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1-Benzyl-5-(1-naphthylmethylene)piperidine-spiro-3,3′-1′-methyl-4′-(1-naphthyl)pyrrolidine-spiro-2′,3″-indole-2″,4-dione–diethyl ether (1/0.75)

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Key indicators

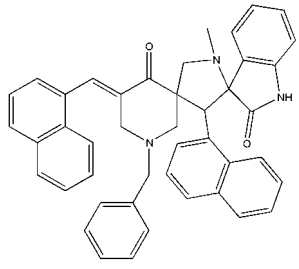
Single-crystal X-ray study
T = 293 K
Mean σ(C–C) = 0.005 Å
Disorder in solvent or counterion
R factor = 0.062
wR factor = 0.188
Data-to-parameter ratio = 13.3

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

The title compound, $C_{44}H_{37}N_3O_2\cdot0.75(C_2H_5)_2O$, was synthesized by the intermolecular [3 + 2] cycloaddition of the azomethine ylide, derived from isatin and sarcosine by a decarboxylative route, and 1-benzyl-3,5-bis-naphth-1-yl-methylene-piperidin-4-one. In the molecule, the two spiro junctions link a planar 2-oxindole ring, a pyrrolidine ring in an envelope conformation and a piperidone in a boat conformation. Molecules are connected into chains along the **a** direction by O—H···N hydrogen bonds, with an O···N distance of 2.944 (3) Å.

Comment

Spiro compounds represent an important class of naturally occurring substances characterized by highly pronounced biological properties (Kobayashi *et al.*, 1991; James & Kunze, 1991). 1,3-dipolar cycloaddition reactions are important in the construction of spiro compounds (Caramella & Grunanger, 1984). The molecular structure of (I) is illustrated in Fig. 1. The two spiro junctions in the molecule link a 2-oxindole ring, a pyrrolidine ring and a piperidone ring. The pyrrolidine ring (N2/C7/C9/C10/C11) is in an envelope conformation; the atoms C7/C9/C10/C11 are almost coplanar, with a mean deviation of 0.031 (3) Å, and N2 is 0.594 (2) Å from this plane, forming the flap of the envelope. However, in the previously reported structure, 1″-benzyl-5″-benzylidene-1′-methyl-4′-phenyl-1*H*-indole-3-spiro-2′-pyrrolidine-3′-spiro-3″- piperidine-2(3*H*),3″-dione, it is the C atom of the —CH_2— group of the pyrrolidine which forms the flap of the envelope (Li *et al.*, 2003), rather than the N atom.



(I)

The dihedral angle between the C7/N2/C11 and C7/C9/C10/C11 least-squares planes is 137.3 (2)°. The dihedral angle between the naphthalene plane (C17/C18/C19/C20/C21/C22/C23/C24/C25/C26) and C7/C9/C10/C11 plane is 108.4 (3)°. The 2-oxindole ring system (C1/C2/C3/C4/C5/C6/C7/C8/N1) is nearly planar, with a mean deviation from the plane of 0.042 (3) Å and a dihedral angle between the 2-oxindole ring and the C7/C9/C10/C11 plane of 82.0 (2)°. The piperidone ring has a boat conformation, which is different from the chair

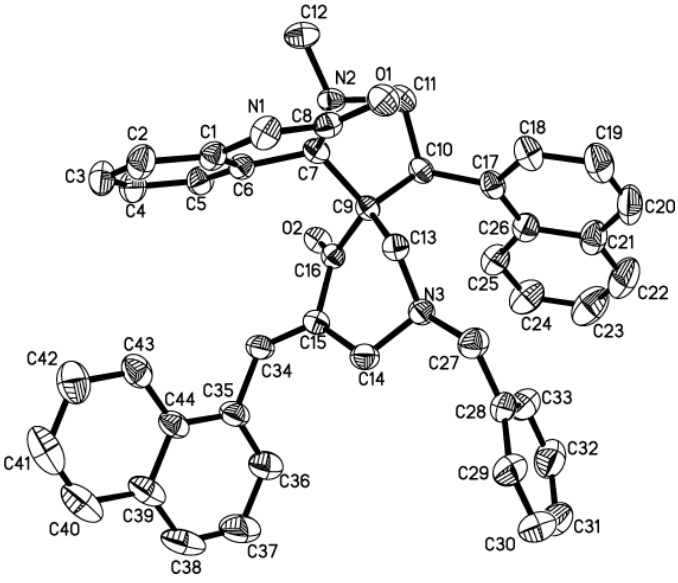


Figure 1
The molecular structure of (I), drawn with 30% probability displacement ellipsoids. H atoms are omitted.

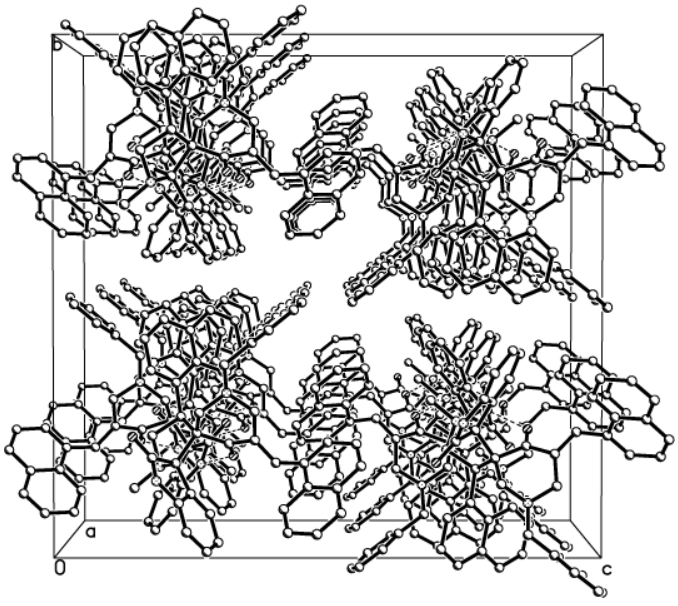


Figure 2
The crystal packing of (I), viewed along the *a* axis, with dashed lines indicating hydrogen bonds. The disordered ether molecules are not shown.

conformation of the piperidone ring in 1″-benzyl-5″-benzylidene-1′-methyl-4′-phenyl-1*H*-indole-3-spiro-2′-pyrrolidine-3′-spiro-3″-piperidine-2(3*H*),3″-dione (Li *et al.*, 2003). Molecules are connected into chains along the **a** direction *via* O—H···N hydrogen bonds. The O···N and H···N distances are 2.944 (3) and 2.11 Å, respectively, and the O—H···N angle is 162°.

Experimental

A mixture of 1-benzyl-3,5-bis-naphth-1-ylmethylene-piperidin-4-one (2 mmol), isatin (2 mmol) and sarcosine (2 mmol) were refluxed in methanol (80 ml) until the starting material had disappeared (as evidenced by TLC). After the reaction was complete, the solvent was removed *in vacuo* and the residue was separated by column chromatography (silica gel, petroleum ether/ethylcetate 5:1) to give the title compound, (I). M.p. 439–440 K; IR (KBr): 3490(N—H), 1734.1, 1718.7 (C═O), 1614.5(C═C) cm^{-1}; ^{1}H NMR ($CDCl_3$, p.p.m.): 1.76 (*d*, *J* = 12.9 Hz, 1H), 2.14 (*s*, 3H), 2.50 (*dd*, *J* = 2.7, 14.7 Hz, 1H), 2.90 (*d*, *J* = 13.5 Hz, 1H), 3.15 (*d*, *J* = 14.7 Hz, 1H), 3.42–3.55 (*m*, 3H), 4.20 (*dd*, *J* = 9.3, 10.8 Hz, 1H), 5.67 (*dd*, *J* = 7.5, 9.9 Hz, 1H), 7.12 (*s*, 1H), 7.01–8.24 (*m*, 23H), 7.94 (*bs*, 1H). 20 mg of (I) was dissolved in 15 ml of a mixture of chloroform and ether and the solution was kept at room temperature for 15 days to undergo natural evaporation. This afforded colorless single crystals of (I) suitable for X-ray analysis.

Crystal data

$C_{44}H_{37}N_3O_2{\cdot}0.75C_4H_{10}O$
M_r = 695.36
Orthorhombic, *Pbca*
a = 12.958 (3) Å
b = 24.556 (5) Å
c = 25.753 (6) Å
V = 8195 (3) Å^3
Z = 8
D_x = 1.127 Mg m^{-3}
Mo *Kα* radiation
Cell parameters from 909 reflections
θ = 3.3–23.9°
μ = 0.07 mm^{-1}
T = 293 (2) K
Block, colorless
0.24 × 0.20 × 0.16 mm

Data collection

Bruker SMART CCD diffractometer
φ and ω scans
Absorption correction: none
20973 measured reflections
7092 independent reflections
3812 reflections with $I > 2\sigma(I)$
R_{int} = 0.050
θ_{max} = 25.0°
h = −11 → 15
k = −25 → 29
l = −23 → 30

Refinement

Refinement on F^2
$R[F^2 > 2\sigma(F^2)]$ = 0.062
$wR(F^2)$ = 0.188
S = 1.02
7092 reflections
534 parameters
H-atom parameters constrained
$w = 1/[\sigma^2(F_o^2) + (0.084P)^2]$ where $P = (F_o^2 + 2F_c^2)/3$
$(\Delta/\sigma)_{max} < 0.001$
$\Delta\rho_{max}$ = 0.59 e Å^{-3}
$\Delta\rho_{min}$ = −0.16 e Å^{-3}

Table 1
Selected geometric parameters (Å, °).

N1—C1	1.398 (3)	O2—C16	1.223 (3)
O1—C8	1.217 (3)		
C8—N1—C1	111.9 (2)	C11—N2—C7	106.0 (2)
C6—C7—C9—C10	144.2 (2)	C11—C10—C17—C18	41.1 (4)

All H atoms were placed in calculated positions, with C—H distances ranging from 0.93 to 0.98 Å and and an N—H distance of 0.86 Å. They were included in the refinement in riding motion approximation, with U_{iso} = 1.2 (1.5 for methyl) times U_{eq} of the carrier atom. The partial occupancy (0.75) solvent ether molecule is disordered over two sites, with occupancies of 0.5 and 0.25.

Data collection: *SMART* (Bruker, 1997); cell refinement: *SMART*; data reduction: *SAINT* (Bruker, 1997); program(s) used to solve structure: *SHELXS*97 (Sheldrick, 1997); program(s) used to refine structure: *SHELXL*97 (Bruker, 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*, Vol. 1, pp. 291–312, edited by A. Padwa. New York: Wiley.
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. & Mikamiy, Y. (1991). *Tetrahedron,* **47**, 6617–6622.
Sheldrick, G. M. (1997). *SHELXS*97 and *SHELXL*97. University of Göttingen, Germany.
Li, X.-F., Feng, Y.-Q., Hu, X.-F. & Xu, M. (2003). *Acta Cryst.* E**59**, o711–o712.