Acta Crystallographica Section E
Structure Reports
Online
ISSN 1600-5368

(*R*,*R*)-(−)-*trans*-1,4-Dioxaspiro[4,4]nonane-2,3-diylbis(diphenylmethanol)–*N*-allyl-*N*-propyl-3-oxocyclohexene-1-carboxamide (2/1)

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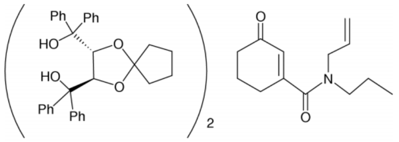
In the title inclusion compound, $2C_{33}H_{32}O_4\cdot C_{13}H_{19}NO_2$, there are two independent host–guest–host assemblies connected by O—H···O hydrogen bonds.

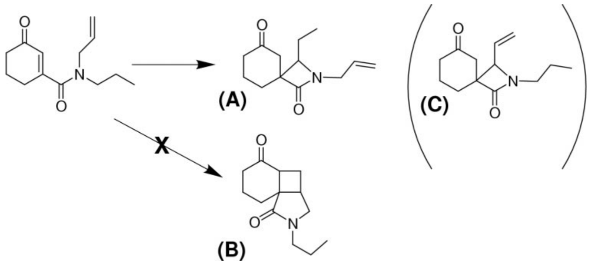
Received 8 December 2003
Accepted 5 January 2004
Online 17 January 2004

Comment

The enantioselective intramolecular photocyclization reaction of *N*-allyl-*N*-*R*-3-oxo-1-cyclohexenecarboxamides (*R* = Me, Et, ^{n}Pr, *etc*) in inclusion crystals was reported by Toda *et al.* (1995). The products obtained were tricyclic compounds, as the result of a [2 + 2]-photoreaction in the guest molecule, except for that with *R* = ^{n}Pr, which gave the spiro *β*-lactam (*A*), not the tricyclic compound (*B*). The crystal structure of the title compound, (I), has been determined to investigate the conformation of the guest amide (*R* = ^{n}Pr).



(I)



The *N*-propyl atoms C30/C31/C32 (C46/C47/C48) and the *N*-allyl group C33/C34/C35 (C43/C44/C45) show extraordinary large displacement parameters, suggesting positional disorder (Fig. 1). The molecular structure of the guest is essentially the same as that observed in the inclusion compound with the corresponding six-membered ring host, differing only in the size of the cycloalkane ring (Ohba & Miyamoto, 2003). The center-to-center distance between the C27═C28 and C34═C35 double bonds and that between bonds C40═C41 and C44═C45 are 4.24 (3) and 3.91 (3) Å, respectively. The plane of the allyl group is nearly perpendicular to the cyclohexene ring. The structure of the guest amide in (I) is not suitable for [2 + 2]-photocyclization and may give product (*C*), not (*A*).

Key indicators

Single-crystal X-ray study
T = 248 K
Mean *σ*(C–C) = 0.013 Å
R factor = 0.077
wR factor = 0.222
Data-to-parameter ratio = 7.0

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

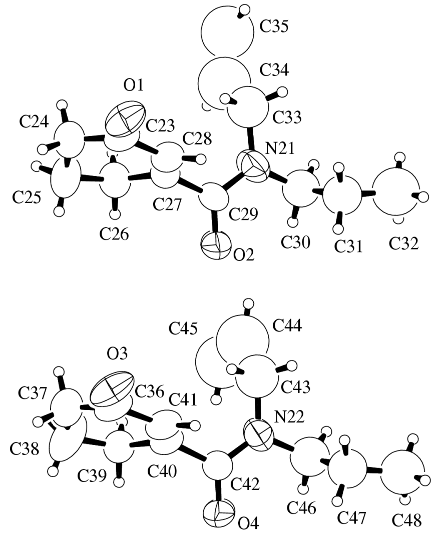


Figure 1
The structure of the two independent guest molecules, with the atom-numbering scheme. Displacement ellipsoids are plotted at the 30% probability level.

There are four independent host molecules in (I), which have similar conformations (Fig. 2). There is an intramolecular O—H···O hydrogen bond in each host molecule (Table 1). The guest molecule is also connected to the two host molecules by O—H···O hydrogen bonds (Fig. 3).

The crystal structure of (I) could not be solved by ordinary direct methods. This might be due to the large number of independent non-H atoms, 180. However, the Patterson-search method for a host molecule using atomic coordinates of the other inclusion compounds (Ohba *et al.*, 2000), and followed by direct methods (*DIRDIF*; Beurskens *et al.*, 1994) was successful.

Experimental

The title inclusion compound, (I), was prepared by Toda *et al.* (1995). Crystals suitable for X-ray analysis were grown from a diethyl ether solution.

Crystal data

$2C_{33}H_{32}O_4 \cdot C_{13}H_{19}NO_2$
M_r = 1206.53
Triclinic, $P1$
a = 34.738 (4) Å
b = 9.5934 (12) Å
c = 10.0715 (19) Å
α = 90.278 (12)°
β = 93.993 (11)°
γ = 90.021 (10)°
V = 3348.2 (9) Å^3
Z = 2
D_x = 1.197 Mg m^{-3}
Cu Kα radiation
Cell parameters from 25 reflections
θ = 25.1–29.1°
μ = 0.62 mm^{-1}
T = 248 K
Prism, colourless
0.68 × 0.30 × 0.20 mm

Data collection

Rigaku AFC-7*R* diffractometer
ω–2θ scans
Absorption correction: by integration (Coppens *et al.*, 1965)
T_{min} = 0.773, T_{max} = 0.889
11840 measured reflections
11148 independent reflections
9004 reflections with $I > 2\sigma(I)$
R_{int} = 0.094
θ_{max} = 75.0°
$h = -43 \rightarrow 43$
$k = -12 \rightarrow 12$
$l = 0 \rightarrow 12$
3 standard reflections every 150 reflections intensity decay: 12%

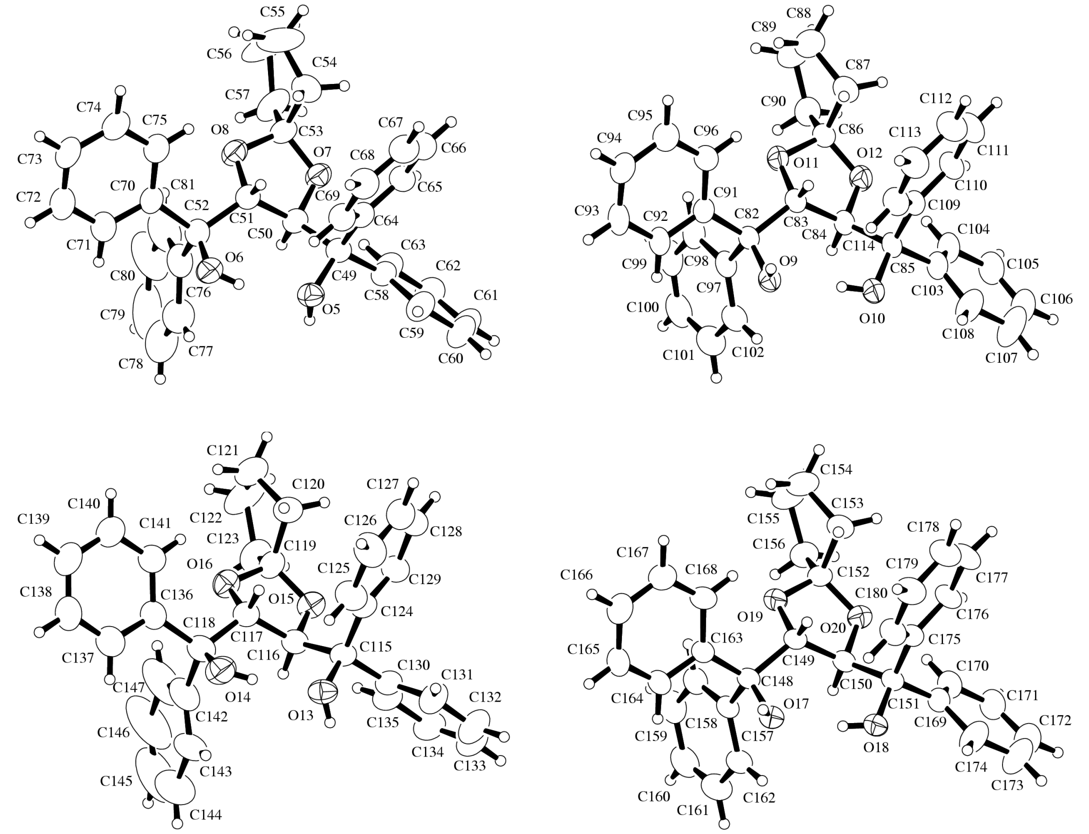


Figure 2
The structures of the four independent host molecules, with the atom-numbering scheme. Displacement ellipsoids are plotted at the 30% probability level.

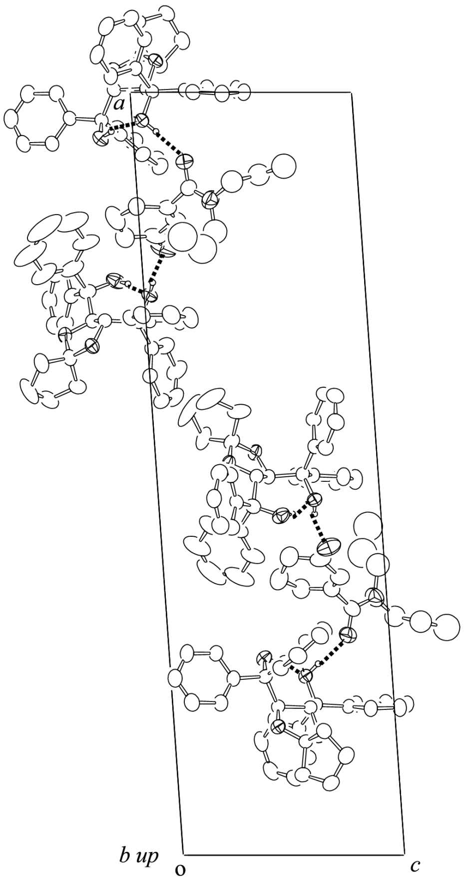


Figure 3
A projection of the crystal structure of (I), viewed along *b*. Broken lines indicate hydrogen bonds.

Refinement

Refinement on F^2
$R[F^2 > 2\sigma(F^2)] = 0.077$
$wR(F^2) = 0.222$
$S = 1.04$
11148 reflections
1588 parameters
H-atom parameters constrained
$w = 1/[\sigma^2(F_o^2) + (0.1612P)^2 + 0.6312P]$
where $P = (F_o^2 + 2F_c^2)/3$
$(\Delta/\sigma)_{\max} = 0.044$
$\Delta\rho_{\max} = 0.38$ e Å^{-3}
$\Delta\rho_{\min} = -0.43$ e Å^{-3}

Table 1
Hydrogen-bonding geometry (Å, °).

D—H···A	D—H	H···A	D···A	D—H···A
O5—H1···O1	0.82	1.95	2.734 (10)	160
O6—H2···O5	0.82	1.87	2.667 (8)	164
O9—H3···O2	0.82	1.94	2.700 (9)	155
O10—H4···O9	0.82	1.83	2.647 (8)	173
O13—H5···O3	0.82	1.91	2.719 (10)	168
O14—H6···O13	0.82	1.87	2.677 (9)	169
O17—H7···O4	0.82	1.89	2.709 (9)	172
O18—H8···O17	0.82	1.83	2.637 (7)	167

A relatively low completeness of 0.81 up to $\theta = 75°$ is due to the blind region ($\theta > 60°$) of the four-circle diffractometer, combined with a cooling apparatus. The C atoms of the *N*-propyl and *N*-allyl groups in the guest molecules, C30–C35 and C43–C48, were refined isotropically, since their extraordinary large displacement parameters suggested positional disorder. The geometry of the allyl groups was restrained, where distances C33—C34 (C43—C44), C34═C35 (C44═C45) and C33···C35 (C43···C45) are 1.50, 1.30, and 2.43 Å, respectively. For the atoms C34/C35/C44/C45, a common $U_{\rm iso}$ parameter was refined. The large, elongated displacement ellipsoids of atoms C55/C56 in the five-membered ring of one of the host molecules suggested positional disorder of these atoms due to the puckering motion of the ring. However, the split-atom refinement for C55/C56 failed. Atoms C78/C79/C80/C81 and C144/C145/C146/C147 of the phenyl rings of the host molecules show large displacement parameters, suggesting orientational disorder of the rings. The geometry of these phenyl rings was restrained. The hydroxyl H atoms were located in difference syntheses or were positioned assuming a hydrogen bond with a neighboring acceptor, and the O—H distances were restrained to be 0.82 Å with $U_{\rm iso}({\rm H}) = U_{\rm eq}({\rm O})$. The other H-atom positional parameters were calculated geometrically (C—H = 0.95 Å) and fixed with $U_{\rm iso}({\rm H}) = 1.2U_{\rm eq}$(parent C-atom). The absolute structure was assigned, based on the known absolute configuration of the host molecule derived from (+)-tartaric acid (Toda & Tanaka, 1988). The Flack (1983) parameter was −2.2 (4) prior to the merging of the Friedel pairs.

Data collection: *WinAFC Diffractometer Control Software* (Rigaku, 1999); cell refinement: *WinAFC Diffractometer Control Software*; data reduction: *TEXSAN* (Molecular Structure Corporation, 2001); program(s) used to solve structure: *DIRDIF* (Beurskens *et al.*, 1994); program(s) used to refine structure: *SHELXL*97 (Sheldrick, 1997); molecular graphics: *ORTEP*II (Johnson, 1976); software used to prepare material for publication: *TEXSAN*.

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