

4-(4-Chlorophenyl)-3-(2,6-dichlorophenyl)-1,7-dioxo-2-azaspiro[4.4]non-2-en-6-one

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Key indicators

Single-crystal X-ray study

T = 293 K

Mean $\sigma(\text{C}-\text{C}) = 0.005 \text{ \AA}$

R factor = 0.049

wR factor = 0.108

Data-to-parameter ratio = 13.6

For details of how these key indicators were automatically derived from the article, see <http://journals.iucr.org/e>.

The title compound, $\text{C}_{18}\text{H}_{12}\text{Cl}_3\text{NO}_3$, was synthesized by the intermolecular [3 + 2]-cycloaddition of 2,6-dichlorobenzonitrile oxide and 3-(4-chlorobenzylidene)dihydrofuran-2-one. A spiro junction in the molecule links an isoxazoline ring and a dihydrofuran-2-one ring. Both rings are non-planar, with envelope conformations.

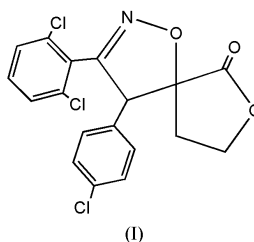
Received 24 March 2003

Accepted 9 April 2003

Online 23 April 2003

Comment

Spiro-compounds represent an important class of naturally occurring substances, characterized by highly pronounced biological properties (Kobayashi *et al.*, 1991; James *et al.*, 1991). 1,3-Dipolar cycloaddition reactions are important processes for the construction of spiro-compounds (Caramella & Grunanger, 1984). The molecular structure of the title compound, (I), is illustrated in Fig. 1.



A spiro junction in the molecule links an isoxazoline ring and a dihydrofuran-2-one ring. The isoxazoline ring (O1/N1/C6/C5/C2) has an envelope conformation. Atoms O1/N1/C6/C5 form an almost perfect plane [mean deviation from this plane is 0.0053 (3) Å]. Spiro-atom C2 lies 0.3309 (3) Å from this plane and forms the flap of the envelope. The dihedral angle between plane C5/C2/O1 and the mean plane O1/N1/C6/C5 is 21.1 (4)°. This is similar to the conformation found for isoxazoline rings in the literature (Li, Feng & Gu, 2003; Li, Feng, Zhuang & Hu, 2003). The O1–N1, C6–N1 and O1–C2 bond lengths are 1.423 (3), 1.276 (4) and 1.466 (3) Å, respectively, and the angles O1–N1–C6 and C2–O1–N1 are 109.2 (2) and 108.3 (2)°, respectively. These values compare with the values of 1.425 (4), 1.268 (4) and 1.440 (4) Å, and 109.0 (3) and 110.5 (2)° reported in the literature (Li, Feng, Zhuang & Hu, 2003). The torsion angle C6–N1–O1–C2 is 12.6 (3)°. The dihedral angle between plane O1/N1/C6/C5 and the phenyl ring (C7–C12) is 99.2 (4)°, and that between plane O1/N1/C6/C5 and the phenyl ring (C13–C18) is 109.9 (4)°. The dihedral angle between phenyl rings C7–C12 and C13–C18 is 95.1 (4)°. The dihydrofuran-2-one ring (C1–C4/O3) has an envelope conformation; atoms C1/C2/O3/C4 lie in a plane, the mean deviation from this plane being 0.0132 (3) Å. Atom C3 lies 0.5384 (4) Å from

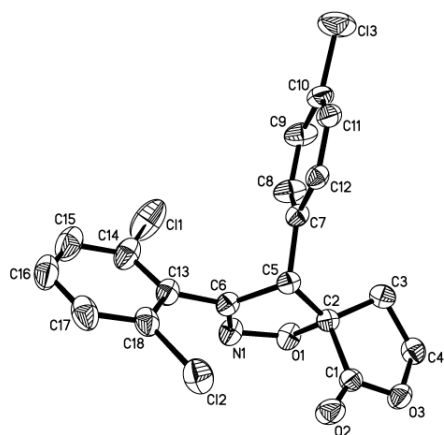


Figure 1
The molecular structure of (I), with displacement ellipsoids at the 30% probability level, and the atom-numbering scheme.

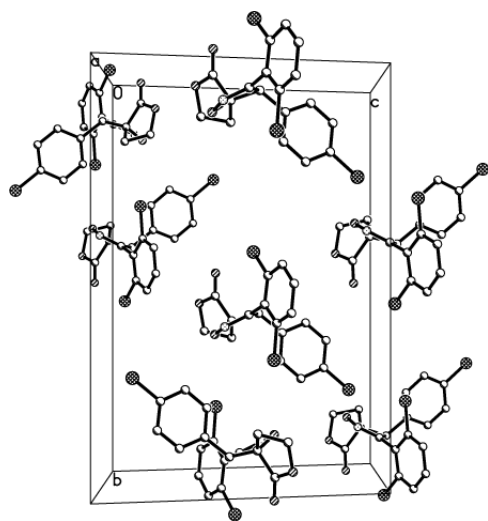


Figure 2
The crystal structure of (I), viewed down the *a* axis.

this plane and forms the flap of the envelope. The dihedral angle between planes C2/C3/C4 and C2/C1/O3/C4 is 34.4 (4)°. The O2—C1, O3—C4 and O3—C1 bond lengths are 1.197 (4), 1.450 (4) and 1.345 (4) Å, respectively, and bond angles O2—C1—O3, C1—O3—C4 and C1—C2—C3 are 121.9 (3), 110.2 (3) and 101.2 (3)°, respectively. These values compare well with those of 1.197 (2), 1.443 (2) and 1.342 (2) Å, and 121.2 (18), 110.9 (15) and 102.48 (15)° reported in the literature (Guzei *et al.*, 2002).

Experimental

A mixture of 2,6-dichlorobenzonitrile oxide (2 mmol) and 3-(4-chlorobenzylidene)dihydrofuran-2-one in dry chloroform (30 ml) was heated under reflux for 4 d. After evaporation of the solvent, the residue was separated by column chromatography (silica gel, petroleum ether/ethyl acetate = 8:1) to give the title compound, (I). M.p. 464–465 K; IR (KBr): 1784 (C=O), 1600, 1581 (C=N, C=C) cm⁻¹; ¹H NMR (CHCl₃) δ = 1.92 (1H, *m*), 2.44 (1H, *m*), 4.26 (1H, *m*), 4.51 (1H, *m*), 5.73 (1H, *s*), 7.17–7.33 (7H, *m*). 20 mg of (I) were dissolved

in 15 ml chloroform, and colourless single crystals, suitable for X-ray analysis, were obtained by slow evaporation at room temperature over 15 d.

Crystal data

C₁₈H₁₂Cl₃NO₃
M_r = 396.64
Monoclinic, C2/c
a = 12.600 (6) Å
b = 20.177 (9) Å
c = 14.202 (6) Å
β = 104.768 (8)°
V = 3491 (3) Å³
Z = 8

D_x = 1.509 Mg m⁻³
Mo Kα radiation
Cell parameters from 699 reflections
θ = 3.5–23.3°
μ = 0.54 mm⁻¹
T = 293 (2) K
Plate, colourless
0.20 × 0.10 × 0.06 mm

Data collection

Bruker SMART CCD area-detector diffractometer
φ and ω scans
Absorption correction: multi-scan (SADABS; Sheldrick, 1997)
*T*_{min} = 0.937, *T*_{max} = 0.968
8872 measured reflections

3078 independent reflections
1702 reflections with *I* > 2σ(*I*)
*R*_{int} = 0.051
θ_{max} = 25.0°
h = -14 → 14
k = -24 → 21
l = -11 → 16

Refinement

Refinement on *F*²
R [*F*² > 2σ(*F*²)] = 0.049
wR (*F*²) = 0.108
S = 1.01
3078 reflections
226 parameters

H-atom parameters constrained
w = 1/[σ²(*F_o*²) + (0.084*P*)²]
where *P* = (*F_o*² + 2*F_c*²)/3
(Δ/σ)_{max} < 0.001
Δρ_{max} = 0.34 e Å⁻³
Δρ_{min} = -0.21 e Å⁻³

Table 1

Selected geometric parameters (Å, °).

Cl3—C10	1.742 (4)	O1—C2	1.466 (3)
N1—C6	1.276 (4)	O2—C1	1.197 (4)
N1—O1	1.423 (3)	O3—C1	1.345 (4)
C6—N1—O1	109.2 (2)	O2—C1—O3	121.9 (3)
N1—O1—C2	108.3 (2)	O1—C2—C5	104.3 (2)
C1—O3—C4	110.2 (3)		
C6—N1—O1—C2	12.6 (3)	O3—C1—C2—O1	89.5 (3)

The H atoms were included in the riding-model approximation with displacement parameters related to the atoms to which they were bonded.

Data collection: SMART (Bruker, 1997); cell refinement: SMART; data reduction: SAINT (Bruker, 1997); program(s) used to solve structure: SHELXS97 (Sheldrick, 1997); program(s) used to refine structure: SHELXL97 (Sheldrick, 1997); molecular graphics: SHELXTL (Bruker, 1997); software used to prepare material for publication: SHELXTL.

References

- Bruker (1997). SMART, SAINT and SHELXTL. Versions 5.10. Bruker AXS Inc., Madison, Wisconsin, USA.
Caramella, P. & Grunanger, P. (1984). 1,3-Dipolar Cycloaddition Chemistry, edited by A. Padwa, Vol. 1, pp. 291–312. New York: Wiley.
Guzei, I. A., Langenhan, J. M. & Chung, Y. J. (2002). Acta Cryst. E58, o65–o66.
James, D., Kunze, H. B. & Faulkner, D. (1991). J. Nat. Prod. 54, 1137–1140.
Kobayashi, J., Tsuda, M., Agemi, K., Shigemori, H., Ishibashi, M., Sasaki, T. & Mikami, Y. (1991). Tetrahedron. 47, 6617–6622.
Li, X.-F., Feng, Y.-Q. & Gu, Y.-F. (2003). Acta Cryst. E59, o330–o331.
Li, X.-F., Feng, Y.-Q., Zhuang, J.-P. & Hu, X.-F. (2003). Acta Cryst. E59, o247–o249.
Sheldrick, G. M. (1997). SADABS, SHELXS97 and SHELXL97. University of Göttingen, Germany.