has been studied by X-ray diffraction (Domiano, Pellinghelli & Tiripicchio, 1972) and more recently by neutron diffraction (Jeffrey, Ruble, Nanni, Turano & Yates, 1985), no X-ray structure of a carbazone has been reported previously. We describe here the intermolecular hydrogen-bonded crystal structure of bis(4-methoxyphenylmethine)carbonylhydrazide (I).



A solution of 762 mg (5.6 mmol) of *p*-methoxybenzaldehyde and 250 mg (2.8 mmol) of carbazide in 20 ml of ethanol was refluxed overnight. The cooled solution yielded a mass of colorless crystals of (I) which were collected by filtration and recrystallized from ethanol to a constant melting point, 479–482 K. The well defined pattern of intermolecular hydrogen bonding in the crystal structure of (I) is shown in Fig. 1 and the spatial orientation of the hydrogen-bonded molecules is illustrated in Fig. 2.

Several important structural features are exhibited. (1) The individual molecules have an essentially planar and symmetrical conformation in which the amidic N atoms are trigonal and the amidic H atoms are *anti* to the carbonyl O atom. (2) Both amidic H atoms of one