2-Diethylamino-4,5-diphenyl-3*H*-pyrrol-3-one

By Hans Preut, Theophil Eicher and Günter Franke

*Abteilung Chemie der Universität Dortmund, Postfach 500500, D-4600 Dortmund 50,
Federal Republic of Germany*

and Carl Krüger

*Max-Planck-Institut für Kohlenforschung, Lembkestrasse 5, D-4330 Mülheim/Ruhr,
Federal Republic of Germany*

Abstract. $C_{20}H_{20}N_2O$, $M_r = 304.4$, monoclinic, $a = 13.195$ (4), $b = 17.389$ (5), $c = 15.542$ (2) Å, $\beta = 108.07$ (2)°, $U = 3390$ Å³; $Z = 8$, $D_c = 1.193$ Mg m⁻³; $F(000) = 1296$, Mo $K\alpha$ radiation, $\lambda = 0.71069$ Å, μ(Mo $K\alpha$) = 0.41 mm⁻¹. Space group $P2_1/a$. Final $R = 0.065$ for 2979 unique X-ray diffractometer data. The molecular structure has been elucidated.

Introduction. Diphenylcyclopropenone (I) reacts with N,N,N',N'-tetraalkyl-substituted guanidine (II) to give the blue 2-amino-4,5-diphenyl-3*H*-pyrrol-3-ones (III) (Eicher, Abdesaken, Franke & Weber, 1979). These five-membered heterocycles exhibit remarkable spectroscopic properties (Eicher & Franke, 1981), especially in their electronic spectra (Klessinger, 1978), in comparison to other azacyclopentadienones, thus indicating a strongly delocalized π system described by means of a mesomeric cyclomerocyanine structure (III A) ↔ (III B). These aspects made it desirable to determine the molecular structure of a compound of type (III) in order to obtain information on the degree of bond delocalization described by (III A) ↔ (III B).
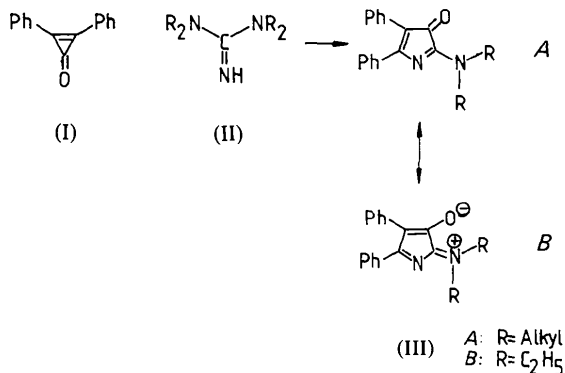
Cell dimensions were obtained by least squares from the θ angles of 58 reflexions. The intensities of 4808 reflexions were measured on a Nonius CAD-4 diffrac-

tometer with graphite-monochromated Mo $K\alpha$ radiation by the $\omega/2\theta$ scan technique up to $2\theta = 46°$. After averaging, 2979 independent observed reflexions ($I > 0$) remained. The intensities were corrected for Lorentz and polarization effects but not for absorption. The structure was solved by direct methods with *MULTAN* 78 (Main, Woolfson, Lessinger, Germain & Declercq, 1978) and refined by blocked full-matrix least squares with *SHELX* (Sheldrick, 1976) with all atoms except H anisotropic. H atoms were placed in geometrically calculated positions (C–H 1.08 Å, H–C–H 109.5° in CH_2 and CH_3 groups, C–C–H angles equal); the phenyl groups were refined as rigid bodies. The scattering factors were taken from Cromer & Mann (1968) and Cromer & Liberman (1970) for C, N and O and from Cromer (personal communication to G. M. Sheldrick) for H. Refinement converged with unit weights to $R = 0.065$.* The figures were drawn with *PLUTO* (Motherwell & Clegg, 1978) and *POP*1 (van de Waal, 1976).

Discussion. The structure of the title compound is shown in Fig. 1; a stereoview of the molecule is given in Fig. 2. The positional parameters and the equivalent values of the anisotropic temperature factors U_{ik} [$U_{eq} = \frac{1}{3}(U_{11} + U_{22} + U_{33} + 2U_{23} \cos \alpha + 2U_{13} \cos \beta + 2U_{12} \times \cos \gamma)$] for the heavy atoms and the isotropic temperature factors U for H atoms are given in Table 1. Bond lengths and angles are given in Table 2. The asymmetric unit contains two molecules with significant differences in the bond lengths and angles. An indication of bond delocalization in the heterocyclic system cannot be derived from these results. Signifi-

---

* Lists of structure factors, thermal parameters and a complete list of atoms have been deposited with the British Library Lending Division as Supplementary Publication No. SUP 36540 (22 pp.). Copies may be obtained through The Executive Secretary, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.

Table 1. *Fractional atomic coordinates* ($\times 10^4$) *and U*
($\text{Å}^2 \times 10^4$)

For each rigid phenyl ring only the coordinates of three C atoms
are given.

	x	y	z	U_{eq} or U
C(1)	9014 (3)	604 (2)	5710 (3)	51 (2)
C(2)	8020 (3)	259 (2)	5395 (3)	51 (2)
C(3)	8850 (3)	−468 (2)	6558 (3)	57 (2)
C(4)	9613 (3)	139 (2)	6445 (3)	52 (2)
C(5)	8247 (4)	−1597 (3)	7204 (4)	91 (3)
H(51)	8634 (4)	−2138 (3)	7446 (4)	201 (13)
H(52)	7708 (4)	−1671 (3)	6527 (4)	201 (13)
C(6)	7629 (5)	−1337 (4)	7811 (4)	114 (4)
H(61)	7036 (5)	−1763 (4)	7816 (4)	201 (13)
H(62)	7242 (5)	−797 (4)	7569 (4)	201 (13)
H(63)	8167 (5)	−1263 (4)	8489 (4)	201 (13)
C(7)	98 (4)	−1059 (3)	7911 (4)	90 (33)
H(71)	462 (4)	−496 (3)	8015 (3)	147 (9)
H(72)	9964 (4)	−1250 (3)	8528 (3)	147 (9)
C(8)	849 (5)	−1616 (4)	7669 (4)	111 (4)
H(81)	1594 (5)	−1636 (4)	8212 (4)	147 (9)
H(82)	993 (5)	−1428 (4)	7055 (4)	147 (9)
H(83)	495 (5)	−2182 (4)	7568 (4)	147 (9)
O(1)	562 (3)	187 (2)	6903 (2)	94 (2)
N(1)	7890 (3)	−390 (2)	5911 (2)	61 (2)
N(2)	9068 (3)	−1008 (2)	7193 (2)	70 (2)
C(11)	9437 (2)	1309 (2)	5417 (2)	54 (2)
C(13)	9316 (2)	2665 (2)	5062 (2)	92 (3)
C(15)	854 (2)	1938 (2)	5006 (2)	88 (3)
C(21)	7120 (2)	499 (2)	4583 (2)	54 (2)
C(23)	6425 (2)	1017 (2)	3097 (2)	78 (3)
C(25)	5233 (2)	387 (2)	3781 (2)	105 (3)
C'(1)	7689 (3)	5236 (2)	9892 (3)	52 (2)
C'(2)	8393 (3)	5447 (2)	9439 (3)	54 (2)
C'(3)	7804 (4)	4374 (3)	8772 (3)	65 (2)
C'(4)	7221 (3)	4528 (2)	9473 (3)	60 (2)
C'(5)	7080 (4)	3047 (3)	8372 (4)	82 (3)
H'(51)	6973 (4)	3061 (3)	9033 (4)	161 (10)
H'(52)	7552 (4)	2551 (3)	8323 (4)	161 (10)
C'(6)	6041 (5)	2991 (3)	7687 (4)	100 (3)
H'(61)	5647 (5)	2472 (3)	7795 (4)	161 (10)
H'(62)	6142 (5)	2974 (3)	7024 (4)	161 (10)
H'(63)	5564 (5)	3484 (3)	7733 (4)	161 (10)
C'(7)	7990 (7)	3833 (4)	7276 (6)	162 (6)
H'(71)	8166 (7)	4412 (4)	7117 (6)	267 (20)
H'(72)	7418 (7)	3578 (4)	6692 (6)	267 (20)
C'(8)	8810 (6)	3425 (5)	7625 (7)	164 (6)
H'(81)	9240 (6)	3354 (5)	7142 (7)	267 (20)
H'(82)	8569 (6)	2868 (5)	7801 (7)	267 (20)
H'(83)	9318 (6)	3702 (5)	8226 (7)	267 (20)
O'(1)	6525 (3)	4130 (2)	9619 (2)	81 (2)
N'(1)	8465 (3)	4942 (2)	8760 (2)	68 (2)
N'(2)	7653 (4)	3762 (3)	8243 (3)	99 (3)
C'(11)	7482 (2)	5541 (1)	709 (2)	53 (2)
C'(13)	8116 (2)	5955 (1)	2269 (2)	78 (2)
C'(15)	6224 (2)	5850 (1)	1503 (2)	85 (2)
C'(21)	9082 (2)	6134 (2)	9564 (2)	48 (2)
C'(23)	692 (2)	6715 (2)	9432 (2)	63 (3)
C'(25)	9489 (2)	7461 (2)	9997 (2)	65 (3)

$U_{ik} \times 10^3 \ \{\exp\left[-2\pi^2(U_{11}h^2 a^{*2} + \ldots + 2U_{12}hka^* b^*)\right]\}$ for the disordered atoms C'(7) and C'(8)

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
C'(7)	143 (7)	113 (6)	215 (10)	33 (6)	−25 (7)	−25 (5)
C'(8)	102 (6)	142 (7)	269 (11)	48 (7)	34 (7)	−2 (5)

Table 2. *Bond lengths* (Å) *and angles* (°)

	Molecule 1	Molecule 2
C(1)—C(2)	1·385 (5)	1·378 (5)
C(2)—N(1)	1·426 (5)	1·398 (5)
N(1)—C(3)	1·357 (5)	1·322 (5)
C(3)—C(4)	1·505 (5)	1·539 (6)
C(4)—C(1)	1·423 (6)	1·439 (6)
C(1)—C(11)	1·477 (5)	1·477 (5)
C(2)—C(21)	1·477 (5)	1·479 (4)
C(3)—N(2)	1·328 (4)	1·321 (6)
N(2)—C(5)	1·496 (6)	1·501 (6)
C(5)—C(6)	1·494 (7)	1·454 (7)
N(2)—C(7)	1·469 (4)	*
C(7)—C(8)	1·514 (7)	*
C(4)—O(1)	1·236 (4)	1·226 (5)
C(2)—C(1)—C(4)	105·5 (3)	105·0 (3)
C(2)—C(1)—C(11)	131·1 (3)	132·6 (3)
C(4)—C(1)—C(11)	123·4 (3)	122·0 (3)
C(1)—C(2)—N(1)	114·2 (3)	115·7 (3)
C(1)—C(2)—C(21)	128·9 (3)	129·4 (3)
N(1)—C(2)—C(21)	116·9 (3)	115·0 (3)
N(1)—C(3)—C(4)	110·4 (3)	110·9 (4)
N(1)—C(3)—N(2)	123·9 (3)	124·5 (4)
C(4)—C(3)—N(2)	125·7 (3)	124·6 (4)
C(3)—C(4)—C(1)	105·4 (3)	103·4 (3)
C(3)—C(4)—O(1)	125·1 (4)	126·9 (4)
C(1)—C(4)—O(1)	129·4 (4)	129·7 (4)
C(3)—N(2)—C(5)	120·2 (3)	124·8 (4)
C(3)—N(2)—C(7)	122·9 (4)	*
C(5)—N(2)—C(7)	116·9 (4)	*
N(2)—C(5)—C(6)	109·8 (4)	110·9 (4)
N(2)—C(7)—C(8)	111·9 (4)	*
C(1)—C(11)—C(12)	121·2 (2)	120·7 (2)
C(1)—C(11)—C(16)	118·7 (3)	119·2 (2)
C(2)—C(21)—C(22)	120·9 (2)	118·0 (2)
C(2)—C(21)—C(26)	119·1 (3)	122·0 (2)

* The abnormal anisotropic temperature factors of the atoms
C'(7) and C'(8) indicate disorder. The corresponding distances and
angles cannot be interpreted as bond distances and bond angles
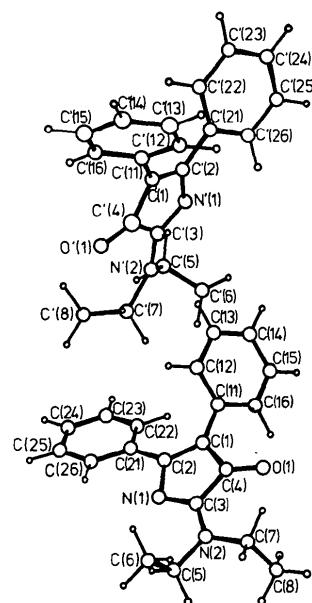and are omitted in this table.
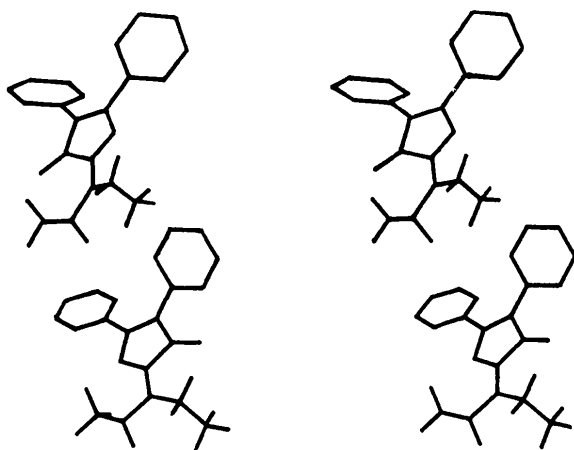

Fig. 1. General view of the molecule.

Fig. 2. Stereoscopic view of the two independent molecules.

cant differences in the orientation of the phenyl groups and the ethyl groups with respect to the five-membered heterocycle and the distances of the nearest inter-molecular neighbours to the five-membered hetero-cycle, $C(3) \cdots C$ 3·547 (6), $C(4) \cdots C$ 3·607 (6), $N(1) \cdots C$ 3·629 (6), $C'(3) \cdots C$ 3·447 (6), $C'(4) \cdots C$

3·499 (6), $N'(1) \cdots C$ 3·480 (6) Å, indicate that the two independent molecules are deformed differently by packing forces.

References

Cromer, D. T. & Liberman, D. (1970). *J. Chem. Phys.* **53**, 1891–1898.
Cromer, D. T. & Mann, J. B. (1968). *Acta Cryst.* **A24**, 321–324.
Eicher, Th., Abdesaken, F., Fanke, G. & Weber, J. L. (1979). *Tetrahedron Lett.* pp. 3915–3918.
Eicher, Th. & Franke, G. (1981). *Liebigs Ann. Chem.* In the press.
Klessinger, M. (1978). *Chem. Unserer Zeit*, **12**, 1.
Main, P., Woolfson, M. M., Lessinger, L., Germain, G. & Declercq, J. P. (1978). *MULTAN 78. A System of Computer Programs for the Automatic Solution of Crystal Structures from X-ray Diffraction Data.* Univs. of York, England, and Louvain-la-Neuve, Belgium.
Motherwell, W. D. S. & Clegg, W. (1978). *PLUTO. Program for Plotting Molecular and Crystal Structures.* Univs. of Cambridge, England, and Göttingen, Federal Republic of Germany.
Sheldrick, G. M. (1976). *SHELX. A Program for Crystal Structure Determination.* Univ. of Cambridge, England.
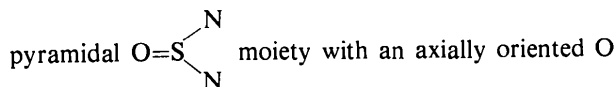Waal, B. W. van de (1976). *POP1. Plot of Packing Program.* Twente Univ. of Technology, The Netherlands.

4-Phenoxy-10H-benzimidazo[2,1-c][1λ⁴,2,4,6]thiatriazine 2-Oxide

By Alajos Kálmán and Gyula Argay

Central Research Institute of Chemistry, Hungarian Academy of Sciences, Budapest 114, POB 17, H-1525, Hungary

Abstract. $C_{14}H_{10}N_4O_2S$, monoclinic, $P2_1/n$, $a = 9\cdot329$ (2), $b = 4\cdot603$ (5), $c = 30\cdot124$ (7) Å, $\beta = 92\cdot57$ (2)°, $V = 1292$ (2) Å³, $D_c = 1\cdot533$ Mg m⁻³, $Z = 4$, $F(000) = 616$. The structure was solved by direct methods and refined by full-matrix least squares to a final $R = 0\cdot068$ for 1089 intensities. A rare trigonal-pyramidal O=S(N)(N) moiety with an axially oriented O atom is embedded in the non-planar thiatriazine ring. However, the bonding and conformation of the thiatriazine S-oxide ring resemble those observed for 1,4-dihydro-3,5-dimethoxy-1-tosylimino-1λ⁴,2,4,6-thiatriazine [Kálmán, Argay, Fischer & Teller (1981). *Acta Cryst.* B37, 164–168]. The S=O distance of

1·452 (7) Å is considerably shorter than the 1·51 Å observed in various sulphoxides.

Introduction. As established by this X-ray analysis the reaction of 2-amino-1-phenoxyformimidoylbenzimi-dazole with N-sulphinyl-p-toluenesulphonamide and a consecutive but unexpected hydrolysis of the inter-mediate* resulted in the title compound. Intensities were collected on an Enraf–Nonius CAD-4 diffractom-eter equipped with a graphite monochromator using Mo $K\alpha$ radiation ($\lambda = 0\cdot71073$ Å). Cell constants were determined by least-squares refinement from the setting angles of 25 reflexions; of the total of 1231 measure-

---

* A protected synthesis of the parent 1-tosyliminothiatriazine carried out by Professor E. Fischer and Dr M. Teller is in progress.